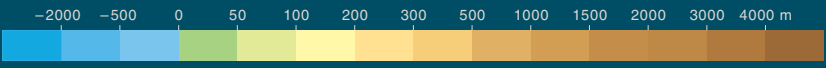
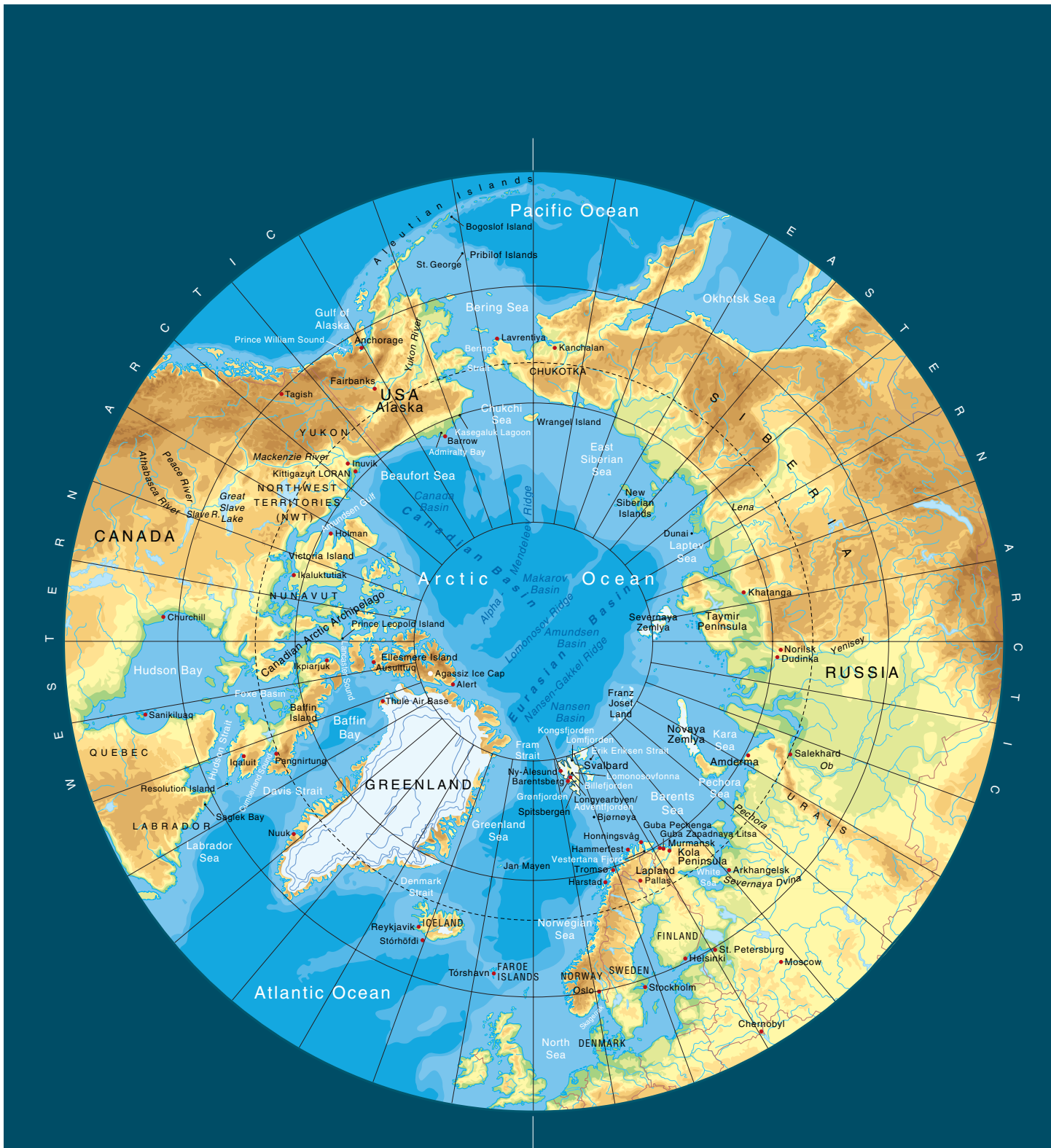


# **AMAP Assessment 2002:**

## Persistent Organic Pollutants in the Arctic



**Arctic Monitoring and Assessment Programme (AMAP)**



# **AMAP Assessment 2002:** *Persistent Organic Pollutants in the Arctic*

*Arctic Monitoring and Assessment Programme (AMAP), Oslo, 2004*



## AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic

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### Production

*Overall volume editors / scientific, technical, and linguistic editing*

Simon J. Wilson, Glen Packman

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---

*AMAP Working Group:*

Helgi Jensson (Chair, Iceland), Yuri Tsaturov (Vice-chair, Russia), David Stone (Canada), Ole Jensen (Denmark), Outi Mähönen (Finland), Gunnar Futsæter (Norway), Cynthia de Wit (Sweden), John Calder (USA), Jan-Idar Solbakken (Permanent Participants of the indigenous peoples organisations).

*AMAP Secretariat:*

Lars-Otto Reiersen, Vitaly Kimstach, Simon Wilson, Inger Utne.

*Indigenous peoples organizations, AMAP observing countries, and international organizations:*

Aleut International Association (AIA), Arctic Athabaskan Council (AAC), Gwitch'in Council International (GCI), Inuit Circumpolar Conference (ICC), Russian Association of Indigenous Peoples of the North (RAIPON), Saami Council.

France, Germany, Netherlands, Poland, United Kingdom.

Advisory Committee on Protection of the Sea (ACOPS), Association of World Reindeer Herders (AWRH), Circumpolar Conservation Union (CCU), European Environment Agency (EEA), International Arctic Science Committee (IASC), International Arctic Social Sciences Association (IASSA), International Atomic Energy Agency (IAEA), International Council for the Exploration of the Sea (ICES), International Federation of Red Cross and Red Crescent Societies (IFRCRCS), International Union for Circumpolar Health (IUCH), International Union for the Conservation of Nature (IUCN), International Union of Radioecology (IUR), Nordic Council of Ministers (NCM), Nordic Council of Parliamentarians (NCP), North Atlantic Marine Mammal Commission (NAMMCO), Northern Forum (NF), OECD Nuclear Energy Agency (OECD/NEA), Oslo and Paris Commissions (OSPARCOM), Standing Committee of Arctic Parliamentarians (SCAP), United Nations Economic Commission for Europe (UN ECE), United Nations Environment Programme (UNEP), World Health Organization (WHO), World Meteorological Organization (WMO), World Wide Fund for Nature (WWF).

*AMAP data centers:*

International Council for the Exploration of the Sea (ICES), Norwegian Institute for Air Research (NILU), Norwegian Radiation Protection Authority (NRPA), University of Alaska – Fairbanks (UAF).



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## Preface

*This assessment report details the results of the 2002 AMAP assessment of Persistent Organic Pollutants (POPs) in the Arctic. It builds upon the previous AMAP POPs assessment that was presented in 'AMAP Assessment Report: Arctic Pollution Issues' that was published in 1998.*

*The Arctic Monitoring and Assessment Programme (AMAP) is a group working under the Arctic Council.*

*The Arctic Council Ministers have requested AMAP to:*

- *produce integrated assessment reports on the status and trends of the conditions of the Arctic ecosystems;*
- *identify possible causes for the changing conditions;*
- *detect emerging problems, their possible causes, and the potential risk to Arctic ecosystems including indigenous peoples and other Arctic residents; and to*
- *recommend actions required to reduce risks to Arctic ecosystems.*

*The Ministers have placed special priority on the potential impacts of contaminants on the health of Arctic residents, including the combined effects of mixtures of contaminants acting together with other potential stressors.*

*This report is one of five detailed assessment reports that provide the accessible scientific basis and validation for the statements and recommendations made in the second AMAP State of the Arctic Environment report, 'Arctic Pollution 2002' that was delivered to Arctic Council Ministers at their meeting in Inari, Finland in October 2002. It includes extensive background data and references to the scientific literature, and details the sources for figures reproduced in the 'Arctic Pollution 2002' report. Whereas the 'Arctic Pollution 2002' report contains recommendations that specifically focus on actions aimed at improving the Arctic environment, the conclusions and recommendations presented in this report also cover issues of a more scientific nature, such as proposals for filling gaps in knowledge, and recommendations relevant to future monitoring and research work, etc.*

*To allow readers of this report to see how AMAP interprets and develops its scientifically-based assessment product in terms of more action-orientated conclusions and recommendations, the 'Executive Summary of the Arctic Pollution 2002 Ministerial Report', which also covers other priority issues (Heavy Metals, Radioactivity, Human Health, and Climate Change Effects on Contaminant Pathways), is reproduced in this report on pages xi to xv.*

*The AMAP assessment is not a formal environmental risk assessment. Rather, it constitutes a compilation of current knowledge about the Arctic region, an evaluation of this information in relation to agreed criteria of environmental quality, and a statement of the prevailing conditions in the area. The assessment presented in this report was prepared in a systematic and uniform manner to provide a comparable knowledge base that builds on earlier work and can be extended through continuing work in the future.*

*The AMAP scientific assessments are prepared under the direction of the AMAP Assessment Steering Group.*

*The product is the responsibility of the scientific experts involved in the preparation of the assessment. Lead countries for the AMAP Persistent Organic Pollutants Assessment under AMAP phase II were Canada and Sweden. The assessment is based on work conducted by a large number of scientists and experts from the Arctic countries (Canada, Denmark/Greenland/Faroe Islands, Finland, Iceland, Norway, Russia, Sweden, and the United States), together with contributions from indigenous peoples organizations, from other organizations, and from experts in other countries.*

*AMAP would like to express its appreciation to all of these experts, who have contributed their time, effort, and data; and especially to the lead experts who coordinated the production of this report, and to referees who provided valuable comments and helped ensure the quality of the report. A list of the main contributors is included in the acknowledgements on page viii of this report. The list is not comprehensive. Specifically, it does not include the many national institutes, laboratories and organizations, and their staff, which have been involved in the various countries. Apologies, and no lesser thanks are given to any individuals unintentionally omitted from the list. Special thanks are due to the lead authors responsible for the preparation of the various chapters of this report.*

*The support of the Arctic countries is vital to the success of AMAP. AMAP work is essentially based on ongoing activities within the Arctic countries, and the countries also provide the necessary support for most of the experts involved in the preparation of the assessments. In particular, AMAP would like to express its appreciation to Canada and Sweden for undertaking a lead role in supporting the Persistent Organic Pollutants assessment. Special thanks are also offered to the Nordic Council of Ministers for their financial support to the work of AMAP, and to sponsors of the project on 'Persistent Toxic Substances (PTS) and Food Security of Indigenous Peoples of the Russian North' and other bilateral and multilateral projects that have delivered data for use in this assessment.*

*The AMAP Working Group that was established to oversee this work and the AMAP persistent organic pollutants assessment group are pleased to present its assessment.*

*Helgi Jensson  
AMAP Working Group Chair*

*Derek C.G. Muir  
AMAP Persistent Organic Pollutants Assessment  
co-lead (Canada)*

*Cynthia A. de Wit  
AMAP Persistent Organic Pollutants Assessment  
co-lead (Sweden)*

*Lars-Otto Reiersen  
AMAP Executive Secretary*

*Oslo, January 2004*

\* AMAP, 1998. AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. xii + 859 pp.

\*\* AMAP, 2002. Arctic Pollution 2002: Persistent Organic Pollutants, Heavy Metals, Radioactivity, Human Health, Changing Pathways. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. xii + 112 pp.

# Acknowledgements

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The AMAP Working Group would like to thank the following persons for their work in preparing the AMAP 2002 Persistent Organic Pollutants Assessment.

*Assessment Leads:*

Derek C.G. Muir, National Water Research Institute, Environment Canada, Burlington, Canada  
 Cynthia A. de Wit, Institute of Applied Environmental Research (ITM), Stockholm University, Stockholm, Sweden

*Editors/Lead Authors:*

Cynthia A. de Wit  
 Aaron T. Fisk, Warnell School of Forest Resources, University of Georgia, Athens, Georgia, USA  
 Karen E. Hobbs, National Water Research Institute, Environment Canada, Burlington, Canada  
 Derek C.G. Muir

*Contributing Authors:*

Geir W. Gabrielsen, Norwegian Polar Institute, Tromsø, Norway  
 Roland Kallenborn, Norwegian Institute for Air Research, Kjeller, Norway  
 Margaret M. Krahn, Northwest Fisheries Science Centre, National Oceanic and Atmospheric Administration/National Marine Fisheries Service, Seattle, WA, USA  
 Ross J. Norstrom, Canadian Wildlife Service, Hull, Canada  
 Janneche U. Skaare, National Veterinary Institute/Norwegian School of Veterinary Science, Oslo, Norway

*Contributors:*

Paul Becker, National Oceanic and Atmospheric Administration/National Institute of Standards and Technology, Charleston, SC, USA  
 Kimberlee Beckmen, Alaska Department of Fish and Game, Fairbanks, AK, USA  
 John Berge, Norwegian Institute for Water Research, Oslo, Norway  
 Terry Bidleman, Meteorological Service of Canada, Environment Canada, Toronto, Canada  
 Anders Bignert, Swedish Natural History Museum, Stockholm, Sweden  
 Pierrette Blanchard, Meteorological Service of Canada, Environment Canada, Toronto, Canada  
 Jennie Bolton, Northwest Fisheries Science Centre, National Oceanic and Atmospheric Administration/National Marine Fisheries Service, Seattle, WA, USA  
 Katrine Borgå, Norwegian Polar Institute, Tromsø, Norway  
 Birgit Braune, Canadian Wildlife Service, Environment Canada, Ottawa, ON, Canada  
 Knut Breivik, Norwegian Institute for Air Research, Kjeller, Norway  
 Eva Bromström-Lundén, Swedish Environmental Research Institute (IVL), Gothenburg, Sweden  
 Guttorm Christensen, Akvaplan-niva, Tromsø, Norway  
 David Cleverly, US Environmental Protection Agency, Washington, D.C., USA  
 Salve Dahle, Akvaplan-niva, Tromsø, Norway  
 Maria Dam, Food and Environmental Agency, Torshavn, Faroe Islands  
 Marlene Evans, National Water Research Institute, Environment Canada, Saskatoon, Canada  
 Thomas Evans, US Fish and Wildlife Service, Anchorage, AK, USA  
 Anita Evenset, Akvaplan-niva, Tromsø, Norway  
 Jesse Ford, Oregon State University, Corvallis, OR, USA  
 Örjan Gustafsson, Institute of Applied Environmental Research (ITM), Stockholm University, Stockholm, Sweden  
 Mark Hermanson, University of Pennsylvania, Philadelphia, PA, USA  
 Dorte Herzke, Norwegian Institute for Air Research, Tromsø, Norway  
 Paul Helm, Freshwater Institute, Department of Fisheries and Oceans, Winnipeg, Canada  
 Brendan Hickie, Trent University, Peterborough, Canada  
 Paul Hoekstra, Department of Environmental Biology, University of Guelph, Guelph,, Canada  
 Hayley Hung, Meteorological Service of Canada, Environment Canada, Toronto, Canada  
 Michael Ikonomu, Institute of Ocean Sciences, Department of Fisheries and Oceans, Sidney, BC, Canada  
 Björn Munro Jenssen, Norwegian University of Science and Technology, Trondheim, Norway  
 Poul Johansen, National Environmental Research Institute, Roskilde, Denmark



Kevin Jones, Lancaster University, Lancaster, UK  
 Even Jørgensen, Norwegian Institute for Nature Research, Tromsø, Norway  
 Lars Kleivane, National Veterinary Institute, Oslo, Norway  
 Markku Korhonen, Finnish Environment Institute, Helsinki, Finland  
 John Kucklick, National Institute of Standards and Technology, Charleston, NC, USA  
 Henrik Kylin, Department of Environmental Assessment, Swedish University of Agricultural Sciences, Uppsala, Sweden  
 Sirkka Leppänen, Finnish Meteorological Institute, Finland  
 Robert Letcher, Great Lakes Institute of Environmental Research, University of Windsor, Windsor, Canada  
 Yi Fan Li, Meteorological Service of Canada, Environment Canada, Toronto, Canada  
 Elisabeth Lie, National Veterinary Institute/Norwegian School of Veterinary Science, Oslo, Norway  
 Rob Macdonald, Institute of Ocean Sciences, Department of Fisheries and Oceans, Sidney, BC, Canada  
 Jaakko Mannio, Finnish Environment Institute, Helsinki, Finland  
 Katie Matthews, University of Pennsylvania, Philadelphia, PA, USA  
 Sandra Meijer, Lancaster University, Lancaster, UK  
 Sergei Melnikov, Regional Centre 'Monitoring of the Arctic', St. Petersburg, Russia  
 Keith Mueller, US Fish and Wildlife Service, USA  
 Madeleine Nyman, Finnish Game and Fisheries Research Institute, Helsinki, Finland  
 Todd O'Hara, University of Alaska Fairbanks, Fairbanks, AK, USA  
 Kristina Olsson, Department of Analytical and Marine Chemistry, Chalmers Institute of Technology, Gothenburg, Sweden  
 Susan Polischuk Blum, University of Saskatchewan, Saskatoon, SK, Canada  
 Frank Riget, National Environmental Research Institute, Roskilde, Denmark  
 Pat Roach, Indian and Northern Affairs, Whitehorse, Canada  
 Neil Rose, Environmental Change Research Centre, University College, London, UK  
 Courtney Sandau, National Center for Environmental Health, Atlanta, GA, USA  
 Vladimir Savinov, Akvaplan-niva, Tromsø, Norway  
 Tatiana Savinova, Akvaplan-niva, Tromsø, Norway  
 Jeff Short, Alaska Fisheries Science Centre, National Marine Fisheries Service, Juneau, AK, USA  
 Detleff Schultz-Bull, Institute of Marine Research, University of Kiel, Kiel, Germany  
 Anna Sobek, Institute of Applied Environmental Research (ITM), Stockholm University, Stockholm, Sweden  
 Trond Skotvold, Akvaplan-niva, Tromsø, Norway  
 Eric Steig, University of Washington, Seattle, WA, USA  
 Gary Stern, Freshwater Institute, Department of Fisheries and Oceans, Winnipeg, Canada  
 William Strachan, National Water Research Institute, Environment Canada, Burlington, Canada  
 Jorundur Svavarsson, Institute of Biology, University of Iceland, Reykjavik, Iceland  
 Sheryl Tittlemier, Food Research Division, Health Canada, Ottawa, Canada  
 Bert van Bavel, Department of Natural Sciences, Örebro University, Örebro, Sweden  
 Sergei Vlasov, Regional Centre 'Monitoring of the Arctic', St. Petersburg, Russia  
 Frank Wania, University of Toronto, Toronto, Canada  
 Carla Willetto, Ilasgavik College, Barrow, AK, USA  
 Hans Wolkers, Norwegian Polar Institute, Tromsø, Norway  
 Xiaowa Wong, National Water Research Institute, Environment Canada, Burlington, Canada

*Reviewers:*

Peter Ross, Institute of Ocean Sciences, Department of Fisheries and Oceans, Sidney, BC, Canada  
 Jeff Vos, National Institute of Public Health and Environment (RIVM), Bilthoven, the Netherlands  
 Barry Hargrave, Bedford Institute of Oceanography, Department of Fisheries and Oceans, Dartmouth, NS, Canada  
 Crispin Halsall, Lancaster University, Lancaster, UK

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## Executive Summary to the *Arctic Pollution 2002* Ministerial Report

The Arctic Monitoring and Assessment Programme (AMAP) was established in 1991 to monitor identified pollution risks and their impacts on Arctic ecosystems. In 1997 the first AMAP report, *Arctic Pollution Issues: A State of the Arctic Environment Report*\* was published.

The assessment showed that the Arctic is closely connected to the rest of the world, receiving contaminants from sources far outside the Arctic region. The report was welcomed by the Arctic Council Ministers, who agreed to increase their efforts to limit and reduce emissions of contaminants into the environment and to promote international cooperation in order to address the serious pollution risks reported by AMAP.

The AMAP information greatly assisted the negotiation of the protocols on persistent organic pollutants (POPs) and heavy metals to the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution (LRTAP Convention). They also played an important role in establishing the need for a global agreement on POPs, which was concluded in 2001 as the Stockholm Convention. Persistence, long-range transport, and bioaccumulation are screening criteria under both the POPs protocol and the Stockholm Convention, to be applied to proposals to add substances to the agreements. Information from AMAP will be useful in this context in showing whether persistent substances are accumulating in the Arctic and are therefore candidates for control, and also in assessing the effectiveness of the agreements.

The Arctic Council also decided to take cooperative actions to reduce pollution of the Arctic. As a direct follow up of the AMAP reports, the Arctic Council Action Plan to Eliminate Pollution of the Arctic (ACAP) was created to address sources identified through AMAP. ACAP was approved in 2000 and several projects have begun. The AMAP information was also used in establishing priorities for the Arctic Regional Programme of Action to Prevent Pollution from Landbased Sources (RPA), developed by the working group on Protection of the Arctic Marine Environment (PAME), and adopted by the Arctic Council in 1998.

After the first assessment, AMAP was asked to continue its activities and provide an updated assessment on persistent organic pollutants (POPs), heavy metals, radioactivity, human health, and pathways in 2002. Five scientific reports and a plain-language report have been prepared. This Executive Summary provides the main conclusions and recommendations of the 2002 AMAP assessments.

### International Agreements and Actions

As described above, the LRTAP Convention protocols and the Stockholm Convention are essential instruments for reducing contamination in the Arctic. However, they cannot have any effect until they are ratified and implemented.

*It is therefore recommended that:*

- The UN ECE LRTAP Protocols on Heavy Metals and POPs be ratified and implemented.
- The Stockholm Convention on POPs be ratified and implemented.

Specific recommendations for monitoring activities in support of these agreements are included in subsequent sections.

### Persistent Organic Pollutants

The POPs assessment addresses several chemicals of concern, including both substances that have been studied for some time and chemicals that have only recently been found in the environment.

The 1997 AMAP assessment concluded that levels of POPs in the Arctic environment are generally lower than in more temperate regions. However, several biological and physical processes concentrate POPs in some species and at some locations, producing some high levels in the Arctic.

The present AMAP assessment has found that the conclusions and recommendations of the first assessment remain valid. In addition:

*It has clearly been established that:*

**Certain Arctic species, particularly those at the upper end of the marine food chain as well as birds of prey, carry high levels of POPs.** Marine mammals, such as polar bear, Arctic fox, long-finned pilot whale, killer whale, harbor porpoise, minke whale, narwhal, beluga, harp seal and northern fur seal, some marine birds including great skua, great black-backed gull and glaucous gull, and birds of prey such as peregrine falcon, tend to carry the highest body burdens.

**Most of the total quantity of POPs found in the Arctic environment is derived from distant sources.** The POPs are transported to the Arctic by regional and global physical processes, and are then subjected to biological mechanisms that lead to the high levels found in certain species. Several potential source regions have now been identified within and outside of the Arctic. A better understanding of local re-distribution mechanisms has also emphasized the important potential role of local processes and sources in determining observed geographical variability.

*There is evidence that:*

**Adverse effects have been observed in some of the most highly exposed or sensitive species in some areas of the Arctic.** Several studies have now been completed on a number of Arctic species, reporting the types of effects that have been associated in non-Arctic species with chronic exposure to POPs, of which there are several examples. Reduced immunological response in polar bears and northern fur seals has led to increased susceptibility

\* AMAP, 1997. *Arctic Pollution Issues: A State of the Arctic Environment Report*. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, xii+188 pp. and AMAP, 1998. *AMAP Assessment Report: Arctic Pollution Issues*. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, xii+859 pp.



to infection. Immunological, behavioral, and reproductive effects as well as reduced adult survival has been found in glaucous gulls. Peregrine falcons have suffered from eggshell thinning and reproductive effects. Reproductive effects in dogwhelks are associated with exposure to tributyltin.

*It is therefore recommended that:*

- AMAP be asked to further enhance studies aimed at detecting effects in Arctic species relating to exposure to high levels of POPs and to integrate this information with an understanding of general population effects and health. Without this understanding, it will not be possible to assess whether proposed and existing controls can be expected to afford the necessary protection (e.g., under the LRTAP and Stockholm agreements).

*There is evidence that:*

The levels of some POPs are decreasing in most species and media in the Arctic, but the rates vary in extent, location and media or species being studied. The decreases can be related to reduced release to the environment. For example, declines in alpha-HCH in air closely follow decreases in global usage, but declines in marine biota are much slower due to a huge reservoir of the substance in the global oceans.

For other POPs, declines are minimal and some levels are actually increasing, despite low current emissions. This illustrates the long period that may pass between the introduction of controls and the resulting decrease in levels in biota, as has been observed for PCBs, toxaphene, and beta-HCH.

*It is therefore recommended that:*

- AMAP be asked to continue trend monitoring of POPs in key indicator media and biota. This will enable assessment of whether the measures taken in the LRTAP Protocol and the Stockholm Convention are being effective in driving down POPs levels in the Arctic.

*There is evidence that:*

POPs substances other than those included in the LRTAP Protocol and Stockholm Convention may be at or approaching levels in the Arctic that could justify regional and global action. For example, levels of the brominated flame retardants such as polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCNs), and some current-use pesticides such as endosulfan have been monitored in Arctic air and biota. PBDEs are increasing in the Canadian Arctic.

*It is therefore recommended that:*

- AMAP be asked to maintain a capacity to detect current-use POPs in the Arctic. This will help ensure that Arctic States have an early opportunity to respond to a trend indicating Arctic accumulation, thus allowing a proactive approach to minimize the contamination rather than having to respond to a more serious situation later.

## Heavy Metals

The heavy metals assessment focuses on mercury, lead, and cadmium.

*It has clearly been established that:*

In the Arctic, mercury is removed from the atmosphere and deposits on snow in a form that can become bioavailable. Enhanced deposition occurs in the Arctic. This recently discovered process is linked to polar sunrise, and is unique to high latitude areas. The resulting enhanced deposition may mean that the Arctic plays a previously unrecognized role as an important sink in the global mercury cycle.

*There is evidence that:*

Some of the deposited mercury is released to the environment at snowmelt, becoming bioavailable at the onset of animal and plant reproduction and rapid growth. Although poorly understood, this process may be the chief mechanism for transferring atmospheric mercury to Arctic food webs.

*It is therefore recommended that:*

- The Arctic Council encourage expanded and accelerated research on critical aspects of the mercury cycle and budget in the Arctic. Such research should include long-range transport, mercury deposition mechanisms, processes leading to biological exposure and effects, and the influence of climate variability and change on these processes.

*There is evidence that:*

Despite substantial mercury emission reductions in North America and Western Europe during the 1980s, global mercury emissions may, in fact, be increasing. Mercury emissions from waste incineration are likely underestimated. The burning of coal in small-scale power plants and residential heaters, principally in Asia, are major potential sources of current mercury emissions. These emissions are likely to increase significantly due to economic and population growth in this region.

*It is therefore recommended that:*

- The Arctic Council promote efforts at global, regional, and national levels to quantify all sources of mercury and report results in a consistent and regular manner to improve emission inventories. Particular efforts should focus on measuring contributions made by the burning of coal for residential heating and small-scale power plants as well as by waste incineration.

*There is strong evidence that:*

There is a trend of increasing mercury levels in marine birds and mammals in the Canadian Arctic, and some indications of increases in West Greenland. The effects of these levels are not well understood. However, there are also examples of stable or decreasing levels in other regions, perhaps indicating the importance of local or regional processes.

*It is therefore recommended that:*

- AMAP be asked to continue temporal trend monitoring and the assessment of effects of mercury in key indicator media and biota. This will enable assessment of whether the measures taken in the LRTAP Protocol are being effective in driving down mercury levels in the Arctic.

*There is evidence that:*

**Current mercury exposures pose a health risk to some people and animals in the Arctic.** These risks include subtle neurobehavioral effects.

*It is therefore recommended that:*

- **In view of the fact that reducing exposure to mercury can only be addressed by regional and global action to reduce worldwide emissions, and acknowledging the assessment for global action undertaken by UNEP and its resulting proposals, the Arctic Council take appropriate steps to ensure that Arctic concerns are adequately addressed and to promote the development of regional and global actions.**

*It has clearly been established that:*

**Dramatic reduction in the deposition of atmospheric lead has occurred in Arctic regions where the use of leaded gasoline is banned.** Arctic-wide elimination of leaded gasoline use will reduce lead exposure in other regions of the Arctic. Although levels in wildlife and fish have not measurably declined, likely reflecting continued uptake from the large reservoir of lead deposited in soils and sediments, lead levels in the environment are expected to diminish over time if current trends continue.

*It is therefore recommended that:*

- **The Arctic Council support continued efforts to eliminate the use of leaded gasoline in all Arctic regions.**

*It has clearly been established that:*

**Certain regions of the Arctic contain elevated lead levels in the environment because of past or current use of lead shot by hunters.** Even though lead shot is banned in Alaska, for example, lead blood levels in endangered US populations of Steller's eiders are above known avian toxicity thresholds for lead poisoning, which may be responsible for observed reduced breeding success. In Greenland, lead shot appears to be a significant source of human dietary exposure to lead.

*It is therefore recommended that:*

- **The Arctic Council encourage a complete ban on the use of lead shot in the Arctic, and that enforcement be improved.**

*There is evidence that:*

**Cadmium levels in some seabirds is high enough to cause kidney damage.** Monitoring data on cadmium in the abiotic and biotic environment to date provide no conclusive evidence of trends or effects. However, cadmium accumulates in birds and mammals and not enough is known about possible effects.

*It is therefore recommended that:*

- **The monitoring of cadmium in the Arctic be continued to support human exposure estimates.**

*There is evidence that:*

**Levels of platinum, palladium, and rhodium have increased rapidly in Greenland snow and ice since the 1970s.** These elements are used in automobile catalytic converters to reduce hydrocarbon pollution. The tox-

icity and bioaccumulation potential of these elements are largely unknown, which prevents assessment of their potential impact in the Arctic.

*It is therefore recommended that:*

- **AMAP be asked to consider the need to monitor trends of platinum, palladium, and rhodium in the Arctic.**

## Radioactivity

The radioactivity assessment addresses man-made radionuclides and radiation exposures deriving from human activities.

*It has clearly been established that:*

**In general, levels of anthropogenic radionuclides in the Arctic environment are declining.** Most of the radioactive contamination in the Arctic land environment is from the fallout from nuclear weapons testing during the period 1945 to 1980. In some areas, the fallout from the Chernobyl accident in 1986 is a major source. For the Arctic marine environment, a major source of radionuclides is the releases from European reprocessing plants at Sellafield and Cap de la Hague.

**However, releases from the reprocessing plants have resulted in increases in levels of some radionuclides in the European Arctic seas during recent years, in particular technetium-99 and iodine-129.** The present doses to the population are low but the present levels of technetium in some marine foodstuffs marketed in Europe are above the EU intervention levels for food to infants and are close to the intervention level for adults.

*The technetium information adds further weight to the recommendation made by AMAP to the Arctic Council in Barrow in 2000 that:*

- **'The Arctic Council encourage the United Kingdom to reduce the releases from Sellafield to the marine environment of technetium, by implementing available technology.'**

*There is evidence that:*

**Radionuclides in sediments are now a source of plutonium and cesium-137 to the Arctic.** Earlier releases such as those from Sellafield that have deposited in sediments in the Irish Sea, especially cesium-137 and plutonium, have been observed to remobilize so that these deposits are now acting as sources to the Arctic. Thus, even if operational releases of these radionuclides from reprocessing plants are reduced, releases from environmental sources such as contaminated sediment in the Irish Sea and the Baltic Sea will be observed in the Arctic.

*It is therefore recommended that:*

- **The Arctic Council support a more detailed study on the remobilization of radionuclides from sediment and its potential effect on the Arctic.**

*It is apparent that:*

**There is continuing uncertainty about the amount of radionuclides present at a number of sources and potential sources in the Arctic.** Access to information about civilian and military sources continues to be a problem.

*It is therefore recommended that:*

- The Arctic Council promote more openness of restricted information from any sources.

*It has clearly been established that:*

Compared with other areas of the world, the Arctic contains large areas of high vulnerability to radionuclides. This is due to the characteristics of vegetation, animals, human diets, and land- and resource-use practices. On land in the AMAP area, there is considerable variation in vulnerability due to differences in these characteristics. In contrast, vulnerability associated with releases of radionuclides to the marine environment is relatively uniform and similar to that for other areas of the world. Maps of vulnerable areas, when combined with deposition maps, can be useful in an accident situation. The information on vulnerability is of importance for emergency planning.

*It is therefore recommended that:*

- AMAP be asked to clarify the vulnerability and impact of radioactivity on the Arctic environment and its consequences for emergency preparedness planning.

*It is apparent that:*

When performing risk reducing actions, close links to assessment programs are important and interventions should be prioritized in relation to the extent and magnitude of threats posed by nuclear activities, especially in respect to accidents. Interventions themselves can also have negative effects for humans and the environment, and careful judgments have to be made together with environmental impact assessments prior to carrying out a project. It is the view of AMAP that this has not always been done in interventions adopted to date.

*It is therefore recommended that:*

- Risk and impact assessment programmes be performed prior to implementation of action to reduce risk.
- Risk and impact assessments, including accident scenarios, be performed with regard to the transport of nuclear waste and fuel within the Arctic and nearby areas and with regard to planned storage and reprocessing within the Arctic and nearby areas.

*It is apparent that:*

The protection of the environment from the effects of radiation deserves specific attention. The current system of radiological protection is entirely based on the protection of human health. This approach can fail to address environmental damage in areas such as the Arctic that have low human population densities. Recently, an international consensus has emerged that the rapid development of a system and a framework for the protection of the environment needs further effort. The International Union of Radioecology (IUR), with support from AMAP, was one of the first international organizations to promote and present such a system and framework.

*It is therefore recommended that:*

- AMAP be asked to take an active part in the continued efforts to address environmental protection, with special responsibility for the Arctic. This should include the task of adding the need for protection of the environment into monitoring strategies and assessment tools.

*It is noted that:*

Since the previous AMAP assessment, nuclear safety programmes have been implemented in Russia at some nuclear power plants and other nuclear installations relevant to the Arctic.

*It is therefore recommended that:*

- The Arctic Council continue its cooperation with Russia to improve the safety and safeguarding of nuclear installations and waste sites.

## Human Health

The human health assessment considered health risks associated with exposure to contaminants in relation to other lifestyle factors determining health. This assessment has extended geographical coverage and confirmed the conclusions and recommendations from the first assessment.

*It has clearly been established that:*

The highest Arctic exposures to several POPs and mercury are faced by Inuit populations in Greenland and Canada. These exposures are linked mainly to consumption of marine species as part of traditional diets. Temporal trends of human exposures to POPs have so far not been observed. Exposure to mercury has increased in many Arctic regions while exposure to lead has declined.

*It is therefore recommended that:*

- The monitoring of human exposure to mercury, relevant POPs, including dioxins and dioxin-like compounds and other chemicals of concern, be continued in order to help estimate risk, further elaborate geographical trends, and begin to establish time trends of exposure.

*There is evidence that:*

Subtle health effects are occurring in certain areas of the Arctic due to exposure to contaminants in traditional food, particularly for mercury and PCBs. The evidence suggests that the greatest concern is for fetal and neonatal development. In the Arctic, human intake of substances with dioxin-like effects is a matter of concern, confirmed by recent results from Greenland. Increasing human exposure to current-use chemicals has been documented, for example for brominated flame retardants. Others such as polychlorinated naphthalenes (PCN) are expected to be found in human tissues. Some of these compounds are expected to add to the total dioxin activity in humans. The AMAP human health monitoring program includes a number of measures of effects, ranging from biomarkers of effects at the molecular level to epidemiological outcomes.

*It is therefore recommended that:*

- The human health effects program developed by AMAP be more extensively applied in order to provide a better base for human risk assessment especially concerning pre- and neonatal exposures.

*It has clearly been established that:*

In the Arctic, diet is the main source of exposure to most contaminants. Dietary intake of mercury and PCBs ex-

ceeds established national guidelines in a number of communities in some areas of the Arctic, and there is evidence of neurobehavioral effects in children in some areas. In addition, life-style factors have been found to influence the body burden of some contaminants, for example cadmium exposure from smoking. In the Arctic region, a local public health intervention has successfully achieved a reduction of exposure to mercury by providing advice on the mercury content of available traditional foods. The physiological and nutritional benefits of traditional food support the need to base dietary recommendations on risk-benefit analyses. The health benefits of breast-feeding emphasize the importance of local programs that inform mothers how adjustments within their traditional diet can reduce contaminant levels in their milk without compromising the nutritional value of their diet.

*It is therefore recommended that:*

- **In locations where exposures are high, carefully considered and balanced dietary advice that takes risk and benefits into account be developed for children and men and women of reproductive age.** This advice should be developed by national and regional public health authorities in close consultation with affected communities.
- **Studies of the nutrient and contaminant content of traditional food items be promoted in order to assess their benefits and to estimate exposures as a basis for public health interventions.**
- **Breast-feeding continue to be recognized as a practice that benefits both mother and child.** Nonetheless, if contaminant levels increase or more information indicates increased risk, the potential need for restrictions should continue to be evaluated.

*It is noted that:*

From the Arctic human health perspective, it is of utmost importance that considerations for global actions against POPs and mercury take into account the concerns for Arctic human health. The Stockholm Convention and the LRTAP protocols should be properly monitored in the Arctic to determine whether their implementation is effective in protecting human health.

*It is therefore recommended that:*

- **AMAP participate in the global monitoring of human exposure to be established under the Stockholm Convention on POPs.**
- **The Arctic Council monitor proposals for global action on mercury being undertaken by UNEP, and contribute as necessary to ensure that Arctic concerns related to human health are adequately addressed.**

## Changing pathways

The assessment of changing pathways provides an introduction to the types of changes on contaminants pathways to, within, and from the Arctic that might be expected as a result of global climate change and variability.

*There is evidence that:*

The routes and mechanisms by which POPs, heavy metals, and radionuclides are delivered to the Arctic are strongly influenced by climate variability and global climate change. These pathways are complex, interactive systems involving a number of factors, such as temperature, precipitation, winds, ocean currents, and snow and ice cover. Pathways within food webs and the effects on biota may also be modified by changes to climate. Studies using global change scenarios have indicated the potential for substantial changes in atmospheric and oceanographic pathways that carry contaminants to, within, and from the Arctic. These effects mean that climate-related variability in recent decades may be responsible at least in part for some of the trends observed in contaminant levels.

*It is therefore recommended that:*

- **AMAP be asked to further investigate how climate change and variability may influence the ways in which POPs, heavy metals, and radionuclides move with respect to the Arctic environment and accumulate in and affect biota.** This will enable Arctic States to better undertake strategic planning when considering the potential effectiveness of present and possible future national, regional, and global actions concerning contaminants.





## Chapter 1

## Introduction

The Persistent Organic Pollutants (POPs) chapter of the previous AMAP Assessment Report (de March *et al.*, 1998) presented the data that were available as of 1996 on POPs in Arctic air, seawater, sediments, soils, and plants as well as terrestrial, freshwater and marine biota. Since then, new data have emerged on spatial and temporal trends, as well as biological effects, which address some of the knowledge gaps identified in that assessment. The focus of this assessment will therefore mainly be on the results of studies conducted since 1996 and any older data that were not in the initial report.

The previous AMAP POPs assessment also helped stimulate regional and global initiatives to identify and ban selected POPs. In 1998, the United Nations Economic Commission for Europe (UNECE) negotiated a Protocol on POPs under the Convention on Long-range Transboundary Air Pollution to which all eight circum-polar countries were signatories ([www.unece.org/env/lrtap/](http://www.unece.org/env/lrtap/)). In 2001, the United Nations Environment Programme (UNEP) completed global negotiations on banning of certain POPs with the signing of the Stockholm Convention on Persistent Organic Pollutants in May 2001 (UNEP, 2001; <http://irptc.unep.ch/pops/>). Particularly relevant to this assessment are the criteria for identifying and listing chemicals under the convention. These are summarized in Table 1.1 and serve to define what is meant by 'POPs'. The initial chemicals listed under the Stockholm Convention were aldrin, chlordane, dieldrin,

dichlorodiphenyltrichloroethane (DDT), endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (UNEP, 2001).

With global agreement that chemicals with characteristics of POPs include presence in locations 'distant from sources' and 'monitoring data showing that long-range environmental transport of the chemical ... may have occurred', the Arctic has become an important indicator region for assessment of persistence and bioaccumulation. The Arctic environment is well suited as a region in which to evaluate POPs. Cold conditions favour persistence of POPs relative to temperate or tropical environments. The presence of fourth level carnivores (i.e., polar bear (*Ursus maritimus*)), and storage of lipid as an energy source, make Arctic food webs vulnerable to bioaccumulative chemicals. Through their utilization of a traditional diet which is high in nutritionally beneficial fat, certain groups of indigenous people in the Arctic receive elevated exposure to some POPs (Hansen *et al.*, 1998).

This assessment also considers many other organic chemicals that are not listed in the Stockholm Convention but that have been detected in the Arctic. Foremost among these are butyl tin compounds, especially tributyltin (TBT), brominated flame retardants, and polyaromatic hydrocarbons (PAHs) from combustion sources. Members of these latter two chemical groups meet some or all of the POPs criteria.

Table 1.1. Criteria for identification of 'new' POPs under the Stockholm Convention (2001).

Chemical identity	Structure, including specification of isomers where applicable, and the structure of the chemical class
Persistence	<ol style="list-style-type: none"> <li>1. The half-life of the chemical in water is greater than two months, or the half-life in soil is greater than six months, or the half-life in sediment is greater than six months.</li> <li>2. Other evidence that the chemical is sufficiently persistent to justify its consideration.</li> </ol>
Bioaccumulation	<ol style="list-style-type: none"> <li>1. Evidence that the bioconcentration factor or bioaccumulation factor in aquatic species is greater than 5000.</li> <li>2. The logarithm of the octanol-water partition coefficient (<math>\log K_{ow}</math>) is greater than 5.</li> <li>3. Evidence that a chemical presents other reasons for concern, such as high bioaccumulation in other species, high toxicity or ecotoxicity.</li> <li>4. Monitoring data in biota indicating that the bioaccumulation potential of the chemical is sufficient to justify its consideration.</li> </ol>
Potential for long-range environmental transport	<ol style="list-style-type: none"> <li>1. Measured levels of the chemical in locations distant from the sources of its release that are of potential concern.</li> <li>2. Monitoring data showing that long-range environmental transport of the chemical, with the potential for transfer to a receiving environment, may have occurred via air, water, or migratory species.</li> <li>3. Environmental fate properties and/or model results that demonstrate that the chemical has a potential for long-range environmental transport through air, water or migratory species.</li> <li>4. The half-life in air is greater than two days.</li> </ol>
Adverse effects	<ol style="list-style-type: none"> <li>1. Evidence of adverse effects to human health or to the environment that justifies consideration.</li> <li>2. Toxicity or ecotoxicity data that indicate the potential for damage to human health or to the environment.</li> </ol>

## 1.1. Physical and chemical characteristics of POPs

The previous AMAP POPs assessment reviewed physical-chemical properties of major chlorinated POPs and TBT (de March *et al.*, 1998). Since then, there have been several new reviews and assessments of POPs, which have included detailed discussions of physical-chemical properties of the chemicals listed in the Stockholm Convention. Notable here are: the *Handbooks on Physical-Chemical Properties and Environmental Fate of Organic Chemicals* by Mackay *et al.* (1991; 1992; 1997); the recent volume of *Handbook of Environmental Chemistry on New Types of Persistent Halogenated Compounds* (Paasivirta, 2000); and, the *Evaluation of Persistence and Long Range Transport of Organic Chemicals in the Environment* resulting from a workshop of the Society of Environmental Toxicology and Chemistry (SETAC) (Klečka *et al.*, 2000). Therefore the emphasis here will be on new physical-chemical characteristics and degradation half-life information that is relevant to understanding the behavior of POPs in the Arctic, and on new or emerging chemicals that may be classified as POPs in the future. Selected chemical and physical properties, and chemical structures of the chemicals considered in this chapter are presented in Annex Table 1.

### 1.1.1. Industrial products and by-products

Chemicals in this category are those that were or are still deliberately produced, or are unintentional by-products from the production of other, mainly halogenated, organics.

#### PCBs

There are 209 chlorinated biphenyl congeners (CBs), with different chlorine substitutions on the biphenyl ring. The number of chlorines, as well as positioning on the rings, influences PCB physical properties and biological activity (Mackay *et al.*, 1991). These are highly lipophilic compounds ( $\log K_{ow} > 5$ ) (Annex Table 1). Most PCB congeners, particularly those lacking adjacent unsubstituted positions on the biphenyl rings (e.g., 2,4,5- or 2,3,5- or 2,3,6-substituted on both rings) are extremely persistent in the environment. They are estimated to have half-lives ranging from three weeks to two years in air and, with the exception of mono- and dichlorobiphenyls, are essentially non-biodegradable in aerobic soils or sediments (Mackay *et al.*, 1991). Highly chlorinated PCBs have been shown to be dechlorinated in anaerobic sediments, but only where present at relatively high concentrations (greater than 10  $\mu\text{g/g dw}$ ) (Brown *et al.*, 1987; Rhee *et al.*, 1993). In vertebrates, some PCBs are metabolized to methylsulfone- and hydroxy-substituted compounds, depending on the chlorine substitution pattern (Letcher *et al.*, 2000a). For example, hydroxy-PCBs are prominent contaminants in polar bear plasma (Sandau, 2000). A recent development in the information available on PCBs is the direct measurement of their reaction with hydroxyl (OH) radicals, the most important atmospheric oxidant (Anderson and Hites, 1996). These results clearly show that OH-radical reactions are the major removal process for PCBs in the atmosphere, with half-lives ranging from two days for biphenyl to 34 days for pentachlorobiphenyl at mid-latitudes. These half-lives could be much longer for PCBs in the Arctic atmosphere because of lower OH radical concentrations at the poles most of the year (Franklin *et al.*, 2000), as discussed in Section 2.1.1.2.

#### Hexachlorobenzene (HCB) and other chlorinated benzenes

HCB and related chlorobenzenes are cyclic aromatic compounds with chlorine substituting for hydrogen atoms on the benzene ring. HCB has a relatively high bioaccumulation potential because of high lipophilicity ( $\log K_{ow} = 5.5$ ) and long half-life in biota (Niimi, 1987). It has an estimated field half-life of 2.7-5.7 years (Howard, 1991). Other di- to penta-substituted chlorinated benzenes (CBz) are also relatively lipophilic, semivolatile, and may be persistent especially in the abiotic environment (Mackay *et al.*, 1991). For example, pentachlorobenzene (PeCBz) has a  $\log K_{ow} = 5.0$  and an estimated half-life in surface waters of greater than 200 days (Howard, 1991; Mackay *et al.*, 1991).

#### Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs)

The polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) are two structurally similar families of compounds that include 75 and 135 congeners, respectively. Most PCDD/F congeners, like PCBs, are extremely hydrophobic and resistant to biodegradation in soils and sediments. PCDD/Fs have vapor pressures from 1.0 to  $10^{-6}$  Pa at 25°C (Mackay *et al.*, 1992) and are distributed in the atmosphere between gas and particle phases. PCDD/Fs encounter similar types of processes in each of these phases: wet and dry deposition; photolysis; and, chemical reactions. It is likely that the lower chlorinated PCDD/Fs are more susceptible to photochemical processes in the gas phase (Brubaker and Hites, 1997). The photodegradation of particle-bound PCDD/Fs in air was found to be negligible (Koester and Hites, 1992). Historical profiles of PCDD/Fs in sediment cores from large lakes show no evidence of transformation of congeners (such as anaerobic dechlorination) over time (Baker and Hites, 2000). Tetra- to octachlorodibenzodioxins/furans have lower vapor pressures and Henry's Law Constants (HLCs) than PCBs and are therefore not expected to undergo long-range transport to the same extent (Mackay *et al.*, 1992). Nevertheless, there is evidence for deposition in Arctic soils and sediments (Oehme *et al.*, 1993; Brzuzy and Hites, 1996; Cleverly *et al.*, 1996; Wagrowski and Hites, 2000). The 2,3,7,8-substituted chlorinated dibenzo-*p*-dioxin/dibenzofuran (CDD/F) congeners are known to bioaccumulate in fish and invertebrates; however, non-2,3,7,8-substituted congeners (which predominate in combustion sources) are readily degraded by vertebrates (Opperhuizen and Sijm, 1990).

#### Polychlorinated naphthalenes (PCNs)

Polychlorinated naphthalenes are planar molecules with structural similarity to PCBs and PCDD/Fs (Falandysz, 1998; Jakobsson and Asplund, 2000). There are 75 PCN congeners. Tr- to OcCNs are very hydrophobic and have high  $\log K_{ows}$  (5.1-6.4). PCNs may photolytically degrade in the atmosphere because they have weak ultraviolet (UV) absorption maxima between 275 and 345 nm and they are also expected to react with hydroxy radicals (Franklin *et al.*, 2000). However, their presence in Arctic air during the summer (Harner *et al.*, 1998) implies a relatively long atmospheric half-life. Little is known about their rates of degradation in other media.

### PAHs

The polycyclic aromatic hydrocarbons constitute a large class of organic compounds with multiple aromatic rings. In the context of this assessment, 2- to 5-ringed unsubstituted PAHs, such as acenaphthene, fluorene, phenanthrene, fluoranthrene, pyrene, benzopyrenes, benzofluoranthrenes, and perylene, formed during combustion and transported long distances in the atmosphere, are the main chemicals of concern because they predominate in Arctic air (Halsall *et al.*, 1997) and snow (Peters *et al.*, 1995; Masclat *et al.*, 2000). These compounds are hydrophobic and have a wide range of vapor pressures. PAHs are degraded rapidly by vertebrates and thus do not bioaccumulate, although they have high log  $K_{ow}$ s. Microbial degradation of PAHs is also significant, especially for 2- and 3-ring compounds, and proceeds via hydroxylation. Susceptibility of PAHs to biodegradation is inversely correlated with the number of aromatic rings in the PAH.

### Octachlorostyrene (OCS)

OCS is a fully chlorinated derivative of styrene, an alkyl-substituted benzene. It is regarded as persistent, based on observations of long half-lives in sediment, and highly bioaccumulative, based on its perchlorinated structure and high log  $K_{ow}$  value (7.46) (CGLI, 1999; USEPA, 1999a).

## 1.1.2. Chlorinated pesticides

### 1.1.2.1. Persistent pesticides

*DDT, chlordane, heptachlor, dieldrin, aldrin, endrin, mirex*  
These seven structurally diverse organochlorine (OC) pesticides are all identified as POPs under the Stockholm Convention. Mackay *et al.* (1997) have summarized the physical property data for these compounds and the previous AMAP POPs assessment also included this information; therefore, they are not reviewed further here. Of these POPs, DDT and chlordane are contaminants of particular concern in the Arctic due to the presence of high levels of their toxic metabolites (*p,p'*-dichlorodiphenyldichloroethylene (DDE) and oxychlordane, respectively) in top predators.

### Toxaphene

Toxaphene is the common name for a mixture of chlorobornanes and chlorinated camphenes containing six to ten chlorines (de Geus *et al.*, 1999; Vetter and Oehme, 2000). In eastern Europe, and possibly in Russia, it was used under the name of 'Melipax' prior to 1990. The octa- and nonachlorobornanes are extremely persistent in aerobic soils but slowly dechlorinate in anaerobic sediments to hexa- and heptachlorobornanes (Fingerling *et al.*, 1996; Stern *et al.*, 1996). Unlike the other seven OC pesticides, there has been a great deal of recent work on the physical properties of toxaphene. Log  $K_{ow}$ s of toxaphene congeners were measured by Fisk *et al.* (1999a), and found to range from 4.8 to 6.6. They were significantly related to chlorine number. The HLC of technical toxaphene was found to be temperature-dependent by Jantunen and Bidleman (2000).

### 1.1.2.2. Less persistent chlorinated pesticides

#### Hexachlorocyclohexanes (HCH)

Hexachlorocyclohexanes are a group of related isomers existing in eight isomeric forms with the  $\alpha$ -,  $\beta$ - and  $\gamma$ -iso-

mers predominating. The latter is the insecticidally active isomer. The isomers differ in their positioning of the chlorines at axial and equatorial positions. The  $\beta$ -isomer has all six chlorines in the equatorial position. The key physical properties that control the ultimate fate of HCH isomers in the environment are high volatility, low HLC, a strong dependency of HLC on temperature ( $\alpha$ -HCH partitions into water about seven times more strongly at 0°C than at 25°C (Kucklick *et al.*, 1991)), low affinity for particles, and resistance to degradation in cold water (Ngabe *et al.*, 1993). The physical properties of HCH isomers are perhaps the most thoroughly studied of any POP. Values of HLC for HCH isomers at 0°C range from 0.06 Pa m<sup>3</sup>/mol for  $\alpha$ -HCH, to 0.03-0.06 Pa m<sup>3</sup>/mol for  $\gamma$ -HCH and 0.003 for  $\beta$ -HCH. The much lower value for  $\beta$ -HCH is very significant. It leads to major differences in the spatial distribution of  $\alpha$ -HCH and  $\beta$ -HCH in oceanic surface waters (Li *et al.*, 2002). Log  $K_{ow}$ s of HCH isomers have been reported in the range of 3.8-4.3. Biological half-lives of the HCH isomers differ significantly.  $\beta$ -HCH has a 25-fold longer half-life in humans than  $\alpha$ -HCH (Willett *et al.*, 1998).

### Endosulfan

Endosulfan is a chlorinated cyclodiene insecticide containing a sulfite (SO<sub>3</sub>) group. It is widely used against a variety of insects especially on high-value crops (NCFAP, 2001). It consists of  $\alpha$ - and  $\beta$ -isomers at a 2:1 ratio in the technical product. The physical properties of endosulfan are characteristic of a compound capable of long-range transport (HLC = 2.98 Pa m<sup>3</sup>/mol; vapor pressure = 5 × 10<sup>-3</sup> Pa) (Suntio *et al.*, 1988).  $\alpha$ -Endosulfan has a higher HLC than  $\alpha$ -HCH but lower vapor pressure. The log  $K_{ow}$  of  $\alpha$ -endosulfan (3.6) is low compared to many chlorinated insecticides but similar to lindane. Endosulfan sulfate is a major, persistent degradation product of endosulfan. Hydrolysis is an important route of degradation for endosulfan. Hydrolysis half-lives for  $\alpha$ - and  $\beta$ -isomers are 11 and 19 days, at pH 7, respectively (USEPA, 2001).

## 1.1.3. Other pesticides

### 1.1.3.1. Organotins including tributyltin (TBT)

TBT is a broad-spectrum algicide, miticide, fungicide, and insecticide (Hoch, 2001). Mono- and dibutyltin (MBT and DBT) are used primarily as heat and light stabilizers in the production of polyvinylchloride (PVC) plastics. All organotins are stable in water and can tolerate high (200°C) temperatures (Zuckerman *et al.*, 1978) and thus, are not subject to thermal decomposition in the environment. Solubility in water decreases with increasing number and length of organic substituents. TBT has water solubilities of 50 mg/L (seawater) and 5-17 mg/L (distilled water), DBT has a water solubility of 4-50 mg/L (seawater) and the solubility of MBT is not known (cited in Hoch, 2001). The log  $K_{ow}$  for TBT-chloride is pH-dependent and ranges from 3.2 (pH 5.8) to 3.85 (pH 7.8) (Tsuda *et al.*, 1990).

Once released to water, TBT is degraded by sequential debutylation to DBT, MBT, and eventually to relatively non-toxic inorganic tin compounds primarily by organisms. Sunlight irradiation may also contribute to TBT degradation. The degradation time in water is short, with half-lives reported from days to a few weeks (Stewart and



de Mora, 1990; Dowson *et al.*, 1993). TBT may be concentrated in the sea surface microlayer (Dobson and Carbrindenc, 1990) and is strongly particle-reactive with partition coefficients reported to be as high as  $10^3$ - $10^4$  (Langston and Pope, 1995). The breakdown of TBT in anaerobic sediments (half-life of two or more years) is much slower than in water (Clark *et al.*, 1988). Therefore, contaminated sediments are potentially an important environmental reservoir for TBT which can continue to provide a source long after the industrial use of TBT has been curtailed.

#### 1.1.4. 'New' chemicals with POPs characteristics

##### *Short-chain chlorinated paraffins (SCCPs)/*

##### *C<sub>10</sub>-C<sub>13</sub> polychlorinated alkanes*

SCCPs are polychlorinated-[C<sub>10</sub>-C<sub>13</sub>]-n-alkanes with chlorine content from 50 to 70% by mass (Tomy *et al.*, 1998). Drouillard *et al.* (1998a; 1998b) reported water solubilities of individual chlorinated alkane congeners ranging from 37 to 994 µg/L, indicating that SCCPs are hydrophobic substances (Annex Table 1). Log K<sub>ow</sub>s for SCCPs, determined by slow-stirring, are in the 5.9 to 6.2 range, classifying them as lipophilic, similar to DDT but less than PCBs of comparable molecular weight. HLCs for C<sub>10</sub>-C<sub>12</sub> SCCPs range from 0.7 to 18 Pa m<sup>3</sup>/mol, which is similar to HLCs for chlorinated pesticides (HCH, toxaphene, *p,p'*-DDT) and implies partitioning from water to air, or from soils to air, depending on environmental conditions and prevailing concentrations in each compartment.

##### *Medium-chain chlorinated paraffins (MCCPs)*

MCCPs are polychlorinated-[C<sub>14</sub>-C<sub>17</sub>]-n-alkanes with chlorine content of 40 to 50% by mass (Tomy *et al.*, 1998). Campbell and McConnell (1980) reported that the solubility of an MCCP (C<sub>16</sub>; 52% Cl) was 10 µg/L in freshwater and 4 µg/L in seawater. Based on this information, it could be assumed that the water solubilities of MCCPs are lower than SCCPs and that solubility in seawater might be somewhat reduced compared to the water solubility in freshwater. Drouillard *et al.* (1998a) found that vapour pressures of C<sub>10</sub>-C<sub>12</sub> SCCPs tended to decrease exponentially with the addition of carbon or chlorine to the molecule. On this basis OSPAR (2000) concluded that the vapor pressures of MCCPs were similar or lower than SCCPs (Annex Table 1).

##### *Tetra- and pentabrominated diphenyl ethers (TeBDE, PeBDE)*

The tetra- and pentaBDEs (TeBDE, PeBDE) have low vapor pressures and are very hydrophobic with log K<sub>ow</sub>s ranging from 5.9-6.2 for TeBDEs, and 6.5-7.0 for PeBDEs (Watanabe and Tatsukawa, 1990). Experimentally determined subcooled vapor pressures for several BDE congeners were found to be lower than for comparably chlorinated PCBs, and decreased with increasing number of bromines (Tittlemeir and Tomy, 2001). Halogen substitution pattern influences vapor pressure such that congeners with bromine substitution in the *ortho* positions to the ether bond have higher vapor pressures (Wong *et al.*, 2001).

##### *Decabrominated diphenyl ether (DeBDE)*

Decabrominated diphenyl ether (DeBDE) is a fully bromine-substituted diphenyl ether. DeBDE and its nona-

and octabrominated BDE impurities are all extremely hydrophobic with log K<sub>ow</sub>s estimated to be 8.4-8.9 for OcBDEs and 10 for DeBDE (Watanabe and Tatsukawa, 1990). These properties imply low water solubility, high binding affinity to particles and a tendency to accumulate in sediments. Several studies have shown that DeBDE is successively and rapidly debrominated by UV light and sunlight to lower brominated PBDEs (down to TrBDEs) (Watanabe and Tatsukawa, 1987; Tysklind *et al.*, 2001; Söderström *et al.*, 2004). If this process is significant in the environment, it could lead to the formation of lower brominated BDEs, which are known to bioaccumulate.

##### *Other brominated flame retardants*

In general, the physical-chemical properties of other brominated flame retardants are not well studied. Log K<sub>ow</sub>s have been estimated based on fragment constant methodology. Tetrabromobisphenol A (TBBPA) has a log K<sub>ow</sub> of 4.5 (WHO/ICPS, 1995). Hexabromocyclododecane (HBCD) has a log K<sub>ow</sub> of 5.8, has low water solubility (IUCLID, 1996), and probably also has an affinity for particles and sediments.

##### *Perfluoroalkylsulfonates and perfluoroalkanoic acids (PFAs)*

'Perfluorinated' is a term used to describe molecules in which all hydrogens have been replaced by fluorine. The most widely known PFAs are perfluorooctane sulfonate (PFOS; C<sub>8</sub>F<sub>17</sub>SO<sub>3</sub>) and perfluorooctanoic acid (PFOA; C<sub>8</sub>F<sub>15</sub>O<sub>2</sub>). However, similar compounds having longer or shorter perfluorinated chains are also produced or exist as impurities within manufactured formulations.

The existing database describing physical properties of perfluorinated acids, including PFOS and PFOA, is severely limited because of their anomalous physical and chemical behavior. Long perfluorinated carbon chains are hydrophobic and oleophobic (i.e., repel both water and oil), and consequently PFAs are markedly surface active because of the hydrophilic anionic head group (i.e., CO<sub>2</sub><sup>-</sup>, perfluorocarboxylates; or SO<sub>3</sub><sup>-</sup>, perfluorosulfonates). The other unique property of PFAs is their thermal and chemical stability, allowing them to be used in high temperature applications or in strongly acidic or basic solutions where hydrocarbon surfactants would otherwise be degraded. This stability results from the numerous covalent carbon-fluorine bonds that act as a protective sheath around the weaker carbon-carbon bonds. PFAs have no known route of degradation or metabolism under normal environmental conditions. Biological means of degradation or transformation have not been thoroughly explored; however, one published report indicated the failure of bacteria (*Pseudomonas* sp.) to degrade PFOS under aerobic, sulfur limiting conditions (Key *et al.*, 1998). PFAs are expected to redefine the term 'persistence' as it relates to organic pollutants (Renner, 2001).

The log K<sub>ow</sub> for PFOS is reported as 10. However, this measurement is prone to error. K<sub>ow</sub> may also be an inherently poor descriptor of bioaccumulation potential because PFOS and PFOA tend to accumulate in blood and liver, not in lipid. PFOS is sparingly volatile (3.3 × 10<sup>-4</sup> Pa) (USEPA, 1999b) and moderately water soluble (1080 mg/L) (USEPA, 2000a). The HLC for PFOS has been estimated at < 2 × 10<sup>-6</sup> Pa m<sup>3</sup>/mol. Taken together, these data suggest PFOS will accumulate in the aquatic environment (USEPA, 2000a,b).

## Chapter 2

## Sources and Pathways of Persistent Organic Pollutants

The previous AMAP assessment summarized the pathways of pollutants in general (Gregor *et al.*, 1998), and POPs (de March *et al.*, 1998) specifically, as they were understood in the mid-1990s. Recent reviews have updated this information (Macdonald *et al.*, 2000; 2003). The general principles are briefly described in the following sections and the reader is referred to the above reviews for more details. In addition we also consider a novel pathway, biotic transport.

## 2.1. Pathways

For pristine regions like the Arctic, generally four possible pathways exist for the transport of POPs (Figure 2-1). The atmosphere, ocean currents, transpolar ice pack, and large Arctic rivers (Ob, Pechora, Yenisey, Lena, Mackenzie) are the main transport routes by which persistent organics enter Arctic ecosystems. The relative importance of each pathway depends on the chemical and physical properties of the substance and its emissions in the source region, which may vary over time. In addition to the above mentioned transport routes, POPs may also be transported into the Arctic via pelagic organisms

(crustaceans, fish, marine mammals) and migratory birds, animals that migrate in large groups throughout different climate zones into the Arctic. These organisms can transfer the pollutants into higher-level organisms via the Arctic food web.

In general, present levels of most POPs cannot be related to known potential sources within the Arctic, and can therefore only be explained by long-range transport from lower latitudes (Hansen *et al.*, 1996; de March *et al.*, 1998; Macdonald *et al.*, 2000). However, there are sources of POPs within the Arctic that have local and possibly regional significance but are minor contributors from a circumpolar perspective. These include PCB emissions from military bases, harbors, and landfills, as well as PCDD/Fs and PAHs from smelters located in high latitudes (Section 2.3.2). Owing to increasing activities related to exploration and production of oil and gas, especially in the Russian and Norwegian Arctic, petroleum hydrocarbon releases are a potential local source of pollution for PAHs in ocean waters and sediments (Robertson, 1998; Hansen *et al.*, 1996). A future AMAP assessment of petroleum hydrocarbons will consider this particular source in more detail.

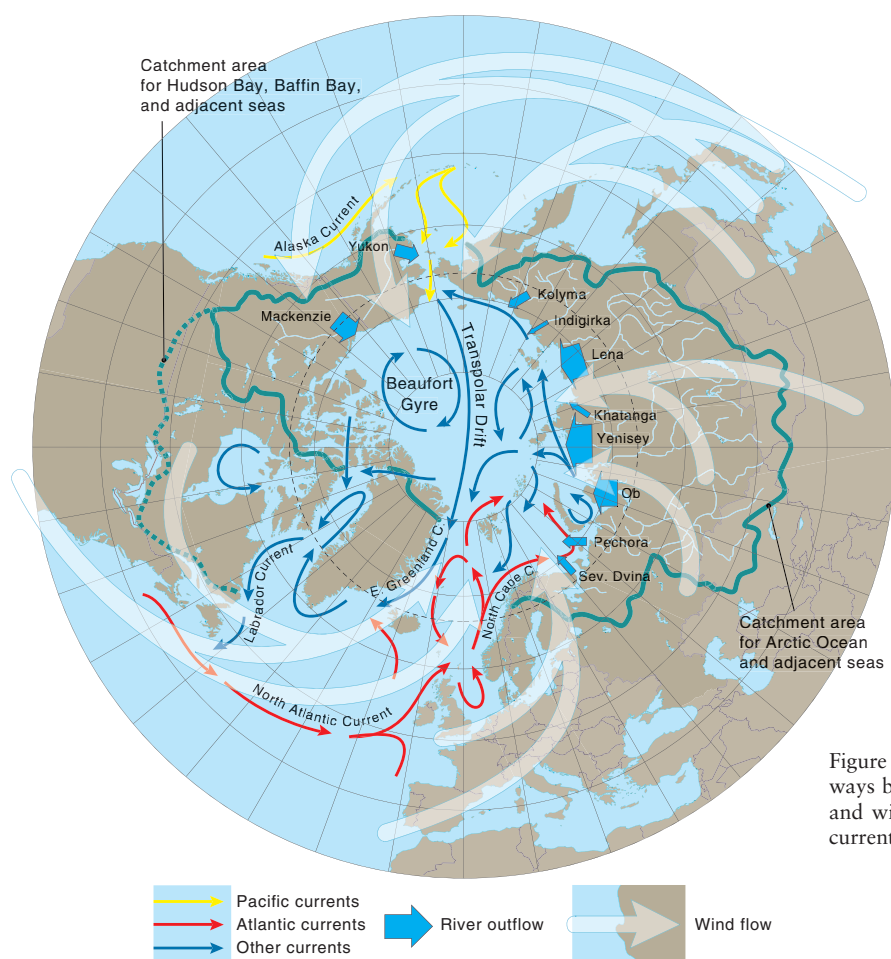


Figure 2-1. Illustration of the different physical pathways by which POPs enter the Arctic. Transport into, and within, the Arctic occurs via air currents, ocean currents, rivers, and transpolar ice movements.

### 2.1.1. Atmospheric transport and deposition of POPs

#### 2.1.1.1. Meteorological conditions

Under favorable meteorological conditions, rapid air transport can take place in a few days or weeks, from the source regions into the Arctic. Almost twenty years ago, it was postulated that the global dispersion of semivolatile POPs occurred mainly via the atmosphere and was dependent on their vapour pressures (Ottar, 1981). Today, this is well documented and models developed show that atmospheric transport is probably the most important long-range transfer route for these compounds (Wania and Mackay, 1993; Strand and Hov, 1996; Wania and Mackay, 1996; Wania *et al.*, 1999a).

In winter, the lower tropospheric circulation is dominated by high pressures over the continents and low pressures over the northern Pacific (Aleutian Low) and Atlantic Oceans (Icelandic Low). The Siberian High tends to force air on its western side into the Arctic acting as an effective atmospheric conduit from industrialized regions of Siberia and eastern Europe into the high Arctic (Figure 2-1). The high-pressure ridge over North America then forces air southward giving a net transport out of Eurasia into the Arctic, across the Arctic and south over North America. The Icelandic Low produces westerly winds over the eastern North Atlantic and southerly winds over the Norwegian Sea providing a conduit for airborne contaminants from eastern North America and Europe to reach the Arctic rapidly. Finally, the Aleutian Low tends to steer air that has crossed the Pacific from Asia up into Alaska, the Yukon, and the Bering Sea (Bailey *et al.*, 2000; Li *et al.*, 2002; Wilkening *et al.*, 2000). During winter, these three routes into the Arctic – southerlies in the Norwegian Sea (40%), Eastern Europe/Siberia (15%), and Bering Sea (25%) account for about 80% of the annual south to north air transport (Iversen, 1996). Thus during winter, the polar front expands into an area encompassing southern regions of the northern hemisphere into central Europe, central U.S., Asia, the European part of Russia, Belarus, and the Ukraine. Eighty percent of the main source areas for persistent pollutants are situated in these regions. Therefore, during the winter season, the polar front is not limiting the atmospheric transport from these sources into the Arctic region.

Summer pressure fields and air-flow patterns are markedly different from those of winter. In summer, the continental high-pressure cells disappear and the oceanic low-pressure cells weaken with the result that northward transport from low latitudes weakens. During summer, the polar front is situated far north and creates a meteorological barrier often difficult to penetrate for air masses transported from southern regions. According to Iversen (1996), summer accounts for only 20% of the annual south to north air transport (southerlies in the Norwegian Sea (10%), eastern Europe/Siberia (5%), and Bering Sea (5%)). Prevailing winds provide a means to transport contaminants from industrialized North America and Europe to the North Atlantic but penetration into the Arctic weakens.

#### 2.1.1.2. Light conditions

The polar night at 80° northern latitude lasts from October until March and then changes into the midnight sun period. These special seasonal light conditions have tremendous effects on the photochemical reactivity and degradation of some persistent pollutants. Lack of photochemical degradation processes may be the main reason why long-range transport of highly- and semivolatile organic compounds such as 2- and 3-ring PAHs is mainly observed during the winter. Photochemical processes during the 24-hour summer light period may quickly degrade chemicals that react rapidly with OH radicals or degrade by direct UV photolysis. In winter, on the other hand, air concentrations of these types of compounds are usually elevated by one or two orders of magnitude. Indications of a seasonally dependent photochemical degradation were also found for the persistent pesticide *trans*-chlordane. Seasonal shifts in the chlordane patterns have been found during year-round ambient air sampling campaigns at Ny-Ålesund (Svalbard, Norway) and at Alert (Ellesmere Island, Canada) (Oehme, 1991; Halsall *et al.*, 1998).

#### 2.1.1.3. Precipitation inputs of POPs

Precipitation is a key factor in contaminant transport. Rain and snow scavenge aerosols and gasses from the atmosphere and deposit them at the Earth's surface (Gregor *et al.*, 1998; Macdonald *et al.*, 2000). Scavenging by precipitation may be relatively weak in the desert-like conditions of the High Arctic. For example, mean precipitation for the Arctic Ocean is estimated at about 25.2 cm/yr and evaporation is about 13.6 cm/yr for a net moisture flux to the ground of 11.9 cm/yr (Barry and Serreze, 2000). Precipitation over land in the Arctic drainage basins is greater, with the runoff yield (precipitation minus evaporation) being estimated at 21.2 cm/yr from the network of gauged discharge by rivers (Lammers *et al.*, 2001).

Due to the low annual average temperature, snow is the dominant form of precipitation in the Arctic. The influence of snow and ice on the fate and air-surface exchange of organic contaminants has been reviewed by Franz *et al.* (1997) and Wania *et al.* (1998). Wania *et al.* (1999b) demonstrated that falling snow serves as an extremely efficient scavenger of both vapour and particulate phase compounds, with vapour scavenging of lower chlorinated PCBs and 2- and 3-ring PAHs being a predominant process. Also the water-air partition coefficient and the ice-air sorption coefficient increase at low temperatures thus increasing the potential for vapour scavenging (Wania *et al.*, 1999b). In addition, a snow crystal possesses a large surface area and adsorbs particles and contaminants to its crystalline surface more efficiently than a water droplet (Franz and Eisenreich, 1998). Therefore snow precipitation transports contaminants more efficiently to the ground (soil, water surface) than rain droplets. In contrast to rainwater, the surface properties of snow crystals change during aging after deposition. The surface of a snow crystal diminishes dramatically after a short time on the ground because of the overall pressure of the surrounding snow, and the weather and climate conditions during the aging process. Thus, due



to surface reduction, the adsorbed contaminants are either released into the ground, stay adsorbed to the snow crystal, or are re-evaporated into the atmosphere. These three possibilities are dependent on the vapour pressure of the respective contaminant and the ambient climatic conditions. Therefore, high POP levels can often be found in upper layers containing new snow surface whereas in aged snow, situated near the ground, the contaminants are already released into the soil.

#### 2.1.1.4. Sea–air gas exchange

Gas absorption and volatilization of POPs are important pathways of deposition and removal in the Arctic Ocean. Evaluation of relative fugacities of PCBs and toxaphene in air and seawater, based on measurements in the mid-1990s, indicated that these compounds are still loading into the Arctic Ocean via the atmosphere, while  $\alpha$ -HCH is volatilizing (Macdonald *et al.*, 2000). Verification of the sea–air exchange process for  $\alpha$ -HCH was accomplished using the distribution of the enantiomers of this chiral pollutant as indicators (Falconer *et al.*, 1995; Jantunen and Bidleman, 1996; Jantunen and Bidleman, 1997). Enantiomeric ratios (ER) found in surface water and in high-volume ambient air samples were similar, implying a seawater source. A significant deviation from the racemic distribution (ER = 1) in environmental samples was also found. This deviation can only occur when biochemical transformation (microbiological or in higher organisms) takes place. Therefore, the authors concluded that  $\alpha$ -HCH found in the ambient air had been subject to earlier microbiological degradation in the water column before being re-evaporated into the atmosphere. This finding represents the first evidence for water–air gas exchange of  $\alpha$ -HCH using an experimental approach.

#### 2.1.2. Ocean transport

POPs are also transported by ocean currents. Ocean circulation is driven by a combination of various forces and a particular force (e.g., tidal forces, wind stress, mixing of water masses) may dominate in a particular area. Within the Arctic Ocean, the main surface circulation features are the clockwise circulation of the Beaufort Gyre and the Transpolar Drift, which flows from Siberia (Russia), across the pole, and then southward, exiting as the East Greenland Current (Gregor *et al.*, 1998; Macdonald *et al.*, 2003). The major current systems whereby water is exchanged between the Arctic Ocean and other oceans, are found in Fram Strait. For example, the West Spitsbergen Current flows northward off the west coast of Spitsbergen (Svalbard, Norway), transporting Atlantic water from the Norwegian Sea into the Arctic Ocean (Gregor *et al.*, 1998). The transport via sea currents, however, may take years. Studies of the transport of radioactive isotopes released from the nuclear power plant in Sellafield (U.K.) indicates a four to five year transport over the 2200 km from southern Norway to northern Spitsbergen (Svalbard). Nevertheless, ocean transport is important for POPs with low HLCs such as  $\beta$ -HCH, which is selectively removed from the air by precipitation-scavenging as it heads northward and is enriched in seawater relative to other

HCH isomers (Li *et al.*, 2002). However, some degradation does occur in seawater. For example,  $\alpha$ - and  $\gamma$ -HCH have recently been shown to undergo biotransformation in Arctic seawater with half-lives estimated to be 19 years for  $\gamma$ -HCH and 5.9 years for the + enantiomer of  $\alpha$ -HCH (Harner *et al.*, 1999).

Prolonged half-lives in abiotic compartments, combined with efficient biomagnification and bioaccumulation in the Arctic food web due to lipophilicity and resistance to biological degradation, give rise to high concentrations of the long-range transported POPs in the higher trophic levels of the Arctic ecosystem, including indigenous people.

The low annual average temperature of the Arctic is the main reason why microbiological degradation of organic material slows to a minimum, especially in terrestrial and freshwater environments. This potentially extends the half-lives of many compounds in the Arctic relative to temperate regions.

#### 2.1.3. Riverine inputs and sea-ice transport

Riverine input is considered to be another important source of contaminants to the Arctic Ocean (Barrie *et al.*, 1992; Pavlov and Pfirman, 1994; de March *et al.*, 1998; Gregor *et al.*, 1998) and can be considered as an important circumpolar source draining environmental pollutants into the Arctic Basin. Particles transported to the coast by large Russian Arctic rivers (e.g., Yenisey, Ob, Lena, and Pechora) during the melting period are contaminated with pollutants originating in the industrial areas of the northern Urals and western Siberia. As a result of various physical processes, the particles are incorporated in the coastal ice which may then carry large loads of contaminated sediments.

The general movement of sea ice from the coast of the Kara Sea is northward to join the Siberian branch of the Transpolar Drift between the Franz Josef Land and Severnaya Zemlya archipelagos. Ice-bound particles with possible contaminants may thus be carried out of the area and released in the main ice-melting areas east of Svalbard and in the Fram Strait (Figure 2·1). It is estimated that ice-rafted material forms up to 80% of the sediment on the ridges and upper slopes in the central Arctic Ocean (Darby *et al.*, 1989).

#### 2.1.4. Biotic transport

Transport of pollutants, particularly POPs in migratory animals may also be a significant pathway in the Arctic, where many important species such as seabirds, cetaceans (beluga, bowhead whales, minke whales), pinnipeds (harp seals), salmon, and Atlantic cod are migratory. Some animals cover long distances, often crossing international boundaries and linking industrialized/agricultural and remote regions during their migration. Some species can be important prey of resident Arctic animals, for example, Barents Sea harp seals consumed by polar bear in eastern Svalbard and Franz Josef Land. A unique example is salmon entering Alaskan rivers to spawn and die. Ewald *et al.* (1998) described the entry of PCBs and DDT into the grayling population of the Copper River in Alaska by migrating sockeye salmon. The grayling in a salmon spawning lake had contami-



nant concentrations more than double those found in grayling from a nearby, but salmon-free lake.

Wania (1998) estimated that the amount of POPs transferred in and out of the Arctic via seabirds migrating annually from the Canadian Arctic to northwest Atlantic waters was in the range of grams to kilograms per year based on the fact that the populations involved are in the hundred thousands to millions of birds. Seabirds also leave behind guano which may contain significant quantities of persistent OC contaminants. Evensen *et al.*, (2002) have shown that elevated PCBs and other OCs in fish and sediments from Lake Ellasjøen on Bjørnøya (74°30'N, 19°E) are due to seabird guano (see Section 4.3.5). The seabirds at Bjørnøya are functioning as a transport link for POPs from the marine ecosystem to the freshwater ecosystem.

While many Arctic whales migrate regionally within the Arctic, the Eastern Pacific stock of gray whales (*Eschrichtius robustus*) spends the summer in the Bering and Chukchi Seas, and the winter off the Pacific coast of Mexico and California (Baker, 1978). Relatively reliable population estimates and recent contaminant concentrations in stranded individuals exist for this stock, allowing for an estimate of the amount of some POPs contained in these whales. Wania (1998) estimated that the total amount of PCBs and DDT contained in the gray whale population and therefore transported annually with gray whales is thus in the range of 20 to 150 kg PCBs, and 1 to 40 kg DDT. Of course only animals that die in the Arctic would leave the contaminants there. However, carcasses of dead or hunted whales are scavenged by Arctic fox and polar bears, thereby transferring contaminants from the temperate to the Arctic environment.

Considering all migratory whale populations, Wania (1998) concluded that the amount of PCBs and DDTs moved around in these whales is likely of the order of tens of tons per year. Especially for DDTs, these gross fluxes with whales may be comparable to those in air and ocean currents. On the other hand, for the more water soluble, more volatile, and less bioaccumulative HCHs the amounts in organisms are relatively low by comparison to the mass in the water column. Gross

fluxes in migrating birds are much lower for PCBs, DDT or HCH than the transport rates in the physical media. The importance of biological transport media obviously increases with decreasing volatility and water solubility, and with increasing bioaccumulation potential of a chemical.

Ewald *et al.* (1998) pointed out that the pollutants in biological transport media are more readily available for bioaccumulation than those in abiotic media. In the case of the investigated Alaska freshwater system, they point out that the 'migrating salmon, the salmon roe, and the carcasses are fed upon directly by predators such as bald eagles, bears, and grayling, allowing the pollutants to be transferred to biota in a direct and efficient way'. Similarly, the studies on migrating salmonids in the Great Lakes (Scudato and McDowell, 1989) concluded that resident fish in salmon-accessible tributaries derived their excess pollutant load by directly ingesting the contaminated salmon eggs. Ewald *et al.* (1998) even suggested that biotransport may provide a transport mode for POPs, whose physical chemical properties prevent them from long-range transport in atmospheric and water currents (extremely low volatility or water solubility, high atmospheric degradability, but resistance to metabolic degradation).

## 2.2. Modeling transport and distribution of POPs in the Arctic

### 2.2.1. Global fractionation

The most widely accepted hypothesis about atmospheric long-range transport and distribution of POPs is 'global fractionation' (Wania and Mackay, 1995; 1996) (Figure 2.2). The 'global fractionation hypothesis' explains atmospheric transport as a complex phenomenon depending on the physical-chemical properties of the transported contaminant (e.g., solubility, vapour pressure, molecule size). The physical-chemical properties of the substance are responsible for the atmospheric transport distance and the subsequent deposition via rain, fog or snow in the water column, sediment or soil. Relatively volatile hydrocarbons, characterized by a high vapour pressure, are taken directly into the gaseous phase and

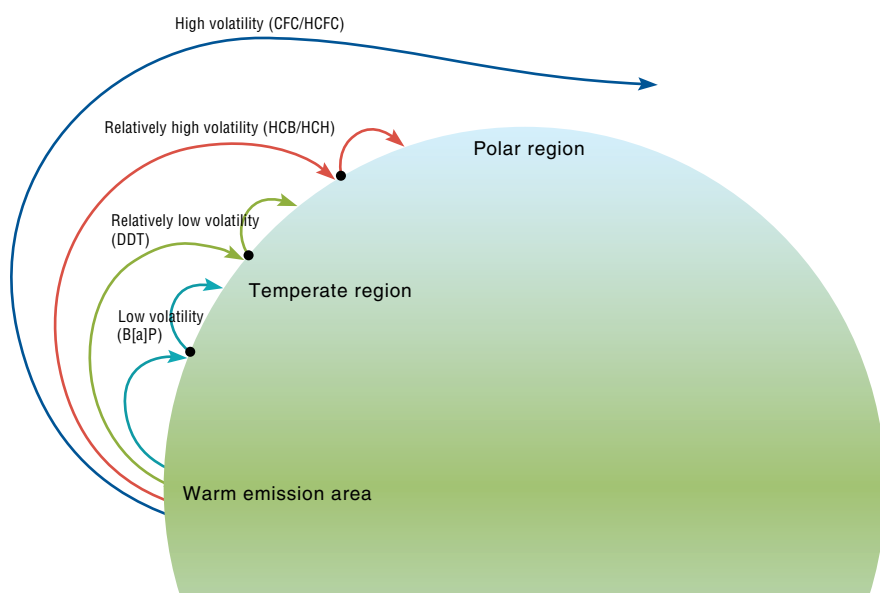


Figure 2.2. Schematic representation of the global fractionation hypothesis and the revolatilization (●) or 'grasshopper' effect. From Wania and Mackay (1996).

immediately transported into the deposition region as shown for lower chlorinated PCBs (CBs 8-99) and CBz. Semivolatile compounds such as lindane ( $\gamma$ -HCH) and chlordanes as well as some highly chlorinated PCBs (CBs 101-153), are distributed between airborne particles and the gaseous phase depending on temperature. These can be washed out via precipitation and temporarily deposited in seawater or soil and can absorb to water, plant and soil surfaces from the gaseous phase. During favorable warm weather conditions, these compounds evaporate again into the atmosphere and undergo further atmospheric transport. This remobilization is also called the 'grasshopper effect'. The role of stormy weather situations in remobilization of semivolatile compounds into the atmosphere is obvious but still scarcely investigated.

As a consequence of these processes, the levels of the more volatile POPs, such as HCHs and HCB, are found at comparable or even higher levels in Arctic biota compared to areas closer to source regions. However, the 'global fractionation hypothesis' and the 'grasshopper effect' are focused mainly on neutral, persistent organics transported via the atmosphere. At the present time, therefore, they are less useful for predicting levels of polar and water soluble persistent compounds that may be associated mainly with seawater.

### 2.2.2. Global transport modeling

A summary of the various modeling approaches for atmospheric and oceanic transport of contaminants in the Arctic was presented in the previous AMAP assessment report (de March *et al.*, 1998). Macdonald *et al.* (2000) also described box models of POP distribution and global transport of POPs using numerical models. The current state of knowledge on modeling the global fate and transport of POPs has recently been reviewed by Scheringer and Wania (2002).

#### 2.2.2.1. Global scale box models

The basic approach for global box modeling is to use models that divide the global environment into a series of zonal bands, each of which is described by a series of well-mixed environmental compartments. This type of global model has seen further development and use during the past five years. In particular, the global distribution model by Wania and Mackay (1993; 1995), has seen further improvement, evaluation, and use, and a version of that model, named Globo-POP, is now available (Wania and Mackay, 2000). A new model of this type was also introduced by Scheringer *et al.* (2000). A detailed example of global modeling of POPs can be found in the recent study by Wania *et al.* (1999a), and Wania and Mackay (1999), on the global chemical fate of  $\alpha$ -HCH. While  $\alpha$ -HCH is no longer a significant POP, the detailed knowledge of its physical-chemical properties and use patterns enabled validation of the model predictions of prevailing and temporal trends of air and water concentrations over time, including contamination of the Arctic.

These global scale box models can be used in two different ways. They can be used to describe the fate of POPs using historical emission estimates, or they can be

used in an evaluative fashion to understand global transport and accumulation processes without the availability of realistic emission scenarios. The first of these uses is primarily limited by the availability of suitable emission estimates, which need to be global in spatial scale and span decades in time. Such estimates only exist for  $\alpha$ -HCH and PCBs at the present time (Section 2.3), and the Globo-POP model has been employed for simulation of the environmental fate of these two POPs (Section 4.2.1.4). The model has also been used to identify chemical property combinations that make a chemical susceptible to accumulation in remote polar regions (Wania *et al.*, 2001).

#### 2.2.2.2. Three-dimensional global atmospheric transport models

Koziol and Pudykiewicz (2001) modeled the global transport of  $\alpha$ - and  $\gamma$ -HCHs in 1993 and 1994 on a  $2^\circ \times 2^\circ$  grid, using a three-dimensional global atmospheric transport model with high spatial resolution, and compared the measured and modeled air concentrations for several Arctic locations. The assumption of constant ocean water concentrations limits the model to fairly short time periods. The model also does not include particle-associated atmospheric transport, which may be important at the low temperatures of high altitudes and high latitudes.

A different modeling approach to quantifying the transport of POPs to the Arctic was used by Commoner *et al.* (2000). Using emission estimates for PCDDs for North America, in combination with air mass trajectories calculated with the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) air transport model, they derived source-receptor relationships for several communities in the Canadian High Arctic. The model estimated the amount of PCDD emitted by each of 44 000 sources that is deposited at each of eight receptor sites in Nunavut over a one-year period (1 July 1996-30 June 1997). A fairly limited number of North American sources outside of Nunavut were found to be responsible for almost all of the PCDDs deposited on that territory.

## 2.3. Global and circumpolar sources of POPs including emission inventories

### 2.3.1. Historical and current uses

The previous AMAP assessment report (de March *et al.*, 1998) gives an overview of sources of POPs in circumpolar countries. Since the preparation and completion of the first assessment report, several regional and global studies have been completed.

One important example is the European emission inventory of heavy metals and POPs for 1990 that has gained substantial interest (Berdowski *et al.*, 1997). This study was prepared based on submissions of emission data from the parties to the Oslo-Paris Commission (OSPARCOM), Helsinki Commission (HELCOM) and Convention on Long-Range Transboundary Air Pollution (LRTAP). For the countries where sources and compounds were lacking in officially submitted data, default emission estimates were prepared to make the inventory

Table 2-1. Estimates of the global historical usage or production of selected deliberately produced POPs, by-products, and potential POPs (thousands of tonnes) (modified after Macdonald *et al.*, 2000). For most chemicals, the major-use areas were, and are at present, the north temperate regions of Europe, North America, and Asia.

Chemical	Use	Production period	Estimated total global usage/production (kt)*	Current annual global emissions (kt)**	Reference
<i>Legacy organochlorine pesticides</i>					
DDT	Insecticide	1950-present	2600	–	Voldner and Li, 1995
Toxaphene	Insecticide	1950-1993	1330	–	Voldner and Li, 1995
Technical HCH	Insecticide	1948-1997	10000	–	Li, 1999a
Chlordane	Insecticide	1945-1988	78	–	Barrie <i>et al.</i> , 1992
Aldrin	Insecticide	1950-1992	500	–	Barrie <i>et al.</i> , 1992
Dieldrin	Insecticide	1950-1992	34	–	Barrie <i>et al.</i> , 1992
<i>Legacy industrial organochlorines and by-products</i>					
PCBs	Various	1930-1992	1320	–	Breivik <i>et al.</i> , 2002a
CB 28			57	–	Breivik <i>et al.</i> , 2002a
CB 52			38	–	Breivik <i>et al.</i> , 2002a
CB 101			31	–	Breivik <i>et al.</i> , 2002a
CB 138			25	–	Breivik <i>et al.</i> , 2002a
CB 153			27	–	Breivik <i>et al.</i> , 2002a
CB 180			14	–	Breivik <i>et al.</i> , 2002a
<i>By-products</i>					
PCDD/Fs (as ITEQs)	By-products	1920-present	–	0.8-3.6×10 <sup>-5</sup>	UNEP, 1999
Hexachlorobutadiene	Intermediate	1920-present	10	0.002-0.02	Van de Plassche, 2001a; Environment Canada, 2000
HCB	Pesticide by-product	1920-present	–	0.012-0.092	Bailey, 2001
Pentachlorobenzene	By-product and dielectric fluid	1920-present	15	0.001-0.005	Environment Canada, 1993
Octachlorostyrene	By-product	1920-present	–	0.0003-0.001	US EPA, 1999a
Polychlorinated naphthalenes	Flame retardants and by-products	1920-1980 (as products) 1920-present (as by-products)	200-400 0.01	– <1×10 <sup>-4</sup>	Van de Plassche, 2001b Van de Plassche, 2001b
<i>Currently produced organohalogen – potential POPs</i>					
Short chain (C <sub>10</sub> -C <sub>13</sub> ) chlorinated paraffins	Cutting oils and flame retardants	1945-present	500-700	50	Tomy <i>et al.</i> , 1998; Muir <i>et al.</i> , 2000a
Medium and long chain chlorinated paraffins (C <sub>14</sub> -C <sub>30</sub> )	Flame retardant plasticizers	1930-present	2000-4000	250	OSPAR, 2000
TeBDE- and PeBDEs	Flame retardants	1960-present	70-120	8.5	BSEF, 2000
Octa- and Deca-bromodiphenyl ether	Flame retardant	1980-present	500-800	55	BSEF, 2000
Hexabromo-cyclododecane	Flame retardants	1980-present	100-200	16	BSEF, 2000
Tetrabromobisphenol-A	Flame retardants	1990-present	600-1000	121	BSEF, 2000
Perfluoroalkyl sulfonates	Surfactant and flame suppressant	1950-present	50•	?	US EPA, 2000b
Perfluoroalkanoic acids	Surfactant and flame suppressant	1950-present	50•	?	US EPA, 2000b
<i>Current-use pesticides</i>					
Lindane	Insecticide	1950- present	720	>2	Voldner and Li, 1995
Endosulfan	Insecticide	1956- present	57	1-2	Barrie <i>et al.</i> , 1992; USEPA, 2001
<i>Butyltins</i>					
Tributyltin	Algicide/fungicide	1960- present	100-500•	<10	Hoch, 2001
Mono- and dibutyltin	Stabilizers	1960- present	200-1000•	approx. 35	Hoch, 2001

\* Estimated global use (to the late 1990s) as reported or inferred from published reports. Values with • are rough estimates assuming linear growth in production over the use period.

\*\* Estimated annual emissions (kilotonnes) include use in consumer products and open applications as a pesticide. Volatilization losses (e.g., from treated soils) are not estimated; thus emissions of legacy OC pesticides are not given. Question mark indicates emissions are presently unknown.

complete. One of the major strengths of this study is thus the wide coverage of different priority substances as identified by OSPARCOM and HELCOM. This inventory covered a wide range of deliberately produced and accidentally formed POPs and related organic chemicals, including PAHs, PCBs, PCDD/Fs, HCB, several pesticides, and solvent chemicals. Another European emission inventory was recently presented by Pacyna (1999). This study focused on a subset of POPs, notably the PCBs, PCDD/Fs, HCHs, HCB, DDTs, and benzo[a]pyrene (B[a]P). One objective of this study was to estimate the historical trend in European emissions from 1970 to 1995 and to facilitate an analysis of the environmental response to changes in emissions. As individual constituents within groups of compounds (e.g., PCDD/Fs, PCBs, HCHs) may behave quite differently in the environment, a particular emphasis was on the emissions of individual isomers and congeners (Pacyna, 1999; Breivik *et al.*, 1999). Similar research efforts have been undertaken at the national level to estimate emissions of other POP constituents. In the United Kingdom, there have been studies on the national emissions for selected PCBs (Harrad *et al.*, 1994) and selected PCDD/Fs (Alcock *et al.*, 2001).

The best possibilities for deriving consistent and reliable source and emission estimates are for the deliberately produced POPs. Several databases have been estab-

lished for the global production and usage of such compounds. Table 2-1 presents selected values compiled in a recent review (Macdonald *et al.*, 2000) along with updated figures for PCBs and 'new' chemicals of concern. Among the insecticides discussed below, most information is available for HCHs (technical HCH and lindane). The global estimates are described in a series of publications (Voldner and Li, 1995; Li *et al.*, 1996; Li *et al.*, 1998a; Li, 1999a; 1999b) and were recently utilized to model the global distribution and fate of  $\alpha$ -HCH (Wania *et al.*, 1999a; Wania and Mackay, 1999).

#### PCBs

The estimates in Table 2-1 account for a reported historical global usage of approximately 1300 kt of PCBs of which more than 70% have been estimated as TrCBs, TeCBs, and PeCBs. The results further suggest that almost 97% of the global historical use of PCBs may have occurred in the northern hemisphere. Figure 2-3 shows the estimated cumulative global consumption pattern for total PCBs (Breivik *et al.*, 2002a). Most (approximately 86%) of the use (and thus emission to air) occurred in the industrialized, northern temperate latitudes, between 30-60°N. France, Germany, Italy, Japan, Spain, the U.K., and the U.S. were responsible for 68% of global PCB usage. The latitudinal distribution of

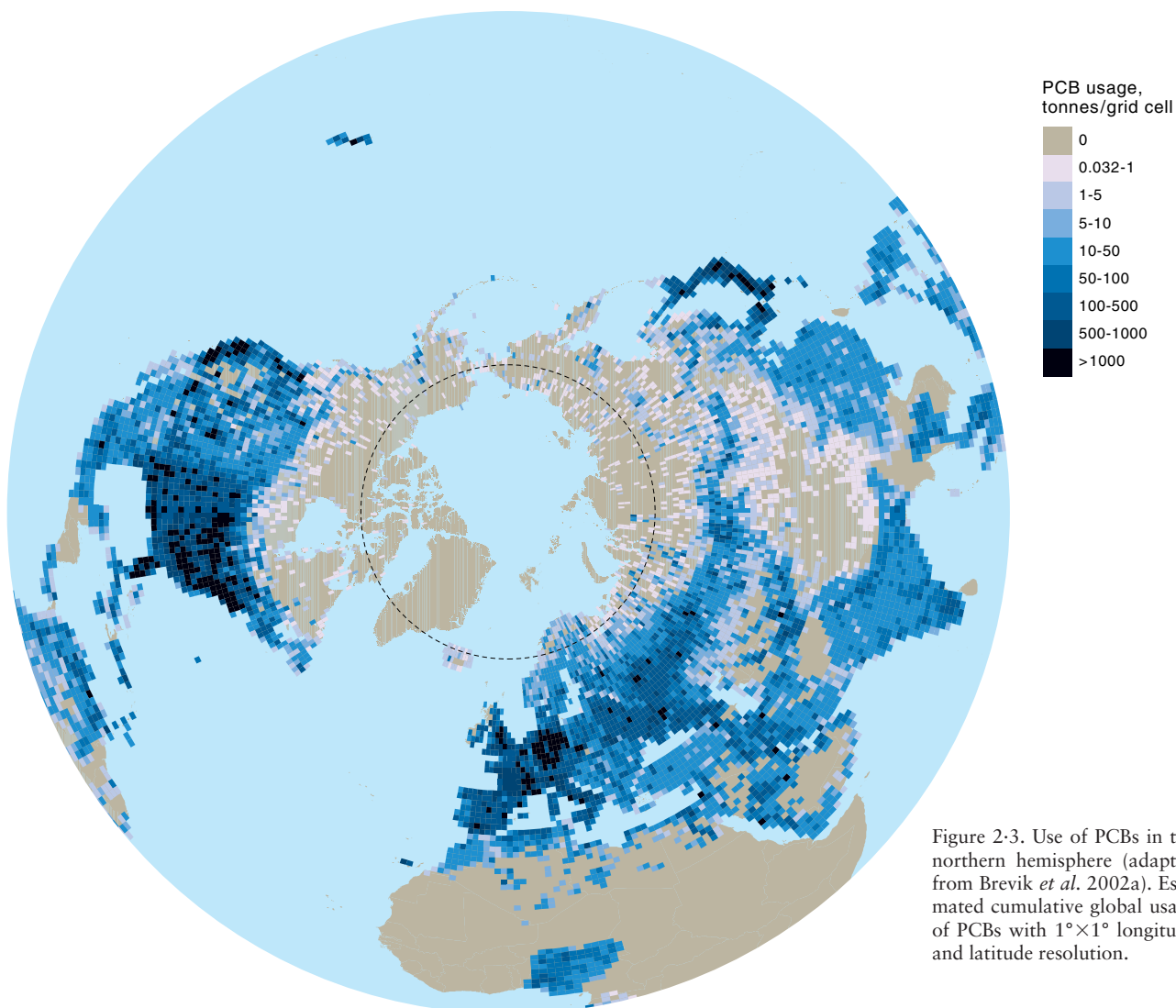


Figure 2-3. Use of PCBs in the northern hemisphere (adapted from Breivik *et al.* 2002a). Estimated cumulative global usage of PCBs with 1°×1° longitude and latitude resolution.



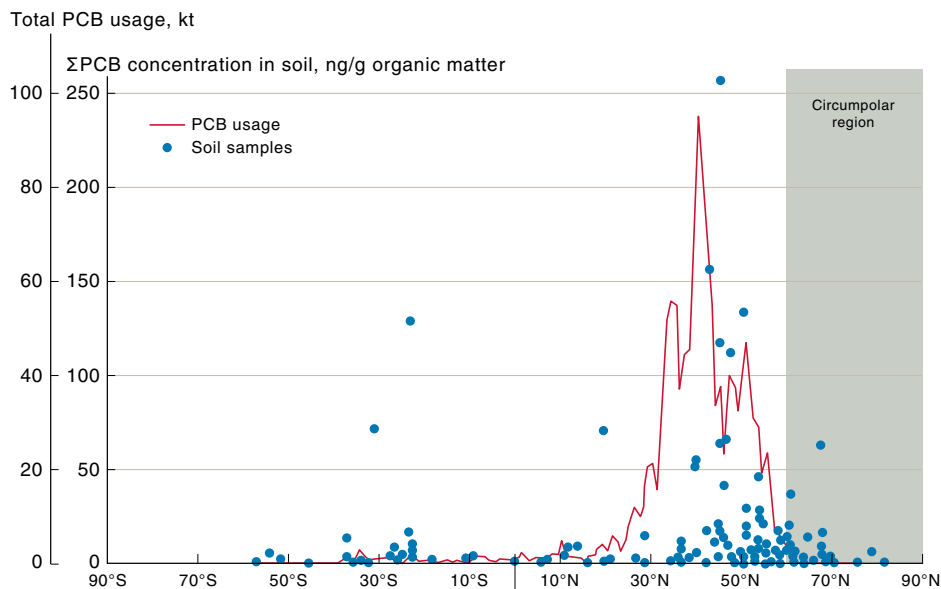


Figure 2-4. The latitudinal distribution of global PCB usage and PCBs in surface soils from Ockenden *et al.* (2002) illustrating the large reservoir in soils of the north temperate zone (45-60°N latitude).

PCBs in surface soils derived from a global survey (Ockenden *et al.*, 2002) is shown in Figure 2-4 along with the latitudinal distribution of PCB usage.

Using the data from the global background soil survey and information on the latitudinal land surface area and soil organic matter content, Ockenden *et al.* (2002) estimated that 45 kt of PCBs were present in background surface (0-5 cm) soils globally. A further burden is present in the sub-surface (>5 cm depth) soils. Adding estimates of the burden in the sub-surface (5-20 cm; 22.5 kt) and urban (0.2 kt) soils increases the estimated burden in soils globally to approximately 67 kt. This is close to recent estimates of the total emissions of PCBs into the environment, of 82-100 kt (Wania, 1999; Axelman and Broman, 2001).

Russia discontinued production of PCB in 1992 (AMAP, 2000). In a recent Russian inventory, it was found that approximately 27 kt of PCB is currently still in circulation in PCB-containing equipment in Russia and therefore represents a source to the environment (AMAP, 2000). Current quantities of PCB wastes and amounts that have been released from equipment or spilled during dismantling were an additional 4.4 kt. The Arctic Council Action Plan against Pollution (ACAP) has initiated a cooperative project to assist Russia in phasing out PCBs and in dealing with PCB-contaminated waste.

#### Technical HCH and lindane

The use of a mixture of  $\alpha$ -,  $\beta$ -,  $\gamma$ -, and  $\delta$ -HCH isomers, known as technical HCH, began in 1943, and global consumption since then has been estimated to be about 10 000 kt (Li *et al.*, 1998a). Technical HCH use was banned in most western countries and Japan in the 1970s but continued in India, Russia and China. China is reported to have been the major world producer accounting for about 4500 kt between 1945 and 1983 (Li *et al.*, 1998b; Li, 1999a). In 1990, India banned the technical product for agricultural use but kept it for public health uses. The circumpolar use pattern for technical HCH in 1980 and 2000, adapted from Li (1999b), is shown in Figure 2-5.

Lindane, containing almost pure  $\gamma$ -HCH (the only insecticidally active isomer), replaced technical HCH in Canada, the U.S., and western Europe during the late-

1970s to early-1980s and in China in 1991. Worldwide, lindane usage between 1948 and 1993 was estimated to be 720 kt (Voldner and Li, 1995). Although lindane is still important on a global scale, many countries have restricted or eliminated its usage. Breivik *et al.* (1999) give the total lindane application in Europe as 81 kt between 1970 and 1996, and 2.2 kt in 1996. According to Centre International d'Études du Lindane (CIEL, 1998), the average lindane consumption in Europe was 2.1 kt/yr from 1992 to 1997. France was the major user of lindane in Europe and in the world during this period, with an annual average consumption of 1.6 kt (CIEL, 1998), more than 76% of total lindane usage in Europe. In July 1998, lindane usage was stopped in France (CIEL, 1998). Lindane use in Sweden, Finland and other countries bordering the Baltic Sea, except Russia, was approximately 0.001 kt in 1995 (HELCOM, 2001). Estimated lindane use in Russia was 0.019 kt in 1995, a decrease from 0.044 kt in 1990 and 0.657 kt in 1985 (HELCOM, 2001). Lindane use was severely restricted in Russia in 1987.

Lindane became one of the top ten insecticides used in Canada during the 1990s (Environment Canada, 1992). However, pesticide information from Canadian companies is proprietary; therefore, surrogate cropland information was used to estimate lindane usage for Canada (Li and Bidleman, 2003). Intensive use of lindane on croplands was concentrated in the prairie region of Canada including the provinces of Alberta, Saskatchewan, and Manitoba where it is used as a seed treatment on canola (Waite *et al.*, 2001). The other major use is as a seed treatment on corn in eastern Canada. Annual lindane applications in Canada to canola and corn seed are estimated to have increased from approximately 0.163 kt in 1970 to 0.477 kt in 2000, with a cumulative usage of about 9 kt over this time period.

Information on lindane use patterns in the U.S. is limited, especially in the case of quantities used for seed treatment (Li and Bidleman, 2003). Direct spray application of lindane to crops was estimated to be 28 kt/yr in the mid-1990s, with 81% on pecans grown in the southeastern U.S. (NCFAP, 2001). Based on the National Center for Food and Agricultural Policy (NCFAP) survey, it seems likely that use of lindane in Alaska during the 1990s was negligible.

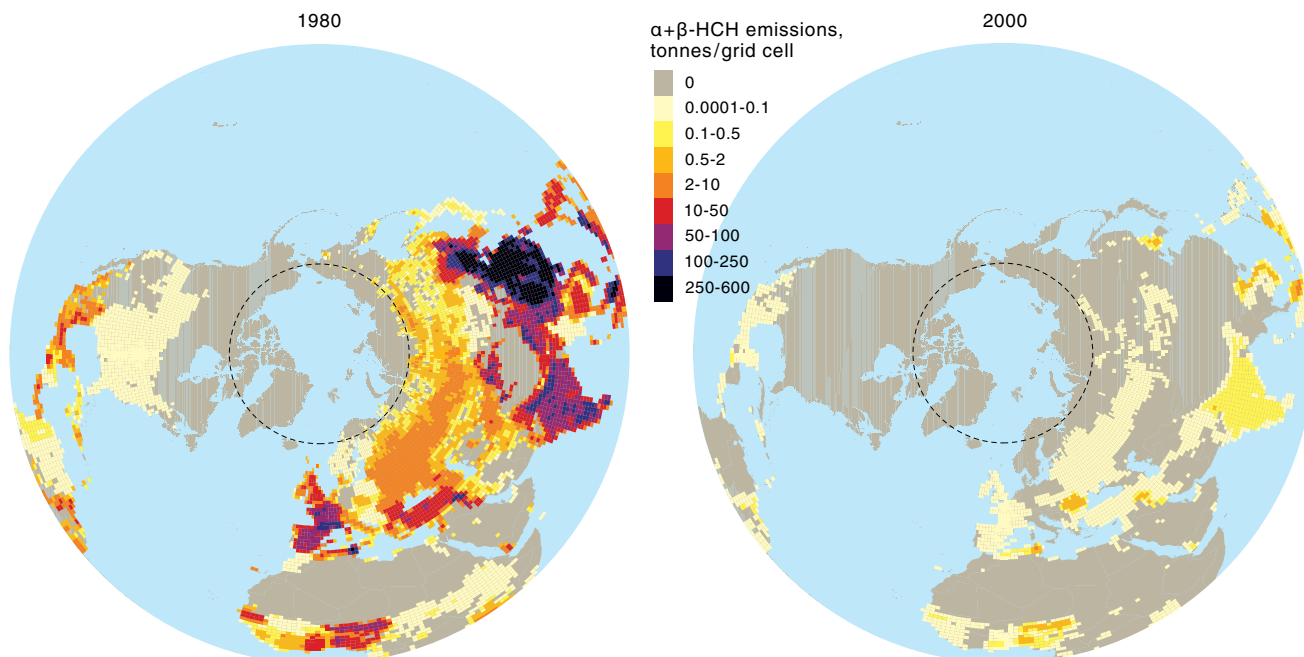


Figure 2-5. Use of technical-HCH in the northern hemisphere in 1980 and 2000 (adapted from Li and Bidleman, 2003).

Only 3.2 kt of lindane was applied in China between 1991 and 2000. The rest was exported or stockpiled. Consumption of lindane in China was 0.50 kt in 2000 (Li *et al.*, 2001a; Li and Bidleman, 2003).

#### DDT

Bans on the use of DDT were introduced in the U.S., Canada, and Japan and in most western European countries in the early-1970s. However, DDT production in China, India, Russia, and possibly other countries continued during the 1970s and 1980s. DDT production peaked in China during the late-1970s and was phased out in 1984 (Li *et al.*, 1999). DDT use in Russia continued until 1990 when 0.084 kt was reportedly used, down from 0.463 kt in 1980 (HELCOM, 2001). The total estimated global usage of DDT in agriculture from

1950 to 1993 is approximately 2600 kt (Voldner and Li, 1995). Between 1945 and 1972, the total cumulative production in the U.S. was 1340 kt, with domestic sales accounting for 645 kt and the rest exported. DDT production was discontinued in 1972 when some producers ceased operations. According to information provided to UNEP as part of the Stockholm Convention negotiations, DDT production continues in China and India, for use in fighting malaria and other insect-borne diseases in over 25 countries. The largest known current producer is India (approximately 0.007 kt/yr).

The gridded global annual usage of DDT on farmland for 1980 and 2000 is shown in Figure 2-6. Total global DDT usage in agriculture for 1980 was around 40 kt, and around 0.1 kt for 2000. Current uses for disease vector control in tropical countries are much larger

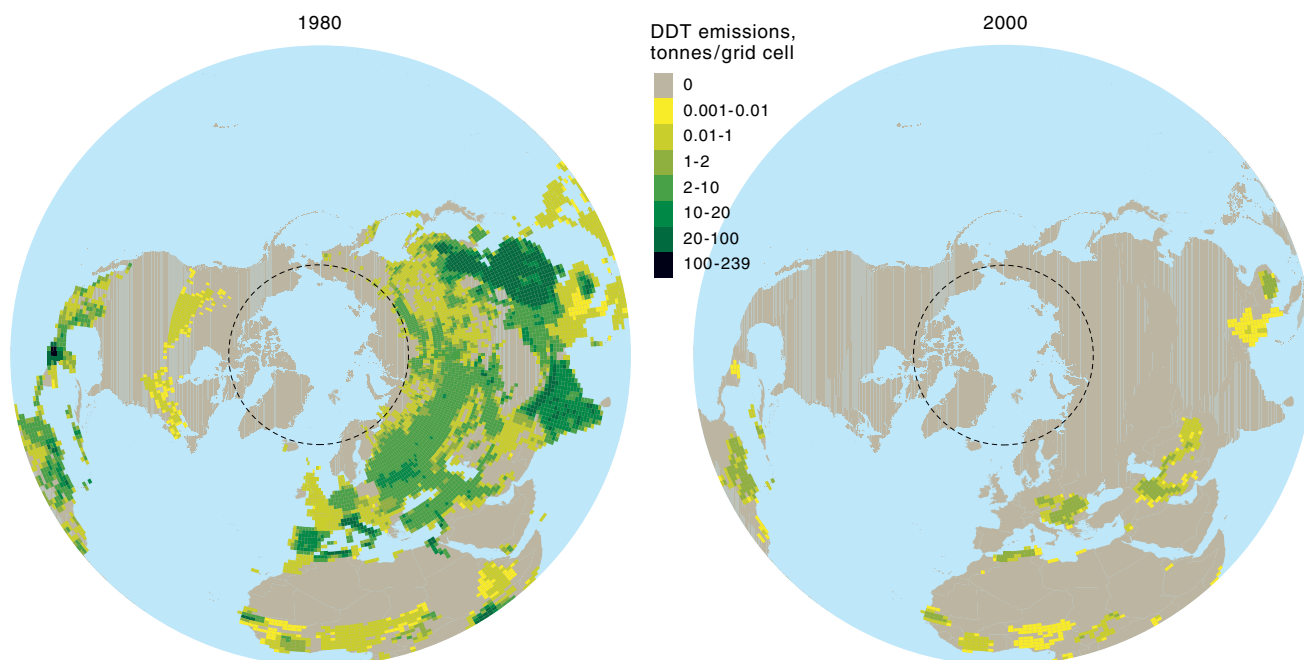


Figure 2-6. Use of DDT in the northern hemisphere in 1980 and 2000 (Li and Bidleman, 2003).

than agricultural uses; however, they are declining (UNEP, 2002). For example Mexico used about 1.80 kt of DDT in 1991. This was reduced to 0.497 kt in 1997 and further reductions are planned (UNEP, 2002). The trade publication *Europa Chemie*, gave India's cumulative DDT usage to 1989 as 280 kt for malaria control and 50 kt in agriculture (Li and Bidleman, 2003).

#### *Polychlorobornanes and polychlorinated camphenes (toxaphene)*

The major use region for toxaphene was the U.S. cotton belt, however, significant uses also occurred on other crop types. In 1983, toxaphene registration was canceled by the U.S. Environmental Protection Agency (EPA) and was banned in the U.S. in 1986. Use of toxaphene continued in Central America and Mexico until the early-1990s. Production for use in this region took place in Nicaragua until 1991. The total toxaphene residues left in agricultural soils in the U.S. at the beginning of 2000 are estimated to be about 29 kt. In 2000, almost 20 years after banning the use of this pesticide, approximately 0.360 kt of toxaphene was estimated to be emitted from agricultural soils in the U.S. (Li, 2001; Li *et al.*, 2001b). Total use in the U.S. and Mexico/Central America was estimated to be 540 kt.

The former Soviet Union was also a significant producer and user of polychloroterpenes. This insecticide was introduced in the former Soviet Union in the 1950s and was still widely used in Russian agriculture at the end of the 1980s. The total toxaphene usage within the former Soviet Union was less than 100 kt (Voldner and Li, 1993; 1995) and was applied mostly in the Ukraine (Kundiev and Kagan, 1993). Toxaphene was still used to control sugar beet pests in 1993 (Kundiev and Kagan, 1993). As of 1992, toxaphene was categorized as a 'severely restricted' pesticide in Russia (Voldner and Li, 1993).

The former East Germany was a major producer and exporter of polychloroterpenes under the name 'Melipax' until 1990 (Heinisch *et al.*, 1994).

China produced toxaphene between 1967 and 1972 with a maximum annual production of approximately 1 kt in 1970 and a total production around 3.6 kt (Li and Bidleman, 2003).

With the signing of the Stockholm Convention, toxaphene is likely to be consigned to the history books. Information supplied to UNEP as part of the Stockholm Convention negotiations suggests it is no longer produced.

#### *Diene-organochlorine insecticides*

Members of the large diene-organochlorine group (including chlordane/heptachlor, dieldrin/endrin/aldrin, mirex and chlordecone) were formerly used as insecticides and use-information was provided in the previous AMAP POPs assessment (de March *et al.*, 1998). Chlordecone, mirex, aldrin, endrin and chlordane are included in the UN ECE POPs Protocol under the LRTAP Convention. Chlordecone has not been measured in Arctic biota or abiotic samples. This chemical was marketed in the U.S. as Kepone and pesticide registration was canceled in the U.S. in 1978 following widespread contamination of the James River in the eastern U.S. from factory releases. Mirex, formed by further chlorination of chlordecone, was more widely used as an insecticide, termiticide, and flame retardant. Total production in the U.S. was esti-

mated to be 1.5 kt, however, global production has not been estimated. The UNEP POPs survey indicated that there is no current mirex production. China, however, requested a production exemption for mirex for use as a termiticide and it is therefore likely that minor uses remain.

#### *Chlordane*

Technical grade chlordane is a mixture of at least 120 compounds, with the major constituents being *cis*- and *trans*-chlordane, heptachlor, *cis*- and *trans*-nonachlor, chlordene, and others (Dearth and Hites, 1991). Chlordane was released into the environment primarily from its application as a soil insecticide and termiticide. The U.S. was the major world producer and user of chlordane. However, there was limited use in western Europe, the former Soviet Union and tropical Asian countries. In the U.S., chlordane was used extensively prior to 1983, and from 1983 to 1988 it was registered for termite control. In 1997, the sole U.S. manufacturer of chlordane voluntarily ceased production at all of its national and international facilities. There are production facilities in Singapore and China, however.

#### *Aldrin, dieldrin, endrin, and heptachlor*

World sales of aldrin and dieldrin ceased in 1991 when the major manufacturer voluntarily stopped production. Endrin production ceased in the mid-1980s. Old stocks of these chemicals, particularly dieldrin, were, however, donated to African countries in the 1980s-90s for insect control, so emissions to the environment have continued (UNEP, 2002). Dieldrin was mainly used as a soil insecticide. In tropical countries it was used for the control of locusts and for disease vector control.

#### *Endosulfan*

Use of endosulfan in the U.S. has recently been estimated to be 0.64-0.95 kt per year (USEPA, 2001). Endosulfan is also used in Canada and Europe. Global use is thus estimated to be in the range of 1-2 kt per year. It is registered for use on a wide variety of speciality crops (pecans, pumpkins, squash). It is applied by boom sprayer and by aircraft, and thus, has the potential for significant dispersal in the environment.

#### *Butyltins*

TBT and other organotin compounds were initially used in agriculture. Subsequently, TBT has had wide application as a marine antifoulant starting in the 1960s. It has also been used as a wood preservative. TBT is found to provide effective protection for boat hulls at release rates of less than 4 µg/cm<sup>2</sup>/day, and has been a popular antifoulant because it maintains its efficacy for up to five years compared to about three years for other conventional applications. TBT's most important entry route to the sea is directly from boats, aquaculture pens, moorings, and industrial cooling pipes to which products containing it have been applied. It may also enter ocean waters from municipal waste water and sewage sludge.

Organotin-stabilized PVC is used in a wide variety of applications including: pipes for drinking water, waste water, and drainage; packaging materials; and, window frames. The organotin constituents have been shown to leach from PVC and other materials leading to contami-



nation of food, drinking water, municipal water and, sewage sludge (Forsyth *et al.*, 1992; 1994; Fent, 1996b; Forsyth and Jay, 1997). In a Japanese study, plastic products such as baking parchments of siliconized paper, polyurethane gloves, dish-washing sponges, and cellophane film for wrapping food bought from a supermarket, were found to contain MBT, DBT, and TBT (Takahashi *et al.*, 1999). No information is available regarding leaching from PVC materials at dump sites, but this is a potential emission source to the environment in Arctic communities.

TBT is degraded by sequential debutylation to DBT, MBT and eventually to relatively non-toxic inorganic tin compounds, primarily by organisms. The breakdown of TBT is much slower in anaerobic sediments than in water (Clark *et al.*, 1988), and therefore, contaminated sediments may act as an important environmental reservoir for TBT as well as MBT and DBT, long after their use has been curtailed.

Most circumpolar and northern European countries now partially regulate the use of TBT (France, 1982; U.S., 1986; U.K., 1987; Canada, 1989; Europe, 1991). Regulations vary, but generally, only controlled release formulations are permitted and TBT-based antifoulants are prohibited for boats smaller than 25 m. The U.N. International Maritime Organization (IMO) has agreed to a global ban on new use of TBT on ship hulls from 1 January, 2003. After 2008, TBT-based antifouling paints must be removed from ship hulls or encapsulated with an impermeable paint so no leakage to the environment can occur.

Global production of organotin compounds was estimated to be 50 kt in 1992 (Mercier *et al.*, 1994). Biocides (primarily trialkyltins) make up approximately 20% of the total annual production (Bennett, 1996) with PVC stabilizers (primarily mono- and dialkyltins) being approximately 70% (Hoch, 2001). Total bis(tributyltin) oxide (TBTO) production in the Federal Republic of Germany, was estimated to be 2.0 kt, of which 70% was used in antifouling paints, 20% as a wood preservative, and 10% as a preservative in textiles, leather, and other materials (WHO, 1990). No estimates are available for current annual global consumption or for use patterns (Hoch 2001).

#### *HCB and pentachlorobenzene (PeCBz)*

HCB is produced as a by-product in the production of a large number of chlorinated compounds, particularly lower chlorinated benzenes, and in the production of several pesticides. In the 1960s, it had limited use as a fungicide. HCB is emitted to the atmosphere in flue gases generated by waste incineration facilities and metallurgical industries. Both HCB and PeCBz have previously been identified as by-products from production of chlorinated solvents (Environment Canada, 1993; USEPA, 1999c).

PeCBz was used in dielectric fluids in PCB-containing transformers. A Canadian survey found that up to 0.2 kt of PeCBz was in use in transformers in the early-1990s. However, this figure is now out of date due to the removal and destruction of most PCB-containing equipment. Nevertheless, it illustrates that older PCB-containing devices in many circumpolar countries are potential sources of PeCBz. PeCBz is also an impurity in technical

HCB used in the manufacture of various substances such as pentachlorophenol, pentachlorothiophenol, and pyrotechnical products (HELCOM, 2001).

A recent assessment of global sources of HCB (Bailey, 2001) concluded that total current emissions could be around 0.023 kt/yr with a possible range between 0.012 and 0.092 kt/yr (Bailey, 2001). A substantial portion of the HCB measured in the atmosphere had unidentified sources and/or was due to revolatilization (i.e., currently known emissions could not account for prevailing global air concentrations). Russia was estimated to emit 0.011 kt/yr of HCB in 1995, with other countries bordering the Baltic Sea accounting for about 0.003 kt/yr (HELCOM, 2001).

#### *OCS*

Maximum emissions of OCS in the Great Lakes region of North America occurred in the 1960s (USEPA, 1999a), probably due to disposal of wastes related to chlorine manufacturing using graphite electrodes, a process abandoned in the 1970s. In Norway, emission of OCS and other chlorostyrenes was traced to production of magnesium using graphite electrodes (Lunde and Ofstad, 1976). Recent measurements have been made in Germany (Bester *et al.*, 1998), however, in general, emissions and use of OCS are not well documented. Little is known about global emissions of OCS, however, magnesium production and chlorine manufacturing were historically important sources (USEPA, 1999a). Hydroxyheptachlorostyrene, a metabolite of OCS, was identified recently as a major hydroxylated metabolite in polar bear and human plasma in the Arctic (Sandau, 2000; Sandau *et al.*, 2000).

#### *PCDD/Fs*

PCDD/Fs enter the environment as by-products of industrial processes. The most significant sources are low-temperature, incomplete incineration of chlorine-containing materials such as plastics. Other major sources include thermal processes, such as motor vehicle fuel combustion in countries where leaded fuel containing chlorine scavengers is still used, and metallurgical industries. Pulp and paper mills using chlorine in the bleaching process have been important sources for inputs to the aquatic environment. PCDD/Fs are also trace contaminants in chlorophenoxy herbicides, PCB formulations, and chlorophenol wood preservatives.

Few studies are available estimating circumpolar emissions of PCDD/Fs. Rough estimates of the global emissions of PCDD/Fs are available from Brzuzy and Hites (1996). In addition, an overview of the current status of national and regional emission inventories for PCDD/Fs globally can be found in UNEP (1999). This report includes estimates of PCDD/F emissions for northern Europe, Canada, the U.S., and Japan. Japan and the U.S. were considered the largest sources of PCDD/Fs (5300 g TEQ/yr and 2700 g TEQ/yr, respectively). Sweden, the only Nordic country in the survey, had very low annual estimated PCDD/F fluxes (22 g TEQ/yr) while Canadian emissions were estimated to be 290 g TEQ/yr.

#### *PAHs*

PAHs can be classified by their source type: petrogenic, biogenic, and pyrogenic. PAHs of petrogenic origin are



related to petroleum, including crude oil and its refined products. The presence of naphthalene and its alkyl-substituted homologues in sediments is characteristic of unweathered petroleum (Robertson, 1998). The naphthalene:phenanthrene ratio is much greater than 1.0 for most petroleum types. PAHs of petrogenic origin are also characterized by homologous families of related PAHs (naphthalenes, phenanthrenes and dibenzothiophenes), where the parent PAH for each family is less abundant than the alkylated homologues (Page *et al.*, 1999). PAHs of biogenic origin are generated by biological processes or by the early stages of diagenesis in marine sediments (e.g., perylene) (Venkatesan, 1988). PAHs with 4- to 6-ring hydrocarbons are generally of pyrogenic origin and are generated by the combustion of fossil fuels and recent organic material. The relative abundance of these PAHs to 2- to 3-ring hydrocarbons can be used to help distinguish between petrogenic and pyrogenic sources (Robertson, 1998). PAHs in the atmosphere are primarily from combustion of fossil fuels to produce electricity and heat, vehicular exhausts, forest fires, as well as industrial activities such as non-ferrous and ferrous metal production, and fertilizer production. PAHs detected in the Arctic atmosphere reflect contributions of emissions from middle and high latitudes in the circumpolar countries (Hoyau *et al.*, 1996; Halsall *et al.*, 1997; Masclet *et al.*, 2000).

#### PCNs

Until the 1970s, PCNs were high volume chemicals, with the worldwide production being approximately 9.0 kt/yr in the 1920s (Jakobsson and Asplund, 2000). Production of PCN decreased significantly after 1977. The production of PCNs in the U.S. stopped in 1980. Given the relatively high production volumes in the 1920-1930s, it seems likely that total PCN production was in the range of 200-400 kt. Crookes and Howe (1993) concluded that it was not possible to derive a global emission estimated for PCNs. Furthermore, Haglund *et al.* (1993) found that PCNs were contaminants in technical PCB formulations. Falandysz (1998) mentions technical PCBs as a potential source of PCNs and estimated a potential release of 0.1 kt based on the worldwide production of PCBs and a median value of 0.0067% PCNs in Aroclor and Clophen mixtures. A significant current source of PCNs is incineration (Crookes and Howe, 1993; Falandysz, 1998). Indeed, PCN congeners associated with combustion are elevated in Arctic air in winter (Harner *et al.*, 1998; Helm and Bidleman, 2003).

#### SCCPs

SCCPs are used mainly as extreme temperature additives in metal working fluids for a variety of engineering and metal working operations such as drilling, machining/cutting, drawing, and stamping (Environment Canada, 1993; Tomy *et al.*, 1998). They are also used in paints and sealants and have a minor but environmentally significant use as fat liquors in the leather working industry. Due to concerns about the toxicity, bioaccumulation, and persistence of SCCPs, use of these chemicals is declining as users switch to alternative products. Use since the 1940s, however, has been very high compared to most aliphatic chlorinated hydrocarbons of similar molecular weight (Table 2.1). Global manufacturing capacity is estimated to be 50 kt/yr.

#### MCCPs

MCCPs are mainly used as flame-retarding plasticizers in polyvinyl chloride plastics and releases to the environment are relatively small compared to production. MCCPs are large-volume production chemicals with global capacity estimated to be 0.25 kt/yr (Muir *et al.*, 2000a; OSPAR, 2000), and a steady production during the 1990s. Thus it is likely that over 5000 kt have been produced since the mid-1970s when production volumes were significantly increased. Annual releases of MCCPs to air and water in western Europe were estimated to be 1.87 kt in 1998, compared with production of 55 kt (OSPAR, 2000). There are, as yet, no reports on these compounds in the Arctic environment.

#### TeBDE and PeBDE

TeBDE and PeBDE compounds have been used since the 1970s as flame retardants in, among other things, polyurethane foams (de Boer *et al.*, 2000). The commercial PeBDE product is comprised mainly of pentabromodiphenyl ether with tetra- and hexabromodiphenyl ether impurities (Hardy, 2001; de Boer *et al.*, 2000). Annual worldwide production of PeBDE in 1990 was estimated to be 4.0 kt (Arias, 1992) and use increased during the 1990s. Global use in 1999 was 8.5 kt, of which >90% was in North America due to a phase-out of use in Europe (BSEF, 2000). Assuming a steady increase in production since the early-1980s, this implies a total production in the range of 70-120 kt. Environmental concentrations were increasing during the 1990s in the Canadian Arctic (See Section 5.4.6).

#### OcBDE and DeBDE

The main use of OcBDE and DeBDE technical products is in high impact polystyrene used to manufacture electrical enclosures (e.g., television sets). The major component of the OcBDE technical product is a heptaBDE as well as hexa-, octa- and nonaBDE impurities. Small amounts of octa- and nonaBDEs are found as impurities in DeBDE. A minor but important use of DeBDE is in flame-retarding of fabrics where it is applied as a fabric back coat encapsulated in latex (Hardy, 2001). Worldwide (i.e., mainly western Europe and North America) use of OcBDE and DeBDE technical products was 6.0 and 30 kt respectively in 1990 (Arias, 1992). Global demand for OcBDE and DeBDE was 3.8 kt and 54.8 kt, respectively, in 1999. Assuming a gradual increase in production since the early-1980s, this implies a cumulative production to date of 500-800 kt Oc- and DeBDEs. Most of this production was used in areas that are potential source regions for atmospheric transport to the Arctic, i.e., countries of the western Pacific/eastern Asia, North America and western Europe.

#### Other brominated flame retardants

Hexabromocyclododecane (HBCD) and TBBPA are other major brominated flame retardants. HBCD has replaced pentaBDEs in Europe, with 8.9 kt used in 1999 (BSEF, 2000). HBCD is produced by bromination of cyclododecane in a batch process and has been used for about 20 years. TBBPA is the major brominated flame retardant in current use primarily in electronic circuit boards (121 kt use globally in 1999; BSEF, 2000). TBBPA is covalently bonded into most materials but

some TBBPA is not bound (Sellström and Jansson, 1995), leading to some environmental releases, though probably minor compared to PBDEs.

#### *Perfluoroalkyl sulfonates*

Perfluorinated surfactants are employed for industrial and commercial applications and are used in lubricants, paints, polishes, food packaging, and fire-fighting foams (Hekster *et al.*, 2002; Key *et al.*, 1997). PFOS is an important perfluorinated surfactant as well as a precursor to other perfluorinated surfactants (Hekster *et al.*, 2002; Key *et al.*, 1997). In 2000, the estimated annual U.S. production of PFOS was 2.9 kt, but as a result of the 3M Company's phase-out, no PFOS production is anticipated in the U.S. in 2003. Total production of all fluorinated surfactants (anionics, cationics and neutrals) was 0.2 kt in the mid-1970s (Fielding, 1979), which suggests that a large increase in PFOS production occurred during the past 25 years. Assuming similar production volumes for the past ten years and smaller volumes since the introduction of these chemicals in the late- 1940s, it seems reasonable to estimate that at least 50 kt have been produced. Most of this production would have been released into the environment due to open uses as surface treatments. Production of PFOS, or its precursors, by other manufacturers outside of the U.S. or European Union may occur but is not presently documented. The properties of PFOS suggest that it is a poor candidate for long-range atmospheric transport. These facts have previously led to speculation that the global dissemination of PFOS must occur via an airborne neutral derivative that yields the free acid upon degradation (Renner, 2001). Long-range transport by ocean currents cannot be ruled out given the stability of PFOS.

#### *Perfluoroalkanoic acids*

Like PFOS, these compounds, particularly perfluorooctanoic acid (PFOA), are employed for industrial and commercial applications and are used in lubricants. PFOA has a unique application as a plasticizer in the fluorinated polymer Teflon®. PFOA was detected at low levels in a global survey of marine and freshwater biota, which included ringed seals (*Phoca hispida*) and polar bears, but results were not reported (K. Kannan, Michigan State University, pers. comm., 2002). No production information is publicly available on the perfluoroalkanoic acids.

### 2.3.2. Local/regional sources within the Arctic

Local sources of POPs are considered to be only minor contributors to contamination of the Arctic environment when considered on a continental or ocean-wide scale. On a local scale, however, household heating in settlements, burning of hydrocarbons for electricity and transport, and incineration and open burning of garbage, contribute significantly to the input of organic pollutants such as PAHs. Emissions of PCDD/Fs from these combustion sources may also be important in Nunavut (Canada) although considered very minor compared to heavily populated regions of North America (Commoner *et al.*, 2000). In several Arctic regions, mineral exploration, coal mining and heavy industry account for the highest input of persistent pollutants such as PAHs and PCBs.

Current and former military bases throughout the circumpolar Arctic, especially those with older radar equipment, have been previously identified as sources and were discussed in the previous AMAP POPs assessment (de March *et al.*, 1998). Many of these sites have undergone cleanup which include removal of contaminated equipment and soils. For example, the cleanup of 21 sites in the Canadian Arctic is scheduled for completion in 2008 (Canada DND, 2001) and there have been similar campaigns in Alaska, Greenland and Norway. The situation in the Russian Arctic regarding PCB use at military sites is unclear. The Russian Ministry of Defence did not take part in the inventory of PCBs or PCB-containing equipment in Russia because their position was that PCB-containing equipment was not in use anymore (AMAP, 2000). Thus no information is available on the disposal of PCBs and PCB-containing equipment from military sites in the Russian Arctic.

#### *Svalbard*

On Svalbard, local coal mining activities and settlements are the major primary sources of POPs (Kovacs, 1997), with coal-fired power plants located in five settlements being sources of PAHs to the atmosphere. Calculation of sulfur- and particle distribution as primary indicators of contaminant sources for the coal mines at Longyearbyen and Svea suggests that mining activities could be a major source of PAHs to the Svalbard environment (Holte *et al.*, 1996; dos Santos *et al.*, 1996). It is also suggested that the Russian coal mines at Barentsburg represent comparable sources. In a study of profiles of pyrogenic PAHs in lake sediments on Spitsbergen, Rose *et al.* (2003) found highest fluxes of PAHs in Tenndammen, a lake within 20 km of the coal mining towns of Barentsburg and Longyearbyen.

The major sources of PAHs in marine sediments are thought to be of natural petrogenic origin resulting from petrogenic seepage from the sea bed and erosion of coal seams (Akvaplan-niva, 1998). Higher  $\Sigma$ DDT concentrations (0.38 ng/g dw) were found at a deep site in Kongsfjorden relative to nearshore sites in eastern Svalbard (Lomfjorden, Erik Eriksen Strait) which had  $\Sigma$ DDT <0.2 ng/g dw. OC concentrations in sediments near the settlements in Svalbard were not assessed, however (Akvaplan-niva, 1998). Hop *et al.* (2001) found higher  $\Sigma$ PCB and PAH concentrations in macrobenthos sampled near the settlements of Billefjorden, Grønfjorden, Adventfjorden, and Kongsfjorden in Svalbard. PCB profiles from fjords with Russian settlements had a higher proportion of lower chlorinated congeners compared to fjords with Norwegian settlements, probably because of local pollution and the different PCB products used in each community.

#### *Coastal Norway and western Russia*

PCBs have been found in surface sediments from harbors in northern Norway (Harstad, Hammerfest, Tromsø, Honningsvåg) and Russia (Kola Bay, Guba Pechenga) (Dahle *et al.*, 2000) with relatively high (10-100 ng/g dw) levels found in comparison with background sites reported previously in the previous AMAP assessment (de March *et al.*, 1998). PCB levels and congener composition among harbors was similar (higher chlorinated PCBs predominated) except for Guba Pechenga, where lower chlorinated PCBs predominated for unknown rea-

sons. No large differences in OC concentrations were found between Norwegian and Russian harbors. Harbors probably constitute hotspots for PCBs in the Arctic marine environment with higher concentrations (up to 10 times) relative to non-harbor areas.

In a follow-up study in Kola Bay (1997), much higher concentrations of most OCs in question were found compared to a control location (Guba Zapadnaya Litsa) (Savinova *et al.*, 2000a). The stations with the highest levels of  $\Sigma$ PCBs, toxaphene and HCB were located close to the harbor of Polarnyy City, situated on Kola Bay, 20 km north of Murmansk. Concentrations of  $\Sigma$ PCB near Polarnyy City ranged from 650 to 8700 ng/g dw while levels were 24 ng/g dw in sediments in the outer reaches of Kola Bay. A surprising observation was the presence of relatively high levels of toxaphene in surface sediments in Kola Bay (3.5-680 ng/g dw) and in Guba Zapadnaya Litsa (19-85 ng/g dw). Although there are few other toxaphene measurements in marine sediments for comparison, the geometric mean levels in both Kola Bay and Guba Zapadnaya Litsa are higher than observed in freshwater surface sediments in the Great Lakes (Pearson *et al.*, 1998) or in northwestern Europe (Rose *et al.*, 2001). This suggests significant use of toxaphene in northern Russia, at least in the Kola Peninsula, possibly for insect control in urban areas or on ships.

#### Jan Mayen

On the island of Jan Mayen, elevated PCB levels were found in soil within and in the immediate proximity of a dump used between 1960 and 1995 (Gabrielsen *et al.*, 1997). The source of the PCBs was seven transformers, each containing approximately 200 L of oil, that were emptied into the dump in the late-1970s. Despite extensive sampling, it was only possible to estimate the amount of PCB in the dump as being between 10 and 2000 kg. There was no evidence of local PCB contamination extending from the dump into the freshwater lake or to other areas on the island. Up until 1989, 2000-3000 L of lubrication oil and other waste products (white spirits, paint) were also dumped. The dump is located 25 m from the edge of a coastal cliff of porous volcanic rock, which is subject to erosion, primarily by seawater. Currently, the rate of erosion is unknown.

#### Greenland

The distribution of persistent halogenated organics was studied during 2000 near the western Greenland villages of Quaqortoq (3500 inhabitants), Igaliko (30 inhabitants) and Usuk (background site 3-5 km from Igaliko) (Christensen *et al.*, 2002; Vorkamp *et al.*, 2002). The animals analyzed were shorthorn sculpin (*Myoxocephalus scorpius*), Greenland cod (*Gadus ogac*), spotted wolffish (*Anarhichas minor*), starry ray (*Raja radiata*) and blue mussels (*Mytilus edulis*). All concentrations of persistent halogenated organics were low in mussels. The highest concentrations of  $\Sigma$ PCBs and PBDEs were observed in fish from Quaqortoq followed by Igaliko and Usuk. This trend was not as pronounced for chlorinated pesticides, which implied that there were local sources of PCBs and PBDEs, but not chlorinated pesticides. The levels of PBDEs were 15-24 times lower than PCB levels measured in the same individuals, except for shorthorn sculpin collected at Quaqortoq, where the level of  $\Sigma$ PCBs

was 40 times higher. The results suggest that villages are sources of legacy OCs such as PCBs, as well as currently used PBDEs, to the nearby marine environment.

Previous studies of scallops (*Chlamys islandicus*) and sediments have shown that Thule Air Base is a local source of PCBs to the marine environment (Kjølholt and Hansen, 1986).

#### Canada

From 1996 to 2001, additional studies and remediation of old military sites contaminated with PCBs were conducted in the Canadian Arctic. Detailed assessments of PCB contamination were conducted at two sites in the eastern Canadian Arctic, Saglek Bay (58°29'N, 62°40'W) in northern Labrador, and Resolution Island at the south-eastern tip of Baffin Island.

At Saglek Bay, high levels of  $\Sigma$ PCBs (more than 50  $\mu$ g/g dw in soils) were found at a former military radar site (ESG, 2002). The cleanup from 1997 to 1999 effectively removed the terrestrial sources of contamination to the surrounding land and to the marine environment of Saglek Bay.  $\Sigma$ PCB concentrations in sediment cores from lakes near the former radar facility declined with increasing distance from the site (Betts-Piper, 2001). Four of eight lakes within 5 km of the site had elevated levels (>100 ng/g dw) compared to background sites 30 km away (0.3-17 ng/g dw) or to other remote lakes in the Canadian Arctic (Muir *et al.*, 1996a). The PCB congener profile in both the near-site and remote lakes had a clear Aroclor 1260 profile similar to the product used at the radar facility. Bright *et al.* (1995a) and Duskenko *et al.* (1996) reported similar results from a study of 24 military radar sites in the Canadian Arctic. They concluded that the radar sites were acting as sources of contamination to nearby lakes via aerial redistribution. This was confirmed at Saglek Bay by the detailed study by Betts-Piper (2001).

Marine investigations were conducted to study PCB contamination in Saglek Bay over three seasons from 1997 to 1999. These studies delineated PCB-contaminated sediments and quantified PCB uptake in the marine food chain, including benthic invertebrates, bottom-feeding fish, pelagic fish, marine mammals, and seabirds. They indicated that the contaminated sediments represented a source of PCBs to local wildlife. Inputs of PCBs to the marine environment occurred from a contaminated beach area associated with the original station. Sediment  $\Sigma$ PCB concentrations near this beach were 500 ng/g to 130 000 ng/g dw in the intertidal sediment, and 600 ng/g to 6200 ng/g dw in the Saglek anchorage. Concentrations declined exponentially with distance, approaching background concentrations (<2 ng/g) within 6-10 km (ESG, 1999; 2002). PCBs were also present in deepwater sediments of the bay, particularly immediately offshore and west of the contaminated beach (Figure 2-7).

The samples of invertebrates collected from the immediate vicinity of the contaminated beach contained PCBs at high concentrations, consistent with the concentrations measured in the beach sediment (ESG, 1998). Snails and scuds from the intertidal area had  $\Sigma$ PCB concentrations of 8000 and 49 000 ng/g ww, respectively. The concentration in sea urchins from this area was slightly lower (5200 ng/g ww). Invertebrate tissue con-



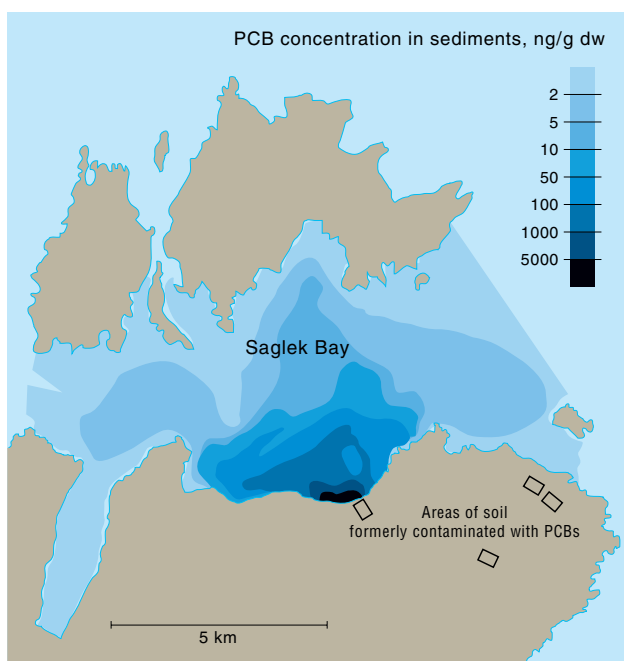


Figure 2-7. PCB contamination in marine sediments of Saglek Bay in northern Labrador based on 243 surface sediment sample results from 1997, 1998 and 1999 indicating the dispersion from a contaminated beach.

taminant concentrations generally return to near-background concentrations within 7-10 km of the formerly contaminated beach.

$\Sigma$ PCB concentrations were relatively high in short-horn sculpin sampled in the nearshore area close to the contaminated beach, exceeding 1000 ng/g ww (whole body minus liver, Aroclor equivalent) in two-thirds of the samples and exceeding 10 000 ng/g ww in a few samples. Concentrations also exceeded 1000 ng/g ww in some shorthorn sculpin samples collected at sites up to 1.5 km east and 3 km west of the beach. PCB concentrations in muscle and liver of Arctic char (*Salvelinus alpinus*) did not vary with the proximity of the fish to the contaminated beach (ESG, 1998). PCB concentrations ranged from a low of 13 ng/g ww to a high of 66 ng/g ww (Aroclor equivalent), with both of these extremes measured in char collected at the Okak reference site (ESG, 1998). These were similar to results for PCBs in anadromous char from the Labrador coast (Muir *et al.* 1999c; 2000c) and are consistent with the largely pelagic feeding habits of char.

$\Sigma$ PCB concentrations in seals from the Saglek Bay area varied over a wide range, from a low of 500 ng/g ww in the adipose tissue of one 11-year-old seal, to a high of 9400 ng/g ww in a 10-year-old seal. The result for this seal is exceptionally high, exceeding the results for the other seals by a factor of 4-6. It is also much higher than results reported for ringed seals from the community of Nain, further south on the Labrador coast, and elsewhere in the Arctic (Section 4.4.6.1).

Black guillemots (*Cepphus grylle*) in Saglek Bay, Labrador also had elevated  $\Sigma$ PCB concentrations due to marine sediment contamination around the former military site.  $\Sigma$ PCB concentrations in liver of nestlings ranged from 15 to 46 ng/g ww in a reference group, 24 to 150 ng/g ww in a group from moderately exposed islands, and 170 to 6200 ng/g ww in the highly exposed beach

group. Biomarker responses were dose-dependent and in some cases, sex-dependent (see Section 6.3.3.3).

Similar contamination by PCBs was investigated at Resolution Island (61°35'N, 60°40'W) at the southeastern tip of Baffin Island in the eastern Canadian Arctic (ASU, 1997; 2001). Initial assessment of the site in the early-1990s showed it to be highly contaminated with PCBs (ESG, 1994). The site contained at least twenty buildings requiring demolition, along with a large amount of visible debris, tanks, and many barrels. Buildings in the communications complex contained electrical equipment suspected of containing PCBs, and other buildings contained asbestos. The total volume of soil which was contaminated with  $\Sigma$ PCBs at concentrations above 50 000 ng/g (exceeding soil guidelines for PCBs under the *Canadian Environmental Protection Act*) was estimated to be 5000 m<sup>3</sup> with about 20 000 m<sup>3</sup> contaminated at the 1000-50 000 ng/g level. Site remediation initially involved building barriers composed of a variety of oil absorbent booms across PCB leachate pathways to prevent further PCB migration. PCB-containing components and liquids from transformers were removed from the main station to the registered PCB storage facility on the site. An environmental assessment of contaminant migration to the marine environment made the assumption that PCB movement was controlled by the barriers and aerial transport of PCBs was insignificant. The assessment concluded that the site does not present unacceptable contaminant exposure conditions to humans, or terrestrial and marine wildlife (Golder, 1997). Remediation work conducted from 1997 to 2001 involved excavation and removal of highly contaminated soils and debris from dump sites on the island (ASU, 2001). This material was stored in sealed containers on-site for future disposal.

In addition to PCBs, some former military sites received significant DDT applications. The bioavailability of this localized DDT contamination to the terrestrial Arctic environment was examined in a study at an abandoned Long Range Aid to Navigation (LORAN) station located at Kittigazuit, Northwest Territories (69°16'56"N, 133°54'32"W) in the western Canadian Arctic (Nirwal, 2001). The study site received applications of DDT between 1948 and 1950. Despite the passage of time, soil concentrations have remained high (maximum  $\Sigma$ DDT = 210 000 ng/g dw), and the composition of  $\Sigma$ DDT compounds in soil still resembled the original pesticide formulation (59% *p,p'*-DDT). In soils, loss and degradation of DDT was less pronounced when compared to temperate and tropical environments. Samples of soil, sediment, willow (*Salix* sp.), grass (*Elymus* sp.), and Arctic ground squirrel (*Spermophilus panyi*) were collected at the LORAN station and at a nearby reindeer herding camp. Concentrations of  $\Sigma$ DDT in soils at the station ranged from 62 to 210 000 ng/g dw. Highest concentrations were found within an area of approximately 4024 m<sup>2</sup>, while a smaller area (386 m<sup>2</sup>) at the herding camp had elevated concentrations as well. Samples of *Elymus* sp. and *Salix* sp. collected from the station had a higher median  $\Sigma$ DDT concentration compared to the area near the herding camp or to background sites. Higher  $\Sigma$ DDT was found in *Salix* sp. (12-10 000 ng/g dw) compared to *Elymus* sp. (3.2-1500 ng/g dw). The concentration and composition of  $\Sigma$ DDT in Arctic



Table 2-2. Examples of sites in Alaska known to be sources of POPs. This does not represent a comprehensive assessment.

Site	POPs are a concern	Contaminated wildlife	Affected subsistence	Reference
Adak Island (Aleutian Islands)	Yes	Likely	Likely	Technical Memorandum, ADEC website <sup>1, 3</sup>
King Salmon Air Station	Yes (solvents)	Likely	Unknown	ADEC website <sup>1, 3</sup>
Pribilof Islands	Fuels, POPs possible	Likely	Likely	ADEC website <sup>1, 3</sup>
St. Lawrence Island	Yes	Potential	Likely	ADEC website <sup>1, 3</sup>
Cape Romanzof Long Range Radar Site	Yes	Yes		ADEC website <sup>1, 3</sup>
Umia (former Air Force Base on Colville River)	Yes	Very Likely	Yes	Army COE website <sup>2, 3</sup>
Operable Unit D, Fort Richardson	Yes	Unknown	Unknown	ADEC website <sup>1, 3</sup>
Dutch Harbor, Unalaska	Yes	Possibly	Possibly	Army COE website <sup>2, 3</sup>
Yakutat Airport	Yes	Unknown	Unknown	Army COE website <sup>2, 3</sup>
Barter Island (Kaktovik)	Transformers (PCBs)	Unknown	Unknown	Army COE website <sup>2, 3</sup>
Wildwood AFS	Dioxins?	Unknown	Unknown	Army COE website <sup>2, 3</sup>

<sup>1</sup> www.state.ak.us/dec/dspar/csites

<sup>2</sup> www.poa.usace.army.mil/fuds

<sup>3</sup> www.akaction.net/pages/mapping

ground squirrel livers were clearly the result of contamination at the study site. Liver concentrations at contaminated areas (maximum  $\Sigma$ DDT = 4300 ng/g lw) declined to background levels (maximum  $\Sigma$ DDT = 4.5 ng/g lw) with increasing distance from contaminated areas. Estimated contaminant exposures were below no-observed effect levels, but a significant relationship between liver size and  $\Sigma$ DDT concentration was found. The concentrations in ground squirrels from background sites in this study were below concentrations reported for Arctic ground squirrels in Alaska (Allen-Gil *et al.*, 1997).

The contribution of atmospheric DDT dispersal and transport at Kittigazuit was negligible because an abrupt transition existed between soil contaminant levels at the sites, and samples collected immediately off-site. For example, the median  $\Sigma$ DDT concentration was 1300 ng/g dw at the LORAN station, and 540 ng/g dw at the reindeer herding camp. In comparison, results for soil samples considered representative of background conditions (3-10 km off site) were below the analytical detection limit of 20 ng/g dw.

A scenario comparable to that at Kittigazuit was also studied in the vicinity of Fort Churchill, Manitoba, on the western coast of Hudson Bay. Three years after aerial spraying for mosquito control, sampling within treated areas showed that the livers of collared lemmings (*Dicrostonyx groenlandicus*) contained 5400-41 000 ng/g lw of  $\Sigma$ DDT and livers of red squirrels (*Tamiasciurus hudsonicus*) contained 7400-17 000 ng/g lw (Brown and Brown, 1970), indicating the persistence and bio-availability of DDT in other terrestrial Arctic environments.

### Alaska

Abandoned sites formally operated by both military and private entities constitute hundreds of known or potential sources of contaminants in Alaska. While most of these sites involve oil-based hydrocarbons (e.g., fuel spills, abandoned wells, etc.), many involve PCBs and OC pesticides as well. Many reports and other informa-

tion on POPs in Arctic and subarctic Alaska exist, but they have not been peer-reviewed and are not generally available. They do serve, however, to describe reliably what contaminants have been detected at these sites of concern. Sources include citations in published manuscripts, reports from a variety of organizations, and websites. Hurwich and Chary (2000) recently outlined and mapped POPs-based research in Alaska and some of these local areas of concern.

Environmental contamination in Alaska is often considered from a general or statewide perspective (i.e., large-scale inputs). Much of the local public awareness of contaminants, however, stems from concern about local sites. Point sources are not typically addressed by researchers studying contaminants, which serves to increase the anxiety of community members. Examples of site-specific concerns in Alaska are the numerous military sites targeted by the *Installation Restoration Program (IRP)* and the *Formerly Used Defense Sites (FUDS)* cleanup programs, which fall under the auspices of the U.S. Department of Defense (Table 2-2).

Table 2-2 is based on a survey of non peer-reviewed documents and websites. Very little effort is made to publish (in scientific journals) the results of studies at these sites and to inform local communities. Examples of documents include *Technical Memorandum: Proposed Biomonitoring Plan for the Nearshore Marine Ecosystems at Adak Island, Alaska*. Adak Naval Complex, Adak, Alaska (Task Order 0204, February 1999) and the *Report of Findings: Cape Romanzof Long Range Radar Site Military Cleanup, USDOJ, Memo to Refuge Manager, Yukon Delta National Wildlife Refuge* (January 17, 1991).

The U.S. EPA Superfund National Priority List sites in Alaska include Adak Naval Air Station, Ketchikan Pulp Company, Fort Richardson (U.S. Army), Elmendorf Air Force Base, Eielson Air Force Base, Fort Wainwright, Standard Steel and Metals Salvage Yard, Alaska Battery Enterprises, Arctic Surplus, and King Salmon (US EPA, 2002).

### 3.1. Toxicokinetics

#### 3.1.1. Distribution

The majority of the substances dealt with in this report are lipophilic, stable, and persistent. They are taken up by aquatic living organisms via diffusion over the gills and from food in the gastrointestinal tract. POPs, particularly organohalogen substances, cross the gill/gut membrane and enter the blood where they are quickly distributed to high lipid tissues such as the liver and adipose tissue. Metabolism and elimination are often slow, leading to a net increase of these substances in the organism over time (bioaccumulation).

There are species differences in the tissue distribution of POPs, partly due to differences in lipid distribution. Several examples of this were given in the first AMAP assessment (de March *et al.*, 1998). Lipid dynamics can also affect the distribution of POPs. For example, many Arctic animals go through dramatic periods of fat accumulation followed by long periods of fasting. Lipophilic POPs will be sequestered in the fat. As the fat is utilized for energy during fasting, POP concentrations in the remaining fat will increase, driving new equilibria to be established between fat, blood lipids, and the lipids of other organs, effectively redistributing POPs to other compartments in the organism. This implies that different tissues in different species will be targets for possible effects from POPs.

For example, lipid dynamics and resultant redistribution of OCS and several PCBs have been studied in wild anadromous Arctic char. Lipid composition was studied in descending (May) fish, ascending (mid-July) fish and in fish caught in mid-July but held in captivity until late September. From May to July, lipid stores increased five-fold with 50% of the lipid content being in the carcass and 35-50% in muscle (Jørgensen *et al.*, 1997a; Jobling *et al.*, 1998). Most of this was triacylglycerols (TAG). Body lipids decreased from mid-July to September in maturing char by 30-40% from all lipid depots, but the major mobilization was of TAG from the carcass and muscle depots. For mature females, the ovaries contained more than 25% of the remaining lipids but for males, the testes only contained 3% of the remaining lipids. Females lost approximately 80% of their body lipids during spawning and overwintering, whereas males only lost 50-55% of body lipids.

The distribution of OCS was studied in lean and fat char. A higher proportion of the body burden was found in extra-adipose organs such as the liver (2 times) and brain (4 times) of lean char than of fat char (Jørgensen *et al.*, 1997b). Similar results were obtained for PCB distribution in a study where wild anadromous char were captured when ascending and treated with PCBs in September (Jørgensen *et al.* 2002a). The fish were not fed during the winter and PCB and lipid analyses were done at three time periods during the winter and spring. From October to May, there was a 20% de-

crease in PCB concentrations in the carcass but brain PCB concentrations increased six-fold and liver concentrations doubled over the same time period. These results show a net redistribution of PCBs from the carcass lipids to the brain and liver during periods of stored lipid utilization.

Significant differences were seen in PCB concentrations with higher concentrations in blubber of molting (lean) harp seals (*Phoca groenlandica*) than in pre-weaning (fat) individuals (Kleivane *et al.*, 2003). When the seals were compared on a total blubber burden basis, however, there were no significant differences (i.e., the same amount of PCB was present in the blubber but was more diluted in the greater blubber mass of the obese seals). In another study on harp seals, POP concentrations were measured in blood and blubber of seals before and after a four-week fast, and in wild seals sampled before the breeding season (fat) and during the molt (lean) (Lydersen *et al.*, 2002). In the fasting experiment, POP concentrations in blubber did not change but blood POP levels increased during the fast. In the wild seals, POP concentrations in both blubber and blood were higher in the lean seals than in the fat seals.

Polar bears show seasonal dynamics in OC concentrations related to fasting (Polischuk, 1999; Norstrom, 2000). The concentrations of  $\Sigma$ CBz,  $\Sigma$ PCB, and  $\Sigma$ chlordanes ( $\Sigma$ CHL) in fat increased and  $\Sigma$ DDT decreased during 47-68 days of fasting and this was entirely due to lipid utilization. For a more thorough presentation of this material, see Section 4.4.7.

#### 3.1.2. Metabolism and elimination

Metabolism of xenobiotics occurs mainly in the liver via a two-phase process. These processes are catalyzed by liver enzymes such as the cytochrome P450 containing monooxygenases (Nebert and Gonzalez, 1987). Lipophilic substances that are resistant to metabolism will be selectively accumulated in living organisms. In addition to detoxification, the enzymatic processes can also create reactive intermediates that may be mutagenic and/or carcinogenic, or metabolites that are stable to further metabolism, that are lipophilic and retained, or that are biologically active, with the ability to bind selectively to proteins and accumulate in the organism.

Many POPs form metabolites that are biologically active. DDT is metabolized in living organisms to DDE, which is lipophilic and toxic, and accumulates in biota (WHO, 1989a). In some cases, a methyl sulfone (MeSO<sub>2</sub>) group is added during metabolism and a number of MeSO<sub>2</sub>-DDE and MeSO<sub>2</sub>-PCB congeners have been identified in animals including several Arctic species (Jensen and Jansson, 1976; Lund *et al.*, 1988; Haraguchi *et al.*, 1990; Bergman *et al.*, 1992b; 1994b; Brandt *et al.*, 1992; Haraguchi *et al.*, 1992; Letcher *et al.*, 1995a; 1995b; 1998; 2000b). MeSO<sub>2</sub>-DDE has a high binding affinity for the adrenal cortex and is highly toxic to this

tissue in mice (Lund *et al.*, 1988; Jönsson *et al.*, 1991; 1992; Brandt *et al.*, 1992; Lindhe *et al.*, 2001). This DDT metabolite is suspected of being one possible cause of adrenal hyperplasia seen in Baltic Sea grey seals (*Halichoerus grypus*) (Jensen and Jansson, 1976; Bergman and Olsson, 1985). MeSO<sub>2</sub>-PCBs may also play a role in hyperadrenocorticism in Baltic Sea seals (Bergman *et al.*, 1992a; Mortensen *et al.*, 1992).

Previously, research has shown that several PCB congeners also form hydroxylated metabolites (Jansson *et al.*, 1975). This type of metabolite has been found to bind selectively to transthyretin (TTR), one of the major transport proteins for thyroid hormones in the blood (Brouwer *et al.*, 1988; 1990; 1998; Bergman *et al.*, 1994a; Letcher *et al.*, 2000a). TTR is complexed to retinol-binding protein (RBP), which transports vitamin A (retinol). Other POPs have also been found to form hydroxylated metabolites, and are suspected of being biologically active in a manner similar to the hydroxylated PCBs. For example, studies have found that some PBDE congeners form hydroxylated metabolites (Klasson Wehler *et al.*, 1996; Örn, 1997; Örn and Klasson Wehler, 1998; Meerts *et al.*, 2000; Klasson Wehler *et al.*, 2001; Mörck *et al.*, 2003; Hakk *et al.*, 2002). 4-Hydroxyheptachlorostyrene has been identified as a metabolite of OCS in polar bear and ringed seal plasma and has been found to bind to human TTR *in vitro* (Sandau *et al.*, 2000). Pentachlorophenol, a metabolite of HCB, also binds to TTR (van den Berg, 1990). Toxaphene congeners Parlar 32 and 62 are metabolized to hydroxylated metabolites by seal liver microsomes (van Hezik *et al.*, 2001).

TBT is metabolized by the cytochrome P450 system in mammals and fish to DBT and MBT, both of which are biologically active (Fish *et al.*, 1976; Kimmel *et al.*, 1977; Lee, 1991; Martin *et al.*, 1989; Fent and Stegeman, 1993).

PAHs are metabolized by the cytochrome P450 system to reactive intermediates that covalently bind to macromolecules such as DNA and proteins, creating adducts.

Chlorinated paraffins (CPs) have been shown to be biotransformed in fish (Fisk *et al.*, 2000), birds (Darnerud and Brandt, 1982), and mammals (Darnerud, 1984), with the susceptibility decreasing with increasing carbon chain length and chlorine content (Tomy *et al.*, 1998). It would appear that there are a number of metabolic pathways that can degrade CPs, but chlorine content and carbon chain length can influence which pathway is utilized. There is little information, however, on the enzymes involved in the degradation. Although older reports suggested that CPs induce phase I (mixed-function-oxygenase enzymes, e.g., CYP450) and phase II enzymes (conjugation reactions, e.g., mercapturic acid synthesis) (Haux *et al.*, 1982), more recent work has failed to find CYP1A induction in fish despite high CP exposures (Fisk *et al.*, 1996).

The major excretion route of POPs and their metabolites is via feces and to some extent, urine. Some of this is passive diffusion over the gut membrane and some from bile excretion of metabolites. In invertebrates and fish, excretion of low log K<sub>ow</sub> compounds may also occur by diffusion through the gill membranes. Female fish and birds excrete lipophilic, organohalogen POPs

via their eggs, and female mammals via placental transfer to the fetus and in breast milk. A particular characteristic of Arctic marine mammals is that most have very high fat contents in breast milk in order to facilitate rapid growth in the young during the short growing season. For example, polar bear milk has a fat content of 20-46% (Derocher *et al.*, 1993; Oehme *et al.*, 1995a; Polischuk *et al.*, 1995; Bernhoft *et al.*, 1997) and various seal species have milk fat contents of 30-60% (Addison and Brodie, 1977; 1987; Bacon *et al.*, 1992; Pomeroy *et al.*, 1996; Beckmen *et al.*, 1999). Therefore, excretion of POPs via milk is more important than placental transfer for adult females of marine mammal species. This in turn enhances POP exposure of young, particularly for polar bears, Arctic foxes (*Alopex lagopus*), whales, and seals.

For example, in Alaska, northern fur seals (*Callorhinus ursinus*), pups have significantly higher blood concentrations of several POPs when compared to their dam's blood and milk (Beckman *et al.*, 1999). The pups of younger dams (primiparous) have much higher concentrations than those of older dams (multiparous). Young harp and hooded seals (*Cystophora cristata*) have as high levels of some POPs as their mothers at the end of the lactation period (Espeland *et al.*, 1997). In Steller sea lions (*Eumetopias jubatus*), 80% of the POP burden may be transferred from a female to her first offspring via lactation (Lee *et al.*, 1996). Young polar bears (1-2 years) have PCB levels similar to adult females with high PCB levels (Bernhoft *et al.*, 1997) and polar bear cubs-of-the-year have higher concentrations of many POPs than their mothers (Polischuk *et al.*, 1995). This is of concern, as young animals may be more sensitive to the effects of POPs than adults.

Polischuk *et al.* (2002) also found clear evidence of sex-specific metabolism in the polar bear. Adult male polar bears continued to lose body burdens of chlordane compounds during a three-month fast, indicating metabolism, while non-lactating females did not lose any of their body burden. On the other hand, lactating females lost PCBs faster than males. This results in higher PCB levels, but lower chlordane levels, in male than female polar bears.

A special case is excretion of TBT, which distributes not only to internal organs but also to bird feathers and seal fur (Tanabe, 1999). The yearly molts of birds and seals may be an important excretion pathway for butyltins. Guruge *et al.* (1996) estimated that up to 25% of the body burden could be excreted in cormorants during a complete molting cycle. Comparable butyltin concentrations are seen in male and female marine mammals, indicating that these are not transferred from mother to fetus/pup to the same degree as other POPs (Tanabe, 1999).

The net result of uptake, distribution, metabolism, and excretion will determine the POP levels found in an organism. This is in turn affected by other factors. Studies carried out to determine the uptake, distribution, metabolism, and excretion of POPs usually investigate one substance at a time. Wildlife and humans, however, are exposed to complex mixtures of POPs.

Very little is known about how different POPs affect each other's toxicokinetics. POPs that induce the hepatic cytochrome P450 system will affect the metabolism of



other xenobiotics, for example. This may lead to an increase in xenobiotic metabolism, thus increasing excretion. An increase in xenobiotic metabolism may also lead to an increase in the formation of reactive intermediates, with increased toxicity and tissue damage (Boon *et al.*, 1992). An example of this is the bioactivation of benzo[a]pyrene (B[a]P) and 7,12-dimethylbenz[a]anthracene (DMBA) in animals after exposure to the CYP1A inducer, CB126. Bioactivated B[a]P and DMBA metabolites were found to bind irreversibly to endothelial cells in certain arteries, veins and capillaries of mice, rats and chicken embryos (Granberg, 2001; Granberg *et al.*, 2000; Annas *et al.*, 1999; 2000).

TBT exposure has been shown to lead to the degradation of cytochrome P450 proteins (CYP1A, CYP2B and CYP3A) and concomitant inhibition of enzyme activities in the liver. This in turn would inhibit the metabolism of organohalogen compounds if an organism was exposed to both substance groups simultaneously, leading to higher accumulation of organohalogens, and possible risk of toxic effects.

Thus, it is very difficult to evaluate the toxicokinetics of environmental exposures to mixtures of POPs. The interactions that have been seen indicate that the relative amounts and the composition of various contaminants in animals may partly be the result of selective effects on the organism's uptake, metabolism, and excretion, and not solely a result of the specific pollution burden of any single contaminant in the area.

### 3.2. Types of effects

In most laboratory experiments studying the toxicological effects of POPs, animals are exposed to single substances or to technical products, often at acutely toxic doses. In a few studies, combinations of a few substances have been used. It should be remembered that wildlife are exposed to complex mixtures of POPs, most often at low doses, and these mixtures may not resemble the original technical products released into the environment because of weathering processes. There are often considerable species differences in sensitivity to specific POPs as well as differences in response. It is, therefore, often difficult to generalize results found in one species to other species. This is particularly difficult when extrapolating effects seen in controlled studies in laboratory species such as rodents to effects seen in marine mammals in the wild. Other factors such as fat dynamics, delayed implantation, differences in physiology, and toxicokinetics may make wildlife more or less sensitive to the effects of POPs. The following is a short, general summary of results from laboratory studies as well as results from field studies where POPs have been quantified and concentrations have been correlated with biological and toxicological effects.

A wide range of effects is seen after exposure to POPs. Some of these types of effects are currently being used as biological markers for POP exposure. These include, among other things, effects on reproduction, development (including the brain), cytochrome P450-dependent enzymes, the immune system, the adrenals, the thyroid gland, thyroid hormone levels, vitamin A levels, formation of DNA adducts, peroxisome proliferation,

and gap junction intercellular communication. Almost all POPs considered in this report also cause visible changes in the liver, including hypertrophy, lesions, and in some cases, tumors.

POPs can cause short-term acute effects when administered in high doses as well as long-term chronic effects at lower doses. In the Arctic, the major concern is long-term chronic exposures as organisms are exposed to POPs over their entire lifetimes. In this context, the major effects of concern are those that may affect reproduction and survival at the individual and population level. Effects at the individual, population or ecosystem level, however, come at a late stage of exposure. It would be more useful to have earlier warning of exposure to POPs and, therefore, biological marker systems based on subtle, low-dose effects are being used or developed. Most biological markers measure effects at the molecular, cellular or organ level; however, it is still not established what these changes might mean at the individual or population level.

#### 3.2.1. Reproduction and development

POPs have a number of effects on the ability of organisms to reproduce and develop normally (for reviews, see Peterson *et al.*, 1993; Bosveld and van den Berg, 1994; Barron *et al.*, 1995; Brouwer *et al.*, 1995). Exposure to some POPs may cause embryo- and fetotoxicity, decreased offspring survival, abnormalities in the estrus cycle and sex hormone levels, reduced sperm production, reduced litter sizes, and even total reproductive failure in mammals. In birds, some POPs cause decreased egg production, retarded egg production, increased embryo mortality, eggshell thinning, embryonic deformities, growth retardation, and reduced egg hatchability, as well as detrimental effects on parental behavior. In fish, some POPs cause decreased egg and larval survival, reduced sexual maturation, and reduced gonad size. Other effects of POPs on organisms may include structural malformations, neurotoxic effects, and neurological and behavioral changes in offspring. Behavioral changes also occur in adult animals, including changes in mating behavior.

Some POPs act as hormones or interfere with endocrine systems and are therefore called endocrine or hormone disruptors (for reviews, see Vos *et al.*, 2000; Damstra *et al.*, 2002). The reproductive effects of embryonic or fetal exposure to these disruptive compounds may only become obvious at later developmental stages or at sexual maturity. The estrogenic and antiestrogenic effects of POPs are the best studied of these effects. Endocrine disruption is also implicated in thyroid and immune system effects, which are treated in Sections 3.2.3 and 3.2.4. POPs may also function as androgens or antiandrogens. Estrogens and androgens are important in the normal sexual differentiation of developing organisms. A number of biomarkers have now been developed for testing the estrogenicity of POPs.

Studies have shown that there is a critical phase in neonatal mouse brain development when the brain is particularly susceptible to effects of low-dose exposure to toxic substances such as PCB, DDT, pyrethroids, organophosphates, paraquat, and nicotine (Eriksson,



1997). This critical phase is known as the 'brain growth spurt' (BGS) and disruption leads to persistent disruption in adult brain function. The BGS occurs at different time points in different mammalian species (Davison and Dobbing, 1968). In rats and mice, it occurs in the first 3-4 weeks of life (neonatal period) whereas in humans, it occurs during the third trimester of pregnancy and throughout the first two years of life. Some POPs, most notably the non-dioxin-like PCBs, may act as neurotoxins by decreasing dopamine content in the brain and altering calcium homeostasis (for a review, see Tilson and Kodavanti, 1998).

### 3.2.2. Cytochrome P450 system and other xenobiotic metabolizing enzyme systems

The most developed of the biological markers is the study of cytochrome P450-dependent liver enzymes (e.g., Förlin *et al.*, 1994; Jensen and Hahn, 2001; Kim and Hahn, 2002). Exposure to OCs and some PAHs induces liver cytochrome P450-dependent enzymes (CYP) known as mixed function oxidases (MFO), which metabolize xenobiotics and endogenous substances (Nebert and Gonzalez, 1987). Exposure to high concentrations of MFO-inducing POPs can affect the metabolism of endogenous substrates, such as steroid hormones, leading to disturbances in critical biological functions (Kupfer and Bulger, 1976). Other methods that can be used to study CYP forms include measuring CYP messenger ribonucleic acid (mRNA), DNA, and protein levels. Protein levels are determined using electrophoresis combined with immunoblotting, which requires the availability of antibodies for each CYP form to be identified.

There are several gene families of cytochrome P450 in vertebrates (Nelson *et al.*, 1993; 1996) and those most relevant for the metabolism of POPs are the CYP 1A, 2B, and 3A gene families. CYP1A forms are induced primarily by planar aromatic hydrocarbons such as PAHs, PCDD/Fs, non-*ortho* and some mono-*ortho* PCBs. Induction of CYP1A is mediated by the aryl hydrocarbon (Ah) receptor, a cytosolic receptor found in all vertebrates studied so far, and for which the most potent ligand is 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). The activated ligand-receptor complex triggers the genetic expression and production of a number of proteins including CYP1A. Induction is often measured as increases in activity of several enzymes including ethoxyresorufin-*O*-deethylase (EROD) and aryl hydrocarbon hydroxylase (AHH) as well as by caffeine demethylation. The efficacy of CYP1A to metabolize POPs that induce these enzymes appears to vary considerably among taxa.

CYP2B forms are induced in mammals by another class of substances, typified by phenobarbital (PB) and measured in laboratory rodents as aminopyrine N-demethylase (APND) activity, aldrin epoxidase (AE), and pentoxyresorufin-*O*-dealkylase (PROD), for example. Considerable caution is needed in interpretation of these activities in wild mammals. For example, PROD activity was shown to be more highly correlated with CYP1A than CYP2B induction in polar bears (Letcher *et al.*, 1996). Birds possess one or more CYP enzymes

that have an activity profile very similar to CYP2B in mammals, especially as judged by the ability to metabolize specific PCB structures (Norstrom, 1988). This enzyme may be in the CYP2 family, but there is no immunochemically recognizable CYP2B in birds. Substances that induce CYP2B are DDT, chlordane, aldrin, endrin, di- to tetra-*ortho* PCBs, and 3-MeSO<sub>2</sub>-PCB. Mono-*ortho* PCB congeners and technical PCBs are mixed type inducers inducing both CYP1A and 2B. There is strong correlative evidence that CYP2B is induced by a combination of PCBs and chlordanes in polar bears (Letcher *et al.*, 1996).

CYP3A forms are among the most versatile iso-enzymes with low substrate specificity. Substances that induce CYP3A include carcinogens, pesticides, some drugs, and steroid hormones such as testosterone. Induction is often measured as the formation of hydroxylated metabolites (e.g., 6- $\beta$  hydroxylation) of testosterone. Recently, CYP3A has been found to be involved in the metabolism of several toxaphene congeners in harbour (*Phoca vitulina*) and grey seals (van Hezik *et al.*, 2001) and implicated in toxaphene metabolism in harp and ringed seals (Wolkers *et al.*, 1998b; 2000). A CYP3A form that is immunologically cross-reactive with anti-rat CYP3A1 was induced in chickens by PB (Ourlin *et al.*, 2000). CYP3A forms may be involved in the metabolism of some POPs in birds.

In the previous AMAP assessment report, de March *et al.* (1998) reviewed species differences in the cytochrome P450 system, particularly with reference to the Arctic situation. At that time, the P450 system had been characterized in a number of Arctic species including harp and hooded seals, polar bears, harbour porpoise (*Phocoena phocoena*), beluga (*Delphinapterus leucas*), short-finned pilot whale (*Globicephala macrorhynchus*), and minke whale (*Balaenoptera acutorostrata*). For terrestrial mammals, they generally have two CYP1A isoforms known as 1A1 and 1A2 as well as functioning CYP2B forms and thus, have a high capacity for metabolizing both groups of POPs.

Harp, grey and hooded seals, and harbour porpoise, like other mammals, seem to have functional CYP1A1 and 1A2 and thus, a higher capacity to metabolize planar compounds, but have weak CYP2B activity and thus, a reduced ability to metabolize xenobiotics that are substrates for these particular MFOs (Boon *et al.*, 1992; Goksøyr *et al.*, 1992; Goksøyr, 1995a). Seals seem to have more CYP2B activity than whales (Goksøyr, 1995a). Polar bears have functional CYP1A1, 2B1, 3A1, and epoxide hydrolase (Bandiera *et al.*, 1995), and have a high metabolic capacity, particularly for PCB and DDT (Norstrom and Muir, 1994).

Previous studies have shown that fish seem to lack CYP2B and have only one version of CYP1A (Nebert *et al.*, 1989; Stegeman, 1989; Stegeman and Hahn, 1994; Goksøyr, 1995b), and thus have a low POP metabolizing capacity. However, one exception to this has been found recently. Deepwater sculpin (*Myoxocephalus thompsoni*) from the Great Lakes, have been found to metabolize PCBs and form 3- and 4-MeSO<sub>2</sub>-PCB metabolites, indicating that they have CYP2B-like activity (Stapleton *et al.*, 2001). With regard to birds, cytochromes P450 1A1 and 1A2 are found in this group of animals (Livingstone and Stegeman, 1989).

Since 1996, new studies have further characterized the cytochrome P450 system in Arctic species. CYP1A is present in Arctic char and is active even at low temperatures (Wolkers *et al.*, 1996; 1998c; Jørgensen and Wolkers, 1999). Recent studies in glaucous gull (*Larus hyperboreus*) have shown some EROD activity implying the presence of CYP1A (Henriksen *et al.*, 1998a; 2000). Ringed seals have been found to have CYP1A and 3A activities, and possibly CYP2B activity (Mattson *et al.*, 1998; Wolkers *et al.*, 1998a; 1998b; Hyyti *et al.*, 2001; Nyman *et al.*, 2001). Besides CYP1A, both grey and ringed seals may have active CYP1B1 (Nyman *et al.*, 2000), which has previously only been found in human, rat and mouse tissues (Nelson *et al.*, 1996). CYP1B1 metabolizes PAHs, often initiating carcinogenesis (Shimada *et al.*, 1996; Baron *et al.*, 1998; Kim *et al.*, 1998a). Harp seals have been found to have active CYP3A (Wolkers *et al.*, 1999; 2000). Steller sea lions have been found to have CYP1A1 and 1A2, whereas minke whales only have CYP1A1 (Teramitsu *et al.*, 2000). Beluga and pilot whales (*Globicephala melas*) both have CYP1A1 activity and show the presence of CYP2B, though it is not clear if this is active (White *et al.*, 2000).

Organisms lacking functional CYP1A, 2B or 3A will not be able to eliminate the POPs metabolized by these enzymes, leading to their bioaccumulation. This is particularly the case for fish, making them amplifiers of many POPs in food webs. The presence of functional cytochrome P450 enzymes means that POPs may be metabolized and eliminated; metabolized to lipophilic and toxic metabolites; metabolized to hydrophilic metabolites that bind to proteins and are retained; and/or, that POP exposure may lead to cytochrome P450 enzyme induction, increasing the amounts of metabolic enzymes present. Those organisms that cannot eliminate POPs may accumulate parent compound concentrations high enough to cause effects. Those organisms that metabolize POPs to lipophilic and toxic metabolites, and/or hydrophilic and toxic metabolites that are retained, may instead accumulate these metabolites to high enough concentrations to cause effects.

### 3.2.3. Immunological effects

Many POPs disrupt both humoral and cell-mediated immune responses of the specific (acquired) arm of the immune system, as well as causing effects on the non-specific (innate) arm. As a result, the resistance to infectious agents may be reduced. Humoral-mediated immunity involves the body's ability to recognize foreign substances (helper T-cells) and mount a response by stimulating the production of antibodies (B-cells). Cell-mediated immunity is involved in delayed hypersensitivity reactions (e.g., skin reactions to allergens) and the production of cytotoxic T-cells against tumors and viruses. Natural killer cells are involved in the non-specific response (i.e., absence of memory) and are a first line of defense against virus-infected cells and tumors. Most POPs cause multiple effects on the immune system.

Some OCs (PCDD/Fs, some PCBs) as well as TBT have direct effects on the thymus, causing atrophy of this lymphoid organ responsible for the maturation of T-cells. The most insidious effect of POPs on the immune system is to decrease an organism's resistance to infec-

tion or cancer. Immunosuppressive effects of POPs have often been studied as: reduced antibody production when exposed to a foreign antigen (e.g., suppression of the anti-sheep red blood cell plaque-forming response); decreased delayed-type hypersensitivity; decreased natural killer cell activity; and, decreased resistance to pathogens (e.g., viral infections) (Vos and Luster, 1989; Tryphonas, 1994; Wong *et al.*, 1992). All of these have been used as indicators of immunosuppressive effects in laboratory and wild animals.

Immunosuppressive effects may be one of the most sensitive and environmentally relevant effects of POPs (Vos and Luster, 1989). For example, immunosuppression has been measured in harbour seals fed Baltic fish in semi-field experiments and was found to correlate with levels of PCDD/F and planar PCBs expressed as toxic equivalents (TEQs) (de Swart *et al.*, 1995; Ross *et al.*, 1995; 1996a). Immunosuppression is also suspected to be the cause of an increasing prevalence of moderate to severe intestinal ulcers in Baltic grey seals (Bergman, 1999). Indications of immunosuppression have also been found in some Arctic species including polar bears, northern fur seals, and glaucous gulls (Chapter 6).

### 3.2.4. Thyroid and retinol effects

Thyroid hormones control metabolism and growth, and are essential for normal reproduction. They are also important for the development of normal brain functions during fetal development (Morse *et al.*, 1993; Morse, 1995). The thyroid gland produces predominantly T4 (thyroxine), which is transported in plasma to target tissues by the transport protein TTR. Once delivered, T4 is deiodinated by T4-monodeiodinase to triiodothyronine (T3), which is the active hormone. Some POP effects on the thyroid may be related to the ability of some POP phenolic metabolites, such as hydroxylated PCBs, hydroxylated PBDEs, pentachlorophenol and 4-hydroxy-heptachlorostyrene, to attach to the binding sites on the transthyretin-retinol-binding protein complex (TTR-RBP) in plasma, thereby disrupting the normal transport of thyroid hormones T3 and T4 as well as vitamin A (retinol) to their target tissues (Rolland, 2000; Simms and Ross, 2000). POPs may also interfere with the enzymes that control thyroid hormone metabolism such as uridine diphosphate glucuronosyl transferase (UDPGT), which is involved in glucuronidation and subsequent excretion of T4 (Brouwer *et al.*, 1998). Recently, TTR has been identified in polar bear plasma (Sandau *et al.*, 2000) and in harbour seals (Simms and Ross, 2000).

The structural requirements for binding of a POP metabolite to TTR are hydroxy-substitution in *para*- or *meta*-positions of one or both phenyl rings with adjacent halogen unsubstituted sites (Lans, 1995). X-ray diffraction studies have shown that several phenolic organohalogen compounds bind with the hydroxy group in the central channel of the TTR molecule (Ghosh *et al.*, 2000). Disruption of normal thyroid hormone transport leads to lowered plasma levels of T3 and T4, which in turn may initiate an increased release of thyroid stimulating hormone (TSH) to stimulate the thyroid gland to secrete more T3 and T4. This disruption of the feedback system for thyroid hormones may lead to thyroid

hyperplasia (goiter), hypertrophy, hypothyroidism, disruptions of metabolism and possibly a tendency to develop thyroid tumors. Another effect may also be related to the ability of some POPs, such as PCBs, to induce the production of liver enzymes involved in the breakdown of thyroid hormones. This results in reduced amounts of thyroid hormones circulating in the plasma.

Imbalance in vitamin A (retinol and its esters) status can cause immunosuppression, susceptibility to cancers, skin lesions, as well as disruption of reproduction, growth, and development. Some POPs (particularly PCDDs and PCBs) affect vitamin A metabolism and transport. Biomarkers used for thyroid and retinol effects include measuring plasma levels of free and bound T3 and T4, TSH as well as vitamin A levels.

### 3.2.5. Mutagenic and carcinogenic effects

Current research supports a two-stage cancer model characterized by a primary mutagenic event (initiation) followed by a long latency period and second event (promotion) that leads to tumor growth. Peroxisomes are cellular organelles mainly found in the liver and kidney. They contain peroxisomal enzymes and are essential for lipid metabolism, cellular respiration and gluconeogenesis among other things (for a review, see Youssef and Badr, 1998). A number of POPs, including perfluorinated compounds such as PFOS and PFOA, are potent peroxisome proliferators. Increased oxidative stress by peroxisome proliferation is thought to be one possible mechanism for tumor promotion (Cattley and Preston, 1995). Another mechanism may be down-regulation of gap junctional intercellular communication (GJIC), which has been linked to tumor-promoting properties of many carcinogens (Trosko and Ruch, 1998). Gap junctions are protein channels that allow for the transport of substances between cells. This communication is necessary for normal growth and function and, if inhibited, may lead to tumor promotion. Various PCBs, DDT, dieldrin, toxaphene and brominated biphenyls have been shown to inhibit GJIC in human breast epithelial cells (Kang *et al.*, 1996) and methyl sulfone PCBs seem particularly potent (Kato *et al.*, 1998). GJIC is measured using a bioassay where a monolayer of cells is scraped after exposure to a contaminant, then exposed to a dye and dye migration into the cells is quantified (Up- ham *et al.*, 1997).

High doses of planar aromatic hydrocarbons induce oxidative stress possibly through induction of CYP enzymes (Toborek *et al.*, 1995; Park *et al.*, 1996; Hennig *et al.*, 1999; Schlezinger *et al.*, 1999; Slezak *et al.*, 1999; Slim *et al.*, 1999). This is manifested by increased production of reactive oxygen species, lipid peroxidation and DNA damage.

Several PAHs, such as B[a]P and DMBA, are well known mutagens and both initiate and promote tumors. Most of the other POPs dealt with in this assessment are not mutagenic, but many are strong tumor promoters. Several POPs are associated with increased tumor prevalence found in highly exposed wildlife from areas outside of the Arctic, including fish (Myers *et al.*, 1998) and beluga from the St. Lawrence Estuary (De Guise *et al.*, 1994; Martineau *et al.*, 1985).

Methods for measuring mutagenicity include the measurement of DNA adducts since there is a positive correlation between a chemical's carcinogenic potency and the extent that it binds to DNA (Kriek *et al.*, 1998). Correlations have also been shown between the level of exposure to PAHs and the amount of adducts formed (French *et al.*, 1996; Shugart and Theodorakis, 1998; Wirgin and Waldman, 1998). Other standard methods include the formation of micronuclei, sister chromatid exchange, chromosome aberration assays in peripheral blood lymphocytes, as well as the Ames test for single nucleotide mutations.

### 3.2.6. Effects of mixtures

In the previous AMAP assessment report, the additive, antagonistic and synergistic effects of mixtures were discussed (de March *et al.*, 1998). Some recent studies indicate that certain POPs singly have little or no endocrine disrupting effects but when tested as mixtures, endocrine disruption occurs, indicating synergism. For example, chlordane, dieldrin and toxaphene singly showed no ability to compete with estradiol when tested against alligator and human estrogen receptors (Arnold *et al.*, 1997; Vonier *et al.*, 1996). However, when tested as a mixture, the combination inhibited estradiol binding by 20-40%. A mixture of 15 POPs mimicking the composition in ringed seal blubber was found to disturb *in vitro* porcine oocyte maturation and development at concentrations comparable to highly exposed humans or mammals in the Arctic (Campagna *et al.*, 2001).

### 3.3. Effects of specific POPs

Because the previous AMAP assessment report contained thorough reviews of the toxicology of legacy OCs and their metabolites (PCDDs, PCDFs, PCBs, MeSO<sub>2</sub>-PCBs, aldrin, dieldrin, chlordanes, DDT, MeSO<sub>2</sub>-DDE, HCB, HCHs, mirex, toxaphene), only those compounds where substantial new information has become available since 1996 are covered in this report. Emphasis has been placed on updates of these few compounds, plus the toxicology of new compounds that have been found in the Arctic. The following descriptions of the toxicology of different POPs are short reviews and not meant to be comprehensive. They mainly cover chronic effects and effects that are relevant to the Arctic discussion. Results for controlled studies in laboratory animal species are discussed first. Where done, controlled studies using wild animal species under laboratory conditions are then presented. If field studies outside of the Arctic have been performed, these are then presented. Finally, studies correlating specific effects with contaminant concentrations found from field studies of wild species, in areas with known high burdens of contaminants such as the Baltic Sea and the Great Lakes, are discussed. This last type of study has inherent problems, as it is never possible to state that the contaminant measured is the cause of the effect, since there may be other contaminants not measured that co-vary with the one measured. Field studies and correlation studies of Arctic species are discussed under Section 6. An overview of toxic effects of the POPs discussed in this report is given in Table 3.1.

Table 3-1. Overview of toxic properties of various POPs. ▼ = suppression or decrease, ▲ = induction or increase.

	<i>Reproductive/developmental effects</i>	<i>Neurotoxic effects</i>	<i>Cytochrome P450 effects</i>	<i>Immune effects</i>	<i>Thyroid/retinol effects</i>	<i>Cancer</i>	<i>Other</i>
Aldrin and dieldrin	▼ Reproduction		Induces cytochrome P450 2B	Suppresses immune system		Non-mutagenic. Increased liver tumors	
Chlordanes	▼ Reproduction		Induces cytochrome P450 2B	Suppresses immune system		Non-mutagenic tumor promoter	
DDT and metabolites	Egg-shell thinning in bird eggs. ▼ Reproduction		Induces cytochrome P450 2B	Suppresses immune system	▼ Thyroid weight		Adrenal cortex hyperplasia
HCBz	Fetotoxic. Teratogenic. ▼ Reproduction		Induces cytochrome P450 1A and 2B	Suppresses immune system	▼ T3 and T4 ▲ TSH ▲ Thyroid weight	Non-mutagenic tumor promoter	▲ Porphyria
α-HCH	No information		Induces cytochrome P450 2B			Non-mutagenic tumor promoter	
β-HCH	Estrogenic		Induces cytochrome P450 2B	Suppresses immune system	▲ Thyroid weight	Non-mutagenic tumor promoter	
γ-HCH (lindane)	Estrogenic and antiestrogenic. ▼ Reproduction		Induces cytochrome P450 1A and 2B			Non-mutagenic tumor promoter	
Mirex	▼ Reproduction		Induces cytochrome P450 2B	Suppresses immune system		Non-mutagenic. Induces tumors	
Toxaphenes	Fetotoxic. ▼ Reproduction		Induces cytochrome P450 1A, 2B and 3A	Suppresses immune system	▲ Thyroid-weight ▲ TSH	Mutagenic, potent carcinogen. Inhibits GJIC	▲ Bone brittleness in fish. Adrenal hypertrophy
Endosulfan	Fetotoxic. ▼ Reproduction		Induces cytochrome P450 1A and 2B	Suppresses immune system		Non-mutagenic	
PCDD/Fs and nPCBs and metabolites	Fetotoxic. Deformities. ▼ Reproduction	Permanent changes in learning, behavior, memory (nPCB)	Induces cytochrome P450 1A	Thymic atrophy. Suppresses immune system	▼ T3 and T4 ▼ Vitamin A	Non-mutagenic tumor promoters. Affects GJIC	▲ Porphyria
Other PCBs	Fetotoxic. Deformities. ▼ Reproduction	Permanent changes in learning, behavior, memory. Decreased dopamine	Induces cytochrome P450 2B	Suppresses immune system	▼ T3 and T4 ▼ Vitamin A	Non-mutagenic tumor promoters. Affects GJIC	▲ Porphyria Hyperadrenocortism
SCCPs	Fetotoxic. Deformities. ▼ Reproduction	▼ Motor performance	Induces cytochrome P450 1A	No information	▼ T4 ▲ TSH	Non-mutagenic. ▲ Peroxisome proliferation. Inhibits GJIC	
PCNs	Embryotoxic. ▼ Reproduction		Induces cytochrome P450 1A				
Octachlorostyrene and metabolites			Induces cytochrome P450 1A and 2B		Binds to TTR <i>in vitro</i>		
PBDEs	Estrogenic and antiestrogenic	Permanent changes in learning, behavior, memory	Induces cytochrome P450 1A and 2B	Suppresses immune system	▼ T4 ▼ Vitamin A	Non-mutagenic	
PFOS/PFOA	▼ Reproduction					Non-mutagenic, tumor promoter ▲ Peroxisome proliferation. Inhibits GJIC	
TBT and metabolites	Imposex in invertebrates. Deformities. ▼ Reproduction		Inhibits liver cytochrome P450 1A, 2B, and 3A	Suppresses immune system		May be carcinogenic	



### 3.3.1. Halogenated industrial chemicals and by-products

#### 3.3.1.1. Update on PCDDs, PCDFs, and PCBs, including PCB metabolites

The 2,3,7,8-PCDD/Fs, as well as PCBs substituted in the 3,3',4,4'-positions with no (non-*ortho* PCB (nPCB)) or one *ortho* chlorine (mono-*ortho* PCB) are among the most toxic POPs. The most toxic and best studied of these planar compounds is TCDD. These substances exert their toxic effects via a common mechanism that requires binding to the Ah receptor. They produce essentially the same spectrum of toxic effects in treated animals as TCDD, differing only in their potencies. Except in some bird species, all are less potent than TCDD. The non-*ortho* PCBs are more toxic than the mono-*ortho* congeners. A thorough review of the effects of PCDDs, PCDFs, and PCBs was presented in the previous AMAP assessment report (de March *et al.*, 1998). The following is an update on relevant effects studies published since 1996.

#### *Reproductive and developmental effects*

Competitive-binding studies using estrogen receptors from humans, green anole (*Anolis carolinensis*), and rainbow trout (*Salmo gairdnerii*) showed that chlorobiphenyls (CBs) 104, 184, and 188 were significant competitors in all three systems and thus have estrogen-like activity (Matthews and Zacharewski, 2000). In the rainbow trout assay, CBs 41, 51, 91, 115, 143, and 173 were also found to compete to some extent. None of these congeners are, however, major components in commercial PCB mixtures, and some are not present at all. The findings are therefore of small relevance to the environment. Hydroxy-PCBs that are found in blood have been shown to be weakly anti-estrogenic in the MCF-7 human breast cancer cell line and in HeLa cells (Kramer *et al.*, 1997; Moore *et al.*, 1997). Andersson *et al.* (1999) found that four hydroxy-PCBs significantly induced vitellogenin synthesis in rainbow trout hepatocytes and induced proliferation of MCF-7 cells, both indicative of the capabilities of these substances to bind to the estrogen receptor and elicit a response. Also shown, was the induced proliferation of MCF-7 cells by CBs 104 and 188.

Experiments have been performed exposing neonatal mice to a single oral dose of specific CB congeners at a critical time point during the brain growth spurt and studying a range of neurobehavioral effects (Eriksson *et al.*, 1991; Eriksson and Fredriksson, 1996a; 1996b; 1998). Permanent changes in spontaneous behavior were seen after neonatal exposure to *ortho*-substituted PCBs (2,4,4'-TrCB (CB28) at 14  $\mu\text{mol}$  (3.6 mg)/kg body weight; 2,2',5,5'-TeCB (CB52) at 14  $\mu\text{mol}$  (4.1 mg)/kg body weight; 2,2',4,4',5,5'-HxCB (CB153) at 14  $\mu\text{mol}$  (5.1 mg)/kg body weight); non-*ortho* PCBs (3,3',4,4'-TeCB (CB77) at 14  $\mu\text{mol}$  (4.1 mg)/kg body weight; 3,3',4,4',5-PeCB (CB126) at 0.14  $\mu\text{mol}$  (0.046 mg)/kg body weight; and, 3,3',4,4',5,5'-HxCB (CB169) at 1.4  $\mu\text{mol}$  (0.51 mg)/kg body weight). The effects worsened with age. As well, learning and memory in adult mice were affected by CB52 and CB153 at the same doses, CB126 at 1.4  $\mu\text{mol}$  (0.46 mg)/kg body weight, and CB169 at 14  $\mu\text{mol}$  (5.1 mg)/kg body weight.

In previous studies reviewed in the AMAP assessment report, Holene *et al.* (1995; 1998) showed signifi-

cant behavioral alterations in rat offspring after pre- and post-natal exposure (including lactation exposure in males) to several specific CB congeners (CBs 118, 126, 153). In a recent study (Holene *et al.*, 1999), female rats were exposed to CB153 through mother's milk. The females showed a significant sex-specific behavioral response, being less sensitive than males studied previously, since only deficient acquisition of time discrimination was seen.

Repeated exposure to Aroclors caused decreases in brain dopamine and the bioaccumulation of several *ortho*-substituted CBs in the brain of rats and non-human primates (Seegal *et al.*, 1986; 1991a; 1991b; Shain *et al.*, 1986; 1991; Seegal and Schantz, 1994). Kodavanti *et al.* (1995; 1996) have since been able to show that the *ortho*-substituted, non-dioxin-like PCB congeners are associated with neurotoxicity. The *ortho*-PCBs were found to disrupt intracellular signal transduction in cells from the cerebellum. Mariussen *et al.* (1999) tested 14 PCBs *in vitro* for their ability to competitively inhibit dopamine uptake into synaptic vesicles and found that only *ortho*-PCB congeners were active. EC<sub>50</sub>s (the concentration affecting 50% of the animals) ranged from 2.3 to 5.6  $\mu\text{g/g}$  (7-30  $\mu\text{M}$ ) for single congeners. In non-human primates, brain concentrations of 2 to 5  $\mu\text{g/g}$  of  $\Sigma\text{PCB}$  were associated with decreased dopamine concentrations (Seegal *et al.*, 1990). In a study of 20 PCB congeners, inhibition of dopamine uptake in synaptic vesicles was found to be most potent for CBs 41, 91, 112, and 143 (all *ortho*-substituted) and no inhibition was seen with the non-*ortho* CBs tested (Andersson, 2000).

Aroclor 1254 has been found to affect bone development in rats (Andrews, 1989) and recently, CB126 (3,3',4,4',5-PeCB) has been shown to cause extensive alterations in the long bones of rats, including decreases in density and strength (Lind *et al.*, 1999; Lind, 2000). TCDD also affects bone structure in rats in a similar manner (Jämsä *et al.*, 2001) and has also been found to impair their molar tooth development (Kattainen *et al.*, 2001; Lukinmaa *et al.*, 2001). A synthetic mixture of 16 MeSO<sub>2</sub>-PCBs, prepared in the same relative concentrations as found in the blubber of Baltic grey seals, was fed to female mink (*Mustela vison*) for one year and reproductive outcome was studied (Lund *et al.*, 1999). Compared to controls, the treated mink had significantly increased litter size, but lower birth weights and reduced kit survival. *In vitro* studies of liver showed increased breakdown of progesterone in the treated mink. The muscle concentrations found in the exposed females and their kits were 18 000 ng/g lw and 21 000 ng/g lw, indicating considerable carry-over from dams to kits.

In mink, continuous exposure to 250 ng PCB/g food caused delayed onset of estrus and lowered whelping rates (Restum *et al.*, 1998). PCB exposure was from a diet made using Saginaw Bay carp as the PCB source. Exposure to 500 ng PCB/g food led to increased litter mortality and reduced kit body weights. Short-term parental exposure reduced kit survival of subsequent generations of mink conceived months after the parents were placed on PCB-free feed.

Kestrels (*Falco sparverius*) were exposed to PCBs *in ovo* via parents that were fed a diet containing a mixture of technical PCBs (7 mg/kg body weight/day) through

breeding and hatching (Ferne *et al.*, 2001). This diet led to an exposure of 34 µg PCB/g whole egg ww. These second-generation kestrels were paired with unexposed, experienced kestrels and compared with non-exposed controls. *In ovo* PCB exposure was found to suppress egg laying in 25% of exposed females, delay clutch initiation and lead to smaller clutch sizes for both exposed sexes. Exposed kestrels also had reduced fledgling success and higher incidence of complete brood mortality. Greater effects were seen in PCB-exposed females than males.

A mixture of 20 PCB congeners administered to zebrafish (*Brachydanio rerio*) via food caused reproductive disturbances (Örn *et al.*, 1998). Egg production and offspring survival time were reduced. CBs 60, 104, and a hydroxy-PCB were highly embryotoxic in zebrafish oocytes exposed via maternal transfer (Westerlund *et al.*, 2000). A significant negative correlation was found between ΣPCB levels above 20 ng/g ww (as Aroclor 1260 in liver) and baculum (penis bone) length in wild juvenile mink (Harding *et al.*, 1999).

Aroclor 1242 caused significant sex reversal in reared sliders (*Trachemys scripta elegans*), a species of turtle, overriding male-producing temperature levels to result in female hatchlings, indicating that this PCB mixture has estrogenic effects (Willingham and Crews, 1999).

Later egg laying, prolonged incubation, and smaller eggs and chicks were correlated with higher yolk sac PCDD/F concentrations in common tern chicks (*Sterna hirundo*) (Murk *et al.*, 1996). Toxic effects in colonial fish-eating birds in the Great Lakes area of North America were correlated with the concentrations of PCDDs/PCDFs and non-ortho PCBs found in the different bird species studied (reviewed in Gilbertson *et al.*, 1991; Giesy *et al.*, 1994a; 1994b). The effects in birds include reduced egg hatching, embryotoxicity, deformities such as crossed bills and clump feet, and impaired parental behavior (Hoffman *et al.*, 1987; Kubiak *et al.*, 1989; Tillitt *et al.*, 1989; 1991; 1992; 1993; Yamashita *et al.*, 1993).

#### Cytochrome P450-dependent monooxygenases

Rats and mice exposed to either Aroclor 1254 or CB105 had induced cytochrome P450 activity measured as EROD, methoxy-O-resorufin deethylase (MROD) and PROD activity, with rats being more sensitive than mice (Hallgren *et al.*, 2001).

In the study of 16 MeSO<sub>2</sub>-PCBs fed to mink, discussed previously, an 11-fold induction of PROD activity was seen in the exposed adult females and a five-fold increase was seen in their five-week-old kits (Lund *et al.*, 1999). This is in accordance with previous research showing high CYP2B induction for MeSO<sub>2</sub>-PCBs in rats (Kato *et al.*, 1995a; 1995b; 1997; 1999).

Mink fed a diet with varying PCB doses made from clean fish or contaminated carp from Saginaw Bay, Lake Huron, had dose-dependent induction of cytochrome P450 activity (Shipp *et al.*, 1998). Young mink were more sensitive than older mink.

Adult male mallard ducks (*Anas platyrhynchos*) dosed orally with Aroclor 1254 had induced cytochrome P450 activity as measured by elevated EROD and PROD activity (Fowles *et al.*, 1997).

In an experimental study, Arctic char were divided into four groups, PCB-exposed and fed, PCB-exposed

and starved, non-exposed and fed, and non-exposed and starved (Jørgensen *et al.*, 1999). The PCB-exposed fish were fed a single oral dose of 1 µg Aroclor 1260/g body weight. Increased EROD activity was found in the PCB-exposed and starved group, which also had the highest PCB concentrations and exhibited fin erosion. The threshold for EROD effects in Arctic char was found to be 1 µg PCB/g ww in liver.

Induction of hepatic CYP1A1 activity was found to be correlated with PCDD/F concentrations in common tern chicks (Murk *et al.*, 1996). EROD induction was found to correlate with ΣPCB and DDT levels in a comparison of ringed and grey seals from the Baltic Sea and from reference sites (Nyman, 2000).

#### Immunosuppression

Beluga leukocytes and splenocytes were exposed to several POPs *in vitro*, and CB138 was found to significantly reduce splenocyte proliferative responses (De Guise *et al.*, 1998). No effects were seen with CBs 153, 180, 169 or TCDD.

Previously, a diet of Baltic Sea herring (*Clupea harengus*) fed to captive harbour seals was found to cause immunosuppression, which was associated with the content of PCDD/PCDF and dioxin-like PCBs (de Swart *et al.*, 1994; 1995; Ross *et al.*, 1995; 1996a). In a laboratory study, using rats as surrogates for harbour seals, pregnant rats were administered extracts from the same diets of Atlantic or Baltic Sea herring as were given to harbour seals, or a positive control of Atlantic herring oil spiked with TCDD, on a daily basis for 41 days (Ross *et al.*, 1997). Immune function was assessed in the offspring and found to be most suppressed in the TCDD-spiked group (reduced thymus weight, thymocyte and splenocyte proliferative response, natural killer cell and specific antibody responses). Similar types of immunosuppression were seen in the Baltic herring group but were less pronounced. The daily intakes for the pregnant rats were 0.3 pg TEQ/g body weight for the cleaner Atlantic herring oil, 2.1 pg TEQ/g body weight for the Baltic Sea herring oil and 134 pg TEQ/g body weight for the spiked oil.

Two juvenile harp seals were exposed experimentally to increasing doses of selected PCB congeners (0.4, 2.0, 4.0, 20.0, and 40.0 mg/day, one week per dose), while two more seals acted as controls (Lohman *et al.*, 2002a). The *in vitro* release of tumor necrosis factor alpha (TNF-α) from isolated monocytes stimulated with *Escherichia coli* (*E. coli*) lipopolysaccharides and β-1,3-glucan were assayed during the treatment period and during a subsequent 30-day fasting period. Changes in the adrenal gland were also studied (Lohman *et al.*, 2002b). No differences were seen between the two groups until after 30 days of fasting. Release of TNF-α was elevated in both groups, with higher levels in the controls than in the PCB-treated seals, indicating possible decrease in monocyte reaction to lipopolysaccharides. Elevated basal levels of serum cortisol and aldosterone were seen in the PCB-exposed seals, compared to the control seals. After 30 days of fasting, both groups had increased levels of cortisol and aldosterone compared to the treatment period, with levels in the PCB-treated seals exceeding those in the controls. Although this study is limited because of small sample sizes, the results may indicate

that short-term exposure to PCB may induce hyperadrenocorticism without noticeable morphological changes in the adrenal gland of seals.

No immunosuppression was seen in rats fed diets containing beluga blubber from the St. Lawrence Estuary (highly PCB contaminated), the Arctic or combinations of the two (Lapierre *et al.*, 1999). Mice fed diets based on varying amounts of beluga blubber from the St. Lawrence Estuary and the Arctic, providing different PCB exposures, showed some indications of immunosuppression but no difference was seen between the treatment groups (Fournier *et al.*, 2000).

Chicken embryos exposed to CB126 *in ovo* had decreased thymus mass, decreased live T-cell numbers in the thymus, declines in some types of thymocytes, and decreased viable B-cell numbers in the bursa of Fabricius (Grasman and Whitacre, 2001).

Glaucous gull chicks with high and low dietary PCB exposure were immunized with antigen. Antibody production, lymphocyte response, mitogen-induced lymphocyte proliferation and hematology were measured after 56 days of age (Larsen *et al.*, 2002d). A significantly higher lymphocyte response to phytohemagglutinin (PHA) in the high PCB group indicated general immune system stimulation. Significantly lower antibody titers in response to influenza virus immunization in the high PCB group indicated impaired ability to produce antibodies and may be associated with decreased resistance to infections.

In the study on Arctic char by Jørgensen *et al.* (1999), previously described in the section on P450-dependent monooxygenases, increased plasma cortisol levels were seen in the PCB-exposed and fed group. In a similar study, Jørgensen *et al.* (2002b; 2002c) exposed Arctic char to varying, single doses of Aroclor 1254 and held them either with or without food for five months before subjecting them to a ten-minute handling disturbance. Starved fish were given doses of 0, 1, 10 or 100 µg/g body weight and fed fish were given doses of 0 or 100 µg/g body weight. Starved control fish had elevated plasma cortisol levels compared with fed fish before handling. These basal cortisol levels were suppressed by PCBs in starved fish and were elevated in fed fish. The cortisol response to handling was suppressed by PCBs in a dose-dependent way in starved fish. Plasma glucose levels were affected in the same manner as cortisol levels. The findings indicate that stress responses in Arctic char are compromised by PCBs and the effect of fasting makes char sensitive to the effects of PCBs.

The char were also subjected to tests of disease resistance (Maule *et al.*, 2002). Mortality after exposure to a disease agent was dose-related (0, 1, 10, and 100 µg/g body weight) among the starved fish, with a significant trend toward higher disease susceptibility with increasing PCB dose. No differences in mortality were seen in the two fed groups (0 and 100 µg/g body weight). However, total mortality was higher in the fed groups compared to the starved groups. The results indicate that PCB reduces immunocompetence in starved Arctic char in a dose-dependent manner, but that lean fish are also more disease-resistant than fed fish. Similarly, juvenile chinook salmon (*Oncorhynchus tshawytscha*) exposed to Aroclor 1254 had higher mortality than controls when exposed to a bacterial pathogen (Arkoosh *et al.*, 2001).

Herring gull (*Larus argentatus*) and Caspian tern (*Sterna caspia*) chicks from five Great Lakes sites were assessed for immune function using the PHA skin test, a sensitive indicator of T-cell-mediated immunity in birds. Suppression of T-cell-mediated immunity was found to correlate most strongly to higher ΣPCB concentrations (Grasman *et al.*, 1996; Luebke *et al.*, 1997; Grasman and Fox, 2001).

Significant correlations have been found between high ΣPCB concentrations, suppressed T-cell function and increased antibody titers after immunization in Caspian terns from Lake Huron (Grasman and Fox, 2001). Significant negative correlations were found between yolk sac levels of PCDD/F, ΣPCB, non-ortho PCBs and TEQs, and plasma corticosterone levels in herring gull embryos from the Great Lakes (Lorenzen *et al.*, 1999). The activities of phosphoenolpyruvate carboxylase and malic enzyme, two metabolic enzymes regulated in part by corticosteroids, were also negatively correlated to yolk sac PCDD/F concentrations.

A significant association was found between ΣPCB concentrations and mortality due to infectious disease in harbour porpoises from England and Wales indicating a possible causal relationship between PCB exposure and immunosuppression (Jepson *et al.*, 1999).

#### *Thyroid and retinol effects*

Until 1996, only a few hydroxy-PCB metabolites had been identified and studied. Since then, up to 30 hydroxy-PCBs have been detected in plasma of various species and 13 such metabolites have been identified (Letcher *et al.*, 2000a). Most of the identified hydroxy-PCB metabolites have the hydroxy-group attached to one of the two *para*-positions with chlorine atoms attached *ortho* to the hydroxy-group. This structural configuration mimics that of T4, the thyroid hormone, except that T4 has iodine atoms instead of chlorines. The affinities of several hydroxy-PCBs for TTR, the plasma protein that transports T4, are up to ten times greater than for the natural hormone (Brouwer *et al.*, 1998; Lans *et al.*, 1993; 1994). 4-OH-3,3',4',5-TeCB (CB77 metabolite) and 4-OH-CB107 reduced total plasma T4 levels in pregnant mice and in the fetuses (Sinjari *et al.*, 1998a) but the effect was less when CB77 was administered and allowed to metabolize *in vivo* (Darnerud *et al.*, 1996). 4-OH-2',3,3',4',5-PeCB (CB105 metabolite) was also found to reduce T4 levels but no effect was seen with 4-OH-2,3,3',4',5-PeCB (CB105 metabolite) (Sinjari and Darnerud, 1998). Hydroxy-PCBs also influence T4 metabolism by inhibiting sulfation, a major regulation pathway in the fetus. Inhibition of sulfation has been shown to occur *in vitro* (Schoor *et al.*, 1996; 1998; 1999). If this occurs *in vivo*, it has implications for fetal brain development (Brouwer *et al.*, 1998). Aroclor 1254 or CB105 treatment significantly reduced free and total plasma T4 levels and hepatic vitamin A in rats and mice but had no effect on levels of TSH (Hallgren *et al.*, 2001).

In the study of 16 MeSO<sub>2</sub>-PCBs fed to mink, discussed previously, plasma concentrations of total T3 and T4 were reduced in the exposed dams (Lund *et al.*, 1999). The authors speculate that the MeSO<sub>2</sub>-PCBs decrease the total T4 concentration via enzyme induction of UDPGT, which is involved in glucuronidation of T4.



Total and free plasma T4 concentrations increased with increasing TEQ levels in mink exposed via a diet of carp from Saginaw Bay, Lake Huron, Michigan, but total and free T3 decreased, indicating a reduction in T4-monodeiodinase activity (Heaton *et al.*, 1995). In a similar study, T4 concentrations were found to be significantly higher in mink fed a fish diet containing PCB than in those not exposed to PCB (Nieminen *et al.*, 2000). Rats fed a diet containing Baltic Sea herring that had previously been used in a semi-field study of immunosuppression in harbour seal (Ross *et al.*, 1995), were found to have lower plasma T4 levels compared to the control group (Ross *et al.*, 1996b).

Chick embryos exposed *in ovo* to Aroclor 1242 or 1254 had reduced plasma T4 concentrations and reduced hepatic monodeiodinase activity (Gould *et al.*, 1999). A correlation was also seen between femur length and plasma concentrations of T3 and T4, indicating a decrease in skeletal growth due to reduced thyroid hormone levels. Adult male mallards dosed with Aroclor 1254 had significantly increased thyroid weights and decreased plasma total T3 concentrations (Fowles *et al.*, 1997). Adult great blue herons (*Ardea herodias*) dosed with 2,3,7,8-TCDD had a significant increase in plasma T4 levels, but no effects were seen on plasma total T3 or the T3:T4 ratio (Janz and Bellward, 1997). White leghorn hens were fed a diet with different PCB levels derived from carp from Saginaw Bay, Lake Huron (Zile *et al.*, 1997). The high PCB intake group had decreased molar ratios of retinol to retinyl palmitate in eggs.

CB77 injected intraperitoneally into brook trout (*Salvelinus fontinalis*) caused decreased plasma retinol in males (Ndayibagira *et al.*, 1994) and exposure with CB77 in rainbow trout caused increased hydroxylation of retinoic acid due to induction of CYP1A enzymes (Gilbert *et al.*, 1995). Oral dosing of lake trout (*Salvelinus namaycush*) with CB126 caused decreased liver retinoids (Palace and Brown, 1994).

In free-living European otter (*Lutra lutra*), a strong negative correlation was found between hepatic vitamin A levels and TEQs calculated from non- and mono-*ortho* PCBs (Murk *et al.*, 1998), and animals with the higher concentrations had a higher incidence of infectious diseases. Immature northern elephant seals (*Miro-unga angustirostris*) with northern elephant seal skin disease had depressed total T3 and T4 levels, depressed retinol levels, and higher concentrations of PCB (and *p,p'*-DDE) compared to unaffected controls (Beckmen *et al.*, 1997). The ratio of T3 to T4 was significantly correlated to the concentration of CB169 in grey seals from the U.K. (Hall *et al.*, 1998). Plasma thyroid hormone concentrations and PCBs were measured in a group of grey seal pups from the Norwegian west coast (Froan, Trondheimfjord) and compared to a more highly exposed group in the Baltic Sea (Sørmo *et al.*, 2002). No relationship was found for T4, but T3 levels were lower in the more contaminated Baltic pups as compared to the Norwegian pups. Disruption of vitamin A and/or thyroid hormones related to high PCB concentrations has also been observed in other captive and free-ranging seal species (Brouwer *et al.*, 1989; de Swart *et al.*, 1994; Rolland, 2000; Simms *et al.*, 2000; Simms and Ross, 2000).

Decreased yolk sac retinoids and plasma thyroid levels, and increased ratios between plasma retinol levels

and yolk sac retinyl palmitate were significantly correlated to higher yolk sac PCDD/F concentrations in common tern hatchlings from Belgium and the Netherlands (Murk *et al.*, 1996). Lower plasma retinol levels in herring gull and Caspian tern chicks from several Great Lakes colonies were associated with high  $\Sigma$ PCB concentrations in eggs from the same colony (Grasman *et al.*, 1996).

Higher PCB concentrations were found to be associated with increased metabolism of retinoic acid by cytochrome P450 enzymes, decreases in hepatic retinoid stores and an increase in developmental deformities in lake sturgeon (*Acipenser fulvescens*) from the St. Lawrence River compared to a reference site (Doyon *et al.*, 1999).

### Cancer

The coplanar PCBs, CB77 and 169, as well as TCDD, inhibit GJIC in mouse Hepa1c1c7 cells (de Haan *et al.*, 1994). Of 20 tested PCB congeners, 14 were found to inhibit GJIC in rat liver white blood cell culture (Anderson, 2000). Most potent were CBs 51, 143, 173, and 184 (tri- and tetra-*ortho* congeners). Other active congeners included CBs 41, 60, 91, 104, 112, 115, 153, 188, 190, and 193. Inactive congeners were CBs 58, 68, 78, 99, 126, and 169.

Glaucous gull chicks from Svalbard were fed a clean (Arctic cod, hen eggs) or a contaminated (Arctic cod, gull eggs) diet (Krøkje *et al.*, 2002). Chromosome aberrations and DNA adducts were measured and higher frequencies of aberrations were seen in both males and females of the exposed group. DNA adduct levels were also higher in males of the exposed group.

### 3.3.1.2. SCCPs/C<sub>10</sub>-C<sub>13</sub> polychlorinated *n*-alkanes

For reviews of the toxicology of SCCPs, see Environment Canada (1993), Willis *et al.* (1994), WHO (1996) and Tomy *et al.* (1998). There are difficulties in assessing the toxicity of SCCPs as most data have been generated using technical SCCP products, which can contain thousands of chemical compounds. This makes it difficult to study the toxicity of individual components, and effects may be due to stabilizers and impurities in the products. As well, degradation, bioaccumulation, and metabolism change the relative amounts of SCCP components found in organisms in the environment, making it difficult to assess exposure. Recently, studies examining the effects of SCCPs on fish have concluded that they have low acute toxicity and a narcotic mode-of-action (Fisk *et al.*, 1999b; Cooley *et al.*, 2001), although histopathological lesions were observed in the livers of exposed rainbow trout (Cooley *et al.*, 2001). In general, SCCPs appear to be much less toxic than other persistent organic pollutants.

### Reproductive and developmental effects

Pregnant rats were dosed orally with 0, 100, 500, and 2000 mg/kg body weight/day of a SCCP (58% Cl) on days 6 through 19 of gestation to study teratogenic effects (WHO, 1996). The high-dose group of dams had 32% mortality and decreased body weight, and there were increased incidences of post-implantation loss, fetal malformations, and decreases in viable fetuses. The



no-observed-effect-level (NOEL) for teratogenic effects was set at 500 mg/kg body weight/day. In pregnant rabbits exposed orally to 0, 10, 30, and 100 mg/kg body weight/day on gestation days 6 through 27, no effects were seen on dams or fetuses, and the no-observed-adverse-effect level (NOAEL) was set at 100 mg/kg body weight/day.

Mallard ducks were exposed to dietary concentrations of 0, 28, 166, and 1000 mg/kg of a SCCP (58% CI) in a one-generation test (cited in Willis *et al.*, 1994). No effects were seen on the adults but some eggshell thinning was seen although the authors questioned the biological significance of this. Hatchlings were fed the same diets for 14 days and those in the high-dose group showed 10% mortality.

#### Neurological effects

Mice exposed intravenously to single doses of 30-300 mg/kg body weight of two SCCPs (49% and 70% CI) were studied for effects on motor performance and thermoregulation (Eriksson and Kihlström, 1985). Dose-dependent decreases in motor performance and thermoregulation were seen with increasing doses of both SCCPs. Mice exposed to 300 mg/kg body weight of the lower chlorinated SCCP showed significantly decreased motor performance 15 minutes after injection. As well, significantly decreased rectal temperature was seen in mice injected with 300 mg/kg body weight of both SCCPs after 60 minutes.

#### Cytochrome P450-dependent monooxygenases

Cytochrome P450 concentrations increased in rats exposed intraperitoneally to 1000 mg/kg body weight/day of two short-chain SCCPs (49% and 71% CI) for four days (Nilsen and Toftgård, 1981). The increase was greater for the more highly chlorinated SCCP. Cytochrome P450 induction potential was studied in rats using five different SCCP formulations dosed orally at 1000 mg/kg body weight/day for four days (Nilsen *et al.*, 1981). Only the higher chlorinated short-chain formulations (59% and 71% CI) led to increased P450 concentrations and EROD activity. A lower chlorinated short-chain (49% CI), a medium-chain (50% CI), and a long-chain formulation (49% CI) had no effect.

High single oral doses (1000 mg/kg body weight) of C<sub>10</sub>-C<sub>13</sub> SCCP (49% CI) caused an increase in benzo[a]pyrene hydroxylase activity in female flounder held in brackish water (Haux *et al.*, 1982). No effects were seen in females kept in seawater, males kept in brackish or seawater, or any fish exposed to a more highly chlorinated product (70% CI). Rainbow trout exposed to two C<sub>12</sub> SCCPs (56% and 69% CI) showed no induction of P450 CYP1A as measured by EROD activity (Fisk *et al.*, 1996).

#### Immunosuppression

There is no information on the immunotoxicity of SCCPs.

#### Thyroid effects

UDPGT is produced in the liver and decreases plasma T4 levels, stimulating TSH release by the pituitary gland. Rats exposed to 1000 mg/kg body weight of two SCCPs (56% and 58% CI) showed two-fold increases in

plasma TSH levels, a 30-40% decrease in total and free T4, a two-fold increase of UDPGT activity but no effect on T3 levels (Wyatt *et al.*, 1993). Elcombe *et al.* (1994) observed a similar increase in UDPGT activity and TSH level and a decrease in T4 levels in male and female rats exposed to a SCCP (58% CI) at similar doses to the above study. They also noted thyroid follicular cell hypertrophy.

#### Cancer

A C<sub>10</sub>-C<sub>23</sub> SCCP (70% CI) was not found to be mutagenic in the Ames test using three different strains of *Salmonella typhimurium* (Meijer *et al.*, 1981). A SCCP (60% CI) was found to increase hepatocellular neoplasms in both sexes of mice and rats; kidney tubular cell adenomas; adenocarcinomas and mononuclear cell leukemia in male rats; and, thyroid follicular cell neoplasms in female rats and mice (Bucher *et al.*, 1987). The rats were given repeated oral doses of 312 and 625 mg/kg body weight/day and the mice, 125 and 250 mg/kg body weight/day and their responses were followed for two years.

Peroxisomal proliferation, as measured by significantly increased peroxisomal fatty acid oxidation, was seen in rats and mice exposed to two SCCPs (56% and 58% CI) (Wyatt *et al.*, 1993). Peroxisome proliferation was confirmed using microscopic methods in rats exposed to the same two SCCPs (Elcombe *et al.*, 1994).

SCCPs (50% and 60% CI) have been found to be potent inhibitors of GJIC in rat liver epithelial cells, indicating that they may be tumor promoters (Kato and Kenne, 1996).

#### 3.3.1.3. PCNs

PCNs are planar molecules, like PCDDs, PCDFs, and non-*ortho* PCBs, and also seem to exert their effects via the Ah receptor. Acute and chronic exposure to PCNs leads to effects similar to those seen for PCDDs, PCDFs and non-*ortho* PCBs (for reviews, see Kover, 1975; Brinkman and Reymer, 1976; Crookes and Howe, 1993; Jakobsson and Asplund, 2000). The most toxic congeners are the PeCNs and HxCNs. Based on *in vitro* studies, several PCN congeners have been assigned TCDD toxic equivalency factors (TEFs) (Hanberg *et al.*, 1990; 1991; Blankenship *et al.*, 2000; Villeneuve *et al.*, 2000).

#### Reproductive and developmental effects

A commercial PCN product (Halowax 1014) as well as a mixture of 1,2,3,5,6,7- and 1,2,3,4,6,7-hexachloronaphthalenes are both toxic to chick embryos (Engwall *et al.*, 1993; 1994). Male and female chickens fed different doses of Halowax 1014 have been mated and egg production studied. At higher doses (20 mg/kg), egg hatchability was reduced and no eggs were produced in chickens fed the highest dose of 100 mg/kg (Pudelkiewicz *et al.*, 1959).

In another study, Halowax 1014, 1013 or 1051 were nano-injected into fertilized medaka (*Oryzias latipes*) eggs at various dose levels (0.3-30 ng/egg) and the embryos allowed to develop to adulthood and sexual maturity (Villalobos *et al.*, 2000). Early life stage and early adult life stage assessments were carried out. Halowax

1014 was found to be more toxic than Halowax 1013 and 1051. The 16-day LD<sub>50</sub> (dose that kills 50% of the exposed animals) for Halowax 1014 in embryos was 4.2 ng/egg and death was caused by cardiovascular abnormalities. The lowest-observed-adverse-effect level (LOAEL) was 3.0 ng/egg with hemorrhage and yolk sac edema as the major effects. Halowax 1014 decreased the gonadosomatic index in adult females. Halowax 1013 caused high mortality at 10 ng/egg and premature hatching of embryos at all doses. Halowax 1051 was the least toxic PCN.

#### *Cytochrome P450-dependent monooxygenases*

Three-spined sticklebacks (*Gasterosteus aculeatus*) fed the commercial PCN mixture Halowax 1014 have shown a dose-related increase in EROD activity as well as lipid accumulation in the liver (Holm *et al.*, 1993). Rainbow trout fry show dose-related increases in EROD activity after microinjection of Halowax 1014 at the embryo stage (Norrgrén *et al.*, 1993). A commercial mixture of tetra-, penta- and hexachlorinated PCN (Halowax 1014) as well as a mixture of 1,2,3,5,6,7- and 1,2,3,4,6,7-HxCNs both caused liver enzyme induction in chick and eider duck embryos (Engwall *et al.*, 1993; 1994). A HpCN congener had much lower EROD induction potency.

Three *in vitro* bioassays were used to test the ability of 18 PCN congeners to induce CYP1A activity (Vileneuve *et al.*, 2000). The PLHC-1 fish hepatoma cell bioassay was fairly insensitive to PCNs but the EROD and luciferase assays using recombinant H-4-II E rat hepatoma cells were more sensitive. The HxCN congeners tested were most potent, followed by the PeCNs. The TeCNs, TrCNs, DiCNs, and MoCNs tested were less active.

Rainbow trout sac fry were treated with Halowax 1014, a mixture of 1,2,3,4,6,7- and 1,2,3,5,6,7-HxCN, or 1,2,3,4,5,6,7-HpCN injected into the yolk sac (Pesonen *et al.*, 2000). After two weeks, immunohistochemical analysis was performed for CYP1A expression and was most pronounced in the hepatocytes. Exposure of a primary cell culture of trout hepatocytes to these PCNs led to increased EROD activity and CYP1A mRNA content, with the HxCN mix being most potent followed by HpCN and then Halowax 1014.

#### 3.3.1.4. OCS

Long-term dietary exposure of rats to OCS causes elevated serum cholesterol and histological changes in the thyroid, liver, and kidney of rats (Chu *et al.*, 1986a).

#### *Reproductive and developmental effects*

OCS was found to have binding affinity for both the androgen and estrogen receptor in an *in vitro* assay (Satoh *et al.*, 2001).

#### *Cytochrome P450-dependent monooxygenases*

OCS significantly induced CYP1A activity in mice (Smith *et al.*, 1994). Long-term dietary exposure to high doses of OCS induced the CYP2B enzymes aniline hydroxylase and aminopyrine N-demethylase (APND) in male and female rats, with males being more sensitive (Chu *et al.*, 1986a).

#### *Thyroid and retinol effects*

Sandau *et al.* (2000) found that 4-hydroxy-heptachlorostyrene (4-OH-HpCS), a metabolite of OCS, had a similar affinity for human TTR as T<sub>4</sub>, the native hormone. The potential of 4-OH-HpCS to bind to TTR makes it capable of disrupting thyroid hormone transport and potentially affecting circulating retinol concentrations.

#### 3.3.1.5. Update on PBDEs

PBDEs are numbered according to the same system as PCBs, based on chlorination degree and placement of the chlorines on the two aromatic rings. Long-term exposure to DeBDE has been found to induce thyroid hyperplasia, hepatocellular and thyroid adenomas, and carcinomas in mice (Great Lakes Chemical Corporation, undated; 1987).

#### *Reproductive and developmental effects*

The estrogenic potency of several BDEs was tested using the estrogen receptor (ER)-CALUX bioassay, and the most potent congeners in descending order were BDEs 100, 75, 51, 30, and 119 (Brouwer *et al.*, 2001; Meerts *et al.*, 2001). Potency was much less than for estrogen. BDEs 166 and 190 were antiestrogenic.

Neonatal exposure to 2,2',4,4'-TeBDE (BDE47) (10.5 µg/g body weight), 2,2',4,4',5-PeBDE (BDE99) (0.8 or 12 µg/g body weight) or 2,2',4,4',5,5'-HxBDE (BDE153) (0.9 or 9 µg/g body weight), administered orally to mice on day 10, induced permanent aberrations in spontaneous motor behavior which worsened with age (Eriksson *et al.*, 2001; Viberg *et al.*, 2001a; 2002). Neonatal exposure to BDE99 (12 µg/g body weight) or BDE153 (0.9 or 9 µg/g body weight) also affected learning and memory functions in the adult animal. BDE209 (2.22 or 20.1 µg/g body weight) administered orally to neonatal mice on day 3 induced permanent aberrations in spontaneous motor behavior, but when administered on day 10, had no effect (Viberg *et al.*, 2001b). BDE99 exposure (0.6, 6 and 30 µg/g body weight/day) during pregnancy to post-natal day 21 in mice caused increased hyperactivity in offspring (Branchi *et al.*, 2002).

In a follow-up study to Eriksson *et al.* (2001), Eriksson *et al.* (2002) investigated whether there is a critical time in neonatal mouse brain development for induction of the neurotoxic effects of BDE99. One single oral dose of 8 µg/g body weight (14 µmol/kg body weight) was administered to 3-day, 10-day and 19-day-old mice. The mice exposed to BDE99 on day 10 showed significant behavior aberrations, as was previously seen in Eriksson *et al.* (2001), and mice exposed on day 3 showed similar aberrations but to a lesser degree. The mice exposed on day 19 showed no significant change from the controls.

Uptake and retention of BDE99 in the brain was also studied by administering <sup>14</sup>C-labelled BDE99 to 3-day, 10-day and 19-day-old mice (Eriksson *et al.*, 2002). The retention of BDE99 in mice exposed on day 3 indicates that the effects observed may be due to the amount of BDE99 still present in the brain on day 10. The neurotoxic effects seem to involve changes in the cholinergic system as mice given BDE99 on day 10 and then challenged as adults with a low dose of nicotine behaved

completely the opposite of controls. From these studies, it was concluded that the window for permanent effects of BDE99 and BDE47 is day 10 in neonatal mice (Eriksson *et al.*, 2001; 2002).

In Sinjari *et al.* (1998b), female rats given BDE47 orally for a period of two weeks were then killed and the choroid plexus of the brain removed, homogenized and incubated with  $^{125}\text{I}$ -T4. Compared to controls, there was a dose-dependent reduction in the binding of  $^{125}\text{I}$ -T4 to the choroid plexus. In contrast, *in vitro* incubation of rat choroid plexus with BDE47 revealed no competitive inhibition of labeled T4 binding. This indicates that BDE47 metabolites can cross the blood-brain barrier and bind to the choroid plexus T4-binding sites. This in turn could cause interference of T4 transport to the brain, with risks for effects on neural development.

BDE99 at 10  $\mu\text{M}$  induced death of 23% of cerebellar neurons in an *in vitro* neurotoxicity test (Llansola *et al.*, 2001). Higher concentrations induced more neuronal death.

Microinjection of BDE47, 2,2',3,4,4'-PeBDE (BDE 85) or BDE99 into newly fertilized rainbow trout eggs in an early life stage mortality bioassay showed no effects compared to TCDD (Hornung *et al.*, 1996). Exposure to low levels of BDE47 affected developmental rates in the invertebrate *Acartia tonsa*, and juveniles were more sensitive than adults (Breitholtz *et al.*, 2001). This may implicate disruption of juvenile hormones or ecdysteroids.

#### *Cytochrome P450-dependent monooxygenases*

Using a recombinant H-4-II E rat hepatoma cell line having Ah receptor mediated expression of a luciferase reporter gene (the dioxin receptor (DR)-CALUX assay) (Aarts *et al.*, 1995; Murk *et al.*, 1998), a number of individual PBDE congeners have been tested for their potency to activate/deactivate the Ah receptor (Meerts *et al.*, 1998). In order to study antagonism, the same PBDE congeners were also tested in the presence of TCDD. Of the 17 PBDE congeners tested, seven (BDEs 32, 85, 99, 119, 153, 166, 190) showed ability to activate the Ah receptor. Potencies could only be determined for BDEs 166 and BDE190 and are in the same range as mono-*ortho* PCB congeners 105 and 118 (Sanderson *et al.*, 1996). Some congeners such as BDEs 85, 99, and 119 showed both agonist and antagonist activities depending on the concentration tested. Nine congeners, including BDEs 15, 28, 47, 77, and 138, showed antagonist activities against TCDD. The observed antagonism may be due to competition between PBDEs and TCDD at the Ah receptor level. In a recent study, more BDE congeners have been tested in the DR-CALUX assay and BDEs 30, 47, 51, 71, 75, and 100 have also been found to activate the Ah receptor, but their potencies could not be determined (Brouwer *et al.*, 2001).

EROD induction was studied in chick and rat hepatocytes, liver cell lines from rainbow trout, rats and humans, and in a human intestinal cell line (Chen *et al.*, 2001). BDEs 77, 100, 119 and 126 induced the greatest EROD activity in all cell types, but were less potent than TCDD. BDEs 153 and 183 were weaker inducers. BDEs 47 and 99 were not inducers in any cell line. The congeners that did not induce EROD also failed to bind to the Ah receptor.

Studies in whole cultured chick embryo liver showed induction of EROD activity after exposure to BDEs 47, 99, and 153 as well as the commercial mixture Bromkal 70-5 DE (Pettersson *et al.*, 2001). BDE99 was most potent but was much less potent than TCDD (TEF of 0.000004).

Microsomal enzyme activities were studied in rats and mice, and results from Bromkal 70-5 DE, and BDE 47 showed induction of EROD, MROD, and PROD in both species (Hallgren and Darnerud, 2002; Hallgren *et al.*, 2001). Rats exposed to Bromkal 70-5 DE orally for 28 days had dose-dependent increases in EROD and PROD activities (Fattore *et al.*, 2001).

Rainbow trout dosed orally with BDE47 or BD99 for 6 and 22 days had significantly inhibited EROD activity in the liver with BDE47 being most powerful in this effect (Tjärnlund *et al.*, 1998).

#### *Immunosuppression*

Mitogen-induced DNA synthesis and immunoglobulin synthesis by human lymphocytes *in vitro* were examined after exposure to purified BDEs 47 and 85. No effects on mitogen-induced proliferation or immunoglobulin synthesis were observed (Fernelöf *et al.*, 1997). The results indicate that proliferation and immunoglobulin synthesis are insensitive to the direct action of PBDEs.

Immunotoxicity was studied after oral treatment with Bromkal 70-5 DE or BDE47 in rats and mice (Darnerud and Thuvander, 1998). In mice, BDE47 caused reduced splenocyte number as reflected in decreased numbers of CD45R+, CD4+, and CD8+ cells in spleens. In mice treated with Bromkal 70-5 DE, absolute numbers of double negative thymocytes were significantly lower than in controls, and mice also showed reduced production of IgG. No effects were seen in rats. Thus, BDE47 and Bromkal 70-5 DE, which contains BDE47, both seem to be immunotoxic in mice.

#### *Thyroid and retinol effects*

Seventeen PBDE congeners (BDEs 15, 28, 30, 32, 47, 51, 71, 75, 77, 85, 99, 100, 119, 138, 153, 166, and 190) were incubated individually with rat hepatic microsomes from rats treated with  $\beta$ -naphthoflavone, phenobarbital or clofibrate (Meerts *et al.*, 2000). The parent congeners and the metabolites formed were then tested for their ability to compete with T4 for binding to human TTR *in vitro*. Results showed no competition with the parent compounds, but considerable potency for several of the metabolites, indicating the metabolism of PBDE to hydroxylated PBDE. The results indicate that hydroxylated metabolites of PBDE may be potent competitors of T4 and could disrupt normal thyroid hormone function in wildlife and humans if present. No binding competition was seen for several of the higher brominated PBDEs such as BDEs 138, 153, 166 and 190 after incubation with the microsomes. This may indicate that these congeners are not readily metabolized.

The Bromkal 70-5DE product causes decreased thymus weight, increased liver/body weight ratios in mice and decreased T4 in rats and mice (Fowles *et al.*, 1994; Darnerud and Sinjari, 1996; Hallgren *et al.*, 2001). Decreases in T4 were also seen when rats and mice were treated with the single congener BDE47, but no effects on TSH were seen for BDE47 or Bromkal 70 (Darnerud



and Sinjari, 1996; Hallgren *et al.*, 2001). Bromkal 70-5 DE and BDE47 also caused significant reductions in hepatic vitamin A concentrations in rats, and Bromkal 70-5 DE caused reductions of hepatic vitamin A concentrations in mice at high doses (Hallgren *et al.*, 2001). Similarly, Bromkal 70-5 DE caused dose-dependent reduction in hepatic vitamin A in rats dosed orally for 28 days (Fattore *et al.*, 2001).

In subsequent studies, the interactive effects of different organohalogen compounds (PCB, PBDE, and chlorinated paraffins (CP)) on T4 levels and microsomal enzyme activities were tested (Hallgren and Darnerud, 2002). Female rats were orally exposed to single compounds or combinations at isomolar concentrations daily over 14 days. The results show that PCBs (Aroclor 1254) and PBDEs (BDE47) significantly reduce the T4 levels in rats, in the actual exposure interval (6-18 mg/kg body weight/day), with Aroclor 1254 resulting in the strongest effect. EROD and MROD, but to a lesser extent PROD and UDPGT, activities were negatively correlated to T4 effects. Regarding the mixed BDE47 + CP group, a synergistic decrease in free T4 levels and increase in EROD activity was observed.

#### Cancer

BDE47 was shown to induce a statistically significant increase in intragenic recombination when studied in one of two tested *in vitro* assays using mammalian cells (Helleday *et al.*, 1999). This may indicate that BDE47 can induce cancer via a non-mutagenic mechanism.

#### 3.3.1.6. PFOS and PFOA

PFOS is a surfactant with both lipophobic and hydrophobic properties. Therefore, it does not accumulate in lipids, but instead accumulates in the liver, gall bladder and the blood (where it binds to proteins). It is speculated that the body treats PFOS as a bile acid. The liver makes bile acids from cholesterol, which are excreted from the gall bladder into the intestine to facilitate the emulsification and uptake of fats in the gut. The bile acids are then recycled back into the liver via enterohepatic circulation. PFOS may also weaken cell membranes (Hu *et al.*, 2000). PFOA is used as a lubricant, detergent, and wetting agent (Guethner and Vietor, 1962).

Liver enlargement and reduced serum cholesterol levels are early responses to exposure to PFOS. Acute toxicity with 100% mortality was seen in rhesus monkeys fed 10 mg/kg/day for three weeks in one experiment and in rhesus monkeys fed 4.5 mg/kg/day for seven weeks in another (Seed, 2000). Even doses of 0.75 mg/kg/day led to changes in cynomolgus monkey (*Macaca fascicularis*) livers, reductions in blood cholesterol, disinterest in food, and death. PFOA also causes liver enlargement and increased liver lipid levels in rats and mice (Kawashima *et al.*, 1995; Kudo and Kawashima, 1997; Kudo *et al.*, 1999).

#### Reproductive and developmental effects

In a two-generation reproductive toxicity study of PFOS in rats, pup-survival in the first generation was significantly decreased in the two highest dose groups receiving 1.6 and 3.2 mg/kg/day (Seed, 2000). All first-generation pups in the high-dose group died within one day of

birth, and close to one-third of first generation pups in the 1.6 mg/kg/day group died within four days of birth. Only pups in the 0, 0.1 and 0.4 mg/kg/day groups were carried to the second generation. For second-generation offspring, reductions in pup weight and reversible delays in reflex and physical development were seen in the high-dose groups. For the second-generation offspring, the NOAEL for reduced pup weight was determined to be 0.1 mg/kg/day and the LOAEL, 0.4 mg/kg/day. These doses corresponded to PFOS liver concentrations of 15 µg/g and 58 µg/g ww, respectively, in the rats.

In rabbits, PFOS caused maternal toxicity (decreased body weight gain) at a dose of 1.0 mg/kg/day or higher (Case *et al.*, 2001). Levels causing maternal toxicity also led to increased abortions and reduced fetal weights.

#### Cancer

PFOA and PFOS are not mutagenic but are known to be liver tumor promoters in rats. Perfluorinated fatty acids such as PFOA and PFOS increase peroxisome levels and inhibit GJIC (Upham *et al.*, 1998). PFOS is almost as potent as PFOA in causing increased peroxisome proliferation (Sohlenius *et al.*, 1993). Several other perfluorinated compounds (perfluorooctanoic sulfonamide, perfluorohexane sulfonate) also affect GJIC (Hu *et al.*, 2001).

### 3.3.2. Persistent organic pesticides

#### 3.3.2.1. Update on toxaphene

A review of the effects of technical toxaphene was given in the previous AMAP assessment report. The toxicity data available at that time were rather limited. The following is an update on relevant effects studies that have been published recently. For a recent review on toxaphene, including toxicology, see de Geus *et al.* (1999).

#### Reproductive and developmental effects

Technical toxaphene, T2 (Parlar 26), and T12 (Parlar 50) were tested for their estrogenicity using the MCF-7 human breast cancer cell model and were found to have weak estrogenic activity (Stelzer and Chan, 1999). T2 and T12 had lower proliferative effects on the cells than technical toxaphene, and T2 was more potent than T12. Effects of mixtures of the three indicated additive effects, and none of the compounds had effects on estrogen receptor or progesterone receptor levels. In another study using MCF-7 cells, technical toxaphene, T2, and T12 were tested for their effects on estrogen receptor function (Jørgensen *et al.*, 1997c). The results indicated that toxaphene and T12 are antiestrogens, and that the effects occur at the gene transcription level. In support of this, toxaphene tested in a battery of assays for estrogenic activity (MCF-7 cells, competitive receptor binding) was found to be weakly antiestrogenic (Arcaro *et al.*, 2000). In a battery of estrogenic screening methods, including mouse uterus, MCF-7 human breast cancer cells, and yeast-based reporter gene assays, toxaphene was found to exert minimal estrogenic effects (Ramamoorthy *et al.*, 1997).

Technical toxaphene and congeners T2 and T12 were tested for their effects on cultured rat embryos during the period corresponding to a critical period of morphogenesis and organogenesis (gestational days 10-12).



Technical toxaphene and both single congeners caused significant changes in total morphology, somite number, head and crown-rump length, and central nervous system scores of the embryos, including a high incidence of central nervous system defects (Calciu *et al.*, 1997). The altered total morphology, including decreases in head and crown-rump lengths indicate that toxaphene and the two single congeners retard growth and morphological development. There were differences in potency and type of toxicity, indicating that specific congeners can produce effects not predicted by the technical mixture.

Technical toxaphene fed to female zebrafish for two weeks at doses of 20, 230, and 2200 ng/g body weight/day was found to have no effects on total number of eggs spawned, percentage of fertilized eggs, percentage of embryo mortality or percentage of hatching (Fåhræus-Van Ree and Payne, 1997). Toxaphene, however, did cause a dose-related decrease in the percentage of oviposition.

The ability of toxaphene to displace native ligands from the estradiol receptor, testosterone receptor and cortisol receptor was tested using rainbow trout liver and brain tissues (Knudsen and Pottinger, 1999). Toxaphene did not bind to any of the receptors and was concluded not to be estrogenic.

In red-eared slider turtle, incubation temperature determines the sex of hatchlings, but male-producing temperatures can be overridden if the eggs are exposed to estrogenic compounds. Toxaphene was tested for its estrogenic activity by application on eggs set to become males (Willingham and Crews, 1999). No estrogenic effects were seen for toxaphene. Similarly, toxaphene showed no estrogenic effects when tested for its ability to displace native estradiol from alligator or human estrogen receptors (Vonier *et al.*, 1996; Arnold *et al.*, 1997). However, Palmer *et al.* (1998) found that water exposure to toxaphene induced significant vitellogenin production in the male African clawed frog (*Xenopus laevis*), which is an estrogenic effect.

#### *Cytochrome P450-dependent monooxygenases*

Subacute levels of toxaphene given to guinea pigs led to induced cytochrome P450 and increased aniline hydroxylase in the liver and kidney (Chandra and Durairaj, 1993). In mice, toxaphene exposure led to increases in CYP2B levels (Hedli *et al.*, 1998). Rats and Japanese quail exposed to single doses of technical toxaphene ranging from 0.012 to 40 mg/kg body weight showed induced P450 systems only at the highest dose (Drenth *et al.*, 2000). These included increased PROD, formation of 15 $\beta$ -hydroxytestosterone and 2-hydroxyestradiol in the rat, and increased formation of 6 $\beta$ -, 15 $\alpha$ - and 16 $\beta$ -hydroxytestosterone in the quail. The doses required to induce the P450 system were close to those known to cause mortality, so it was concluded that P450 activity induction is probably unlikely in wildlife exposed to toxaphene.

In cynomolgous monkeys, toxaphene treatment induced aminopyrene, MROD, and EROD activities, indicating that toxaphene is a mixed-type inducer that induces both CYP1A and 2B (Bryce *et al.*, 2001). Liver enzyme induction occurred at doses that did not cause any toxic effects in the monkeys, indicating that they may be less sensitive to the toxic effects of toxaphene than laboratory rodents.

#### *Immunosuppression*

Cynomolgus monkeys were treated with toxaphene at 1 mg/kg body weight/day for 52 weeks to study immune effects (Tryphonas *et al.*, 2000). Effects were seen that were not statistically significant, but which indicated possible negative effects of long-term exposure to toxaphene on humoral immunity.

#### *Thyroid and retinol effects*

Toxaphene caused thyroid follicular cell hypertrophy, intrafollicular hyperplasia and increased production of TSH in rats given 100 mg/kg/day for three days (Waritz *et al.*, 1996). No changes were seen for T3 or T4. The mechanism proposed for this effect was that toxaphene induced cytochrome P450 enzymes (CYP2B type) which led to increased excretion of T4, thereby stimulating the pituitary gland to produce and excrete more TSH. This, in turn, led to thyroid hypertrophy and hyperplasia.

#### *Cancer*

Toxaphene is a potent carcinogen in rats and mice (reviewed in Reuber, 1979; Saleh, 1991; de Geus *et al.*, 1999). Toxaphene induces malignant liver tumors, reticulum cell sarcomas, uterine sarcomas, reproductive system tumors, mammary gland tumors, and tumors in the pituitary, adrenal and thyroid glands.

Toxaphene is mutagenic in the Ames test (Hooper *et al.*, 1979; Mortelmans *et al.*, 1986). When toxaphene and four single congeners (Parlars 26, 50, 62, and 32) were tested for mutagenicity using two different bacterial (*Salmonella typhimurium*) strains, toxaphene was mutagenic but the single congeners were not (Steinberg *et al.*, 1998). Schrader *et al.* (1998) found that technical toxaphene was mutagenic in all five *S. typhimurium* strains tested but high concentrations were required. However, no mutagenesis was seen for toxaphene when tested in Chinese hamster V79 lung fibroblasts. Toxaphene and Parlar 32 were found to be genotoxic using the Mutatox assay, but Parlars 26, 50, and 62 were not genotoxic (Boon *et al.*, 1998).

Low concentrations of toxaphene induce micronuclei formation *in vitro* in beluga skin fibroblasts (Gauthier *et al.*, 1999).

Non-cytotoxic concentrations of toxaphene inhibited GJIC in normal human breast epithelial cells, in a dose-dependent manner (Kang *et al.*, 1996), indicating it may be a tumor promoter. No DNA adducts were found in the livers of mice treated with toxaphene (Hedli *et al.*, 1998). Toxaphene has been found to down-regulate the retinoblastoma gene, a tumor suppressor gene, indicating that toxaphene could promote tumors by turning off tumor suppression (Rought *et al.*, 1999).

### 3.3.3. Other pesticides

#### 3.3.3.1. TBT and its metabolites (DBT, MBT)

TBT is one of the most toxic substances deliberately introduced into natural waters (Goldberg, 1986). TBT is moderately lipophilic and bioconcentrates and bioaccumulates in the marine environment (Tanabe, 1999; Maguire, 2000; Hoch, 2001), and is a classic endocrine disrupter (Matthiessen and Gibbs, 1998). For recent reviews on TBT toxicology, see Fent (1996a), Maguire (2000) and WHO (1999). Chronic effects are observed

at exposure levels of 1000 ng/L or less for oysters, mussels and crustaceans (Rexrode, 1987), while the most sensitive species (dogwhelk snails, e.g., *Nucella lapis*) show sublethal effects at concentrations of only a few ng/L or less (Bryan and Gibbs, 1991; Gibbs, 1993; Stewart and Thompson, 1994).

Recently (since the mid-1990s), studies have been published showing the occurrence of TBT and other butyltins (MBT, DBT) in fish, birds, and terrestrial and marine mammals from the Pacific Ocean (Japan, Australia, Taiwan, India, Bangladesh, Thailand, Vietnam, Indonesia, Alaska, U.S., and open ocean areas) (Iwata *et al.*, 1995; Kannan *et al.*, 1995a; 1995b; Guruge *et al.*, 1996; 1997; Kim *et al.*, 1996a; 1996b; 1996c; Takahashi *et al.*, 1997; 1999; Tanabe *et al.*, 1998), in the Baltic Sea (Kannan and Falandysz, 1997), on the U.S. Atlantic and Gulf coasts (Kannan *et al.*, 1997), in a freshwater lake in the Netherlands (Stäb *et al.*, 1996), and along the coast of Italy (Kannan *et al.*, 1996).

Butyltins concentrate in the liver, blubber, and muscle of vertebrates. Higher relative amounts of MBT and DBT are found in birds and mammals as compared to fish, due to metabolism of TBT.

#### *Reproductive and developmental effects*

TBT has been found to have high binding affinity to the androgen receptor in an *in vitro* assay but shows no affinity for the estrogen receptor (Satoh *et al.*, 2001).

At concentrations of 1-2 ng/L, dogwhelk snails exhibit imposex. Imposex is caused by TBT interference with the biosynthesis of steroid hormones (i.e., the synthesis of 17 $\beta$ -estradiol from testosterone and the synthesis of estrone from androstenedione) (Bettin *et al.*, 1996). High levels of testosterone result in the development of a penis and vas deferens in female neogastropods. At levels of 7-10 ng/L, the vas deferens can overgrow the genital opening of the female, resulting in reproductive failure of the species (Gibbs *et al.*, 1987). Thresholds for other species are poorly known. Studies with cod (*Gadus morhua*) (Granmo *et al.*, 2002) indicate that threshold levels for cod embryos are higher than found in the Arctic environment.

Imposex has been observed most frequently in coastal areas near obvious TBT sources such as marinas or harbors, and has been associated with TBT paints on both pleasure boats and commercial shipping. Sediments in particular seem to be reservoirs for TBT especially after use has stopped, leading to continued exposure (Maguire, 2000). Open ocean areas are exposed to TBT from large vessels that are still allowed to use TBT on their hulls, with imposex being found in whelks (*Buccinum undatum*) collected from the open North Sea along shipping routes, indicating that the problem is not confined to coastal areas (Ellis and Pattisina, 1990; Bryan and Gibbs, 1991; Ten Hallers-Tjabbes *et al.*, 1994).

TBT causes developmental effects in the early life stages of fish. Water concentrations of 690-820 ng/L cause scoliosis and an inability to swim in minnows (*Phoxinus phoxinus*) (Fent, 1992) and notochord length is significantly reduced in larval striped bass (*Morone saxatilis*) at 514 ng/L (Pinkney *et al.*, 1990). Long-term exposure of rainbow trout yolk sac fry to 1000 ng/L resulted in decreased numbers of red blood cells, increased

liver weight, and liver cell hyperplasia (Seinen *et al.*, 1981). A NOAEL of 40-50 ng/L for rainbow trout yolk sac fry has been proposed (De Vries *et al.*, 1991). TBT exposure in medaka embryos on day 0 led to concentration-related lethality, reductions in hatching success, increases in gross abnormalities (bent, curled and/or shortened tails caused by reduced number of somites) and slowed developmental rates (Bentivegna and Piatkowski, 1998). The LOAEL for TBT combined chronic effects in medaka embryos was 12 500 ng/L.

TBT-oxide (TBTO) exposure (0, 2.7 and 9 ng/ml) for four days led to significant changes in spatial position, response to predator attack, recovery time and latency time in three-spined stickleback (Wibe *et al.*, 2001).

Dietary exposure of Japanese quail (*Coturnix coturnix japonica*) to 60 000 and 150 000 ng TBTO/g food led to reduced egg hatchability and an increase in the percent of chicks found dead in the shell (Coenen *et al.*, 1992; Schlatterer *et al.*, 1993). The no-observed-effect concentration (NOEC) for reduced egg weight and hatchability was 60 000 ng/g food (Schlatterer *et al.*, 1993).

#### *Cytochrome P450-dependent monooxygenases*

TBT exposure to 3.3, 8.1 and 16.3 mg/kg (intraperitoneal injection) led to the concentration-dependent degradation of CYP1A and loss of EROD activity *in vitro* and *in vivo* in scup (*Stenotomus chrysops*) (Fent and Stegeman, 1991; 1993; Fent *et al.*, 1998). The lowest dose level resulted in liver TBT concentrations of 8000 ng/g. At the high dose levels used, CYP2B (responsible for testosterone 15 $\alpha$ -hydroxylase activity) and CYP3A (responsible for testosterone 6 $\beta$ -hydroxylation) proteins were also destroyed, which could lead to effects on steroid metabolism and possible endocrine disruption.

TBT exposure *in vitro* in rainbow trout, bullhead (*Cottos gobio*), and eel (*Anguilla anguilla*) hepatic microsomes led to strong inhibition of EROD activity and reduction in total cytochrome P450 protein levels (Fent and Bucheli, 1994), with rainbow trout microsomes being most sensitive. Rainbow trout hepatocytes also showed reduced cytochrome P450 levels after exposure to 1  $\mu$ M TBT (290 000 ng/L) (Reader *et al.*, 1996). In the fish hepatoma cell line PLHC-1, TBT exposure led to inhibition of EROD, and decreased levels of CYP1A and DBT exposure also led to inhibition of EROD, but was less potent than TBT (Brüschweiler *et al.*, 1996). These studies indicate that TBT is metabolized by the P450 monooxygenase system but that it also inhibits this system, affecting its own metabolism and that of other substances. TBT and DBT both inhibit carboxylesterases in the tropical marine fish *Siganus canaliculatus*, with DBT being most potent (Al-Ghais *et al.*, 2000). The IC<sub>50</sub>s (concentrations at which 50% inhibition occurs) were 180-385  $\mu$ M (52 200-112 000 ng/g) for TBT and 17-49  $\mu$ M (4000-11 400 ng/g) for DBT.

*In vitro* inhibition of hepatic cytochrome P450 in Dall's porpoise (*Phocoenoides dalli*) and Steller sea lion hepatic microsomes by TBT has also been shown (Kim *et al.*, 1998b). Total P450 levels decreased and EROD and PROD activity decreased with increasing doses. The apparent effect threshold concentration was estimated to be 100  $\mu$ M TBT (29 000 ng/g). Comparison of the composition of TBT and its metabolites DBT and MBT

in marine mammals and their prey indicates that pinnipeds (seals, sea lions) have a higher metabolic capacity for organotins than cetaceans (dolphins, porpoises, whales) (Tanabe *et al.*, 1998; Tanabe, 1999). Both TBT and DBT are hepatotoxic in mice, with DBT being more potent than TBT (Ueno *et al.*, 1994). TBT toxicity in this study was attributed to the metabolism of TBT to DBT and subsequent accumulation of DBT. The effect threshold for hepatotoxic effects in mice was 2600 ng DBT/g ww in liver.

#### *Immunosuppression*

Both TBT and DBT have been found to be immunosuppressive in a range of animals, causing thymic and splenic atrophy, reductions in thymic, circulating and splenic lymphocytes, suppression of T-cell-dependent immunity, and suppression of tumoricidal activity (Seinen *et al.*, 1977; Krajnc *et al.*, 1984; Vos *et al.*, 1984; Snoeij *et al.*, 1985; 1988). For a review of organotin immunotoxicity, see Snoeij *et al.* (1987). DBT and TBT also suppressed concanavalin-A-induced mitogenesis in peripheral blood monocytes of marine mammals (Nakata *et al.*, 2002). *In vitro* immunotoxicity is seen in rat thymocytes (Snoeij *et al.*, 1986) and rabbit polymorphonuclear leucocytes (Elferink *et al.*, 1986) with an effect threshold less than 1.0  $\mu$ M TBT (290 ng/g ww) in both studies. A NOEL of 500 ng/g body weight has been proposed for TBTO immunotoxicity in rats (Verdier *et al.*, 1991). The most sensitive *in vivo* effect of TBT-induced immunosuppression is seen in weanling rats, as measured by reduced IgE titers and reduced resistance to parasitism by *Trichinella spiralis* in muscle (Vos *et al.*, 1990). The NOAEL was determined to be 25  $\mu$ g/kg/day (equivalent to 500 ng/g in diet), and the LOAEL was 250  $\mu$ g/kg/day (equivalent to 5000 ng/g in diet).

One possible mechanism that has been proposed for TBT immunotoxicity is the induction of apoptosis, or programmed cell death, in the thymus (Pieters *et al.*, 1994). This may be related to alterations of Ca<sup>2+</sup> home-

ostasis, formation of reactive oxygen species and DNA fragmentation (Gennari *et al.*, 2000).

TBT is also immunotoxic in rainbow trout immune cells, but DBT has been found to be more potent as an immunotoxin (O'Halloran *et al.*, 1998). Both TBT and DBT were found to suppress mitogenic activity in trout head kidney and splenic cells at exposure concentrations of 50 ng/g or greater (154 nM). Studies in rainbow trout *in vivo* also show similar types of effects to those seen in mammals, such as lymphoid depletion and immune modulation (Schwaiger *et al.*, 1992). In channel catfish (*Ictalurus punctatus*), TBT affected humoral immunity as measured by response to *Edwardsiella ictaluri* infection (Rice *et al.*, 1995). The effects were significant for all doses (10, 100, and 1000 ng/g administered intraperitoneally), but the strength of the response was dose-dependent. Flounder (*Platichthys flesus*) exposed to fairly high TBT concentrations ( $\mu$ g/L) had a significant decrease in thymus volume (Grinwis *et al.*, 1998).

In Canada, studies have shown effects of TBT, DBT, and MBT on the *in vitro* phagocytic activity of hemocytes from three marine bivalve species, *Mytilus edulis*, *Mya arenaria*, and *Mactromeris polynyma*, using flow cytometry (Bouchard *et al.*, 1999). Phagocytosis was reduced with increasing doses of TBT and DBT, and the toxicity of butyltins on hemocytes decreased in the order DBT > TBT > MBT. The comparison of the relative sensitivity of the three species showed that blue mussels (*M. edulis*) were more tolerant of butyltin compounds than both clam species.

#### *Cancer*

Generally, negative results have been obtained when TBT is tested for mutagenicity in various test systems (WHO, 1990). However, Hamasaki *et al.* (1993) have shown that MBT, DBT and TBT were mutagens in *S. typhimurium* TA100. Carcinogenicity has not been well investigated. Since the data on genotoxicity of TBT are not consistent, it is not possible to draw definite conclusions.

## Chapter 4

# Regional and Circumpolar Levels and Trends in Abiotic and Biotic Media

This section reviews new information on the levels and spatial trends of POPs in Arctic abiotic and biotic media available since 1996. Some earlier data which were omitted from the previous assessment are also included. Most interpretations in this chapter, including comparisons and conclusions about spatial trends, were made by scientists in charge of projects described (most are listed as contributing authors) and by the four editors. In some cases interpretation was not possible due to limited sample numbers. The new data are primarily entered as summary data in the Annex Tables, and interpretations were kept to a minimum. It should also be noted that sample sizes for results given in the Annex Tables vary greatly. Readers should be aware of the risk of spurious conclusions drawn from small data sets. Within the AMAP monitoring program, QA/QC criteria have been defined, and these have been followed by almost all laboratories that have contributed data to the assessment. All laboratories providing data within their national monitoring programs have also taken part in intercalibration exercises. Therefore, the data presented here are comparable across laboratories. The only major exceptions occur when varying numbers of congeners have been analyzed and quantified. This is most prevalent for PCBs and toxaphene, both of which have been reported as differing numbers of congeners as well as total sums based on quantification against a technical product (total PCB, total toxaphene).

Readers are also reminded that concentrations are expressed in several ways in this report. In biological samples concentrations are expressed on a wet weight basis,

giving the concentration of OC in the total weight of tissue analyzed; on a lipid weight basis, giving the concentration in the lipids in that particular tissue; or as a body burden, giving the contaminant concentration in the whole organism. OC levels in abiotic samples and some biotic samples, most often plants, are expressed on a dry weight basis. The following abbreviations are used throughout the text to distinguish lipid weight concentrations (that is 'ng/g lw'), from results reported on a wet weight ('ww') or dry weight ('dw') basis. All lipid weight concentrations are calculated from the wet weight concentrations unless otherwise indicated. Concentrations of OCs in air samples expressed as  $\text{pg}/\text{m}^3$  are presented for gas phase and particle phase separately or are summed to give a total air concentration. Fluxes of OCs in sediments are expressed as  $\text{ng}/\text{m}^2/\text{yr}$  (i.e. concentration in  $\text{ng}/\text{g} \times \text{sedimentation rate } \text{g}/\text{m}^2/\text{yr}$ ).

## 4.1. Atmospheric environment

### 4.1.1. Air

#### 4.1.1.1. Introduction

Measurements of POPs in Arctic air have continued on a weekly basis at locations in Canada, Iceland, Norway, Finland, and Russia, building on the datasets discussed in the first assessment. In addition, over the past five years, a number of ship-based studies have also measured air concentrations of POPs over shorter time periods to address air-water exchange.

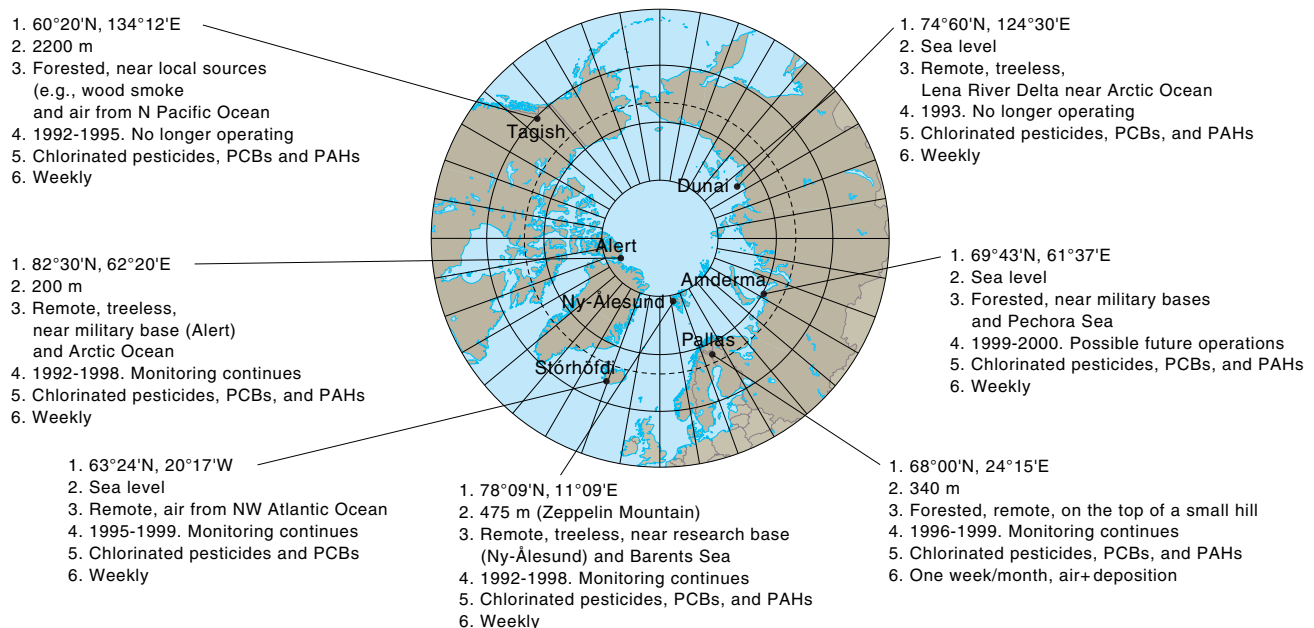


Figure 4-1. Locations and site-specific information for each POPs air monitoring station. 1. Latitude/longitude; 2. Elevation; 3. Description; 4. Sampling period and status as of 2002 (this is the total operational period for which data were available for this assessment. Sampling period varies with chemicals measured); 5. POPs monitored; 6. Sampling schedule.



Data were available for this assessment from the six land-based stations: Tagish (Yukon, Canada); Alert (Nunavut, Canada); Pallas (Finland); Stórhöfði (Iceland); Dunai (eastern Russia); and, Ny-Ålesund (Svalbard, Norway). The data from the Norwegian, Icelandic, Finnish and Canadian stations were made available from the AMAP Thematic Data Center for atmospheric contaminants at the Norwegian Institute for Air Research (Kjeller, Norway). In addition, selected results from the air monitoring station at Amderma in northwest Russia, jointly operated (since March 1999) by Russia and Canada, were available for the assessment.

The locations of these stations relative to possible local, as well as long-range transported POPs, constitute an important consideration when evaluating spatial trends (Figure 4-1). Most stations are at remote but well-established research stations. Some are influenced by local events such as forest fires, wood burning for domestic heating, and garbage burning. This may be the case for the Tagish site in particular, as discussed further in Section 4.1.2.5.3. Samplers at Alert and Ny-Ålesund have experienced contamination due to presence of OC pesticides in building materials (Alert) and PCBs (Ny-Ålesund). Altitude may also be a factor. Tagish is much higher than all other sites and receives air from the northeast Pacific Ocean.

Alert, Tagish, Dunai, Amderma, and Ny-Ålesund operate on a weekly sampling basis, while at Pallas sampling is conducted one week per month, and Stórhöfði reports data from samples collected over a two- to three-week period. In order to compare results with the weekly samples from the rest of the stations, the week of the sampling period was chosen for the comparison and the other weeks omitted. Using this approach, it was possible to compare the data from all stations; however, the resolution of the Icelandic data is reduced and of limited use for the elucidation of long-range transboundary events.

In the discussion that follows, average annual concentrations of selected POPs are compared among the stations to examine prevailing spatial trends in the late 1990s. Results from shipboard measurements in the late 1990s are also discussed. An assessment of long-term temporal trends in air from many of the same stations is presented in Section 5.1.1.

#### 4.1.2. Air concentrations – spatial trends

##### 4.1.2.1. OC pesticides

A full list of the OC chemicals monitored is given in Annex Table 2. Major OC pesticides in Arctic air are  $\alpha$ - and  $\gamma$ -HCH,  $p,p'$ -DDT, chlordane compounds, endosulfan, pentachloroanisole, as well as HCB (a multisource chemical) (Halsall *et al.*, 1998; Hung *et al.*, 2002b). Unfortunately, not all of these compounds are measured at all locations. HCH and DDT-related compounds are compared in Figure 4-2 at five locations using average, minimum and maximum concentrations in the gas + particle phase ( $\text{pg}/\text{m}^3$ ) for different years. Results suggest rather uniform average concentrations of  $\gamma$ -HCH isomers and greater regional variation of  $\alpha$ -HCH. In all Arctic samples,  $\gamma$ -HCH represents about 15-20% of the total  $\alpha$ - and  $\gamma$ -HCH burden. This distribution seems to be independent of the geographic location of the station. The lowest  $\Sigma$ HCH average concentrations were measured in the Stórhöfði and Amderma samples. The highest average values were found at Ny-Ålesund and Alert in 1996 (73 and 62  $\text{pg}/\text{m}^3$   $\Sigma$ HCHs, respectively).

Highest average and maximum values of DDT-related compounds were found at Stórhöfði. The contribution of the parent compounds  $o,p'$ - and  $p,p'$ -DDT, accounts for more than 60% of the total DDT burden suggesting fresh sources. The DDT group pattern at Amderma also had a high proportion of  $p,p'$ -DDTs (50% of  $\Sigma$ DDTs). However, overall levels of  $\Sigma$ DDTs at Amderma were lower than Stórhöfði and similar to the other sites (Figure 4-2). At Alert, summertime DDT/DDE ratios of the order of 1-1.5 have been observed, whereas for Amderma, the DDT/DDE ratios were about 3. A larger DDT/DDE ratio has also been observed at Tagish in western Canada and is linked to trans-Pacific transport from Asia (Bailey *et al.*, 2000). Higher DDT/DDE ratios are indicative of fresh sources.

Elevated maximum concentrations, indicating incursions of southern air with higher levels of  $\gamma$ -HCH and DDT compounds, occurred in spring and summer months particularly at Ny-Ålesund, Alert, and Stórhöfði. Halsall *et al.* (1998) also reported relatively uniform mean annual concentrations of HCH and DDT isomers in Arctic air collected in 1993 from Alert, Dunai, Tagish, and Ny-Ålesund. Back-trajectory results for the 1996 and 1998

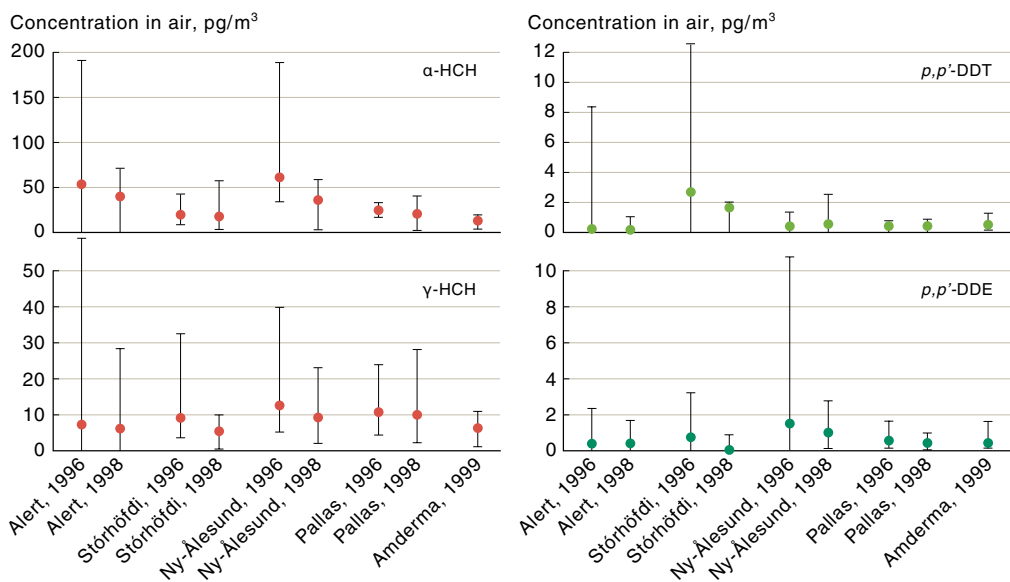


Figure 4-2. Comparison of average annual concentrations of major HCH- and DDT-related compounds in air (gas + particle phase) at monitoring stations in Canada (Alert), Iceland (Stórhöfði), Norway (Ny-Ålesund, Zeppelin Mountain), Finland (Pallas), (all 1996 and 1998), and western Russia (Amderma) (1999). Bars represent the ranges.

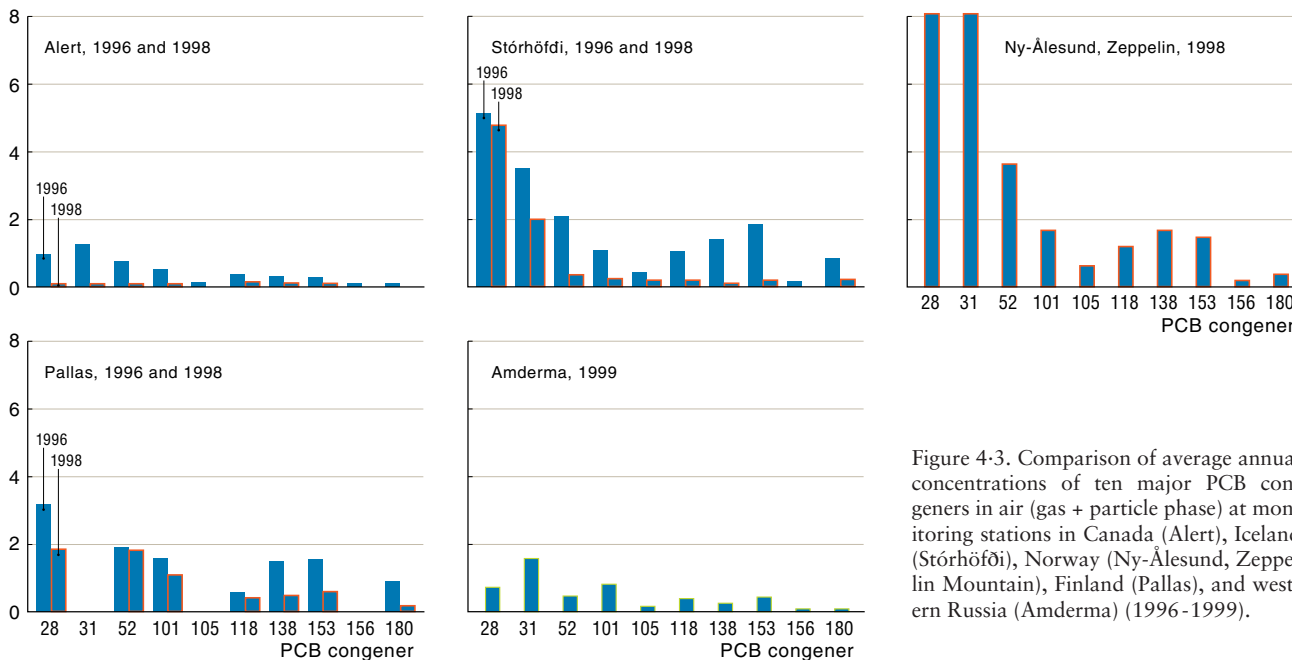
Concentration in air, pg/m<sup>3</sup>

Figure 4-3. Comparison of average annual concentrations of ten major PCB congeners in air (gas + particle phase) at monitoring stations in Canada (Alert), Iceland (Stórhöfði), Norway (Ny-Ålesund, Zeppelin Mountain), Finland (Pallas), and western Russia (Amderma) (1996-1999).

data shown in Figure 4-2 are not available. Harner *et al.* (1999), however, measured HCH isomers during a cruise in the Barents Sea and the eastern Arctic Ocean in July-September 1996 and found that concentrations and proportions of  $\gamma$ -HCH increased in air masses that had passed over central Europe. This is consistent with similar observations at Lista, Norway (Haugen *et al.*, 1998) and the southern Baltic Sea (Wiberg *et al.*, 2001). In the Barents Sea and eastern Arctic Ocean, summertime, above-ocean concentrations of  $\alpha$ -HCH (11-68 pg/m<sup>3</sup>) and  $\gamma$ -HCH (6-68 pg/m<sup>3</sup>) measured by Harner *et al.* (1999) were within the range observed at Alert and Ny-Ålesund at the same time of year, but higher than at Pallas and Stórhöfði.

#### 4.1.2.2. PCBs

Figure 4-3 presents the distribution of ten PCB congeners (PCB<sub>10</sub>) (seven congeners for Pallas) measured at five stations in 1996 and/or 1998 or 1999. Results were not available from all stations for all years. In particular, there were blank problems at the Ny-Ålesund site so that the 1996 data could not be used. Earlier measurements of PCBs in air (1993-94) from Alert, Tagish, and Dunai have been published (Fellin *et al.*, 1996; Stern *et al.*, 1997) and, along with results from Ny-Ålesund and Stórhöfði (Heimaey Island, Iceland), were discussed in the previous AMAP assessment (de March *et al.*, 1998). The congener distribution is derived from annual average concentrations. In general, PCBs 28, 31, and 52 form the majority of the  $\Sigma$ PCB<sub>10</sub> burden. Reporting of only ten PCB congeners seriously underestimates the total PCBs in air and makes source identification problematic. For example,  $\Sigma$ PCB<sub>10</sub> represented only 11-27% of total PCB (based on 88 congeners) at Alert (1994), Dunai, and Tagish (Stern *et al.*, 1997). Major contributors to total PCBs at Alert, and probably at all Arctic sites, are the mono- and dichlorobiphenyls (CBs 3, 4, 5, 6, 8, 16, 18). Nevertheless,  $\Sigma$ PCB<sub>10</sub> was highly correlated ( $r^2 = 0.80$ ) with total PCBs at Alert, which suggests that overall trends can be assessed.

Highest  $\Sigma$ PCB<sub>10</sub> concentrations were found at Ny-Ålesund compared to other sites (Figure 4-3). Results for Alert were consistently lower than other sites. Amderma, in northwest Russia, had average  $\Sigma$ PCB<sub>10</sub> of 4 pg/m<sup>3</sup> in 1999-2000 (the first year of site operations), similar to Alert and lower than Pallas or Stórhöfði. Homologues at Amderma were more evenly distributed than at sites such as Alert and Tagish. There were similarities between Amderma and Dunai, another Russian site operated in the mid-1990s. The concentration range of 5 to 30 pg/m<sup>3</sup> for  $\Sigma$ PCB<sub>10</sub>, or about 20-100 pg/m<sup>3</sup> for total PCBs, probably represents background levels independent of the station location. Air mass back-trajectories for Alert and Dunai, reported by Stern *et al.* (1997), showed that the PCB profile of air masses passing over Russia differed from air masses from the North American sector by having higher proportions and concentrations of penta- and hexachlorobiphenyls. Samples originating over the northern Pacific had higher proportions of trichlorobiphenyls.

#### 4.1.2.3. PCDD/Fs

During the winter of 2000/2001, 15 weekly filter samples from Alert (particulate phase) were analyzed for PCDD/Fs (Hung *et al.*, 2002a). The sampling period coincided with the occurrence of Arctic haze, when airborne particulate levels are high. Since PCDD/Fs have similar properties to PAHs (e.g., both originate from combustion sources and have low volatility with high tendencies to partition to aerosols), they are expected to follow similar seasonal cycles, and thus maximum concentrations of PCDD/Fs were anticipated during winter months. Table 4-1 (next page) compares the atmospheric concentrations observed at Alert (week 49 of 2000, weeks 3 and 7 of 2001) with those found at other locations. The concentrations of both  $\Sigma$ PCDDs and  $\Sigma$ PCDFs observed were lower than those found at Ny-Ålesund in the mid-1990s (Schlabach *et al.*, 1996) and lower than most other locations worldwide except the

Table 4-1. Comparison of reported concentrations of PCDD/Fs in Arctic and temperate air samples.

Location/dates	Total PCDD (fg/m <sup>3</sup> )	Total PCDF (fg/m <sup>3</sup> )
Alert, filter only (11/00-02/01)	2.1-13	2.4-46
Ny-Ålesund (21/4-17/5/95) <sup>a</sup>	28	76
Ny-Ålesund (21/7-23/8/95) <sup>a</sup>	16	51
Trout Lake, Wisconsin (mid-1990s) <sup>b</sup>	240	180
Bloomington, Indiana – suburban <sup>b</sup>	2500	2600
Nordrhein-Westfalen, Germany – urban and industrial <sup>b</sup>	3200	5500
Barbados (18/3/96-13/8/97) <sup>c</sup>	6.8	12
Bermuda (4/6/96-20/8/97) <sup>c</sup>	25	16

<sup>a</sup> Schlabach *et al.* (1996).

<sup>b</sup> Adapted from Table 3 of Luger *et al.* (1996).

<sup>c</sup> Calculated from supplementary material of Baker and Hites (1999).

ocean island locations of Bermuda and Barbados. The low PCDD/F levels are attributable to the remoteness of Alert and the absence of anthropogenic activities nearby. At the time of sampling, during the first week of 2001, Alert was mainly affected by air originating from the North Atlantic and North America. During the second and third weeks, when the air concentrations of PCDD/Fs peaked at Alert, the origin of the air mass shifted eastward further into Russia and Eurasia. After that time, the influence from Eurasia decreased while the North Atlantic sector regained dominance when the concentrations tapered off during the fourth and fifth weeks.

#### 4.1.2.4. PAHs

All stations reported up to 25 PAH compounds in atmospheric samples to the AMAP database (see list in Annex Table 2); however, only 11 PAH compounds (PAH<sub>11</sub>) were reported from all stations. Thus, a level comparison for the compounds anthracene, benzo[a]anthracene, benzo[a]pyrene, benzo[b,j,k]fluoranthene, benzo[g,h,i]perylene, chrysene, dibenzo[a,h]anthracene, fluoranthene, indeno[1,2,3-cd]pyrene, phenanthrene, and pyrene was performed (Figure 4-4). Airborne PAHs were not reported in the previous assessment of POPs or petroleum hydrocarbons (Robertson, 1998). Results from Alert, Dunai, and Tagish from 1993-1994 were, however, reported by Halsall *et al.* (1997). Therefore, results

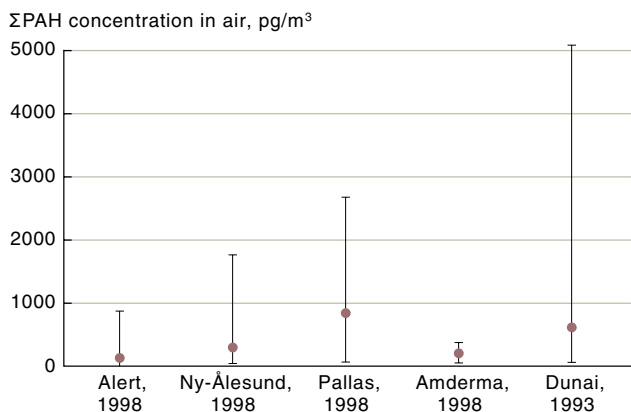


Figure 4-4. Average and range of concentrations of PAHs in air (gas + particle phase) from five Arctic air monitoring stations during the 1990s. Results are based on 11 PAH compounds that are monitored at all stations.

from 1993 for Dunai were included in Figure 4-4 for comparison with more recent results from Alert, Ny-Ålesund, Pallas, and Amderma to give a broader perspective of prevailing levels in air. Temporal trends of PAHs in air, using the long-term data, are presented in Section 5.1.1.3.

All stations reported wide ranges of air concentrations of ΣPAH<sub>11</sub>, ranging from less than 10 pg/m<sup>3</sup> (the approximate reporting limit) to 5000 pg/m<sup>3</sup> (vapor phase and particles). Highest average annual concentrations of airborne ΣPAH<sub>11</sub> were found at Pallas and lowest at Alert (Figure 4-4). PAH concentrations in air from the Ny-Ålesund station were similar overall to those at Alert and Pallas, although some high maxima have been recorded (maximum value of 5800 pg/m<sup>3</sup> in 1997). The data from Amderma were missing results from winter time and therefore, were not suitable for comparison.

Halsall *et al.* (1997) found clear seasonality in the PAH concentrations at Alert, Dunai, and Tagish (1993-94), with highest concentrations occurring during the colder months. Highest concentrations of ΣPAHs (18 unsubstituted PAH) were found at Dunai (2580 ± 2230 pg/m<sup>3</sup>) during the winter period (November-March) compared to Alert (714 ± 579 pg/m<sup>3</sup>) and Tagish (312 ± 236 pg/m<sup>3</sup>). Air mass back-trajectories revealed that long-range transport from Eurasian sources gave rise to elevated PAH concentrations at Dunai and Alert. These elevated concentrations were also characterized by high levels of dibenzothiophene, a marker of coal and oil combustion, and by high ratios of benzo[a]pyrene (B[a]P) to benzo[e]pyrene (B[e]P) (0.6-0.8) which resemble urban air sources. Air at Ny-Ålesund was also characterized by high B[a]P/B[e]P ratios (average 2.7) suggesting long-range transport, as well as possible influences from local combustion on Svalbard. This suggests very limited breakdown of B[a]P in the troposphere during the winter months. The B[a]P was 100% in the particulate phase during the winter period at all locations.

Halsall *et al.* (2000) developed a simple model to describe the removal processes for four PAHs: fluorene, phenanthrene, fluoranthene and benzo[a]pyrene, transported over a five-day period from the U.K. to the Arctic. They noted that U.K. urban air was dominated by phenanthrene (52% of the sum of four compounds) and had a relatively high proportion of B[a]P (3%). By contrast, air at Dunai had much higher fractions of fluorene. The more rapid degradation of phenanthrene by

Table 4-2. Current-use pesticides and other semi-volatile organochlorine compounds that are now being measured in Arctic air (gas phase) during 1993-1994, pg/m<sup>3</sup> (Halsall *et al.*, 1998; Bidleman *et al.*, 1999; 2000; 2001; Helm and Bidleman, 2003).

Compound	Alert			Tagish			Dunai		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
<i>Current-use pesticides</i>									
Endosulfan	4.22	0.02	16.2	7.05	0.08	88.6	2.99	0.05	7.18
Methoxychlor	0.27	0.07	1.43	0.36	0.09	6.53	0.41	0.22	0.73
Trifluralin	0.12	0.03	0.64	0.16	0.04	2.92	0.18	0.09	0.13
Pentachloroanisole	3.03	0.1	20.5	3.28	0.04	73.4	2.92	0.95	6.92
<i>By-products</i>									
Trichloroveratrole	0.93	0.05	10.1	1.85	0.07	20.6	1.49	0.15	3.65
Tetrachloroveratrole	0.19	0.05	0.99	0.25	0.07	4.52	0.44	0.15	2.19
Octachlorostyrene	0.79	0.04	2.96	0.67	0.07	11.9	0.60	0.11	1.74
<i>Other halogenated compounds</i> <sup>a</sup>									
Polychlorinated naphthalenes	0.50	<0.01	1.00	0.30	<0.01	1.40	0.79	<0.01	2.60
Coplanar PCBs <sup>b</sup>	0.085	0.03	0.20	0.047	<0.01	0.11	0.41	0.19	0.82
Polybrominated diphenyl ethers	282	10	868	424	27	2127	14	0	62
Chlorinated paraffins (C10-C13)	2.0	<0.4	7.3	–	–	–	–	–	–

<sup>a</sup> Based on retrospective analysis of 28-day composite samples from 1994 and January 1995 only.

<sup>b</sup> Coplanar PCB = Sum of CBs 81, 77, 118, 114, 105, 126, 156, and 169.

OH radicals, combined with a shift to the particulate phase as air masses pass into the polar region, probably accounted for the removal of phenanthrene and other higher molecular weight PAHs.

#### 4.1.2.5. 'New' chemicals in the Arctic atmosphere

##### 4.1.2.5.1. Current-use pesticides and chlorinated by-products

Halsall *et al.* (1998) reported the presence of the current-use pesticides endosulfan, methoxychlor, and trifluralin, as well as the pentachlorophenol metabolite, pentachloroanisole (PeCA), in air at Alert, Tagish, and Dunai during 1993-94. PeCA and endosulfan were among the top five pesticide-related compounds at all three sites, exceeded in concentration only by total PCBs, HCB, and  $\alpha$ - and  $\gamma$ -HCH (itself a current-use pesticide). Methoxychlor and trifluralin were also detected at sub-pg/m<sup>3</sup> concentrations at all three sites (Table 4-2). Trifluralin volatilization from agricultural soils has been well documented (Majewski *et al.*, 1998), and it has a very short atmospheric half-life (Mackay *et al.*, 1997). Its appearance in Arctic air is therefore, surprising, especially at all three locations. However, more than  $5 \times 10^6$  kg was applied annually in western Canada and U.S. in the mid-1990s for weed control in soybean, cotton, and cereal crops (NCFAP, 2001). Concentrations at Alert in 1994 (Halsall *et al.*, 1998) were more than 1000 times lower than observed over the Mississippi River by Majewski *et al.* (1998).

Methoxychlor had limited agricultural use on fruit crops (NCFAP, 2001) as well as in home-gardening products. Methoxychlor is more stable to sunlight photolysis than its chlorinated analog DDT, and thus may have a relatively long atmospheric half-life (Zepp *et al.*, 1977).

Two other methylated chlorophenolics, trichloroveratrole and tetrachloroveratrole, were reported by Halsall *et al.* (1998) in air from all three sampling locations at sub-pg/m<sup>3</sup> concentrations (Table 4-2). These compounds may originate from bleaching of wood pulp and chlorination of wastewaters. However, their widespread detection at all three locations suggests other unknown sources. Führer *et al.* (1996) have reported a series of chlorinated

anisoles and tetrachlorodimethoxybenzene in air from waste treatment plants and in marine air over the tropical Atlantic Ocean, suggesting that these chlorinated methoxylated aromatics are widespread in the troposphere.

OCS, a semi-volatile by-product of magnesium and chlorine manufacturing, was detectable at the three Arctic stations studied by Halsall *et al.* (1998). While mean concentrations of OCS were low relative to many OC pesticides, maximum concentrations were up to 15 times higher, especially at Tagish, illustrating that long-range transport events from source regions are contributing to the presence of OCS. Actual source regions have not been identified for OCS.

##### 4.1.2.5.2. PCNs and coplanar PCBs

Polychlorinated naphthalenes, mono- and non-*ortho*-substituted (coplanar) PCBs, polybrominated diphenyl ethers, and chlorinated paraffins were determined in retrospective analyses of pooled archived extracts from Alert and Dunai stations from 1994, and reported by Bidleman *et al.* (1999; 2000; 2001), Harner *et al.* (1998), and Helm and Bidleman (2003). The extracts represented 28-day composites (based on four weekly samples). Although use of the archived extracts meant that extraction conditions for these compounds were not optimized, the study nevertheless provided the first look at their presence in the Arctic as well as some information on seasonal variation.

The  $\Sigma$ PCN concentrations in 1994-1995 ranged from <0.01 to 1.00 pg/m<sup>3</sup> at Alert, <0.01 to 1.4 pg/m<sup>3</sup> at Tagish (both Canadian sites), and from <0.01 to 2.60 pg/m<sup>3</sup> at Dunai, Russia (Figure 4-5). The  $\Sigma$ PCN levels at Alert were within the range reported for 1993 (0.003-0.077 pg/m<sup>3</sup>), while those at Dunai (0.001-0.009 pg/m<sup>3</sup>) were comparable (Harner *et al.*, 1998).  $\Sigma$ PCN concentrations at these High Arctic sites were lower than  $\Sigma$ PCNs observed at lower latitudes over the eastern Arctic Ocean, Norwegian Sea, and Barents Sea (Harner *et al.*, 1998). Elevated  $\Sigma$ PCNs over the Barents Sea were associated with air trajectories from the North Sea – central Europe region, and, in subsequent work, Harner *et al.* (2000) found especially high  $\Sigma$ PCNs in urban air



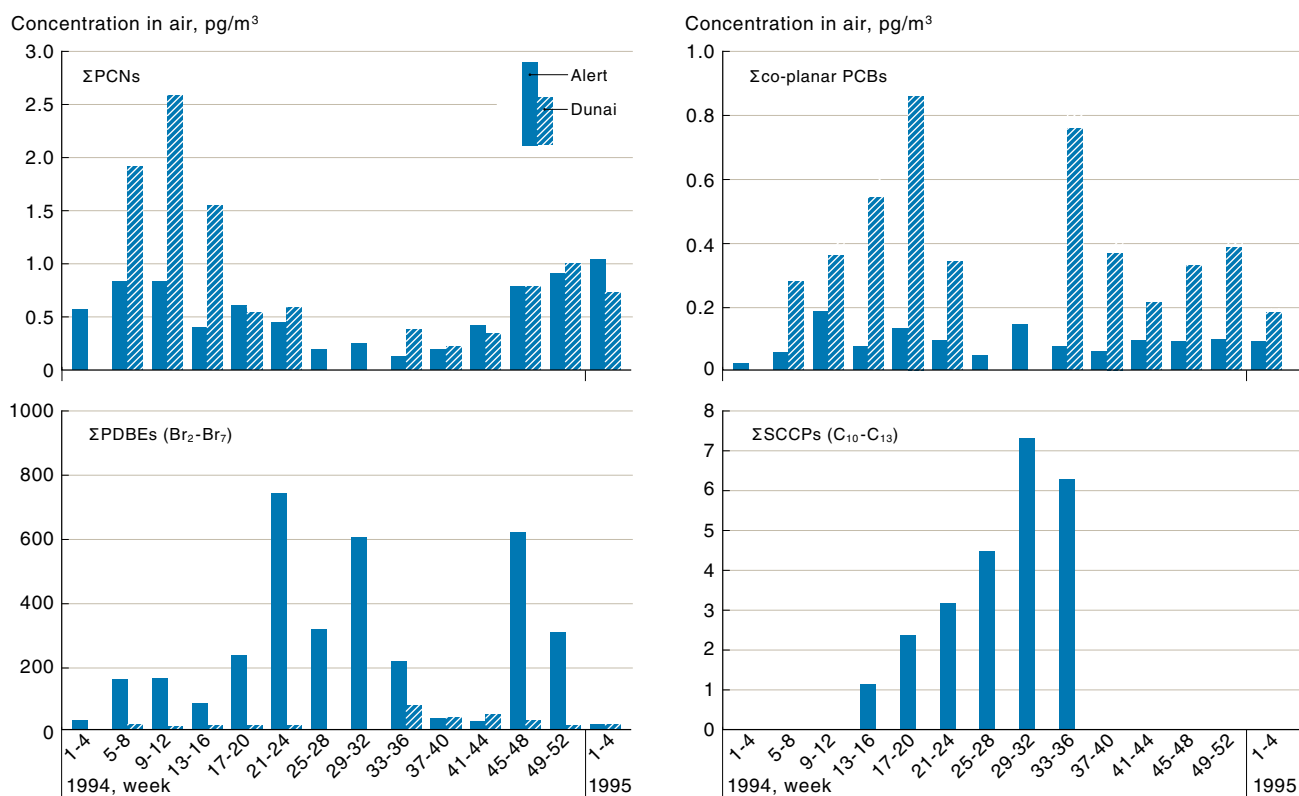


Figure 4-5. Concentrations of PCNs, co-planar PCBs, PBDEs and SCCPs in archived air samples (4-week composite samples from 1994 and early 1995).

from the U.K. Higher  $\Sigma$ PCN concentrations were found at Alert, Tagish, and Dunai in winter months (October-April) than in summer months (May-September) (Figure 4-5). This is similar to the seasonality observed for PAHs (Halsall *et al.*, 1997), and suggests that PCNs, like PAHs, may be combustion-related and associated with Arctic haze during winter/spring, or lost due to effects of photolysis in summer. In support of the former explanation, certain PCN congeners associated with combustion (e.g., CNs 13, 26, 29, 44, and 54) were enriched in the winter air samples (Helm and Bidleman, 2003).

The coplanar PCBs (non-*ortho* and mono-*ortho* substituted PCBs: CBs 81, 77, 118, 114, 105, 126, 156, and 169) were also measured in the archived samples from 1994 to early 1995 (Helm and Bidleman, 2003) and in an earlier set from 1993 (Harner *et al.*, 1998). With the exception of CBs 105, 118, and 156, these congeners have not been determined previously in Arctic air. The sum of these five congeners averaged  $0.085 \text{ pg/m}^3$  at Alert,  $0.047 \text{ pg/m}^3$  at Tagish, and  $0.41 \text{ pg/m}^3$  at Dunai for the 1994-95 set. Pentachlorobiphenyl congeners 118 and 105 were the dominant congeners at all sites. Seasonal variation was observed at the Dunai site with higher coplanar PCB concentrations during the summer months, while no trend was evident at Alert and Tagish. This agrees with the findings of Stern *et al.* (1997) that the contribution of PeCB congeners to  $\Sigma$ PCBs increased in the summer, particularly at Dunai. The authors suggested that emissions of Russian PCB mixtures with higher percentages of pentaCBs (PeCBs) could account for this observation. Coplanar PCB concentrations were elevated in shipboard samples collected from the Barents Sea in 1996 and, like PCNs, were associated with air transport from Europe (Harner *et al.*, 1998).

The contribution to dioxin TEQs in air samples was calculated for PCNs and coplanar PCBs using relative potencies or toxic equivalency factors determined by H-II-4 E bioassays for both PCNs (Blankenship *et al.*, 2000; Villeneuve *et al.*, 2000; Kannan *et al.*, 2001b) and PCBs (Giesy *et al.*, 1997). Average  $\Sigma$ TEQs (sum of TEQs for PCNs, mono- and non-*ortho* PCBs) during the cold season of 1994-1995 were  $0.022 \text{ fg/m}^3$  at Alert,  $0.0075 \text{ fg/m}^3$  at Tagish, and  $0.061 \text{ fg/m}^3$  at Dunai. PCNs contributed approximately 70-74% of TEQs relative to the PCBs in air at Alert and Dunai and 31% at Tagish. PCN congeners 66/67, 64/68, and 73 contributed most of the TEQs due to PCNs, while PCB congener 126 was by far the dominant contributor to coplanar PCB TEQ.

#### 4.1.2.5.3. PBDEs

PBDEs have been determined in archived air samples from the Canadian Arctic (Bidleman *et al.*, 2001) and in a short air monitoring campaign on Bjørnøya (Norway) in 1999-2000 (Kallenborn, 2002a). In the Canadian study, PBDEs were measured in the same archived extracts as the PCNs and coplanar PCBs (Bidleman *et al.*, 2001) (Table 4-2). Results for Dunai and Alert are shown in Figure 4-5. A significant fraction of the PBDEs (10-25% for BDE47; 10-90% for BDE99) were present in the particulate phase during the winter months. Levels of PBDEs in air samples from Dunai were much lower than at Alert. Much higher concentrations of PBDEs were found in air samples from Tagish (Table 4-2). The concentrations at Alert and Tagish were higher than air concentrations reported by Strandberg *et al.* (2001) for the city of Chicago (mean of  $52 \text{ pg/m}^3$  for 1997-1999) and much higher than concentrations at air monitoring sites

in the Great Lakes (means of 5-23 pg/m<sup>3</sup> at three sites). In the case of Tagish, there is the possibility that incineration of household items in the region could contribute PBDEs to the air at the sampling site. At Alert, the source has not been identified.

In contrast, the air concentrations of total PBDEs observed at Bjørnøya ranged from 3-10 pg/m<sup>3</sup> for the period of mid-December 1999 to mid-April 2000. The same suite of PBDE congeners as Alert were detected, with BDE47 predominating. Di- and TrBDEs (BDEs 13, 15, and 33) also were prominent. While no summertime concentrations are available from Bjørnøya, the results suggest that levels are much lower than at Alert or Tagish but similar to levels in rural areas of the Great Lakes. Thus, the reported results for Alert are probably not typical of concentrations of PBDEs in Arctic air, although they could be typical of low-temperature burning which is common in Arctic communities in Canada and Alaska. Further measurements are being conducted in an effort to better understand the levels and potential sources at Alert. The main PBDE congeners observed at all sites were BDEs 47, 99, 100, 153, and 154 with BDE99 having the highest concentrations. BDE47 predominated in all Great Lakes samples (Strandberg *et al.*, 2001), as well as in air samples in Sweden (Bergander *et al.*, 1995; de Wit, 2002). Mono-, di- and triBDEs were found in air samples from Alert, which may indicate photodegradation (debromination) of PBDEs during long-range atmospheric transport. The PBDE results, if typical, imply significant air contamination by these compounds. Air concentrations are higher than for ΣPCBs at the same sites.

#### 4.1.2.5.4. SCCPs

SCCPs were analyzed in the same archived extracts from Alert that were analyzed for PCBs, coplanar PCBs, and PBDEs. SCCPs were above method detection limits only

in samples from the summer months, and were highest in August. Individual homologue concentrations indicated that the chlorododecanes (C<sub>12</sub>), accounted for approximately 50% of the total, the bulk being from the hexa- and heptachlorododecanes. Levels of SCCPs were below detection limits in polyurethane foam plugs (PUFs) for periods other than summer months and also in all filters (Bidleman *et al.*, 2001).

#### 4.1.3. Air and fog water measurements at Bjørnøya

In 1994, very high levels of PCBs in Arctic char and sediment from the lake Ellasjøen on Bjørnøya (Bear Island, 74°N, 19°E), situated about 500 km southwest of Svalbard (Skotvold *et al.*, 1999), prompted a comprehensive research study which was started in 1999 (Evenset *et al.* 2002; Kallenborn, 2002a; 2002b). The studies included investigation of the influence of atmospheric long-range transport on POPs at the Ellasjøen area on Bjørnøya. This included the measurement of POPs in ambient air, snow, and fog water (Annex Table 3). Fog water was included based on information provided by the Norwegian Meteorological Institute that, on average, fog events were occurring more than 30% of the time in summer, and to assess fog as medium for transport and deposition pathways at Bjørnøya. A total of 20 air samples (average volume: 1000 m<sup>3</sup>) were collected every second week in 1999 and spring 2000. In the Ellasjøen catchment area and at the meteorological station on Bjørnøya, four fog water samples (approximately 50 L) were collected during the 2000 summer sampling campaign.

Continuous POP monitoring in ambient air at the 'Zeppelin Mountain' atmospheric research station (Ny-Ålesund, Svalbard) provided an excellent comparison for the Bjørnøya air samples. Typical POP distributions in air samples from Bjørnøya (meteorological station) and the Ny-Ålesund measuring station are presented in Figure 4-6. In general, the air concentrations of

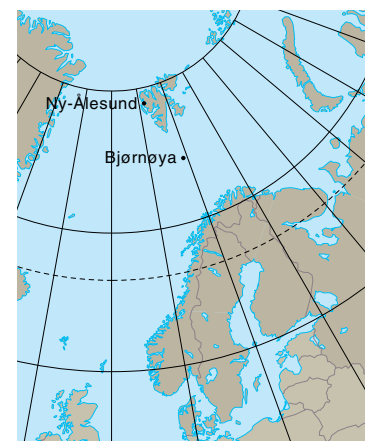
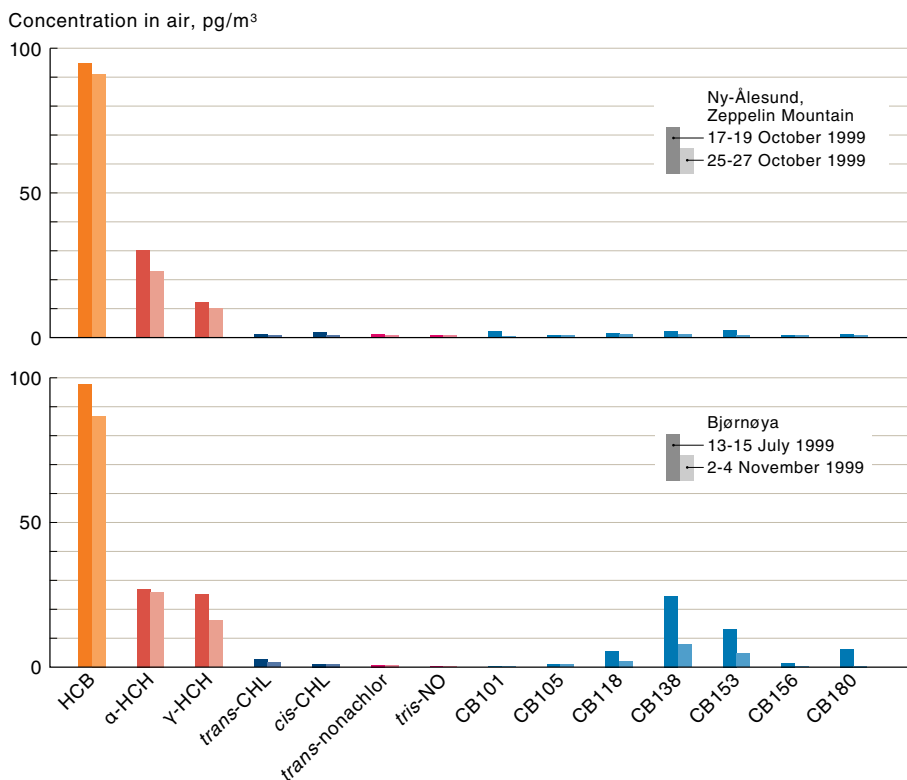


Figure 4-6. POP concentrations in ambient air from the Bjørnøya (Bear Island) meteorological station and the 'Zeppelin Mountain' atmospheric research station at Ny-Ålesund (Svalbard) in 1999.

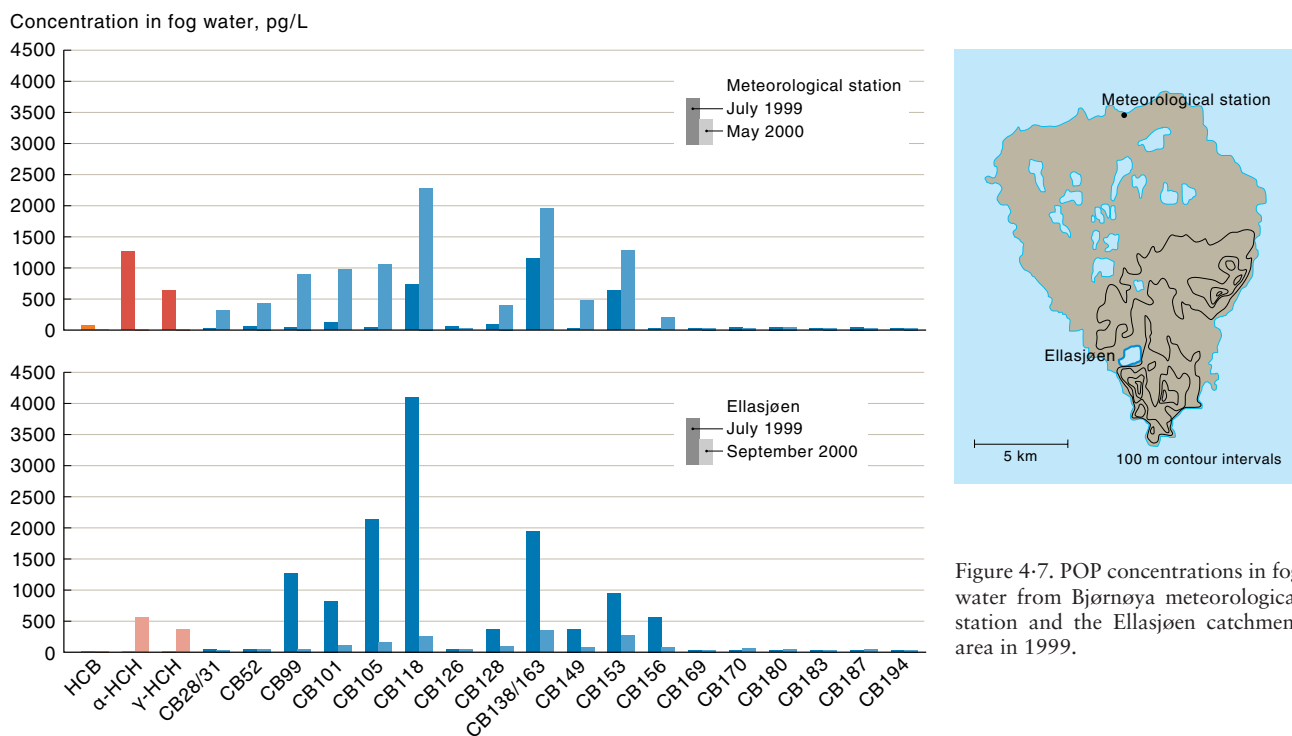


Figure 4-7. POP concentrations in fog water from Bjørnøya meteorological station and the Ellasjøen catchment area in 1999.

POPs were similar in ambient air collected at the Ny-Ålesund station and at Bjørnøya, with HCB dominating in all air samples from both stations. The comparison of the PCB patterns, however, reveals remarkable differences.

PCB concentrations in air samples from Ny-Ålesund are dominated by the lower chlorinated congeners. However, the medium chlorinated PCB congeners dominate the Bjørnøya air samples. Usually, the latter PCB pattern is characteristic for biological samples. The dominance of the medium chlorinated PCBs (CBs 153 and 138) is particularly evident in the sample taken in summer (13-15 July 1999). During this period of the year, the cliffs, mainly south of Ellasjøen, are populated by thousands of nesting seabirds. Therefore, one hypothesis for the unusual POP pattern is that the guano of seabirds redistributes into the air around Bjørnøya, and contributes significantly to the elevated POP air levels at Bjørnøya as the dominating local source during summer time.

Medium chlorinated PCB congeners also predominated in fog water samples collected at Bjørnøya, indicating the strong influence of local biological sources (e.g., seabird guano) on POP patterns (Figure 4-7). The influence of possible biological sources is also supported by the predominance of  $\alpha$ -HCH in Bjørnøya fog. Seasonal differences are even more pronounced in fog samples compared to ambient air, probably due to the proximity of seabird cliffs and hatching areas in the Ellasjøen catchment area. The September sample from Ellasjøen (2000) showed low PCB concentration levels, whereas fog samples collected in July 1999 at the end of the nesting season at Bjørnøya had concentrations of PCBs (e.g., CB118) that were approximately ten times higher.

Comparable seasonal differences could not be found in the fog water samples from the meteorological station (16 km north of Ellasjøen), probably due to the absence of large seabird colonies.

#### 4.1.4. Passive sampler measurements

Airborne concentrations of PCBs and HCB were measured by passive air samplers (semipermeable membrane devices, SPMDs) along a latitudinal transect from the south of the U.K. to the north of Norway during 1998-2000 (Meijer *et al.*, 2003b). This work is part of an ongoing air sampling campaign in which data were previously gathered for 1994-96 (Ockenden *et al.*, 1998). The SPMDs were exposed for two years in Stevenson screen boxes at remote sites in the U.K. and Norway. Sequestered amounts generally decreased between the two sampling periods by a factor 2-5 over four years, suggesting half-lives of 1.7-4 years. Spatial trends of the 1998-2000 data show a decrease in absolute sequestered amounts of the heavier PCBs with increasing latitude/distance from the source area, whereas the lighter PCBs were more equally distributed along the transect (Figure 4-8). However, relative sequestered amounts (expressed as a ratio to PeCBs) show a clear latitudinal trend with the relative contribution of the lighter congeners increasing with increasing latitude, providing evidence of latitudinal fractionation. Absolute amounts of HCB were found to increase with increasing latitude, suggesting that this compound is undergoing cold condensation and global fractionation (Meijer *et al.*, 2003a).

#### 4.1.5. Precipitation

##### 4.1.5.1. Background

It is well known that snow and ice play an important role in the hydrological cycle of the Arctic, but they also play a strong role in the behavior of POPs in the environment by modifying chemical cycling between the atmosphere and the Earth's surface. Snow deposition was studied extensively in the Canadian Arctic in the early 1990s and results were reviewed in the previous AMAP

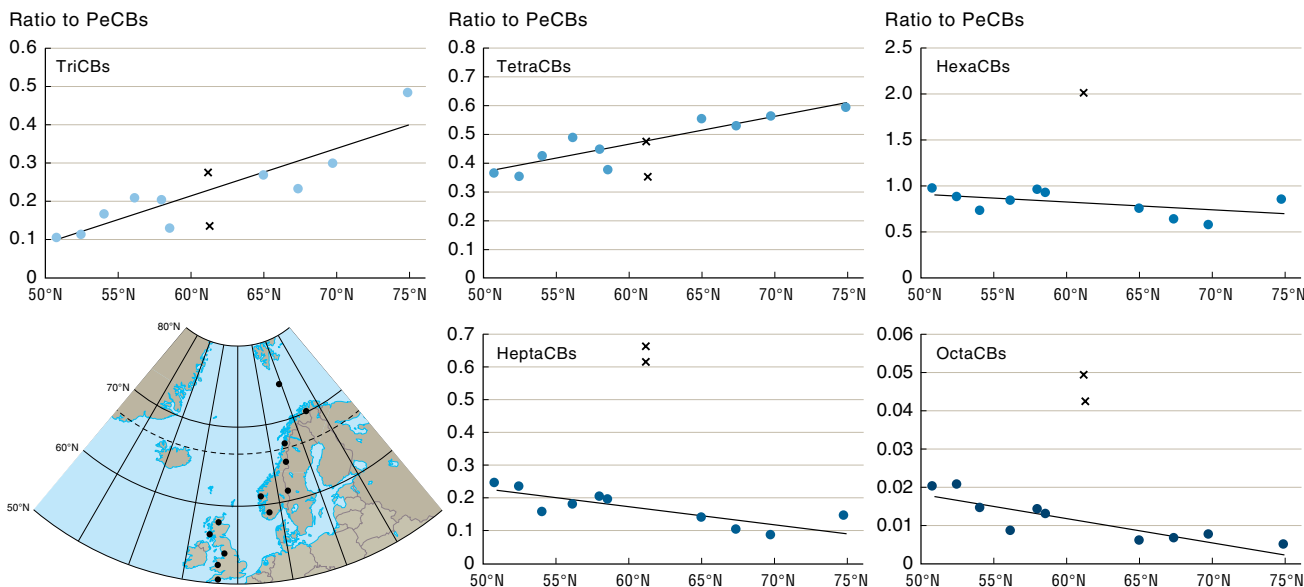


Figure 4-8. Latitudinal trends of PCB homologues in air in northwestern Europe determined using semi-permeable membrane devices (SPMDs) (Meijer *et al.*, 2003b). Results are expressed as ratio to PeCBs. Sites 7 and 8 (crosses) represent outliers omitted from the regression.

POPs assessment (de March *et al.*, 1998) and in Macdonald *et al.* (2000). Studies of POPs in snow on sea ice in the Russian Arctic were also assessed in the previous AMAP assessment report. No additional work, however, appears to have been conducted on POPs in snow in the Canadian or Russian Arctic during the second phase of AMAP. However, sampling and analysis of snow was conducted in northern Alaska, Svalbard and northern Norway.

4.1.5.2. Wet deposition

Bulk deposition of PAHs (31 congeners), PCBs (seven congeners) and HCHs (three congeners) was studied at a site in northern Finland (Pallas and Oulanka) and southern Finland (Evo) in the summers 1993-2001 (Figure 4-9) (Korhonen *et al.*, 1998; Korhonen *et al.*, 2002). In 1993-1994, the sampling in northern Finland was conducted at Oulanka, approximately 100 km south of Pal-

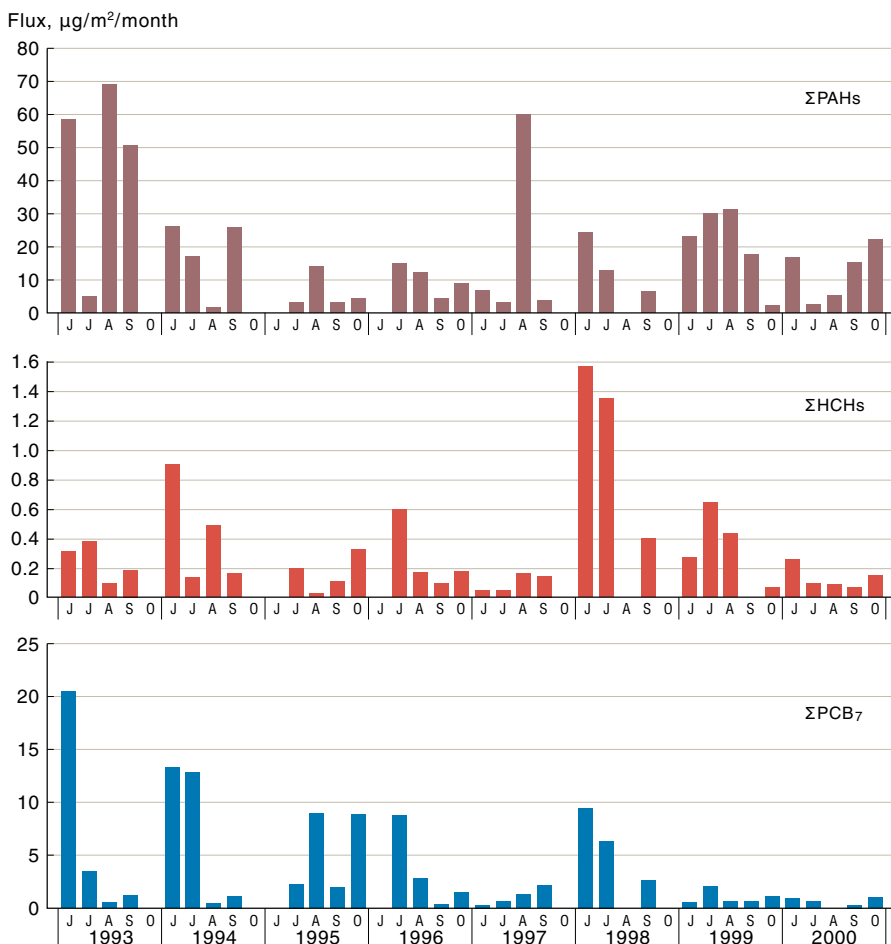


Figure 4-9. Fluxes of PAHs (sum of 31 compounds), ΣHCHs, and total PCB (estimated from analysis of seven congeners) from rainfall at Oulanka (1993-1994) and Pallas (1995-) in northern Finland during the summer months (June-October), 1993-2000.



las. This program represents the longest continuous precipitation monitoring program within the Arctic. Rainfall was collected using a glass funnel bulk sampler (30 cm in diameter) (Korhonen and Kiviranta, 2002). The mean deposition values of PAHs were 27.7  $\mu\text{g}/\text{m}^2/\text{month}$  in southern Finland and 17.9  $\mu\text{g}/\text{m}^2/\text{month}$  in northern Finland. The main PAH components in both southern and northern Finland were phenanthrene, fluoranthene, and pyrene. PCBs averaged 1.6  $\mu\text{g}/\text{m}^2/\text{month}$  in the south and 1.1  $\mu\text{g}/\text{m}^2/\text{month}$  in the north. CBs 153 and 138 predominated among the seven congeners measured. Mean  $\Sigma\text{HCH}$  fluxes were 0.5  $\mu\text{g}/\text{m}^2/\text{month}$  and 0.3  $\mu\text{g}/\text{m}^2/\text{month}$  in the south and north respectively, and  $\gamma\text{-HCH}$  was the predominant isomer reflecting lindane use in the Baltic region (HELCOM, 2001). Overall, the deposition of all chemical groups was about 30% higher in southern Finland than in northern Finland. The temporal trends of PCBs in wet deposition at Pallas are discussed in more detail in Section 5.1.2.

#### 4.1.5.3. Fluxes of POPs in surface snow

Garbarino *et al.* (2002) analyzed snow cores taken over the sea ice from two northwestern Alaska estuaries, Kasegaluk Lagoon, and Admiralty Bay, in 1994-95. They found non-detectable levels of most persistent OC pesticides. However, quantitation limits in the study were high (e.g., 4 ng/L for  $\alpha\text{-HCH}$ ) which precluded detection of most compounds. POPs were measured in surface snows and snow cores (see Section 5.1.3) from Summit (Greenland) and on the Lomonosovfonna and Austfonna glaciers in Svalbard, for 'legacy' OC pesticides and PCBs, by Hermanson *et al.* (2002) and Matthews (2001) (Table 4-3). Lomonosovfonna is the highest ice sheet on Svalbard at approximately 1250 m above sea level (asl). It is about 35 km from Pyramiden, 75 km from Longyearbyen and 110 km from Barentsburg, all coal-mining towns on western Svalbard. Austfonna is approximately 600 m asl, and is located 180 km northeast of Lomonosovfonna. Summit (3230 m asl), the highest point on Greenland, is roughly 1300 km west-southwest of Svalbard.

The PCB profile in near-surface snow layers from Lomonosovfonna is shown in Figure 4-10. The volatility of PCB declines with increasing chlorine substitution (from left to right in the diagram). The homologue distribution of PCBs suggests that the more volatile Di- and TrCBs remain largely in the atmosphere or are revola-

tilized and accumulate in small amounts in snow. The less volatile Te- and PeCB homologues accumulate in relatively large amounts in the upper meter of snow, but significant amounts of them may evaporate back into the atmosphere before final burial, resulting in larger proportions of Hp- and OcCB homologues in the deeper snow layers. The high snow surface percentage of the Te- and PeCB homologues, and the lower percentages of Hp- through NoCB homologues suggests that some post-depositional volatilization may be occurring. This is plausible if the homologue distribution of PCB deposited to Lomonosovfonna has not changed. The overall pattern of homologues at the Lomonosovfonna Ice Cap shows a higher proportion of more highly chlorinated PCBs than observed by Gregor *et al.* (1995) on the Agassiz Ice Cap on Ellesmere Island.

The snow accumulation rate at Austfonna is nearly twice as high as Lomonosovfonna, and is more than twice as high as Summit. Thus, fluxes are the most appropriate way to compare among sites. The proximity of Lomonosovfonna to population centers on Svalbard makes it more likely to accumulate local contaminants than Austfonna, as indicated by the pesticide data. The high flux of  $\Sigma\text{DDTs}$  at Lomonosovfonna, nearly nine times greater than Summit, suggests a significant, possibly local source. Other pesticides ( $\Sigma\text{HCHs}$ , dieldrin, and endosulfan ( $\alpha$ - and  $\beta$ -isomers)) are all higher at Lomonosovfonna than at Austfonna and Summit. Samples from Summit show the highest flux of  $\Sigma\text{PCBs}$ , about 25% greater than Lomonosovfonna. The flux of HCB is the lowest of any observed compound, but is 70% greater at Summit than at Lomonosovfonna. By comparison, surface fluxes for  $\Sigma\text{PCBs}$  on the Agassiz Ice Cap were about 200  $\text{ng}/\text{m}^2/\text{yr}$  in 1992/1993, and ranged from 50 to 650  $\text{ng}/\text{m}^2/\text{yr}$  in lower layers (Gregor *et al.* 1995). PCB deposition at Mould Bay in the Canadian High Arctic ranged from  $400 \pm 300$  to  $600 \pm 500$   $\text{ng}/\text{m}^2/\text{yr}$  in 1990-1991 and 1992-1993, respectively (Franz *et al.*, 1997).

Although flux measurements for PCBs in snow from mainland Norway are not available for comparison, Carrera *et al.* (2001) reported concentrations of  $\Sigma\text{PCBs}$  at Ovre in western Norway ( $63^\circ\text{N}$ ,  $7^\circ\text{E}$ ) of 730  $\text{ng}/\text{L}$ .  $\Sigma\text{DDTs}$  and  $\Sigma\text{HCHs}$  were below detection limits in the same samples. In northern Norway at a site near Tromsø, Enge *et al.* (1998) collected snow in stainless steel lysimeters and reported  $\Sigma\text{PCB}$  (eight congeners) concentrations ranging from 1500 to 3600  $\text{pg}/\text{L}$  sam-

Table 4-3. Concentrations ( $\text{pg}/\text{L}$ ) and fluxes ( $\text{ng}/\text{m}^2/\text{yr}$ ) of chlorinated pesticides and PCBs in Arctic glacial snow from Lomonosovfonna and Austfonna (on Svalbard) and Summit (Greenland). (Matthews, 2001; Hermanson *et al.*, 2002).

Site	Snow accumulation $\text{L}/\text{m}^2/\text{yr}$	Collection year	HCB	$\Sigma\text{HCHs}$	$\Sigma\text{CHLs}$	$\Sigma\text{DDTs}$	Dieldrin	Endo sulfan	$\Sigma\text{PCBs}$
			$\text{pg}/\text{L}$	$\text{pg}/\text{L}$	$\text{pg}/\text{L}$	$\text{pg}/\text{L}$	$\text{pg}/\text{L}$	$\text{pg}/\text{L}$	$\text{pg}/\text{L}$
Lomonosovfonna	340	2000	3.4	169	137	550	87.7	85.2	575
Austfonna	657	1998	n.m.	14.1	n.m.	n.m.	11.4	13.2	n.m.
Summit	293	2000	8.2	36.2	105	83.6	94.2	25.2	994
			$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$	$\text{ng}/\text{m}^2/\text{yr}$
Lomonosovfonna	340	2000	1.2	57.4	46.6	187	29.8	29.0	195
Austfonna	657	1998		9.3			7.5	8.7	
Summit	293	2000	2.4	10.6	30.7	24.5	27.6	7.4	291

n.m. = not measured.

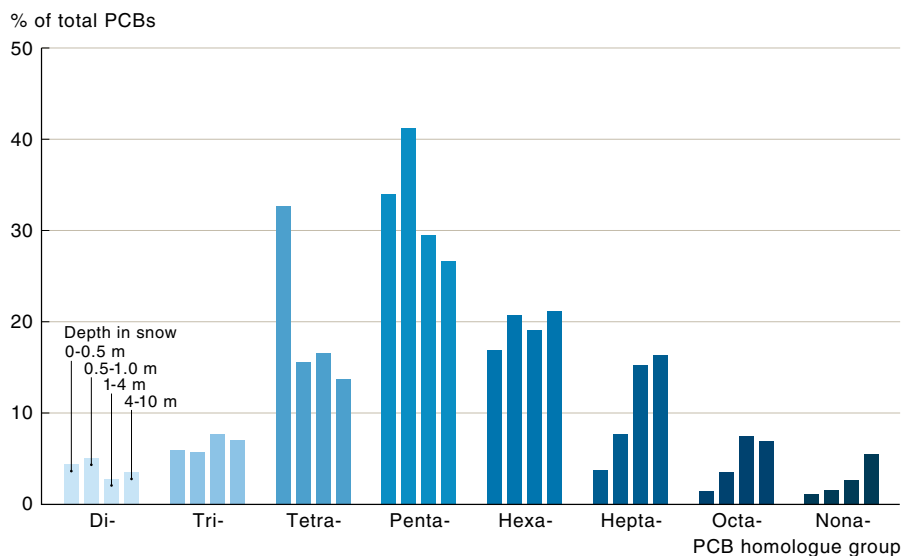


Figure 4.10. PCB homologue distribution at various near-surface depths in snow cores from the Lomonosovfonna glacier on Svalbard (Hermanson *et al.*, 2002).

pled over a three-month period. PCB concentrations in surface snow from the same site were 1040 pg/L, reflecting volatilization from the snow pack.

#### PAHs

Summit, at the centre of the Greenland Ice Cap (3230 m asl), has been the main location for investigations of PAHs in Arctic glacial snow cores. Jaffrezo *et al.* (1994) determined dissolved and particulate bound PAHs in a snow core from Summit, collected in 1991, representing four years of deposition. They observed a strong correlation between sulfate and total PAH in the snow with maxima in winter/spring and summer minima. Total PAH averaged 1360 ng/g (range 600-2370 ng/g) in the combined dissolved and particulate phases. The pattern of PAH compounds in the surface snow indicated that fossil fuel combustion was the major source as well as biomass burning. Major PAH compounds were naphthalene, phenanthrene, fluoranthene, pyrene, benzo[a]pyrene and benzo[g,h,i]perylene. The latter compound comes mainly from combustion of coal and petroleum (Mascelet *et al.*, 1986). Peters *et al.* (1995) analyzed snow collected from the Agassiz Ice Cap in 1993 as part of a study of a snow core. They found naphthalene (NAP) was by far the major PAH present, representing 88% of ΣPAHs (16 compounds). Similar to the observations of Jaffrezo *et al.* (1994), phenanthrene, fluoranthene and pyrene were the major 3- and 4-ring PAHs present. Excluding NAP, ΣPAH concentrations at Agassiz were 19 ng/L, significantly higher than at Summit. Peters *et al.* (1995) estimated an annual deposition of ΣPAHs (16 unsubstituted PAH compounds) to the Canadian Arctic and subarctic of 37 t per year in the 1980-1990s, based on a deposition flux of 11 μg/m<sup>2</sup>/yr. Masclet *et al.* (2000) found PAH concentrations of 1.2 ng/g in particulate matter filtered from melted snow (5-12 L volumes) in surface snows from the Greenland Ice Cap. The PAH levels had pronounced seasonal trends coinciding with Arctic haze events (see Section 5.1.3.2). Measurements of PAHs were performed on a 122 m ice core record from the Lomonosovfonna glacier on Svalbard, but only NAP was detectable (Vehviläinen *et al.*, 2002). This was attributed to the very small (average 56 g) sample sizes. The results suggested that prior to the 1930s, NAP concentrations were below the detection

limit, but increased until the 1980s. In general, NAP concentrations (5000-53 000 ng/g) at Lomonosovfonna were six times lower than in the Agassiz Ice Cap (Peters *et al.*, 1995), but are about fifty times higher than in Greenland (Jaffrezo *et al.*, 1994).

#### 4.1.5.4. 'New' chemicals in snow and ice

Garbarino *et al.* (2002) reported detection of the insecticide chlorpyrifos in snow from two northwestern Alaska estuaries at concentrations ranging from <10 to 80 ng/L as well as the herbicide dacthal (detectable near the quantitation limit of 4 ng/L). Chernyak *et al.* (1996) had previously reported chlorpyrifos at 0.17 ng/L in melted sea ice samples from the Bering Sea. Dacthal has not been reported previously in Arctic samples but has been shown to be transported regionally in North America (Rawn *et al.* 1999).

Laniewski *et al.* (1998) analyzed ice samples from the Mårma glacier in the Swedish Arctic (68°10'N, 18°40'E) and detected a number of chlorinated organics not previously reported in snow or wet deposition in the Arctic. The tri(chloroalkyl)phosphates (TCAPs) were present in the ice sample from the Mårma glacier and snow from southern Sweden and Poland, but were not present in ice from Queen Maud's Land in Antarctica. The isomers detected were tris(2-chloroethyl)phosphate, tris(1-chloro-2-propyl)phosphate, and bis(1-chloro-2-propyl)(3-chloro-1-propyl)phosphate. These compounds are semivolatile (vapor pressures 1-10 Pa at 25°C; (WHO/IPCS, 1998)) and have come into increasing use during the 1990s for use as flame retardants in plastics. Also detected in the glacier were dichlorobenzenes and tetrachlorobenzene (Laniewski *et al.*, 1998).

#### 4.1.5.5. Snow and rain deposition at Bjørnøya

Snow and meltwater were collected in 1999/2000 in order to assess transport and deposition pathways of POPs within the Bjørnøya ecosystem (Evenset *et al.*, 2002; Kallenborn, 2002a). This was in connection with the measurement of POPs in ambient air and fog (see Section 4.1.3) in a study addressing high POP levels in sediment and Arctic char. During a first, preliminary sampling campaign in 1999/2000, two meltwater samples and one deposition snow sample were collected for

Table 4-4. POP levels in snow deposition and meltwater samples from Bjørnøya (pg/L)<sup>a</sup>

Sampling Period	Meteorological station			Ellasjøen					
	Sample type	ΣPCBs	ΣHCHs	HCB	Sampling period	Sample type	ΣPCBs	ΣHCHs	HCB
May 2000	Snow	7220	78	2	Summer 2000	Snow	3200	32	1
July-Sept. 2000	Meltwater 1 <sup>b</sup>	1180	490	i.c.	June-Sept. 2000	Meltwater 4 <sup>b</sup>	391	921	i.c.
Sept.-Nov. 2000	Meltwater 2 <sup>b</sup>	845	1800	i.c.	July 1999	Meltwater 3 <sup>b</sup>	3740	740	n.a.

<sup>a</sup> ΣPCBs = 17 congeners, ΣHCHs = α- and γ-HCH, HCB = hexachlorobenzene.

<sup>b</sup> During the summer period of the year, a substantial amount of rain deposition was collected in addition to snow. i.c. = interference in the chromatogram, n.a. = not analyzed.

POP analyses at the meteorological station. At the lake Ellasjøen, two meltwater and three snow deposition samples were quantified. Meltwater was sampled at the meteorological station and Ellasjøen using a 1-m<sup>2</sup> meltwater collector. Surface snow was collected over a long period of time (1-2 months) and allowed to melt under natural conditions. Thus, the meltwater sample represented an integrated sample over a long period of time. The POP pattern of the meltwater sample was influenced by remobilization and volatilization processes.

PCBs were the dominant OC contaminants in snow (Table 4-4), while HCHs were the main contributors for the chlorinated pesticides in snow and meltwater from Bjørnøya. In two meltwater samples (Meltwater 2 and Meltwater 4), HCH was the predominant compound class (Table 4-4). Due to relatively high volatility, HCB probably re-volatilizes into the atmosphere and therefore dominated in Bjørnøya ambient air samples but not in meltwater and snow. The highest PCB concentrations were found for snow deposition, collected close to the meteorological station.

These findings indicate characteristic differences found for the distribution of HCHs and PCBs in snow deposition and meltwater. In all snow deposition samples, only minor contribution from HCHs was found. However, the contribution of HCH to the overall contamination is significantly higher in meltwater samples (Figure 4-11). This may partly be caused by re-evaporation of lower chlorinated PCBs into the atmosphere and the parallel continuous uptake of water soluble α- and γ-HCH from the atmosphere. However, the number of samples analyzed is too small to draw final conclusions about this hypothesis regarding snow surface/atmosphere exchange processes. It should be noted that during the summer season, both rain and snow deposition were collected and therefore, a direct correlation between meltwater properties and POP contamination is difficult.

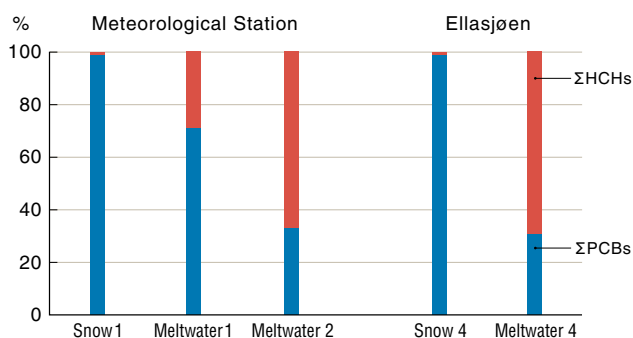


Figure 4-11. Percentage distribution of ΣHCHs and ΣPCBs in snow and meltwater samples from Bjørnøya (Bear Island).

The same analytical method was used for meltwater samples collected at the valley Dividalen in Norway and at Bjørnøya (Ellasjøen and meteorological station). Concentrations of ΣHCHs and ΣPCBs in Bjørnøya meltwater were comparable with those from the Norwegian mainland site. ΣPCB levels of 3600 and 1500 pg/L were found in two meltwater samples from Dividalen. The two meltwater samples taken at the Bjørnøya meteorological station had slightly lower levels of PCBs than the Ellasjøen and the Dividalen samples. No distinct difference in the PCB levels between the Bjørnøya samples and the meltwater taken at the Norwegian mainland was found. This is a rather surprising result, since no local PCB source is known at Bjørnøya, whereas possible sources close to the Dividalen sampling site are known. The extent to which biological sources such as seabird guano, influence the PCB levels, patterns, and distribution of other POPs in snow remains to be determined, however.

#### 4.1.6. Summary and conclusions – air and precipitation

Measurements of POPs in Arctic air have continued on a weekly basis at locations in Canada, Iceland, Norway, Finland, and Russia. A large temporal-trend dataset is being developed as discussed in Section 5.1. With six stations reporting results during the 1990s (although not all operational in all years), it is possible to examine geographical differences in concentrations of major OCs and PAHs among sites and to compare these with results from monitoring stations in southern Canada/U.S. and Sweden. At three stations, results were also available for some current-use pesticides and flame retardants.

In this assessment, spatial comparisons were limited to a relatively small suite of OCs that have been measured at all stations and reported to the AMAP data center. Thus, only ΣPCB<sub>10</sub> congeners have been measured at all stations, although data are available for up to 102 congeners at some sites. The reporting of ΣPCB<sub>10</sub> underestimates total PCBs in air by four to five times because mono- and dichlorobiphenyls, which predominate in Arctic air, are underrepresented in the list.

Results for ΣHCHs, chlordane, and DDT-related compounds suggest uniformly low concentrations of these pesticides in Arctic air during the mid- to late 1990s. This is expected because, with the exception of γ-HCH, all others have been banned for at least ten years or more in circumpolar countries, although there may be some use of DDT continuing in Russia (see AMAP, 2003). The concentrations of these three OC pesticides in Arctic air were lower than in air at a rural site in the Great

Table 4-5. Comparison of average concentrations of persistent OCs and PAHs in the Great Lakes and at several Arctic locations for approximately the same time period. Concentrations (pg/m<sup>3</sup>) represent combined gas and particulate (filter) phases.

	Great Lakes rural <sup>a,c</sup> 1996-98		Great Lakes urban <sup>b,c</sup> 1996-98		Alert 1996-98		Ny-Ålesund 1996-98		Stórhöfði 1996-98		Amderma 1999-2000
	Mean	SE	Mean	SE	Mean	SE	Mean	SE	Mean	SE	Mean
ΣPAHs	1160	100	113000	15500	274	34.3	2750	320			
ΣHCHs	97	7.2	131	10	37.2	3.1	55.7	2.8	22.2	1.3	
ΣCHLs	9.4	1.1	151	15	2.0	0.2	1.9	0.1	0.5	0.01	
ΣDDTs	4.5	0.66	80.2	7.9	0.8	0.1	2.1	0.2	2.6	1.2	
HCB	68	2	110	6	40.8	3.2	94.0	6.6	9.8	1.2	
ΣPCBs <sup>d</sup>	63	6	1800	170	7.3	0.6	125.0	35.9	67.4	19.1	4.1

<sup>a</sup> Eagle Harbor – western Lake Superior.

<sup>b</sup> Chicago (downtown).

<sup>c</sup> In the Great Lakes samples, HCH represents the sum of  $\alpha$ - and  $\gamma$ -HCH. ΣCHLs represents the sum of *cis*- and *trans*-chlordane and *trans*-nonachlor. ΣDDTs represents the sum of the *p,p'*-isomers of DDT and its two metabolites, DDE and DDD.

<sup>d</sup> Sum of 100 PCB congeners in Great Lakes samples and ΣPCB<sub>10</sub>×4 for Alert, Ny-Ålesund, Stórhöfði, and Amderma.

Lakes region near the shore of Lake Superior (Eagle Harbor) during the same time period (Table 4-5). However, the differences between this temperate North American site and the Arctic stations were less than a factor of 2 for ΣDDTs at Ny-Ålesund, Stórhöfði, and Amderma.

Concentrations of PCBs and HCB at Ny-Ålesund are higher than in rural air in the Great Lakes (Buehler *et al.*, 2001). All other Arctic sites have lower average levels than the rural Great Lakes. The air sampling building at Ny-Ålesund was replaced in 1998 (completed by August 1998) and the elevated CB28 and 31 concentrations observed during the remainder of 1998 may be related to contamination during startup of the new location, although similar concentrations were found in 1999 and 2000 (see Section 5.1). Urban air (Chicago) has much higher concentrations of PCBs and PAHs than any Arctic sites as expected (Table 4-5). Combined with results from Dunai and Stórhöfði, where ΣPCB concentrations were also higher than at Alert and in the upper Great Lakes, the results suggest that, during the period of 1996-1998, the European Arctic continued to receive air with elevated PCB concentrations compared to the North American Arctic.

Although there are fewer data for PAH, higher PAH levels are seen at Dunai and Svalbard than at Alert, Tagish (Halsall *et al.*, 1997) or Lake Superior (Buehler *et al.*, 2001), which also suggests that the European Arctic is receiving elevated atmospheric inputs in comparison to the western Canadian and High Arctic archipelago. Further assessment of the importance of European and particularly Russian sources of PAHs for air concentrations over the Barents Sea awaits more complete datasets from the station at Amderma in northwestern Russia and Pallas in northern Finland.

Concentrations of pesticides and PCBs in Table 4-5 can also be compared with those found at Tenerife (Canary Islands) (28°N, 16°W) during 1999-2000 (Van Drooge *et al.*, 2002). Samples were taken in the free troposphere at 2367 m asl altitude and near sea level at 47 m asl. Concentrations of all substances were higher within the marine boundary layer than at high altitude, except for HCB, which appears to be very well mixed throughout the troposphere. The free tropospheric concentrations were similar to those found at Arctic stations for HCB (51 pg/m<sup>3</sup>), ΣHCHs (17 pg/m<sup>3</sup>) and ΣDDTs (5 pg/m<sup>3</sup>). ΣPCB concentrations (sum of 19 congeners) at

Tenerife were similar to levels measured at Ny-Ålesund and Stórhöfði (78 pg/m<sup>3</sup> at 2367 m and 190 pg/m<sup>3</sup> at sea level), taking into account the difference in the number of congeners.

The past five years has seen a major increase in the number of halogenated organic chemicals detected in Arctic air. Among the current-use pesticides, the presence of trifluralin is surprising because of its short atmospheric half-life. Trifluralin is a common air contaminant in agricultural areas of the U.S. and Canada and is present at ng/m<sup>3</sup> concentrations during application periods in May-June (Grover *et al.*, 1988; Hoff *et al.*, 1992; Majewski *et al.*, 1998). Thus, the high volume of trifluralin use results in detectable levels of this pesticide, and possibly others such as methoxychlor, even though removal processes in the atmosphere are quite rapid. The presence of similar levels of trifluralin at Dunai implies Asian and Russian uses of this pesticide.

The presence of PCNs in air at Alert and over the Barents/Kara Seas found by Harner *et al.* (1998) is an important new finding. The PCNs contribute significantly to total TEQs in air and are correlated with PAHs, suggesting that they are combustion by-products associated with Arctic haze. The Arctic haze association is important because the annual PCN profiles seem to be meteorologically driven, with maxima in winter. A full evaluation of their importance requires concurrent measurements of PCDD/Fs as well as coplanar PCBs. These compounds have not yet been routinely measured and to do so may require modification of sample extraction methodology to optimize their recovery from the particulate phase, as well as increased air volumes for some sampling stations.

A pilot study conducted at Alert revealed that although levels of PCDD/Fs found in air were low, there are still possibilities of significant accumulation of these compounds in other Arctic environmental media due to the low temperatures in the Arctic and slower degradation processes. The PCDD/Fs also have to be considered in the context of coplanar PCBs and PCNs. Further measurements of PCDD/Fs are needed in order to gain more insight into the fate of these compounds in the Arctic given predictions of significant deposition in Nunavut. A report by Commoner *et al.* (2000) has focused attention on the current levels, pathways, and sources of PCDDs and PCDFs in the Canadian Arctic.



In their study, Commoner *et al.* (2000) predicted dioxin TEQ deposition of about 4 to 53 pg TEQ/m<sup>2</sup>/yr to terrestrial surfaces near eight communities in Nunavut. Highest deposition in the south and east was predicted because of the preponderance of dioxin sources in the eastern U.S. and Canada. Russian and northern European sources were regarded as insignificant for Nunavut in comparison to those in the mid-west U.S. and Ontario and Quebec (Commoner *et al.*, 2000).

The presence of PBDEs in Arctic air has been demonstrated at Alert and Tagish, as well as Dunai in the Russian Arctic and at Bjørnøya in the European Arctic. The elevated levels of PBDEs found at Alert and Tagish must be viewed with caution given that they are far higher than levels found in the Great Lakes region, including within the city of Chicago, and higher than levels found at Bjørnøya during winter time. The results for PBDEs illustrate a general problem with all air sampling for widely used products such as flame retardants, surfactants, and plasticizers (including chlorinated paraffins, perfluorinated acids, and phthalates), which is that sampling media or emissions from building materials or use of materials near the site could inadvertently contaminate the samples. Further work is urgently needed to confirm the levels of PBDEs in air given their increasing presence in Arctic biota (Section 5.4.6.1).

Toxaphene, a major contaminant in Arctic biota, which has also been widely measured in Arctic seawater during the period of 1996-2001, remains relatively unstudied in Arctic air. No new measurements have been reported from any of the monitoring stations. Given the importance of this contaminant in both the North American and European Arctic, additional data are needed in order to examine levels and sources.

The very limited data for PCBs and OC pesticides in snow and precipitation, with the exception of detailed studies at Bjørnøya, illustrates a key knowledge gap about the fate of airborne OCs. For example, the proportion of the contaminants measured in air that is actually entering the snow or soil environment and then re-volatilized is not known. A full understanding of the pathways is essential for predicting future trends and also for evaluating other chemicals that have been detected in Arctic air.

The concentrations of PCBs in snow at Bjørnøya were similar to those in northern Norway (Enge *et al.*, 1998) and much higher than on the Lomonosovfonna and Austfonna glaciers in Svalbard or at Summit, on the Greenland Ice Cap. Air concentrations of PCBs were also higher at Bjørnøya than at Ny-Ålesund. These results suggest a strong influence of local biological sources (e.g., seabird guano) on the POP patterns. This is a surprising new finding which may also be the case on other islands colonized by seabirds.

## 4.2. Terrestrial environment

Levels of OCs have been shown to be generally lower in the terrestrial Arctic ecosystem than in the aquatic ecosystem (de March *et al.*, 1998). For this reason, the amount of research focused on, and the available data for, the terrestrial Arctic is much more limited, both historically (see Thomas *et al.*, 1992; de March *et al.*, 1998) and in recent years. A number of terrestrial mammals

and birds are however important as food species (e.g., caribou), and levels of OCs could be an issue for human consumption. Temporal trends in birds of prey remain an important data set but temporal trend studies in other species, such as lichen or caribou, have not been carried out or continued. More volatile, less bioaccumulative OCs, in particular HCH, are the most common individual compounds found in soil and lower trophic-level terrestrial organisms. At higher trophic levels, such as in wolf and birds of prey, the more bioaccumulative OCs, in particular DDTs and PCBs, become more prevalent.

### 4.2.1. Soils and plants

There has been a significant amount of new work on concentrations of POPs in soils and plants since the previous AMAP POPs assessment. Global surveys of PCDD/Fs in soils and tree bark by Wagrowski and Hites (2000), and PCBs in soils (Ockenden *et al.*, 2002), have included Arctic sites. An extensive survey of OCs in soil and plants in Russia has also been carried out (RAIPON/AMAP/GEF Project, 2001). This latter study is important because the Russian Arctic was previously identified as a knowledge and data gap for assessment of circumpolar trends of OCs. A project that examined levels of OCs in vegetation and soil from Alaska and Siberia during the years 1991-1993 (Ford *et al.*, 2000) that was not available for the first AMAP report is also considered.

#### 4.2.1.1. PCBs and OC pesticides in vegetation

Samples of vegetation were collected from four regions of the Russian Arctic: Kola Peninsula; Pechora Basin; Taymir Peninsula (Dudinka and Khatanga); and Chukotka (Kanchalan and Lavrentiya) in 2000/2001 for analysis of OCs (RAIPON/AMAP/GEF Project, 2001). The four regions cover a wide geographical area (Figure 4-12). Pooled samples of three types of vegetation were collected, including berries, lichens, and mosses; the species included for each type of vegetation varied with the region (see Annex Table 4 for species). PCBs were the predominant OC in vegetation, followed by DDT, HCH, and chlorobenzenes. Toxaphene (sum of Parlars 26, 52, and 60; detection limits approximately 0.2 ng/g in biota) and PBDEs (detection limits approximately 0.5 ng/g in biota) were not detected in any vegetation samples, but the detection limits of this study are above the levels normally found for these compounds in vegetation in the Arctic.

Across all regions, OC concentrations were greatest in mosses, followed by lichens and then berries, which had very low concentrations. In general, strong geographical trends in OC concentrations were not observed between regions for any of the three vegetation groups (Figure 4-12 and Annex Table 4). PCB concentrations (3.1-3.9 ng/g dw) in lichens are in the same range reported previously for lichens in the Russian Arctic, collected in 1994 (Melnikov *et al.*, 1995), but are an order of magnitude higher than levels reported for lichen from the Canadian Arctic in 1992-1993 (de March *et al.*, 1998). HCHs were the only OC group with higher concentrations in Canadian Arctic lichens compared to the Russian Arctic. The fact that the Canadian lichens were collected in the early 1990s, and that ΣHCH levels have

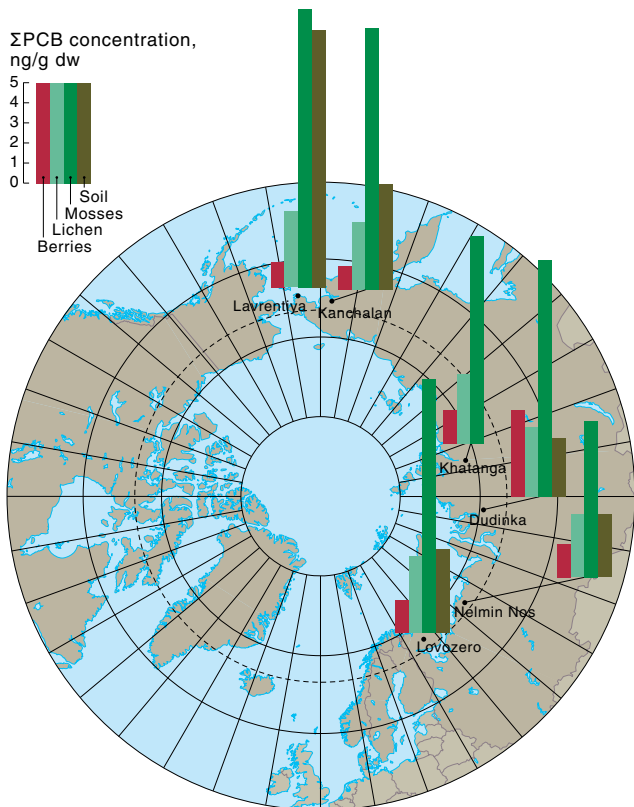


Figure 4-12. Concentrations of ΣPCBs in berries, lichens, mosses, and soils from regions in Arctic Russia (RAIPON/AMAP/GEF Project, 2001). All samples are pools of numerous samples collected in 2000-2001. Species included in each vegetation group are given in Annex Table 4.

decreased in the atmospheric environment may explain part of this trend.

In an older study, a total of 209 vegetation samples (11 species across all sites) were collected from Alaska and Russia in 1991-93 and analyzed for 51 OC pesti-

cides, industrial compounds, and PCB congeners (see Figure 4-13 for locations, and Annex Table 4 for species names and concentrations) (Ford *et al.*, 2000). Vegetation samples were primarily two species of ground-dwelling lichens (*Masonhalea richardsonii* and *Cetraria cucullata*) and two mosses (*Racomitrium lanuginosum* and *Hylocomium splendens*). All four of these species were collected in Alaska but only *C. cucullata* was commonly found at the Russian study sites. Samples of other taxa, most notably blueberries (*Vaccinium uliginosum*), were also taken opportunistically and predominantly in Alaska. Concentrations of OCs in lichens and mosses from all Alaskan and Russian locations were low (low ng/g dw) and in the range reported for Russian lichens, mosses and berries recently collected and discussed above (RAIPON/AMAP/GEF Project, 2001), and previously for Arctic plants (de March *et al.*, 1998).

Analysis of the Alaskan data set demonstrated that concentrations for commonly encountered OCs were generally highest in the lichen *C. cucullata*, followed by the other lichens and mosses (Annex Table 4). This differs from the results of the recent Russian study where the highest OC concentrations were found in mosses. Concentrations of OCs in other Alaskan vegetation, such as blueberries, were very low, and for many species OCs were not detected, consistent with the Russian study.  $\alpha$ -HCH and PeCA were the individual OCs with the highest concentrations; ΣPCB concentrations were the highest among OC groups.

OC concentrations in *C. cucullata* collected on the Kola Peninsula/northern Urals (where sampling sites were close to industrial centers) were similar to those measured in the same species from Alaska and Taymir (Figure 4-13). However, PCB concentrations in *C. cucullata* from the Taymir Peninsula were approximately twice as high as those observed at other sites. This is

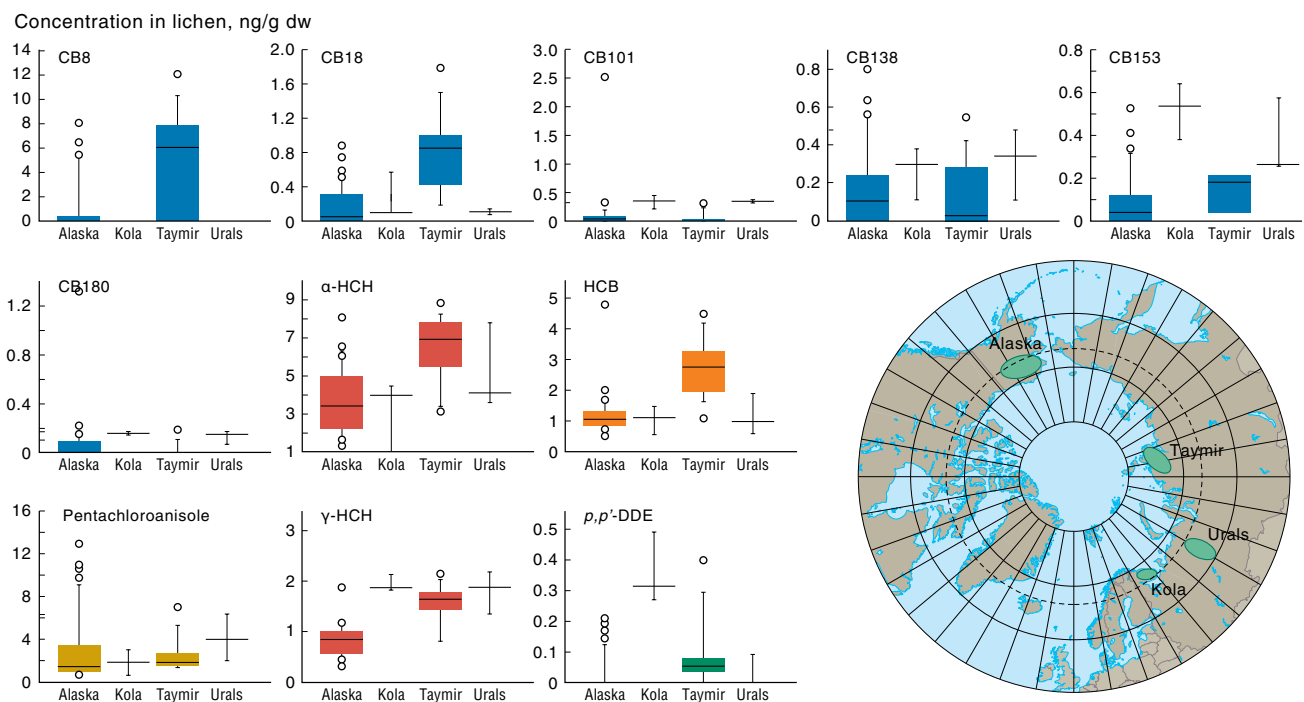


Figure 4-13. Areas where lichens, mosses and soils were sampled in Alaska and Russia between 1991 and 1993, and concentrations of selected OCs in lichen (*Cetraria cucullata*) from Alaskan and Russian locations (Ford *et al.*, 2000).

due, in part, to much higher concentrations of lower chlorinated congeners, in particular CB8. Concentrations of higher chlorinated congeners show less spatial differences, which is consistent with the recent Russian study (RAIPON/AMAP/GEF Project, 2001).

Comparison of individual OC concentrations in *C. cucullata* from the Kola Peninsula/northern Urals, with regionally dispersed samples from Alaska and the Taymir Peninsula, show a diversity of patterns (Figure 4-13). Several analytes (aldrin, endrin, heptachlor and mirex) were not detected in Russian samples but sporadically encountered in Alaskan samples. Lower chlorinated PCBs (CBs 8 and 18) and semi-volatile OCs ( $\alpha$ -HCH, HCB) were present in higher concentrations on the Taymir Peninsula (Figure 4-13). Taymir is considerably further north than the sites in the other regions and, this finding is consistent with the predicted global fractionation of semi-volatile persistent organics (Wania and Mackay, 1996). In contrast, higher chlorinated PCBs and several pesticides were present at higher concentrations in samples from Kola/Urals sites than in those from Alaskan and Taymir sites (Figure 4-13). No regional differences were evident for PeCA, *cis*-nonachlor, and CBs 29, 187, and 206 (although outliers were present for some of these analytes in a few Alaskan samples).

#### 4.2.1.2. PCDD/Fs in tree bark

Tree bark was used by Wagrowski and Hites (2000) to study the global distribution of PCDD/Fs. The bark samples (collected at 1 m height) were from locally important tree species from sites in Alaska (1 site), Yukon/Northwest Territories (7 sites) and Norway (1 site), along with samples from 54 other sites in the northern and southern hemisphere. Concentrations of total PCDD/Fs in bark were low in the Arctic, ranging from not-detected (approximately 1 ng/g lw) to 685 pg/g lw with a median of 131 pg/g lw (Annex Table 4). No geographical differences were discernible among the Arctic samples. These concentrations were, however, at the low end of the range found globally in tree bark from temperate zones (approximately 100-227 000 pg/g lw). Most tree bark samples from the Arctic were characterized by high proportions of tetrachlorodibenzofurans. Octachlorodibenzo-*p*-dioxin, which predominated in Arctic soils, was near detection limits in bark. This pattern is consistent with the bark absorbing primarily lower chlorinated PCDD/Fs from the gas phase in areas remote from major sources.

#### 4.2.1.3. OC pesticides and PCBs in soils

##### *Arctic soils in a global survey*

Ockenden *et al.* (2002) conducted a global survey of soils from rural and remote locations, which included 22 sites in the Arctic (Canada, eastern Greenland, northern Norway, and northwestern Russia). Samples (0-5 cm depth) were collected from 208 undisturbed sites throughout the world. Sites were more than 2 km from the nearest town/city/busy road and more than 500 m from small dwellings or tracks. The global latitudinal distribution of PCBs in soils is plotted in Figure 2-4. The Arctic soil results are presented in Annex Table 4.  $\Sigma$ PCB

concentrations (27 congeners) in the 22 Arctic samples ranged from 0.004 to 48 ng/g dw with highest concentrations in or near urban areas in northwestern Russia (Monchegorsk) and lowest concentrations in Greenland. After adjusting for soil organic matter (SOM), the range of concentrations was somewhat narrower (0.1-81 ng/g SOM). Ockenden *et al.* (2002) concluded that the SOM content and turnover governs PCB behavior because of their strong affinity for organic carbon. Degradation of POPs in soil is slow, of the order of tens of years (or more) for many compounds (Mackay *et al.*, 2000). As such, physical processes, bioturbation/ploughing, and carbon burial/sequestration in forest soils/peat become key factors. These processes, together with physical occlusion/partitioning into the SOM (Luthy *et al.*, 1997), physically remove the bulk of the atmospherically-derived POPs available for air-surface exchange by taking it below the all-important surface 'skin' of soil which is in active exchange with the atmosphere (Harner *et al.*, 2001).

##### *Russian and Alaskan soils*

Samples of surface soil were collected from four regions of the Russian Arctic: Kola Peninsula; Pechora Basin; Taymir Peninsula (Dudinka); and, Chukotka (Kanchalan and Lavrentiya), in 2000-2001 for the analysis of OCs (RAIPON/AMAP/GEF Project, 2001). Samples were collected in the same locations as plants and terrestrial mammals, and covered a wide geographical area. Each sample analyzed comprised a pool of the samples collected. PCBs were the dominant OC measured in soils at all locations. Similar to results for vegetation from these regions, there were no strong geographical trends in OC concentrations in Russian soils (Figure 4-12), and OC concentrations were within a factor of two (Annex Table 4). A single soil sample from Lavrentiya in Chukotka had higher concentrations of  $\Sigma$ PCBs (5.2  $\pm$  3.5 ng/g dw) but, since this is a single sample, caution is warranted about interpreting this as a hot spot. PCB concentrations reported by the RAIPON/AMAP/GEF Project (2001) were generally lower than reported by Ockenden *et al.* (2002); however, the latter soils were generally collected close to urban areas while the former study focused on background levels in remote areas. Concentrations of the OCs are in the range previously reported for Russian soils (Melnikov *et al.*, 1995). Toxaphene (sum of Parlars 26, 52 and 60, detection limits approximately 0.1 ng/g in soil) was not detected in any soil sample, but low concentrations (0.16 to 0.23 ng/g dw) of PBDEs were observed in three soil samples.

In an earlier study, concentrations of OCs in the surface soils of Alaska and Russia, collected between 1991 and 1993, were low (low ng/g dw) (Annex Table 4) (Ford *et al.*, 2000) and in the range of levels reported for Arctic soil in the first AMAP assessment report (de March *et al.*, 1998).  $\Sigma$ PCBs and  $\Sigma$ DDTs were the predominant OCs found, with concentrations of  $\Sigma$ CBz and  $\Sigma$ HCHs being similar to  $\Sigma$ PCB and  $\Sigma$ DDT concentrations in the Alaskan soil only. PeCA, CB28, HCB, and  $\alpha$ -HCH were among the more common individual OCs found. Concentrations were much higher in Alaska than on Kola and Taymir Peninsulas in Russia. However, the Alaskan and Taymir samples included both surface vegetation and litter, whereas Kola soils only included the



A-horizons (no vegetation or litter), making comparisons with the Kola samples difficult. OC concentrations in the Alaskan soils are similar, but the older Taymir samples have much lower concentrations than the recently collected Russian soils (RAIPON/AMAP/GEF Project, 2001). Differences in the collection methods and the organic content of the soils may explain some of the differences between the Russian data, but this information is not currently available.

Concentrations of individual OCs in the Kola Peninsula samples, which had no vegetation or litter, rarely exceeded 1 ng/g dw. Due to the removal of vegetation and litter, it is not possible to compare these results with recent results from Russian soil samples. Concentrations of *p,p'*-DDT in Kola soils were one to two orders of magnitude higher than most other analytes, suggesting recent DDT use in this area at the time of collection (1991-1993).

Specific information on A-horizon soils is not available for Alaska and the Taymir Peninsula, although several soil cores from both those regions were taken. When A-horizon and 0-5 cm increment samples are collectively compared to the results for *C. cucullata* lichen, some results were similar and some surprisingly dissimilar. As with the lichen results, endrin, heptachlor, and mirex were found only in Alaskan soils. Surprisingly, concentrations of HCB,  $\alpha$ -HCH and CB18 were quite low in the Taymir soil relative to the Kola and even Alaskan soils, although concentrations of CB8 were high. Concentrations of PeCA were high only in Taymir and Alaska, where they reached concentrations that exceeded those of most other analytes by one to two orders of magnitude. Also as with the lichens, the Kola signals for *p,p'*-DDE and CBs 101 and 138 were high relative to Alaska and Taymir. Five of the six DDTs (except *o,p'*-DDE), as well as CB153, were routinely found in Kola/Urals soils, but not in Alaska or Taymir soils.

Inorganic, cryogenically exposed surface soils were collected only in Alaska and the Taymir, and POPs concentrations rarely exceeded 1 ng/g dw in this matrix. Where QA considerations permitted comparison, Alaskan samples of surface inorganic soils had higher analyte concentrations than those from the Taymir, which is opposite to the pattern found for heavy metals and trace elements (Ford *et al.*, 2002).

#### 4.2.1.4. PCDD/Fs in Arctic soils

As part of a global survey of PCDD/Fs in soils and tree bark, Wagrowski and Hites (2000) reported PCDD/Fs in soils from Alaska, the Yukon and NWT in the Canadian Arctic, western Greenland, and Norway. For this assessment, these data were combined with results from Brzuzy and Hites (1996) who conducted an earlier, smaller-scale soil survey that included samples in Alaska and northwestern Russia (Annex Table 4). Fluxes of total PCDD/Fs estimated from the mass of chemicals in a 10×10 cm (15 cm depth) core ranged from 1.2 to 143 ng/m<sup>2</sup>/yr. There was no consistent geographical trend in the 12 samples; however, higher fluxes were seen in samples from Alaska and western Greenland. By comparison, the range of fluxes of total PCDD/Fs for sites in the mid-latitudes and tropics was 5-8100 ng/m<sup>2</sup>/yr. The authors concluded that PCDD/Fs did not

move appreciably from warm to cold latitudes. Most Arctic soil samples were characterized by high proportions of octachlorodibenzo-*p*-dioxin, although tetrachlorodibenzofurans were also important contributors to total PCDD/Fs.

Using the PCDD/F emission inventory for Canada and the U.S., Commoner *et al.* (2000) predicted 'dioxin' TEQ deposition of about 4-53 pg TEQ/m<sup>2</sup>/yr for terrestrial surfaces near eight communities in Nunavut. Highest deposition was predicted for the most southerly location (Sanikiluaq, 53 TEQ/m<sup>2</sup>/yr). Eastern locations such as Broughton Island (9 TEQ/m<sup>2</sup>/yr) had higher deposition than western locations (Ikaluktutiak, 4 TEQ/m<sup>2</sup> year). Highest depositions were predicted to occur in the southern and eastern Arctic because of the preponderance of dioxin sources in the eastern U.S. and Canada. Russian and northern European sources were regarded as insignificant in comparison to those in the mid-west U.S., Ontario, and Quebec (Commoner *et al.*, 2000). A direct comparison with the PCDD/F results from Wagrowski and Hites (2000) was not possible because of differences in geographical area and lack of data for specific 2,3,7,8-substituted PCDD/Fs in the soil survey.

#### 4.2.1.5. Simulation of the global fate of PCBs in soils

The Globo-POP model has been used to describe the global fate of PCBs over a time scale of several decades (Wania *et al.*, 1999c; 2000). PCBs have been used as mixtures consisting of individual substances which differ substantially in their physical-chemical characteristics and persistence. It is also likely that the temporal and spatial pattern of release into the environment has been different for different congeners (Breivik *et al.*, 2002b). Large differences in the simulated fate of the various PCB congeners reaffirm the need to perform calculations for individual chemicals rather than chemical mixtures. Thus, calculations were performed for a selection of congeners that vary in the number of chlorine substitutions.

One of the motivations for modeling PCBs globally is to identify the major global-loss processes in order to assess the likely rate of future concentration decline in the Arctic environment and elsewhere (Wania *et al.*, 1999c). The model calculations showed that, historically, atmospheric degradation and transfer to the deep sea contributed most to the loss of PCBs from the global environment, whereas burial in freshwater sediments was of little significance on a global scale. Reaction of the gaseous compounds with OH radicals is the loss process of primary importance for the lighter congeners, whereas the deep-sea transfer process increases in relevance with the degree of chlorination. The model further predicts that the relative importance of the various loss processes has been changing over time, with degradation in soils taking over as the major loss process in the past twenty years. As primary emissions decreased, the concentrations (and thus, loss rates) of PCBs in the atmosphere and ocean water have declined quickly, whereas soils have retained a high pollutant load due to their slow response time (large capacity, but slow evaporation and degradation). Unfortunately, this implies that the future rate of purification of the global environment will be determined by the slow and poorly quantified degra-



PCB homologue composition in soil, %

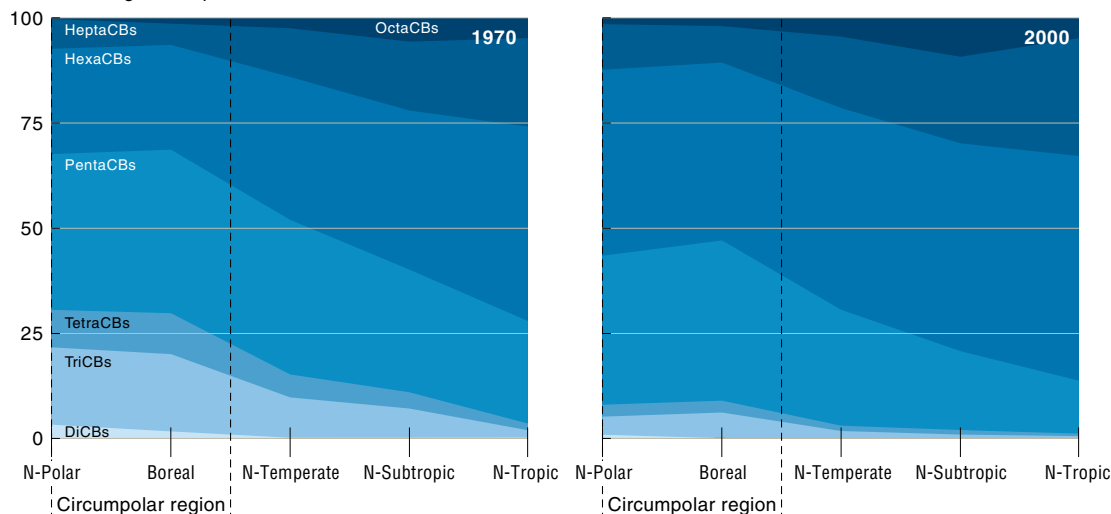


Figure 4-14. Modeled PCB homologue composition in soil in 1970 and 2000 in different zones of the northern hemisphere. The model predicts a shift to increasingly more volatile homologues with increasing latitude, and increased importance of the less volatile homologues over time, as the lighter homologues are more readily degraded (Wania, 1999).

duction rate in the soil environment. Figure 4-14 shows the homologue composition of PCBs in soil in 1970 and 2000 predicted by the Globo-POP model. The composition shifts to increasingly more volatile homologues with increasing latitude. In the past thirty years of steadily declining PCB emissions, the less volatile congeners have increased in relative importance as the lighter homologues are degraded more readily.

The global distribution model can also be used to investigate compositional shifts among the PCB congeners between compartments, zones and different time periods. Due to their wide range of physical-chemical properties, PCBs have played an important role in the derivation of the concept of global fractionation, which results in compositional shifts of compound mixtures with latitude (Wania and Mackay, 1993; 1996). The model succeeded in reproducing observed shifts toward lighter (lower chlorinated) PCB congeners with increasing latitude (Figure 4-15) found in various measurement cam-

PCB homologue concentration in soil, normalized to global average

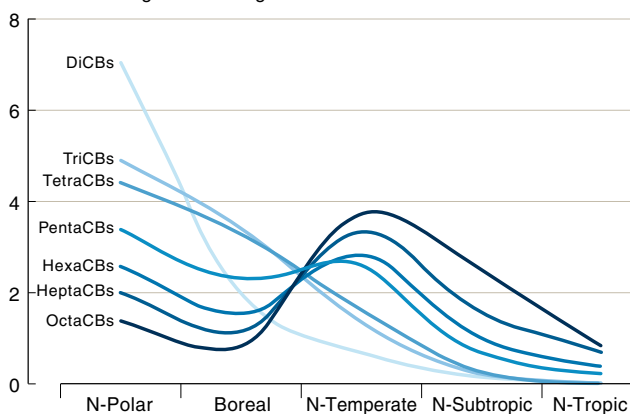


Figure 4-15. Soil concentrations of various PCB homologues as a function of (northern hemisphere) climate zone for the year 2000, predicted using the 'Globo-POPs' model of Wania and Mackay (2000). The soil concentrations are normalized to the global average concentration of a particular homologue. The concentrations of the lighter homologues, with up to four chlorines, increase steadily with latitude.

paigns (Muir *et al.*, 1996a; Ockenden *et al.*, 1998; 2002). The model further suggests that these shifts differ between various environmental media, change over time and are rather complex.

#### 4.2.2. Terrestrial herbivores

The previous AMAP assessment reported a large amount of OC data on caribou/reindeer (*Rangifer tarandus*). This species is an important food item in most northern societies and represents an excellent species to monitor terrestrial contamination, due to their diet of plants and lichen. Geographical differences were observed previously in OC concentrations in caribou/reindeer (de March *et al.*, 1998) but, in general, levels were low compared to Arctic marine mammals. For this reason, there are few new data on OC levels in caribou/reindeer and studies of terrestrial species have mainly focused on concentrations of radionuclides and heavy metals. Since 1997, there have been new studies on OCs in terrestrial mammals from Greenland (Muir and Johansen, 2001), Faroe Islands (Larsen and Dam, 1999) and Russia (RAIPON/AMAP/GEF Project, 2001), and a study on OC levels in reindeer from Finland (Hirvi and Henttonen, 2002).

##### *Russian terrestrial herbivores*

OC analyses were conducted on samples of various tissues (kidney, liver, and muscle) from four species of terrestrial herbivores (reindeer, mountain hare (*Lepus timidus*), ptarmigan (*Lagopus mutus*), and willow grouse (*Lagopus lagopus*)) collected in 2000-2001 from four regions of the Russian Arctic: Kola Peninsula (Lovozero); Pechora Basin (Nelmin Nos); the Taymir Peninsula (Dudinka and Khatanga); and, Chukotka (Kanchalan and Lavrentiya) (RAIPON/AMAP/GEF Project, 2001). Samples were collected in the same regions as the vegetation and soil samples (see Sections 4.2.1.1 and 4.2.1.3). Across all species, OC concentrations were highest in liver followed by kidney and muscle, which is likely due to higher lipid content of the liver, although no lipid data are currently available for these samples. Across all re-

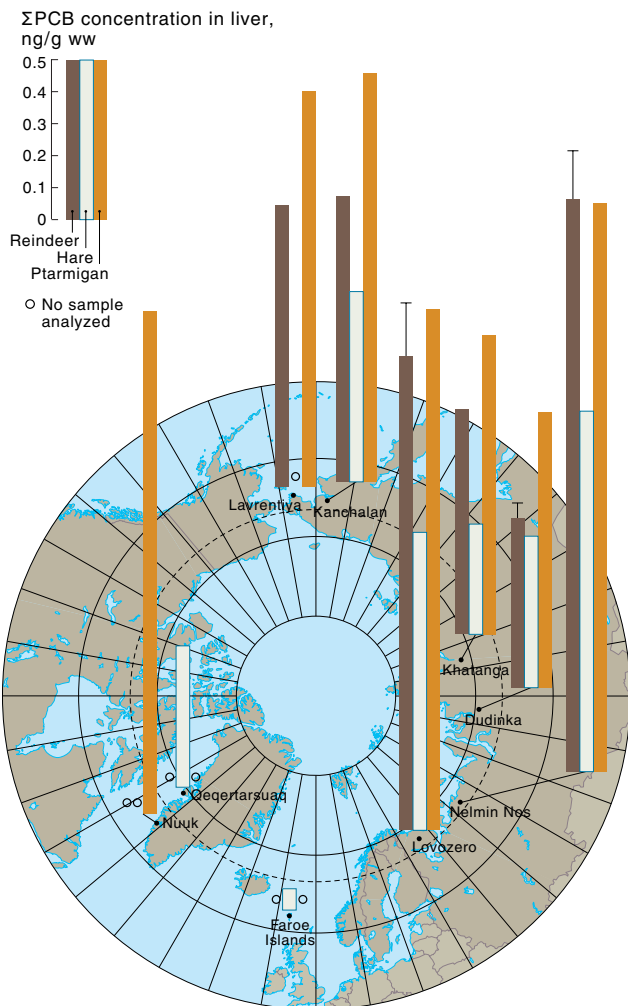


Figure 4-16. Concentrations of  $\Sigma$ PCBs in reindeer, Arctic hare and ptarmigans/willow grouse from regions within Russia (RAIPON/AMAP/GEF Project, 2001), and from Greenland and the Faroe Islands. Bars are means (with  $\pm 1$  SE indicated when more than two pooled samples are available). All samples are pools of numerous samples collected in 2000-2001.

regions and OC groups, OC concentrations were similar in the reindeer, ptarmigan, and willow grouse but lower in the mountain hares (Figure 4-16 and Annex Table 5). Some geographical trends were found. OC concentrations were higher in the regions of Lovozero (Kola Peninsula) and Nelmin Nos (Pechora Basin), but for each species, OC concentrations were within a factor of four across all regions. OC concentrations in these Russian terrestrial herbivores are in the same range as those observed recently in Greenland terrestrial herbivorous mammals (Muir and Johansen, 2001), and reported for Russian reindeer collected in 1994 (Melnikov *et al.*, 1995). Toxaphene (sum of Parlars 26, 52 and 60; detection limits approximately 0.2 ng/g in biota) and PBDEs (detection limits approximately 0.5 ng/g in biota) were not detected in any terrestrial herbivore samples, but the detection limits of this study are above the levels normally found for these compounds in Arctic biota.

Concentrations of  $\Sigma$ PCDDs,  $\Sigma$ PCDFs, and  $\Sigma$ TEQs were reported for muscle of reindeer, hares, ptarmigan and willow grouse collected in 2000-2001 from four regions of the Russian Arctic (RAIPON/AMAP/GEF Project, 2001) (Annex Table 16). Liver was also analyzed in reindeer and hares (the same samples analyzed for OCs,

discussed above). Liver concentrations were higher than muscle for  $\Sigma$ PCDDs,  $\Sigma$ PCDFs, and  $\Sigma$ TEQs and  $\Sigma$ PCDFs were greater than  $\Sigma$ PCDDs. Concentrations of  $\Sigma$ PCDDs,  $\Sigma$ PCDFs, and  $\Sigma$ TEQ were generally below 0.5 pg/g ww, with the exception of reindeer liver from the western sites Kola Peninsula (6.5 pg/g ww), Pechora Basin (2.5 pg/g ww), and the Taymir Peninsula (0.7 pg/g ww). These locations are downwind of large cities (Murmansk, Monchegorsk, Norilsk) which have non-ferrous smelters, a known source of airborne PCDFs.

#### Greenland terrestrial herbivores

Levels of OCs were assessed in a variety of tissues (liver, kidney, muscle, and fat) of terrestrial mammals (mountain hare, domestic lambs (*Ovis* spp.), and musk-ox (*Ovibos moschatus*)), and ptarmigan of Greenland (Annex Table 5). These species are important foodstuffs (Muir and Johansen, 2001). PCBs (concentration range of 0.5-7.5 ng/g ww) were by far the predominant OCs in all species across all tissues. The ranking of the other OCs varied with the tissue, but, in general, DDT and chlordanes were the next most common OC. Wet weight concentrations were greatest in fat, but lipid-corrected concentrations were often highest in liver or kidney. OC concentrations were similar between species, reflecting similar diets. These concentrations are in the range reported previously for herbivorous terrestrial mammals from the Canadian and European Arctic, but are orders of magnitude less than those observed in marine mammals such as the ringed seal (de March *et al.*, 1998). Compared to the Russian data, PCB concentrations in hares are similar (Figure 4-16) but ptarmigan have much higher concentrations. Interestingly, concentrations of chlorobenzenes observed in these terrestrial samples are similar to those in marine mammals.

#### Faroe Islands herbivores

Levels of OCs were determined in Faroe Islands terrestrial mammals, sheep, and mountain hare, which are part of the human diet (Larsen and Dam, 1999). In hare liver ( $n = 13$ ), the predominant OCs were HCB and oxy-chlordane, both occurring at mean concentrations near 30 ng/g lw. The concentrations of HCB are similar to those found in sculpin from the Faroe Islands, but are one fifth of those detected in other marine species such as black guillemot eggs and pilot whale tissues. The concentration of PCB (as CB153) was low and near the detection limit (approximately 4 ng/g lw), and other congeners were rarely detected. Of the six DDT isomers, only *p,p'*-DDE was regularly detected with a mean concentration of 7 ng/g lw. Toxaphene, mirex, and the other chlordanes were normally not detected. Liver and tallow taken from around the kidney of eight sheep and 17 lambs at each site were sampled from two locations in 1997. In most cases, PCB congeners, chlordanes, toxaphene, and *p,p'*-DDT were not detected. In lamb and sheep liver, the concentration of CB153 and CB138 were approximately 5 ng/g lw and 2 ng/g lw, respectively, with slightly lower concentrations in tallow, especially in sheep. The concentrations of *p,p'*-DDE in sheep and lamb liver were approximately 6 ng/g lw and 3 ng/g lw, respectively. Concentrations of OCs in the Faroe Islands terrestrial herbivores are similar to values reported in similar species from Greenland.

### Finland reindeer

OCs were found to be very low in the fat of reindeer collected in northern Finland in 2000, with most OCs below the detection limits (<5 ng/g) (Hirvi and Hentonen, 2002). However, these detection limits are high and above OC levels normally seen in Arctic terrestrial animals (Annex Table 5). Chlorobenzenes were found at the highest concentration ( $\Sigma\text{CBz}$   $14.3 \pm 4.4$  ng/g ww), followed by  $\Sigma\text{DDTs}$  ( $6.1 \pm 1.9$  ng/g ww) and  $\Sigma\text{PCBs}$  ( $3.5 \pm 8.8$  ng/g ww). These concentrations are at the low end of those reported previously for Arctic caribou/reindeer (de March *et al.*, 1998). High levels of chlorobenzenes, relative to other OCs, are often found in caribou/reindeer (de March *et al.*, 1998).

### 4.2.3. Birds of prey

Birds of prey have been of particular importance in the study of OCs in the Arctic due to their susceptibility to the effects of OCs and the potential for accumulation of OCs from winter habitats. A number of species were examined in the past (see de March *et al.*, 1998) but most recent work has focused on peregrine falcons (*Falco peregrinus*).

#### 4.2.3.1. North American peregrine falcon

Subspecies in Alaska are: the Arctic peregrine (*F. p. tundrius*), which nest in northern tundra; the American peregrine (*F. p. anatum*), which nests in the forested interior; and, the Peale's peregrine (*F. p. pealei*), which nests along the southern coast from the Aleutian Islands to southeast Alaska.

Persistent OC contaminants were measured in American and Arctic peregrine falcon eggs from Alaska during the period of 1979-1995 (Ambrose *et al.*, 2000) (Annex Table 5). This dataset was not included in the first AMAP POPs assessment, and a more detailed discussion of the data is provided in the temporal trends section (Section 5.2). Dieldrin, *p,p'*-DDE, heptachlor epoxide, oxychlor-dane, mirex, and total (Aroclor) PCBs were consistently detected and measured in all samples. HCB, *p,p'*-dichlorodiphenyldichloroethane (DDD), *p,p'*-DDT,  $\beta$ -HCH, and *trans*-nonachlor were detected in more than 50% of samples, and  $\alpha$ -HCH,  $\gamma$ -HCH, *trans*-chlordane, *cis*-chlordane, *o,p'*-DDD, *o,p'*-DDE, *o,p'*-DDT, endosulfan II, and endrin were detected in less than 50% of samples.

#### 4.2.3.2. European Arctic birds of prey

To assess the current level of OCs in Arctic European birds of prey, 44 egg samples from eight different raptor species (Table 4-6) were collected from throughout Nor-

way, including northern parts, during the period 1991-1997 and analyzed for OCs, including toxaphene (Herzke *et al.*, 2002). The DDT-transformation product *p,p'*-DDE still dominates the pesticide burden in all predatory bird eggs (70-90% of the pesticide burden) 30 years after the ban of these chemicals in western countries. PCBs accounted for 70-80% of the total OC burden in predatory birds, confirming the high ecotoxicological potential of this type of contaminant for birds of prey. The highest average concentrations for PCBs were found in eggs from white-tailed sea eagle (*Haliaeetus albicilla*) and peregrine falcon (average  $\Sigma\text{PCB}$  concentration: 8.9  $\mu\text{g/g}$  ww and 9.1  $\mu\text{g/g}$  ww, respectively). Merlin (*Falco columbarius*) and sparrowhawk (*Accipiter nisus*) eggs had the highest concentrations of chlorinated pesticides (average sum pesticide concentration: 3.0  $\mu\text{g/g}$  ww and 4.3  $\mu\text{g/g}$  ww, respectively). Toxaphenes were determined, and concentrations were low compared to PCBs and OCs. The highest toxaphene levels were found for bird species feeding on fish or migratory passerines. The highest toxaphene concentration was found in white-tailed sea eagle eggs (0.09  $\mu\text{g/g}$  ww). No toxaphene was found in osprey (*Pandion haliaetus*) and merlin eggs. No spatial or regional specific trends or patterns in OCs were found for the egg samples analyzed.

Osprey, merlin, sparrowhawk and peregrine falcon migrate to southern winter habitats where many pesticides are in use, and feed on local prey available in these regions. Gyrfalcon (*Falco rusticolus*), goshawk (*Accipiter gentiles*), golden eagle (*Aquila chrysaetos*), and white-tailed sea eagle are usually more confined to a specific habitat, and not as apt to migrate. The high pesticide levels found in merlin, sparrowhawk, and peregrine falcon may thus, in part, be explained by their migratory habits.

#### 4.2.3.3. 'New' chemicals in European Arctic birds of prey

High  $\Sigma\text{PBDE}$  concentrations have been found in predatory birds feeding on terrestrial mammals and birds, particularly peregrine falcons in northern Sweden (Sellström *et al.*, 2001; Lindberg *et al.*, 2004) and Norway (Herzke *et al.*, 2001a). The mean  $\Sigma\text{PBDEs}$  (BDEs 47, 99, 100, 153, and 154) in the two populations are 224 and 260 ng/g ww (4500 and 4700 ng/g lw), respectively, with levels ranging from 43 to 1580 ng/g ww (680-39000 ng/g lw) in the Swedish population. The  $\Sigma\text{PBDE}$  levels in Norwegian golden eagles, gyrfalcons, and merlins were 7, 18, and 36 ng/g ww (approximately 140, 360, and 720 ng/g lw). The congener pattern in all these bird species is quite different from that seen in fish and fish-eating birds and mammals, dominated by BDE153 and

Table 4-6. Habitat and diet information for Norwegian bird of prey species from which eggs were collected between 1991 and 1997 for OC analyses (Herzke *et al.*, 2002).

Species	Habitat description	Main food objects
Merlin	Terrestrial	Passerines and waders
White-tailed sea eagle	Marine	Fish and seabirds
Goshawk	Terrestrial	Medium-sized birds, small mammals
Golden eagle	Terrestrial	Mammals and game birds
Peregrine falcon	Terrestrial/marine	Medium-sized birds
Osprey	Limnic	Freshwater fish
Gyrfalcon	Terrestrial	Medium-sized birds
Sparrowhawk	Terrestrial	Passerines

99. The Swedish peregrine falcons also had measurable levels of BDE183 (HpBDE) and BDE209 (DeBDE).  $\Sigma$ PBDE levels are only 3-4 times less than  $\Sigma$ PCB levels in Swedish peregrine falcons. HBCD concentrations ranged from 2.2 to 28 ng/g ww (34-2400 ng/g lw) in the Swedish peregrine falcons. The burden of PBDEs in peregrine falcons may be linked to their migratory habits as the northern population in Sweden overwinters along the coast and estuaries of central and southern Europe. In addition, many of the birds that they prey on when in the Arctic are also migratory and may have PBDE burdens from their overwintering sites further south.

#### 4.2.4. Other carnivores

There are a number of terrestrial mammalian carnivores in the Arctic including the mink, wolf (*Canis lupus*), and wolverine (*Gulo gulo*). The Arctic fox is often considered a terrestrial carnivore but has been included in the marine mammal section of this report (Section 4.4.8). Through the process of biomagnification, these organisms generally have higher concentrations of OCs than terrestrial herbivores (de March *et al.*, 1998). However, the generally lower levels of OCs in the Arctic terrestrial environment results in OC concentrations in terrestrial carnivores that are much lower than in their marine counterparts. Some terrestrial mammals (e.g., the wolf) are also considered to have more efficient biotransformation ability based on the very small numbers of PCB congeners found in their tissues (Gamberg and Braune, 1999; Shore *et al.*, 2001). There have been few studies on OCs in terrestrial carnivores since the original AMAP report, in part because of low concern based on low levels in the terrestrial Arctic environment, and because few terrestrial carnivores are included in the diet of humans.

##### Wolves

There have been two studies on OC levels and patterns in wolves collected in the subarctic and Arctic in the past five years. Levels of OCs were measured in the liver of 58 wolves collected in northwest Russia (Shore *et al.*, 2001) and in the liver of wolves collected in the Canadian Yukon (Gamberg and Braune, 1999). Both studies found concentrations to be very low (mean or median below 50 ng/g ww) but were slightly higher in the Russian wolves (Annex Table 5). For many samples, common OCs, such as *p,p'*-DDE, were not detected, likely due to a combination of low levels and efficient biotransformation. A small number of highly chlorinated PCB congeners dominated the PCB load in both studies. OC concentrations were not found to vary between sexes and, with the exception of PCBs, did not vary with age. Both studies concluded that the levels of OCs do not pose a threat to the health of wolf populations.

##### Wolverines

Wolverines are omnivorous terrestrial mammals that live throughout the alpine and Arctic tundra of Canada and Scandinavia. Livers from 12 wolverines were collected at Kugluktuk, Nunavut, in the western Canadian Arctic to determine, for the first time, the residue patterns of OCs in this species (Hoekstra *et al.*, 2002a). The ranking of hepatic concentrations for sum ( $\Sigma$ ) OC groups in wolverines was  $\Sigma$ PCBs >  $\Sigma$ CHLs >  $\Sigma$ DDTs >  $\Sigma$ HCHs >  $\Sigma$ CBz.

The most abundant OC analytes detected in wolverine liver (in order from highest to lowest) were CB153, CB180, oxychlorane, CB138, *p,p'*-DDE, CBs 170/190, CB99, CB118, and dieldrin. These OCs are among the most recalcitrant and are also the predominant OCs in other terrestrial Arctic mammalian predators, such as wolves (Gamberg and Braune 1999). Wolverine age and sex did not influence OC concentrations.

Levels of chlordane and HCHs were much lower in the wolverines compared to the Arctic fox collected to the north in Ulukhaqtuuq (Hoekstra *et al.*, 2002a) but are in the range of levels in other Arctic terrestrial mammals. This was expected because coastal populations of Arctic fox are known to scavenge marine mammals, resulting in elevated OCs (Klobes *et al.*, 1998a). However, levels of PCBs and DDTs were similar or higher in the wolverine compared to the Arctic fox. Concentrations of these OCs in the wolverine are much higher than other Arctic terrestrial mammals (Annex Table 5). Although these PCB and DDT levels are below most threshold effects levels (Hoekstra *et al.*, 2002a) an explanation for these unexpected high levels is not evident and merits further research.

##### Common shrew

OCs were measured in the common shrew (*Sorex araneus*) liver collected in 1999 and 2000 in the area of Pallas, northern Finland (Hirvi and Henttonen, 2002). This small mammal feeds on insects, worms, and other similar small animals. The mean levels of OC pesticides in immature shrews were 0.2-13.0 ng/g ww and in adults 0.5-25.0 ng/g ww, which are similar but slightly higher than levels observed in reindeer from the same region of Finland.  $\Sigma$ PCB concentrations were 0.3-5.0 ng/g ww. The highest levels were measured for oxychlorane followed by  $\alpha$ -HCH,  $\gamma$ -HCH and *trans*-nonachlor. The concentrations of these contaminants were 10-20 times higher in shrews than in the humus layers, reflecting biomagnification up this short food web.

#### 4.2.5. Summary and conclusions – terrestrial environment

The first AMAP POPs assessment concluded that there had been a significant amount of research on OC levels in the Arctic terrestrial environment, although it found that there was insufficient spatial coverage of media other than in caribou and reindeer (de March *et al.*, 1998). Since that report (1997 to 2001), studies on the global distribution of PCBs and PCDD/Fs in soils have included sites in the Arctic, thus providing a much clearer perspective of the levels of these contaminants in Arctic soils. With the exception of work in Russia, these studies were not conducted as part of the AMAP implementation programs of various circumpolar countries. While the global survey results show that levels of PCBs and PCDD/Fs are much lower in the Arctic than in the northern temperate zone, there are urban areas within the Arctic, especially in Russia, with elevated PCB levels. The results of global modeling and soil measurements of PCBs imply that the future rate of purification of the global environment will be determined by the slow and poorly quantified degradation rate in the soil environment. Thus, despite low levels, future surveys of Arctic



soils will be important for tracking long-term trends in the elimination of PCBs and related POPs.

The limited amount of research that has been carried out in the terrestrial Arctic confirms what was reported in the first AMAP POPs assessment. It is apparent that Arctic terrestrial biota, with the exception of predatory birds and animals near local sources, have among the lowest OC concentrations of any biota in the world. Concentrations of OCs in this environment are orders of magnitude less than what is observed in the freshwater and marine environments. In soil and lower trophic-level organisms, HCB and HCHs are the most prevalent OCs, but PCBs, chlordane, and DDTs become more prevalent at higher trophic levels.  $\Sigma$ CBz concentrations in caribou/reindeer are similar to  $\Sigma$ PCBs and  $\Sigma$ DDTs.

The extensive spatial survey of OCs in soil, vegetation and terrestrial herbivores carried out very recently in the Russian Arctic (2000 and 2001), provided data that fill a knowledge gap identified in the first AMAP POPs assessment (de March *et al.*, 1998).  $\Sigma$ PCB concentrations were the highest of any OC group, followed by  $\Sigma$ DDTs and  $\Sigma$ HCHs. Toxaphene and PBDEs were rarely detected and only in soil; however, the detection limits in this study are above the levels normally found for these compounds in the Arctic. This survey found that there were minor geographical trends in OC concentrations across Russia in the terrestrial environment, although slightly elevated levels of OCs were observed in terrestrial herbivores from two regions and PCDD/F concentrations were high in reindeer in areas near smelters. The concentrations found were similar to those found in Russian Arctic samples collected in the mid-1990s and in Greenland terrestrial herbivorous mammals, but were slightly higher than those observed in older Canadian Arctic terrestrial studies. There have been no recent studies on OCs in terrestrial herbivores in the Canadian or Alaskan Arctic. Two independent studies on wolves in Canada and Russia found similar levels of OCs.

Contamination of the terrestrial environment by OCs appears to be close to uniform across the circumpolar Arctic, with slightly lower (2-3 times) concentrations in Canada. Lower OC concentrations in the Canadian terrestrial Arctic conform to spatial trends of OCs observed in Arctic air and aquatic biota. However, there has been no recent extensive circumpolar survey of OCs in any one matrix or biota species and therefore, conclusions about spatial trends of OCs in the terrestrial Arctic are difficult and should be made with caution. Although this represents a knowledge gap, the generally low OC levels found in the Arctic terrestrial environment indicate that this not a major issue.

High levels of PBDEs were found in Arctic terrestrial-feeding birds of prey. Congener patterns were quite different from those observed in biota feeding within the aquatic environment. There is very little data for other 'new' chemicals, such as PFOS, in the terrestrial environment. There is a need to assess the levels and spatial trends of 'new' chemicals, particularly in birds of prey.

#### 4.3. Freshwater environment

A fairly substantial dataset on OCs in Arctic freshwater ecosystems was summarized in the first AMAP POPs assessment (de March *et al.*, 1998). That report concluded

that 'With the exception of river and lake waters, the original minimum monitoring objectives of AMAP for POPs in freshwater matrices have been met in all circumpolar countries.' The freshwater ecosystems of the Arctic are contaminated almost exclusively by atmospherically transported POPs. There are exceptions to this, such as rivers and the systems fed by these rivers, which flow through northern cities and/or drain more southerly agricultural watersheds.

OC concentrations in Russian Arctic river waters collected in the mid-1990s were found to be approximately ten times higher than levels observed in Canada and Norway (de March *et al.*, 1998). These results were originally questioned but have since been verified as discussed in Section 4.3.1. There were insufficient data to examine circumpolar trends of POPs in freshwaters in detail in the first AMAP assessment report. There was however moderately comprehensive coverage for surface sediments. With the exception of  $\Sigma$ PCB concentrations in water from a few Canadian lakes, levels of OCs in water and sediment did not exceed guideline levels for protecting aquatic wildlife (de March *et al.*, 1998) (see also Table 6.1). OC levels in freshwater biota were found to be higher than in terrestrial biota, but lower than in comparable trophic levels of marine biota (de March *et al.*, 1998). Good circumpolar coverage of OCs was available for Arctic char, which showed similar levels throughout the Arctic except in lakes where some char had become cannibalistic. PCBs were the most common OC group, although toxaphene was shown to be a major OC in Canadian freshwater biota.

Since the first AMAP POPs assessment, there have been a number of studies on POPs (specifically PCBs and OC pesticides) in the freshwater environment, but the amount of data produced is small compared to that produced for the marine environment. As with the terrestrial environment, concentrations of OC contaminants are relatively low in most freshwater biota. Nevertheless, studies of OCs in freshwater fish in Alaska, Canada, Greenland, and Russia have continued, in part because they are important dietary items. Extensive studies on OCs in the food webs of Bjørnøya have also been carried out, and long-term temporal trends studies of freshwater fish in Sweden have continued (Section 5.3.2).

##### 4.3.1. Concentrations and loadings in surface waters

###### 4.3.1.1. Verification of older Russian river water data

###### *Background*

The previous AMAP POPs assessment compared concentrations of the OC pesticides HCH and DDT in major north-flowing rivers in Russia to results from Canada and Norway from sampling and analysis campaigns conducted in the mid-1990s. In the past five years, there have been no new studies of POPs in river waters in the Canadian Arctic, northern Norway or in northern Russia. A project was conducted in Russia in 2001-2002 to study contaminants in the Yenisey and Pechora rivers (RAIPON/AMAP/GEF Project, 2001), however the results were not available in time for this assessment. The data from Russian rivers reported in de March *et al.* (1998) were received late in the assessment process and not thoroughly evaluated. Since then, several papers have been published which give a more com-

prehensive description of the quality assurance program followed, as well as the levels and trends of OC pesticides in major Russian rivers (Zhulidov *et al.*, 1998; Alexeeva *et al.*, 2001). Therefore, the available results are re-examined here. Zhulidov *et al.* (2002) have also reported temporal trends of HCH and DDT isomers from the late 1980s to early 1990s in major rivers and these are discussed in Section 5.3.1.

All sampling and analyses of POPs in Russian rivers during the period 1990-1996 were carried out by regional laboratories of ROSHYDROMET (Federal Service of Russia on Hydrometeorology and Environment) with method development and quality assurance carried out by a central co-ordinator (the Hydrochemical Institute, located in Rostov-on-Don). Methods used for the OC pesticides involved solvent extraction (hexane) of 1 L of unfiltered water and gas chromatographic determination. An improved clean-up method involving H<sub>2</sub>SO<sub>4</sub> became available in 1995 and was in use in 1996 in all UGMS labs. The internal UGMS results for both precision and accuracy for all contaminants were very good (<30% deviation) for the six OC compounds determined. These six compounds were  $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH, *p,p'*-DDE, *p,p'*-DDT, and dihydroheptachlor (DHH; a heptachlor analog not determined in European or North American studies of OC pesticides).

#### Sampling program

Sampling took place at stations along major Russian rivers that are tributaries to the Arctic Ocean. Most attention was focused on the stations located further downstream in each large river in order to estimate loadings to the Arctic Ocean. Some of the stations were several hundred kilometers upstream of the geographical river mouth, as defined by headlands on the mainland. In some cases, this was because the tidal/saline characteristics of the river extend many kilometers upstream; in others, it was a matter of logistical convenience for sample collection. In some cases, there are cities with significant populations located upstream of the river mouth monitoring stations. Such cities and rivers include: Onega on the Onega River; Novodvinsk and Archangelsk on the Severnaya Dvina River; Naryan-Mar on the Pechora river; and, Dudinka on the Yenisey River.

Estimation of OC pesticide loadings was made difficult by the presence of many non-detect (nd) values. This does not mean that the OCs were not present, only that they were less than the detection limit. In the data sets evaluated, a zero was substituted for 'nd' for spreadsheet purposes whenever, e.g., the mean for an analyte was calculated. This biases the mean result toward a low value; however, substitution of the detection limit would bias the results toward a high value. During examination of the data, it was seen that an occasional maximum value (outlier) represented the determining value in the mean, usually when there were only a small number of data points. Where a reason to reject such points was determined by the group responsible for this study, the mean was recalculated; where there was no reason for rejection, the data were included. All data presented have been 'vetted' in this manner. The extent of the 'nd' values in the data reported in their study is unknown, but it is believed to be extensive so the means and loadings presented here are considered conservative.

Table 4-7. Seven-year weighted mean concentrations ( $\mu\text{g/L}$ ) of contaminants in Russian northern rivers: 1990-1996<sup>a</sup>.

River	$\alpha$ -HCH	$\gamma$ -HCH	<i>p,p'</i> -DDT	<i>p,p'</i> -DDE
Kola	0.003	0.003	<0.001	<0.001
Onega	0.003	0.003	<0.001	<0.001
Sev. Dvina	0.001	0.001	<0.001	<0.001
Mezen	0.004	0.004	0.004	<0.001
Pechora	0.002	0.012	0.001	0.001
Ob	0.030	0.053	0.020	0.002
Nadym	0.029	0.065	0.020	0.007
Pur	0.069	0.107	0.027	0.006
Taz	0.058	0.114	0.038	<0.001
Yenisey	0.009	0.016	<0.001	<0.001
Anabar	<0.001	<0.001	<0.001	<0.001
Olenek	0.001	<0.001	<0.001	<0.001
Lena	<0.001	0.001	<0.001	0.008
Kolyma	<0.001	0.003	<0.001	<0.001

<sup>a</sup> Weighted average calculated for all data for the study period, weighted for the number of contributing samples for each annual mean.

Comparability of OC pesticide concentrations for the rivers Ob, Yenisey, Lena, and Kolyma were assessed by Alexeeva *et al.* (2001) using independent data reported by Zhulidov *et al.* (1998). The mid-range of the data from the 'independent specialists' part of that report was employed for comparison since means were not presented. The mid-range values from the independent specialists and the concentration means in this section (Table 4-7) gave inconsistent ratios. In the Ob, values from the specialists were 3-6 times higher than those reported by UGMS laboratories for both  $\alpha$ -HCH and  $\gamma$ -HCH; for the Yenisey, the concentrations given in both reports were similar (within 20%), while levels for the Lena and Kolyma were below those of the specialists.

#### OC pesticide levels and spatial trends

The available concentration data were limited to annual arithmetic means, ranges, and the number of samples (log normal distributions were reported for the larger data sets). This data set for 15 rivers and six regularly monitored OC contaminants over seven years, was examined for each river to assess whether temporal or geographic pattern(s) existed. A significant temporal trend was apparent only for  $\gamma$ -HCH concentrations in the Pechora River (1990-1996), but not for any other pesticide or river. In the case of  $\gamma$ -HCH in the Pechora, the origin of the contaminant must be relatively local because similar patterns were not observed in other nearby rivers. There has, however, been no reported agricultural use of  $\gamma$ -HCH in this basin during the years reported. It may be that the source is from urban waste waters due to domestic uses in towns and cities within the watershed, or from use for biting-fly control in urban and other non-agricultural settings. As a consequence of there being no consistent temporal patterns, the concentration data for the rivers were averaged for the study period, and weighted for the number of contributing samples for each annual mean (Table 4-7)

The  $\alpha$ -HCH and  $\gamma$ -HCH concentrations in the rivers draining into the White/Barents Seas (Table 4-7) were in the low  $\mu\text{g/L}$  range (means 0.0023 and 0.0041  $\mu\text{g/L}$ , respectively). DDT and DDE were non-detectable except for the Pechora in 1993 and 1996. Even the maximum contaminant levels for the rivers do not appear to change

in any consistent fashion over the reporting period.  $\gamma$ -HCH in the Pechora seemed to display higher mean and maximum levels in the 1990-1994 time period, but these were only 2-3 times those found elsewhere in the same region. DDT and DDE detection limits were high and, therefore, it is not surprising that almost no residues were found to be above the low  $\mu\text{g/L}$  range.

Four of the five monitored rivers flowing to the Kara Sea (the Ob, Nadym, Pur, and Taz) were characterized by much higher levels of reported OC pesticides than were rivers flowing to the White and Barents Seas. The fifth, the Yenisey River, is significant for loadings but less so for concentrations. The averages of the annual means for  $\alpha$ -HCH,  $\gamma$ -HCH,  $p,p'$ -DDT, and  $p,p'$ -DDE in these four rivers were, respectively: 0.028; 0.050; 0.015; and, 0.0022  $\mu\text{g/L}$ , although the contributing means had a high level of variability, and the Ob had more samples contributing to its annual means than did the other rivers. Babkina (1999) reported that agricultural usage and soil concentrations of  $\gamma$ -HCH in the river basins and sub-basins studied here were significant only in the upper Ob-Irtysh Basin, and to a lesser extent, in the upper Yenisey Basin. The Ob River (and its tributary, the Irtysh River) drain agricultural areas in the south of Russia and in Kazakhstan, which have received high pesticide applications including  $\gamma$ -HCH. The observation that the shorter, non-agricultural Nadym, Pur and Taz rivers exhibited  $\gamma$ -HCH levels as high as or higher than those of the Ob lends weight to the premise that forestry, mining or other usage was significant in those basins.

For the HCHs in the Ob, there appears to be a transition around 1993-1994 as concentrations before this period are much higher than those after. Alexeeva *et al.* (1997) reported total HCH usage in Russia at roughly 5000 t/yr for the period 1970 to the mid-1980s, and falling to 600-700 t/yr by the late 1980s. HELCOM (2001) reports lindane use in Russia at 13.7 t in 1990 and 5.9 t in 1996. Unpublished data (Babkina, 1999) show that there was a further sharp reduction in use of HCH in the upper Ob-Irtysh Basin around 1992 and that collectively, the usage in this basin before this time was a significant fraction (approximately  $\frac{1}{3}$ ) of the amounts reported for all of the former Soviet Union (fSU) in the late 1980s. The reductions in HCH usage in these upper reaches of the river are roughly reflected in the water concentrations at the river mouth, indicating a fairly rapid system response time.

There were few observations of OC pesticides above the detection limits for any of the rivers flowing into the Laptev and East Siberian Seas (only for HCHs; mainly in 1990). This part of Russia is characterized by tundra over much of these basins, and there is less reason to expect the use of  $\gamma$ -HCH in these regions. It is noted that the 1990 observations were during the period when HCH usage for agriculture was still high. While there are no significant agricultural activities in the region, mining is extensive, and the extent of pesticide use in support of such activities, as with forestry, is unknown.

#### Other chlorinated pesticides in Russian Arctic rivers

$\beta$ -HCH was analyzed in water from northern Russian rivers but was not detected at any site. The lack of observations was expected given that the  $\alpha$ - and  $\gamma$ -HCH isomer levels are near their limits of detection. In Canadian Arctic rivers, the levels of  $\beta$ -HCH were also less than detection limits (approximately 0.00002  $\mu\text{g/L}$ ) (Jeffries *et al.*, 1996). In Norway, they were not reported (Holtan *et al.*, 1994).

The rivers flowing to the Kara Sea were the only ones in which the insecticide degradation product DHH was investigated. This compound may have been used in forests against biting insects, as well as in the agricultural industry. The overall mean of the reported years/rivers was 0.0042  $\mu\text{g/L}$  and, at least for the Ob, the sample numbers were considerable. For all of fSU/Russia, 580 and 520 t/yr of DHH were used in 1990 and 1991, respectively (Alexeeva *et al.*, 1997), and the Ob Basin accounted for 12% of these amounts in both years. Data for later years are not available. This pesticide has not been investigated in Canadian or Norwegian rivers.

#### Loadings of OC pesticides in the northern Russian seas

The loadings of  $\Sigma\text{HCHs}$  and  $\Sigma\text{DDTs}$  (1990-1996) are illustrated in Figure 4.17. The Kara Sea received, by far, the largest fraction of OC pesticide discharges going to the northern seas; well over 90% of the totals for three of the four measured organics.  $\alpha$ - and  $\gamma$ -HCHs and  $p,p'$ -DDT flowed to the Arctic largely via the Ob River ( $\alpha$ -HCH 50%,  $\gamma$ -HCH 53%,  $p,p'$ -DDT 72%); DDE largely (76%) entered the Laptev Sea via the Lena River in 1990. Since DDT has been banned for more than 25 years, this material may be partly from soil and deposited material applied earlier (Harner *et al.*, 1998),

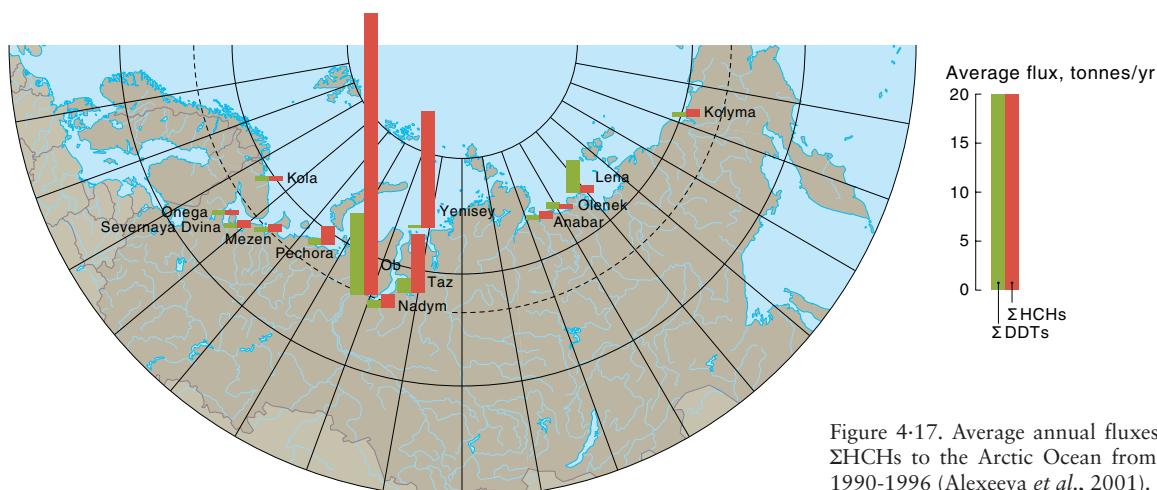


Figure 4.17. Average annual fluxes of  $\Sigma\text{DDTs}$  and  $\Sigma\text{HCHs}$  to the Arctic Ocean from Russian rivers, 1990-1996 (Alexeeva *et al.*, 2001).



although a higher proportion of DDE would be expected than was observed. The loadings of HCHs were greater than those of DDT and DDE, except for DDE in the Lena in 1990, for which there is no known explanation.

The Ob carried the major loadings of  $\alpha$ - and  $\gamma$ -HCHs which, in the case of  $\alpha$ -HCH, was 50% of the total for all of the rivers studied. The Yenisey was responsible for a further 24% of the total. The Ob carried 53% of the  $\gamma$ -HCH to the Kara Sea, while the Yenisey carried 18% of this isomer. A major fraction ( $1/3$  based on data for pre-1990 use) of Russia's agricultural use of HCH occurred in the upper reaches of these two rivers (and these are the only reported uses in the northern river basins). The observation that the Nadym, Pur, and Taz rivers also have significant loadings (and high concentrations) of both of these isomers relative even to the Ob, indicates other anthropogenic sources in these non-agricultural watersheds. Forestry and mining are prominent industries in the area, and both use significant quantities of pesticides to provide better working conditions in the region.

These rivers represent the major loadings from the northern flows, but data are incomplete. Using the AMAP data for total flows to the various seas (Gregor *et al.*, 1998), the basin loadings from the monitored rivers in each northern sea were scaled up to account for the rivers not monitored (those measured represented 58-80% of the combined basin flows). The total annual loadings of the OCs were estimated to be 25 t for  $\alpha$ -HCH, 44 t for  $\gamma$ -HCH, 13 t for *p,p'*-DDT, and 6 t for *p,p'*-DDE. The HCH values can be compared with the various flux estimates previously reported for the Arctic (Macdonald *et al.*, 2000). Ocean current delivery via the Bering Strait was 52 and 12 t/yr for  $\alpha$ -HCH and  $\gamma$ -HCH, respectively and, via the atmosphere, 53 and 10 t/yr.

#### 4.3.1.2. Recent studies of OCs in Russian lake and river waters

Samples of freshwater (15-45 L) were collected from lakes and rivers in four regions of the Russian Arctic: Kola Peninsula (Lake Lovozero); Pechora River mouth at Nelmin Nos; Taymir Peninsula (Yenisey River at Dudinka and Khatanga River at Khatanga), and at Kanchalan on the Kanchalan River in Chukotka, in 2000-2001 for analysis of POPs (RAIPON/AMAP/GEF Project, 2001). Samples from Lake Lovozero were collected at various depths (0-30 m). There did not appear to be trends in OC concentrations with depth, and therefore, these data have been combined. Samples from all other sites were collected from rivers near the surface. PCBs were the predominant OC followed by DDTs and chlorobenzenes (Figure 4-18 and Annex Table 6). Toxaphene (sum of Parlars 26, 52, and 60; detection limits approximately 0.1 ng/L) and PBDEs (detection limits approximately 0.5 ng/L) were not detected in any water sample but the detection limits of this study are above the levels that would normally be anticipated for these compounds in the water column.

Except for HCHs, no geographical trends were obvious for any OC group, although levels were slightly higher in Lake Lovozero (Figure 4-18). Whether this is due to these being lake, versus river, samples cannot be

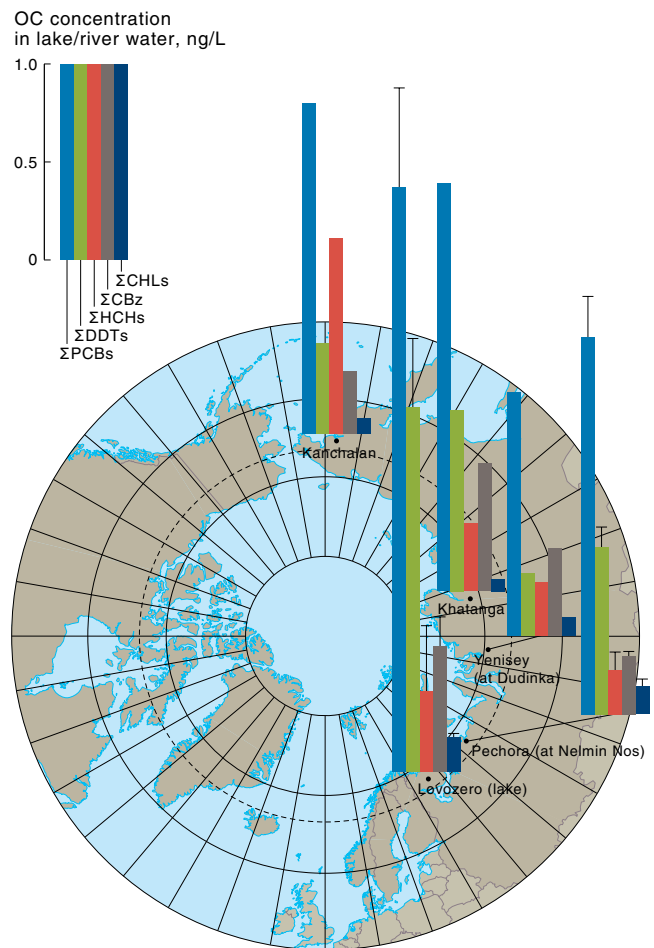


Figure 4-18. Concentrations of OCs in lake (Lovozero) and river water in northern Russia (RAIPON/AMAP/GEF Project, 2001). Bars are means (with  $\pm 1$  SE indicated when more than two pooled samples are available). All samples are pools of numerous samples collected in 2000-2001. Samples from Lovozero included surface and depth samples. OC concentrations varied little with depth in Lake Lovozero.

assessed from the information available. OC concentrations are in the same range as levels reported previously for Russian rivers.

#### 4.3.1.3. OCs in Canadian Arctic lake waters

A very limited number of new measurements of POPs in freshwater have been carried out in the Canadian Arctic since the previous AMAP POPs assessment. Law *et al.* (2001) summarized results for  $\alpha$ - and  $\gamma$ -HCH in lakes from Cornwallis Island (central Canadian Arctic Archipelago) and the Yukon, and compared the results for Arctic lakes with the Great Lakes and small lakes in south-central Ontario. Concentrations of  $\alpha$ -HCH ranged from 0.64 to 1.7 ng/L and  $\gamma$ -HCH ranged from 0.13 to 1.3 ng/L. Sampling dates for the lake waters ranged from 1993 (Yukon) to 1998 (Cornwallis Island). Declining airborne levels of  $\alpha$ -HCH over this time period (Li *et al.*, 1998a) may have influenced the observed levels in surface waters. Highest concentrations of  $\alpha$ -HCH were found in large, cold and oligotrophic lakes, such as those in the Arctic, subarctic, and the upper Great Lakes, and this was attributed to greater inputs from atmospheric deposition and slower loss rates relative to warmer, temperate lakes.



Helm *et al.* (2000) analyzed water from Amituk, Char, and Meretta Lakes for enantiomers and concentrations of  $\alpha$ -HCH to estimate the extent of biodegradation in watersheds in the Canadian High Arctic.  $\alpha$ -HCH concentrations ranged from 0.839-1.021 ng/L in Char Lake and 1.205-1.264 ng/L in Meretta Lake. By comparison,  $\alpha$ -HCH was present at concentrations of 0.520-0.688 ng/L in the water column of Amituk Lake (Falconer *et al.*, 1995) and at 0.119-0.529 ng/L in inflowing streams. The  $\alpha$ -HCH ERs declined with increasing stream temperatures indicating greater rates of biodegradation of  $\alpha$ -HCH. As stream flows declined during the summer so too did stream volume-to-surface ratios, resulting in greater contact time between water and bed surfaces. The results suggest that enantioselective degradation was enhanced by the greater contact time between the chemical in the water and stream or slope substrates, the presumed site of microbial communities. Approximately 7% of  $\alpha$ -HCH in the Amituk Lake Basin was enantioselectively degraded prior to entering the lake. ERs within Amituk Lake were controlled by meltwater inputs rather than within-lake degradation, and clearly illustrate the riverine-like nature of High Arctic lakes.

Differences in the lake  $\alpha$ -HCH inventory from the end of summer 1993 to spring 1994 indicate that from 33 to 61% of  $\alpha$ -HCH within the lake may have been lost via non-enantioselective microbial degradation at a rate ranging from 0.48 to 1.13/yr. In comparison to the average ER of 0.75 in Amituk Lake, the average ERs in the oligotrophic Char Lake and the more productive Meretta Lake were 0.65 (range 0.62-0.69) and 0.88 (range 0.84-0.90), respectively. These differences are likely attributable to within-lake degradation that is a function of lake water residence times, which are 9-14 yr for Char Lake, 3 yr for Amituk Lake, and <3 yr for Meretta Lake, rather than degradation within streams or lake productivity. Law *et al.* (2001), in a survey of 24 lakes, including the lakes studied by Helm *et al.* (2000), also noted an inverse relationship between enantioselective degradation and lake trophic status inferred by phosphorus and nitrogen concentrations.

#### 4.3.2. Levels and fluxes of OCs in lake and river sediments

A significant number of studies on OCs in sediment were carried out prior to, and summarized in, the original AMAP POPs assessment, allowing for examination of circumpolar trends (de March *et al.*, 1998). A limited number of studies of POPs in freshwater sediments have been conducted since the previous AMAP POPs assessment. In this section, we examine studies of concentrations and fluxes in surface sediments and their spatial variation. Historical profiles are examined in Section 5.3.1. Most of the studies in this assessment involved lakes and rivers that are remote and where POPs inputs are likely due to long-range transport and deposition. Urban/industrial sources of contamination, however, may be important in rivers in Russia as well as in the Northwest Territories and Yukon in Canada and are also considered. The first AMAP assessment concluded that there were no global latitudinal or longitudinal trends of OCs in Arctic lake sediment, although fluxes of OCs declined with latitude if temperate lakes were included.

##### 4.3.2.1. Russian river and lake sediments

Sediment samples were collected from the same sites as surface waters in four regions of the Russian Arctic (Kola Peninsula (Lake Lovozero); Pechora River mouth; on the Taymir Peninsula, the Yenisey River at Dudinka and Khatanga River at Khatanga; and, at Kanchalan on the Kanchalan River in Chukotka) in 2000/2001 and were analyzed for POPs (RAIPON/AMAP/GEF Project, 2001). A single pooled sample was analyzed from each region based on samples collected at either 0-2 cm (Lovozero) or 0-5 cm depth (all others). PCBs were the dominant OCs in the freshwater sediments followed by DDT (Annex Table 6). Toxaphene (sum of Parlars 26, 52, and 60; detection limits approximately 0.1 ng/L) and PBDEs (detection limits approximately 0.5 ng/L) were not detected in any freshwater sediment sample.

OC concentrations were similar in all regions of Russia with the exception of DDT concentrations in Pechora sediment (Annex Table 6). Concentrations of DDT in this region were 70 times higher than the other regions. This sample was a pool of ten subsamples from the Nelmin Nos area of the river. The ratio of DDT isomers to  $\Sigma$ DDT in the Nelmin Nos sediment is 0.67 compared to 0.2-0.24 in the other sediments analyzed in the RAIPON/AMAP/GEF Project. DDT isomers predominate in newly synthesized DDT, but are converted to DDE and DDD isomers in the environment. The high concentrations of  $\Sigma$ DDTs and the high ratio of DDT to  $\Sigma$ DDTs in the Pechora sediment suggest recent use of DDT in this region.

Skotvold and Savinov (2003) reported  $\Sigma$ PCB<sub>10</sub> (sum of ten major congeners) in surface (0-2 cm) sediment samples from 11 coastal tundra ponds collected along the Russian Arctic coast from the southeastern Barents Sea to the East Siberian Sea in 1994-95. Most of these data were previously reported in the AMAP POPs assessment (de March *et al.* 1998) but not discussed in detail. Mean  $\Sigma$ PCB concentrations ranged from  $0.38 \pm 0.22$  ng/g dw for three ponds on the Laptev Sea coast to  $7.9 \pm 9.9$  ng/g dw for three ponds on the Kara Sea coast. Only one sample collected in a pond located on Baydaratskaya Bay coast had a relatively high  $\Sigma$ PCB<sub>10</sub> concentration (19.3 ng/g).

Overall the  $\Sigma$ PCB concentrations in Russian freshwater lake/river sediments analyzed by the recent RAIPON/AMAP/GEF Project are at similar low ng/g (dw) levels when compared to results from the major north-flowing rivers in Russia reported in the first AMAP POPs assessment (de March *et al.*, 1998). Concentrations in Lake Lovozero sediments were similar to those reported by Skotvold and Savinov (2003) for lakes in northern Finnmark and on the southeastern Barents Sea coast. However, the  $\Sigma$ DDT concentrations in the pooled sediment sample from the Pechora mouth exceeded concentrations previously reported in the first AMAP POPs assessment for the major north-flowing rivers in Russia (de March *et al.*, 1998). In the present dataset (Annex Table 6) one site, Watson Lake, in the Yukon in the Western Canadian Arctic had higher  $\Sigma$ DDTs than the sample from the Pechora. The Watson Lake area was known to have received DDT applications for biting-fly control in the 1950's (Rawn *et al.*, 2001) and use of DDT for this purpose may be the source of the DDT in the Pechora as well.

#### 4.3.2.2. Lake sediments in northern Norway and Sweden

Skotvold and Savinov (2003) determined  $\Sigma\text{PCB}_{10}$  in surface bottom sediments (0-2 cm depth) collected from 26 remote lakes in northern Norway in 1994 and 1995 and previously reported, but not discussed in detail, in the AMAP POPs assessment (de March *et al.*, 1998).  $\Sigma\text{PCB}_{10}$  levels in sediments from one lake on Svalbard (Barentsvann) and four in Nordland (northern/central Norway) were significantly higher compared to other areas to the east. Significantly lower  $\Sigma\text{PCB}_{10}$  levels were found in southwestern and northern Finnmark and the coast of the East Siberian and southeastern Barents Seas compared to the lakes in Nordland, Svalbard, and Bjørnøya. The highest PCB concentrations were found in the lake Ellasjøen on Bjørnøya as discussed in Section 4.3.2.3.

Rose *et al.* (2003) found  $\Sigma\text{PCB}_{10}$  concentrations for surface sediments from five lakes along the west coast of Svalbard sampled in 1997 that ranged between 2.5 and 13.5 ng/g (Annex Table 6). This study involved the analysis of surface (0-1 cm) and pre-industrial sediments from dated cores. Highest concentrations and fluxes were recorded in the surface slice of a core from the lake Tenndammen located near the coal mining towns of Barentsburg and Longyearbyen. This lake also showed elevated spherical carbonaceous particles and PAHs in surface slices compared to the other lakes on Svalbard, although levels of contamination were low compared to European sites (Rose *et al.*, 2003).

A latitudinal study of organochlorine contamination in bottom sediments of 100 Swedish lakes, many of which were part of the National Swedish Environmental Monitoring program (Swedish Monitoring Program, 2002), included two lakes from the Swedish Arctic mountains and other other high latitude lakes (Söderström *et al.*, 2002). This study compared concentrations of PCBs (seven congeners), three DDT-related compounds, HCB, and three HCH-isomers in surface (0-2 cm) samples of each lake. The six Swedish Arctic lakes had significantly lower mean concentrations of  $\Sigma\text{PCB}$  ( $0.9 \pm 0.5$  ng/g dw),  $\Sigma\text{DDTs}$  ( $1.0 \pm 1.0$  ng/g dw), and  $\Sigma\text{HCH}$  ( $0.1 \pm 0.05$  ng/g dw) than southern lakes ( $9.2 \pm 6.3$  ng/g dw,  $14.6 \pm 12.1$  ng/g dw, and  $1.3 \pm 1.5$  ng/g dw, respectively). For HCB the six northern lakes had about three-fold lower concentrations than the 60 southern lakes in the study, reflecting the longer atmospheric half-life of HCB than the other compounds and lack of major past local uses in the south.

These studies in Sweden and the Norwegian and Russian Arctic illustrate how a large array of lakes can be used to assess broad geographical trends in deposition of POPs. Similar approaches have been used in North America (e.g., Muir *et al.*, 1995a; 1996a), but with far fewer and less well characterized lakes than in Sweden or Norway. A limitation of this approach is that, generally, the sediment cores were not dated, thus sedimentation rates and the degree of particle focusing are not known. Given generally lower sedimentation rates, especially in the Arctic mountain lakes and in other lakes above the tree line, it is reasonable to assume that a much longer period of deposition was represented by the 2-cm layer in the high latitude lakes

than in those from southern Sweden or northern/central Norway. This complicates the geographical comparison since only recent deposition may be measured in high sedimentation sites versus longer-term deposition at low sedimentation sites.

#### 4.3.2.3. Sediments from lakes on Bjørnøya

Sediment cores were taken from two small lakes (Ellasjøen and Øyangen) on Bjørnøya in 1996 for OC analysis as part of a larger study (Section 4.3.5). Concentrations of all OCs were much higher in Ellasjøen compared to Øyangen. The concentration of PCBs in surface sediment from Ellasjøen ( $\Sigma\text{PCB}_7 = 60$  ng/g dw) was an order of magnitude higher than in Øyangen ( $\Sigma\text{PCB}_7 = 4.4$  ng/g dw). As previously noted, the concentrations found in the sediments of Ellasjøen are the highest found for a remote lake in the Arctic. In Ellasjøen, guano from seabirds was identified as a potential source of contaminants (Evenset *et al.*, 2002). Levels reported for Øyangen are in the range generally reported for lakes in northern Norway and Sweden (Skotvold and Savinov, 2003).

DDT isomers make up a small percentage of the total  $\Sigma\text{DDTs}$  in lake sediments from Bjørnøya indicating that atmospheric input (and/or guano inputs in the case of Ellasjøen) was the primary source.

The only chlordanes with concentrations above the detection limit in the surface sediment from Øyangen were *cis*-chlordane and *trans*- and *cis*-nonachlor at 0.08, 0.06, and 0.03 ng/g dw, respectively. These same compounds were present at concentrations two to four times higher (0.17, 0.22, and 0.10 ng/g dw, respectively) in surface sediments from Ellasjøen. Oxychlordane, *trans*-chlordane, and *cis*-chlordane were also detected in Ellasjøen (0.11, 0.05 and 0.04 ng/g dw, respectively). Heptachlor and its oxidation product, heptachlor epoxide, were below the detection limit in all sediment samples. Concentrations of  $\gamma$ -HCH and  $\alpha$ -HCH in the surface sediments from Ellasjøen and Øyangen were relatively low (<0.2 and <0.4 ng/g dw, respectively), as were the concentrations of HCB (<0.8 ng/g dw). Dieldrin was present at 0.51 ng/g dw in the surface sediment in Ellasjøen. None of these pesticides could be detected in the sediment from Øyangen.

#### 4.3.2.4. North American Arctic lake sediments

Concentrations and fluxes of POPs in lake sediments have been recently reported for about 14 lakes in the North American Arctic (Cleverly *et al.*, 1996; Lockhart, 1996; 1997; Macdonald *et al.*, 2000; Muir *et al.*, 2002b; Rawn *et al.*, 2001; Stern and Evans, 2003). When combined with previous studies in Alaska and northern Canada that were reviewed in the AMAP POPs assessment (de March *et al.*, 1998) results are available for about 30 lakes. Similar to the approach used in studies in Norway and Sweden, sediment cores from the deepest point in each lake have been used in almost all studies in the North American Arctic. However, most of the sediment cores have also been dated using the  $^{210}\text{Pb}$  or  $^{137}\text{Cs}$  techniques (Oldfield and Appleby, 1984) which yields an estimate of sedimentation rates and particle focusing factors. Thus fluxes (concentration  $\times$  sedimentation

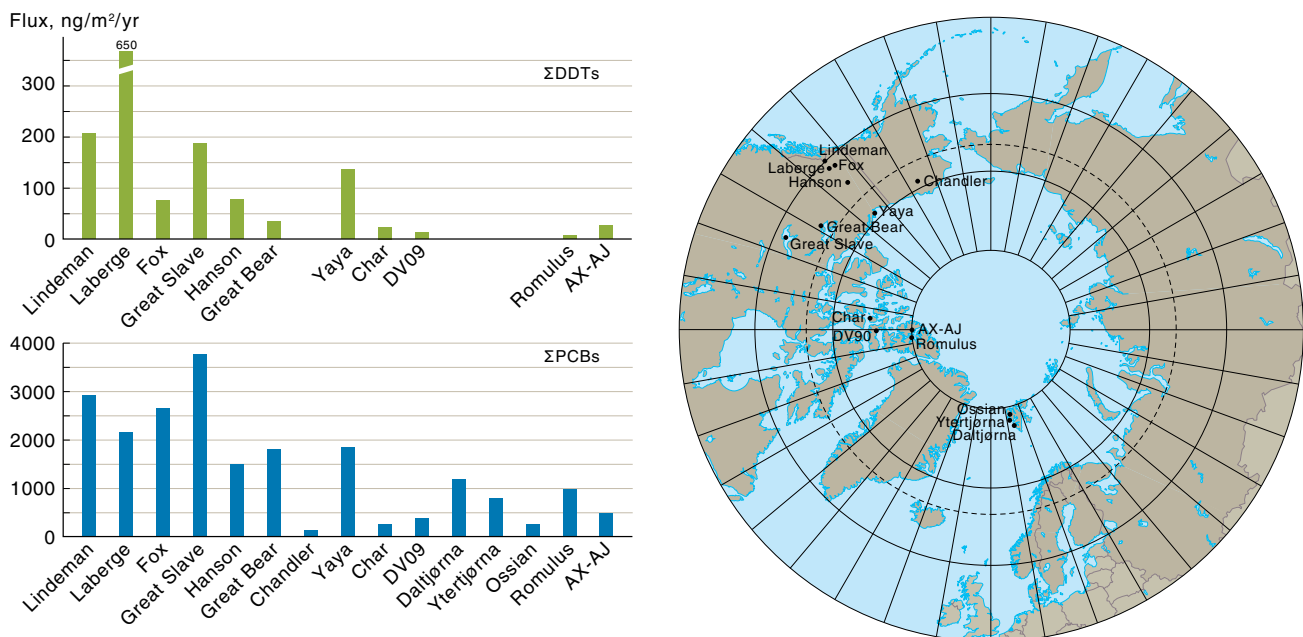


Figure 4-19. Fluxes of  $\Sigma$ DDTs and  $\Sigma$ PCBs in lake sediment cores (averages for slices dated to the 1990s) from the Canadian Arctic and Svalbard. Lakes are arranged by increasing latitude from left to right. A significant decline with increasing north latitude was observed for both DDT and PCB. Lakes with significant influence of local inputs are omitted.

rate) of POPs to lake sediments can be calculated over a wide geographic area in the North American Arctic. Fluxes (post-1990) of  $\Sigma$ DDTs and  $\Sigma$ PCBs reported recently in 14 lakes in the North American Arctic and, for PCBs, in three lakes on Svalbard (Rose et al., 2003) are shown in Figure 4-19.

#### Alaska

Cleverly *et al.* (1996) measured fluxes of PCBs, including coplanar PCBs, as well as PCDD/Fs in a dated sediment core from Chandler Lake in northern Alaska. These data were not included in the previous AMAP POPs assessment. Very low concentrations of PCBs were detected (Annex Table 6) and fluxes were among the lowest observed in Arctic lake sediments (Figure 4-19). Gubala *et al.* (1995) found similar low fluxes of PCBs and OC pesticides in a sediment core from Wonder Lake and lower fluxes in Schrader Lake in northern Alaska. Overall, knowledge of fluxes of PCBs and OC pesticides to lakes in Alaska is very limited.

#### Canadian Arctic lakes

Rawn *et al.* (2001) reported fluxes and historical profiles of PCBs and OC pesticides in sediment cores from six lakes and from one alpine lake in northern British Columbia within the Yukon River watershed. Surface concentrations and maximum concentrations in the core are presented in Annex Table 6. Several lakes showed evidence of local contamination. The highest  $\Sigma$ PCB and  $\Sigma$ DDT levels were measured in deep sediments from Watson Lake, although levels were much lower in surface sediments. Both chemicals showed increases in the mid-1940s consistent with the use of pesticides for biting-fly control during the building of the Alaska Highway. Hanson Lake was treated with toxaphene, and Lockhart *et al.* (1997) and Vetter *et al.* (1999) have reported the profile of toxaphene congeners in sediments from this lake. Sediment cores also revealed evidence for past use of DDT in the region, especially in Lake

Laberge (Rawn *et al.*, 2001).  $\Sigma$ PCB concentrations in Lindeman and Kusawa Lake surface sediments, both very remote from urban areas in the Yukon, were low relative to the other Yukon lakes that were studied (Annex Table 6).

Concentrations of  $\Sigma$ HCHs and  $\Sigma$ CHLs in surface sediments of the seven lakes studied by Rawn *et al.* (2001) were generally in the low ng/g range, consistent with a predominantly atmospheric input source (Annex Table 6). Subsurface maximum  $\Sigma$ HCH concentrations were observed in all but Lindeman Lake.  $\alpha$ - and  $\gamma$ -HCH were the dominant isomers contributing on average, 39 and 52%, respectively, to  $\Sigma$ HCHs. Elevated levels of  $\Sigma$ CHLs were observed in slices dated to the 1970s and early 1980s in Fox, Kusawa, Hanson, and Lindeman Lakes. Peak concentrations were measured in the surface sediment from Atlin Lake (3.87 ng/g dw).

Evans *et al.* (1996) examined concentrations and fluxes of PCBs, OC pesticides, and PCDD/Fs in Great Slave Lake. This study was not reported in the previous AMAP POPs assessment. Great Slave Lake, which is the fourth largest lake in Canada in terms of both surface area (27 000 km<sup>2</sup>) and volume (2088 km<sup>3</sup>), is a relatively pristine ecosystem with low concentrations of organic contaminants in all but the most industrialized areas (Mudroch *et al.*, 1992). Greater than 50% of the population in the Northwest Territories resides along the shores of Great Slave Lake. The Slave River, which receives the majority of its water from the Peace and Athabasca rivers, is the major river entering Great Slave Lake and contributes some 87% of the total annual inflow of 135 km<sup>3</sup> (Evans *et al.*, 1996).

Sedimentation rates were highest near the mouth of the Slave River and decreased with increasing distance from the river mouth (Figure 4-20 a). Sedimentation rates were very low at the westernmost sampled station and roughly comparable to that in the East Arm of Great Slave Lake. Most sediments (and associated contaminants), therefore, are likely to be deposited in the



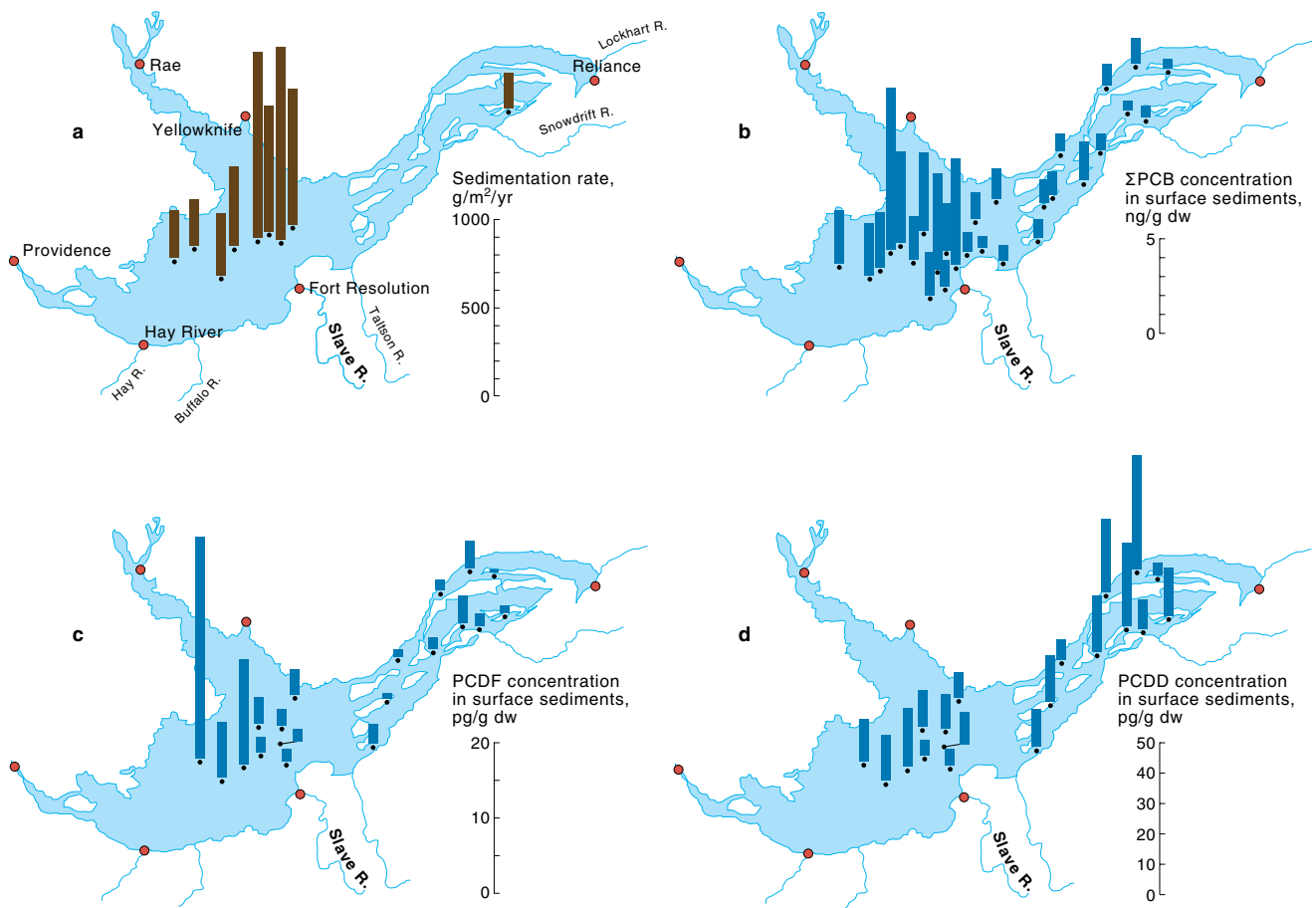


Figure 4-20. a) Sedimentation rates, b)  $\Sigma$ PCB concentrations, c) PCDF concentrations, and d) PCDD concentrations in surface sediments of Great Slave Lake (Northwest Territories, Canada).

deeper regions immediately offshore of the mouth of the Slave River.

$\Sigma$ PCB (70 congeners) and PCDDFs (major 2,3,7,8-substituted tetra- to octachlorinated congeners) were highest in sediments offshore of the river mouth and decreased with increasing distance from the river mouth (Figure 4-20 b,c,d). This, combined with the higher sedimentation rates in this region, indicates that contaminant fluxes of these compounds to the sediments were substantially greater offshore of the river than with increasing distance away from the river mouth. The converse pattern was observed for PCDFs, which tended to occur in high concentrations in the East Arm of Great Slave Lake.

Lockhart (1997) analyzed a sediment core from Yaya Lake, an isolated lake in the Mackenzie Delta (Annex Table 6). Surface concentrations of major OC pesticides and  $\Sigma$ PCBs in this core were very low, consistent with its isolated location relative to Great Slave Lake (Figure 4-19). Graf Pannatier (1997) examined concentrations and fluxes of PCBs and OC pesticides in a series of 'closure' lakes (connected to the Mackenzie River and flooded annually in the spring freshet) in the Mackenzie Delta. These lakes were characterized by very high sedimentation rates of 2300-12 200 g/m<sup>2</sup>/yr due to annual sediment deposition from flooding. This was much higher than nearby Yaya Lake (480 g/m<sup>2</sup>/yr) which is not flooded as frequently (Lockhart, 1997). Concentrations of  $\Sigma$ PCBs and major OC pesticides in sediment cores from Lakes 6 and 7, two middle delta lakes, were low and similar to those in Yaya Lake (Annex Table 6).

Surface and maximum concentrations of major OC pesticides and PCBs in sediment cores from five isolated lakes in the Canadian Arctic Archipelago (Stern and Evans, 2003; Muir *et al.*, 2002b) are presented in Annex Table 6. Surface  $\Sigma$ PCB concentrations (0.5 cm depth) were relatively similar in all lakes ranging from 0.96 to 3.7 ng/g dw.  $\Sigma$ HCHs had the widest range of concentration in surface sediments (0.01-0.69 ng/g dw). Historical profiles of POPs in several of these lakes are discussed in Section 5.3.

#### *Geographic trends in deposition of PCBs and DDT in lake sediments*

Recent (1990s) fluxes (ng/m<sup>2</sup>/yr) of  $\Sigma$ PCBs and  $\Sigma$ DDTs in sediment, corrected for particle focusing, for selected lakes in the Canadian Arctic, Alaska, and Svalbard are shown in Figure 4-19. Lakes for which past local inputs were thought to be significant, such as Watson and Little Atlin Lakes in the Yukon (Rawn *et al.*, 2001), and the lake Tenndammen near mining towns in Svalbard, were omitted, as were Lakes 6 and 7 due to direct inputs of sediments from Mackenzie River flooding (Graf Pannatier, 1997).  $\Sigma$ DDT fluxes in all of the high latitude lakes, were two-fold or more lower than reported for Yukon or NWT lakes, but similar to fluxes found in northern Alaska (Gubala *et al.*, 1995), and to previous reports for cores from four High Arctic lakes collected in the late 1980s and early 1990s (Muir *et al.*, 1995a; 1996a).  $\Sigma$ DDT fluxes ranged widely; Fox, Lindeman, and Hanson Lakes in the Yukon had similar fluxes to Great



Slave and Yaya Lakes, while Lake Laberge had a much higher flux reflecting some continued input from old sources within the watershed.  $\Sigma$ PCB fluxes in all of the high latitude lakes in Canada and in Svalbard (Rose *et al.*, 2003) were also lower than lakes in the Yukon and Northwest Territories, but higher than in most Alaskan lakes (Cleverly *et al.*, 1996; Gubala *et al.*, 1995). A statistically significant decline in fluxes of  $\Sigma$ PCBs and  $\Sigma$ DDTs with latitude was observed, reflecting the higher fluxes in the western Canadian Arctic lakes (Figure 4-19). This decline with increased northern latitude was also observed for higher chlorinated PCBs by Muir *et al.* (1996a) and for  $\Sigma$ DDTs by Muir *et al.* (1995a) when remote lakes from northwestern Ontario were included in the regression.

#### 4.3.2.5. PCDD/F fluxes to lake sediments

Cleverly *et al.* (1996) reported planar PCBs and PCDD/Fs in a sediment core from Chandler Lake in northern Alaska. The flux of total TCDD TEQs, based on PCBs and PCDD/Fs were 8.7 pg TEQ/m<sup>2</sup>/yr in the surface slice from this core. The TEQs were mainly due to CB126, 2,3,7,8-TCDD, and 1,2,3,7,8-pentachlorodibenzo-*p*-dioxin. PCDD/Fs have also been determined in sediments from a laminated core from Lake DV09 on Devon Island (Stern and Evans, 2003). The surface and maximum deposition flux values for PCDDs to Lake DV09 were calculated to be 0.01 and 0.36 pg TEQ/m<sup>2</sup>/yr, respectively. The historical trends in PCDD/F deposition in this core, which had much higher temporal resolution than the one from Chandler Lake, are discussed in Section 5.3.

Using the PCDD/F emission inventory for Canada and the U.S., Commoner *et al.* (2000) predicted 'dioxin' toxic equivalents (TEQ) deposition of about 4-53 pg TEQ/m<sup>2</sup>/yr to terrestrial surfaces near eight communities in the eastern Canadian Arctic which is in reasonable agreement with observations of Cleverly *et al.* (1996) but an order of magnitude higher than reported for the Devon Island core.

Evans *et al.* (1996) determined PCDDs and PCDFs in surface sediments and sediment cores from Great Slave Lake (Figure 4-20). PCDD concentrations were highest in sediments offshore of the Slave River mouth and decreased with increasing distance. This, combined with the higher sedimentation rates in this region, indicates that contaminant fluxes of these compounds to the sediments were substantially greater offshore of the river than with increasing distance away. The converse pattern was observed for PCDFs, which tended to occur in high concentrations in the East Arm of the lake.

No reports were found on PCDD/Fs or coplanar PCB deposition to sediments in the European Arctic.

#### 4.3.2.6. 'New' chemicals in Arctic sediments

Tomy *et al.* (1999) determined SCCPs in three Arctic lake sediment cores previously analyzed by Muir *et al.* (1996a) and Lockhart (1997). Surface concentrations (0-1 cm) ranged from 257 ng/g dw in Fox Lake (Yukon) to 1.6 ng/g dw in Yaya Lake (Mackenzie Delta). Lake Hazen had intermediate concentrations of SCCPs. Stern and Evans (2003) reported maximum concentrations of

SCCPs (17.6 ng/g dw) in the surface slice of a core from Lake DV09 on Devon Island. The possibility of contamination from SCCPs, which are widely used as flame retardant plasticizers and in cutting oils for metal machining cannot be ruled out, especially where there is a significant amount of small-boat traffic such as in Fox Lake. However, the results also indicate that long-range atmospheric transport and deposition were probably the major pathways for SCCPs to the very remote lakes (Yaya, DV09, and Hazen).

No reports were found on SCCPs or other 'new' POPs in lake sediments from the European Arctic.

#### 4.3.2.7. PAHs in Arctic lake sediments

PAHs in freshwater sediments were not assessed in the previous AMAP assessment. Here we confine the discussion primarily to pyrogenic PAHs in dated lake sediment cores. Fernández *et al.* (1999; 2000) calculated fluxes of 23 PAHs for the lake Arresjøen on northwestern Svalbard (79°40'N) and Øvre Neådalsvatn (62°46'N) in the Caledonian mountain range of central Norway. Major PAHs in Arresjøen were primarily pyrolytic in origin (i.e. resulting from combustion of coal and hydrocarbon fuel burning) such as phenanthrene, fluoranthene, pyrene, chrysene/triphenylene, indeno[1,2,3-*c,d*]pyrene and benzo[*g,h,i*]perylene. Fernández *et al.* (1999) concluded that the PAH deposition pattern in high altitude mountain lakes, including Arresjøen, paralleled sulfate deposition, pointing to combustion particles as the main input pathway. Rose *et al.* (2003) determined 15 PAHs in surface and prehistorical slices of sediment cores from four lakes in Svalbard. Highest fluxes were found in the lake Tenndammen (360 µg/m<sup>2</sup>/yr) a lake within 20 km of the coal mining towns of Barentsburg and Longyearbyen. PAH fluxes in the four Svalbard lakes appeared to decline with distance from the Barentsburg/Longyearbyen area.

Fluxes of  $\Sigma$ PAH (the same 23 PAHs as analyzed by Fernández *et al.* (1999) minus perylene and retene) have also been reported in a series of studies of dated sediment cores from the Canadian Arctic (Lockhart, 1994; 1996; 1997; Muir and Lockhart, 1994; Lockhart *et al.*, 1995; 1997). Total PAH fluxes in Yukon lakes ranged from 9.1 µg/m<sup>2</sup>/yr in remote Kusawa Lake to 174 µg/m<sup>2</sup>/yr in Little Atlin Lake (Lockhart *et al.*, 1997). Two remote lakes in the Mackenzie River Basin, Lac Sainte Therese and Yaya Lake, had high  $\Sigma$ PAH fluxes (68 and 140 µg/m<sup>2</sup>/yr, respectively) (Lockhart, 1997).

Graf Pannatier (1997) determined PAHs (unsubstituted as well as methyl naphthalenes and phenanthrenes) in sediment cores from four 'closure' lakes in the Mackenzie River Delta. Concentrations of  $\Sigma$ PAHs in surface sediments (0-1 cm) ranged from 650 to 900 ng/g dw. Due to high sedimentation rates in these lakes resulting from annual deposition in the spring freshet, fluxes of  $\Sigma$ PAHs in these lakes were in the range of 4-11 mg/m<sup>2</sup>/yr, well above fluxes for nearby Yaya Lake which is flooded less frequently. High  $\Sigma$ PAH values have also been found in Mackenzie River suspended particulates and they are thought to be mainly of petrogenic origin (Yunker and Macdonald, 1995).

Concentrations and fluxes of PAHs were also determined in sediment samples from Great Slave Lake

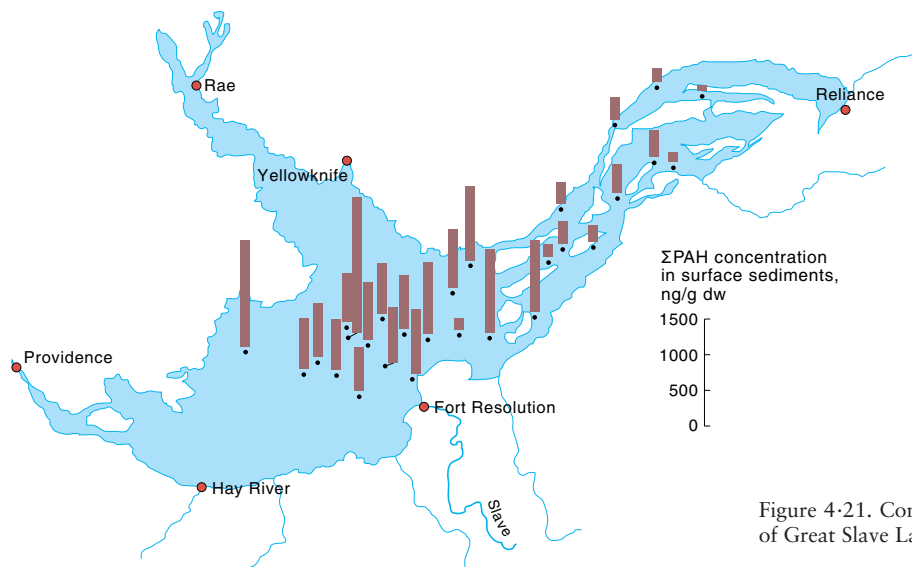


Figure 4-21. Concentrations of PAHs in surface sediments of Great Slave Lake (NWT, Canada) (Evans *et al.*, 1996).

(Evans *et al.*, 1996). PAH concentrations, like those for PCDD/Fs, were highest in sediments offshore of the mouth of the Slave River and decreased with increasing distance (Figure 4-21).

The recent (late 1980-1990s) PAH fluxes for the European and Canadian Arctic are compared in Figure 4-22. In general, excluding the lake Tenndammen, higher fluxes of substituted PAHs have been found in the Mackenzie valley and Yukon lakes compared with the European Arctic lakes. The large range of PAH fluxes in Canadian Arctic lake sediment cores demonstrates that sources of PAHs are complex and could be due to natural petrogenic sources within the basin while at the same time showing some characteristics of pyrolytic inputs.

#### 4.3.3. Freshwater fish and invertebrates

Freshwater fish are an important component in the diet of many northern communities, and a variety of tissues may be consumed. OC concentrations observed in freshwater invertebrates and fish appear to be predominantly controlled by exposure (i.e. regional differences) and trophic level, with levels in various tissues determined by lipid content. OCs have been shown to biomagnify in Arctic freshwater food webs, which results in the greatest concentrations in the highest trophic-level fish (Kidd *et al.*, 1998). To accurately assess spatial and temporal trends of OCs in fish, the length, age and growth rates need to be considered because all can have an impact on observed OC concentrations (Johnston *et al.*, 2002). In light of this, observed relationships in this section need to be viewed with caution because these parameters were not considered in most studies, making comparisons of different studies difficult.

A fairly large dataset on OCs in freshwater fish and invertebrates was assessed for the first AMAP POPs assessment (de March *et al.*, 1998); since 1996, a smaller amount of additional data has been produced. This is likely due, in part, to the low concentrations of OCs generally found in Arctic freshwater systems when compared to temperate freshwater and Arctic marine systems. In general, concentrations (lipid corrected) of OCs in freshwater fish of the same species are similar throughout the Arctic (within an order of magnitude), with a

few important exceptions that can be attributed to increased exposure or unique biological variations. For example, elevated levels of OCs in the lake Ellasjøen on Bjørnøya, north of Norway, have resulted in high OC

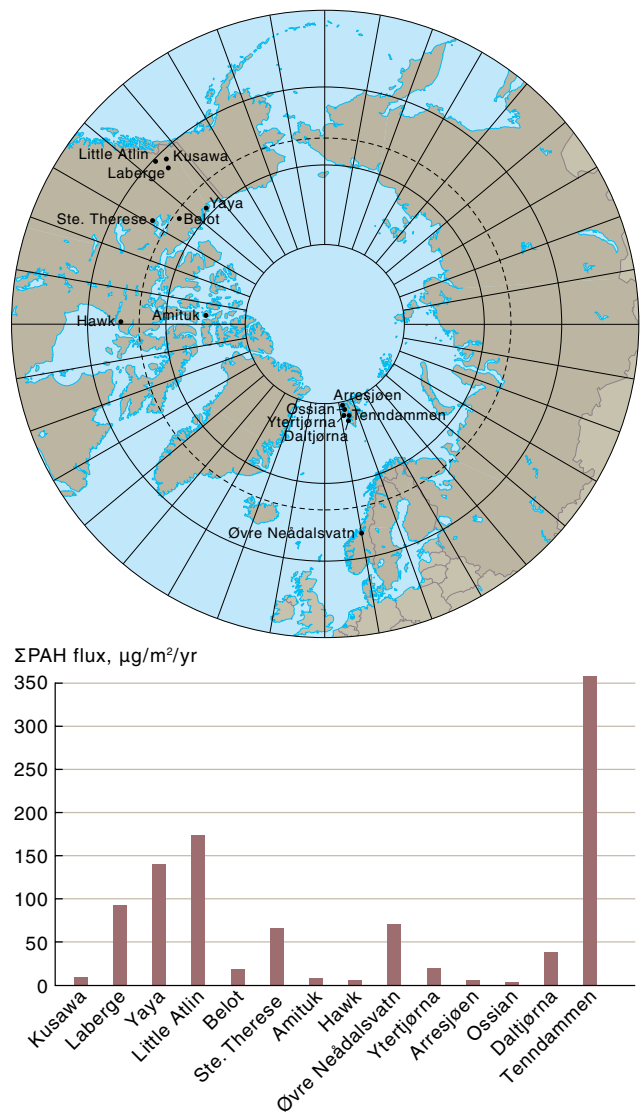


Figure 4-22. Recent fluxes of total PAH (23 PAHs minus perylene and retene) in dated lake sediment cores from the Canadian and Norwegian Arctic. Lakes are organized longitudinally from west to east.

concentrations in landlocked Arctic char from this lake (Section 4.3.5). PCBs and toxaphene continue to be the predominant OCs in freshwater invertebrates and fish, although data on toxaphene in the European Arctic are limited, and some exceptions do exist.

#### 4.3.3.1. Invertebrates

A fairly comprehensive dataset on OCs in zooplankton from the Canadian Arctic was reviewed in the first AMAP assessment report. Observed spatial trends were attributed to a combination of differences in water concentrations and lake plankton biomass. Since this first AMAP assessment, there have been few studies on OCs in freshwater zooplankton (Annex Table 7). This is likely due to the low concern about OCs in freshwater zooplankton because of low concentrations and the fact that they are not a human diet item.

Data on OC concentrations in freshwater invertebrates were generated as part of a large study of OCs in the lakes of Bjørnøya (see Section 4.3.5). Zooplankton and benthic invertebrate samples were collected from three lakes (Skutillen, Ellasjøen, and Øyangen) on Bjørnøya in 1996 and in 1999. Chironomid larvae and pupae were also collected from fish stomachs in 1996 and in 1999. The levels of  $\Sigma$ PCBs, HCB and  $\Sigma$ DDTs in pooled samples of zooplankton and benthic organisms (*Chironomidae* sp. and *Lepidurus arcticus*) were greatest in Ellasjøen, consistent with results in sediment and fish (Figures 4-27 and 4-28, page 76). Sampled zooplankton from Ellasjøen had higher concentrations of all the measured contaminants than zooplankton from Skutillen, followed by Øyangen with the lowest concentrations. Chironomids from Ellasjøen had higher contaminant levels than the *Lepidurus* samples from the two other lakes. *L. arcticus* from Øyangen had higher levels of contaminants than *L. arcticus* from Skutillen. The lipid content was very low in *Lepidurus* from Skutillen. Calculated on a lipid weight basis, the contamination levels were higher in *Lepidurus* from Skutillen than from Øyangen.

#### 4.3.3.2. Freshwater fish

##### Russian Arctic fish

POP analyses including PCDD/Fs were conducted on samples of various tissues (liver and muscle) from a number of fish species collected in 2000-2001 from four regions of the Russian Arctic: Kola Peninsula; Pechora Basin; Taymir Peninsula (Dudinka and Khatanga); and, Chukotka (Kanchalan and Lavrentiya) (RAIPON/AMAP/GEF Project, 2001) (Annex Table 16). Species analyzed for each location are found in Annex Tables 7 and 16. This project addresses a major data gap for OCs in Russian freshwater biota identified in the first AMAP assessment report (de March *et al.*, 1998). However, the somewhat limited sample numbers, current lack of biological data (e.g., body size), and lack of lipid data confounds efforts to statistically compare the sites within Russia with other locations, and such comparisons should be viewed with caution.

PCBs and DDTs were the predominant OCs found across all species in Russian freshwater fish, followed by chlordane and HCH (Annex Table 7). The highest con-

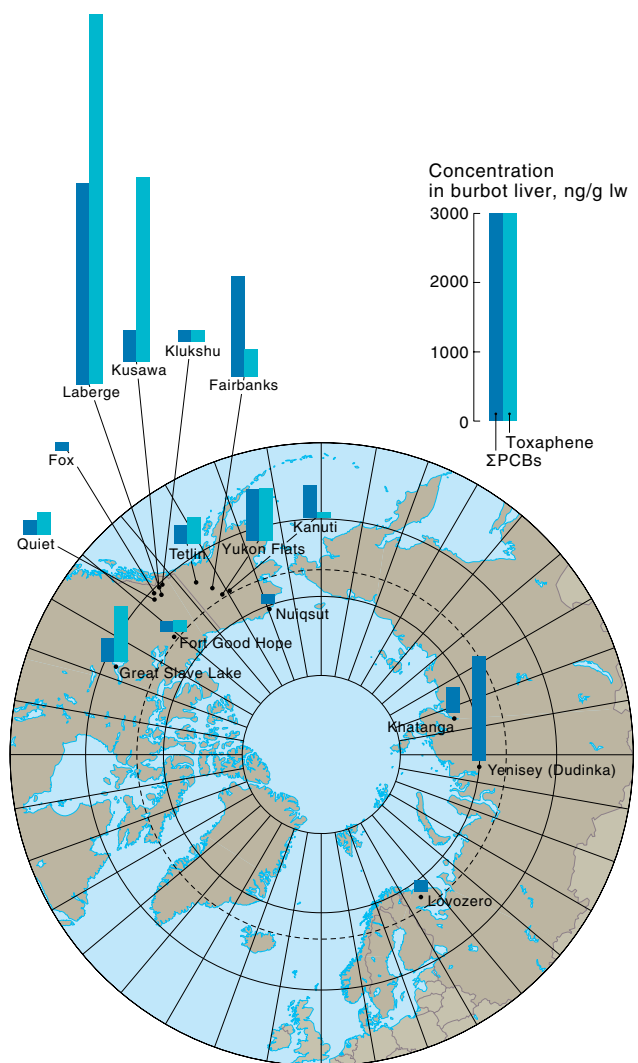


Figure 4-23. Spatial trends of  $\Sigma$ PCBs and toxaphene in burbot collected in Arctic Canada, Alaska, and Russia. Data for Lake Lovozero, Dudinka and Khatanga (Russian samples) were the sum of ten PCB congeners (no lipid content data was available for these samples, and this was therefore assumed to be 30%); for Fairbanks, Kanuti, Tetlin, and Yukon Flats, data were generated by the Aroclor method; all other data by the summation of >50 PCB congeners. Russian data from RAIPON/AMAP/GEF Project (2001); Fairbanks, Kanuti, Tetlin, and Yukon Flats data are from Mueller and Matz (2000); Nuiqsut data from Hoekstra (2002a), Great Slave Lake data from Evans and Muir (2001), and Fort Good Hope and Fox Lake data from Stern *et al.* (2001a); all other data are from Roach (2002).

centrations were observed in burbot (*Lota lota*) livers. This was attributable to their predatory nature as well as the high lipid content of their liver. The lowest concentrations were found in the whitefish species (*Coregonus clupeaformis*), a non-predatory fish. The relative differences in concentrations between species in Russia are similar to those observed in the Canadian Arctic.

Regional comparison of POP levels in Russian Arctic fish is difficult because no one species was collected in all regions. In burbot, the highest concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs were observed in samples collected from Dudinka on the Taymir Peninsula, which were 5-10 times higher than concentrations observed on the Kola Peninsula and Khatanga on the Taymir Peninsula (Annex Table 7). In general, concentrations of PCBs in burbot from Dudinka are among the highest found in



Arctic burbot, but levels on the Kola Peninsula and at Khatanga are among the lowest (Figure 4·23). Again, without information on lipid levels, it is difficult to draw conclusions about spatial trends. Since these data were only recently generated, in-depth evaluation will be a future priority.

Comparison of PCDD/F levels between fish species in different regions of Russia is also difficult due to limited sample size and a lack of biological data (e.g., lipid contents) (Annex Table 16). In general, PCDD/F concentrations were similar between species with the exception of Arctic char from Lavrentiya, where concentrations of PCDD were ten times higher. These fish are likely sea-

cies (Evans and Muir, 2000; 2001). In addition to water, the Peace and Athabasca rivers, which flow into the Slave River, transport tremendous amounts of sediments from the south into the Great Slave Lake ecosystem. This raises the possibility that increased development to the south may be resulting in increased contaminant transport to the lake through the Slave River, in addition to input via the atmosphere.

OC concentrations were similar between four species of predatory fish (northern pike (*Esox lucius*), walleye (*Stizostedion vitreum*), burbot, and inconnu (*Stenodus leucichthys*)) collected in the same region of Great Slave Lake (Annex Table 7), which is likely due to the fact that all of these species predate predominantly on fish. OC concentrations (on both a wet and lipid weight basis) tended, however, to be slightly higher in fish collected in the East Arm of Great Slave Lake compared with the West Basin in 1993 and 1999, although this varied somewhat with the contaminant and/or fish species. The variation in concentrations between basins could be due to differences in the characteristics of the fish (e.g., length and/or age) collected for the two regions and/or the characteristics of the two basins. OC concentrations and sedimentation rates tended to be higher in lake sediments from the West Basin than in the East Arm. Lake water in the East Arm is clear with low particulate concentrations, and invertebrates such as mysids and amphipods are strongly benthic. In contrast, in the West Basin, waters are turbid with high concentrations of suspended silts and clays, and invertebrates spend less time in close contact with the sediments than in the East Arm. Biota inhabiting the East Arm of Great Slave Lake may be accumulating higher body burdens of persistent OCs than in the West Basin because these contaminants are more bioavailable to them.

#### Lake trout

Concentrations of OCs were determined in lake trout muscle from a number of lakes in the Yukon and Northwest Territories in the late 1990s (Stern *et al.*, 2000; Evans and Muir, 2001; Roach, 2002). Previous work on the Yukon lakes in 1993 found that OC concentrations were particularly high in Lake Laberge, which was attributed to local point sources and a long food web (Kidd *et al.*, 1995). Since that time, concentrations in Lake Laberge lake trout have decreased up to four-fold, over the five-year period from 1993 to 1998 (see Section 5.3.3). When the lipid-adjusted OC levels in lake trout from Lake Laberge are compared to the other Yukon lakes (1998 and 1999 collections only), levels in the Lake Laberge trout do not stand out as they did in the early 1990s. However, caution is warranted in interpreting these results as no effort was made to account for differences in biological characteristics (e.g., size or age) between the lake trout sampled in each lake. In some lakes, much smaller trout were sampled.

Concentrations of the more lipophilic contaminants, such as toxaphene and PCBs, were much higher in lake trout from Kusawa Lake (1998-1999), while lake trout from Atlin Lake had the highest concentrations of the less lipophilic OCs ( $\Sigma$ HCH and  $\Sigma$ CBz) (Figure 4·25). The lakes Kusawa and Atlin receive water from glacial melt, which may, in part, explain high OC levels in fish from these lakes. Glacial melt has been implicated as a

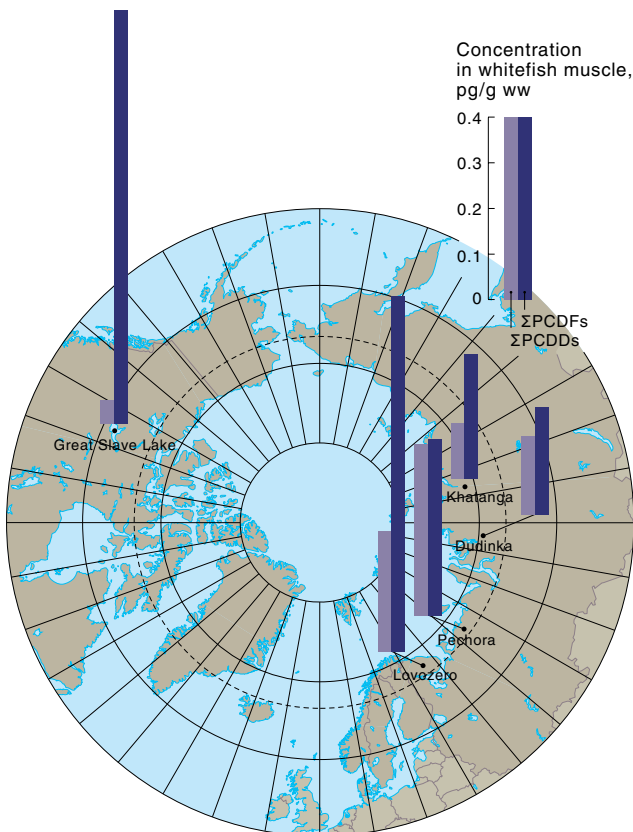


Figure 4·24 Spatial trends of PCDD/Fs in muscle of whitefish collected from Russian lakes and rivers in 2000-2001 (RAIPON/AMAP/GEF Project, 2001), and Great Slave Lake, NWT, Canada in 1994 (Evans and Muir, 2001).

run, which may explain the higher level observed. In whitefish, PCDD/F concentrations were slightly higher in samples from the Kola Peninsula and Pechora Basin compared to eastern sites in Russia (Figure 4·24). Levels of PCDDs are higher and PCDFs are lower in the Russian Arctic whitefish compared to levels observed in Great Slave Lake whitefish in 1995 (Figure 4·24). PCDDs in the Russian fish were dominated by octachlorodibenzo-*p*-dioxin, and 2,3,7,8-tetrachlorinated dibenzofuran (TCDF) had the highest concentrations among PCDFs.

#### Great Slave Lake study

A variety of studies have been conducted investigating contaminants in biota in Great Slave Lake, Canada. Most of these studies were conducted in the mid-1990s, but monitoring has continued for a number of fish spe-



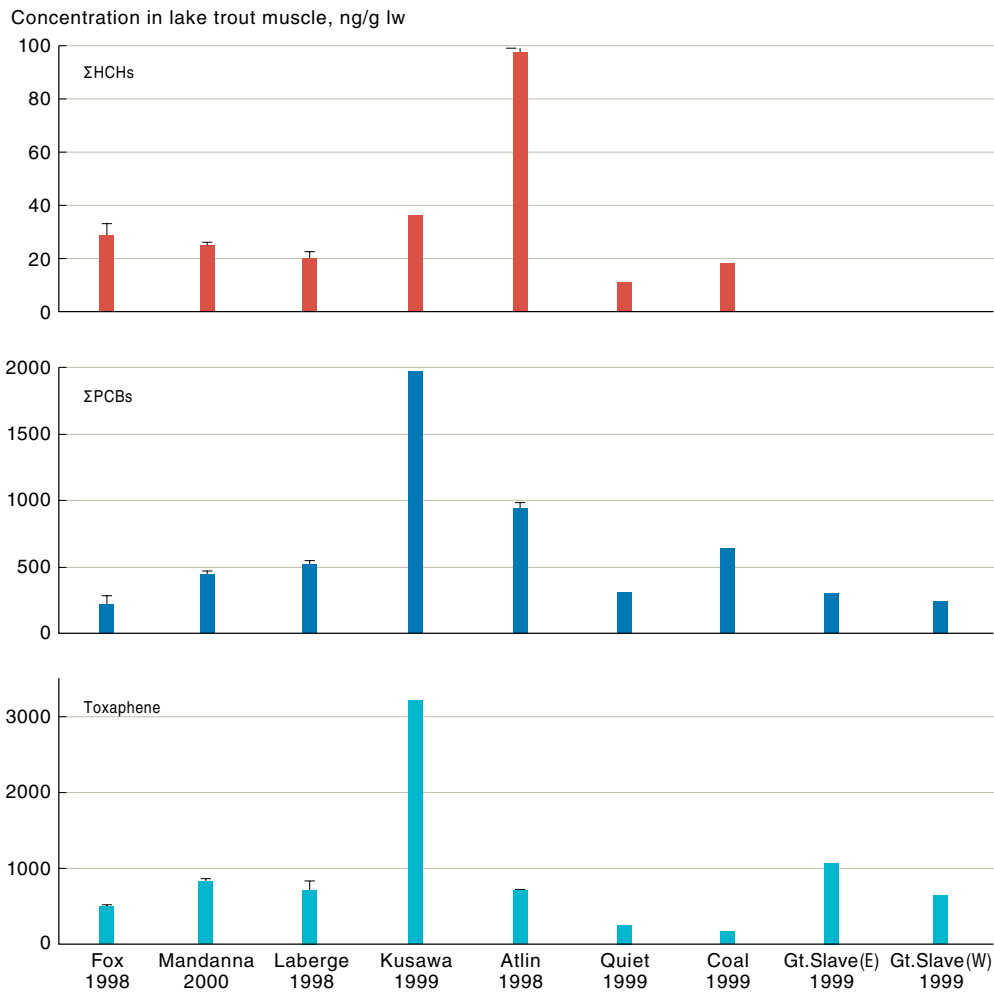


Figure 4-25.  $\Sigma$ HCH,  $\Sigma$ PCB, and toxaphene concentrations in lake trout from the Canadian Arctic. Great Slave Lake data from Evans and Muir (2001), all other data from Roach (2002).

source of higher levels in fish from some lakes in Alberta (Blais *et al.*, 2001). The levels of the different OC groups are not consistent between Kusawa Lake and Atlin Lake, however, and OC levels in burbot liver from these two lakes are not higher than those observed in other Yukon and NWT lakes. These data need to be assessed in light of variable biological characteristics of the fish collected from each lake. For example, lipid was found to be a significant co-variate of OC concentrations in lake trout from the Yukon lakes (Stern *et al.*, 2000). Concentrations (lipid corrected) of most OCs were higher in the Yukon lakes compared to Great Slave Lake (Figure 4-25).

Lake trout from several sites in northern Norway had  $\Sigma$ PCB levels of 0.23-3.2 ng/g ww in muscle (Annex Table 7) (Schlabach *et al.*, 2001). PCDD/F and non-ortho PCB levels expressed as TEQs were 0.17-0.84 pg/g ww (13-34 pg/g lw) (Annex Table 16).

#### Burbot

There is a fairly large dataset on OCs in burbot that was reported in the previous AMAP assessment (de March *et al.*, 1998). Levels of OCs in burbot have received attention because indigenous people consume their high-lipid liver, and the species has a wide distribution. In addition to Russian studies discussed above, a small number of studies have been carried out on POPs in burbot in North America since the previous AMAP assessment, and all were focused on Alaska and the western Canada Arctic.

Levels of POPs were assessed in burbot livers from Alaska (Fairbanks and Yukon Flats, Tetlin, and Kanuti National Wildlife Refuges on the Yukon, Koyukuk, and Tanana rivers) in 1998, in part, to assess the influence of long-range transport of POPs on the freshwaters of the Alaskan interior (Mueller and Matz, 2000) (Figure 4-23). This study highlighted the influence of biological variability on OC concentrations. Lipid concentrations were significantly positively correlated with dieldrin and heptachlor epoxide concentrations. Weight was significantly positively correlated with  $\alpha$ -chlordane, dieldrin, HCB, heptachlor epoxide, mirex, toxaphene, and *trans*-nonachlor concentrations. This study also showed that OC concentrations could vary between fish populations that are fairly close geographically. General patterns of greater OC contamination in burbot livers from Yukon Flats and Fairbanks compared to Tetlin and Kanuti were consistent, with significant differences in concentrations for some analytes. Analysis of the data was complicated by differing lipid concentrations in samples, differing fish weights among sites, and by a small sample size at Yukon Flats. The greater concentrations of DDT and its metabolites and  $\Sigma$ PCBs found at Fairbanks compared with other sites likely reflect historical use of these compounds within the city of Fairbanks and at nearby military bases. Toxaphene concentrations were generally low.

There were variations in OC concentrations in the liver of burbot collected in the Alaskan, Russian, and Canadian Arctic (Figure 4-23). The highest OC concentrations, including  $\Sigma$ PCBs and  $\Sigma$ DDTs, were found in

Lake Laberge burbot, but these OC levels are similar to previous measurements in burbot from this lake (Kidd *et al.*, 1995; 1998). Burbot from Fairbanks and Yukon Flats had somewhat lower mean  $\Sigma$ PCB concentrations than at Lake Laberge. The  $\Sigma$ PCB concentrations observed in burbot of Fairbanks and Yukon Flats are likely due to local sources. Mean  $\Sigma$ PCB concentrations in Russian burbot from Dudinka were similar to levels found in burbot from Fairbanks and Yukon Flats, but  $\Sigma$ DDT levels were higher. Information is lacking to completely evaluate the Russian data.

For the remaining lakes, with the possible exception of Lake Laberge, atmospheric deposition is the dominant route of OC delivery (Kidd *et al.*, 1995). For these lakes, OC levels varied much less and were generally within a factor of two of each other. The OC concentrations in burbot from these lakes are lower than those previously reported for burbot collected from the Mackenzie River in the Canadian Arctic in the 1980s (Muir *et al.*, 1990a). For Canada, all of these sites are fairly close to the Mackenzie, so the lower concentrations in the recently collected burbot most likely reflect temporal changes as opposed to spatial differences between the sites (see Section 5.3.3).

Burbot liver from northern Norway had TEQ levels based on PCDD/Fs and non-*ortho* PCBs (see Annex Table 16) of 12.8 pg/g ww (110 pg/g lw) (Schlabach *et al.*, 2001).

#### Landlocked Arctic char

Landlocked char have been collected over a number of years from lakes near the community of Qausuittuq (Resolute) in the Canadian High Arctic (Muir *et al.*, 2001a). The effect of diet and trophic level on OC levels was investigated using stable isotopes of carbon and nitrogen. A few fish have  $\delta^{15}\text{N}$  values that were up to 3.8 parts per thousand (‰) higher than others. This reflects differences in trophic level because concentrations of the heavier isotope of nitrogen,  $^{15}\text{N}$ , are progressively enriched from prey to predator at an average of 3 to 5 ‰ (Peterson and Fry, 1987). Landlocked char populations are known to develop a behavioral dichotomy wherein some individuals at a certain 'escape size' adapt to a cannibalistic feeding habit, while other individuals may not. In Char Lake, a system containing only char, Hobson and Welch (1995) associated  $\delta^{15}\text{N}$  values of 13.7 ‰ with piscivory in char. They also found a significant increase in the  $\delta^{15}\text{N}$  of these fish with size that they attributed to cannibalism within the population. There were no significant correlations of  $\delta^{15}\text{N}$  with length or weight in Resolute Lake char.  $\Sigma$ PCB,  $\Sigma$ DDT, and  $\Sigma$ CHL concentrations (lipid-normalized) were significantly correlated with  $\delta^{15}\text{N}$  in char from Resolute Lake collected in 1997 and 1999, suggesting that biomagnification of OCs is occurring within the char population due to the presence of piscivorous char. These results support the hypothesis (Muir *et al.*, 2001a) that the reason for higher levels seen in char sampled in 1993 in Char Lake was that piscivorous char were analyzed.

In 1998, dorsal axial muscle from landlocked (non-anadromous) Arctic char from the lakes Ellasjøen and Øyangen (all caught in 1996) located on Bjørnøya, were analyzed for stable isotopes of nitrogen, PCBs, and a number of pesticides. An overview of the Bjørnøya lake

studies is provided in Section 4.3.5. The  $\delta^{15}\text{N}$  values in muscle tissue of Arctic char were generally higher in Ellasjøen (mean 18.1‰) than in Øyangen (mean 8.8‰), suggesting a different food web structure between the lakes or an input of N that was enriched in  $^{15}\text{N}$ . Consistent with the higher  $\delta^{15}\text{N}$  values, the mean concentrations of OCs were greater in the Ellasjøen char compared with the Øyangen char. The largest concentration differences were found for the heavier PCB congeners,  $\Sigma$ DDTs, and oxychlordane (Figures 4-27 and 4-28). There was no significant difference for HCHs.  $\Sigma$ PCB concentrations (seven congeners) in Arctic char were 694 ng/g ww for fish from Ellasjøen and 49 ng/g ww for fish from Øyangen. The congener pattern was generally similar for both lakes, with CB138 and CB153 having the highest levels, followed by CBs 180, 118, 156, 105, and 101. However, CB138 co-eluted with CB160 and 163, leading to an overestimation of this value. The mean concentration of *p,p'*-DDE in char muscle tissue was 57.7 ng/g ww in Ellasjøen and 3.4 ng/g ww in Øyangen. Over 97% of the  $\Sigma$ DDT in Arctic char from Ellasjøen was *p,p'*-DDE. In Øyangen, the corresponding number was 79%. The concentrations of chlordane, heptachlor, heptachlor epoxide, aldrin, and trifluralin were below detection limits in almost all fish samples.

In 1999, small Arctic char (from 7-64 g, presumed to be prey fish) from the two lakes (Ellasjøen and Øyangen,  $n=3$  for both lakes) were analyzed in order to supplement the already analyzed material (consisting mostly of larger fish). In these smaller Arctic char,  $\delta^{15}\text{N}$  values in Ellasjøen char were higher than in the char from Øyangen (average 17.42 and 8.65‰, respectively) but were very similar to the larger Arctic char sampled previously for these two lakes. Not surprisingly, OC concentrations were very different between the two lakes for the small char but very similar to results in the larger char. The average  $\Sigma$ PCB<sub>7</sub> concentration in small Arctic char from Ellasjøen was 642.7 ng/g ww (range 373.6-906.6 ng/g ww) and the corresponding value for small Arctic char from Øyangen was 37.1 ng/g ww (range 18.2-55.9 ng/g ww). For  $\Sigma$ PCB, concentrations were 1139.1 ng/g ww (range 596.0-1629.6 ng/g ww) for small Arctic char from Ellasjøen and 64.8 ng/g ww (range 32.2-97.5 ng/g ww) for small Arctic char from Øyangen.

In another study in Ellasjøen, Arctic char were found to have PCDD/F and non-*ortho* PCB concentrations (see Annex Table 16) expressed as TEQs of 7.4 pg/g ww (566 pg/g lw) (Schlabach *et al.*, 2001). The PCBs contributed most to the TEQs.

Arctic char were sampled from a mountain lake in the Faroe Islands in 2000 and 2001 ( $n=25$  and 40, respectively) and muscle tissue was analyzed for OCs (Hoydal *et al.*, 2001). The single OC occurring with the overall highest concentration was *p,p'*-DDE, with a mean of approximately 1 ng/g ww in a char size group of 36-38 cm fork length. The next highest concentrations were for HCB and CB153 at 0.9 ng/g ww for both. Concentrations of CB153 and DDE increased with fish length but HCB did not.

Arctic char from a freshwater lake on the island of Jan Mayen were analyzed for OCs (Gabrielsen *et al.*, 1997). The lake is near cliffs with nesting seabirds. The char have relatively high liver concentrations of  $\Sigma$ PCB (155 ng/g ww) and  $\Sigma$ DDT (71 ng/g ww), which may be

indicative of seabird guano inputs, similar to the situation at Ellasjøen on Bjørnøya.

#### *Faroe Islands brown trout*

In 1997, liver from landlocked brown trout (*Salmo trutta*) were collected for OC analyses from two lakes (Fjallavatn and Leitisvatn) in the Faroe Islands (Larsen and Dam, 1999). Fish from Fjallavatn were combined into two pools, one containing the larger specimens (n=9, mean length 28.6 cm) and one containing the small individuals (n=19, mean length 23.4 cm). The fish from Leitisvatn were of similar length but were combined into two, mixed-size pools. HCB, *p,p'*-DDE and *trans*-nonachlor were detected in the four liver pools at 0.5 ng/g ww, 3-4 ng/g ww and at 1-2 ng/g ww, respectively. For PCBs, only CBs 153, 138 (+163), and 180 were detected in all samples at approximately 2 ng/g ww, 1 ng/g ww and 1 ng/g ww, respectively. These concentrations are similar to those observed in Arctic char from the Faroe Islands (Hoydal *et al.*, 2001).

#### *Yukon River study*

A number of fish species have been sampled and analyzed for OCs from headwater lakes in the Yukon River system since 1996 in response to detection of high concentrations of toxaphene (Palmer and Roach, 2001). Results for lake trout and burbot are covered previously in this section. OC concentrations in whitefish collected in 1998 from Lake Laberge are lower than those observed in lake trout on a wet weight basis but similar when concentrations are normalized to lipid content (Annex Table 7). OC concentrations in Lake Laberge are lower than those observed in Watson Lake (collected in 1997 and 1998), although the Watson Lake whitefish are larger. OC concentrations in inconnu collected from Peel River in 1999 are low compared to most other species of fish from the Yukon region, but are similar to inconnu collected in Great Slave Lake in 1996 (Annex Table 7). Concentrations of OCs in fish from the Yukon region are consistently below levels that result in any health advisories (Palmer and Roach, 2001).

#### *Finnish Arctic fish*

Recently a study was conducted on levels of OCs in the muscle tissue of four species of freshwater fish (Arctic char, northern pike, European perch (*Perca fluviatilis*), and whitefish) in Finland (Mannio, 2002) (Annex Table 7). A majority of OC concentrations were less than 1 ng/g ww, one to two orders of magnitude below concentrations observed in similar species in the Canadian Arctic. The lipid content of the muscle tissue of these fish was very low (<1%), which explains some of the differences with the Canadian samples. One group of whitefish collected in the river Tornionjoki had much higher OC concentrations than all other fish collected from Finland (see Annex Table 7) but this was because these fish were anadromous.

#### *Alaskan slimy sculpin*

Slimy sculpin (*Cottus cognathus*) from twelve locations within the Cook Inlet Basin were collected to assess levels of OCs in this region (Frenzel, 2000). These twelve locations included sites along roadways and in remote

areas. This region has a population of approximately 350 000 people but with minimal industrial activity. The slimy sculpin is a non-migratory, bottom-feeding fish, which makes it suitable for assessing levels of OCs in sediment. OCs, including PCBs, were detected in just three samples from the 12 analyzed. DDE and DDT concentrations (whole body) of 9.0 and 6.1 ng/g were found at one site; HCB concentrations of 5.7 ng/g at another; and, ΣPCB concentrations of 79 ng/g at a third. These concentrations are low and in the range found for other similar benthic fish from this region (see Annex Table 7 for other benthic species).

#### 4.3.4. 'New' chemicals in freshwater fish

There has been limited work on 'new' chemicals in Arctic freshwater fish and most of it has focused on PBDEs (Annex Table 17).

In the European Arctic, data are only available for burbot and trout from northern Norway, and for Arctic char from Ellasjøen on Bjørnøya (Schlabach *et al.*, 2001). Trout muscle ΣPBDE levels (BDEs 47 and 99) are low at 0.10-0.36 ng/g ww (8-14 ng/g lw). Burbot liver had ΣPBDE levels (BDEs 47 and 99) of 20 ng/g ww (175 ng/g lw). For both species, the PBDE levels are a factor of 2-10 times lower than ΣPCB<sub>7</sub> levels. The Arctic char at Bjørnøya have elevated levels of ΣPBDE of 16.3 ng/g ww (1250 ng/g lw), but these are a factor of 30-40 times lower than ΣPCB concentrations. The congener pattern in all three species is evenly distributed between BDE47 and BDE99, and resembles the PeBDE technical product. Measurable amounts of PCNs and SCCPs were also found in these fish. PCN concentrations were 8.6-16 ng/g ww in trout muscle, 13 ng/g ww in Arctic char muscle and 643 ng/g ww in burbot liver. SCCP concentrations were 3.3 ng/g ww in trout, 6.9 ng/g ww in Arctic char, and 38 ng/g ww in burbot.

PBDEs were measured in the livers of burbot collected in the Mackenzie River (Canada) in 1988, 1999, and 2000 as part of a project on temporal trends (Stern *et al.*, 2001a) (see Section 5.3.3). BDEs 47 and 99 were the predominant congeners in the burbot, but levels (<2 ng/g lw per congener) were below those observed in the Norwegian fish (Annex Table 17). BDE congeners 100, 153, and 154 were also detected in the burbot but at levels lower than congeners 47 and 99.

#### 4.3.5. Bjørnøya lake study

In 1994, an investigation of contaminants in sediments and fish from Ellasjøen on the island of Bjørnøya was initiated (Evenset *et al.*, 2002). This pilot study was based on a very limited number of samples, but found some of the highest PCB and DDT concentrations in freshwater sediments and fish ever seen in the Arctic. Other studies from the same area have shown high PCB levels in glaucous gulls (Gabrielsen *et al.*, 1995; Bustnes, 1998). In a follow-up study, the contamination level in Ellasjøen was compared to Øyangen, another lake on Bjørnøya. Plankton, benthic animals and landlocked Arctic char from both lakes were analyzed for contaminants and stable isotopes to provide information on contaminant levels and bioaccumulation in the food chain.

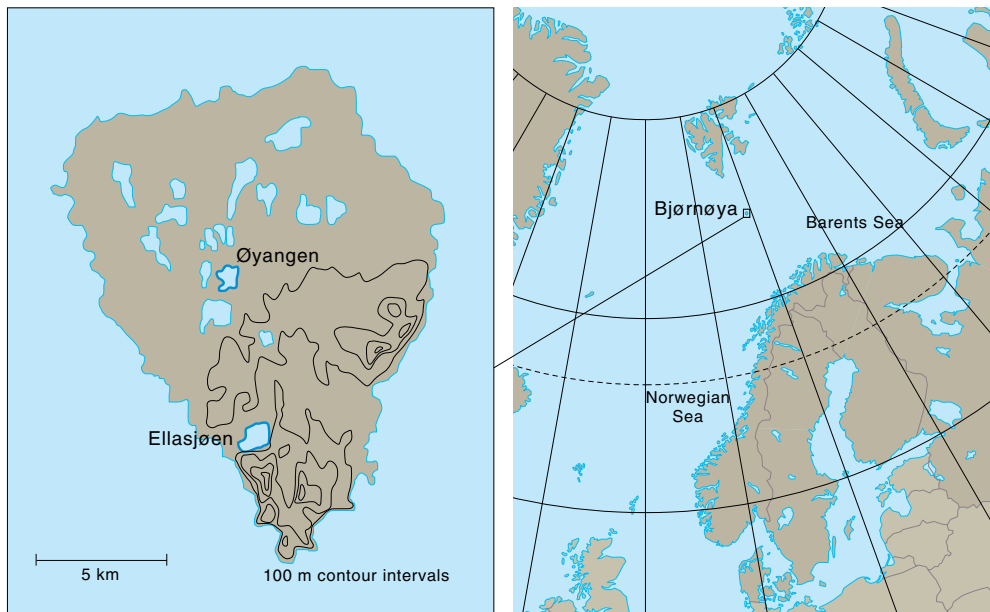


Figure 4-26. The locations of Bjørnøya, and the lakes Ellasjøen and Øyangen (from Evenset *et al.*, 2002).

Bjørnøya (74°30'N, 19°E) is situated halfway between the mainland of Norway and Svalbard (Figure 4-26). Bjørnøya has a surface area of 178 km<sup>2</sup> and measures 20 km from north to south. The annual average temperature is -3.8°C. Precipitation is low, with an annual rate of 367 mm (information from Det Norske Meteorologisk Institutt). The southern part of the island is mountainous with altitudes up to 536 m asl. Seabirds breed on the steep cliffs along the south coast. The main species are common guillemot (*Uria aalge*), Brünnich's guillemot (*Uria lomvia*), kittiwake (*Rissa tridactyla*), little auk/dovekie (*Alle alle*), and glaucous gull. The terrain in most of the middle and north parts of Bjørnøya is flat (30-40 m asl). The island has about 740 lakes which cover 11% of the surface area. More than 600 of these lakes are found on the central and northern plains, and their depths seldom exceed 2 m. Fewer than ten lakes reach depths between 5 and 10 m and these are found in the southern parts of the island. Ellasjøen is the deepest lake on the island (maximum depth of 34 m). Water temperatures are low, typically below 7°C in summer. There is no summer stratification, but the lakes are cold monomictic (i.e. sub-polar). The upper littoral zone generally comprises a rocky margin, 0.5-1 m deep.

Ellasjøen, with an area of 0.72 km<sup>2</sup> (21 m asl), is situated in the southern, mountainous part of Bjørnøya. The catchment area is 6.1 km<sup>2</sup>, and the lake has an estimated retention time of 6.5 years. In the southern part of the catchment area (Alfredfjellet), there is a large colony of little auks. Large flocks of kittiwakes (hundreds to thousands) can be observed on the lake during the summer period. At the outlet of the lake, there is a colony of glaucous gulls (approximately 100 pairs). The birds present in the catchment area seem to have a large impact on the lake, as indicated by the growth of green algae on the shores directly below the colony. Mosses and lichens occur around the northern and western shore close to the lake, although in general, the catchment area is devoid of vegetation. The lake has a very well developed profundal and pelagic zone, and is inhabited by landlocked Arctic char (Klemetsen, 1985).

The littoral zone in the northern part of the lake is also well developed but in the southern part it is very steep, and near the shore the depth is 15 to 20 meters.

The lake Skutilen is situated in the same catchment area as Ellasjøen. The lake has an area of 0.08 km<sup>2</sup> and a maximum depth of 2 m. The primary outlet flows to Ellasjøen. Birds are seldom seen on Skutilen, but Arctic char have been observed in this lake. There is almost no vegetation in the catchment area, except for some mosses. The lake has no pelagic or profundal zone.

Øyangen (33 m asl) is situated on the central plains of Bjørnøya. The lake has an area of 0.35 km<sup>2</sup> and a maximum depth of 5 m. Most of the lake, however, is less than 1 m deep. The total catchment area is approximately 2.2 km<sup>2</sup>, and the estimated retention time is 0.9 years. The area around Øyangen is very flat and consists mainly of boulders. On Øyangen, flocks of kittiwakes can be observed, but the flocks are smaller and not seen as frequently as on Ellasjøen. There is almost no vegetation in the catchment area except for some mosses. The lake has no pelagic or profundal zone, but Øyangen has a population of landlocked Arctic char.

The studies carried out in the limnic ecosystems on Bjørnøya have shown that Ellasjøen is significantly more contaminated by OCs than Øyangen (Figures 4-27 and 4-28), and that Ellasjøen has a higher 'basic trophic level' (Skotvold *et al.*, 1999). Possibly, the higher 'basic level' of isotopes is caused by guano-input to the lake. Seabirds feed in the marine environment and may bring in nutrients (through guano) with different isotopic composition than other nutrients that are transported to the lake (i.e. runoff from land, decomposed material). Guano can also function as a transport medium for contaminants from the marine environment to the limnic environment in Ellasjøen. This theory was also supported by the fact that there was a similarity between the CB99:CB101 ratios in guano samples of seabirds and char from Ellasjøen, but none in char from Øyangen.

Analysis of toxaphene congeners provided further evidence of the influence of seabird guano. From the nine chorobornane congeners analyzed, only three, Parlars 26, 40, and 50, were found in considerable amounts in



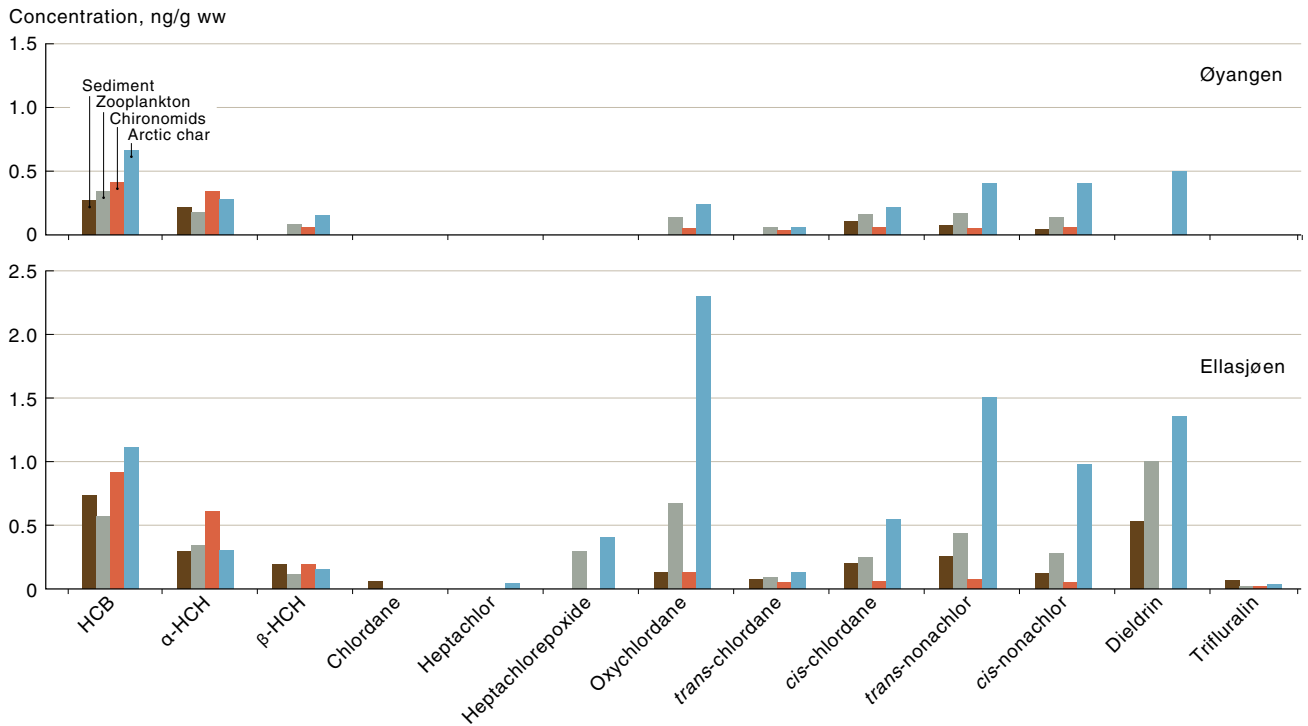


Figure 4-27. Concentrations of pesticides in sediment and biota from the lakes Ellasjøen and Øyangen on Bjørnøya (from Evenset *et al.*, 2002).

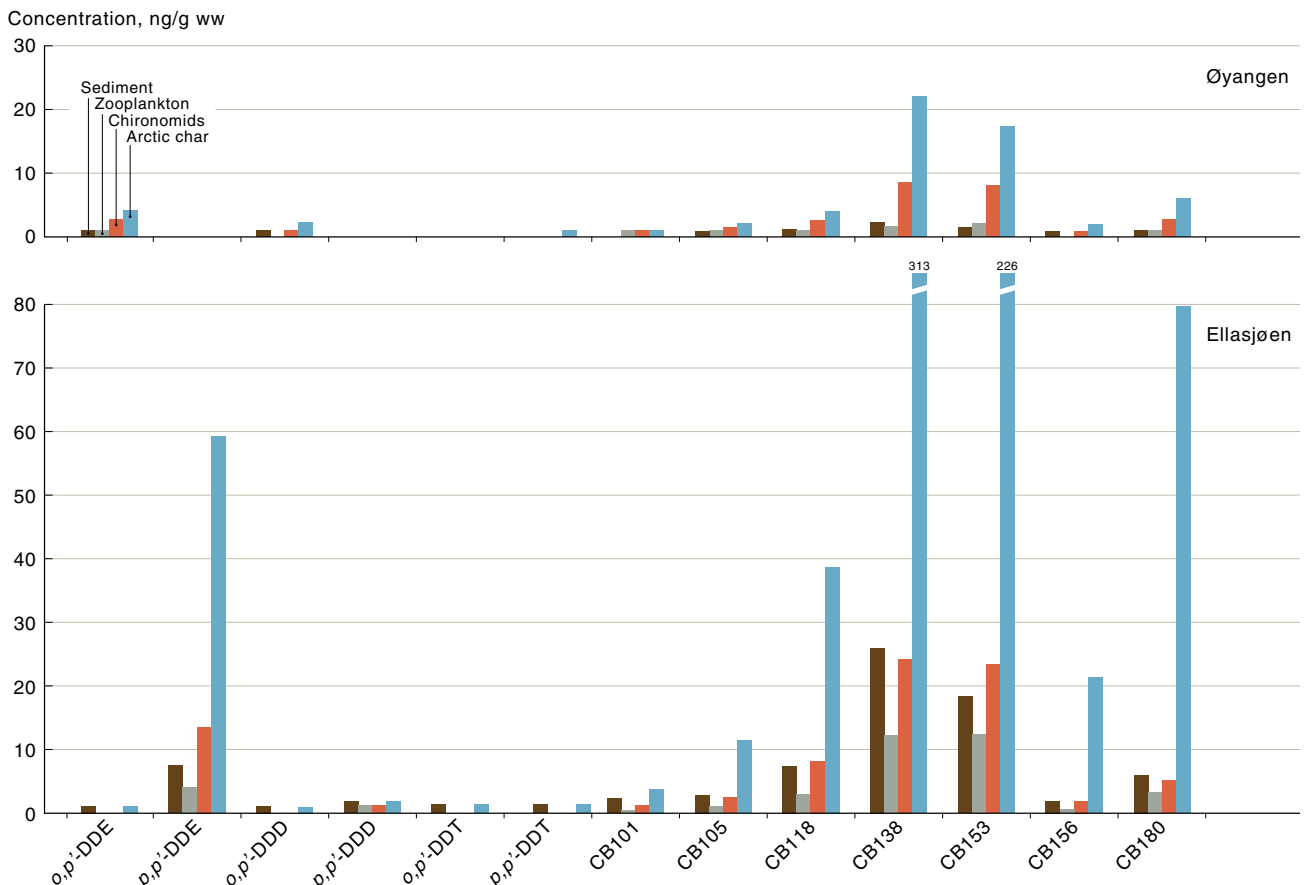


Figure 4-28. Concentrations of DDT (including metabolites) and seven PCB congeners in sediment and biota collected from the lakes Ellasjøen and Øyangen on Bjørnøya in 1996 (from Evenset *et al.*, 2002). Note that CB138 may have co-eluted with CB160 and 163, inflating its value.

the analyzed biota samples from Bjørnøya. Levels found for all samples from Øyangen were very low, often not exceeding the detection limits. The generally low levels found at Øyangen were consistent with low levels of PCBs found in this lake. As with the PCB results, con-

siderably higher chlorobornane levels were found in biota samples from the Ellasjøen catchment area. Almost no chlorobornanes were observed in zooplankton from Øyangen. In Ellasjøen, Parlar 50 concentrations of PCBs found in this lake. As with the PCB results, con-

cant increase from zooplankton (sum of Parlars 26, 40 and 50: 1.2 ng/g ww) to Arctic char (average sum of Parlars 26, 40 and 50: 8.1 ng/g ww) was found. Glaucous gull gut from Ellasjøen had the highest toxaphene concentrations (sum of Parlars 26, 40, and 50: 84.2 ng/g ww). Although significant differences were found between Øyangen and Ellasjøen samples, the highest chlorobornane concentrations, found in the pooled Ellasjøen glaucous gull gut sample, cannot be considered unusually high compared to published literature data reported for Arctic marine top predators. A comprehensive review article presents levels up to 4000 ng/g ww ( $\Sigma$ chlorobornanes) for marine mammals (de Geus *et al.*, 1999). However, based on the results of the present study, glaucous gull guano was identified as one of the major chlorobornane sources for the Ellasjøen freshwater system.

The Bjørnøya lake study has highlighted the important influence of locally defined and restricted ecological, geographical, and meteorological environmental factors on overall contaminant levels (e.g., precipitation, presence of local seabird nesting colonies, and resting areas). Based on these results, seabirds at Bjørnøya seem to function effectively as a transport link for POPs from the marine ecosystem to the freshwater ecosystem. It can also be assumed that in other locations seabird guano may be a potential contamination source for persistent pollutants, for example on Jan Mayen. Not only in Arctic regions can such a seabird linkage between freshwater and marine ecosystems result in elevated OC concentrations.

#### 4.3.6. Summary and conclusions – freshwater environment

The years 1996-2001 saw few new studies of POPs in river water in the Canadian Arctic, northern Norway or in northern Russia, although a major project was initiated in Russia in 2001-2002 to study contaminants in the rivers Yenisey and Pechora (RAIPON/AMAP/GEF Project, 2001). However, results reported in the previous AMAP POPs assessment were independently verified by several researchers, confirming the original results and deriving flux estimates for  $\Sigma$ DDTs and  $\Sigma$ HCHs. The flux estimates suggest that, at least until 1996, rivers such as the Ob and Yenisey contributed significant amounts of  $\Sigma$ DDTs and lindane to the Arctic Ocean, assuming that what was measured at downstream sampling stations was eventually carried to the northern Russian seas.

New measurements of OC pesticides and PCBs in freshwater sediments were available for lakes in Canada, on Svalbard and Bjørnøya, and on the Kola Peninsula. The limited number of studies provided evidence for contamination from both local sources and long-range transport. In the case of Ellasjøen, levels of  $\Sigma$ PCBs and  $\Sigma$ DDTs were up to ten times higher in its surface sediments compared to Øyangen surface sediments due to inputs of seabird guano. This unusual input pathway was not important in Canadian Arctic lake sediments; however, local sources from past uses of DDT, toxaphene, SCCPs, and PCBs were clearly important in explaining some differences in fluxes of these compounds in Yukon lakes. The significant declining trend of PCBs and DDT fluxes with latitude in lake sediments within the Cana-

dian Arctic may be partly attributable to the influence of local sources in the Yukon and the complete absence of these sources in most of the High Arctic lakes.

The recent data for POPs in freshwater are too limited to draw any additional conclusions about spatial trends or the presence of new contaminants. Results from studies of Canadian Arctic lakes did provide new information on degradation pathways of  $\alpha$ -HCH, indicating relatively rapid degradation compared to ocean waters and an inverse relationship between enantioselective degradation and lake trophic status. These findings may be useful for interpreting the fate of other POPs in Arctic lake waters.

There has been a considerable amount of research focused on OCs in Arctic freshwater biota since the first AMAP POPs assessment, although less than that carried out in the marine environment. Similar to the terrestrial environment, levels of OCs in most freshwater biota of the Arctic are low compared with the marine environment but are similar to terrestrial predators. The exceptions are freshwater systems that receive additional POP contamination via non-atmospheric routes. An excellent example is the high levels of POPs found in the sediment and biota of Ellasjøen on Bjørnøya. This lake receives a large amount of seabird guano, which is likely acting as a vector to transport and concentrate POPs from the marine environment and deposit them in a freshwater environment.

Toxaphene and PCBs continue to be the predominant OCs measured in freshwater biota, as was observed for Canada and Greenland in the first AMAP POPs assessment. New data from Bjørnøya suggest that toxaphene is a major OC contaminant in freshwater invertebrates and fish of the European Arctic. Toxaphene and PBDE were not found in Russian water or sediments, but the detection limits were above levels normally found in the Arctic. PBDEs were found to be higher in freshwater fish from Bjørnøya lakes and northern Norway compared with fish from the Mackenzie River in Canada. Levels were well below legacy OCs and lower than levels of PBDEs in marine biota. Lack of data on 'new' chemicals, particularly perfluorinated compounds, is a significant gap for the freshwater environment.

The Bjørnøya study and results from Alaskan and Russian burbot studies all highlight the potential for OC levels to vary significantly between geographically close locations. Elevated levels of OCs can occur due to local point sources, although these are rare in the Arctic, or through unique ecological or biological scenarios. The results of the Alaskan burbot study also highlight the importance of biological characteristics on fish OC levels. Proper statistical analysis of spatial and temporal trends of OCs in freshwater fish requires biological data, in particular size and age. Other factors, such as meltwater from glaciers, may also influence OC levels in fish.

$\Sigma$ PCB concentrations in many of the predatory freshwater fish of the Canadian, Russian, and Alaskan Arctic, as well as fish from Ellasjøen on Bjørnøya exceeded the most conservative guidelines for the protection of aquatic wildlife of 15-48 (Canada) to 160 (USEPA) ng/g ww (de March *et al.*, 1998) (Table 6.1). Few fish exceeded the U.S. guidelines. Information on the health of these fish populations is limited but there is no evidence to suggest that the populations are currently compro-

mised. No fish muscle samples were found to exceed the PCB guideline limit for human consumption and export of fish of 2 µg/g ww, which is widely used in Europe and North America.

Circumpolar trends of OCs in freshwater fish were evaluated using the Arctic char in the first AMAP assessment report (de March *et al.*, 1998). Circumpolar OC data for freshwater fish have not become available since that report, are not available even for Arctic char and, in particular, are lacking for invertebrates. However, evaluation of OC levels in different species of predatory fish suggest that concentrations of OCs in freshwater biota of the circumpolar Arctic are similar with a few notable exceptions (e.g., Ellasjøen on Bjørnøya), and lakes in the Yukon Territory in the western Canadian Arctic. New data for OC concentrations in Russian freshwater fish are difficult to assess as little biological information is available, but they suggest that some freshwater systems of the Russian Arctic are among the most contaminated in the circumpolar Arctic (see Section 4.3.3.2). Slight differences in OC levels of predatory fish within Great Slave Lake, one of the largest lakes within the Arctic, provide evidence that abiotic environment and/or food web characteristics can influence OC levels.

#### 4.4. Marine environment

The marine environment has historically received the most attention with regard to OC and PAH contaminants in the Arctic environment. It was also the first Arctic system to be examined for the presence of OC contaminants (Holden, 1970). The level of research on OC contamination of the marine environment has continued to outdistance efforts in the freshwater and terrestrial Arctic. This is clearly due to the higher levels observed in this system and the role of marine organisms in the diet of northern populations. Since the first AMAP POPs assessment (i.e. post 1996), there has been a large amount of data produced for the marine environment, in particular for seals and whales, and a number of comprehensive studies on dynamics and mechanisms of OCs in this environment. There have also been new initiatives to produce data for regions that were under-represented in the previous AMAP assessment, in particular the Alaskan and Russian Arctic.

This review and assessment of the post-1996 data will focus on persistent organohalogen compounds (mainly OCs, as well as recent measurements of brominated and fluorinated organics and TBT). However, PAHs are not included because sources in the aquatic marine environment are considered to be predominately of petrogenic origin (either from natural sources or anthropogenic uses) rather than from combustion and atmospheric deposition sources (Robertson, 1998). A future review of petrogenic PAHs will be conducted under the updated AMAP assessment of 'petroleum hydrocarbons' that is due to be published in 2006.

##### 4.4.1. Seawater

###### 4.4.1.1. Overview

A large number of measurements of OC pesticides and PCBs have been made in Arctic Ocean waters in the period of 1996 to 2001 (Annex Table 8) mainly as a result

of scientific cruises organized by circumpolar countries. During this period, there were also several peer-reviewed publications on results of measurements of OCs in seawater pre-1996 (Rice and Shigaev, 1997; Hargrave *et al.*, 1997; Jantunen and Bidleman, 1998; Strachan *et al.*, 2001); however, those results were included in the previous assessment (de March *et al.* 1998) and reviews (Macdonald *et al.*, 2000), and are included only for comparison here.

The new measurements have, for the most part, been conducted from oceanographic research vessels in a series of cruises (Figure 4·29). The Swedish *Oden* cruise in 1996 conducted sampling from the ice edge to the North Pole in the northern Barents Sea (Harner *et al.*, 1999). In 1997, a Canadian supply trip (JOIS) for the Surface Heat Budget of the Arctic (SHEBA) study (Perovich *et al.*, 1999) was used to obtain seawater from the western Canadian Arctic Archipelago in September. In May-June 1998, the Northwater Polyna study in Northern Baffin Bay (NOW '98) provided another platform for sampling. In 1997-1998, the SHEBA study permitted continuous sampling of seawater over the Beaufort/Chukchi Seas. The Swedish Tundra Northwest study (TNW '99) traversed the Canadian Arctic Archipelago during July/August, 1999. An Arctic to Antarctic cruise by the RV *Polarstern* provided seawater samples from the East Greenland Sea in 2000 (Lakaschus *et al.*, 2002). Schultz-Bull *et al.* (1998) reported PCBs in large-volume samples collected in the North Atlantic Ocean around Iceland which are also relevant to this discussion and Sobek and Gustafsson (2002) presented preliminary results from the *Oden* cruise to the North Pole in 2001. Studies have also been conducted in the Marginal Ice Zone of the Barents Sea north of Svalbard (Olsson, 2002) and in the Laptev Sea (Utschakowski, 1998).

The new measurements have filled in some of the information gaps on spatial trends of OCs in the Arctic Ocean. Sampling and extraction techniques were more uniform than in the 1980s and early 1990s, with large-volume (20 to >100 L) samples collected and extracted using resin columns to minimize airborne contamination. Some investigators used filtration (Harner *et al.*, 1999) or continuous centrifugation (Strachan *et al.*, 2001) to remove particles; however, samples for HCH analyses were often unfiltered (Lakaschus *et al.*, 2002). HCH isomers were the most commonly measured chemicals in the Arctic Ocean and adjacent seas. These compounds are present at ng/L concentrations and there are few problems with contamination on ships or in the laboratory. PCBs were the next most prominent contaminants (Annex Table 8); however, there were far fewer measurements. PCB measurements in seawater are challenging because of low levels and potential for shipboard as well as laboratory contamination. Comparison among different studies and with previous work on PCBs in Arctic and northern temperate seawaters (Iwata *et al.*, 1993; Schultz-Bull *et al.*, 1998; Sobek and Gustafsson, 2002) raises questions about contamination and the effects of different sampling techniques. These include possible differences between whole (unfiltered) water versus filtered samples, and the use of *in situ* collection using remotely deployed samplers versus submersible pumping onto the ship.

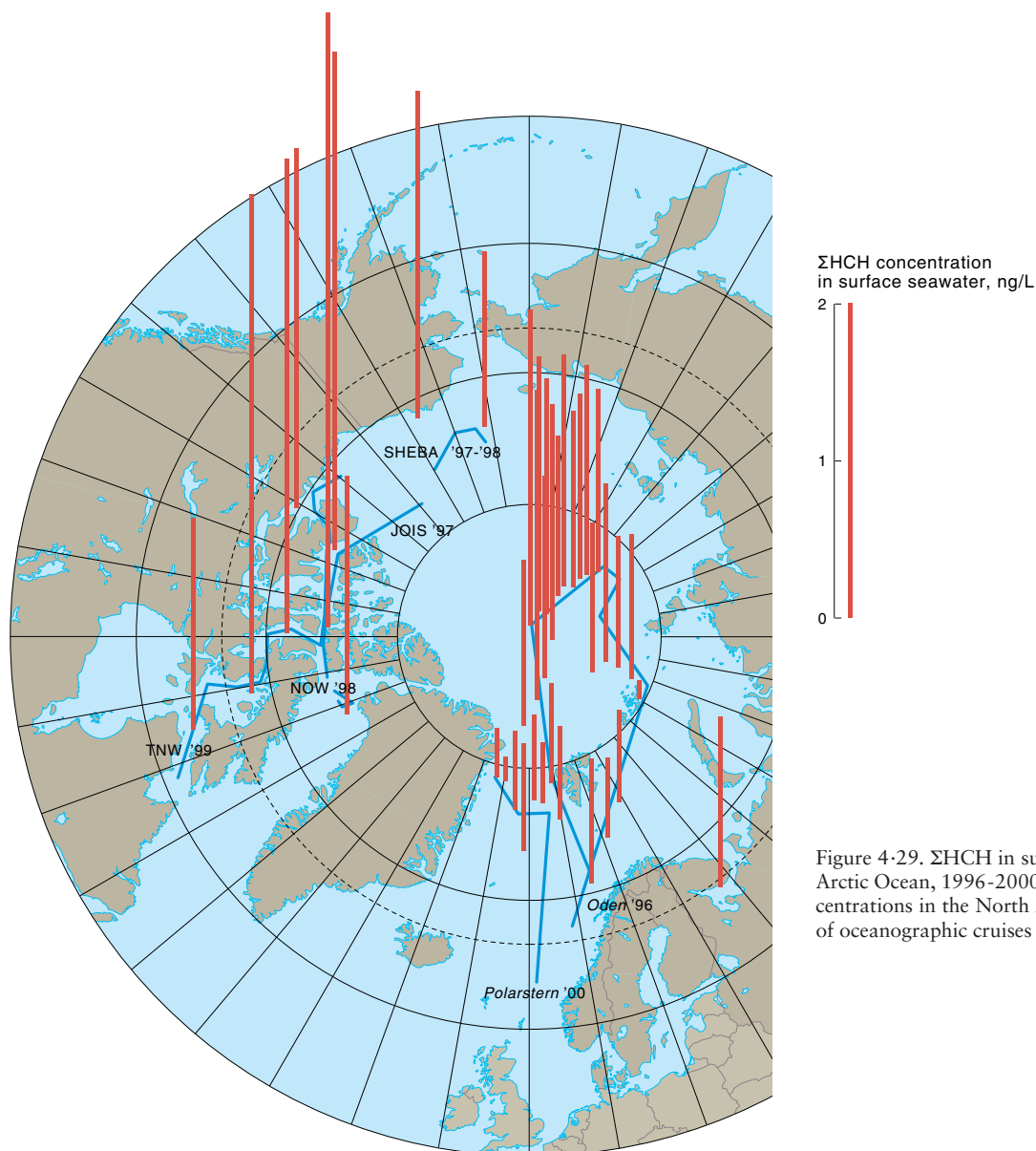


Figure 4-29.  $\Sigma$ HCH in surface seawater from the Arctic Ocean, 1996-2000, illustrating higher concentrations in the North American Arctic. Tracks of oceanographic cruises are shown in blue.

#### 4.4.1.2. HCHs

Harner *et al.* (1999) determined HCH isomers and ERs of  $\alpha$ -HCH in a large series of surface and deepwater samples from the northern Barents Sea and central Arctic Ocean (80-88°N, 11-143°E) (Annex Table 8). The mean concentrations for surface water were  $1.18 \pm 0.408$  ng/L for  $\Sigma$ HCHs. These agreed well with measurements by Strachan *et al.* (2000) on the same cruise and also with values reported by Gaul (1992) for a 1985 survey of the northeast Atlantic ( $\Sigma$ HCHs = 1.25 ng/L). Both  $\alpha$ - and  $\gamma$ -HCHs increased with latitude between 74-88°N ( $r^2 = 0.58$  and  $0.69$  for  $\alpha$ - and  $\gamma$ -HCH, respectively). Values similar to those on the *Oden* cruise were found during 1994 in the eastern Arctic Ocean north of Spitsbergen ( $\Sigma$ HCHs = 1.17 ng/L) and in the Greenland Sea ( $\Sigma$ HCHs = 0.83 ng/L) (Jantunen and Bidleman, 1996; 1998).

Vertical profiles at 11 *Oden* cruise stations indicated that concentrations at depths of 250-1000 m were approximately 40% of surface values for  $\alpha$ -HCH and approximately 60% for  $\gamma$ -HCH (summarized as  $\Sigma$ HCHs in Annex Table 8). Tracer studies in this region (Nansen Basin) indicate that the water masses at 250-1000 m have

ages on the order of 12-20 years, which coincides with the time of heaviest usage of HCH (Li *et al.*, 1998a).

Lakaschus *et al.* (2002) determined  $\alpha$ - and  $\gamma$ -HCH in surface (11 m depth) seawater along an east-west transect in the Greenland Sea. Concentrations in this region ( $\Sigma$ HCHs =  $0.57 \pm 0.20$  ng/L east; 0.13-0.698 ng/L west) were similar to those in surface water and deeper water in the Barents Sea (Harner *et al.*, 1999), but two to four times lower than in the Beaufort Sea (Annex Table 8). However,  $\Sigma$ HCH concentrations in the Greenland Sea (68-75°N) were three to five times higher than in the eastern Atlantic near the Bay of Biscay because of much higher levels of  $\alpha$ -HCH in Arctic waters (Lakaschus *et al.*, 2002).

$\Sigma$ HCH concentrations in the White Sea (Kandalaksha Bay and central basin) in northwestern Russia were similar to those in the Beaufort Sea ( $1.1 \pm 0.11$ ) and two-fold higher than in the Barents Sea near Svalbard (Harner *et al.*, 1999; Muir *et al.*, 2002a). In general, surface concentrations of HCHs are highest in the central Canadian Arctic Archipelago, intermediate in the Beaufort/Chukchi Seas and at the North Pole, and lowest in the Greenland Sea and northern Barents Sea (Figure 4-29).



The large number of measurements of HCH during the 1980s and 1990s in the Bering/Chukchi/Beaufort Seas, as well as in the Greenland Sea, also permits an examination of mass balances over time and temporal trends. These are discussed in Section 5.1. Other measurements in the nearshore waters of the southern Beaufort Sea also show higher  $\Sigma$ HCH levels (Hoekstra *et al.*, 2002b) than in the eastern Canadian Arctic (northern Baffin Bay).

The lowest  $\Sigma$ HCH concentrations (mean 18 pg/L) were found by Utschakowski (1998) in seawater collected annually in the summers of 1993 to 1996 in the southern Laptev Sea near the Lena River Delta. These concentrations were lower than previously reported levels for the same region (20-150 pg/L; de March *et al.*, 1998).

$\beta$ -HCH, which is not as abundant in the Arctic abiotic environment, has not been as well studied. This is partly due to its low concentrations, which have posed analytical challenges. However, there is growing concern over its potential risks as it is the most bioaccumulating of the HCH isomers and may be an environmental estrogen (Willett *et al.*, 1998). Due to its greater water solubility (low Henry's law constant),  $\beta$ -HCH is more efficiently scavenged by rain and snow than  $\alpha$ -HCH and is preferentially deposited closer to the source regions in high-precipitation areas in the northern North Pacific. Ocean currents then transport  $\beta$ -HCH farther north into the Bering Sea through the Bering Strait (Li *et al.*, 2002). Direct atmospheric transport of  $\beta$ -HCH is a minor pathway. This means that, unlike  $\alpha$ -HCH,  $\beta$ -HCH concentrations in seawater peak in the Bering-Chukchi region and then decrease northward into the Arctic Ocean interior. This indicates that  $\beta$ -HCH that was maximally deposited in the North Pacific during 1970-1985 is now entering the Arctic Ocean as a pulse to the Bering Sea, delayed by some unspecified time, perhaps a decade or more (Li and Bidleman, 2003).

#### 4.4.1.3. PCBs

After the HCHs, PCBs are the next most prominent group of persistent OCs in Arctic seawater. As noted above, comparison among different studies and with previous work on PCBs in the Arctic Ocean and nearby northern seas (Iwata *et al.*, 1993; Schultz-Bull *et al.*, 1998; Sobek and Gustafsson, 2002) raises important questions about contamination and effects of different sampling techniques. Schultz-Bull *et al.* (1998) and Sobek and Gustafsson (2002) have reported concentrations of PCBs which are about ten times lower than other measurements summarized in Annex Table 8. These latter groups have used ultra-clean techniques. Schultz-Bull *et al.* (1998) employed the 'Kiel' *in situ* sampler, a remotely deployed filtration/extraction system (Petrick *et al.*, 1996) to sample large volumes of seawater (230-1070 L) through glass fiber filters (GFF) and XAD-2 resin. Sobek and Gustafsson (2002) employed a stainless steel seawater intake system directly into the *Oden* followed by on-line collection using GFF and polyurethane foam (PUF) in a 'clean' room on board. Seawater sampling on the earlier *Oden*, TNW, and JOIST cruises for PCB analysis involved use of a stainless steel lined 'Go-Flo' bottle followed by GFF and XAD-2 resin but did not employ clean room techniques

during the sampling phase. Sampling on the SHEBA study and nearshore samples at Barrow, Holman, and in the White Sea used *in situ* samplers (Axys Instruments, Sidney BC) and GFF/XAD-2 resin deployed from ships or through sea ice.

The results in Annex Table 8 summarize the current state of the art of PCB measurements in the Arctic Ocean. Up to 100 PCB congeners were determined by some investigators; however, for this assessment results for the sum of ten major PCB congeners were also included, where possible, to make comparisons more equitable. Although all studies employed solid phase extraction (mainly XAD-2 resin) for collection of PCBs, and followed strict protocols to avoid contamination, there is nevertheless major disagreement between measurements, especially between those made using samples collected with 'Go-Flo' bottles or other large-volume containers and those with *in situ* or direct online sampling of water. There are also major differences between results for PCBs determined by *in situ* sampling with 'Axys' systems and the 'Kiel' sampler.

There are similar irreconcilable differences between results from Strachan *et al.* (2001) for the PCBs in the Bering Sea and results from Iwata *et al.* (1993) from the Bering Sea (previously reported in the 1998 AMAP assessment). The latter study used GFF/PUF sampling from an online water system while the former used a submersible pump, a continuous centrifuge to separate particles and solvent extraction with dichloromethane. Iwata *et al.* (1993) reported mean concentrations of 0.0084 ng/L at three locations in the Chukchi Sea and 0.012 ng/L for four locations in the Bering Sea from their 1990 cruise. Strachan *et al.* (2001) reported dissolved phase concentrations averaging 1.0 ng/L in the Bering Sea (seven sites) and means of 0.6-0.9 ng/L in the Chukchi Sea. Given that both cruises covered the same area and sampled approximately the same depth, it is difficult to attribute the differences except to contamination introduced from the ship. The results of Iwata *et al.* (1993) for the Chukchi Sea are consistent with those of Sobek and Gustafsson (2002), Schultz-Bull *et al.* (1998), and Utschakowski (1998) in terms of the range of concentrations but cannot be compared directly because of different numbers of congeners analyzed.

With these caveats in mind we have limited the discussion of results to data from Schultz-Bull *et al.* (1998), Utschakowski (1998), Olsson (2002), and Sobek and Gustafsson (2002); and to the 'Axys' *in situ* sampler work conducted on the SHEBA study and in nearshore waters (Annex Table 8) recognizing that even the latter results may be confounded by contamination introduced on handling and storage or in the laboratory.

There are too few PCB measurements to assess spatial trends of PCBs, nevertheless some general conclusions can be drawn from the data of Schultz-Bull *et al.* (1998) and Sobek and Gustafsson (2002).  $\Sigma$ PCB concentrations in the upper 50 m water column under the permanent ice cap are much lower (mean  $\Sigma$ PCB<sub>10</sub> = 0.00055  $\pm$  0.00043 ng/L) than in the northern North Atlantic (range 0.0014-0.0021 ng/L). Similar  $\Sigma$ PCB concentrations (0.0016-0.0021 ng/L; dissolved + particulate) were found in an ice-covered and a pelagic sampling site in the marginal ice zone north of Svalbard (Olsson, 2002). Somewhat higher  $\Sigma$ PCB concentrations

(mean in the dissolved phase of 0.0034 ng/L (range 0.0001-0.011 ng/L)) were found by Utschakowski (1998) in the southern Laptev Sea near the Lena River Delta. Utschakowski (1998) found that PCBs were present at similar concentrations in the particulate phase at this location.

*In situ* sampling during the SHEBA study (Macdonald *et al.*, 2001) and in nearshore waters (Hoekstra *et al.*, 2002b) also found higher  $\Sigma$ PCB concentrations in nearshore waters of the Canadian Arctic Archipelago and southern Beaufort Sea ( $\Sigma$ PCB<sub>10</sub> means of 0.040-0.060 ng/L).  $\Sigma$ PCB<sub>10</sub> levels in the central basin of the White Sea (0.028±0.014 ng/L) were lower than in the Canadian Arctic Archipelago, using the same sampling equipment and analytical techniques, despite the proximity of urban areas of Dvina Bay (eastern White Sea) and Kandalaksha Bay (western arm).

Although the PCB measurements in the northern Chukchi Sea (Figure 4-30) may be confounded by con-

tamination they are internally consistent in that they show a remarkable transition from the Canada Basin to the Chukchi Plateau. East of the plateau (about 77°N, 160°W), the PCB profile consisted mainly of mono-, di- and trichlorinated CB congeners (Figure 4-30 a). Over the southern edge of the Chukchi Plateau (April), the profile appears far more strongly influenced by the technical mixture component (Figure 4-30 b). As SHEBA drifted back toward the interior ocean, the profiles in July (Figure 4-30 c) and September (Figure 4-30 d) revealed a strengthened lower chlorinated PCB congener profile. The PCB profile in waters west of the Chukchi Plateau bore a greater resemblance to technical PCB mixtures such as the Russian 'Sovol' (Schulz *et al.*, 1989; Ivanov and Sandell, 1992) (Figure 4-30 e) and Aroclor 1254. Given the stratification of the water column and ice cover, these changes were not due to ice melt or biological processes. It seems much more plausible that the PCB compositions along the track reflect the various

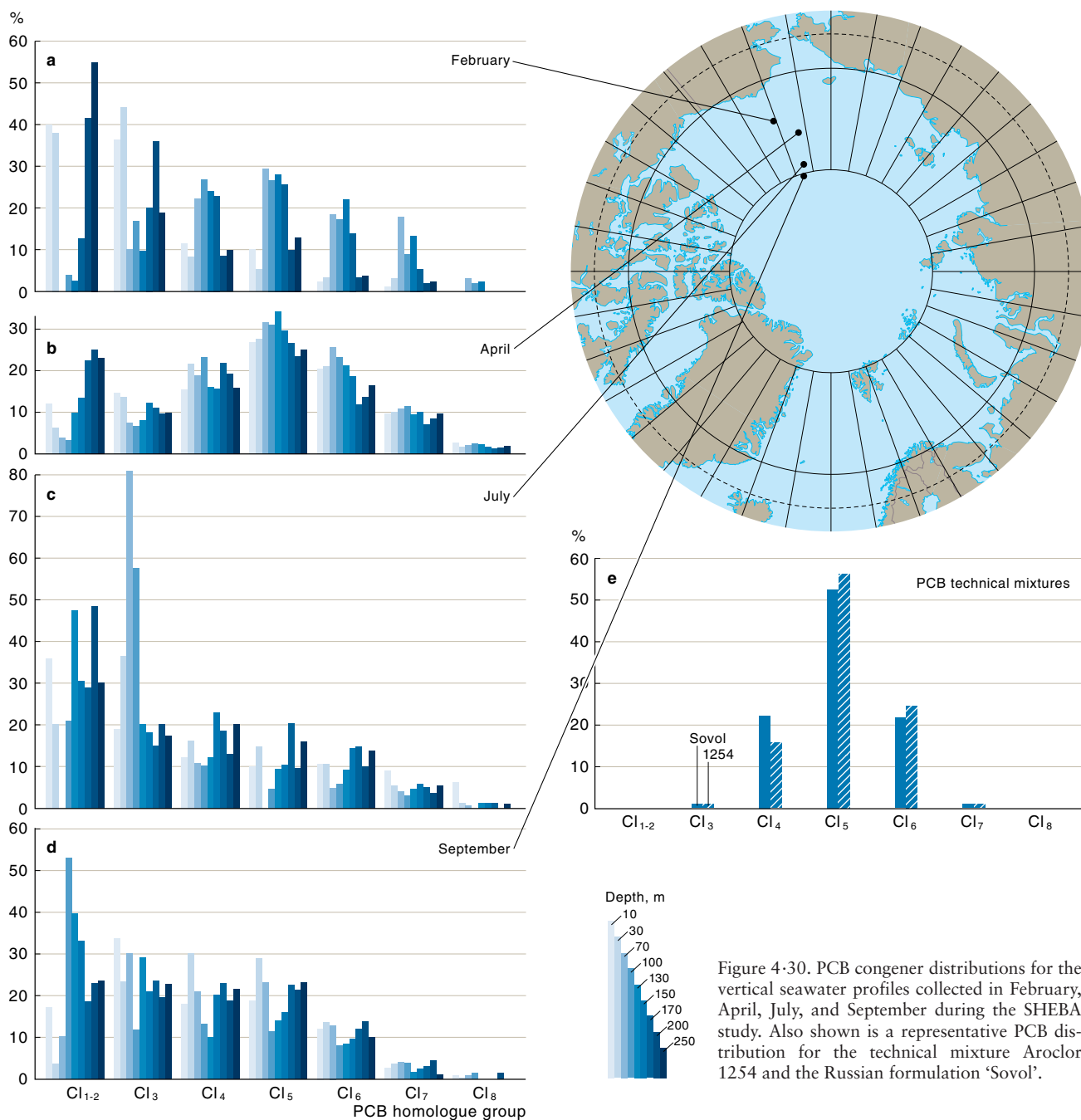


Figure 4-30. PCB congener distributions for the vertical seawater profiles collected in February, April, July, and September during the SHEBA study. Also shown is a representative PCB distribution for the technical mixture Aroclor 1254 and the Russian formulation 'Sovol'.

water masses through which SHEBA passed. The dominant contribution from technical mixtures seen in April (Figure 4-30 b), strongly suggests that this profile has been imported from the Pacific Ocean/Bering Sea. The profiles collected in February, July, and September, on the other hand, probably reflect a stronger influence of Canada Basin interior water that has accumulated an important component of its PCB inventory from the atmosphere. The inference to be drawn here is that PCBs have been delivered via the Bering Sea in water directly contaminated through, for example, runoff or spills, whereas the interior Arctic Ocean owes a significant portion of its PCB content to condensation and/or air-sea exchange which favors the lighter components as one progresses northward (Wania and Mackay, 1993; Muir *et al.*, 1996a).

#### 4.4.1.4. Toxaphene

Only a limited number of new measurements of toxaphene in seawater have been reported since the previous AMAP POPs assessment, and they are too few to fully assess spatial trends. Total toxaphene levels in sea-

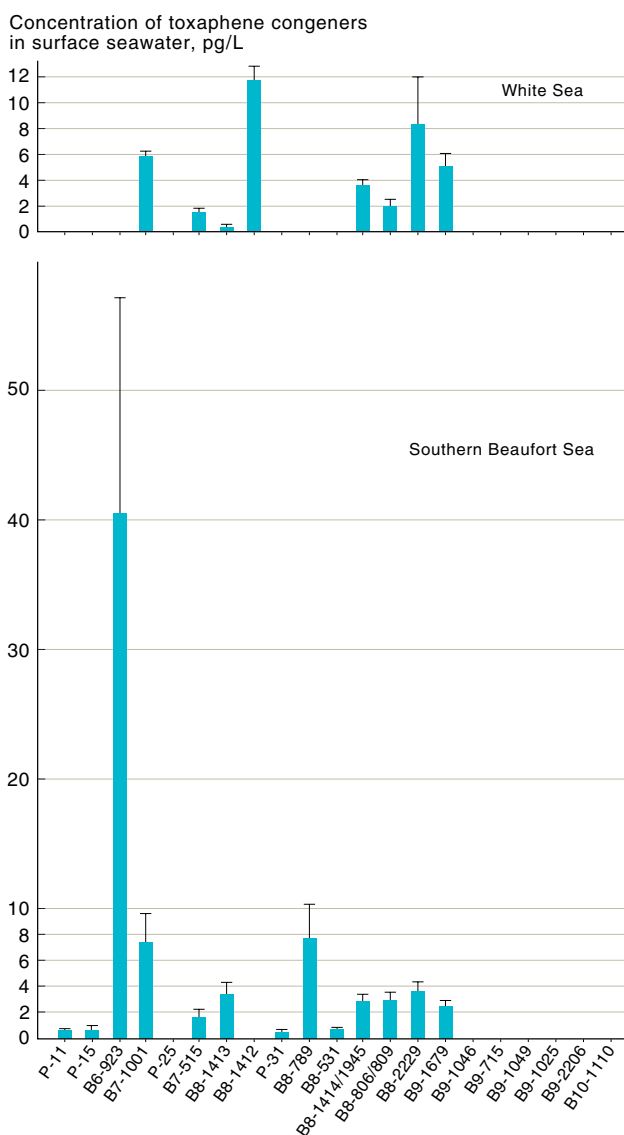


Figure 4-31. Comparison of mean ( $\pm$  SE) concentrations (ng/L) of individual toxaphene congeners in surface waters from the White Sea ( $n=5$ ) and the southern Beaufort Sea (Hoekstra *et al.*, 2002b).

water in the North American Arctic were higher in northern Baffin Bay than in the southern Beaufort Sea (Hoekstra *et al.*, 2002b). Lower toxaphene levels were found in seawater in the White Sea and in the northern Chukchi Sea than in the southern Beaufort Sea or northern Baffin Bay (Annex Table 8). The recent measurements have included chlorobornane congeners, thus allowing some insights into the degradation of toxaphene. Levels of individual chlorobornanes are in the low pg/L range (Figure 4-31). The congener profile of toxaphene in the Beaufort Sea was dominated by hexa- and heptachlorobornanes, especially B6-923 and B7-1001 (these have no Parlax numbers). These are terminal residues from dechlorination of many toxaphene congeners (Fingerling *et al.*, 1996) and are prominent in lake water in the Great Lakes (Muir *et al.*, 1999a; Muir *et al.*, 2001b). The White Sea had a different congener pattern with undetectable levels of B6-923 and more prominent octa- and nonachlorinated congeners. This pattern suggests a fresher, less degraded source of the technical product. Polychlorocamphene or toxaphene was widely used in Russian agriculture at least until the late 1980s (McConnell *et al.*, 1996). Toxaphene was also present in sediments of a harbor area in Kola Bay (near Murmansk) (Section 4.4.2) indicating other possible uses such as insect control on ships (Savinova *et al.*, 2000a).

#### 4.4.1.5. Other 'legacy' OC pesticides

$\Sigma$ DDT and chlordane-related compounds, as well as HCB were determined in several studies (Annex Table 8) and were present at much lower concentrations than toxaphene, PCBs or HCH isomers. Similar to the pattern for PCBs, DDT residues were higher (by 5 to 20 times) in seawater from the Canadian Arctic Archipelago and nearshore waters of the Beaufort Sea than over the Chukchi Plateau or in the central Arctic Ocean.  $\Sigma$ CHL had a similar pattern although differences were not as pronounced. This suggests that gas exchange and melting are important sources for  $\Sigma$ DDTs and  $\Sigma$ CHLs. Hargrave *et al.* (1997) found generally higher levels of *cis*- and *trans*-chlordane and toxaphene under ice-covered conditions in the central Canadian Arctic Archipelago compared to the open-water season. These authors concluded that net deposition by seawater-air exchange of toxaphene and chlordanes during the open-water period was equivalent to 50 to more than 100% of the surface layer inventory. Thus, lower concentrations of the more hydrophobic OCs, such as PCBs and DDT under the ice pack, are probably due to slow removal by sedimentation and lack of resupply from the atmosphere.

$\Sigma$ DDT levels in White Sea seawater were lower than in the southern Beaufort Sea or in the northern Barents Sea (Annex Table 8). Despite possible ongoing use in urban areas in the White Sea and Kola regions (Savinov *et al.*, 2003; AMAP, 2003), there was relatively little spatial variation of  $\Sigma$ DDT among ten sampling sites (0.002-0.005 ng/L) ranging from Dvina Bay (eastern White Sea) to the western end of Kandalaksha Bay (western arm).

Utschakowski (1998) found  $\Sigma$ DDT concentrations ranging from 0.0001 to 0.00178 ng/L in the dissolved phase and from 0.00036 to 0.00087 ng/L on particles in seawater from the southern Laptev Sea collected in 1993

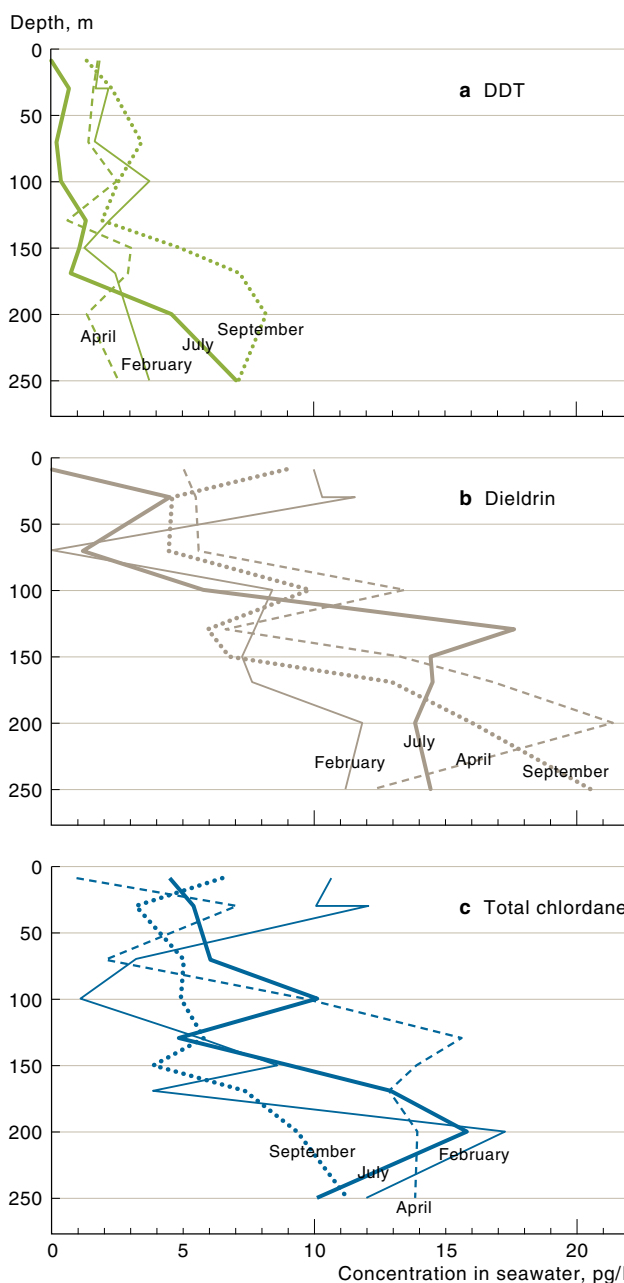
to 1994. The DDT residues in this region were predominantly in the form of *p,p'*-DDE.

Macdonald *et al.* (2001) reported on the vertical distribution of DDT, dieldrin and chlordane over the Canada Basin and Chukchi Plateau (Figure 4.32 a-c) as part of the SHEBA study. The vertical distributions of these OC pesticides were very different from those observed for the HCHs where concentrations are found to decrease from their highest values near the surface to very low values below depths of 200-300 m. This may have been due to changes in water mass. Taking DDT as perhaps the best example, however, it seems likely that there is also a seasonal signal related to the biological cycle. In April and February of 1998, the DDT profiles were somewhat noisy, but reasonably uniform at about 2 pg/L through the water column down to 250 m (Figure 4.32 a). By July, however, this uniformity was disrupted, with much lower ΣDDT concentrations evident in the top 100-150 m of the water column and higher concentrations at about 200-250 m. In September, the surface concentrations of DDT were again uniform at about 2 pg/L, but higher concentrations are evident at or below approximately 150 m. Although water mass changes may contribute partly to the observed behavior, there appears also to be a transfer of DDT from surface to deeper waters in spring when biological production produces a vertical flux of organically rich particles.

#### 4.4.1.6. Current-use chemicals

Endosulfan and lindane ( $\gamma$ -HCH) are the only pesticides in current use in circumpolar countries that are reported to date in Arctic Ocean water. Levels of lindane are generally highest in the Canadian Arctic Archipelago and southern Beaufort Sea, and lowest in the central Arctic Ocean (Strachan *et al.*, 2000), over the Chukchi Plateau and in the White Sea. Lakaschus *et al.* (2002) found lindane concentrations in the North Sea (0.33 ng/L) within the range of concentrations observed in the Greenland Sea (0.072-0.50 ng/L). White Sea waters were characterized by higher  $\gamma$ -HCH than in the Barents Sea, reflecting use in urban areas within the watershed. Within the White Sea, higher lindane concentrations were found in Kandalaksha Bay (0.19-0.41 ng/L) compared with the Central Basin (0.16-0.37 ng/L) (Muir *et al.*, 2002a). However, in general,  $\gamma$ -HCH levels in the White Sea were lower than those in the southern Beaufort Sea.  $\alpha/\gamma$ -HCH ratios averaged  $1.8 \pm 0.05$  in the White Sea, compared with  $2.2 \pm 0.52$  in the Greenland Sea,  $3.4 \pm 0.77$  in the northern Barents Sea/central Arctic Ocean, and  $2.3 \pm 0.77$  in the Beaufort Sea (Hoekstra *et al.*, 2002b; Macdonald *et al.*, 2001).

Endosulfan is a widespread contaminant in Arctic seawater although present at much lower levels than lindane. Endosulfan (sum of  $\alpha$ - and  $\beta$ -isomers) concentrations ranged from  $<0.0005$  to 0.003 ng/L with no clear spatial trends. Hargrave *et al.* (1997) found endosulfan concentrations were higher during open water periods in the central Canadian Arctic Archipelago reflecting inputs from gas exchange and freshwater. During a twelve month study in 1993, endosulfan concentrations increased three-fold during the late summer and autumn months, paralleling increasing air concentrations. This was possibly related to summer use of endosulfan in mid-latitude North America.



Figures 4.32. Vertical profiles of 'legacy' OC pesticides (DDT, dieldrin, and chlordane) in seawater sampled during four intervals (February, April, July, and September) over the Canada Basin and Chukchi Plateau (see Figure 4.30) in 1998 (Macdonald *et al.*, 2001).

Chlorobenzenes are prominent contaminants in Arctic seawaters; however, the number of measurements of chlorobenzenes is limited. Di- to hexachlorobenzenes have been reported (Strachan, 2002). HCB concentrations appear to be relatively uniform compared to other compounds. Concentrations ranged from 0.006 ng/L in the southern Beaufort Sea to means of 0.005 ng/L in the White Sea. There are no data for chlorobenzenes in seawater from the European Arctic except for the White Sea (Annex Table 8).

#### 4.4.1.7. Modeling latitudinal trends of $\alpha$ -HCH in ocean waters

It is of particular interest, from an Arctic perspective, to examine the global fate of  $\alpha$ -HCH. By the 1990s, the concentration levels in the Canadian Arctic Ocean were



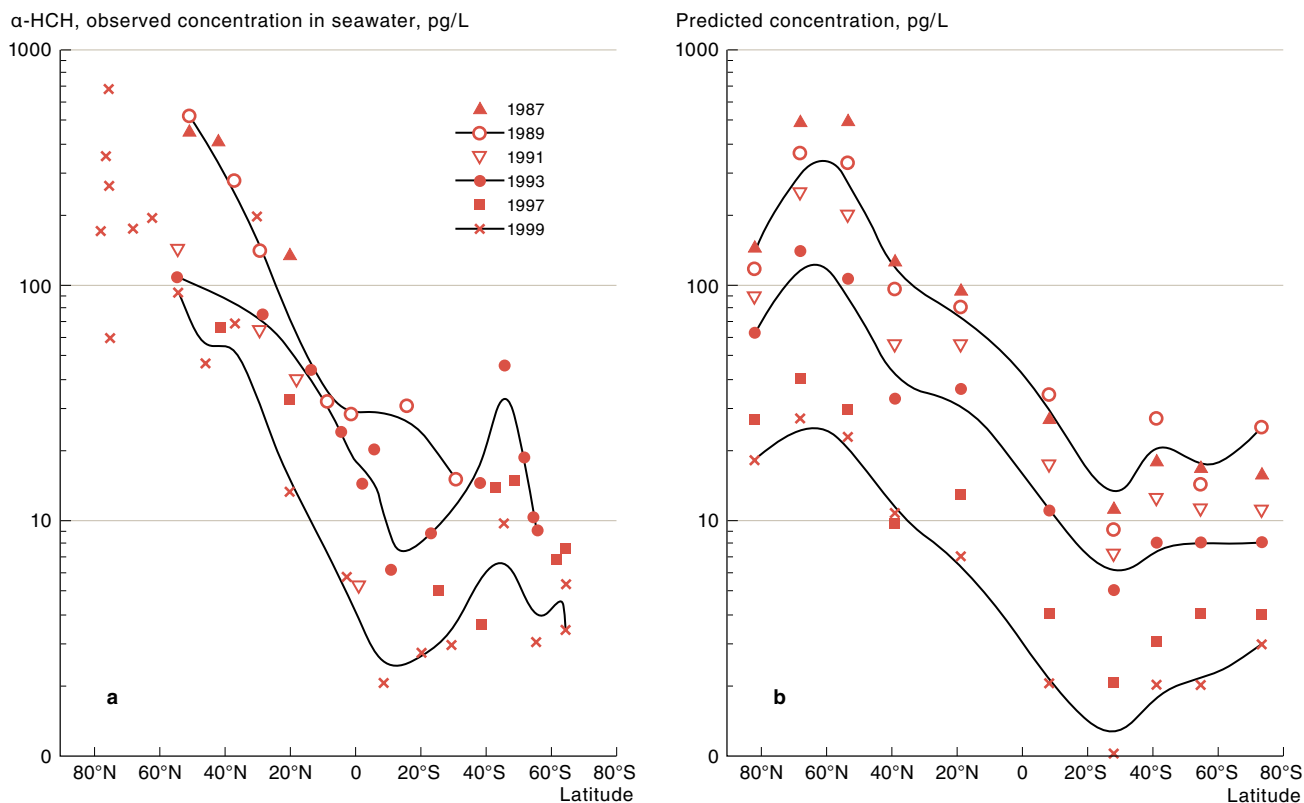


Figure 4-33. Latitudinal trends in a) observed and b) predicted seawater concentrations of  $\alpha$ -HCH in the Atlantic Ocean (Lakaschus *et al.*, 2002). Concentrations were predicted with the 'Globo-POP' model of Wania and Mackay (2000).

higher than anywhere else in the global marine environment, notably an order of magnitude higher than in the Indian Ocean and the western Pacific, which are much closer to the major source regions (Iwata *et al.*, 1993; Wania and Mackay, 1996; de March *et al.*, 1998).  $\alpha$ -HCH, the major component of technical HCH, was used in massive quantities in Asia during the 1970s. There has been a steady decline in use since then, and it is now virtually out of use worldwide (Li *et al.*, 1996; 1998a; Li 1999a; 1999b). Compared to most other OC pesticides, it is relatively water-soluble, relatively volatile, and less persistent. Calculations using the 'Globo-POP' global distribution model (Wania and Mackay, 2000) suggested that this inverted latitudinal concentration gradient developed during the two decades of declining emissions and was the result of  $\alpha$ -HCH being trapped and preserved in the cold Arctic Ocean, whereas the levels in source regions declined as a result of degradation and volatilization. This is illustrated in Figure 4-33. Thus, the Arctic Ocean constitutes the last global refuge of  $\alpha$ -HCH (Macdonald *et al.*, 2000). Interestingly, this highlights the fact that inverted concentration gradients can be established even if only a small percentage of the globally emitted amount is transferred to Arctic latitudes. In fact, the model predicted that the bulk of the  $\alpha$ -HCH never left the agricultural systems in which it had been applied. Relatively small amounts can result in high concentrations in the Arctic because of the relatively small size of the Arctic as a whole and of the environmental phases with high capacity for adsorbing hydrophobic organic chemicals (organic soils, vegetation, organic sediments) within the Arctic. By binding substances such as HCH, these phases reduce the amount present in the atmosphere and in aquatic systems. The

model also suggested that ocean currents are as important in the northbound transport of  $\alpha$ -HCH as the atmosphere. Most of the  $\alpha$ -HCH in the Arctic originated in the northern temperate zone, which also was a net exporter of  $\alpha$ -HCH to low latitudes. While  $\alpha$ -HCH accumulated in the north, it was rapidly degraded under tropical conditions. Comparisons showed that calculated and measured  $\alpha$ -HCH concentrations in the atmosphere and seawater generally agree within an order of magnitude (Wania *et al.*, 1999a; Lakaschus *et al.*, 2002) (Figure 4-33). Deviations are due to the zonal averaging characteristics of the model and uncertainties associated with the environmental degradation rates of  $\alpha$ -HCH.

#### 4.4.2. Marine sediments

The previous AMAP assessment report presented a large dataset on  $\Sigma$ PCBs,  $\Sigma$ DDTs, HCB, and  $\Sigma$ HCHs in marine sediments based on results from the early 1990s (de March *et al.*, 1998). Those results showed relatively uniform low levels of OCs in marine surface grab samples (all concentrations <10 ng/g dw). Thus, a general understanding of the distribution of the different contaminants in the Arctic was established during AMAP Phase I. No widescale regional geographic trends were apparent. However, there were distinct differences between nearshore and offshore sampling sites especially in Norway, which pointed to the need for further understanding of 'hot spots' and their importance as a source for contamination of adjacent areas. Previous studies in the Canadian Arctic have identified harbors in communities with military radar bases as having higher  $\Sigma$ PCB concentrations in sediment (Bright *et al.*, 1995b).

### Svalbard

POPs were determined in surficial sediments collected in the Svalbard area of the Barents Sea in 1997 (Akva-plan-niva, 1998). Sampling sites included Kongsfjorden (western Svalbard), Questrenna (north of Svalbard), Lomfjorden and Hinlopen Strait (northern Svalbard), Erik Eriksen Strait (eastern Svalbard), and Strofjorden (southern Svalbard) (Figure 4-34). The samples were from relatively deep stations offshore (56-363 m depth, and 2224 m at Questrenna) and did not include harbor areas.  $\Sigma$ PCB levels were very low, ranging from 0.025 to 0.61 ng/g dw (sum of seven congeners multiplied by two to estimate total PCBs). Levels were similar to those reported around Franz Josef Land (Killie *et al.*, 1997; de March *et al.*, 1998). The deep sediment sample (Questrenna) had higher concentrations than other sites and a distinctly different PCB pattern, with higher proportions of hepta- and octachlorinated biphenyls. No influence on samples was evident from possible local PCB sources in harbors of towns on Svalbard.

$\Sigma$ DDTs were present at similar levels as  $\Sigma$ PCBs with highest levels at the deep site (0.62 ng/g dw). DDE predominated at the deep site, while higher proportions of DDT and DDD isomers were found in most nearshore samples, suggesting some past use of DDT on Svalbard.

### Northern Norway/Kola Peninsula and the White Sea

A study was conducted of OCs in sediments from six harbors in northern Norway and northwest Russia, an area that has been determined to be a key monitoring area by AMAP. The results allowed comparison of concentrations and the composition of congeners of the different OCs in the two countries (Dahle *et al.*, 2000).

When comparing the results for the six different harbors, some differences in study set-up should be noted. The most polluted areas in the Norwegian harbors were generally known, and the stations were positioned in order to confirm the local 'hot spots' and to check the degree to which contaminants had been spread to other parts of the harbor areas. The study in the Russian harbors was a screening study based on a limited number of stations.

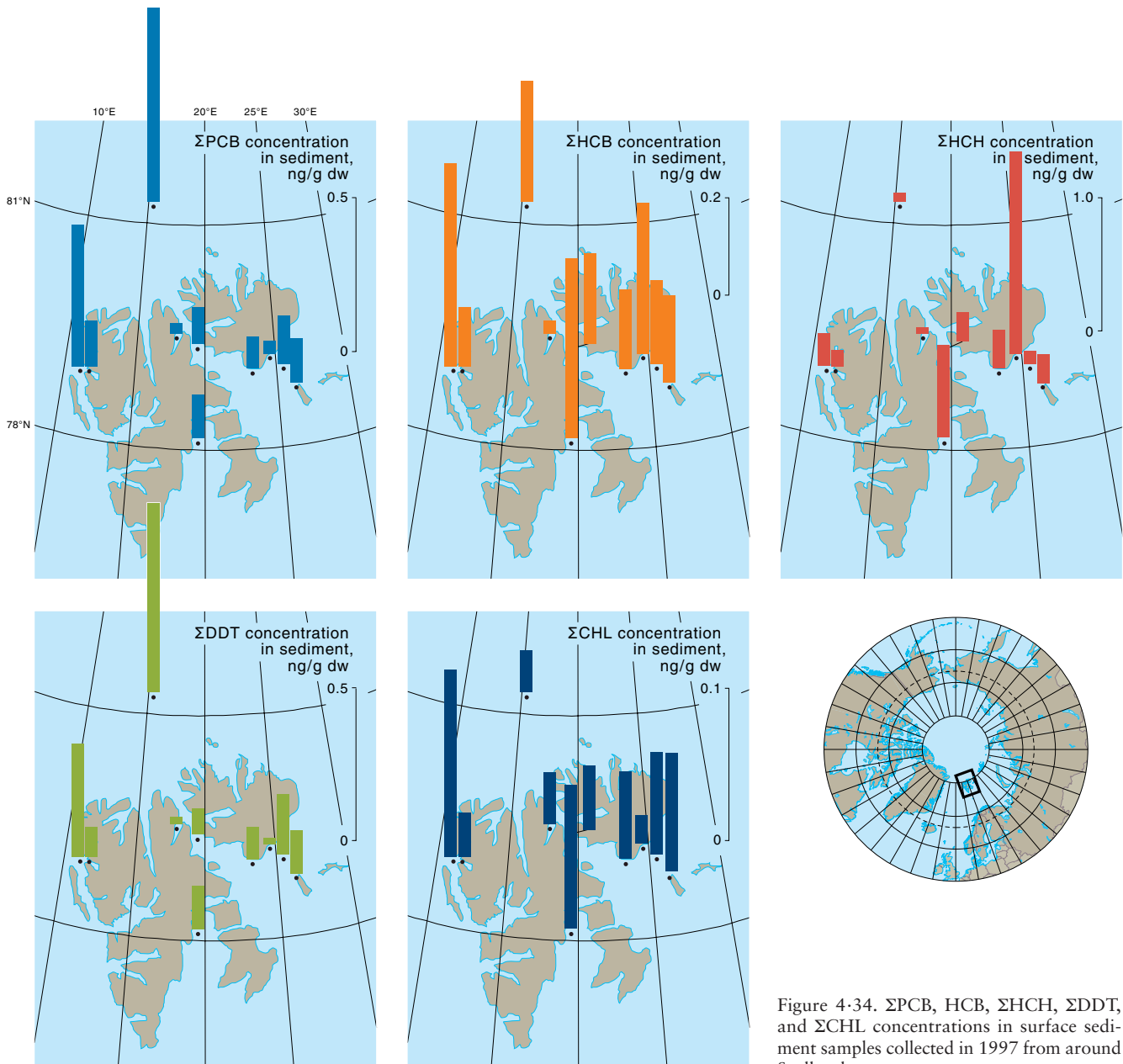


Figure 4-34.  $\Sigma$ PCB, HCB,  $\Sigma$ HCH,  $\Sigma$ DDT, and  $\Sigma$ CHL concentrations in surface sediment samples collected in 1997 from around Svalbard.

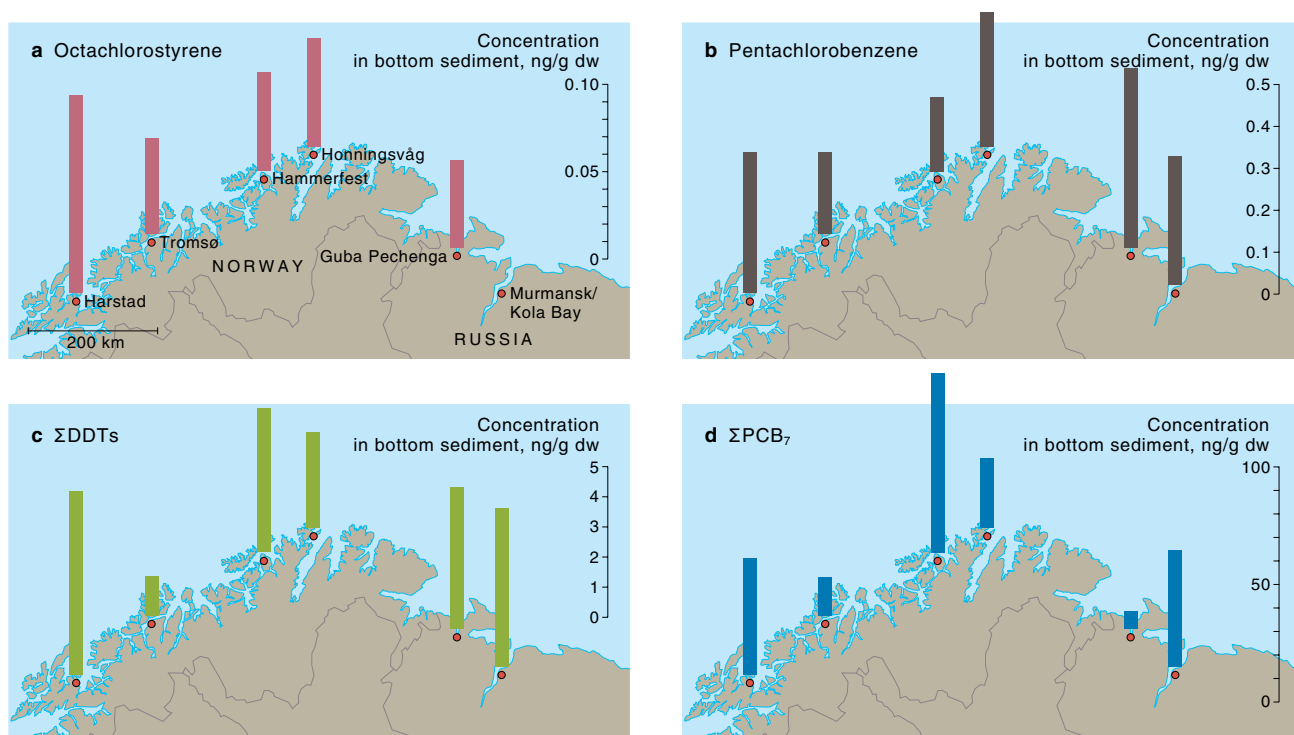


Figure 4-35. Geometric mean concentrations of octachlorostyrene, pentachlorobenzene,  $\Sigma$ DDT, and  $\Sigma$ PCB<sub>7</sub> in bottom sediments of some harbors of northern Norway and the Kola Peninsula.  $\Sigma$ DDT = *p,p'*-DDE + *p,p'*-DDD + *p,p'*-DDT;  $\Sigma$ PCB<sub>7</sub> = CB28 + CB52 + CB101 + CB118 + CB138 + CB153 + CB180.

The highest concentration of  $\Sigma$ PCB<sub>7</sub> was found in the harbors of Harstad, Hammerfest, and the Murmansk area of Kola Bay (Figure 4-35). Detailed results are presented in Annex Table 9. All these harbors had PCB concentrations, which according to guidelines from Norwegian State Pollution Control Authority (SFT) (Molvær *et al.*, 1997) are classified as 'strongly polluted'. In all the harbors, locations were found having  $\Sigma$ DDT concentrations classified as 'strongly polluted'. The concentrations of pentachlorobenzene and OCS were also elevated in all the harbors compared to previous reports for background areas at offshore locations (de March *et al.*, 1998).

There were also statistically significant differences ( $p < 0.05$ ) between the distributions of HCB and  $\Sigma$ HCHs in the six harbors. For OCS, Harstad harbor was significantly different from the four others (no data from Kola Bay), and for pentachlorobenzene, both Harstad and Pechenga were significantly different compared to the other four harbors. For  $\Sigma$ DDTs, Tromsø had a significantly different pattern, and for  $\Sigma$ PCB<sub>7</sub>, the most polluted harbors (Harstad, Hammerfest, Honningsvåg, and Kola Bay) comprised a homogeneous group different from the two others (Figure 4-35).

In sediments from all these harbors and also from Honningsvåg and Kola Bay areas, the higher chlorinated CBs predominated in samples where the highest  $\Sigma$ PCB<sub>7</sub> concentrations were found. Only sediments from Guba Pechenga with maximum  $\Sigma$ PCB<sub>7</sub> levels had high percentages of lower chlorinated CBs.

Savinova *et al.* (2000a) analyzed samples collected in 1997 from Kola Bay and Guba Zapadnaya Litsa on the western Kola Peninsula. In general, levels of OCs were similar to those reported by Dahle *et al.* (2000). However, high concentrations of toxaphene were found at locations in Kola Bay (up to 681 ng/g dw) and intermedi-

ate concentrations in Guba Zapadnaya Litsa (19-95 ng/g dw). This was the first report of toxaphene in marine sediments in the European Arctic and it suggests use and/or spills in the harbor area of Polamyy north of Murmansk.

Savinov *et al.* (2003) reported OCs in sediments collected in 1997 in Guba Pechenga and adjacent marine coastal areas: Varangerfjord, Guba Malaya Volokovaya, and Guba Bol'shaya Volokovaya on the western Kola Peninsula. Slightly elevated  $\Sigma$ DDT concentrations (37 ng/g dw) were found in Liinakhamari harbor in Guba Pechenga and a high DDT:DDE ratio (23.4) indicated a possible local DDT source in this area. However, the overall average  $\Sigma$ DDT levels in Guba Pechenga sediments were comparable with those in harbor sediments of northern Norway (Dahle *et al.*, 2000) and outer Kola Bay (Savinova *et al.*, 2000a). PCB concentrations in Guba Pechenga were slightly elevated in Liinakhamari harbor as well. However, overall values were significantly lower in comparison with those found in harbors of the northern Norway and Kola Bay (Dahle *et al.*, 2000; Savinova *et al.*, 2000a).

The studies of harbors in northern Norway and northwestern Russia indicate that harbors may be 'hot spots' for contaminants in the Arctic. Steps need to be taken to understand the extent of the pollution in the harbors, the potential for spreading to adjacent waters including the Barents Sea, and the impact on regional fauna.

Analysis of a sediment core from the profundal area of Kandalashka Bay in the White Sea revealed relatively low levels of persistent OCs (Annex Table 9). Maximum concentrations of most OCs were found in deeper layers indicating greater inputs in the recent past (Muir *et al.*, 2002a).  $\Sigma$ PCB concentrations were higher than back-

ground locations in the southwest Barents Sea (Savinov *et al.*, 2003) but lower than in Kola Bay and Guba Zapadnaya Litsa.

#### Kara and Laptev Seas

Sericano *et al.* (2001) reported POPs in surficial sediments from the southern Kara Sea adjacent to the Ob and Yenisey. The samples were collected in 1993. However, results were not available for the previous AMAP POPs assessment.  $\Sigma$ DDT and  $\Sigma$ PCB concentrations in sediments ranged from <0.1 to 1.2 ng/g dw and <0.1 to 1.5 ng/g dw, respectively. Chlordane compounds were not detected (<0.1 ng/g dw). Despite the low levels, a distinct distribution pattern was observed with higher concentrations in the lower part of the Yenisey below the salt wedge. The concentrations of  $\Sigma$ DDTs and  $\Sigma$ PCBs in marine sediments were within the range reported previously for the Ob and Yenisey Gulf (0.03-0.8 ng/g dw and 0.01-2.2 ng/g dw, respectively) (Vlasov and Melnikov, 1995, *in de March et al.*, 1998).

Relatively high proportions of *p,p'*-DDT and *o,p'*-DDT in sediments suggested local or fresh sources of DDT at the time of sample collection (1993). The PCB homologue profile in the sediments was dominated by Pe- and HxCBs, suggesting local or nearby sources of PCBs to the Kara Sea.

Utschakowski (1998) determined PCBs in surface sediment samples collected in 1993 from the southern Laptev Sea offshore of the Lena River Delta.  $\Sigma$ PCB concentrations averaged 0.53 ng/g dw (range 0.14-1.99 ng/g dw) and were correlated with organic carbon content of the sediments (Annex Table 9).

#### Canadian Arctic

In 1997 and 1998, sediment cores and grab samples were collected from various regions of the Canadian Arctic Archipelago including northern Baffin Bay (Annex Table 9). A number of these grab samples have been analyzed for OC pesticides, PCBs, and current-use contaminants such as short-chain ( $C_{10}$ - $C_{13}$ ) chlorinated par-

affins and PBDEs (Stern and Lockhart, 2001). Concentrations of the major OC groups were lower than reported previously for Hudson Bay surface sediments taken from a core (Lockhart, 1997). A generally decreasing trend from south to north and from east to west is apparent from the results, although this could be influenced by variation in sedimentation rates (Figure 4-36).

$\Sigma$ PCB concentrations in the sediments from the archipelago waters ranged from 0.3 to 2.9 ng/g dw. These are at the low end of the range for marine sediments found in the previous AMAP POPs assessment (de March *et al.*, 1998). DDT and total chlordane-related compounds were present at extremely low levels (0.04-0.44 ng/g dw). SCCP concentrations in the surface sediments ranged from 4.8 to 77.4 ng/g dw and were among the predominant OC compounds in these sediments. As with the OC pesticides and PCBs, a decreasing trend in SCCP concentrations was observed from southern (Barrow Strait) to northern (Peary Channel) and western Arctic (McClure Strait/Viscount Melville). Sediment concentrations were lower than in the eastern Arctic (Strathcona Sound/Lancaster Sound) sediments (Annex Table 9; Figure 4-36).

#### 4.4.3. Marine phytoplankton and invertebrates

Marine invertebrates provide a link between phytoplankton and fish/seabirds/mammals in Arctic marine food webs. They are important in the transfer of carbon and nutrients as well as POPs to upper trophic-level organisms. An understanding of the trends and dynamics of POPs in marine invertebrates is a key to understanding the trends of POPs in Arctic marine ecosystems.

A number of studies have been recently carried out to examine OC concentrations in marine invertebrates, an area identified as a knowledge gap in the first AMAP assessment report. These studies covered a wide geographical area and have provided information on spatial trends. There was also a single study on levels of OCs in plankton, predominantly phytoplankton. Many of these stud-

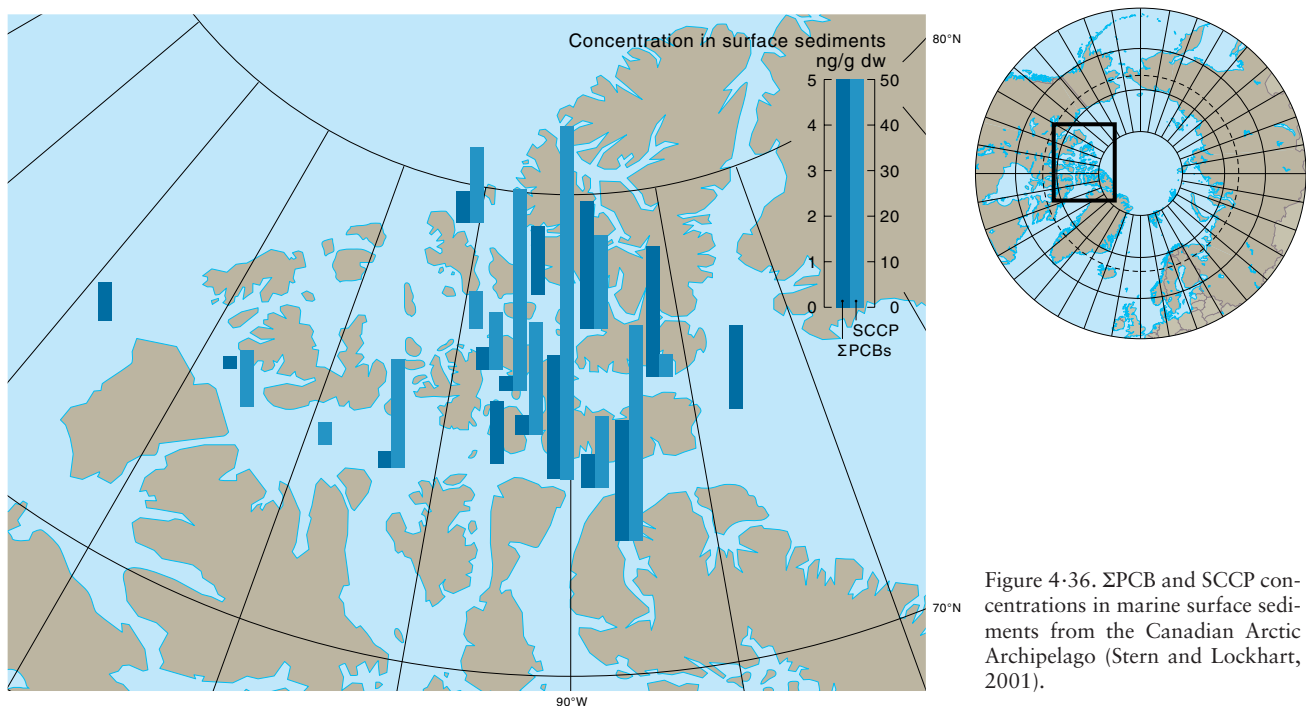


Figure 4-36.  $\Sigma$ PCB and SCCP concentrations in marine surface sediments from the Canadian Arctic Archipelago (Stern and Lockhart, 2001).



ies were designed to examine factors that influence OC concentrations in these lower trophic-level organisms. A large number of samples from single species, particularly in zooplankton, were analyzed for OCs. In a number of cases, these data were used in food web studies, discussed in Section 4.4.9.

In general, OC concentrations in marine phytoplankton and invertebrates are low, consistent with their generally lower trophic levels and smaller body sizes. Invertebrates that are longer-lived, larger-bodied or that scavenge have higher POP levels and are generally benthic.

#### Phytoplankton

Marine samples were collected to the west of Bjørnøya (74°22'N, 18°55'E) as part of a marine food web study (Evenset *et al.*, 2002). This work was part of the larger study examining elevated levels in Ellasjøen (see Section 4.3.5). Twenty-three vertical hauls (net diameter 25 cm, mesh size 5 µm) were taken from 20 m depth to the surface. These samples were predominantly phytoplankton. Low concentrations of PCBs and pesticides (mostly below detection limits for pesticides) were found in the marine phytoplankton sample taken west of Bjørnøya (Annex Table 10).

#### Zooplankton

A number of studies have examined OCs in pelagic zooplankton. The most common OC compounds in pelagic zooplankton are the more water-soluble compounds, such as the HCH isomers and lower chlorinated PCB congeners. This reflects the smaller size, lower trophic level, and lack of biotransformation capacity generally found in zooplankton as compared to fish, mammals, and birds.

#### Spatial trends in calanoid copepods

OC concentrations were determined in calanoid copepod samples from a number of studies in the Beaufort Sea and Hudson Bay (Hoekstra *et al.*, 2002b), northern Baffin Bay (Fisk *et al.*, 2001a), and in the ocean around Svalbard (Borgå *et al.*, 2001) (see Figure 4-37 for locations). Calanoid copepods are dominant components, in terms of both number and biomass, in high-latitude marine zooplankton communities, and play an important role in polar food webs, as their high lipid reservoirs and biomass provide organisms at higher trophic levels with a high-energy diet (Springer *et al.*, 1996). Concentrations (lipid-normalized) of  $\Sigma\text{PCB}_{10}$  (sum of ten congeners) in the Beaufort Sea samples were slightly lower than those in northern Baffin Bay and Hudson Bay, which were similar (Figure 4-37). OC concentrations in the North American copepods were greater than those measured in oceans to the east (Greenland Sea) and north of Svalbard but lower than those observed to the south of Svalbard (Barents Sea) (Figure 4-37). Similar trends were seen with other OC groups including toxaphene,  $\Sigma\text{DDTs}$ , and  $\Sigma\text{CHLs}$ . Levels of OCs were also higher in the northern Baffin Bay predatory amphipod, *Themisto libellula* (Fisk *et al.*, 2002a) compared with the same species collected east and north of Svalbard (Borgå *et al.*, 2001). These spatial trends do not agree with that observed in marine mammals (Muir *et al.*, 2000b) and some seabird species (Fisk *et al.*, 2001b). Concentrations of most OCs, excluding HCH, in upper

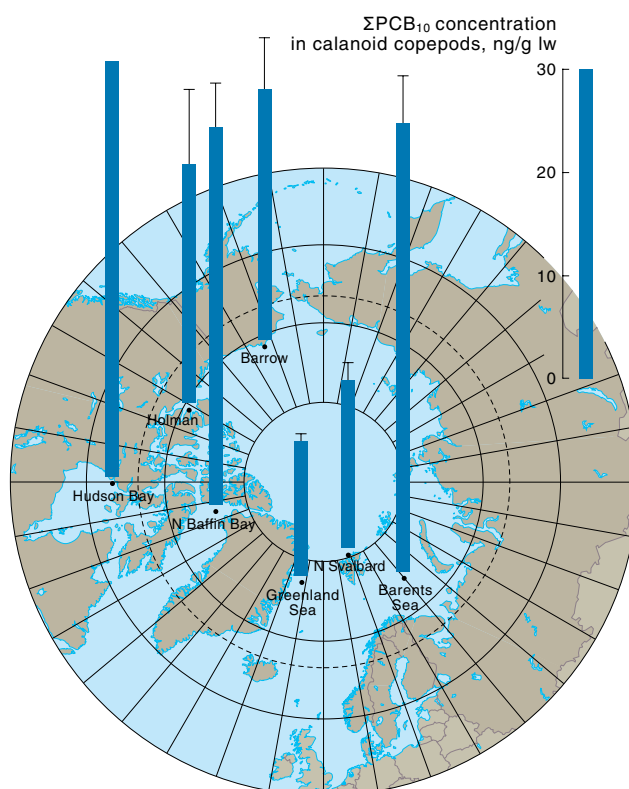


Figure 4-37. Concentrations of  $\Sigma\text{PCB}_{10}$  in calanoid copepods from Barrow, Alaska; Holman, NWT and Hudson Bay (Hoekstra *et al.*, 2002b); northern Baffin Bay (Fisk *et al.*, 2001a); and regions around Svalbard ( $\Sigma\text{PCB}_9$ , Borgå *et al.*, 2001).

trophic-level organisms are much higher in the European compared to the North American Arctic. Thus, concentrations found at the base of these two food webs cannot explain the high POP levels seen in upper trophic-level organisms in the European Arctic compared to northern Baffin Bay.

The low OC concentrations in the copepods from east (Greenland Sea) and north of Svalbard may be due to timing of the sample collection, seasonal changes in water concentrations of OCs, and/or the source of water in these locations. As a small organism, the copepod could reflect subtle changes of OCs in water that a large organism would not. The source of water for these copepods would come predominantly from under the ice cap to the north, which would explain, in part, the lower concentration of OCs compared to the Barents Sea copepods. The collection of the copepods from the east and north of Svalbard also occurred in September and October after the open-water period. Hargrave *et al.* (2000) found that OC concentrations were highest in Arctic zooplankton during periods of ice cover.

Concentrations of HCB and HCH isomers in the Beaufort Sea copepods were relatively higher than the more easterly sampling locations. The abundance of HCB and  $\Sigma\text{HCHs}$  in Alaskan and western Canadian zooplankton reflects the long-range atmospheric transport of the chemicals and geographic proximity to areas of recent application in Asia (Li, 1999a; 1999b; Bailey *et al.*, 2000). The concentrations of OC compounds in zooplankton samples, including *Calanus hyperboreus*, collected in the late 1980s in the Arctic Ocean were similar to the Beaufort Sea values (Bidleman *et al.*, 1989). This suggests that OC concentrations in the western

Arctic zooplankton have remained constant during the 1990s in the Beaufort Sea region.

The ranking of OC group concentrations in the Alaskan and Canadian copepods from highest to lowest were: toxaphene  $\geq$   $\Sigma$ PCBs  $>$   $\Sigma$ HCHs  $>$   $\Sigma$ DDTs  $>$   $\Sigma$ CHLs  $>$   $\Sigma$ CBz (Fisk *et al.*, 2001a; Hoekstra *et al.*, 2002b). There are no toxaphene data for the Barents Sea copepods, but the general ranking of OCs is similar, except for lower relative levels of  $\Sigma$ HCHs (Borgå *et al.*, 2001). Recent work on mixed zooplankton from the west of Bjørnøya found toxaphene to be the predominant OC although at concentrations lower than PCBs (Annex Table 10) (Evenset *et al.*, 2002). The higher abundance of toxaphene congeners relative to other OCs in North American zooplankton is consistent with OC concentrations in zooplankton collected in the Arctic Ocean offshore from Axel Heiberg Island in the Canadian Arctic (Bidleman *et al.*, 1989). The  $\alpha$ - and  $\gamma$ -HCH isomers and lower chlorinated PCB congeners were the most common OCs found in the Arctic calanoid copepods.

#### Levels in other zooplankton species

OC data for a number of other pelagic zooplankton have been recently generated in northern Baffin Bay (Fisk *et al.*, 2001c; 2002a; Moisey *et al.*, 2001) and the Barents Sea region (Borgå *et al.*, 2001; 2002b). A majority of these samples were analyzed as part of food web studies, discussed in Section 4.4.9. Seven species of zooplankton were collected in northern Baffin Bay and analyzed for OCs, including: *C. hyperboreus* (herbivorous copepod, discussed above); *Euchaeta glacialis* (omnivorous copepod); *Metridia longa* (omnivorous copepod); *Mysis oculata* (detritus-feeding and predatory mysid); *Themisto libellula* (predatory amphipod); *Sagitta* sp. (predatory arrowworm); and *Pandalus* sp. (predatory shrimp). Lipid content varied between species, ranging from 2.1 to 7.0%. OC concentrations (lipid corrected) varied between species and appeared to be related, in part, to a combination of trophic position (as determined by  $\delta^{15}\text{N}$ ) and body size (Figure 4-38). The relative ranking of OC groups also varied with the species.

In general, for these zooplankton species,  $\Sigma$ PCB concentrations were highest and  $\Sigma$ CBz concentrations were lowest. Borgå *et al.* (2001) measured OCs in the herbivorous pelagic copepod *Calanus* sp. (copepods), the herbivorous euphausiid *Thysanoessa* sp., and the predacious pelagic crustacean *Parathemisto libellula* (amphipod). OC concentrations were similar to those observed in the northern Baffin Bay zooplankton.

Concentrations of PCBs (both as sum of 11 congeners and Aroclor 1254) were determined in mixed zooplankton (*Calanus* sp., *T. libellula*, ctenophores, gastropods) and shrimp (*Pandalus borealis*) collected in 1991 from various sites within the Barents Sea region (Joiris *et al.*, 1997).  $\Sigma$ PCB levels were not related to lipid levels of the zooplankton. Levels of  $\Sigma$ PCB were higher in the shrimp samples compared to the mixed zooplankton using lipid corrected values, but lower when wet weight concentrations were examined. Concentrations for both sample types did not vary between sites. Concentrations were found to be lower than levels measured in similar species from the Atlantic Ocean but higher than levels observed in the Canadian Arctic.

#### Factors influencing OC levels in zooplankton

The relative abundance of hydrophobic chemicals such as  $\Sigma$ DDTs and  $\Sigma$ CHLs in copepod samples of Alaska and Canada was greater than that observed in water (Fisk *et al.*, 2001a; Hoekstra *et al.*, 2002b), and reflects their greater potential for bioaccumulation from the abiotic environment into marine zooplankton near the base of the food chain than less hydrophobic OCs. The relationships observed between the bioconcentration factor (BCF) and  $K_{ow}$  for the Beaufort Sea and northern Baffin Bay calanoid copepods were linear and near, but slightly higher than, 1:1 for OCs with a log  $K_{ow}$  between 3 and 6, but curvilinear when hydrophobic OCs (log  $K_{ow}$   $>$  6) were included (Fisk *et al.*, 2001a; Hoekstra *et al.*, 2002b). These results suggest that for hydrophobic OCs (log  $K_{ow}$  3.8-6.0) dietary accumulation may play a small role in elevating OC levels above equilibrium in zooplankton.

Studies of OC levels in zooplankton collected to the west of Bjørnøya found that zooplankton size was an important variable in OC concentrations (Evenset *et al.*, 2002). The concentrations in zooplankton seemed to increase from small zooplankton ( $<$ 1 mm, mainly copepods) to larger zooplankton ( $>$ 1 mm, mainly comb jell-

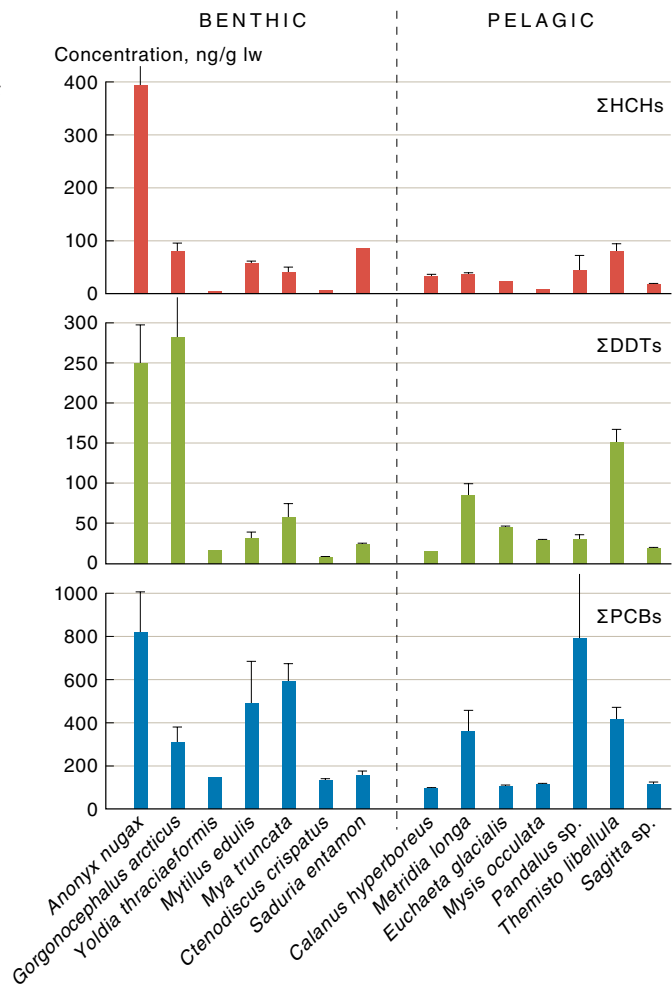


Figure 4-38. Concentrations of  $\Sigma$ HCHs,  $\Sigma$ DDTs, and  $\Sigma$ PCBs in Arctic benthic and pelagic invertebrates. All samples were collected in 1998 in northern Baffin Bay except *M. truncata* (1993 in Iqaluit harbor), *M. edulis* (1998 at seven locations in northern Quebec) and *S. entamon* (1998 in northern Alaska near Barrow). Results of *A. nugax* and *G. arcticus* include samples from Cumberland Sound. All data from Fisk *et al.* (2002a). Bars are mean  $\pm$  1 SE; see Annex Table 10 for sample numbers.

lies, arrow worms, and medusae) to larger crustaceans. The exception was PCBs, concentrations of which were highest in the zooplankton >1 mm.

To investigate the levels of OCs in sympagic (i.e. associated with ice edges) fauna, ice-associated amphipods were collected in the marginal ice zone north of Svalbard and in the Fram Strait in September 1998 and 1999 (Borgå *et al.*, 2002a; 2002b). Amphipods dominate the sympagic fauna (Melnikov, 1997), and represent a direct link between ice-algae and sympagic meiofauna and higher trophic levels (Lønne and Gulliksen, 1989; Lønne and Gabrielsen, 1992). Due to the general sea-ice drift pattern (Colony and Thorndike, 1985) and the possible transport of contaminants by sea ice (Pfirman *et al.*, 1995), the contaminant occurrence in organisms in melting areas, such as the marginal ice zone north of Svalbard and in the Fram Strait, is of special interest and may explain elevated OC concentrations in Svalbard marine mammals and seabirds. Three species of amphipods, representing a number of feeding strategies, were analyzed for OCs (Borgå *et al.*, 2002a; 2002b) including *Apherusa glacialis*, *Onisimus* sp. and *Gammarus wilkitzkii*. OC concentrations increased from the more herbivorous *A. glacialis* to the more carnivorous *G. wilkitzkii* and the more necrophagous *Onisimus* sp. (Borgå *et al.*, 2002a). The composition of compound classes of HCHs, chlordanes, and DDTs was similar between the amphipod families, whereas the profiles of PCBs differed.

Even though marine invertebrates have direct uptake by passive diffusion of contaminants across their gills, these results imply that the species' ecology, such as diet, is important in the bioaccumulation process of OCs. In addition, the results show that the sea-ice drift route influences the concentrations of OC pollutants in sympagic organisms.

In sympagic amphipods, the levels and patterns of OCs differed spatially, with higher  $\alpha$ -HCH concentrations in amphipods from the Fram Strait in comparison to amphipods collected north of Svalbard (Borgå *et al.*, 2002c). This could be related to the sea-ice drift route, since sea ice in the Fram Strait had a drift route across the central Arctic Ocean, while the sea ice north of Svalbard had a western drift route to the sampling stations.

The largest and longest-living sympagic amphipod, *G. wilkitzkii*, lives up to five years, reaching a maximum body length of 62 mm (Lønne and Gulliksen, 1991; Poltermann, 1997; 2000). With increasing age and size, *G. wilkitzkii* switches gradually from herbivory to a more carnivorous diet (Werner, 1997; Poltermann, 2000; Scott *et al.*, 2001). The OC burden in *G. wilkitzkii* may increase with age due to bioaccumulation over time, and due to increased biomagnification associated with the amphipod's age and size-related shift in trophic position. In addition, the contaminant burden in gilled organisms may be related to increasing size and body mass due to changes in lipid content, total metabolism, and the surface-to-volume ratio of gills which influences the exchange rate of OCs with water (Landrum, 1988; Walker *et al.*, 1996). The OC concentrations in two size-classes (small <29 mm and large >29 mm) of *G. wilkitzkii* were analyzed. The concentrations of more lipophilic compounds (e.g., PCBs) were lower in the smaller size-class, whereas concentrations of more hydrophobic com-

pounds (e.g., HCHs) did not differ by size-class (Borgå *et al.*, 2002c).

#### *Benthic marine invertebrates*

Benthic invertebrates have a larger range of sizes, feeding ecology, and ecological niches than pelagic zooplankton, and therefore, have a greater range of OC concentrations (Fisk *et al.*, 2002a). Scavenging benthic invertebrates have the potential to have very high OC concentrations. PCBs are the OC group with the greatest concentrations in benthic invertebrates, with chlordanes and DDT having high concentrations in certain species.

A number of Arctic benthic invertebrates have been analyzed for OCs since the previous AMAP assessment (see Annex Table 10). As with other invertebrates,  $\Sigma$ PCB concentrations were highest in benthic species, but the concentrations of the other OC groups varied with the invertebrates' feeding strategy (Figure 4-38). Scavenging invertebrates, such as the amphipod *Anonyx nugax* and basketstar *Gorgonocephalus arcticus*, have  $\Sigma$ HCH,  $\Sigma$ CHL and  $\Sigma$ DDT concentrations that are within 30-50% of  $\Sigma$ PCB concentrations. In filter-feeding and detritus-feeding invertebrates, such as *Yoldia thraciaeformis* or *Ctenodiscus crispatus*, the concentrations of other OC groups are much lower (approximately 20%) than  $\Sigma$ PCB concentrations. The highest concentrations of OCs in invertebrates are found in *A. nugax*, which have concentrations in the same range as found in Arctic cod and the little auk (seabird) (Fisk *et al.*, 2001c). OC concentrations in filter- and detritus-feeding invertebrates are among the lowest found in any biota in the world and are similar to pelagic zooplankton (Figure 4-38).

The blue mussel is a monitoring species that has been commonly used throughout the world, including the Arctic, to monitor OCs. PCBs and other OCs were present at very low levels in blue mussels from five locations in Nunavik, in the Ungava Bay and Hudson Strait areas, and two locations in Labrador, all from the eastern Canadian Arctic (Fisk *et al.*, 2002a) (see Annex Table 10 for locations). PCBs were present at much higher concentrations (range of means 3.7-46 ng/g ww) in mussel tissues compared to other OCs. HCH isomers were the next most prominent contaminants, with means ranging from 1.1 to 2.9 ng/g ww. The greater sorption of PCBs to sediment particles filtered by mussels, compared to most other OCs, especially HCH, may account for the much higher concentrations of PCBs. Doidge *et al.* (1993) found low ng/g ww levels of PCBs and OC pesticides in a survey of blue mussels from six communities in Nunavik. Levels of PCBs in mussel samples from Kuujuaq were considerably higher than in the same species at most other locations, suggesting that local contamination sources might be a factor.

Levels of PCBs were determined in mussels (*Mytilus trossulus*) from 39 sites within the Aleutian Islands and five sites from southeast Alaska in 1994/1995 to determine whether high PCB levels in sea otter (*Enhydra lutris*) were due to local sources (Reese, 1998). Very high PCB concentrations were found at Amchitka, Adak, and Unalaska Islands (83, 430, and 2800 ng/g dw, respectively) but low levels were found elsewhere (ranging from 7.1 to 51 ng/g dw). This is a clear example of the influence of point source contamination, probably from military facilities, in the Arctic, which results in concentra-



tions that rank with the most contaminated sites in North America. Spatial patterns of OC pesticides suggest that these compounds come from sources that are more distant.

In 1997, soft parts of blue mussels collected in the Faroe Islands were analyzed for OCs (Larsen and Dam, 1999). The highest concentrations detected were for CB153 and *p,p'*-DDE, both at 0.7 ng/g ww and CB138 and *p,p'*-DDT, both at 0.4 ng/g ww. These concentrations are in the range of those measured in blue mussels in the eastern Canadian Arctic (Fisk *et al.*, 2002a) but are much lower than those measured close to the military sites in southeastern Alaska.

To assess the influence of point sources of pollutants on Svalbard (Spitsbergen), five species of benthic invertebrates were collected at various distances from possible point sources in 1998 and 1999 and analyzed for HCB, PAHs and PCBs (Hop *et al.*, 2001). Stable isotope analysis revealed that these species were all feeding at a low trophic level. Local inputs of PAHs and PCBs seemed to be present in some of the island's harbors and were elevated in benthic fauna compared to samples from Bjørnøya, which has little industry. The PCB profiles were similar to technical PCB, suggesting a local source. Concentrations were higher at sites closer to settlements. HCB did not appear to have local sources. This study provides more evidence that point sources in the Arctic can have a significant influence on the observed OC levels in wildlife.

In a recent study (Evenset *et al.*, 2002), spider crabs (*Hyas araneus*) were collected by diving in the littoral zone off the southwest part of Bjørnøya (late June 2000). Spider crabs had slightly higher PCB and pesticide concentrations than zooplankton collected from the same region. This study produced the first toxaphene results for Arctic benthic invertebrates and showed that toxaphene is present, although at lower concentrations than  $\Sigma$ PCBs,  $\Sigma$ DDTs and  $\Sigma$ HCHs.

#### 4.4.3.1. TBT

TBT received limited attention in the first AMAP POPs assessment, as only a few invertebrates had been analyzed for TBT and its metabolites (DBT, MBT). TBT and its degradation products have now been measured in the blue mussel in Greenland, the Faroe Islands, Norway, and Iceland. Most of the studies dealing with TBT in the western part of the Atlantic are from animals living south of the AMAP area (Chau *et al.*, 1997; St-Jean *et al.*, 1999). In Greenland, TBT levels have only been examined near Nuuk, the largest town ( $\approx$ 13 000 inhabitants) in central western Greenland (Jacobsen and Asmund, 2000). Levels there were quite low (close to 1 ng/g ww) despite the fact that Nuuk harbor hosts several large shrimp trawlers. In the Faroe Islands, TBT was measured in the blue mussel at eight sites in 1996. The levels ranged from 49 to 372 ng Sn/g dw (Følsvik *et al.*, 1998). Highest levels were found near Tórshavn, the capital of the Faroe Islands. In northern Norway, the levels of TBT were generally very high in 1993 and 1994 (Berge *et al.*, 1997), in particular near harbors, where levels as high as 4407 ng/g dw have been reported. In Iceland, the levels of TBT in the blue mussel were on average 122.6 ng/g ww  $\pm$  92.5 (SD) near the large harbor

area of Reykjavík. At sites in southwestern, western, and northwestern Iceland, the levels were usually low (14.7 ng/g ww  $\pm$  13.2 (SD)) (Skarphéðinsdóttir *et al.*, 1996). High levels of TBT are still found in the blue mussel near larger harbors. Near Reykjavík harbor, the level of TBT was around 4420 ng/g dw in 2000 (Hall-dórsson, 2002).

TBT levels were examined in the dogwhelk along the Norwegian coast from 1993 to 1995 (Følsvik *et al.*, 1999) and again in 2000 (Green *et al.*, 2002). The concentrations ranged from 8 to 141 ng Sn/g dw at sites within the AMAP area in 1993 to 1995. Out of ten sites north of 65° N, TBT was at or below the detection limits at three of the sites. The most recent studies of TBT levels show that at eight locations in northern Norway, the levels are lower than found in the earlier study (Green *et al.*, 2002). Two sites had less than 7 ng Sn/g dw, while at the rest of the sites, the levels ranged from approximately 35 to 100 ng Sn/g dw.

TBT levels were examined in the limpet, *Patella vulgata*, at eight sites in the Faroe Islands (Følsvik *et al.*, 1998). At only one site, near Tórshavn, were the levels above detection limits (90 ng Sn/g dw). Skarphéðinsdóttir *et al.* (1996) measured TBT levels in the blue mussel and the dogwhelk at a single site near Reykjavík harbor at different times of the year. The levels of TBT in the tissue of the blue mussel and the dogwhelk seem to vary considerably over the year at northern latitudes. High levels (approximately 60-70 ng/g ww) were found in the blue mussel from July to January, while from February to April, the levels were less than 15 ng/g ww. In the dogwhelk, the summer and autumn levels were approximately 30-40 ng/g ww, while during late winter and early spring the levels were comparable to those found in the blue mussel. Skarphéðinsdóttir *et al.* (1996) concluded that monitoring of TBT in these animals at high latitudes should be confined to late summer or early autumn.

#### 4.4.4. Marine and anadromous fish

Marine and anadromous fish occupy a range of trophic positions in Arctic marine ecosystems, and hence concentrations of OCs are quite variable. The Arctic cod (polar cod, *Boreogadus saida*) is considered a key link in marine food webs between invertebrates and seabirds/marine mammals, including the ringed seal and beluga. A number of these fish species are important components of the traditional human diet (e.g., Arctic char) and several have become important commercially (e.g., Greenland halibut (turbot, *Reinhardtius hippoglossoides*). There was a somewhat large dataset available for OCs in marine fish in the first AMAP POPs assessment (de March *et al.*, 1998). Less data have been produced on these organisms since that report; however, the data produced recently are of improved quality with a greater range of contaminants measured, particularly PCB congeners. Six marine fish species were recommended for inclusion in the AMAP Phase II monitoring program, including Arctic char, whitefish/cisco (*Coregonus* spp.), Arctic cod, Atlantic cod, sculpin (*Myoxocephalus* spp.) and long rough dab (*Hippoglossoides platessoides*). Data on 'new' POPs have only been produced for a few of these species.



OCs have generally been measured in the liver or muscle tissue of fish. The amount of lipid found in these tissues can vary significantly within and between species and needs to be taken into account when comparing levels of OCs between species. For example, the Atlantic and Arctic cod have high-lipid livers and low-lipid muscle, whereas the Greenland halibut has similar lipid content in liver and muscle. Differences in size, growth rate (Johnston *et al.*, 2002), and trophic position (Fisk *et al.*, 2001c) can all influence observed OC levels in fish and need to be considered when comparing different populations, locations or studies. For most of the OC data produced for Arctic fish, these variables have not been adequately accounted for.

Concentrations of OCs in marine fish are generally higher than those observed in freshwater fish and marine zooplankton. For some larger species of fish, OC levels are similar to ringed seals, likely due to feeding at a higher trophic level (Fisk *et al.*, 2002b).

#### Sea-run Arctic char

Low levels of OCs were detected in anadromous Arctic char muscle (including skin) from two locations in Labrador and three locations in Nunavik in the Canadian Arctic (Muir *et al.*, 2000c) (Annex Table 10). PCBs were the most prominent contaminants at Labrador sites, with concentrations averaging 30 ng/g ww in samples from Makkovik and 63 ng/g ww in samples from Nain.  $\Sigma$ PCB levels in char muscle samples from the three locations in Nunavik were lower, with means ranging from 13 to 18 ng/g ww. Statistical analysis showed that there were no significant differences in  $\Sigma$ PCB concentrations among locations. Lipid content, length, and age were not significant co-variates.  $\Sigma$ PCB levels were, however, influenced by the sex of the fish, with males having significantly higher levels than females. This may be due to the timing of sampling near the spawning period for the char. Females may have mobilized fat from muscle to reproduce and, subsequently, transferred contaminants to their eggs as the fat is mobilized, thus lowering the concentrations observed in muscle. The next most prominent groups of OCs in char muscle were the HCH and DDT groups, which were present at low ng/g levels.  $\Sigma$ DDT levels were higher at Nain and Lavrentiya than other locations. In general, these levels of persistent OCs are similar to or lower than those reported in char muscle from other locations in the Canadian Arctic (Muir *et al.*, 1999b). Concentrations of all OCs were much lower in muscle samples from sea-run char from Finland.

#### Arctic (polar) cod

PCBs was the dominant OC group, followed by CHLs in Arctic cod collected in northern Baffin Bay in 1998 (Fisk *et al.*, 2002c), Barrow, Alaska in 1998 and 1999 (Hoekstra *et al.*, 2003a), and the Barents Sea in 1995 (Borgå *et al.*, 2001). For Arctic cod collected around Jan Mayen,  $\Sigma$ DDTs was the next dominant OC group after PCBs (Gabrielsen *et al.*, 1997). Concentrations (lipid corrected) of  $\Sigma$ PCBs were quite similar in cod from the Barents Sea and around Jan Mayen but were somewhat higher in northern Baffin Bay and Alaskan cod (Figure 4-39).  $\Sigma$ DDTs was somewhat higher in the northern Baffin Bay Arctic cod than other locations.  $\Sigma$ HCHs was highest in the Canadian cod, reflecting the higher con-

Concentration in Arctic cod, ng/g lw

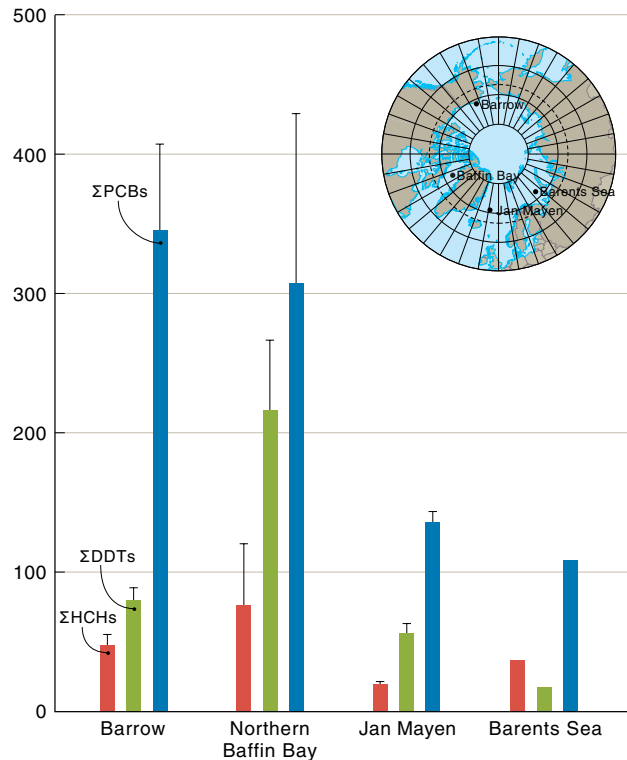


Figure 4-39. Concentrations ( $\pm$ SD) of  $\Sigma$ HCHs,  $\Sigma$ DDTs, and  $\Sigma$ PCBs in Arctic cod collected in Barrow, Alaska (Hoekstra, 2002b), northern Baffin Bay (Fisk *et al.*, 2001c), Jan Mayen (Gabrielsen *et al.*, 1997), and that Barents Sea (Borgå *et al.*, 2001).

centrations also seen in seawater. These spatial trends generally follow those of zooplankton (Section 4.4.3) but are not consistent with spatial trends observed in marine mammals and seabirds at higher trophic levels (Muir *et al.*, 2000b) (Sections 4.4.5 and 4.4.6).

#### Atlantic cod

OC data were also generated recently (1998-2000) for Atlantic cod from the Barents Sea (Borgå *et al.*, 2001), locations around Iceland (Yngvadóttir and Halldórsdóttir, 2002), and southwestern Greenland (Muir and Johansen, 2001). Concentrations of OCs were consistent at all locations. Concentrations (ng/g lw) of  $\Sigma$ HCHs ranged from 15 (Iceland) to 41 ng/g lw (Barents Sea) and  $\Sigma$ PCB concentrations from 199 (Iceland) to 281 (southwestern Greenland) ng/g lw. In the Barents Sea, concentrations in Atlantic cod were about twice as high as those observed in Arctic cod, and could be due to feeding at a higher trophic level, larger size or longer life span of the Atlantic cod. The Icelandic data were part of a continuing monitoring program in Iceland, which also includes the dab (*Limanda limanda*), and is discussed under temporal trends in Section 5.4.2.

#### Faroe Islands fish

In the Faroe Islands, livers from shorthorn sculpin sampled in 1999 and 2000 ( $n=13$  and  $15$ ) were analyzed (Hoydal *et al.*, 2001). The single OC occurring in highest mean concentrations was *p,p'*-DDE at 84 and 134 ng/g ww in 1999 and 2000, respectively. The second highest mean concentration was CB153 at 58 and 94 ng/g ww in 1999 and 2000, respectively. The concentrations of chlordanes, toxaphene, and mirex were less by a fac-

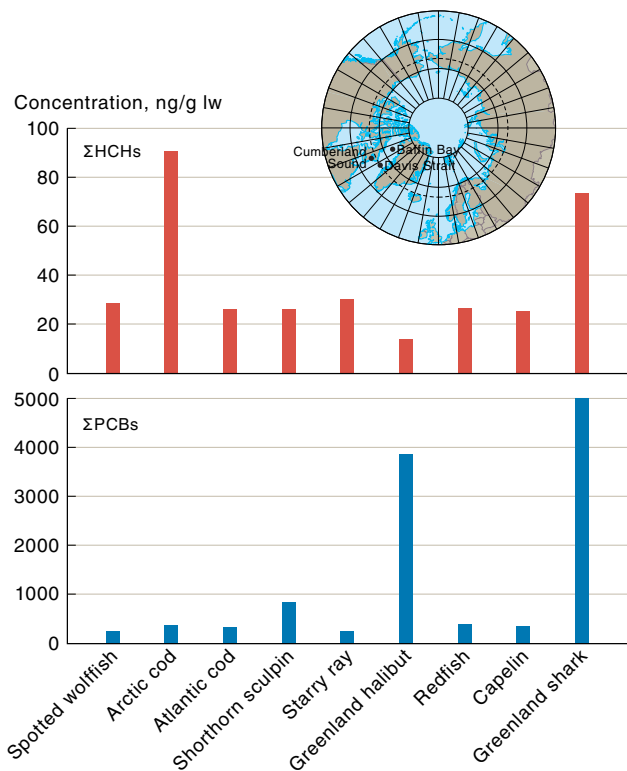


Figure 4-40. Concentrations of  $\Sigma$ HCHs and  $\Sigma$ PCBs in Arctic marine fish collected in Davis Strait and Baffin Bay (all species except Greenland shark, which were collected in Cumberland Sound). Arctic cod data from (Fisk *et al.*, 2001c), Greenland shark data from Fisk *et al.* (2002b), all other data from Denmark (2002).

tor of 8 or more. There was large variability in OC concentrations between individual fish. No significant correlations between length or age and OC concentrations were found. Overall, the concentrations of OCs were higher in the year 2000 samples than in 1999. The reason may be related to the fact that the sampling stations were not in identical locations between years. A local pollution source at one of these sites as an explanation for between-year differences is not likely, due to the similar ratios of concentrations for PCB, toxaphene, DDTs, and mirex.

Results are also available for Atlantic cod sampled in 1997 at the Faroe Islands shelf (Larsen and Dam, 1999). Liver samples from 45 fish were analyzed in two pools with lipid contents of 56.9 and 53.5%. As for sculpin, the single OC occurring in highest concentrations was *p,p'*-DDE, at 40 and 37 ng/g ww, with CB153 having the second highest concentration at 25 and 26 ng/g ww, respectively. In contrast to sculpin, the concentration of *trans*-nonachlor was similar to that of CB153, near 20 ng/g ww and *p,p'*-DDD was almost half the concentration of *p,p'*-DDE in Atlantic cod. In sculpin, the ratio of *p,p'*-DDD to *p,p'*-DDE was close to 1/100.

#### Greenland marine fish

Concentrations of OCs have been measured in a range of marine fish around Greenland (Figure 4-40 and Annex Table 10). A majority of these were collected as part of a larger study on OCs in foodstuffs. There is a lack of data from other Arctic regions to use for spatial comparisons. Concentrations of OCs were fairly consistent across species with the exception of the Greenland shark (*Somniosus microcephalus*) and Greenland halibut, which

had higher levels. The Greenland halibut is a large, benthic fish, which may account for the higher levels. OC levels in the Greenland shark are discussed below in this section. PCBs were the predominant OCs followed by  $\Sigma$ DDTs and chlordanes in these fish, reflecting their generally higher trophic level, as compared with zooplankton.

#### Greenland shark and Greenland halibut

Concentrations of OCs in the Greenland sharks collected in the Davis Strait and Cumberland Sound region in 1997 and 1999 (Fisk *et al.*, 2002b) were in the range of other top Arctic marine predators, the polar bear, and glaucous gull (Figure 4-41). The Greenland shark is the only known shark to routinely inhabit Arctic waters, and limited evidence indicates that these animals may be long-lived (>100 years) (see Hansen, 1963) and potentially numerous. Concentrations (lipid basis) of OCs in Greenland sharks in the study of Fisk *et al.* (2002b) were not related to fork length, sex,  $\delta^{13}\text{C}$  or  $\delta^{15}\text{N}$ . Greenland sharks had higher OC concentrations than black dogfish (*Centroscyllium fabricii*), a small (<1 m in length), deep-water shark collected in Davis Strait (see Berg *et al.*, 1997). Lower OC concentrations in the black dogfish were not unexpected because this species feeds lower in the food web, mainly on crustaceans and small fish (Compagno, 1984). Concentrations of OCs (lipid basis) in Greenland sharks were 10-100 and 3-10 times higher than those observed in Greenland halibut and ringed seals, respectively, suggesting a very high trophic position. Marine mammals have been found in the stomachs of Greenland sharks (Fisk *et al.*, 2002b). Concentrations of  $\Sigma$ DDTs in the study samples of Greenland sharks are among the highest in Canadian Arctic biota, which may be related to the low metabolism and long life span of these sharks. Reported concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs in Greenland halibut collected in the Davis Strait in 1992 (Berg *et al.*, 1997) were five and ten times higher, respectively, but concentrations of  $\Sigma$ CHL and  $\Sigma$ HCH were very similar to those reported for Greenland halibut in the study of Fisk *et al.* (2002b). The higher PCB and DDT in Greenland halibut reported in Berg *et al.*, (1997) could be due to the larger size and the deeper-water habits of these Greenland halibut.

CB153 concentration, ng/g lw

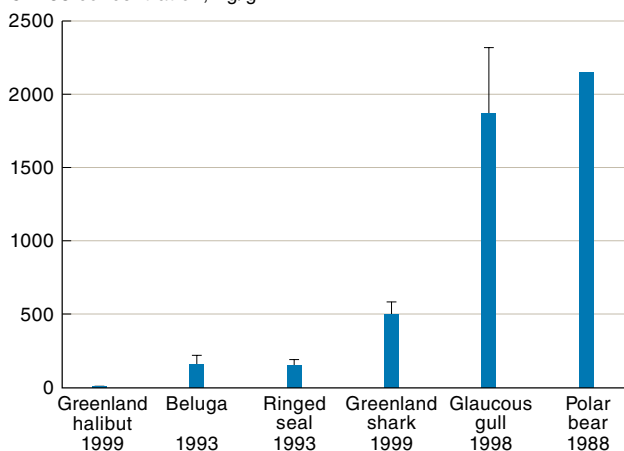


Figure 4-41. Mean ( $\pm$ SE) concentrations of CB153 in Greenland halibut (Fisk *et al.*, 2002b), beluga (Muir *et al.*, 1999b), ringed seal (Muir *et al.*, 1999c), Greenland shark (Fisk *et al.*, 2002b), glaucous gull (collected in northern Baffin Bay, Fisk *et al.*, 2001c), and polar bear (mean only) (Norstrom *et al.*, 1998) from Cumberland Sound.

*Jan Mayen fish*

Long rough dab, capelin (*Mallotus villosus*), grey gurnard (*Eutrigla gurnardus*), coalfish (*Pollachius virens*), Atlantic poacher (*Leptagonus decagonus*), daubed shanny (*Lumpenus maculatus*), and checkered eelpout (*Lycodes vabli*) were collected from around Jan Mayen, and liver samples were analyzed for OCs (Gabrielsen *et al.*, 1997). ΣPCBs were the predominant contaminants followed by ΣDDTs. On a lipid weight basis, the highest OC concentrations were seen in long rough dab and checkered eelpout.

*Toxaphene in fish from European waters*

Concentrations of three toxaphene congeners (Parlars 26, 50 and 62) were determined in 221 fish samples, covering 23 species from northern European waters (McHugh *et al.*, 2000). Levels of the three congeners were highest in the Barents Sea and Norwegian Sea regions compared with the coast of Iceland, the North Sea, and the Baltic Sea. However, some of the differences between regions could be due to the use of different fish species, which will feed differently and influence exposure levels. Toxaphene concentrations in these fish are in the range reported for other marine fish from this region (see Annex Table 10).

## 4.4.4.1. 'New' chemicals in marine and anadromous fish

*PBDEs in marine fish*

PBDEs have been determined in the liver of a few species of marine fish: Atlantic cod and tusk (*Brosme brosme*) from Norway (Herzke, 2002a); and, Atlantic cod (liver and muscle) and Greenland halibut (liver) from Greenland (Muir and Johansen, 2001) (Annex Table 17). ΣPBDE levels in the liver of the Norwegian fish ranged from 24 to 109 and 60 to 300 ng/g lw in the Atlantic cod and tusk, respectively. These concentrations are much higher than levels observed in the fish off southwestern Greenland. Shorthorn sculpin, cod, spotted wolffish, and starry ray were studied during 2000 near the western Greenland villages of Quaortoq (3500 inhabitants), Igaliko (30 inhabitants), and Usuk (background site 3-5 km from Igaliko) (Christensen *et al.*, 2002; Vorkamp *et al.*, 2002). The highest concentrations of PBDEs were observed in fish from Quaortoq (46 ng/g lw) followed by Igaliko (18 ng/g lw) and Usuk (12 ng/g lw). The levels of PBDEs were 15-24 times lower than PCB levels measured in the same individuals, except for shorthorn sculpin collected at Quaortoq, where the level of ΣPCBs was 40 times higher. For all fish, BDE47 dominated the PBDE congener pattern.

## 4.4.5. Seabirds

There are approximately fifty species of Arctic seabirds (de March *et al.*, 1998). They have a variety of feeding and migration strategies, and accordingly, POP concentrations can vary considerably among species. Species, such as the glaucous gull, great skua (*Stercorarius skua*), and great black-backed gull (*Larus marinus*), that may migrate to more contaminated regions and/or that scavenge, particularly on dead marine mammals, have the highest POP concentrations (Gabrielsen *et al.*, 1997; Borgå *et al.*, 2001; Fisk *et al.*, 2001b; 2001c). Biological

effects related to POP exposure have been seen in highly contaminated glaucous gulls (Bustnes *et al.*, 2000), and concerns about POP effects remain for some other species of Arctic seabirds. Seabirds also have the potential to biologically transport POPs from one region to another (e.g., marine to the freshwater ecosystems) through the production and release of guano (see Section 4.3.5).

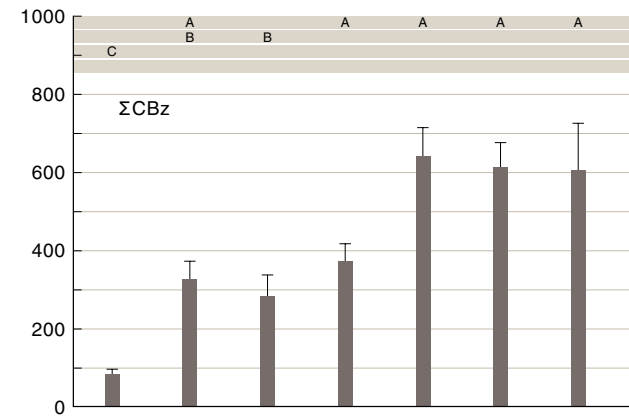
There has been a considerable amount of work done on POPs in Arctic seabirds since the first AMAP assessment report, encompassing a wide range of species (Annex Table 11). Continued monitoring of POPs in Canadian Arctic seabird eggs from 1975 to 1998 and beyond, has provided a strong dataset for examining temporal trends between different species (Section 5.4.3). A comprehensive study of OCs in Barents Sea seabird eggs (Barrett *et al.*, 1996) was not included in the first AMAP POPs assessment. A number of studies have also been carried out examining OC concentrations and dynamics in adult seabirds (Henriksen *et al.*, 2000; Borgå *et al.*, 2001; Fisk *et al.*, 2001b), some of which were part of larger studies examining spatial trends (Gabrielsen *et al.*, 1997) or food web dynamics of OCs, discussed in Section 4.4.9. New data on PCDD/Fs, toxaphene, and 'new' POPs have also been generated for Canadian seabirds (Braune *et al.*, 2001a) and on mono-*ortho* and non-*ortho* PCBs, toxaphene, and 'new' POPs in glaucous gulls on Bjørnøya and Svalbard (Daelemans *et al.*, 1992; Burkow *et al.*, 2001; Herzke *et al.*, 2003).

PCBs are the most common OCs measured in Arctic seabirds (see Figure 4.42) (Barrett *et al.*, 1996; Borgå *et al.*, 2001; Braune *et al.*, 2001b; Buckman *et al.*, 2004), although the relative amounts are related to the trophic position of the seabird (Borgå *et al.*, 2001). Higher trophic-level seabirds generally have a larger proportion of PCBs as a percentage of total OCs. This is due to the greater biomagnification potential of PCBs and the ability of seabirds to metabolize other OCs, such as α-HCH (Moisey *et al.*, 2001) and some chlordanes components (Fisk *et al.*, 2001b). Other recalcitrant and biomagnifying OCs, such as *p,p'*-DDE and oxychlordanes, also become more prevalent (Borgå *et al.*, 2001; Fisk *et al.*, 2001b), although the relative proportions can vary dramatically between species, even within the same family (Fisk *et al.*, 2001b). Of particular note are the relative proportions of HCH isomers, which in seabirds are dominated by β-HCH (Barrett *et al.*, 1996; Moisey *et al.*, 2001). Seabirds are able to efficiently metabolize α- and γ-HCH but not β-HCH (Moisey *et al.*, 2001).

*Species comparison*

OC concentrations in seabirds were, in general, related to trophic position and secondarily to migration, with the highest concentrations found in great skuas, great black-backed gulls, and glaucous gulls. These are species that scavenge, prey on other seabird species, and migrate to more southerly habitats in the winter (Barrett *et al.*, 1996; Gabrielsen *et al.*, 1997; Borgå *et al.*, 2001; Fisk *et al.*, 2001c). Glaucous gulls, however, do not migrate as far as the other two species. These concentration-trophic level relationships in seabirds hold throughout the Arctic, including the Barents Sea area, and around the Jan Mayen, northern Baffin Bay, and the Iceland regions (Barrett *et al.*, 1996; Gabrielsen *et al.*, 1997; Borgå *et al.*,

Concentration in liver, ng/g, lw



Concentration in liver, ng/g, lw

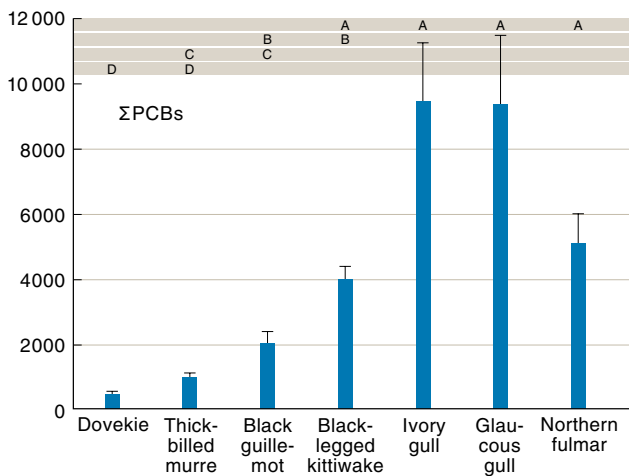
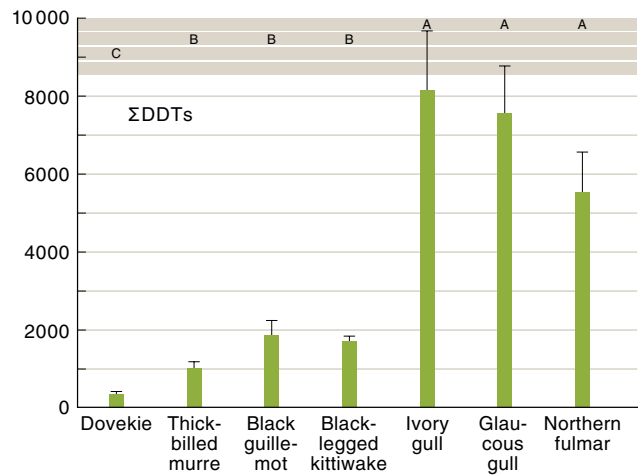
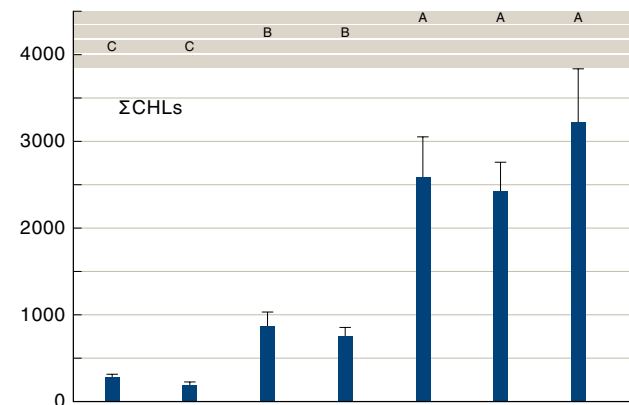
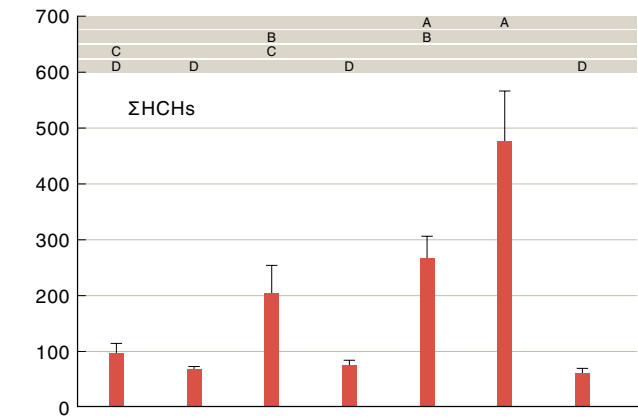


Figure 4.42. Mean ( $\pm$  SE) concentrations of  $\Sigma$ CBz,  $\Sigma$ HCHs,  $\Sigma$ CHLs,  $\Sigma$ DDTs, and  $\Sigma$ PCBs in liver tissue of Northwater Polynya seabirds (Buckman *et al.*, 2003). Concentrations in males and females did not significantly differ ( $p < 0.05$ ) except for  $\Sigma$ DDTs, and data were therefore combined. Bars with the same letter do not differ significantly ( $p > 0.05$ ).

2001; Fisk *et al.*, 2001c; Ólafsdóttir *et al.*, 2001). In northern Baffin Bay, ivory gull (*Pagophila eburnean*) and northern fulmar (*Fulmarus glacialis*), both of which scavenge, had slightly lower OC concentrations than glaucous gulls from the same area but higher concentrations than other seabirds (Figure 4.42) (Buckman *et al.*, 2004). OC concentrations in black-legged kittiwakes (*Rissa tridactyla*), a non-scavenging species, were lower than scavenging seabird species but were higher than the fish- and zooplankton-eating alcids (guillemots, murres, and little auks) in both Baffin Bay and the Barents Sea area. On Jan Mayen, dovekies had the lowest OC concentrations; black-legged kittiwakes, fulmars, and black guillemots had intermediate concentrations; and, highest concentrations were found in glaucous gulls, great black-backed gulls, and great skuas (Gabrielsen *et al.*, 1997).

Black-legged kittiwakes have been shown to feed at a lower or similar trophic level as the black guillemot (Hobson *et al.*, 2002). Kittiwakes are known to migrate longer distances (e.g., to southern regions) than black guillemot, which probably only move locally during the winter (Anker-Nilssen *et al.*, 2000). Therefore, the higher OC concentrations in black-legged kittiwakes are likely to be a result of uptake at their winter habitat, highlighting the impact, in both the Canadian and European Arctic, of migration to more contaminated regions.

OC concentrations can also vary within a single population of seabirds. Sagerup *et al.* (2002) observed a large range of OC concentrations in the livers of glaucous gulls collected in the western Barents Sea in 1996. For example,  $\Sigma$ PCB concentrations ranged from 16 000 to 292 000 ng/g lw. Concentrations of OCs were signifi-



cantly correlated to  $\delta^{15}\text{N}$  ( $r^2 = 0.18$ ), and therefore, diet accounted for some of the variation within this population. Bustnes *et al.* (2000) studied two colonies of glaucous gulls on Bjørnøya that nested only a few kilometers from each other but had different food habits. The two colonies had different OC concentrations, with the cliff colony having much higher concentrations than the sea-level colony. The colony that nested on cliffs fed more on other seabird eggs and chicks, whereas indications were that the sea-level colony fed more on fish (i.e. fed at a lower trophic level).

#### *Influence of sex and tissue*

Lipid-corrected concentrations of OCs have rarely been found to vary significantly between sexes or tissues of seabirds (Ólafsdóttir *et al.*, 1998; Buckman *et al.*, 2004). For example, concentrations of OCs were found to be similar between liver and muscle, tissues with similar lipid content, of the common eider (*Somateria mollissima*) collected in Iceland (Ólafsdóttir *et al.*, 1998). This is a commonly observed phenomenon and reflects the hydrophobic nature of OCs and accumulation in lipid. Lipid-normalized concentrations of OCs generally did not vary to a large extent between liver and fat in seabirds (Buckman *et al.*, 2004; Fisk *et al.*, 2001b), but lipid-corrected concentrations of chlordane compounds were significantly lower in the liver compared with the fat of little auks collected in Baffin Bay in May/June. At that time, little auks are feeding heavily on low trophic-level zooplankton (e.g., copepods) in anticipation of reproduction and egg production. Ólafsdóttir *et al.* (1998) observed large seasonal changes in OC concentrations in the muscle and liver tissue of the common eider, which were attributed to relocation of OCs to these tissues due to the shrinking of body fat. Therefore, changes in diet, the effects of migration and/or reproduction can, in certain cases, result in differences between tissues and in seasonal changes in OC concentrations in seabirds.

#### *Spatial trends*

Spatial trends of OCs in Arctic seabirds varied with the seabird species and the chemical, and generally were in agreement with spatial trends observed previously in seabirds (de March *et al.*, 1998) and marine mammals (Muir and Norstrom, 2000; Muir *et al.*, 2000b). Concentrations of most OCs in seabirds are highest in the Russian part of the Barents Sea and lowest in the North American Arctic, with the exception of HCH, which is higher in the North American Arctic (Annex Table 11). There is a lack of data for the eastern Russian Arctic.

$\Sigma\text{PCB}$  data for four seabird species (thick-billed murre or Brünnich's guillemot, black guillemot, black-legged kittiwake, and glaucous gull) are available from a number of locations, and highlight the geographical trends despite different tissues having been analyzed. The first three of these seabird species are non-scavengers, feeding on zooplankton and fish, although the black guillemot is more of an inshore feeder. Black guillemots do not migrate, but thick-billed murrens do. Black-legged kittiwakes migrate long distances, exposing themselves to higher levels of OCs in more southerly habitats.  $\Sigma\text{HCH}$  concentrations were highest in the Canadian seabirds (no HCH data are available for the Bering Sea murrens), which reflects the higher concentrations of these chemicals in the

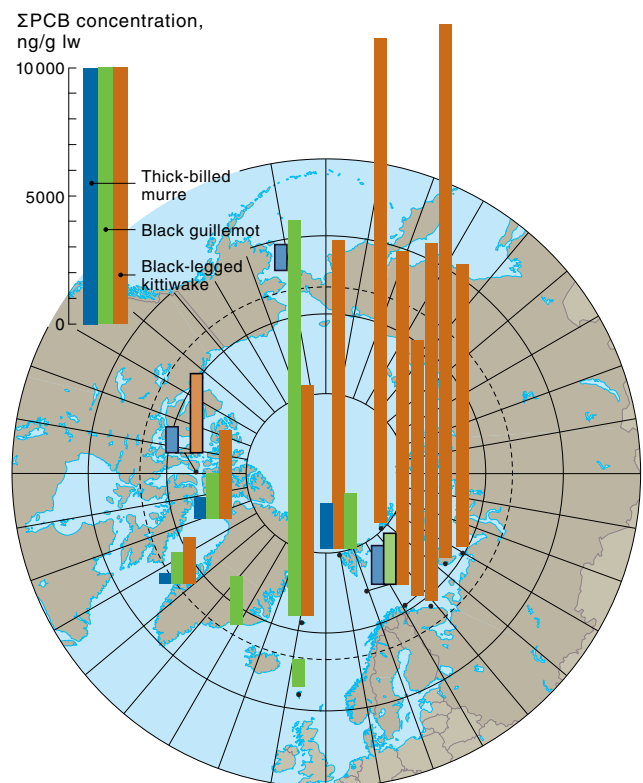


Figure 4.43. Concentrations of PCBs in liver (solid columns) and eggs (lighter-shaded, outlined columns) of thick-billed murre, black guillemot, and black-legged kittiwakes from regions throughout the Arctic. Data for the Bering Sea (year of collection: 2000; Vander Pol *et al.*, 2002), Prince Leopold Island (1998; Braune *et al.*, 2001a), northern Baffin Bay (1998; Buckman *et al.*, 2002), West and East Greenland (1999, 2000; Muir and Johansen, 2001), Barents Sea (1995; Borgå *et al.*, 2001 and; 1995; Gabrielsen *et al.*, 1997), Bjørnøya (1999; Evensen *et al.*, 2002), Faroe Islands (1995/96; Hoydal *et al.*, 2001), and Jan Mayen (1995; Gabrielsen *et al.*, 1997).

Canadian Arctic due to the closer proximity to Asia, where HCH has recently been used.  $\Sigma\text{DDT}$  concentrations were similar within and between species in the Canadian and Barents Sea thick-billed murrens, black guillemots, and kittiwakes but were lower in the Greenland seabirds. There was much more variation for  $\Sigma\text{PCB}$  concentrations (Figure 4.43). For black guillemots, highest  $\Sigma\text{PCB}$  concentrations were found on Jan Mayen (15 300 ng/g lw in liver) followed by Iceland (4560 ng/g lw in muscle, not shown in figure). The higher concentrations on Iceland could in part be due to the inclusion of birds collected in 1991 and 1992 and/or due to muscle being analyzed instead of liver. For thick-billed murrens, highest  $\Sigma\text{PCB}$  concentrations were seen in the Barents Sea area and lowest concentrations in the North American Arctic. Black-legged kittiwakes had the highest  $\Sigma\text{PCB}$  concentrations of the three non-scavenger species, with highest concentrations found in the eastern Barents Sea around Novaya Zemlya and Franz Josef Land.  $\Sigma\text{PCB}$  (10 000–21 000 ng/g lw) and  $\Sigma\text{DDT}$  concentrations (500–1900 ng/g lw) in the kittiwakes from around the Barents Sea region, including Svalbard, Bjørnøya, Franz Josef Land, Novaya Zemlya, and the Kola Peninsula, were fairly similar and are probably indicative of exposure due to migration more than to local sources.

Concentrations of  $\Sigma\text{PCBs}$  and  $\Sigma\text{DDTs}$  in glaucous gulls, a high trophic-level scavenging seabird, were higher in the Barents Sea and Jan Mayen than in Baffin Bay,

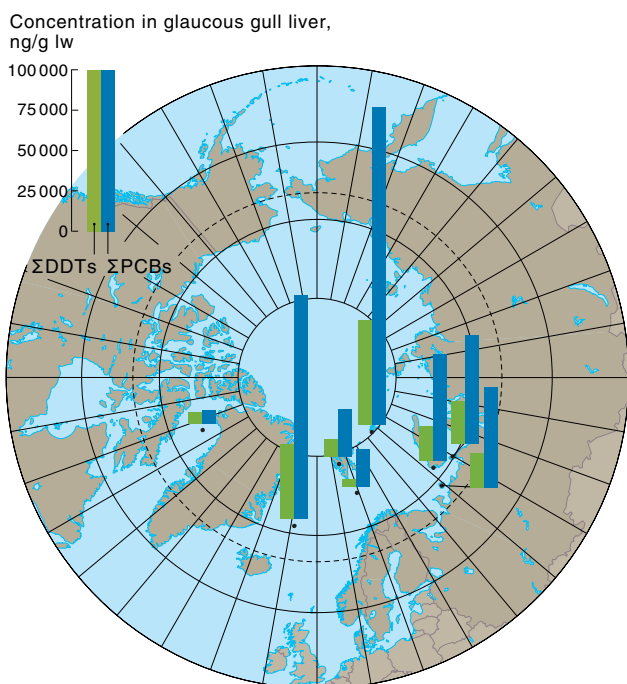


Figure 4-44. Concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs in glaucous gull liver. Data for Baffin Bay are from Fisk *et al.* (2001b), for all other sites data are from Gabrielsen *et al.* (1997).

with highest concentrations around Franz Josef Land (Figure 4-44). Higher  $\Sigma$ PCB and  $\Sigma$ DDT concentrations in seabirds from the Barents Sea region, relative to seabirds from more westerly regions, have been observed previously (de March *et al.*, 1998). OC levels, including  $\Sigma$ PCBs and  $\Sigma$ DDTs in zooplankton and fish from the Barents Sea were within the range of those recorded in similar species in the Canadian Arctic. The low OC levels in selected taxa at lower trophic positions were unexpected, based on the higher levels of  $\Sigma$ PCB and  $\Sigma$ DDT, in particular, in higher trophic-level predators in the Barents Sea compared to the Canadian Arctic (Borgå *et al.*, 2001). Therefore, a diet of pelagic organisms does not seem to explain the elevated concentrations in top predators in the Barents Sea area, here exemplified by glaucous gulls. A diet of ice-associated fauna may, however, be of importance in the bioaccumulation of OCs (Norstrom *et al.*, 1998). As suggested by Pfirman *et al.* (1995), contaminated particles in the sea ice may be released in the melting areas in the Fram Strait and the Barents Sea, followed by bioaccumulation in ice-associated fauna. However, ice-associated fauna from the marginal ice zone near Svalbard have low OC levels (Borgå *et al.*, 2002b; 2002c), which correspond to levels in zooplankton at similar trophic positions from the Barents Sea (Borgå *et al.*, 2001).

#### Canadian seabird eggs

As part of a continuing temporal-trend study of contaminants in seabird eggs (see Section 5.4.3), thick-billed murre, northern fulmar, and black-legged kittiwake eggs were collected on Prince Leopold Island in Lancaster Sound in 1998 and analyzed for OCs (Braune *et al.*, 2001b). Concentrations and the relative proportions of OC groups were in general agreement with those observed in tissues of adult birds of the same species collected in northern Baffin Bay (Fisk *et al.*, 2001b; Buck-

man *et al.*, 2004). Lipid content in the eggs of the three species was similar, allowing comparisons of wet weight concentration data. PCB was the predominant OC in all three species and was similar in black-legged kittiwakes and northern fulmars ( $280 \pm 20$  and  $270 \pm 20$  ng/g ww, respectively) and nearly twice that observed in thick-billed murre ( $130 \pm 10$  ng/g ww). This reflects the higher trophic level of northern fulmars and black-legged kittiwakes and migration to more contaminated habitats by the kittiwakes.  $\Sigma$ DDTs was highest in the northern fulmar ( $210 \pm 20$  ng/g ww), followed by the thick-billed murre ( $100 \pm 10$  ng/g ww) and the black-legged kittiwake ( $60 \pm 10$  ng/g ww), and similar trends were observed for  $\Sigma$ CHLs.

#### Faroe Islands black guillemots

A large study of black guillemots in the Faroe Islands in 1995 and 1996 (Dam, 2000) revealed that the black guillemot had distinctive winter and summer diets, where fish (primarily *Amodytidae* or sandeel type) made up a substantial dietary component in spring and early summer along with *Galathea* spp. crustaceans. In winter, the diet was dominated by *Galathea* spp. and gastropods, in particular the banded chink shell, *Lacuna divericata*. Indications of elevated OC concentrations in liver of adult males and females in spring hints to a possible link to a fish-dominated diet at that time. The single OC occurring in highest concentrations was most often CB153, but the concentration differences between this PCB congener and *p,p'*-DDE were small, and in some samples, nonexistent. In April 1996, the CB153 concentrations in adult females and males were 37 and 42 ng/g ww and *p,p'*-DDE concentrations were 24 and 30 ng/g ww, respectively. In August 1996, the respective CB153 concentrations were 20 ng/g ww and 24 ng/g ww, and *p,p'*-DDE concentrations were 9 ng/g ww and 10 ng/g ww in females and males, respectively.

#### Grey heron and shag from Norway

$\Sigma$ PCB concentrations (34 congeners) were measured in the yolk sac of grey heron (*Ardea cinerea*) hatchlings from two populations in Norway, at Frøya (mid-Norway) and Finnfjordøy (northern Norway) (Jenssen *et al.*, 2001). No statistically significant differences were seen in  $\Sigma$ PCB concentrations between the two populations. Mean PCB concentrations were 2070 (Frøya) and 2450 ng/g ww (Finnfjordøy) with ranges of 570-4520 ng/g ww and 1360-4910 ng/g ww, respectively. Similarly, yolk sacs from newly hatched shag chicks (*Phalacrocorax aristotelis*), from the island of Sklinna on the central Norwegian coast were analyzed for 29 PCB congeners (Murvoll *et al.*, 1999). The mean  $\Sigma$ PCB concentration was 1200 ng/g ww.

#### Alaskan bald eagle eggs

Levels of OCs were measured in the eggs of bald eagles (*Haliaeetus leucocephalus*) from four islands of the Aleutian Archipelago to assess their possible role in low reproductive productivity on one of the islands, Kiska Island (Anthony *et al.*, 1999). Bald eagles are resident on these islands and do not migrate. Samples were collected in 1993 and 1994, and OC pesticides were elevated in eggs from all four islands. No statistically significant differences were seen between the four islands for  $\beta$ -HCH,

dieldrin, HCB or heptachlor epoxide concentrations. *p,p'*-DDE concentrations ranged from 300 to 4100 ng/g ww, but were significantly higher in the eggs collected on Kiska Island (geometric mean of 2750 ng/g ww) as compared to Adak, Tanaga, and Amchitka Islands (geometric means of 750-950 ng/g ww). *Trans*-nonachlor, mirex, oxychlorane, and mercury concentrations followed the same general pattern as DDE, with highest concentrations on Kiska Island. Kiska Island is furthest west, and the authors concluded that the east-west increase in concentrations suggested Asiatic sources of these contaminants. ΣPCB concentrations (as Aroclor 1260) ranged between 100 and 9900 ng/g ww on the four islands and were highest on the three islands (Adak, Kiska, and Amchitka) that had military facilities (geometric means of 2100, 2090, and 1700 ng/g ww, respectively). PCB concentrations on Tanaga Island were much lower (geometric mean of 700 µg/g ww). The Kiska Island bald eagles had a higher percentage of seabirds, such as northern fulmars and glaucous-winged gulls, in their diet, which could explain their higher OC concentrations. This study highlights the potential for local OC contamination from military activities to result in elevated OC concentrations in wildlife.

*PCDD/Fs, mono- and non-ortho PCBs, and toxaphene in seabirds*

Recently, compounds such as PCDD/Fs, non-ortho PCBs and toxaphene were analyzed in thick-billed murre, northern fulmars, and black-legged kittiwakes collected on Prince Leopold Island in Lancaster Sound (Braune *et al.*, 2001a) (Annex Table 16). These compounds have not been measured previously in Canadian Arctic seabirds. Samples included livers from 1975 and 1993 and egg samples from 1993. This study was set up as a pilot study to determine if temporal changes had occurred, and is examined in more detail in Section 5.4.3. PCDD/Fs and non-ortho PCBs were found in all Arctic seabird samples analyzed in this study. In particular, concentrations of HxCDDs, PeCDFs and HxCDFs found in Arctic seabird livers exceeded levels reported for marine mammals in the Canadian Arctic (de March *et al.*, 1998) by several orders of magnitude. Ringed seals, polar bears, and walrus (*Odobenus rosmarus*) in the Canadian Arctic tend to have congener profiles dominated by higher TCDD and lower PeCDD and PeCDF levels (de March *et al.*, 1998), whereas Arctic seabirds show the reverse profile. Concentrations of PCDD/F homologues in the Arctic seabirds are in the range reported for seabirds from temperate North America and Europe (Hebert *et al.*, 1994; van den Berg *et al.*, 1987). Toxaphene was detected in every seabird sample analyzed, except for the pool of kittiwake livers from 1975, and concentrations were one to two orders of magnitude lower than reported for marine mammals from the Canadian Arctic (Muir *et al.*, 1999b).

PCDD/F concentrations in common guillemot eggs from the Faroe Islands sampled in 2000 were 10 pg/g ww (Mikkelsen, 2002). These concentrations were lower than seen in pilot whale blubber from the Faroe Islands, indicating a similar relationship as seen in the Canadian Arctic between seabirds and mammals. However, the difference between the TEQs in pilot whale and guillemot were less pronounced, approximately six times higher on a lipid weight basis (Mikkelsen, 2002).

Yolk sacs from grey heron hatchlings from two sites in Norway (Frøya, Finnfjordøy) had TEQs based on mono-ortho PCBs of 79.4 pg/g ww and 93.2 pg/g ww, respectively (Jenssen *et al.*, 2001). Ranges were 21-179 and 55-177 pg/g ww, respectively. Yolk sacs from shag hatchlings from Sklinna (central Norwegian coast) had TEQs based on mono-ortho PCBs of 44 pg/g ww (Murvoll *et al.*, 1999).

Liver samples from glaucous gulls from Longyearbyen on Svalbard were analyzed for PCBs including non- and mono-ortho PCBs (Daelemans *et al.*, 1992). The TEQ concentration based on these congeners was 2500 pg/g ww. Toxaphene was measured in the liver of eighteen glaucous gulls collected from Bjørnøya and four from Longyearbyen in 1999 (Burkow *et al.*, 2001; Herzke *et al.* 2003). Additionally, toxaphene was analyzed in the intestinal contents of fifteen of the glaucous gulls from Bjørnøya. Toxaphene Parlars 50 and 26 had average concentrations of 10 and 19 ng/g ww, respectively, in liver and were the dominant toxaphene congeners. For glaucous gulls from Bjørnøya, total toxaphene concentrations in gut contents were 84 ng/g ww, in posterior colon contents 53 ng/g ww, and in muscle 175 ng/g ww (Evenset *et al.*, 2002). Levels of toxaphene were up to 100 times lower than PCBs and some of the legacy OC pesticides.

#### 4.4.5.1. 'New' chemicals in seabirds

A number of very recent studies have examined levels of 'new' chemicals in Arctic seabirds, in particular brominated compounds (Annex Table 17).

##### *Canadian seabirds*

A range of 'new' chemicals was measured as part of the PCDD/F and toxaphene analyses in Arctic seabirds described in Section 4.4.5 (Braune *et al.*, 2000; 2001a). These include livers (1975 and 1993) and eggs (1993) from thick-billed murre, northern fulmars, and black-legged kittiwakes collected on Prince Leopold Island in Lancaster Sound. These results are discussed in more detail in the temporal-trends section (Section 5.4.3).

PBDEs were present at low ng/g levels in most of the samples analyzed. The highest estimated PBDE levels (3 ng/g ww or about 60-70 ng/g lw) were detected in the 1993 egg and liver samples of kittiwakes, and the 1993 murre and fulmar samples also contained ng/g levels. BDE47 was the major PBDE congener in all the samples. Although the data suggest that exposure to PBDEs has increased from 1975 to 1993, the highest level reported is an order of magnitude lower than levels reported for Swedish common guillemots from the Baltic Sea in 1993 (720 ng/g lw) (Sellström *et al.*, 1993; 2003; Sellström, 1999), but is still higher than levels reported for Canadian marine mammals (Stern and Addison, 1999; Stern and Ikonou, 2000; 2001). However, PBDE concentrations in Baltic Sea guillemots have declined since their peak in the mid-1980s and were lower (143 ng/g lw) in 1999 (Sellström *et al.*, 2003). Temporal trends of PBDEs in marine mammals indicate increasing levels in the Canadian Arctic during the period of decline in the Baltic Sea (see Section 5.4.6.1), implying that concentrations in Canadian Arctic seabirds may be approaching those of guillemot in the Baltic Sea.



Halogenated dimethyl bipyrrroles (HDBPs), which are naturally-occurring, mixed bromine/chlorine persistent organic compounds, have been shown to bioaccumulate in seabird eggs (Tittlemier *et al.*, 1999). HDBPs were detected in all of the 1993 samples and only one of the liver samples from 1975. The major contaminant found was 1,1'-dimethyl-3,3',4,4'-tetrabromo-5,5'-dichloro-2,2'-bipyrrrole (DBP-Br<sub>4</sub>Cl<sub>2</sub>). The highest estimated level (a total of 5 ng/g ww) was found in the kittiwake egg sample from 1993. Trace amounts (<2 ng/g ww) of chlorinated terphenyls, hexabrominated biphenyls, and tris(4-chlorophenyl)methane (TCPMe) were also detected in most of the Arctic seabird samples. No chlorinated diphenyl ethers, TCPMe or PCNs were detected in any of these samples (detection limit approximately 2 ng/g ww).

In a separate study, Tittlemier *et al.* (2001) examined levels of HDBPs in four seabird species (dovekie, black guillemot, black-legged kittiwake, and glaucous gull) collected in northern Baffin Bay. This is significant because it represents the first time a naturally produced halogenated compound has been measured in Arctic species. This was part of a larger study on the food web dynamics of these chemicals in marine food webs (Section 4.4.9). HDBPs were detected in all of the samples studied from the Northwater Polynya. Levels of DBP-Br<sub>4</sub>Cl<sub>2</sub> in the seabirds were generally similar to those recorded in seabird samples from Atlantic Canada (Tittlemier *et al.*, 1999). A comparison cannot be made for the other congeners measured since this study contains the first reported concentration data for these congeners. Little auks provide the single clear exception where DBP-Br<sub>4</sub>Cl<sub>2</sub> concentrations (1.76 ng/g ww) were approximately ten times lower than those of the Atlantic puffin (*Fratercula artica*) (20 ng/g ww), a species that has a similar winter habitat but a more piscivorous diet. The difference in concentrations is likely driven by the little auks feeding at a lower trophic position just prior to their collection in early summer (Fisk *et al.*, 2001b). In the majority of the bird species, DBP-Br<sub>4</sub>Cl<sub>2</sub> was the predominant congener. The exception occurred with black guillemots, where DBP-Br<sub>6</sub> was the most abundant congener. This may be due to black guillemots feeding on benthic organisms during part of the year (Gaston and Jones 1998).

#### *Bjørnøya seabirds*

Brominated flame retardants were measured in the liver and intestinal contents of fifteen glaucous gulls collected on Bjørnøya in 1999 (Burkow *et al.*, 2001; Herzke *et al.*, 2003). Only two PBDE congeners were detected, BDEs 47 and 99, at concentrations between 2 and 25 ng/g ww (27-450 ng/g lw). Analysis of the samples using gas chromatography/high resolution mass spectroscopy revealed a number of other PBDE and polybrominated biphenyl (PBB) congeners. BDE<sub>47</sub> dominates the PBDE congener pattern in seabirds. ΣPBDE levels in most species are approximately 20 times lower than ΣPCB levels, but are up to 1000 times lower for glaucous gulls. For most glaucous gulls, these concentrations are similar to what has been measured in seabird eggs from the Canadian Arctic (Braune, 2000). However, a few individual glaucous gulls had higher PBDE levels (480-560 ng/g lw), which were higher than levels seen in Baltic Sea

guillemots for the same year. Levels of PBDE were up to 100 times lower than levels of PCBs and some of the legacy OC pesticides.

Butyltins have recently been measured in glaucous gulls from Bjørnøya (Table 4-8, page 111). Liver samples were found to contain the TBT metabolites, DBT (detection limit to 51 ng/g ww), and MBT (detection limit to 14 ng/g ww), but no TBT (Berge *et al.*, 2002).

#### *Greenland seabirds*

ΣPBDE levels were determined in the liver of thick-billed murre and black guillemot collected from southwestern Greenland in 1999 (Muir and Johansen, 2001). Concentrations of ΣPBDE in the thick-billed murre (1.7±1.6 ng/g ww; 32 ng/g lw) and guillemot (3.0±2.8 ng/g ww; 46 ng/g lw) were similar to levels observed in Canadian Arctic seabird eggs but lower than observed in glaucous gulls from Bjørnøya.

#### *Northern Norway*

New data on dioxin-like substances, PCNs, PBDEs, and toxaphene in herring (*Larus argentinus*) and great black-backed gull eggs from northern Norway were recently produced (Gabrielsen, 2002). Eggs were collected in 2001 from four sites: Alta, Kongsfjord (Finmark), Sommarøy, and Vardø. Twenty eggs from each site were pooled into one sample. Concentrations of PCNs, PBDEs, and toxaphene and total TEQs were similar between regions, although levels were somewhat lower in Alta (Annex Tables 15, 16 and 17). TEQ values in these gull eggs are similar to levels measured in Arctic Canadian seabird eggs (Braune, 2001). PBDE and toxaphene levels in these seabirds are slightly higher than measured in Bjørnøya glaucous gulls (Burkow *et al.*, 2001; Herzke *et al.* 2003).

#### *Chiral pesticides*

Chiral pollutants exist as two mirror-image forms or optical isomers called enantiomers. Enantiomers have identical physical-chemical properties and abiotic degradation rates, but can have different rates of biotransformation (Buser and Müller, 1992). The chemical manufacturing process results in a mixture containing approximately fifty percent of each chiral compound, called a racemic mixture. Selective biotransformation of one chiral component over another can occur and result in an enantiomeric enrichment (Buser and Müller, 1992). This selective enrichment originates from one enantiomer being more easily biotransformed. The resulting selective accumulation of a single enantiomer can provide information on fate and dynamics of the chemical and may have significant toxicological ramifications. It has been proposed that comparison of enantiomer ratios (ERs) or enantiomer fractions (EFs) may provide information on biotransformation capacity of species and the trophic transfer of contaminants in a food web (Wiberg *et al.*, 2000).

Enantiomeric fractions of chiral chlordane components were examined in the liver and fat of seven Arctic seabird species collected in northern Baffin Bay in 1998 (Fisk *et al.*, 2001b). EFs of chiral components failed to predict concentration or trophic level, but did identify biotransformation differences between species and chlordane components. The relative proportions of chlordane components in seabirds were related to taxonomy and



the magnitude of EF values; the northern fulmar and gulls (black-legged kittiwake, ivory gull, and glaucous gull) had a greater percentage and higher EFs for oxychlordanes than the alcids (little auk and black guillemot). The exception was the thick-billed murre, an alcid, where oxychlordanes made up a significant percentage of its  $\Sigma$ CHLs. Thick-billed murre appeared to have a greater capacity to metabolize and eliminate chlordanes, based on high proportions of oxychlordanes and the highest EFs for oxychlordanes and heptachlor epoxide.

#### 4.4.6. Marine mammals

There is an extensive database on OCs in Arctic marine mammals. This includes data that were produced before 1997 and summarized in the first AMAP POPs assessment and further studies since that time. The focus on marine mammals comes in part because these animals, in particular seals and whales, are important components of the diet of many Arctic societies. Marine mammals also occupy a range of trophic positions, and a number of species (e.g., polar bears) are apex predators, and thus have high levels of OCs. Therefore, more than for any other group of animals, is concern about OCs greatest in marine mammals, both with respect to effects on the animals themselves and transfer to humans.

All relevant data on OCs and organotin compounds in marine mammals that have become available since the previous AMAP assessment (de March *et al.*, 1998) are reviewed here. It must be emphasized that the spatial trends of persistent OCs discussed below in this section are often qualitative because they are based on evaluations of means and ranges of concentrations from different studies. In the case of the sums of groups of compounds, such as PCBs and chlordanes, some laboratories have included more congeners or components than others in  $\Sigma$ PCB and  $\Sigma$ CHL results. Rigorous comparisons between locations also require information on age, sex, blubber thickness, nutritional status, collection season, and reproductive status of the animals, all of which can have an important influence on contaminant concentrations. This information has been collected for many locations, but used only qualitatively in this assessment of spatial trends between studies.

A considerable body of data has been collected on pinnipeds and cetaceans (both mysticetes and odontocetes) since the previous AMAP POPs assessment (de March *et al.*, 1998). All relevant data that have become available since the previous AMAP assessment are included here for pinnipeds (Annex Table 12) and cetaceans (Annex Table 13). Data for individual toxaphene congeners are reported in Annex Table 15, while data for individual PBDEs are reported in Annex Table 17, and data for PCDD/Fs, non-*ortho* PCBs (nPCBs) and toxic equivalents (TEQs) are reported in Annex Table 16. Although the current assessment primarily includes data and publications since 1996, some reports included here do contain data collected prior to this time, if they were not considered in the previous AMAP assessment.

As in the previous AMAP assessment, by far the most frequently determined OC compounds in marine mammals are still PCBs (as congeners) and DDT-related compounds. Chlordane-related compounds, HCHs and HCB, are somewhat less well represented, while other chloro-

benzenes, toxaphene components, dieldrin, mirex, and PBDEs are not well represented in overall contaminant measurements in marine mammals across the Arctic. PCDD/Fs have also only been quantified to a limited extent across marine mammal species or geographical locations in the Arctic (Annex Table 16).

##### 4.4.6.1. Pinnipeds

###### Ringed seals

Ringed seals are the most abundant and widely distributed resident Arctic pinniped. Their diet consists of fish, mainly schooling gadids, and crustaceans (amphipods, mysids, and euphausiids). They have a broad circumpolar distribution and prefer annual, landfast ice, but are also found in multi-year ice. No clear-cut boundaries are known to separate ringed seal stocks in marine waters. Adults are believed to be relatively sedentary, but sub-adults sometimes disperse over long distances (Reeves, 1998). Ringed seals are a key component of the diet of polar bears and the Inuit of Canada and Greenland.

Results of studies completed since the previous AMAP POPs assessment are in general agreement with geographical trends reported previously for ringed seals (de March *et al.*, 1998). That is,  $\Sigma$ PCB and  $\Sigma$ DDT concentrations increase from west to east, with levels being lowest in ringed seals from the Chukotka Peninsula (Lavrentiya) (RAIPON/AMAP/GEF Project, 2001) and Alaska (Hoekstra *et al.*, 2003a; Kucklick and Krahn, 2002), intermediate in ringed seals from the eastern Canadian Arctic (Muir *et al.*, 2000b; Fisk *et al.*, 2002d), higher in seals from Svalbard and the White Sea (Wolkers *et al.*, 1998b; Kostamo *et al.*, 2000; Muir *et al.*, 2002b), and highest in seals from the Kara Sea near Dikson (Nakata *et al.*, 1998a) (Figure 4-45). Between Chu-

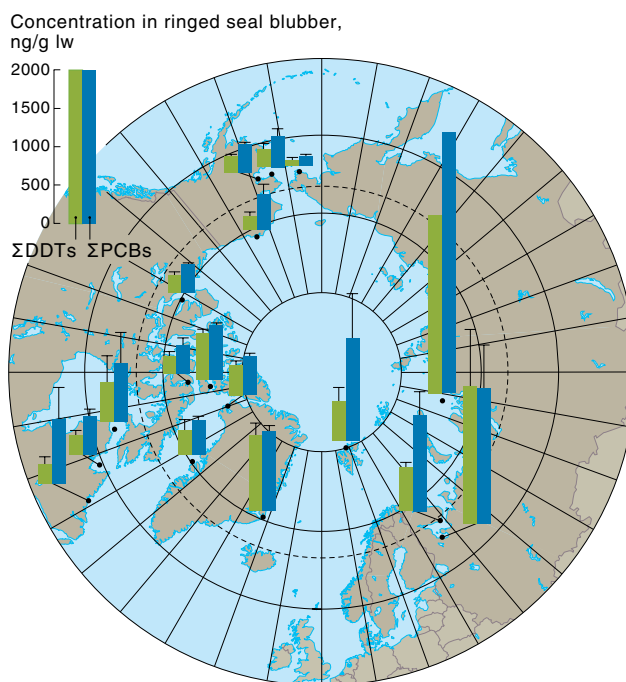


Figure 4-45. Mean ( $\pm$  95% CI) concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs in female ringed seal blubber (Krahn *et al.*, 1997; Muir *et al.*, 1999c; 2001c; 2002c; Muir and Johansen, 2001; Denmark, 2002; Fisk *et al.*, 2002c; Hoekstra, 2003a; Kucklick and Krahn, 2002; Nyman *et al.*, 2002; Nakata *et al.*, 1998a; RAIPON/AMAP/GEF Project, 2001).

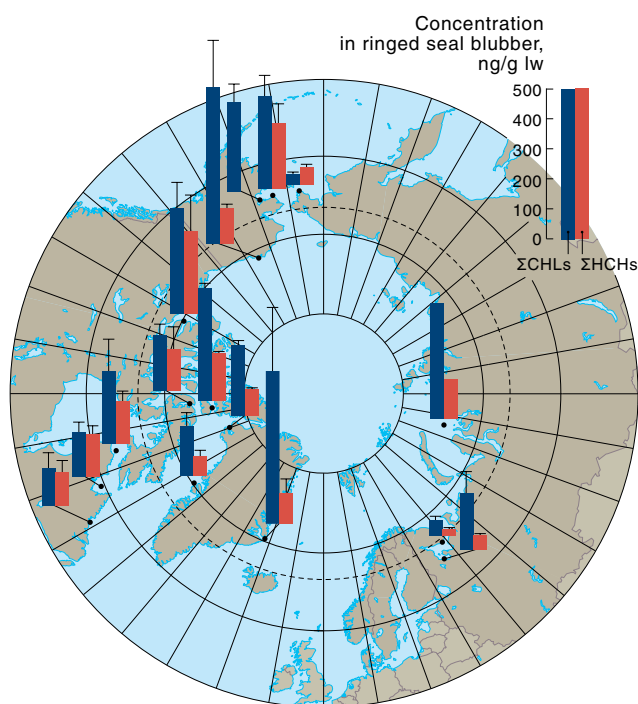


Figure 4-46. Mean ( $\pm$  95% CI) concentrations of  $\Sigma$ CHLs and  $\Sigma$ HCHs in female ringed seal blubber (Krahn *et al.*, 1997; Muir *et al.*, 1999c; 2001c; 2003; Muir and Johansen, 2001; Denmark, 2002; Fisk *et al.*, 2002c; Hoekstra, 2003a; Kucklick and Krahn, 2002; Nyman *et al.*, 2002; Nakata *et al.*, 1998a; RAIPON/AMAP/GEF Project, 2001).

katka and the Kara Sea, there are no data for seals in the Russian Arctic, representing a large geographical gap. Levels of  $\Sigma$ CHLs are fairly consistent across the Arctic, while  $\Sigma$ HCH concentrations are highest in Alaska and decrease from west to east (Figure 4-46).

Within Alaska, Kucklick and Krahn (2002) reported that  $\Sigma$ PCB and OC pesticide concentrations measured in ringed seals collected between 1988 and 1998 generally decreased from Barrow to the more westerly site at Nome (Annex Table 12). After adjusting for the effects of age and blubber thickness, females from Barrow were found to have higher concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs, and dieldrin, while in males, only  $\Sigma$ HCH concentrations were higher in seals from Barrow. Only HCB and mirex were not significantly different between the two locations for either sex. There was general agreement amongst studies between  $\Sigma$ PCB and OC pesticide levels observed in Barrow ringed seals (Hoekstra *et al.*, 2003a; Kucklick and Krahn, 2002), and these levels were comparable to those measured recently in ringed seals from the western Canadian Arctic (de March *et al.*, 1998; Muir *et al.*, 1999b) (Annex Table 12). For example, the geometric mean of  $\Sigma$ DDTs in female seals from Resolute was 340 ng/g ww (range of 163-540 ng/g) (Muir *et al.*, 2000b) versus 408 ng/g ww in the Barrow seals (range of 274-652 ng/g) and 178 ng/g ww (range of 143-387 ng/g) in the seals from Nome.

Further east, ringed seals from Holman, NWT, (Hoekstra *et al.*, 2003a) had levels of PCBs and OC pesticides that were in general agreement with, and intermediate between, levels found in ringed seals from further west (Alaska) (Hoekstra *et al.*, 2003a; Kucklick and Krahn, 2002) and further east (eastern Canadian Arctic) (Muir *et al.*, 2000b; Fisk *et al.*, 2002d). A variety of PCB and

OC pesticides were quantified in the blubber of ringed seals from seven sites within Nunavik and Labrador, in 1998 and 1999 (Muir *et al.*, 1999c; 2000c) (Annex Table 12). For comparison among regions, three sites within Ungava Bay were combined (Kuujjuaq, Kangirsuk and Kangiqsualujjuaq), as were two sites in the Hudson Strait (Salluit and Quaqtaq). The two sites in Labrador (Nain and Makkovik) were treated separately. Average blubber concentrations of  $\Sigma$ PCBs, the major OC contaminants in seals from these four locations, ranged from 572 to 1042 ng/g ww in males, and from 512 to 730 ng/g ww in females (Muir *et al.*, 2000c). The highest levels in males were found in samples from the more westerly locations of Ungava Bay and Hudson Strait, while the highest average levels in females occurred in samples collected at Nain. DDT-related compounds were also prominent contaminants in ringed seal blubber, with average concentrations ranging from 198 to 884 ng/g ww in males. Statistical analysis did not reveal any significant differences in concentrations of  $\Sigma$ PCBs or  $\Sigma$ DDTs among the four locations for females. For males,  $\Sigma$ PCB,  $\Sigma$ DDT, and  $\Sigma$ HCH concentrations were significantly higher in samples from Ungava Bay, after adjusting for age. There was a significant interaction, however, between age and location (i.e. a differing relationship of age with concentrations of the OCs) for males, which may have affected the results. In general, levels of  $\Sigma$ PCBs and  $\Sigma$ DDTs in ringed seal blubber from this study were comparable to those found in ringed seal blubber at other eastern Canadian Arctic locations (Weis and Muir, 1997; Muir, 1998; Muir *et al.*, 1999b), including those from the more northerly region of the Northwater Polynya in northern Baffin Bay (Fisk *et al.*, 2002d). Results for Fisk *et al.* (2002d) were also in the range reported for ringed seals from western-central and northeastern Greenland (Muir and Johansen, 2001; Denmark, 2002) (Annex Table 12). After removing the influence of age, sex, and blubber thickness,  $\Sigma$ PCB and OC pesticide concentrations did not differ in ringed seals from the east and west side of the Northwater Polynya, likely due to the relatively small distance between these two sites (Fisk *et al.*, 2002d). Concentrations of  $\Sigma$ DDTs and  $\Sigma$ PCBs in seals from this region were lower than those reported for this species in Svalbard, Norway (Wolkers *et al.*, 1998b; Kleivane *et al.*, 2000; Severinsen *et al.*, 2000) (Annex Table 12). In contrast,  $\Sigma$ HCH concentrations in Canadian and Alaskan ringed seals were higher than those reported for ringed seals from the European Arctic. These geographical trends are consistent with past compilations of circumpolar data for ringed seals (Muir *et al.*, 2000b) and polar bears (Norstrom *et al.*, 1998).

Levels of  $\Sigma$ PCBs found in Svalbard ringed seals (Wolkers *et al.*, 1998b; Kleivane *et al.*, 2000; Severinsen *et al.*, 2000; Nyman *et al.*, 2002) were in good agreement, and were at least twice as high as those in seals from the Canadian Arctic (Annex Table 12), despite the smaller number of congeners included in  $\Sigma$ PCBs for the Svalbard seals. The predominant PCB congeners in the Kongsfjorden (Svalbard) ringed seals were 153, 138, 99, 180 and 101 (Wolkers *et al.*, 1998b). The observed PCB patterns were very similar to patterns in seals from other studies (Muir *et al.*, 1988; Luckas *et al.*, 1990; Beck *et al.*, 1994), suggesting a similar biotransformation capacity.

Further east, concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs, and HCB, quantified in the blubber of ringed seals from the White Sea (Kostamo *et al.*, 2000; Muir *et al.*, 2003) (Annex Table 12), were comparable to or higher than in ringed seals from Svalbard, but lower than reported for this species in the more easterly Kara Sea (Nakata *et al.*, 1998a). Within the White Sea, Muir *et al.* (2003) found higher levels in seals sampled at Dvina Bay, near the city of Archanglesk, in comparison to those from the more northerly and less industrialized region of Gorlo Basin (Annex Table 12). The high levels in the White Sea ringed seals are likely influenced by local point sources, as some parts of the White Sea are heavily industrialized. Levels in White Sea ringed seals were also much lower than levels in two freshwater sub-species of ringed seals living in Lake Saimaa, Finland, (*P. b. saimensis*) and Lake Ladoga, Russia, (*P. b. ladogensis*), (Kostamo *et al.*, 2000). Kostamo *et al.* (2000) found that the PCB congener composition in blubber from the White Sea seals resembled a mixture of Aroclor 1254 and 1260. They also reported that the  $\Sigma$ DDT: $\Sigma$ PCB ratio in the White Sea ringed seals ranged from 0.96 to 1.49 (Kostamo *et al.*, 2000), which is similar to the results from ringed seals in the east-central Canadian Arctic (Muir *et al.*, 1988). The higher levels found in the Kara Sea seals may be due to OC inputs from the Ob and Yenisey Rivers, which have been shown to be major sources of OCs to the Arctic Ocean via river water and sediments (de March *et al.*, 1998) (Section 4.3.1.1).

Total toxaphene levels in ringed seal blubber were generally lower, moving eastward from Barrow, Alaska, (mid- to high hundreds of ng/g lw) to the Hudson Strait and Ungava Bay (Canada) (low hundreds of ng/g), Greenland and the Barents Sea (tens of ng/g), and then slightly higher (low hundreds of ng/g) in White Sea ringed seals (Annex Table 12) (Wolkers *et al.*, 1998b; Muir and Johansen, 2001; Denmark, 2002; Hoekstra *et al.*, 2003a; Muir *et al.*, 2003). On a congener basis, Parlar 26 levels were highest in ringed seals from Hudson Strait and Ungava Bay in the Canadian Arctic, followed by those in seals from Alaska and the White Sea and then Svalbard, with the lowest levels in seals from western Greenland. Levels of Parlar 50 were highest in seals from Svalbard and Hudson Strait and lower in seals from Ungava Bay and western Greenland (Annex Table 15; Figure 4.47).

In ringed seal blubber from northern Quebec, octa- and nonachlorobornanes were the major homologue groups (Muir *et al.*, 2000d) (Annex Table 15). Toxaphene Parlars 39, 40, and 42 were also identified in the blubber of ringed seals from Arviat, Nunavut (formerly in the NWT) (Loewen *et al.*, 1998), marking the first time that Parlar 42, the most toxic congener in technical toxaphene, has been found in any significant concentrations in a marine mammal. Ringed seals from Kongsfjorden, Svalbard, (Wolkers *et al.*, 1998b) showed no effect of sex, age or total blubber on toxaphene levels. Slightly higher concentrations of toxaphene Parlars 26, 50, and 62 were found in male than female ringed seals from Kongsfjorden (Føreid *et al.*, 2000).

Blubber from ringed seals collected in 1993 near Pangnirtung, Nunavut, were analyzed for non- and mono-*ortho* PCBs (Helm *et al.*, 2002). Total non- and mono-*ortho* PCB concentrations (sum of CBs 77, 81,

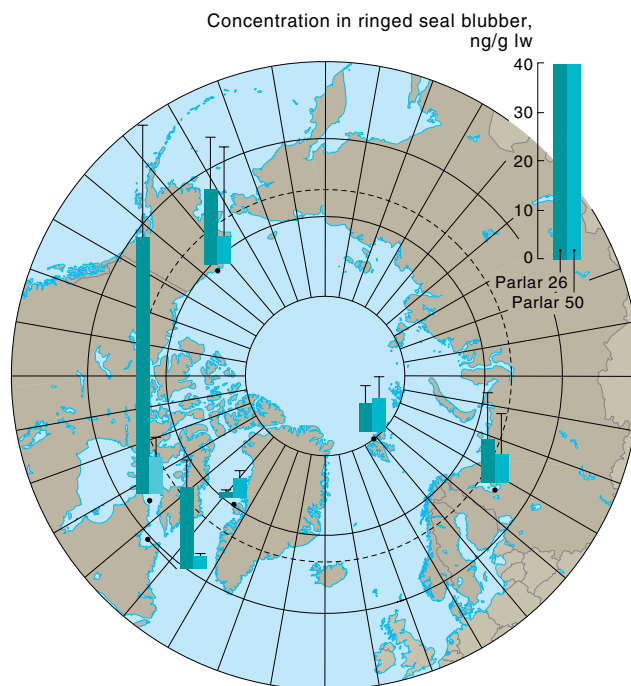


Figure 4.47. Mean ( $\pm$  95% CI) concentrations of toxaphene Parlar 26 and Parlar 50 in female ringed seal blubber (Wolkers *et al.*, 1998b; Muir *et al.*, 1999c; 2003; Hoekstra, 2003a).

105, 126, 118, 114, 156, and 169) ranged from 14.9 to 32.7 ng/g ww, with congeners 118, 105 and 156 being the most predominant. Mean ( $\pm$  standard deviation) levels of these were lower in female ( $15.4 \pm 0.8$  ng/g ww) than male ( $26.7 \pm 8.0$  ng/g ww) ringed seals. TEQs for non- and mono-*ortho* PCBs in males and females were  $0.624 \pm 0.144$  pg/g lw and  $0.508 \pm 0.088$  pg/g lw, respectively, most of which was accounted for by congener 126 (Helm *et al.*, 2002) (Annex Table 16). For ringed seals from Holman, TEQs based on non- and mono-*ortho* PCBs in blubber from 2000 ranged from 4.3 to 91 pg/g ww, with the PCBs contributing most to the TEQs (Ikonomou, 2002). Ringed seals from the Kara Sea had total non- and mono-*ortho* PCB concentrations (sum of CBs 77, 105, 118, 126, 156, and 169) of approximately 670 ng/g ww in blubber (Nakata *et al.*, 1998a). The TEQ for these CB congeners was approximately 144 pg/g ww (160 pg/g lw), and CB126 contributed most, followed by CB118. Blubber from ringed seals and spotted seals (*Phoca largha*) collected from the Chukotka Peninsula (Lavrentiya) area were analyzed for PCDD/Fs (RAIPON/AMAP/GEF Project, 2001). TEQs were 1.1 pg/g ww for ringed seals and 1.3 pg/g ww for spotted seals.

#### Harp seals

Harp seals are an Atlantic species, inhabiting Arctic and subarctic waters. They feed primarily on small marine fish and, secondarily, on crustacean macroplankton. Three separate harp seal populations undergo annual migrations between southerly breeding sites and their northern feeding grounds, both at the edge of the pack ice. Harp seals are not a common dietary item of northern human populations.

Little information has become available on contaminants in harp seals since the previous AMAP assessment. However, recent studies are available for seals



from Svalbard and the White Sea. Data are available for PCBs (Wolkers *et al.*, 1999), DDTs, HCB, HCHs, as well as cyclodiene pesticides such as dieldrin, endrin, and the chlordanes (Wolkers *et al.*, 2000) (Annex Table 12). The levels of PCBs and most pesticides, especially the DDTs, for sub-adult ( $\leq 4$  years) harp seals from the ice edge, east of Svalbard, were relatively low compared to those reported in other harp seal studies. This was probably, in part, because animals were sampled during the summer, when the total blubber content is high compared to the spring (Wolkers *et al.*, 1999; 2000). In contrast to this, levels of PCBs and OC pesticides quantified in older adult males from the same area (Kleivane *et al.*, 2000) were substantially higher than previously reported in this species (Oehme *et al.*, 1995c; Kleivane *et al.*, 1997).

Levels of PCBs and OC pesticides were quantified in harp seals from the southern ice area located north of the White Sea in 1993 and were also elevated, but to a lesser extent than in the mature Svalbard males. All animals in the Svalbard group were lean specimens in late molt, while those from the White Sea were sampled early in the molt. The Svalbard seals had significantly thinner blubber than the seals sampled outside of the White Sea (Kleivane *et al.*, 2000). Thus, the dramatic decrease in the fat reservoir in adult seals during the molting period, when feeding is at a minimum or non-existent, clearly resulted in a significant concentration of PCBs, DDTs, and chlordanes in the remaining blubber of these seals. To the south, blubber concentrations of PCBs, DDTs, chlordanes, and HCB in harp seals from the Gorlo Basin of the White Sea in 1998 (Muir *et al.*, 2003) were comparable to those reported for harp seals sampled east of Svalbard in 1997 (Wolkers *et al.*, 1999; 2000) and were on the low side of the range of levels reported for this species in the Kleivane *et al.* (2000) study. Mean POP levels in young harp seals sampled in 1992 (Muir *et al.*, 2003) were generally higher compared to those sampled in 1998, and were comparable to those reported for seals north of the White Sea in 1993 (Kleivane *et al.*, 2000). It is not clear if this is an indication of declining levels with time or variability in biological parameters, such as blubber thickness.

Toxaphene levels in juvenile harp seals from eastern Svalbard in 1997 (Wolkers *et al.*, 2000) were comparable to those found in adult females and juveniles from the White Sea in 1998 (Muir *et al.*, 2003). Interestingly, in contrast to results from seals in other areas (Muir *et al.*, 1992a; Wolkers *et al.*, 1998b), toxaphenes were the predominant compounds in young harp seals from eastern Svalbard, exceeding PCB concentrations. Toxaphene concentrations in these young harp seals (Wolkers *et al.*, 2000) were also more than 20 times higher than in juvenile ringed seals from the west coast of Svalbard (Wolkers *et al.*, 1998b). Although there may be species-specific differences, the high toxaphene levels found in the seals from the east coast of Svalbard (Barents Sea) as compared to the seals from the west coast of Svalbard, may indicate that, in spite of a ban on the use and production of these compounds in the western world (Voldner and Li, 1993), the Barents Sea area is continuously exposed to toxaphenes. Thus, in addition to atmospheric long-range transport, another source of toxaphenes may contribute to the high levels found. Furthermore, a study of

harp and ringed seals in the White Sea has demonstrated that levels of toxaphene are higher there than in west Svalbard or Greenland (Savinova *et al.*, 2000b). Studies of sediments in the Kola Bay area have demonstrated that elevated toxaphene is present in sediments in Polamy, a harbor north of Murmansk (Savinova *et al.*, 2000a), possibly associated with past use for insect control on ships.

#### Harbour and grey seals

Harbour seals are one of the most broadly distributed pinnipeds, inhabiting temperate, subarctic, and in some cases Arctic waters, in the north Atlantic and north Pacific. They are a relatively sedentary species that feed opportunistically on a wide variety of fish, cephalopods, and crustaceans (Bigg, 1981). Grey seals are a more mobile pinniped that occur in Atlantic Canada, around Iceland, the Faroe Islands, Great Britain, Norway, the Kola Peninsula, and the Baltic Sea (Bonner, 1981). They consume a wide variety of fish, as well as some crustaceans and cephalopods, and generally feed further offshore and in deeper waters than do harbour seals.

Blubber concentrations of PCBs, DDTs, chlordanes, dieldrin, and HCB in harbour seals from Prince William Sound (Alaska) in 1993 (Krahn *et al.*, 1997) (Annex Table 12) were similar to corresponding data for this species from the same region in 1989-1990 (Varanasi *et al.*, 1993). Their levels of PCBs and DDTs were about twice those of bearded (*Erignathus barbatus*) and ringed seals from the more northerly region of Norton Sound in the Bering Sea (Alaska) (Krahn *et al.*, 1997). These differences may be explained by the higher trophic position occupied by harbour seals compared to ringed or bearded seals, or by the greater anthropogenic activity in Prince William Sound compared to Norton Sound.

Ruus *et al.* (1999) quantified  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs,  $\Sigma$ HCHs, and HCB in harbor and grey seals from Jarfjord, northern Norway (Annex Table 12). Levels of  $\Sigma$ PCBs,  $\Sigma$ DDTs, and  $\Sigma$ CHL levels were up to an order of magnitude higher than in harbour seals from Alaska, while HCB levels were comparable between the two regions (Krahn *et al.*, 1997). Interspecies differences in levels of  $\Sigma$ HCHs and HCB, as well as PCB congener patterns (e.g., CBs 101, 118, 153, and 180) between Norwegian harbor and grey seals, may be explained by different dietary preferences and metabolic capacities.  $\alpha$ -HCH was the most abundant of the HCH isomers in both harbor and grey seals from Jarfjord.

Grey seals sampled in 1993-1995 at the Faroe Islands, in connection with a study of their summer diet (Mikkelsen, 1998), were analyzed for OCs in pooled samples initially, and later separately in 30 individuals (Larsen and Dam, 1999; Dam, 2001). The pooled samples were composed of a total of 45 individuals: the pools were composed of adult males ( $n=4$ , age  $>8$  yr); adult females ( $n=20$ , pregnant individuals); and, juveniles ( $n=21$ , 15 females and six males, age  $>2$  yr and  $<4$  yr). As in the Jarfjord study,  $\alpha$ -HCH was the dominant HCH isomer, but was pronounced only in the group of juveniles. In the adult female group, the HCH isomers were found in similar or slightly decreasing concentrations in the following order:  $\gamma$ -HCH  $\geq$   $\alpha$ -HCH  $\geq$   $\beta$ -HCH. The single OC occurring in the highest concentration among the adults was CB153, followed by  $p,p'$ -



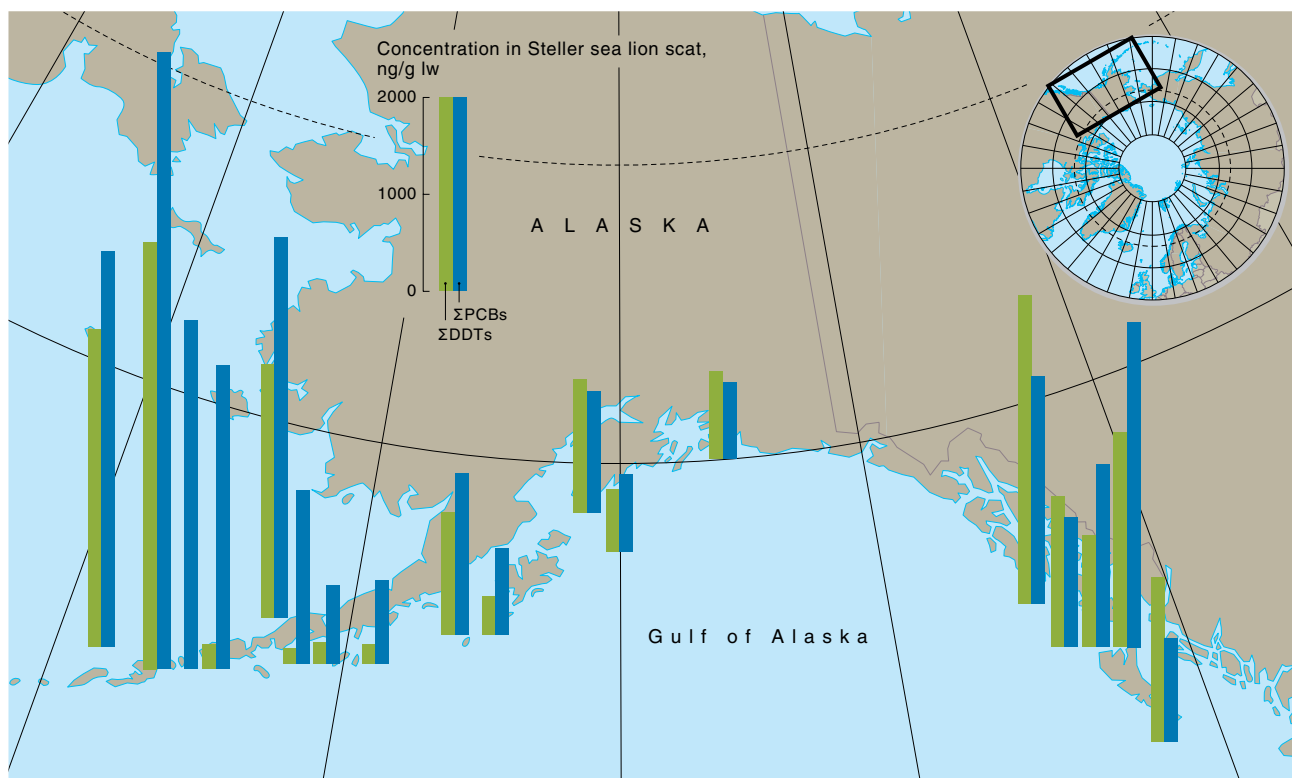


Figure 4-48. Concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs in Steller sea lion scat composites collected from rookeries or haulouts in 1998 and 2000 (Beckmen, 2002).

DDE and CB180. CB153 concentrations in blubber were 4900 ng/g ww and 540 ng/g ww in males and females, respectively. In the juveniles, this sequence was different with  $p,p'$ -DDE > CB153 > CB 138+163, with CB153 at 1200 ng/g ww in blubber. The analyses of the 30 individuals gave the 'typical' pattern of decreasing OC level in females with age and a parallel increase in males. The results further revealed that in the female seals there were significant correlations between toxaphene (as Parlar 50 concentrations) and CB 153 but not among the males. There were, however, highly significant correlations between CB 153 and  $p,p'$ -DDE and *trans*-nonachlor in both sexes.

#### Bearded seals

Bearded seals from Norton Sound and Barrow, Alaska had relatively low OC concentrations in blubber compared to other pinniped species from that region and in the broader Arctic context (Krahn *et al.*, 1997; Hoekstra *et al.*, 2003a) (Annex Table 12). This may be primarily because bearded seals remain in the northern Alaskan seas, a region with limited anthropogenic activity, and consume lower trophic-level prey, including a wide variety of invertebrates, such as decapod crustaceans and mollusks (Kelly, 1988; Krahn *et al.*, 1997). Blood samples from bearded seals from Svalbard had comparable OC concentrations on a lipid weight basis to the Alaskan seals, except for HCH, but on the Chukotka Peninsula (Lavrentiya), blubber samples had generally lower OC concentrations compared to Alaska and Svalbard (RAIPON/GEF/AMAP Project, 2001). In contrast to bearded seals from Alaska, Chukotka, and Svalbard, those from the White Sea (Muir *et al.*, 2003) had very high  $\Sigma$ PCB and  $\Sigma$ DDT levels (approximately 3000 ng/g

ww), which is an order of magnitude higher than the other regions.  $\Sigma$ CHL and mirex concentrations were similar in seals from all regions. HCB and HCH concentrations were highest in seals from Barrow, Alaska. HCB levels were comparable in seals from Chukotka, Svalbard, White Sea, and Norton Sound (Annex Table 12). Toxaphene levels in White Sea bearded seals were comparable to those in ringed and harp seals from the same region, as well as levels in northwestern Greenland walrus, Barents Sea harp seals, and ringed seals from Barrow, Alaska (Muir *et al.*, 2000d; 2003; Wolkers *et al.*, 2000; Hoekstra *et al.*, 2003a). The high levels in the White Sea bearded seals are an example of the effects of point sources in the Arctic, as some parts of the White Sea are heavily industrialized. Blubber from bearded seals collected from the Chukotka Peninsula (Lavrentiya) area was analyzed for PCDD/Fs; the TEQ was 0.97 pg/g ww (RAIPON/AMAP/GEF Project, 2001) (Annex Table 16).

#### Steller sea lions

Based on samples from eighteen rookeries in 1998 and 2000, Beckmen (2002) determined that Steller sea lions sampled from the eastern Aleutian Islands (part of the endangered western Alaskan stock) excrete significantly higher levels of PCBs and DDTs in their feces (mainly adult females) compared to Steller sea lions in southeast Alaska (Figure 4-48). Levels of POPs in blubber biopsies were not significantly different between pups and juveniles in southeast Alaska and Prince William Sound in the Gulf of Alaska (Annex Table 12). Unfortunately, no blubber biopsies were available from the eastern Aleutian Islands or further west in the range. These findings indicate that PCBs are present in the food web ex-

exploited by Steller sea lions in Alaska, at least as far west as the eastern Aleutian Islands. In southeast Alaska where  $\Sigma$ PCB levels were intermediate, the population has increased in recent years. The relatively high levels for the eastern Aleutian Islands, in comparison to the nearby Gulf of Alaska, are of interest, as they suggest either a local source or a strong influence from the Bering Sea.

#### Northern fur seals

Breeding rookeries for more than 72% of the world's population of northern fur seals are located on the two largest Pribilof Islands, St. Paul and St. George (Alaska) in the Bering Sea (Loughlin *et al.*, 1994). A preliminary study demonstrated that young northern fur seal dams (presumably primiparous) in the early post-partum period on St. George and St. Paul Islands, had significantly higher OC levels in their milk and blood than old (multiparous) dams (Beckmen *et al.*, 1999). Higher milk OC exposure to the suckling pups was correlated with significantly higher OC levels in blood of the neonatal pups (Beckmen *et al.*, 1999). In 1996, 50 perinatal pups were captured for blood sample collection (during the ten-day perinatal period and referred to as neonates), and 43 were re-sampled 29 to 51 days later ('pups'). Mean blood OC levels were higher in neonates than at recapture, and again, neonates of young dams had higher mean blood OC levels than neonates of older dams (Annex Table 12). The traditionally harvested and consumed subadult males have lower concentrations of PCBs and DDTs than do adult females or pups (Annex Table 12). PCB and DDT concentrations in subadult males from another study (Krahn *et al.*, 1997) were comparable to levels reported by Beckmen *et al.* (1999). OC concentrations are lowest in fetal blubber but data were only available from two individuals. Pups showed some remarkably high OC concentrations in blubber, especially *p,p'*-DDE, but there is a large variance in blubber lipid content due to nutritional status.

PCB, DDT, chlordane, HCB, and dieldrin levels in subadult northern fur seals from St. Paul Island were comparable to, or higher than, concentrations in ringed and bearded seals from the Bering Sea and harbour seals from Prince William Sound (Gulf of Alaska) (Krahn *et al.*, 1997). This can be partially explained by the fur seals' consumption of higher trophic-level prey such as pollock, herring, and squid (Wynne, 1993; Krahn *et al.*, 1997), and, perhaps more importantly, by their extensive annual migration. This species migrates as far south as California and through the eastern Pacific as far as Japan, where these animals potentially feed on prey that are much more highly contaminated than prey in the waters of the southern Bering Sea or the Gulf of Alaska.

Concentrations of non- and mono-*ortho* PCBs followed the general pattern for PCBs with highest TEQs found in pups (32 pg/g ww) as compared to adults and subadults (17-21 pg/g ww) (Beckmen, 1999) (Annex Table 16).

#### Walrus

Walrus are long-lived benthic feeders and, as such, an important indicator species for bioaccumulating contaminants in benthic marine food webs. Although they have an important role in native traditional hunts, rela-

tively little is known about levels of OCs in walrus compared to seals and beluga. The previous AMAP report noted that although walrus generally have low OC concentrations in blubber, some individuals feed at higher trophic levels than others and, as a result, have much higher contaminant concentrations (de March *et al.*, 1998). These walrus are believed to be including ringed seals in their diet (Muir *et al.*, 1995b). Since then, studies have examined levels of OCs in walrus from the Russian and Alaskan Bering Sea, eastern Hudson Bay, and eastern and western Greenland (Muir and Kwan, 2000; Muir *et al.*, 2000e; Seagars and Garlich-Miller, 2001; Kucklick and Struntz, 2002;).

Levels of PCBs and OC pesticides such as DDTs, HCHs, and chlordanes in the blubber of Pacific walrus (*Odobenus rosmarus divergens*) sampled across the Russian and the Alaskan Bering Sea (Seagars and Garlich-Miller, 2001; Kucklick and Struntz, 2002) were comparable to the relatively low levels previously reported in Bering Sea walrus (Galster and Burns, 1972; Taylor *et al.*, 1989) (Annex Table 12). As in earlier studies, DDT compounds were essentially absent from walrus blubber in this region, while heptachlor epoxide was detected for the first time at low ng/g levels. In the Bering Sea walrus, oxychlordane dominated  $\Sigma$ CHLs, while  $\beta$ -HCH dominated  $\Sigma$ HCHs (Kucklick and Struntz, 2002). Additionally, significantly higher levels of  $\Sigma$ PCBs occurred in adult males (450 ng/g ww) than females (160 ng/g ww), and significantly higher oxychlordane levels occurred in males than females of any age. No differences were detected between sexes for  $\Sigma$ HCH and dieldrin (Seagars and Garlich-Miller, 2001).

OC levels in walrus from eastern Hudson Bay (northern Quebec) (Atlantic walrus, *O. r. rosmarus*) were much lower than those found in earlier studies from the same area (Muir and Kwan, 2000; Annex Table 12), most likely because the earlier studies sampled walrus that were seal-eaters (Muir *et al.*, 1995b). Nonetheless, concentrations of many OCs were comparable to those in recent reports for walrus from other nearby regions such as Foxe Basin (Muir *et al.*, 1995b). Levels of  $\Sigma$ PCBs were similar to those in walrus from the Bering Sea and northwestern Greenland, while  $\Sigma$ DDT levels were similar to those in walrus from northwestern Greenland (Muir *et al.*, 2000e).  $\Sigma$ HCHs and HCB were also detected in concentrations similar to those in walrus from the Bering Sea and eastern and northwestern Greenland (Muir *et al.*, 2000e) (Annex Table 12). In the case of  $\Sigma$ CHLs, levels in eastern Hudson Bay walrus were higher than those in the Bering Sea and northwestern Greenland, but lower than in eastern Greenland walrus (Muir *et al.*, 2000e).

An examination of spatial trends in levels of OC compounds in two separate stocks of walrus from Greenland found lower concentrations in the northwestern Greenland animals, and much higher levels of all OCs, except HCH isomers and mono/dichlorobiphenyls in samples from eastern Greenland (Muir *et al.*, 2000e) (Annex Table 12).  $\Sigma$ PCB levels averaged 246 ng/g ww in male walrus from northwestern Greenland, and 2860 ng/g ww in samples from eastern Greenland. However, DDT isomers showed the most difference between the two sites, with *p,p'*-DDE and *p,p'*-DDT being 50 and 69 times higher, respectively, in the eastern Greenland

walrus. Toxaphene levels in walrus in northwestern Greenland were lower than those from eastern Greenland, but were consistent with levels reported in ringed seals from Barrow, Alaska, ringed, harp, and bearded seals from the White Sea, and harp seals from the Barents Sea (Wolkers *et al.*, 2000; Hoekstra *et al.*, 2003a; Muir *et al.*, 2003). The eastern Greenland walrus showed a pattern of OCs characteristic of seal-eating animals, although they may have been consuming other prey as well. Despite the apparent differences in prey, the higher levels of OCs in eastern Greenland compared to the northwestern Greenland animals are consistent with results for polar bears, seals and gulls from the same regions.

#### *Butyltins in pinnipeds*

Mean butyltin ( $\Sigma$ BTs) concentrations of 17 ng/g ww were found in liver tissue from Steller sea lions sampled at the Aleutian Islands (Alaska) between 1976 and 1985 (Kim *et al.*, 1996a). In these animals, TBT concentrations ranged from 1.9 to 5.6 ng/g ww, while DBT and MBT concentrations ranged from below detection limits to 20 ng/g ww and 7.1 ng/g ww, respectively. DBT was the dominant component in the liver samples, which is in contrast to fish (their prey), in which TBT levels are highest, irrespective of the species and sampling site (e.g., Suzuki *et al.*, 1992; Takayama *et al.*, 1995). This implies that Steller sea lions are capable of metabolically transforming at least some of the TBT residues they consume into DBT and MBT (Kim *et al.*, 1996a). There was no evidence of age- or sex-dependent accumulation of butyltin residues in Steller sea lions, and no evidence of increasing concentrations of DBT and TBT between 1976 and 1985 (Kim *et al.*, 1996a). Butyltin concentrations in Svalbard ringed seals were 1.5 ng/g ww for MBT, 3.1 ng/g ww for DBT, and TBT was not detected (Berge *et al.*, 2002). TBT and its metabolites, DBT and MBT, were non-detectable (sub ng/g) in ringed seal blubber and liver from Labrador and northern Quebec (Muir *et al.*, 1999c; 2000c).

### 4.4.6.2. Cetaceans

#### 4.4.6.2.1. Mysticetes

##### *Minke whales*

North Atlantic minke whales are trans-Atlantic, as well as polar to north temperate in range, and are capable of large-scale migrations. They feed at a lower trophic level than polar bears, odontocetes, and some seals, eating primarily capelin, herring, Atlantic cod, and krill (Larsen and Kapel, 1981; Nordøy and Blix, 1992; Skaug *et al.*, 1997).

Concentrations of PCBs and major OC pesticides quantified in the blubber of minke whales from seven regions across the north Atlantic and European Arctic ranged widely, but generally increased from west to east (Hobbs *et al.*, 2003) (Annex Table 13, Figure 4.49). Contaminant concentrations suggested that western and southeastern Greenland minke whales represent one group of whales, which are distinct from both Jan Mayen minkes and those from other more easterly regions. Although some differences were detected in PCB and OC pesticide concentrations between Jan Mayen whales and those from other regions, overall,

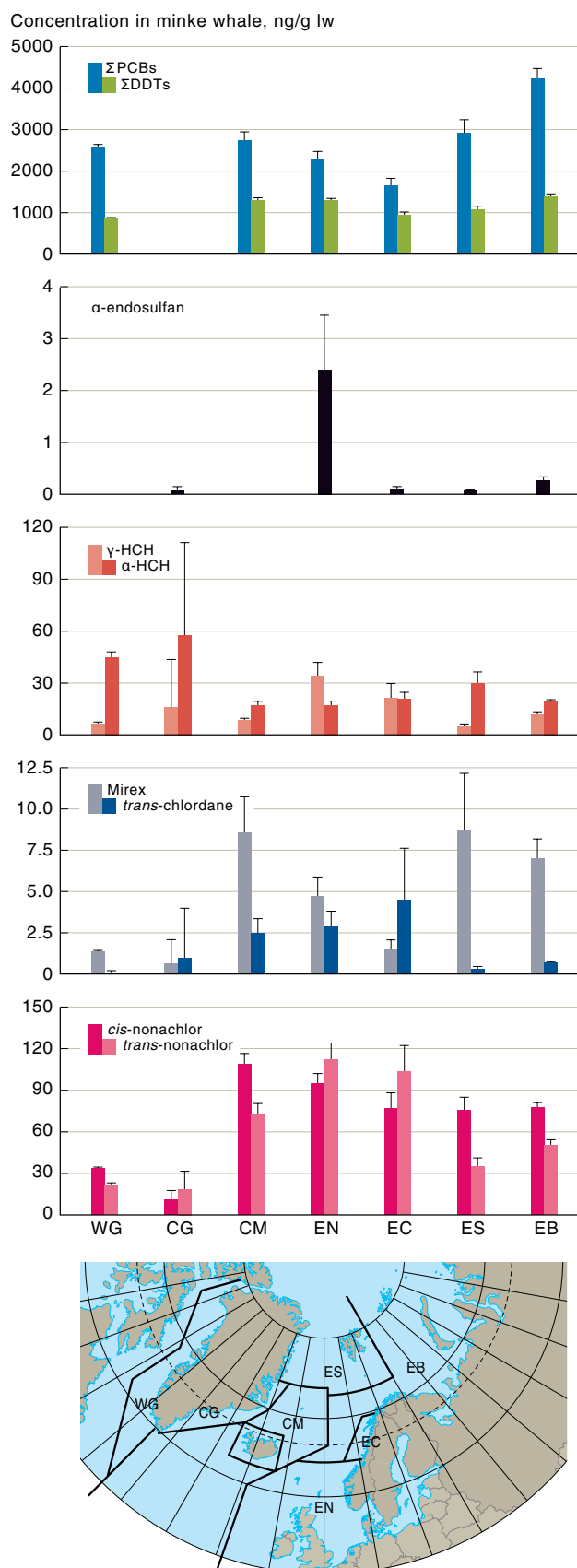


Figure 4.49. Sex-adjusted, geometric mean ( $\pm$  95% CI) concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs, and geometric mean concentrations of  $\alpha$ -endosulfan,  $\alpha$ -HCH,  $\gamma$ -HCH, mirex, trans-chlordane, cis-nonachlor, and trans-nonachlor in minke whales from several areas in the North Atlantic region. Areas are IWC 'small areas' (WG: west Greenland, CG: southeast Greenland, CM: Jan Mayen, EN: North Sea, EC: Vestfjorden/Lofoten, ES: Svalbard, EB: eastern Barents Sea) (Hobbs *et al.*, 2002a).

Jan Mayen whales were more similar to minke whales from the easterly regions, and generally appeared to be most similar to those from Svalbard and Vestfjorden/Lofoten.

Contaminant levels in minke whales from the North Sea and the Kola Peninsula region of the Barents Sea were relatively distinct from other groups of whales, and were often higher compared to other areas. For example, North Sea minkes had higher concentrations of the current-use pesticides, lindane ( $\gamma$ -HCH),  $\alpha$ -endosulfan, and *trans*-nonachlor, compared to Svalbard whales. Minke whales from the Kola Peninsula region of the Barents Sea had higher concentrations of PCBs, and a lower  $\Sigma$ DDT: $\Sigma$ PCB ratio compared to those from Vestfjorden/Lofoten, as well as marginally higher  $\Sigma$ PCB concentrations compared to the North Sea and Svalbard whales. Therefore, although some mixing probably occurs between whales from the Kola Peninsula and Svalbard and between whales from North Sea and Vestfjorden/Lofoten, there appears to be some separation between whales from these two sets of regions. Furthermore, Skaare *et al.* (2001c) found significantly lower concentrations of PCBs in the dorsal blubber of adult female minkes sampled near Spitsbergen (461 ng/g ww) compared to adult females sampled in the North Sea (2190 ng/g ww) (Annex Table 13). Despite these distinctions, there was a general blurring in levels and patterns of PCBs and OC pesticides in minkes from Jan Mayen, Svalbard, Vestfjorden/Lofoten, the North Sea, and the Kola Peninsula region of the Barents Sea, suggesting that minke whales are quite mobile and feed in multiple areas (Hobbs *et al.*, 2003).

Mean  $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHL,  $\Sigma$ HCH, and HCB concentrations reported in minke whales from the northeast Atlantic (western Spitsbergen, Lofoten/Vesteralen, Finmark, Bjørnøya, and the Kola Peninsula) in 1992 (Kleivane and Skaare, 1998) were about two to three times higher than mean levels reported in the same general regions by Hobbs *et al.* (2003). The lower contaminant levels in more recent minke samples (Hobbs *et al.*, 2003) may be attributable to changes in the availability of northeast Atlantic minke whale prey species. For example, in 1992, the diet of Svalbard minke whales was dominated by capelin, while in subsequent years, following the collapse of the capelin stocks in 1992/1993, their diet was almost 100% krill (Haug *et al.*, 2002).

Concentrations of PCDD/Fs and non- and mono-*ortho* PCBs were also analyzed in dorsal blubber of female minkes sampled near Svalbard and in the North Sea (Annex Table 16). Total mean TEQs based on these compounds were 24 pg/g ww (range 10-37 pg/g ww) for Svalbard and 67 pg/g ww (range 25-103 pg/g ww) for the North Sea whales (Skaare *et al.*, 2001c). The major contribution to the TEQs came from the non- and mono-*ortho* PCBs.

Levels of total toxaphene showed considerably more geographic variability than  $\Sigma$ PCBs (Hobbs *et al.*, 2002a). Levels were highest in the North Sea whales, followed by those from Jan Mayen and west Svalbard. The lowest levels occurred in whales from either side of Greenland and from the Vestfjorden/Lofoten region of Norway. Total toxaphene levels in minkes from the Kola Peninsula region of the Barents Sea were intermediate in comparison to the other sites (Hobbs *et al.*, 2002a).

#### *Grey whales (Eschrichtius robustus)*

The eastern grey whale makes an annual round-trip migration between their breeding grounds in subtropical waters (e.g., off Baja California and the southern Gulf of California) and their predominant feeding grounds in the northern Pacific Ocean. Although the majority of feeding occurs in the Bering and Chukchi Seas around Alaska (Highsmith and Coyle, 1992; Moore *et al.*, 2000), some animals spend extended periods in the spring and summer opportunistically feeding in the coastal waters of Washington, California, Oregon, and British Columbia. Grey whales feed primarily on benthic prey, such as ampeliscid amphipods, using suction to engulf sediments and prey from the bottom, then filtering out water and sediment through their baleen plates and ingesting the remaining prey. This unique feeding method often results in the ingestion of sediment and other bottom materials. Thus, exposure to sediment-associated contaminants is possible if they feed in areas containing contaminated sediments and benthic invertebrates.

$\Sigma$ PCB and  $\Sigma$ DDT blubber concentrations of 17 grey whales sampled from the Chirikov Basin of the Russian Bering Sea in October 1994 ranged from 110 to 1300 and 30 to 540 ng/g ww, respectively (Krahn *et al.*, 2001; Tilbury *et al.*, 2002) (Annex Table 13). All of the whales sampled were juveniles, thus minimizing the influence of length (i.e. related to age and developmental stage) on contaminant concentrations. Furthermore, no significant differences in concentrations (lw) occurred by sex within each group of whales sampled. Blubber from one grey whale from the Chukotka Peninsula (Lavrentiya) was analyzed for OCs including PCDD/Fs.  $\Sigma$ PCB and  $\Sigma$ DDT concentrations were 196 ng/g ww and 93 ng/g ww, respectively, and TEQ based on PCDD/F was 1.8 pg/g ww (Annex Table 16) (RAIPON/AMAP/GEF Project, 2001). No other grey whale data are available for comparison. However, on a wet weight basis, these levels are similar to  $\Sigma$ PCB and  $\Sigma$ DDT levels in other mysticetes. For example, mean concentrations of  $\Sigma$ PCBs in blubber of bowhead whales (*Balaena mysticetus*) from Alaska (Hoekstra *et al.*, 2002c), fin whales (*Balaena physalus*) from the north Atlantic (Aguilar and Borrell, 1994), and minke whales from western Greenland (Hobbs *et al.*, 2003) were 359, 732, and 2290 ng/g ww, respectively. Similarly, mean concentrations of  $\Sigma$ DDTs in bowhead-, fin-, and minke whales were 331, 633, and 650 ng/g ww, respectively.

#### *Bowhead whales*

The bowhead whale is a large mysticete found in Arctic waters. The largest population, the Bering Sea stock, migrates annually between the eastern Beaufort Sea-Amundsen Gulf in summer and the Chukchi and northern Bering Sea in the winter (Schell *et al.*, 1989; Lowry, 1993; Moore and Reeves, 1993). Blubber and liver tissue were collected during the Native subsistence harvest of this species, which coincides with the migration between the Beaufort and Bering Seas.

OC concentrations in Alaskan bowhead whales reported by Hoekstra *et al.* (2002c) (Annex Table 13) were similar to values reported by Mössner and Ballschmiter (1997) and O'Hara *et al.* (1999). Hoekstra *et al.* (2002c) also reported mean concentrations of toxaphene in bow-



head whales for the first time. The bioaccumulation of PCBs and OC pesticides in bowhead whale blubber appeared to change seasonally and was characterized by differences in analyte metabolism. The proportions of individual OCs in bowhead blubber samples were related to the harvest season. Principal components analysis showed that whales harvested in the autumn of 1997-2000 had higher loadings for less chlorinated PCBs, toxaphene, and chlorobenzene congeners, which distinguished them from spring harvest specimens collected during 1998-2000 that had higher proportions of the more recalcitrant PCBs (CBs 153 and 180), *p,p'*-DDE, HCB and  $\beta$ -HCH. The patterns of OC loadings found in bowhead whale blubber generally reflect the different levels of persistent OCs in the surface waters of the Bering and Beaufort Seas (Iwata *et al.*, 1993; Hoekstra *et al.*, 2002c). Toxaphene levels in the Beaufort Sea bowheads were comparable to those measured in minke whales from western Greenland and the Norwegian Sea (Hobbs *et al.*, 2002a). Results reported by Hoekstra *et al.* (2002c) demonstrate that bowhead whales are exposed to a variety of OC pollutants from lower trophic-level prey items, and that seasonal variation in OC profiles coincides with the annual migration between the Beaufort and Bering Seas. The influence of collection season on OC bioaccumulation in blubber seen in their study suggests that OC concentrations change annually in the whales, and that spatial differences in contaminant levels within the Bering-Chukchi-Beaufort region may be reflected in marine biota.

#### 4.4.6.2.2. Odontocetes

##### *Belugas*

The beluga or white whale is a small (up to 4.5 m long) toothed cetacean (odontocete) that is circumpolar in distribution in the Arctic. Belugas feed near the top of the marine food web on a variety of fish and invertebrates, such as cephalopods and shrimp (Banfield, 1974), and are relatively long lived (>35 yr). They have a nearly continuous distribution across the Russian Arctic coast, but in the NE Atlantic Ocean they are limited to the north coast of Norway, and in the Pacific Ocean, to the Okhotsk Sea (Kleinenberg *et al.*, 1964). They are present along the east and west coasts of Greenland and in North America, from Alaska across the Canadian western Arctic to a large population in Hudson Bay, and among islands in the eastern Canadian Arctic. Beluga movements are extensive, seasonal and generally predictable. They come into coastal waters and estuaries in mid-summer, and spend the winter offshore in pack ice and polynyas (Brodie, 1989).

In a recent study by Krahn *et al.* (2000), samples of beluga blubber from three of the five different Alaskan stocks (eastern Beaufort Sea, eastern Chukchi Sea and Cook Inlet) were analyzed for concentrations and patterns of OC contaminants.  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs and HCB levels in Point Lay (eastern Chukchi Sea) beluga blubber (Wade *et al.*, 1997; Hoekstra *et al.*, 2003a) (Annex Table 13) were in good agreement with those reported for the same region by Krahn *et al.* (2000). Blubber of Alaskan belugas contained PCBs and OC pesticides in ranges similar to those found in belugas from the Canadian Arctic (Annex Table 13) (Sang *et al.*,

2000). The Cook Inlet stock generally had the lowest concentrations, while the eastern Beaufort Sea stock had the highest concentrations. This is somewhat surprising given that whales from the Cook Inlet stock reside in one of the most 'urban' areas of Alaska, where anthropogenic contamination results from relatively higher density of human residents and commercial activities. The eastern Chukchi Sea and eastern Beaufort Sea stocks could clearly be distinguished from the Cook Inlet stock by patterns of individual OC analytes, and although there was some overlap between the males and females of the Cook Inlet stock, these groups were also generally separated (Krahn *et al.*, 2000). These differences in contaminant patterns are indicative of differences in contaminant accumulation, which likely reflects differences in habitat use and prey forage among the stocks.

In the Canadian Arctic, PCBs were quantitatively the most predominant POPs in the blubber of eastern Hudson Bay belugas, followed by DDT and chlordane-related compounds (Sang *et al.*, 2000) (Annex Table 13). Higher concentrations of all major groups of OCs, except for  $\Sigma$ HCHs, were observed in blubber of males (n=8) as compared to females (n=2). Data were too limited to evaluate effects of age or animal condition on contaminant levels. No other contaminant data for beluga blubber samples from the Hudson Strait area (Kangiqsujuaq) have been analyzed for comparison. Levels of major OC groups in male beluga blubber sampled by Sang *et al.* (2000) were, however, similar to previous reports for male beluga from eastern Hudson Bay (Nastapoka River) in the mid-1980s (Muir *et al.*, 1990b), and much lower than levels in beluga from the southeast Baffin beluga stock. Muir *et al.* (1990b) concluded that belugas sampled from the Kangiqsujuaq area of the Hudson Strait are from a population that inhabits eastern Hudson Bay and southern Hudson Strait, and not the southeast Baffin Island area. Innes *et al.* (2001) also concluded that there are distinct differences in the OC signature of southeast Baffin and Hudson Bay belugas. Comparisons between east Hudson Bay and west Hudson Bay beluga are complicated by missing age estimates and lipid content data for these whales (Hobbs *et al.*, 2002b) (Annex Table 13). With this in mind, it seems that the western Hudson Bay beluga may generally have had higher PCB and OC pesticide concentrations than the east Hudson Bay whales. This may be at least partially a result of the west Hudson Bay beluga having been sampled six years before the east Hudson Bay whales. OC levels for beluga from Kimmirut, Baffin Island were similar to, but somewhat higher than, levels in the east Hudson Bay beluga (Annex Table 13). Non- and mono-*ortho* PCBs in beluga from the same region were analyzed by Helm *et al.* (2002), and levels ranged from 14.4 to 294 ng/g ww, with CBs 118, 105, and 156 being the most dominant congeners. Total non- and mono-*ortho* PCBs were lower in females ( $60.2 \pm 40.3$  ng/g ww) than males ( $228 \pm 113$  ng/g ww). TEQs in males and females were  $1.73 \pm 0.627$  pg/g lw and  $1.32 \pm 0.909$  pg/g lw respectively, most of which was accounted for by congener 126 (Helm *et al.*, 2002) (Annex Table 16). Stern and Addison (1999) analyzed non-*ortho* PCBs (CBs 77, 126, and 169) in beluga from Cumberland Sound. The TEQ in blubber from 1997 was 6.1 pg/g lw.

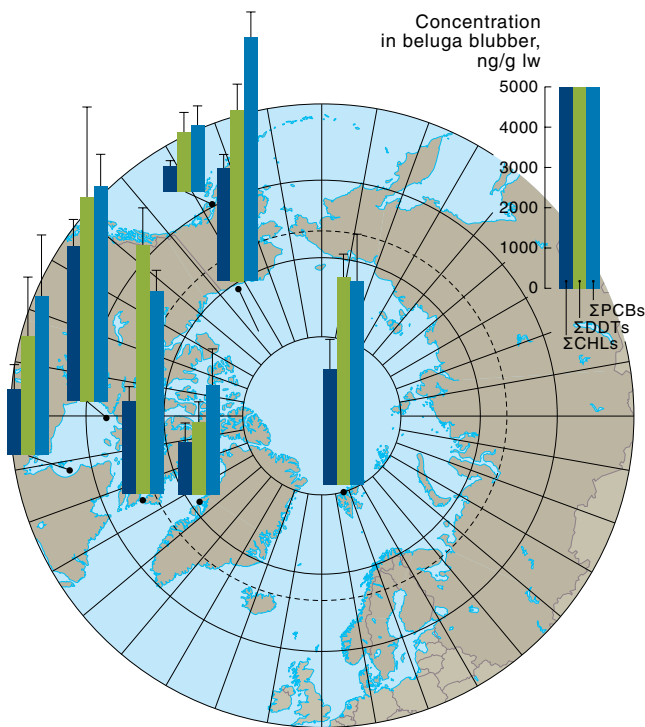


Figure 4-50. Mean ( $\pm$  95% CI) concentrations (ng/g lw) of  $\Sigma$ PCBs,  $\Sigma$ DDTs and  $\Sigma$ CHLs in male beluga whale blubber (Stern and Addison, 1999; Krahn *et al.*, 2000; Sang *et al.*, 2000; Andersen *et al.*, 2001a; Muir and Johansen, 2001; Hobbs *et al.*, 2002b).

Further east, Andersen *et al.* (2001a) and Wolkers (2002) reported levels and patterns of OC pollutants in blubber biopsies taken from belugas from western Svalbard (Annex Table 13). Levels reported in the two studies were comparable, and the slightly lower levels in whales examined by Wolkers (2002) were consistent with their younger age (immature) compared with those reported in Andersen *et al.* (2001a). Andersen *et al.* (2002) also reported high levels of toxaphene ( $11\,400 \pm 4210$  ng/g lw) in the same ten Svalbard belugas they examined for OC pollutants (Annex Table 13). Overall, levels of  $\Sigma$ PCBs,  $\Sigma$ DDTs and  $\Sigma$ CHLs in belugas from the Arctic are generally highest in the Beaufort and Barents Seas and western Hudson Bay, somewhat lower in the rest of the Canadian Arctic, and lowest in Cook Inlet, southern Alaska (Figure 4-50).

As is typical of top predators in Arctic marine food chains (Borgå *et al.*, 2001; Fisk *et al.*, 2001c), the major compounds detected in beluga blubber were  $\Sigma$ PCBs,  $\Sigma$ DDTs, toxaphene, and  $\Sigma$ CHLs. OC levels in the Svalbard belugas were generally similar to, or slightly higher than, values in other more westerly Arctic beluga stocks (Annex Table 13) (Muir *et al.*, 1990b; Stern *et al.*, 1994; Muir *et al.*, 1999b), but were lower than in those from the St. Lawrence River (Canada) (Muir *et al.*, 1990b; Muir *et al.*, 1996b). In whales examined by Andersen *et al.* (2001a),  $\Sigma$ HCHs was dominated by  $\beta$ -HCH in contrast to reports for belugas from the Canadian Arctic (Muir *et al.*, 1990b). Patterns in relative prevalence of various OC compounds to their compound groups were consistent with what is seen in other marine mammals in the Arctic.

Toxaphene levels in beluga from Alaska and Svalbard were quite variable, with mean values ranging from roughly 1000 to 10 000 ng/g ww (Wade *et al.*, 1997;

Hoekstra *et al.*, 2003a), while levels in Pangnirtung beluga were consistently on the high end of this range (Stern, 1999; Stern and Addison, 1999) (Annex Table 13).

#### *Long-finned pilot whales (Globicephala melas)*

Screening for OCs was performed on blubber samples from 420 long-finned pilot whales sampled in 1997 at the Faroe Islands (Dam and Bloch, 2000). The analyses were done on pooled samples of whales from nine separate schools with each school represented by a subsample of 50 individuals randomly sampled from a total of 805 individuals. In general, concentrations of all OCs were of similar magnitude in the juveniles and adult males, but were approximately half as high in adult females. *p,p'*-DDE dominated the DDT group, accounting for  $60 \pm 6\%$ ,  $57 \pm 7\%$ , and  $65 \pm 3\%$  of the  $\Sigma$ DDTs in the young, females and males, respectively. Chlordane was dominated by *trans*-nonachlor, which accounted for approximately 50% of  $\Sigma$ CHLs. There is a general scarcity of data on these compounds, but it may be noted that in a study of five male and nine female long-finned pilot whales stranded in Newfoundland in 1980 (Muir *et al.*, 1988), concentrations of *trans*-nonachlor and  $\Sigma$ CHLs were half of what was found in the Faroese study. The concentrations of HCB in the Newfoundland study (Muir *et al.*, 1988) were very similar to what was found in the Faroese study (Dam and Bloch, 2000). The much higher PCB concentrations found in pilot whales of the Faroe Islands compared to stranded individuals in New Zealand (Schröder, 1998) is noteworthy, and in agreement with what is seen for baleen whales (O'Shea and Brownell, 1994). In the Faroese study, the mean  $\Sigma$ PCBs for all 417 individuals was 11 900 ng/g ww, whereas the mean for the 61 individuals from New Zealand was 310 ng/g ww.

In the Faroe Islands, samples from 100 individuals from two pilot whale schools (Sandavágur, August 26, 1997 and Tórshavn, September 24, 1997) were analyzed in order to determine the variability in pollutant load within the groups of adult males and females, and juveniles (Dam, 2001). No strong relationships between  $\Sigma$ PCB<sub>7</sub> and *p,p'*-DDE and pilot whale length were found although older males tended to have higher concentrations. Toxaphene was also determined in these samples including Parlars 26, 32, 50, 62, and 69, but of these, Parlars 32 and 69 were not detected in any sample. Juvenile males had the highest concentrations of toxaphene and levels were lower in adults.

#### *Killer whales (Orcinus orca)*

Killer whale populations that inhabit Washington's Puget Sound (U.S.), the inside waters of British Columbia, southeast Alaska, and Kenai Fjords/Prince William Sound, Alaska, have been extensively studied over the past 30 years. Two eco-types of killer whales, 'transient' and 'resident', occur in all of these regions. These eco-types are genetically distinct and differ in various aspects of morphology, vocalization patterns, diet, and habitat use. For example, transient killer whales feed on marine mammals, while resident killer whales are fish-eaters. Various genetic and photo-identification studies of eastern North Pacific killer whales have provided information on the male-female composition of most of these resident pods and transient groups, as well as the ap-

proximate age, reproductive status, and putative recruitment order (birth order) of the individual whales.

Concentrations of PCBs and OC pesticides, including dioxin-like PCB congeners and DDTs, in blubber biopsies from free-ranging transient killer whales were much higher than those found in resident animals sampled from the Kenai Fjords/Prince William Sound areas between 1994 and 1999 (Annex Table 13). Mean blubber  $\Sigma$ PCB concentrations were 3900 ng/g ww in residents (range 270-27 000 ng/g ww) and 59 000 ng/g ww (range 4900-140 000 ng/g ww) in transients. These differences are apparently due to the differences in diets of these two killer whale eco-types (Ylitalo *et al.*, 2001). The concentrations of PCBs and DDTs that were measured in blubber of the Alaskan killer whales were much higher than the concentrations in blubber of various other cetaceans and pinnipeds that reside and feed in Alaskan waters (Annex Tables 12 and 13) (de March *et al.*, 1998). The  $\Sigma$ PCB levels measured in these Alaskan transient killer whales are similar to those recently reported in biopsy blubber samples of transient killer whales from the more contaminated coastal waters of British Columbia (Ross *et al.*, 2000). Mean mono-*ortho* PCB TEQs in residents were 29 pg/g ww (range 1.5-150 pg/g), and in transients, 220 pg/g ww (range 14-580 pg/g) in blubber (Annex Table 16).

#### Harbour porpoises

Concentrations of PCBs, HCB, *p,p'*-DDD, *p,p'*-DDE,  $\gamma$ -HCH, and PCDD/Fs were lower in immature harbour porpoises from southwestern Greenland in comparison with levels in immature, non-Arctic dwelling porpoises from the North and Baltic Seas (Bruhn *et al.*, 1999).  $\alpha$ -HCH values were highest in porpoises from southwestern Greenland. *p,p'*-DDT was only detected in the Greenland porpoises, possibly due to a higher degree of metabolic induction in the non-Arctic animals. The higher proportion of lower chlorinated PCBs in the Arctic samples is likely associated with a predominance of these more volatile compounds in the atmosphere, or perhaps to a lesser degree, metabolic induction, and subsequent excretion of these compounds, relative to the non-Arctic animals. The concentrations of PCDD/Fs, given as TEQs in the Arctic porpoises, ranged from 0.2 to 0.9 pg/g lw with a median of 0.41 pg/g lw (Annex Table 16). Mono-*ortho* PCBs were also determined, and TEQs based on these were stated to be 40-70 times higher than for PCDD/F TEQs in the three populations, but actual TEQ values were not given in the publication. CB 118 contributed most to the total TEQs.

Further east, Berggren *et al.* (1999) found that PCB and DDT levels in blubber sampled from mature male harbour porpoises from the Norwegian west coast were relatively high compared to other Arctic-dwelling cetaceans, and were comparable to levels in samples taken from the Baltic and Kattegat-Skagerak Seas (Annex Table 13). They were an order of magnitude higher than in porpoises from southwestern Greenland (Bruhn *et al.*, 1999). Porpoises from the west coast of Norway had the most variability in non-*ortho* PCB concentrations (96-7624 pg/g lw) compared to the other more southerly sites, which may be caused by exposure of some porpoises to a local contaminant source. Their  $\Sigma$ PCDD/F concentrations ranged from 7.3 to 19 pg/g lw, lower

than in mature porpoises from the more southerly locations. The mean TEQ based on PCDD/Fs, non- and mono-*ortho* PCBs was 111 pg/g lw (Annex Table 16).

#### Narwhal (*Monodon monoceros*)

Narwhal are deep-water, benthic feeders and are an important Arctic species that has received little attention in terms of contaminants studies. In the Canadian Arctic, levels of PCBs, DDTs and chlordanes (not corrected for age) were generally higher in male narwhal from Pond Inlet (northeastern Baffin Island), than those from Broughton Island (eastern Baffin Island), or Grise Fjord (southern Ellesmere Island). Similar trends occurred for toxaphene in narwhal from these sites (Stern, 2001) (Annex Table 13). Concentrations of PCBs, DDTs, and chlordanes were generally higher in narwhal from Svalbard (Wolkers, 2002) as compared to those from Greenland (Denmark, 2002), while HCHs, HCB, and toxaphene levels were comparable between these regions (Annex Table 13). Overall, levels of POPs in narwhal were quite similar in the Canadian Arctic and western Greenland, while levels of PCBs, DDTs and chlordanes were considerably higher in the Svalbard narwhal. Levels of  $\Sigma$ HCHs, HCB and toxaphene were relatively consistent across the sites sampled.

#### Organotins in cetaceans

Studies indicate that marine mammals are exposed to organotins but data from the Arctic area are lacking. Harbour porpoise, harbour seal, and ringed seal are species that have been analyzed for organotins in Norwegian territories (Berge *et al.*, 2002). Relatively high concentrations were observed in liver from harbour porpoises caught in 1988 (Table 4·8), just before restrictions on the use of TBT (mainly on small boats) were introduced in several other European countries. The concentrations were significantly reduced 11 years later (Table 4·8), possibly as a consequence of the introduced restrictions. Mean concentrations in porpoise from the west coast of Norway in 1998/1999 were higher than in samples collected further north in the Barents Sea. Considerably lower concentrations were observed in seals compared to porpoises. The lowest concentrations were found in seals from Spitsbergen, where only traces of DBT and MBT (degradation products of TBT) were found. The degradation products in all samples were generally more predominant than TBT itself, and probably indicated metabolic capacity to degrade TBT. Triphenyltin (TPHT) was observed in all porpoise samples and in common seals but not in ringed seals. The limited data available indicate low to moderate exposure to organotins in Arctic areas like Spitsbergen and Bjørnøya. Marine mammals are more exposed, however, along the Norwegian coast. It is anticipated that exposure will decline further as a consequence of additional restrictions on the use of organotin in antifouling paint for ships.

Butyltin levels were quantified in liver tissue from male Dall's porpoises sampled from the Aleutian Islands chain, the Bering Sea, and the northwestern North Pacific between 1979 and 1984 (Tanabe *et al.*, 1998). Low levels ( $\Sigma$ BTs = 41-180 ng/g ww) were found in Dall's porpoises from these sites, compared with cetaceans and pinnipeds examined in other regions, including Japan, China, the Philippines, the west Pacific and India (17-



Table 4·8. Concentrations of mono-, di- and tributyltin and triphenyltin (in ng Sn/g ww) in marine mammals and seabirds from Norway and the Faroe Islands.

Species/year	Location	dw %	MBT	DBT	TBT	TPhT	Reference <sup>a</sup>
Harbour porpoise, 1988	Norwegian coast of the Barents Sea	–	35	285	98	27	1
Harbour porpoise 1999	Norwegian coast of the Barents Sea	–	11	67	34	6	1
Harbour porpoise, 1999	West coast of Norway	–	19	122	40	10	1
Common seal ,1998/1999	West coast of Norway	–	11.7	8.6	1.2	3.3	1
Ringed seal, 2000	Spitsbergen (Ny-Ålesund)	–	1	1.6	<1	<1	1
Glaucous gull, 1998	Bjørnøya	–	<1	11.7	<3	<3	1
Pilot whale, adult male, 2000	Faroe Islands	36	<0.1	1.3	3.0	<0.1	2
Pilot whale, adult female, 2000	Faroe Islands	22	<0.1	1.4	1.8	<0.1	2
Pilot whale, adult female, 2000	Faroe Islands	29	<0.1	1.8	2.2	<0.1	2
Pilot whale, fetus, 2000	Faroe Islands	19	<0.1	1.8	1.2	<0.1	2
Pilot whale, fetus, 2000	Faroe Islands	21	<0.1	<0.1	<0.1	<0.1	2
Pilot whale, fetus, 2000	Faroe Islands	14	<0.1	<0.1	<0.1	<0.1	2

<sup>a</sup> 1: Berge *et al.* (2002); 2: Mikkelsen (2002).

3000 ng/g ww). DBTs were predominant among the butyltins. MBT, DBT and TBT concentrations in the Dall's porpoises ranged from 22 to 33, 29 to 59, and 12 to 26 ng/g ww, respectively. Further east, butyltin (MBT, DBT, and TBT) was not detectable in livers from five Hudson Strait (Canada) belugas sampled in the summer of 1998 (de Mora *et al.*, 1999). The authors concluded that the limited maritime shipping activities in northern Quebec were not sufficient to provide an appreciable input of TBT into the marine environment of northern Quebec, and that since these same whales were contaminated with OCs but not butyltins, it was unlikely that aerial input could be a source of organotin compounds to Arctic whale populations. In contrast to this, butyltins were detected in all 21 samples examined from the more southerly St. Lawrence River population of beluga whales located in eastern Canada (de Mora *et al.*, 1999).

Organotins, such as mono-, di- and tri-substituted butyl- and phenyltin compounds, were analyzed in pilot whale kidney and blubber from three adults (two females, one male) and three fetuses sampled in the Faroe Islands in 2000 (Mikkelsen, 2002). The results showed no detectable concentrations of organotins in blubber tissue and kidney (detection limit 0.1 ng Sn/g ww) except some low concentrations of DBT and TBT near the detection limit in the adults and in one of the fetuses (Table 4·8).

#### 4.4.6.3. 'New' chemicals in pinnipeds and cetaceans

Of the data produced on 'new' chemicals in the Arctic, the greatest amount has been generated for marine mammals, in particular seals and whales. These are logical animals in which to check for 'new' chemicals, as seals and whales have among the highest levels of OCs in the Arctic.

##### PBDEs

PBDE data have been produced for seals and whales from the Canadian and European Arctic (Annex Table 17). There have been two temporal trends studies of PBDEs, in beluga (Stern and Ikononou, 2000; 2001) and ringed seals (Ikononou *et al.*, 2002), which have shown that the concentrations of these compounds are increasing in Arctic marine mammals (see Section 5.4.6.1). In general, concentrations of PBDEs are orders of magnitude less than legacy OCs such as PCBs and

DDTs. However, PBDEs consist of a much smaller number of individual compounds, so the difference between individual PBDE and individual PCB congeners is less. BDE47 is the most common congener measured, followed by BDE congeners 99 and 153. Other BDE congeners, such as 100 and 49 have been measured in Canadian Arctic beluga (Stern and Ikononou, 2000; 2001), but others, such as BDEs 85 and 138, have been reported as non-detectable in Svalbard beluga and Faroe Islands pilot whales (van Bavel *et al.*, 2001). These congeners are found at lower concentrations in the technical PBDE products, but may also be less prevalent due to biotransformation.

Although it is too early to draw conclusions about spatial trends of PBDEs in Arctic marine mammals, there are sufficient data to suggest that concentrations are higher in the European Arctic compared with the North American Arctic. Concentrations of ΣPBDEs were  $92.9 \pm 56.5$  ng/g ww in Svalbard beluga blubber collected in 1998 (van Bavel *et al.*, 2001) and were higher, compared to concentrations of 15.5 ng/g ww in beluga from the western Canadian Arctic (Stern and Ikononou, 2000; 2001). ΣPBDE concentrations in the blubber of ringed seals from northeastern Greenland ( $58 \pm 23$  ng/g ww) were an order of magnitude higher than levels reported from western Greenland ( $3.6 \pm 1.1$  ng/g ww) (Muir and Johansen, 2001) and the western Canadian Arctic (4.6 ng/g, ww) (Ikononou *et al.*, 2002).

Minke whales from the Barents Sea had ΣPBDE levels of 0.15–0.45 ng/g ww in muscle, while minke whales from the Norwegian Sea had higher levels at 3.1–15 ng/g ww in muscle (Herzke, 2002b).

The greatest concentrations of PBDEs measured in the Arctic are those observed in Faroe Islands long-finned pilot whales (144–1620 ng/g ww) (Annex Table 17) (van Bavel *et al.*, 2001). Concentrations are an order of magnitude greater than in any other Arctic marine mammal examined to date. Due to a general lack of PBDE data in the Arctic, it is difficult to determine if these high levels are due to spatial trends or the behavior of the pilot whales (i.e. trophic level). Additionally, as in beluga whales from Svalbard (van Bavel *et al.*, 2001), surprisingly high levels of methoxylated PBDE (Me-O-PBDE), almost comparable to the most abundant BDE47, were found in the pilot whale. Results for long-finned pilot whales from Tórshavn, Faroe Islands, also revealed large differences between PBDE concentrations with age



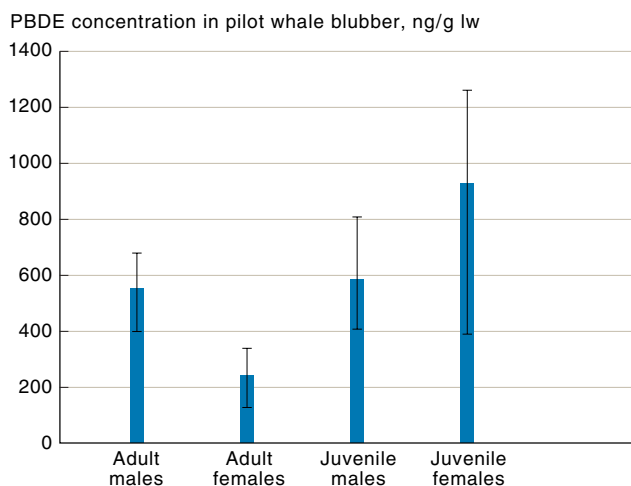


Figure 4-51. Mean (and range) of concentrations of PBDEs in long-finned pilot whales (adult males and females compared to juvenile males and females) caught at the Faroe Islands (van Bavel *et al.*, 2001).

and sex (van Bavel *et al.*, 1999). However, the highest levels were found in juveniles (402-1246 ng/g lw), with somewhat lower values in adult males (397-669 ng/g lw) and the lowest values in adult females (126-326 ng/g lw) (Figure 4-51) (van Bavel *et al.*, 1999). This indicates transfer of the PBDEs from mother to calf by lactation. Results from a mother and fetus also indicated the presence of a partial transplacental barrier, as the level in the fetus was generally half the amount of the mother (van Bavel *et al.*, 2001).

#### Other 'new' chemicals

In addition to the PBDEs, a number of studies have also examined several other 'new' OCs, including PFOS, tris(4-chlorophenyl) methanol (TCPM-OH), SCCPs, PCNs, endosulfan sulfate, and a number of chiral compounds.

PFOS was detected in the blood of ringed seals, and blood and liver of northern fur seals (Table 4-9) (Giesy and Kannan, 2001; Kannan *et al.*, 2001a). Concentrations were greater in liver compared to blood. No obvious trends between the Canadian and European Arctic in PFOS in ringed seals were observed, although this is a very small dataset.

Table 4-9. Concentrations of PFOS in liver and/or blood of Arctic marine mammals (ng/g ww or ng/mL) from Kannan *et al.* (2001a) and Giesy and Kannan (2001).

Species/tissue	Location	n	Collection year	Sex	Age class	PFOS <sup>a</sup>
Ringed seal blood plasma	Cumberland Sound	24	1998	M and F		<3-12
Ringed seal blood plasma	Spitsbergen	10	1996	4 M; 6 F	3.0-20 yr	8.1 ± 2.5
Ringed seal blood plasma	Spitsbergen	8	1998	3 M; 5 F	2.0-12 yr	10.1 ± 2.7
Northern fur seal liver	Pribilof Islands (Bering Sea)	13	1995 and 1998	11 M; 2 F	3 pups (<4 m); 10 subadults (2-4 yr)	<10-122 [38]
Northern fur seal blood	Pribilof Islands (Bering Sea)	10	1995	10 F	adult (>3 yr)	<6
Northern fur seal blood	Pribilof Islands (Bering Sea)	7	1995	7 M	subadult (2-4 yr)	<6
Polar bear liver	Northwestern Alaska (Barrow; Nuiqsut; Point Lay; Gambell; Shishmaref; Little Diomedea; Savoonga)	17	13 Dec. 1997- 15 Jun. 1999	14 M; 3 F	13 adults (>5 yr); 4 subadults (3-4 yr)	175-678 (350)
Polar bear blood	Beaufort Sea	14	1999	7 M; 7 F	n.a.	26-52 (34)

<sup>a</sup> Values in brackets [ ] indicate the percentage of detectable observations. Values in parentheses ( ) indicate the mean.

Lower chlorinated CBz have been detected in Arctic seawater and in lower trophic organisms from northern Quebec (Muir *et al.*, 2000c; Muir *et al.*, 2000f).

Hexachlorobutadiene (HCBd) is mainly created as a by-product in the manufacture of chlorinated hydrocarbons like tri- and tetrachloroethene. This hydrophobic (log  $K_{ow}$  4.78) and volatile product (vapor pressure = 19.96-20.00 Pa (20-25°C)) (Mackay *et al.*, 2000) has been detected at low levels in Arctic biota (Muir *et al.*, 2000f) and in the north Atlantic Ocean (WHO/IPCS, 1994).

TCPM-OH was measured in five ringed seals from northern Russia, near the Yenisey River, and had a mean concentration of about 47 ng/g lw, in 1995 (Watanabe *et al.*, 1999).

Blubber from beluga whales collected in 1994 near Kimmirut, Nunavut, was analyzed for PCNs and non- and mono-*ortho* PCBs (Helm *et al.*, 2002). ΣPCN concentrations ranged from 40 to 384 pg/g (n=6), on a wet weight basis (approximately 90% lipid), and were dominated by PeCNs. ΣPCN levels were lower in female belugas (n=3) than in males (n=3). Total non- and mono-*ortho* PCB concentrations were much higher than ΣPCNs, ranging from 14.4-294 ng/g ww in beluga whale blubber with the CBs 118, 105, and 156 having the highest concentrations. PCNs contributed an extra 11% (0.026-0.40 pg TEQ/g ww) to total TEQs (0.30-2.6 pg TEQ/g ww) calculated using non- and mono-*ortho* PCBs and PCNs. Most of the TEQs in beluga were accounted for by CB 126.

Blubber from ringed seals from Pangnirtung, Nunavut, collected in 1993 also contained PCNs (Helm *et al.*, 2002), but concentrations were lower than in beluga, ranging from 29-63 pg/g ww. The PCNs added insignificantly to the total TEQs.

Endosulfan sulfate was found in blubber of beluga from Pangnirtung collected in 1996/1997, with approximate mean concentrations of 13.5 ng/g lw (Stern and Addison, 1999).

#### Chiral contaminants

As discussed in the seabird section (Section 4.4.5.1), chiral pollutants exist in two forms as optical isomers called enantiomers. Enantiomers have identical physical-chemical properties and abiotic degradation rates, but can

have different rates of biotransformation, providing information on the ability of species to biotransform OCs.

Wiberg *et al.* (2000) examined ERs of  $\alpha$ -HCH and several chlordane compounds in the blubber and liver of ringed seals from Resolute Bay in the Canadian Arctic. The ERs in ringed seals were frequently nonracemic (ER  $\neq$  1), due to enantiomer-specific biotransformation; however, cod from the same region showed near-racemic mixtures (ER = 1) for most compounds. (+)- $\alpha$ -HCH was more abundant than (-)- $\alpha$ -HCH in ringed seals. There was no uniform trend for the ER changes in the various chlordane compounds examined. It was also determined that oxychlordane was formed in ringed seals and metabolized by polar bears that preyed on them, and the ER had an important role in the class separation of male/female seals and fat/liver tissues.

EFs of  $\alpha$ -HCH and other chiral contaminants in seal blubber may not reflect the metabolic capability of seals. Wiberg *et al.* (1998; 2000) noted near racemic  $\alpha$ -HCH (EFs = approximately 0.52) in blubber of ringed seals but non-racemic values in liver (EFs = approximately 0.6). This phenomenon was observed with other chiral pollutants, such as *trans*-chlordane, but in some cases, the EF was greater in blubber (Wiberg *et al.*, 2000). Wiberg *et al.* (1998) attributed this difference to greater metabolic activity in the liver as compared to the blubber. This would imply that the proportion of the  $\alpha$ -HCH body burden that is transformed is small, and consequently, the EF in ringed seal blubber is closer to that in the diet than in the liver. This is not always the case, since EFs of many chiral OCs in ringed seal blubber have been found that do not match their main prey item, Arctic cod (Wiberg *et al.*, 2000; Moisey *et al.*, 2001). In seabirds, which do not retain as large a reserve of fat as found in ringed seals, there were no differences in EFs of chiral chlordanes between liver and fat (Fisk *et al.*, 2001b). Differences in EFs of chiral pollutants between tissues of seals, and potentially other marine mammals, require further study.

Harbour and grey seals from Iceland showed an  $\alpha$ -HCH ER > 1 (Klobes *et al.*, 1998a). The ER of CB149 was comparable in the two species, but for oxychlordane, ER < 1 was observed in harbour seals, while the oxychlordane ER in grey seals was > 1. The differing ER for oxychlordane was consistent with results for blubber from two harbour seals from the German North Sea coast (König *et al.*, 1994) and a Baltic Sea grey seal (Müller and Buser, 1994). An excess of (+)  $\alpha$ -HCH (EFs = 0.58) was found in the blubber of harbour and grey seals, although no data were provided for their food.

EFs of chiral contaminants and stable isotopes of nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) were measured along with OCs in ringed seals collected from the east and west side of the Northwater Polynya in northern Baffin Bay (Fisk *et al.*, 2002d). *Cis*- and *trans*-chlordane, oxychlordane, and heptachlor epoxide were all non-racemic in the ringed seal blubber but did not vary with age, sex or collection site.  $\alpha$ -HCH appeared racemic (EF =  $0.50 \pm 0.01$ ) in the seals, although this EF is different from those previously observed in their prey species, and was found to vary significantly with age. An overall food web assessment of  $\alpha$ -HCH in the Northwater Polynya, which included the ringed seal data of the study by Fisk *et al.* (2002d), concluded that ringed seals do not meta-

bolize  $\alpha$ -HCH efficiently (Moisey *et al.*, 2001). EF values in the ringed seals varied considerably from other Arctic marine mammals and seabirds, providing additional evidence that the type(s) and characteristic(s) of the enzymes involved in biotransformation of chiral OCs vary between these organisms.

#### 4.4.6.4. Persistent OCs in other pinniped and cetacean tissues

Compared to blubber, fewer analyses of brain, liver, kidney, muscle, and blood have been made in pinnipeds and cetaceans. However, in all these tissues, OC concentrations are generally lower than in blubber because of their lower lipid content. Low ng/g ww levels of PCBs and OC pesticides were found in liver, kidney, and muscle from ringed seals sampled from Greenland (Muir and Johansen, 2001) and in blood sampled from ringed and bearded seals from Svalbard, Norway, and Alaskan northern fur seals (Bang *et al.*, 2001; Beckmen, 2002) (Annex Table 12). Similarly, low ng/g ww levels of PCBs and OC pesticides were found in brain, liver, and muscle from beluga from Hendrickson Island in the western Canadian Arctic (Metcalf *et al.*, 1999) and in bowhead whales from the Bering-Chukchi-Beaufort Seas (Hoekstra *et al.*, 2002c) (Annex Table 13). When lipid concentrations were compared to wet weight concentrations of OCs in brain, liver, kidney, and muscle from grey whales sampled from the Chirikov Basin of the Russian Bering Sea, lipid levels were significantly correlated to  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs, and HCB (Krahn *et al.*, 2001; Tilbury *et al.*, 2002). This relationship is consistent with a study by Aguilar and Borrell (1985) reporting that lipid content is an important factor in controlling accumulation of lipophilic OCs in marine mammals. In both beluga and grey whales, concentrations of OCs in these tissues, with the exception of brain, were generally more comparable when the values were calculated on a lipid weight rather than on a wet weight basis (Krahn *et al.*, 2001; Tilbury *et al.*, 2002).

In brain tissue, total lipid-normalized concentrations were significantly lower than in all other tissues. This is in agreement with previous reports that the blood-brain barrier controls the transport of certain contaminants to brain tissue (Norton, 1980). Beluga brain samples could be distinguished from other tissues by differences in PCB congener patterns and higher concentrations of  $\Sigma$ HCHs (primarily  $\alpha$ -HCH), as was reported in an earlier study for northern fur seals and harbour porpoises (Mössner *et al.*, 1992). In addition, the lipids in the brain of marine mammals consist of high proportions of polar lipids (i.e. phospholipids and cholesterol) (Fukushima and Kawai, 1980; Aguilar and Borrell, 1985; Tilbury *et al.*, 1997) that have a lower affinity for OCs than neutral lipids. Thus, the greater proportion of neutral lipids (i.e. triglycerides and non-esterified fatty acids) found in tissues other than the brain favors the accumulation of OC compounds in these tissues (Kawai *et al.*, 1988). These variations in patterns of POPs in the different tissues may also be influenced by differences in contaminant metabolism or the degree of blood perfusion in the various tissues (Kiceniuk *et al.*, 1997; Jenssen *et al.*, 1996; Metcalfe *et al.*, 1999). The need for caution in interpreting OC concentration data from different tissues is

further demonstrated by findings reported for a study examining how concentrations of OCs in blood and blubber vary with nutritional condition in captive and wild, fasting ringed seals (Lydersen *et al.*, 2002). The study demonstrated that extreme variability occurs in the concentrations of OCs in seal blood in response to change in body condition as a result of fasting, and that the responses of blubber OC concentrations are also very different compared to those in blood. The authors concluded by recommending that, since the natural variation in body condition is extreme during annual cycles of phocid seals, blood should not be used in studies of OCs, where the aim of the study is to monitor OC levels for comparative purposes or time-trend analyses.

#### 4.4.6.5. Effects of age and sex on OC levels in pinnipeds and cetaceans

As mentioned previously, age and sex are important factors that must be taken into account to ensure accurate comparisons of contaminant levels in pinnipeds and cetaceans. Results of recent studies are consistent with previously recorded trends showing that most OCs occur at lower levels in juveniles than adults, and lower in adult females than in adult males (Krahn *et al.*, 1997; Wade *et al.*, 1997; Kleivane and Skaare, 1998; Severinsen *et al.*, 2000; Bang *et al.*, 2001; Ylitalo *et al.*, 2001; Fisk *et al.* 2002d; Hoekstra *et al.*, 2002c; Krahn *et al.*, 2001). PCB and OC pesticide concentrations also increase with age in males, and may either decrease, remain relatively constant or increase with age in females, although in the latter case, generally at a slower rate than in males (Wolkers *et al.*, 1998b; Muir *et al.*, 2000b; Fisk *et al.*, 2002d; Hoekstra *et al.*, 2002c). Lower levels of contaminants in females are normally primarily attributable to the transfer of compounds to offspring during gestation and lactation. Whether or not females experience age-related changes in contaminant levels is variable, and regardless of the rate of change, this age-concentration relationship likely depends largely on their level of contaminant exposure, as well as how often they successfully produce and wean offspring. Nonetheless, it is important to remember that age- and sex-related trends in contaminant concentrations may vary both within and between species, depending on the compound being examined.

Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs, and  $\Sigma$ CHL compounds often increase with age in marine mammals. This was the case in both sexes of ringed seals from the northern Baffin Bay region (Fisk *et al.*, 2002d), and in male, but not female, ringed seals from Alaska (Kucklick and Krahn, 2002), as well as those in a circumpolar study that statistically examined ringed seal data from the previous AMAP assessment (Muir *et al.*, 2000b). The significant relationships observed in the female ringed seals from northern Baffin Bay are likely due to the inclusion of very old female seals (>40 years) that have probably stopped reproducing (Fisk *et al.*, 2002d). Wolkers *et al.* (1998b) found PCBs increased with age in Svalbard ringed seals, but that sex was not an important variable, and suggested that in the Svalbard seals, continued feeding by females during lactation may compensate for loss of OCs during lactation. The authors also

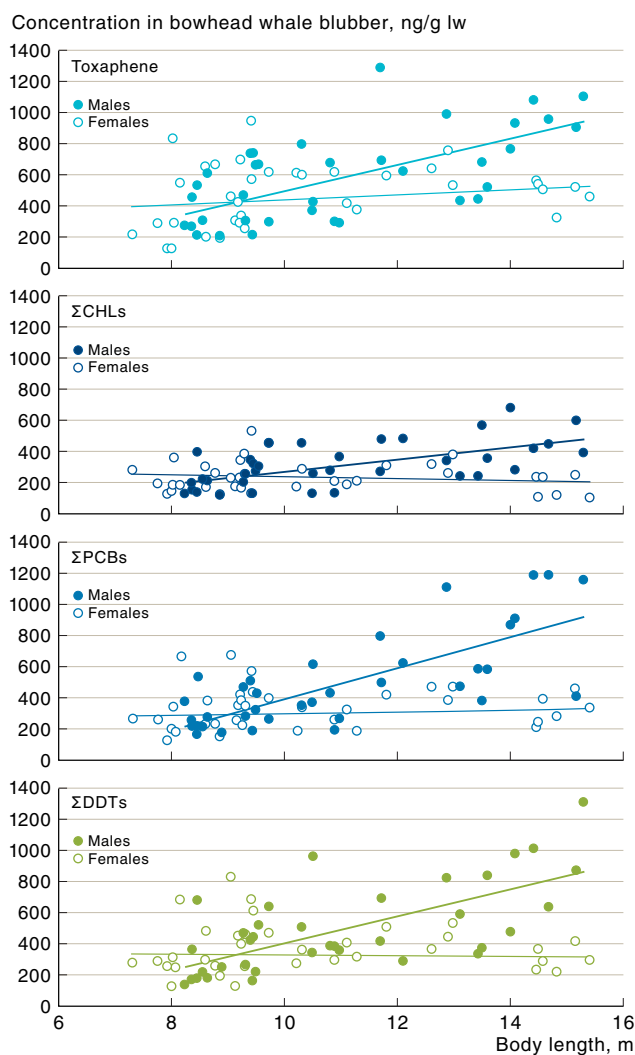


Figure 4-52. Body length versus  $\Sigma$ OC concentrations in blubber samples from male and female bowhead whales. Significant relationships between increasing body length and  $\Sigma$ OC concentrations ( $p < 0.05$  for all comparisons) were found for male bowhead whales (Toxaphene,  $r^2 = 0.44$ ;  $\Sigma$ CHLs,  $r^2 = 0.38$ ;  $\Sigma$ PCBs,  $r^2 = 0.56$ ;  $\Sigma$ DDTs,  $r^2 = 0.45$ ). Source: Hoekstra *et al.*, 2002c.

found no difference in PCB congener pattern, relative to CB153, between the sexes in adults, while juveniles showed significantly higher relative concentrations of the lower chlorinated PCBs (28, 52, 74, 99, 101, and 118) and lower relative concentrations of the higher chlorinated PCBs (170 and 180) as compared to the adults (Wolkers *et al.*, 1998b).

In male Alaskan bowhead whales, levels of  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs, and toxaphene significantly increased with body length, and thus presumably with age (Figure 4-52) (Hoekstra *et al.*, 2002c). In female bowheads, this relationship applied until they reached approximately 13 m in length, while those longer than 13 m had generally lower concentrations of these compounds than shorter, younger females. Similar decreasing OC concentrations with age occurred in female belugas from Point Lay, Alaska (Wade *et al.*, 1997). However, in contrast to this, concentrations of  $\Sigma$ HCHs and HCB or  $\Sigma$ CBz often do not show sex- or age-related differences in pinnipeds and cetaceans. For example, no sex-related differences were observed for HCB or dieldrin concentrations in Alaskan ringed seals of comparable ages (Kucklick and Krahn, 2002), or for  $\Sigma$ CBz and  $\Sigma$ HCHs in seals from the



Nunavut region (Fisk *et al.*, 2002d). Similarly,  $\Sigma$ HCH and  $\Sigma$ CBz or HCB levels were independent of age and sex in Alaskan bowhead whales and Barents Sea minke whales (Kleivane and Skaare, 1998; O'Hara *et al.*, 1999; Hoekstra *et al.*, 2002c). Notably, much lower levels of OCs occurred in reproductive female Alaskan killer whales than sexually immature whales or mature male animals of the same age class (Ylitalo *et al.*, 2001), and a single beluga fetus from Point Lay, Alaska, had about 10% higher concentrations for all OCs compared to its mother (Wade *et al.*, 1997).

Recruitment order (birth order) can also influence the concentrations of OCs in pinnipeds and cetaceans. For example, in northern fur seals from St. George Island, Alaska, pups of young (presumably primiparous) dams had significantly elevated levels of PCBs in their blood as compared with pups of older (multiparous) dams (Beckmen *et al.*, 1999). Similarly, in adult male resident Alaskan killer whales, first-recruit whales contained much higher OC concentrations than those measured in non-first-recruited (e.g., second-recruited, third-recruited) resident animals from the same age group (Ylitalo *et al.*, 2001).

#### Faroe Islands study

A recent study in the Faroe Islands examined the relationship between OC concentrations and length in pilot whales (1997), white-sided dolphins (*Lagenorhynchus acutus*) (1997), and grey seals (1993-1995) (Dam, 2001). The following comparisons of the OC concentrations in the three species are based on females because they maintain constant OC concentrations that are not dependent on age, and because there were a greater number of females to sample, especially in pilot whale pods. Overall, the single OC occurring at the highest concentration was *p,p'*-DDE, which was found at 10 800 ng/g lw in the juvenile females of one pod of pilot whales, with decreasing concentrations among juveniles in the other pilot whale pod, followed by white-sided dolphins and grey seals. The OCs occurring in the next highest concentration were Parlar 50 (a component of toxaphene) at 3900 ng/g lw and CB 153 at 3600 ng/g lw in the same group of juvenile pilot whales that had the highest *p,p'*-DDE concentrations. *p,p'*-DDT and *trans*-nonachlor were both found at similar concentrations in the odontocete juveniles, but were markedly lower in grey seals.

When comparing the juvenile and adult female grey seals, white-sided dolphins, and pilot whales, it appeared that the difference in OC burden between the two age groups was highest for white-sided dolphins and lowest for grey seals. This could be a consequence of different criteria used to sort individuals into the age groups for each species. Only grey seals were sorted based on actual inspection of reproductive state, and the odontocetes were sorted according to body length. The difference between  $\Sigma$ PCBs in the juvenile and adult white-sided dolphins was less than the average seen for the other OCs. Some of the chlordanes and  $\beta$ -HCH exhibited a similar pattern. This generally occurred for the two other species. The OC with greatest difference amongst the two age groups was oxychlordane, which occurred at an exceptionally low concentration (27 ng/g lw) in adult female white-sided dolphins. Similarly, the

mean value for sum toxaphene for the adult female white-sided dolphins was only one tenth that of the juveniles (Parlar 50 at 1872 ng/g lw,  $n=7$  juveniles).

#### 4.4.7. Polar bear

Polar bears (*Ursus maritimus*) are widely distributed throughout the Arctic, including some subarctic regions, and range over large areas in search of food. They move south with the ice in the autumn and winter and then north as the pack ice melts in the spring and summer. These seasonal movements of the sea ice also influence the distribution and concentration of their primary prey, ringed and bearded seals (Stirling *et al.*, 1982; Kingsley *et al.*, 1985). Polar bears are top Arctic predators, and often eat only the blubber from a seal (Stirling and McEwan, 1975), where the highest concentrations of OCs are found. Polar bears have superior biotransformation capacity and have high levels of OC metabolites.

OC levels in polar bears were covered extensively in the previous AMAP assessment (de March *et al.*, 1998) with good spatial coverage. Much like other marine mammals, OCs in polar bears were found to be generally highest in the European and lower in the North American Arctic (de March *et al.*, 1998). Some spatial trends were also observed within the Canadian Arctic (Norstrom *et al.*, 1998).

Since the first AMAP assessment, monitoring of OCs in polar bears has continued in western Hudson Bay (Norstrom 2000; 2001), Alaska, Svalbard, and Greenland. There have also been studies on OC levels in polar bear plasma from the Russian Arctic (Andersen *et al.*, 2001b; Lie *et al.*, 2003). Temporal-trend studies have been carried out in western Hudson Bay and Svalbard polar bears and are discussed in detail in Section 5.4.5.2.

#### Alaska polar bears

Although elevated OC concentrations have been documented in Canadian, eastern Greenland and Norwegian polar bear populations, relatively little information is available for populations in Alaska. Lentfer (1976) documented elevated OC concentrations in polar bears prior to the major oil and gas development on the North Slope. Data collected through satellite telemetry indicate that there are two distinct polar bear population stocks in Alaska: one in the southern Beaufort Sea and the other in the Chukchi/Bering Seas (Amstrup, 1995), with an area of overlap from Point Barrow to Point Hope. Differences in the feeding ecology of polar bears between the Beaufort and the Chukchi/Bering Seas may affect OC concentrations found in the two stocks. Specifically, polar bears in the Chukchi/Bering Seas feed more heavily on Pacific walrus carcasses.

Levels of  $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHL, and  $\Sigma$ HCH determined in adult male polar bears sampled from the Barrow and St. Lawrence Island regions of Alaska between 1996 and 1998 (Krahn *et al.*, 2002), were consistent with levels in adult males from the southern Beaufort Sea population in northern Alaska and the Chukchi/Bering Sea population in western Alaska (Evans, 2001) around the same time (Annex Table 14). These data provide an important addition to contaminant level data in Alaska. Levels of PCBs in these Alaskan bears are relatively low compared to levels found in polar bears in



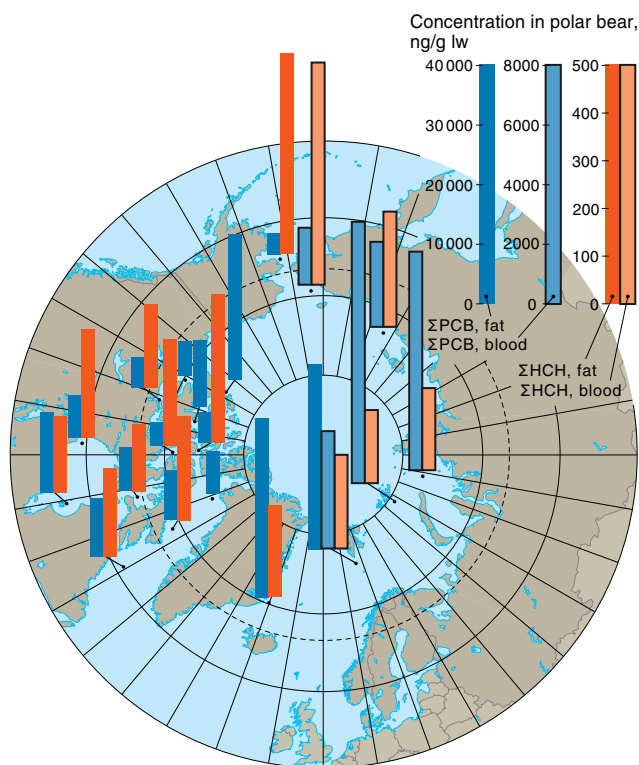


Figure 4-53. Concentrations (of  $\Sigma$ PCBs and  $\Sigma$ HCHs in blood from adult female polar bears (age 5-28 yr) from Svalbard and the Russian Arctic (Andersen *et al.* 2001b), and in fat from polar bears (adjusted to levels expected in 11-year-old males after correction for age and sex) from the Bering Sea, Canada, eastern Greenland, and Svalbard (de March *et al.*, 1998). At Svalbard, lipid-weight PCB concentrations measured in fat are approximately five times higher than in blood from the same animals. Assuming that this relationship is the same in other areas, high fat PCB concentrations can be expected in bears in the Russian Arctic.

eastern Hudson Bay, Canada, eastern Greenland, and Svalbard, Norway (Figure 4-53).  $\Sigma$ PCB levels for males from Alaska were higher than those reported by Norstrom *et al.* (1998) for three bears taken west of Barrow, Alaska. Significantly higher levels of  $\Sigma$ PCBs were found in bears from the southern Beaufort Sea population than in the Chukchi/Bering Seas population. Average  $\Sigma$ HCH levels in bears sampled by Evans (2001) (Annex Table 14) are among the highest levels reported in the Arctic, and were similar to the high levels reported for the Chukchi and Bering Seas by Norstrom *et al.* (1998). There were no significant differences between  $\Sigma$ HCH levels in the Chukchi/Bering Seas and southern Beaufort Sea populations.  $\beta$ -HCH, the most persistent HCH isomer, constituted about 93% of  $\Sigma$ HCHs. Although  $\alpha$ -HCH contributed only a small fraction of the  $\Sigma$ HCHs in fat, unlike  $\beta$ -HCH, it occurred in significantly greater concentrations in bears from the southern Beaufort Sea region. The chlordanes compounds in these bears were the next most abundant compounds, and although, the levels were not significantly different between the two populations ( $p < 0.07$ ), there was a trend suggesting higher levels in the southern Beaufort Sea population.

#### Greenland polar bears

OC levels were recently determined in the fat of Greenland polar bears collected in 1999/2000 (Sonne-Hansen, 2002; Dietz *et al.*, 2003) (Annex Table 14). These bears ranged in age from two to ten years. As observed in

other polar bears,  $\Sigma$ PCBs dominated the OC loads with concentrations that were an order of magnitude greater than any other OC group. Concentrations of OCs were greater in the males except for chlordanes, which have been observed in polar bears from other regions (Norstrom *et al.* 1998). Concentrations of  $\Sigma$ PCBs were much lower than previously reported for East Greenland bears whilst levels of DDE were within the range previously reported (see Section 5.4.5).

#### Iceland polar bears

Although polar bears are not naturally occurring in Iceland, they do appear occasionally on drifting pack ice. Klobes *et al.* (1998b) analyzed PCB and OC pesticide levels in the adipose tissue and liver of one such polar bear. They found distinctly different contaminant profiles in the two tissues, where the liver had patterns similar to those found in the Icelandic Arctic fox, with oxychlordanes as the predominant compound. In the adipose tissue, CB 153 predominated, followed by C180 and oxychlordanes. Oxychlordanes and  $p,p'$ -DDD were the only compounds found to be more abundant in liver than in adipose tissue, and toxaphene Parlars 26 and 50 were present in the adipose, but not the liver tissue. The sum concentration of Parlars 26 and 50 found in adipose tissue was within the range of the levels of toxaphene Parlars (26, 52, and 62) quantified in the adipose tissue of polar bears from Svalbard in an earlier study (Bernhoft *et al.*, 1997). Due to the high levels of oxychlordanes relative to PCBs, the authors concluded that it was likely that the bear originated from Greenland, not Svalbard.

#### OCs in Norwegian and Russian polar bear plasma

Geographical variations in PCB (Andersen *et al.*, 2001b) and OC pesticide concentrations (Lie *et al.*, 2003) were studied in blood samples from ninety adult female polar bears from Svalbard, Franz Josef Land, Kara Sea, East Siberian Sea and Chukchi Sea, between 1987 and 1995 (Annex Table 14). Regional differences in levels and patterns of PCBs, oxychlordanes, *trans*-nonachlor,  $\alpha$ -HCH,  $\beta$ -HCH, and  $p,p'$ -DDE were found. Bears from Franz Josef Land and the Kara Sea had similar  $\Sigma$ PCB levels and these were higher than all other populations (Figure 4-53). Svalbard polar bear PCB levels were similar to those from the East Siberian Sea, but higher than those from the Chukchi Sea. Svalbard polar bears had relatively lower proportions of CB99 and higher proportions of CB194 than bears from other regions. Bears from Franz Josef Land had higher proportions of CB180, but lower CB153 levels compared to all other regions. Of the PCB congeners investigated by Andersen *et al.* (2001b), the lower chlorinated CBs increased, and the higher chlorinated CBs decreased from Svalbard eastward to the Chukchi Sea. In all regions, oxychlordanes was the dominant OC pesticide, and the highest levels of oxychlordanes, *trans*-nonachlor, and DDE were found in bears from Franz Josef Land and the Kara Sea. Polar bears from the Chukchi Sea had the highest levels of  $\alpha$ - and  $\beta$ -HCH. The lowest  $\alpha$ -HCH concentration was found in bears from the Kara Sea and was lower than in bears from all the other circumpolar regions. In all the bears,  $\Sigma$ HCHs was dominated by  $\beta$ -HCH. HCB levels did not differ between regions.

Results from Andersen *et al.* (2001b) and Lie *et al.* (2003) combined with earlier findings (Bernhoft *et al.*,

1997; Norstrom *et al.*, 1998), indicate that polar bears from Franz Josef Land and the Kara Sea have the highest  $\Sigma$ PCB,  $\Sigma$ CHL and  $\Sigma$ DDT levels in the Arctic. Decreasing trends were seen eastward and westward from these regions, and may imply the presence of significant pollution sources in the Russian Arctic area. Regional differences in pollution sources, contaminant transport, and prey preferences could also explain the variation in PCB and OC pesticide levels and patterns between regions.

Ringed seals are assumed to be the most important and common prey of polar bears. However, Kleivane *et al.* (2000) found an unexpectedly high number of bears on the ice east of Svalbard feeding on harp seals in June 1995. Significantly higher OC concentrations were found in blubber samples from adult male harp seals from this area during their June 1995 molt (when they fast and live off their blubber), as compared to ringed seals sampled at the same time, which marks the very beginning of their molting season (Kleivane *et al.*, 2000). Significant species-specific differences were, however, detected only for  $\Sigma$ HCHs and HCB, while differences in  $\Sigma$ PCBs,  $\Sigma$ DDTs, and  $\Sigma$ CHLs were ascribed to age, xiphosternal blubber thickness, and possible differences in xenobiotic metabolizing capacity (Boon *et al.*, 1992; Wolkers *et al.*, 1999). These results indicate that it is not necessarily the species, but the time, availability, and biological condition of polar bear prey that may play a major role in the biomagnification of OCs at the top of the Arctic ecosystem (Kleivane *et al.*, 2000).

#### OCs in Canadian polar bear plasma

Concentrations of OCs were determined in the plasma of Resolute Bay polar bears (Norstrom, 2000; Sandau, 2000) (Annex Table 14). This work also included plasma samples from Svalbard polar bears.  $\Sigma$ CHLs and  $\Sigma$ PCBs were the dominant OC groups found in the plasma of polar bears from both regions. OC concentrations were two times higher in subadults than adults except for  $\Sigma$ DDT concentrations, which were similar. These results were in line with previous findings in polar bear adipose tissue. The exceptions were chlordanes, which were 30-60% lower in males, but concentrations were similar comparing the same sex in both areas.  $\Sigma$ PCB concentrations were similar in males and females from both areas, and two to three times higher in the Svalbard bears, in line with previous analyses of adipose tissue from these areas (Norstrom, *et al.* 1998).

#### Influence of age and sex on OC levels in polar bears

The effects of age and sex are also important considerations with respect to PCBs and OC pesticides in polar bears. In 1995, Norstrom (1999a) examined concentrations of OCs in male and female polar bears from Hudson Bay to test the effect of sex over a single year. Concentrations of OCs in males were similar to those in females; however, there was a (generally non-significant) tendency for most residues to be lower in males, except PCBs and DDTs, which were slightly higher in males. The difference in PCBs between males and females was not as large as observed in more extensive data sets. Norstrom *et al.* (1998) showed that males had 40% higher  $\Sigma$ PCB concentrations than females on average. Younger male bears have  $\Sigma$ PCB concentrations closer to those in females, which may explain the similarity be-

tween males and females in 1995. Males had 30% lower levels of chlordanes than females, which is the same as that found by Norstrom *et al.* (1998). Polischuk *et al.* (2002) showed that males are capable of metabolizing chlordanes during a seasonal fast, while females are not. The enhanced metabolic capability of males therefore, explains the lower levels of chlordanes and possibly some of the other compound groups in males. Lower levels of  $\Sigma$ PCBs and lack of an age effect in females are presumed to be due to the additional losses from lactation.

#### Influence of reproduction on OC levels in polar bears

To study the effect of fasting, gestation and lactation on toxicokinetics, POP concentrations were determined in adipose tissue, plasma, and milk samples from seven female polar bears and their cubs near Cape Churchill, Hudson Bay, between 1992 and 1996 (Polischuk, 1999; Norstrom, 2000). Pregnant females were captured from August 7 to October 7, and the same females with cubs were captured from March 2 to March 17 of the following year before they had moved onto the ice to begin hunting seals. All females had therefore been fasting five to seven months by the time of their second capture. Body composition of females was determined from  $^2$ H dilution in blood, and body weight. The total body mass of females declined by  $43 \pm 5\%$ , and total fat mass declined by  $42 \pm 3\%$ . The proportion of mass lost as fat, ranged between 55 and 66%.

During gestation and early lactation, the mean concentrations of  $\Sigma$ PCBs,  $\Sigma$ CHLs, and  $\Sigma$ CBz in female adipose tissue increased significantly by  $2096 \pm 1292$  ng/g ww,  $1600 \pm 1349$  ng/g ww, and  $49 \pm 23$  ng/g ww, respectively. Adipose tissue concentrations of  $\Sigma$ DDT declined by  $91 \pm 82$  ng/g ww, while concentrations of  $\Sigma$ HCH remained the same. Despite these increases in concentration, POP body burdens in female polar bears declined during gestation and the early lactation period due to loss of fat mass (Figure 4.54). In descending order, the mean proportional decrease in body burdens was

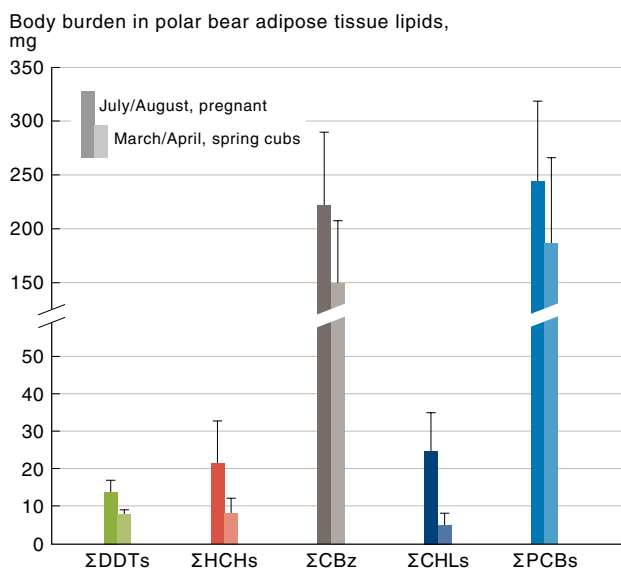


Figure 4.54. Mean ( $\pm$  SD) body burden (mg) of major POPs in adipose tissue lipids of polar bear females on land, in the summer (pregnant) and the following March (with cubs-of-the-year, COYs) in the Cape Churchill area, Hudson Bay (1992-1996). In all cases, the difference in body burden between the two periods was statistically significant.

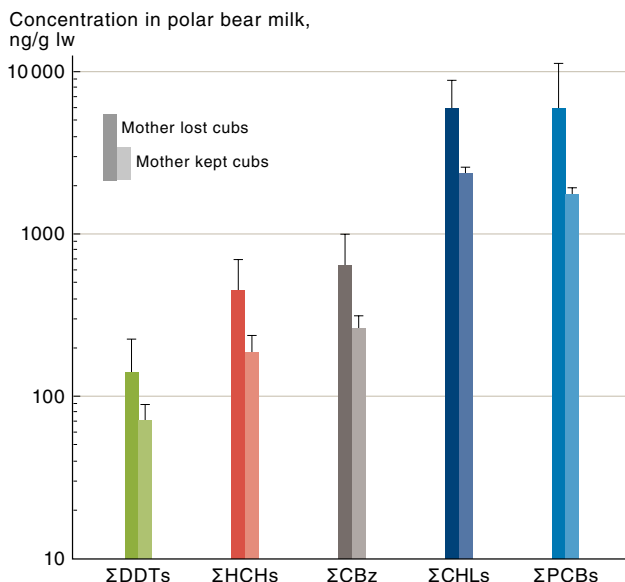


Figure 4-55. Mean ( $\pm$  SD) concentrations of major POPs in milk of female polar bear with cubs, after emerging in March from dens in the Cape Churchill area, Hudson Bay (1992-1996). The data are grouped according to whether the female still had her cubs the following fall, or had lost them. In all cases, the difference in concentrations between the two groups was statistically significant.

$\Sigma$ DDTs ( $75 \pm 8\%$ ) >  $\Sigma$ HCHs ( $61 \pm 8\%$ ) >  $\Sigma$ CBz ( $45 \pm 12\%$ ) >  $\Sigma$ CHLs ( $29 \pm 20\%$ )  $\approx$   $\Sigma$ PCB ( $24 \pm 16\%$ ), while the mean mass of POPs lost was  $\Sigma$ CHLs ( $71 \pm 57$  mg) >  $\Sigma$ PCBs ( $56 \pm 34$  mg) >  $\Sigma$ DDTs ( $20 \pm 8$  mg) >  $\Sigma$ HCHs ( $14 \pm 7$  mg) >  $\Sigma$ CBz ( $7 \pm 2$  mg). Biotransformation during gestation and lactation probably accounts for the greater proportional decrease in  $\Sigma$ CBz,  $\Sigma$ HCHs, and  $\Sigma$ DDTs compared to  $\Sigma$ PCBs and  $\Sigma$ CHLs.

The mean period between first and second capture (mostly in their dens) was  $188 \pm 22$  days, and the mean number of days of lactation prior to sampling was approximately  $79 \pm 4$  days. It is likely that the toxicokinetics of OCs during the first 100 days were similar to that during the subsequent summer fast. The polar bear cub weighs only about 0.7 kg at birth (I. Stirling, pers. comm., Canadian Wildlife Service, Environment Canada, Edmonton AB) and two newborn cubs represent about 0.4% of the mother's weight. Therefore OC transfer to the fetus is unlikely to be a significant part of the mother's body burden. Thus, most of the 24-29% loss of body burdens of the poorly biotransformed  $\Sigma$ CHLs and  $\Sigma$ PCBs from the mother during the 188-day fast must have been transferred to the cub in milk during the 79-day lactation period prior to capture, at which time the cubs weighed  $12.7 \pm 0.9$  kg. However, on average, only about 25-50% of this body-burden loss could be accounted for by  $\Sigma$ CHL and  $\Sigma$ PCB burdens in the cubs. Furthermore, a crude estimate of lactational transfer based on cub growth rates could only account for half of the body-burden loss from the females. These discrepancies are difficult to reconcile and require further study.

Mothers that were recaptured in the autumn without cubs had higher OC concentrations in their milk when emerging from their dens in spring. By comparison, mothers recaptured in the autumn and still accompanied by cubs had lower OC concentrations in their milk the previous spring (Figure 4-55). The differences in concentrations were significant for all residue classes. For example, PCBs were approximately three times higher

(5780 ng/g lw) in females that lost their cubs than in females that kept their cubs (1830 ng/g lw). It is not known how much significance can be attached to this finding in terms of reproductive performance, but it is suggestive, at least, that cub survival may be dependent on the degree of exposure to OCs in milk.

In a study on Svalbard, 35 mother/cub pairs were sampled during late March to mid-May in 1995-1998 (Lie *et al.*, 2000). Blood plasma samples were analyzed for six CBs (99, 118, 153, 156, 180, and 194), which make up 78% of total PCB concentrations in polar bears based on 28 congeners. Samples were grouped according to age and reproductive status: cubs (cubs-of-the-year); yearlings; females with cubs; and, females with yearlings. Geometric mean  $\Sigma$ PCB concentrations in plasma were 12 300 ng/g lw in cubs, 5820 ng/g lw in females with cubs, 6820 ng/g lw in yearlings, and 2945 ng/g lw in females with yearlings (Annex Table 14, Figure 4-56). Cub  $\Sigma$ PCB concentrations were significantly higher than the other three groups, and  $\Sigma$ PCB concentrations in fe-

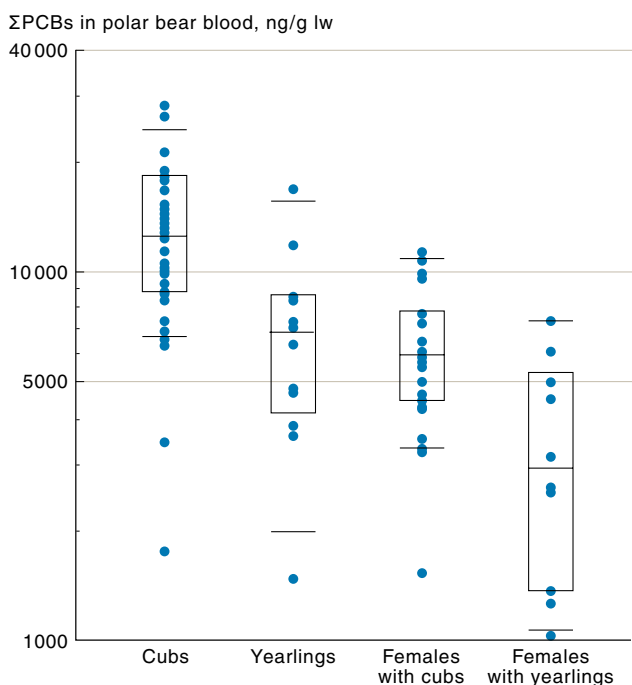


Figure 4-56.  $\Sigma$ PCB concentrations in blood plasma of polar bears: cubs (three to four months old), yearlings, females with cubs, and females with yearlings (Lie *et al.*, 2000). Dots represent individual values.

males with yearlings were significantly lower than the other three groups ( $p < 0.05$ ).

The  $\Sigma$ PCB concentrations found in the adult bears and yearlings were comparable to those found in earlier studies on Svalbard (Bernhoft *et al.*, 1997; 2000). No previous studies of 3-4 month old cubs on Svalbard have been done. The results show that cubs and yearlings have higher  $\Sigma$ PCB concentrations than their mothers, probably due to lactational transfer. This has also been seen previously in Canadian polar bears (Polischuk *et al.*, 1995; Polischuk, 1999). Cub survival in the Svalbard population is lower than in other populations, and the high  $\Sigma$ PCB concentrations found in cubs compared to older bears could be a possible explanation. There may be other causes, however, such as high population density.



*Tissue distributions of OCs in female polar bears*

In the vicinity of Cape Churchill, Hudson Bay, between 1992-1996, distribution of POPs in plasma relative to adipose tissue was determined in the following stages: pregnant females in the summer at the beginning of the on-land fast; females with cubs emerging from the den; females with cubs in summer; and, females with cubs after 1-2 months fasting (Polischuk, 1999; Norstrom, 2000). At all of these stages, the ratio of concentrations of  $\Sigma$ CHLs and  $\Sigma$ PCBs in whole plasma to adipose tissue lipids was the same as the fraction of lipids determined in plasma, 0.01. That is, these POPs were equally distributed in adipose and plasma lipids. Plasma:adipose tissue ratios of  $\Sigma$ CBz in pregnant females in summer and in females the following spring with cubs, and  $\Sigma$ HCHs in pregnant females were also distributed according to lipid content. The plasma:adipose tissue ratios of  $\Sigma$ CBz and  $\Sigma$ HCHs were two to three times higher in plasma of females with cubs in the summer and autumn than predicted by lipid content, suggesting that components of the plasma other than extractable lipids were responsible for some of the carrying capacity of plasma at these times. The plasma:adipose tissue ratio of  $\Sigma$ DDT was close to two times that of the lipid fraction in female plasma at all times. This is probably indicative of binding to proteins in plasma, as well as lipids. There was no indication that physical-chemical properties, such as water solubility or  $\log K_{ow}$ , were governing the partitioning of POPs between plasma and adipose tissue.

Ratios of POP concentrations in milk lipids:female adipose tissue lipids were determined in spring, summer and autumn. The ratio was greatest for  $\Sigma$ HCHs and lowest for  $\Sigma$ DDTs and  $\Sigma$ PCBs. On a lipid weight basis, the ratio ranged from close to 1:1 for  $\Sigma$ PCBs and  $\Sigma$ CHLs to approximately 2:1 for  $\Sigma$ CBz and  $\Sigma$ HCHs, indicating that milk lipids were not at equilibrium with adipose tissue lipids for these compounds. The milk:adipose ratios for  $\Sigma$ CBz were significantly greater than  $\Sigma$ CHLs,  $\Sigma$ DDTs, and  $\Sigma$ PCBs. Similarly, ratios for  $\Sigma$ CHLs were significantly greater than  $\Sigma$ DDTs and  $\Sigma$ PCBs.

The ranking of the milk:adipose ratios for the chemical groups was similar to that of the  $K_{ows}$  for the chemicals (Hawker and Connell, 1988; Mackay *et al.*, 1992), unlike what was seen for plasma. Since it is improbable that concentrations in extractable milk lipids would be higher than those in adipose tissue on a thermodynamic basis, low  $K_{ow}$  OCs must partition from adipose tissue to constituents of milk other than extractable lipids, such as lipoproteins.

*Effect of seasonal fasting on whole-body toxicokinetics in Hudson Bay polar bears*

POP concentrations were determined in adipose tissue from 47 fasting polar bears in the Cape Churchill area of western Hudson Bay during the summer to autumn of 1992-1995 (Polischuk, 1999; Norstrom, 2000). Body burdens (mg/animal) were also determined based on body composition determined from  $^2\text{H}$  dilution in blood and body weight. Adipose tissue, milk, and blood samples were taken when the bears came on land in July/August. The same bears were recaptured and sampled in September/October (47-68 days apart) during the fasting period. Five categories of bears were defined: cubs-of-the-year (COYs); yearlings (YRLGs); females with COYs

(Fem/COYs); females with yearlings (Fem/YRLG); and, males. There were no single females sampled.

Concentrations of  $\Sigma$ CBz,  $\Sigma$ CHLs, and  $\Sigma$ PCBs in adipose tissue lipid increased, and concentrations of  $\Sigma$ DDTs decreased in all bears during the 47-68 day fast. The changes in concentration of  $\Sigma$ CHLs and  $\Sigma$ PCBs in adults were mostly significant. In COYs, the concentrations of  $\Sigma$ CHLs and  $\Sigma$ PCBs increased 30%, while those of  $\Sigma$ HCHs and  $\Sigma$ DDTs decreased by 20% and 34%, respectively, during the fasting period. Increases in concentration in adipose tissue in adults were entirely due to utilization of lipids by the bears. Most of the increases in concentration in COYs were due to decreased adipose tissue, not uptake from nursing. The body burden of  $\Sigma$ CHLs and  $\Sigma$ PCBs in females with COYs did not decrease during fasting. Taken together, these data suggest that the COYs were nursing very little during the 47-68 day period.

Biotransformation of *p,p'*-DDE and HCHs occurred in all bears during the fast, as did biotransformation of oxychlordan and some other chlordan compounds in adult males. Chlorobenzenes, oxychlordan (except males), and the major PCB congeners (CBs 60, 99, 138, 153, 156, 170, 180, and 194) were not metabolized or cleared by any other mechanism at a measurable rate over 47-68 days of fasting.

In males, no age-related increases were found in concentrations of the compounds with slow biotransformation rates during fasting, except highly chlorinated PCBs (CBs 180 and 194). That is, average annual rate of intake and excretion of the POPs are balanced. Clearance of all POPs by a mechanism other than biotransformation is, therefore, occurring for at least part of the year. For most of the POPs, there was no significant difference in concentrations or trends in males and females. Therefore, lactation is not governing the rate of excretion in females, except perhaps for octa- and nonachlorinated PCBs. The only clearance mechanism for slowly biotransformed POPs that remains available to both sexes, is partitioning to gut contents and excretion in feces during periods when they are feeding.

Toxicokinetics of POPs in polar bears are therefore likely to be variable with season, sex/reproductive status, and area. Slowly biotransformed PCBs, oxychlordan (except in adult males), and chlorobenzenes will be taken up and excreted in feces only during periods of active feeding. On the other hand, oxychlordan in males and HCHs and *p,p'*-DDE in all bears are taken up during feeding periods, but biotransformed throughout the year regardless of whether the bear is feeding or not. Due to lack of ice from which to hunt seals during summer and autumn in southwestern Hudson Bay, feeding may be restricted to 7-8 months of the year for males, solitary females and females with cubs or yearlings, and as short as 4 months for pregnant females. In more northerly areas, polar bears (except pregnant females) may not fast at all, if they can remain on the ice to hunt ringed seal or have marine mammal carcasses to scavenge. In this case, uptake and clearance will occur year-round.

## 4.4.7.1. 'New' chemicals in polar bears

A number of 'new' chemicals have been measured in polar bears. As an apex predator, the polar bear has among the highest levels of POPs and is a logical organism to



utilize when searching for 'new' chemicals. However, the superior biotransformation capacity of polar bears may also reduce levels of some chemicals below detection limits.

#### Perfluorinated compounds

PFOS has been detected in liver of polar bears from northern Alaska and in blood plasma of ringed seals from Pangnirtung (Nunavut) and Svalbard (Giesy and Kannan, 2001; Kannan *et al.*, 2001a). Other PFAs were detected at lower levels but not reported (Kannan *et al.*, 2001a; K. Kannan, pers. comm.). The levels of PFOS in polar bear liver are in the hundreds of ng/g ww range, suggesting that this compound is one of the most prominent individual organohalogen chemicals in polar bear, when considering levels of PCBs, chlordane, and HCH-related chemicals (Norstrom *et al.*, 1998). Levels in the polar bear were higher than levels found in ringed seals suggesting that PFOS can biomagnify (Table 4-9).

#### PBDEs

Mean  $\Sigma$ PBDE concentrations in polar bears from Svalbard are 17.5 ng/g ww (14-144 ng/g lw), in the same range as minke whales and beluga from the Barents Sea area (Annex Table 17).  $\Sigma$ PBDE concentrations are more than 100 times lower than  $\Sigma$ PCB levels. The congener pattern was dominated by BDE47 and a methoxy-TeBDE (van Bavel *et al.*, 2001). This indicates that polar bears may metabolize PBDEs, and therefore, the concentrations of parent compounds may not give an accurate picture of total exposure. No other data on PBDEs in polar bears could be found in the literature for comparison.

#### Chiral compounds

ERs of  $\alpha$ -HCH and several chlordane compounds were examined in the fat and liver of polar bears and the ringed seals that they preyed on (Wiberg *et al.*, 2000). The authors found that polar bears frequently showed non-racemic (ER  $\neq$  1) mixtures of most compounds due to enantiomer-specific biotransformation. As (+)- $\alpha$ -HCH was transferred up the food chain from ringed seals to polar bears, it became more abundant relative to (-)- $\alpha$ -HCH. There was no uniform trend for the ER changes in the various chlordane compounds examined. It was also determined that oxychlordane was formed in ringed seals and metabolized by polar bears. In addition, the ERs of some highly recalcitrant chlordanes in polar bear fat showed linear relationships with age (Wiberg *et al.*, 2000).

#### Metabolites of OCs in polar bears

There have been a number of comprehensive studies on OC metabolite formation, mainly MeSO<sub>2</sub>- and OH-PCBs, in polar bears (Letcher *et al.*, 1996; Norstrom, 1997; Sandau *et al.*, 2000). Polar bears have superior ability to biotransform OCs. For example, the PCB burden in polar bears is dominated by a small number of congeners, much less than that observed in their major prey item the ringed seal (Muir *et al.*, 1988).

MeSO<sub>2</sub>-PCB and -*p,p'*-DDE metabolites were examined in the tissues of polar bears shot in 1993 in Resolute Bay (Letcher *et al.*, 1996; Norstrom, 1997). Concentrations of the  $\Sigma$ MeSO<sub>2</sub>-PCB were highest in liver (3049  $\pm$  1290 ng/g lw) and represented 11% of the concentrations of  $\Sigma$ PCBs. In fat, testes, lung, and brain,

$\Sigma$ MeSO<sub>2</sub>-PCB had, respectively, 2, 9, 13, and 60 times lower levels than liver on a lipid weight basis. The major congeners in all tissues were 3- and 4-MeSO<sub>2</sub>-CB87 and 3- and 4-MeSO<sub>2</sub>-CB 101. MeSO<sub>2</sub>-PCB uptake from seal appears to be the most important source of MeSO<sub>2</sub>-PCBs in bears.

3-MeSO<sub>2</sub>-*p,p'*-DDE concentrations (303  $\pm$  85 ng/g lw) in liver were nearly half those of DDE and were 126 to 337 times higher than found in testes, adipose, and lung tissues. The highly asymmetric tissue distribution of  $\Sigma$ MeSO<sub>2</sub>-*p,p'*-DDEs may be due to several factors, such as the liver being the site of its formation or because of its highly selective binding in liver cells. It is not possible to resolve the relative importance of MeSO<sub>2</sub>-DDE bioaccumulation versus formation in the polar bear based on adipose tissue concentrations.

MeSO<sub>2</sub>-PCBs are efficiently transferred from polar bear females to cubs via milk, resulting in concentrations that are approximately three times higher in the cubs than their mothers, compared to approximately two times higher for PCBs. 3-MeSO<sub>2</sub>-*p,p'*-DDE was not preferentially transferred to cubs. Decreased cub survival is emerging as one of the major issues in ecotoxicology of polar bears. It is assumed that PCBs are the cause. However,  $\Sigma$ MeSO<sub>2</sub>-PCBs are present at 8-10% of  $\Sigma$ PCBs in cubs, and their involvement in possible effects should be considered in any future studies.

Phenolic compounds and neutral POPs were identified and determined in polar bear blood plasma from the Resolute Bay area in the Canadian Arctic and Svalbard (Figure 4-57) (Norstrom, 2000; Sandau *et al.*, 2000). Thirty-five compounds in the phenolic fraction were identified as hydroxy-PCBs (OH-PCBs) in the two polar bear populations. The OH-PCB identity number system follows that of the PCB system with the same chlorine substitution pattern, with addition of the OH-group. Note that the precursor PCBs to the OH-PCBs frequently do not have the same chlorine substitution pattern, since chlorines shift on the ring during metabolism. In addition to OH-PCBs, a previously unidentified phenolic metabolite of OCS, 4-hydroxyheptachlorostyrene (4-OH-HpCS) was identified in polar bear plasma (Sandau *et al.*, 2000). The compound was synthesized, and the binding affinity to human TTR was determined to be about 1.1 times that of T4. Therefore, it is most likely that 4-OH-HpCS is present in polar bear plasma bound to TTR. Traces of pentachlorophenol at approximately 0.2 ng/g ww were also found.

Mean concentrations of  $\Sigma$ OH-PCBs in polar bear plasma ranged from 57 ng/g ww in males from Resolute to 218 ng/g ww in females from Svalbard. Females had significantly higher concentrations of  $\Sigma$ OH-PCBs than males. The ratio of  $\Sigma$ OH-PCBs/ $\Sigma$ PCBs in plasma was also significantly lower in females (mean 1.49) than males (mean 4.08). It appears that females either have a higher binding capacity for OH-PCBs in plasma (i.e. higher TTR concentration) than males, or a higher capacity to form OH-PCBs. The concentration of  $\Sigma$ OH-PCBs was two to three times higher than any other residue class in female polar bear plasma. In the other age groups and males, the concentration of  $\Sigma$ OH-PCBs was equal to, or higher than, the concentration of the next highest residue class,  $\Sigma$ PCBs. The concentration of  $\Sigma$ OH-PCBs in subadults was the same as that in females.

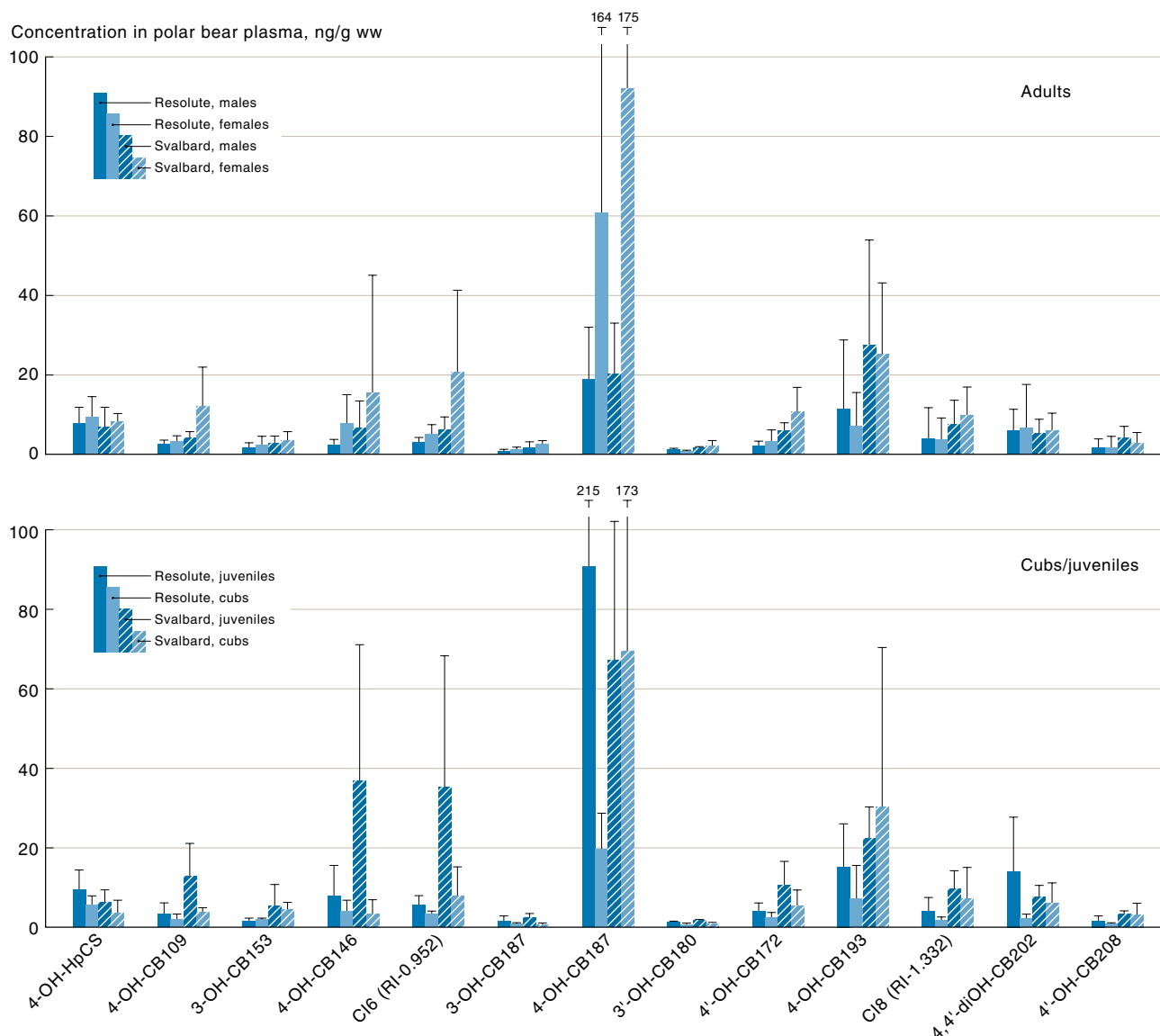


Figure 4.57. Mean ( $\pm$  SD) concentrations of major OH-PCB congeners and OH-HpCS in plasma from polar bears from Resolute Bay in the Canadian Arctic, collected in April-May 1997, and from Svalbard, collected in April-May 1998. Adult males and females,  $n = 12-15$  in each category; cubs (0-2 yr) and juveniles (3-4 yr),  $n = 2-5$  in each category.

Thus, it appears that there is no selective transfer of OH-PCBs in polar bear milk, unlike MeSO<sub>2</sub>-PCBs.

To put the importance of OH-PCBs and other phenolic compounds in polar bear plasma into context, the ranking of concentrations of the first 50 individual POPs, both neutral and phenolic, was determined in the combined Resolute Bay/Svalbard data set. Out of the ten highest POPs in plasma, six are phenolic compounds, three are PCBs, and the last one is oxychlorodane. These six phenolic compounds, including 4-OH-HpCS, constituted 42% by weight of all POPs in polar bear plasma. Of the remaining forty compounds, nine are chlordanes-related compounds, seventeen are PCBs, five are OH-PCBs, and the rest are chlorobenzenes, DDTs, HCHs, and pentachlorophenol (PCP).

In order to determine the potential for bioaccumulation of hydroxy metabolites of PCBs and OCS, patterns of accumulation of these metabolites and their precursors were studied in ringed seal and compared to those in polar bear. Concentrations of  $\Sigma$ OH-PCBs were 1000 times lower in ringed seal than in polar bear plasma, whereas  $\Sigma$ PCBs were only two times lower. Considering that most of the hydroxy metabolites in ringed seal are

probably in blood, and at low concentrations, it would seem that the potential for the polar bear to bioaccumulate OH-PCBs and 4-OH-HpCS is very low.

#### 4.4.8. Arctic fox and sea otter

##### *Arctic fox (Alopex lagopus)*

The Arctic fox is one of the few species that constitutes an important component of both the terrestrial and marine ecosystems (Hiruki and Stirling, 1989). On land, Arctic foxes feed mainly on lemmings, birds, and their eggs, as well as scavenging on caribou remains (Kennedy, 1980; Stickney, 1991; Kapel, 1999) whereas coastal foxes will also eat marine invertebrates (Fay and Stephenson, 1989; Kapel, 1999) and fish (Banfield, 1987) in summer. In winter and spring on the sea ice, Arctic foxes scavenge the remains of ringed seals and bearded seals killed by polar bears (Stirling and Smith, 1977; Andriashek *et al.*, 1985; Fay and Stephenson, 1989; Hiruki and Stirling, 1989; Kapel, 1999), and they will also actively prey on newborn seal pups and eat placental remnants (Smith, 1976; Andriashek *et al.*, 1985; Lydersen and Gjertz, 1986). Populations feeding at dif-

ferent trophic levels (Peterson and Fry, 1987; Hobson and Welch, 1992) and in marine or terrestrial/freshwater environments (reviewed by Hobson *et al.*, 1997) can be distinguished using stable isotope analyses. The  $^{15}\text{N} : ^{14}\text{N}$  isotope ratio, often reported as  $\delta^{15}\text{N}$ , increases in a predictable manner between trophic levels (Kelly, 2000), allowing trophic positions and OC concentrations to be interpreted along a continuous variable. Carbon isotopes ( $^{13}\text{C}/^{12}\text{C}$ ) are not enriched significantly between trophic positions and can be used to evaluate energetic pathways between regions with differing carbon sources. As a result, stable carbon isotope signatures can differentiate an organism's dependence on in-shore/benthic and offshore/pelagic regions (Hobson *et al.*, 1995).

Arctic fox muscle and liver tissues were collected at Barrow, Alaska, (n=18) and Holman, NWT, (n=20) from 1998 to 2000, to elucidate the feeding ecology of this species, and relate these findings to body residue patterns of OC contaminants (Braune *et al.*, 2001d; Hoekstra *et al.*, 2003b) (Annex Table 14). At both sites, PCBs and chlordane-related compounds were the predominant OCs present in muscle, while in liver, chlordanes were the predominant group, followed by PCBs. The most abundant OC analytes extracted from Arctic fox muscle and liver were oxychlordane, the principal metabolite of chlordane-related compounds, CB153 and CB180. While mean concentrations of all major OC groups were less than 250 ng/g ww, a few individuals had very high levels of total PCBs and oxychlordane. The concentrations of OCs were relatively elevated on a lipid basis. For example,  $\Sigma\text{PCBs}$  ranged from 110 to 14 600 ng/g lw in liver and from 76 to 8047 ng/g lw in muscle in the Holman foxes. The mean, lipid-adjusted  $\Sigma\text{PCB}$  concentrations in foxes from Holman and Barrow ( $1853 \pm 730$  ng/g lw and  $1516 \pm 370$  ng/g lw, respectively) are lower than  $\Sigma\text{PCBs}$  in Arctic fox liver from Svalbard, the Norwegian mainland, and Iceland (Norheim, 1978; Wang-Andersen *et al.*, 1993; Skaare, 1996; Klobes *et al.*, 1998b;), which is consistent with the west-east gradient of  $\Sigma\text{PCB}$  concentrations in ringed seals (Muir *et al.*, 2000b). The OC profile in the Arctic fox suggests a similarity in metabolism with the polar bear, and that the capacity of this species to biotransform OCs dramatically influences accumulation profiles.

PCB, DDT, and chlordane levels measured in fat of young Arctic foxes from the Pribilof Islands, Alaska, (Krahn *et al.* 2002) (Annex Table 14) were higher, on a wet weight basis, than liver and muscle concentrations in Canadian foxes from Holman, NWT. On a lipid weight basis, OC concentrations in the foxes from the Pribilof Islands were also slightly higher than concentrations in the Holman foxes (Hoekstra *et al.*, 2003b). TEQs based on mono-*ortho* PCB concentrations range from 3.4 to 146 pg/g ww, with means of 22.7 and 69.5 pg/g ww for females and males, respectively.

Concentrations of several OC groups in both liver and muscle samples were not significantly correlated with increasing trophic position based on stable nitrogen isotope ( $^{15}\text{N}/^{14}\text{N}$ ) values from the Holman and Barrow foxes (Hoekstra *et al.*, 2003b).  $\Sigma\text{HCHs}$  had the strongest positive relationship with trophic position (Figure 4-58). The stable isotope data suggest that as the Hol-

$\Sigma\text{HCH}$  concentration in Arctic fox, ng/g ww

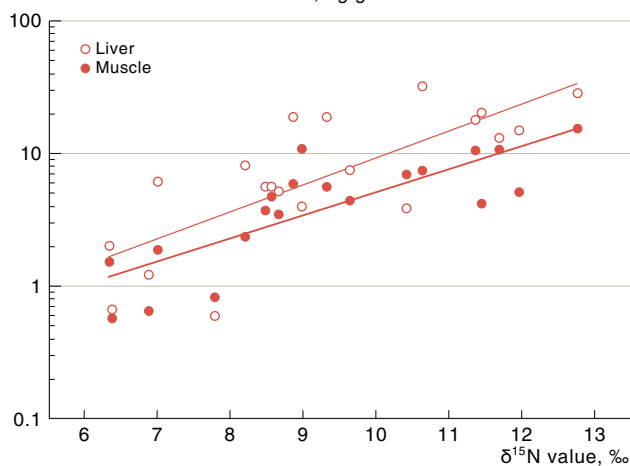


Figure 4-58.  $\delta^{15}\text{N}$  isotope ratio versus  $\Sigma\text{HCH}$  concentrations in liver and muscle tissue of Arctic fox (Hoekstra *et al.*, 2003b). Relationship for muscle and liver samples are:  $\log \Sigma\text{HCH} = 0.175 \delta^{15}\text{N} - 1.04$  ( $r^2 = 0.67$ ), and  $\log \Sigma\text{HCH} = 0.204 \delta^{15}\text{N} - 1.07$  ( $r^2 = 0.59$ ), respectively.

man Arctic fox population shifts its feeding from the terrestrial/freshwater systems to the marine environment, its relative trophic position significantly increases. However, most OC concentrations were not strongly correlated to trophic position. The comparison of  $\delta^{15}\text{N}$  with OC concentrations indicated that trophic position does not serve as an accurate predictor for OC bioaccumulation in the Arctic fox, possibly due to different turnover rates of OCs and stable isotopes in metabolically active tissues, and/or the capacity of the Arctic fox to readily biotransform OCs.

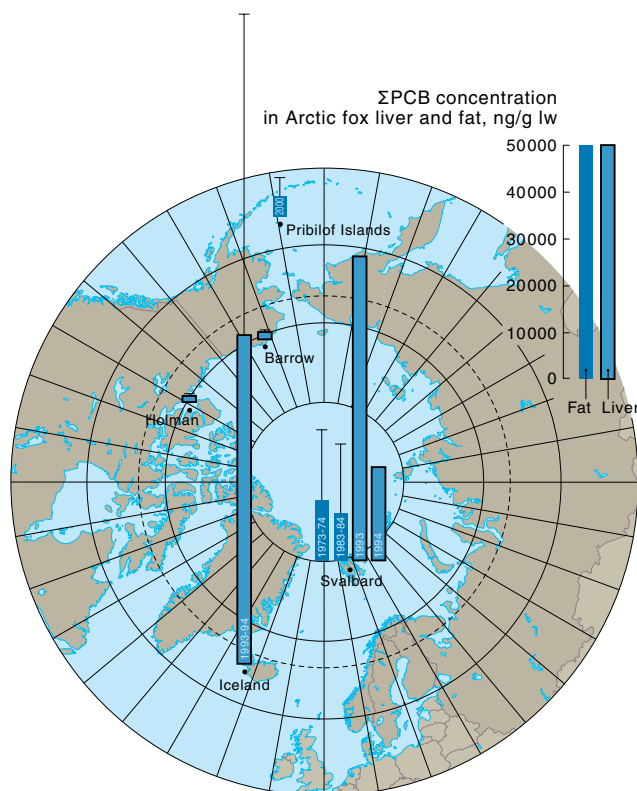


Figure 4-59. Concentrations of  $\Sigma\text{PCBs}$  in Arctic fox liver and fat from Pribilof Islands, Barrow, Holman, Iceland, and Svalbard (previous AAR data).

Concentrations of PCBs and a variety of OC pesticides were measured in the livers of foxes from an inland and coastal region of Iceland (Klobes *et al.*, 1998b) (Annex Table 14). In both regions, the foxes' OC patterns were dominated by oxychlorane. PCB was dominated by CB 180, *p,p'*-DDE was dominant in the DDT group of contaminants, and  $\gamma$ -HCH and  $\beta$ -HCH were the only detectable HCH compounds.  $\Sigma$ PCB levels were much higher in the coastal foxes (approximately 72 500 ng/g lw) than in inland foxes (approximately 1000 ng/g lw). The  $\Sigma$ PCB concentrations in coastal foxes are comparable to concentrations previously seen on Svalbard (20 000–64 000 ng/g lw) (Skaare, 1996). This was explained by the significant differences in trophic levels of the two groups of foxes, where the coastal foxes consume prey items such as piscivorous birds and carcasses of seals, and the inland foxes consume relatively more herbivorous species such as ptarmigan and geese. This is consistent with the findings of Hoekstra *et al.* (2003b) for foxes from Alaska and the western Canadian Arctic.  $\Sigma$ PCB concentrations for all Arctic foxes, including those studied previously on Svalbard, are presented in Figure 4.59.

#### *Sea otter (Enhydra lutris)*

Sea otters are an endangered marine mammal that inhabits the north Pacific Rim. These non-migratory animals are part of the mink family feeding on marine shellfish, and therefore, may be particularly sensitive to the effects of OCs. Members of the mink family have been shown to be highly sensitive to the effects of OCs (Hornshaw *et al.*, 1983). OCs were measured in the liver of sea otters collected in southeastern Alaska, the Aleutian Islands, and California between 1988 and 1992 (Bacon *et al.*, 1999). OC concentrations were much lower in the Alaskan otters compared with the Californian otters with the exception of PCBs in the Aleutian otters (Bacon *et al.*, 1999).  $\Sigma$ PCB levels in the Aleutian otters were 310 ng/g ww, 1.7 and 38 times higher than those measured in the California (180 ng/g ww) and southeastern Alaska otters (8 ng/g ww), respectively. PCDD and PCDF levels were very low in all otter populations. The source of the high levels of PCBs in the Aleutian islands is unknown and remains a concern for the health of the sea otter population in this region. As a non-migratory animal, the source of these high PCBs is likely local (Estes *et al.*, 1997).

#### 4.4.9. Food web studies

A number of studies examining the food web transfer of OCs in marine food webs have been carried out since 1997, filling a knowledge gap identified in the previous AMAP POPs assessment (de March *et al.*, 1998). These studies incorporated a larger number of species and trophic levels than was previously available for a single Arctic marine food web, and also incorporated stable isotopes of nitrogen to discern trophic positions. They provide an advantage over relationships developed for the original AMAP POPs assessment, in that all samples were collected at the same time and in the same region, and the analytical methods were consistent for all samples.

#### *Barents Sea food web study*

To study the bioaccumulation of OCs in the Barents Sea food web, and to examine whether biomagnification could partially explain the high burden of contaminants in top predators near Svalbard (Wang-Andersen *et al.*, 1993; Gabrielsen *et al.*, 1995; Andersen *et al.*, 2001b), selected taxa of zooplankton, fish, and seabirds were collected in the Barents Sea near Bjørnøya in June 1995 (Borgå *et al.*, 2001). OC concentrations increased along the food web in a manner that mainly reflected the organisms' trophic position in the food web, as predicted by previous dietary studies. The concentrations of OCs ( $\Sigma$ HCHs, HCB,  $\Sigma$ CHLs,  $\Sigma$ DDTs, and  $\Sigma$ PCBs) were low in zooplankton and fish, but biomagnified by one to three orders of magnitude in seabirds, with the highest concentrations being found in glaucous gulls. The low OC levels in selected taxa at lower trophic positions were unexpected considering the high concentrations of particularly  $\Sigma$ DDTs and  $\Sigma$ PCBs in top predators. Since a diet of pelagic organisms does not seem to explain the elevated concentrations found in top predators near Svalbard, in this case exemplified by glaucous gulls, a diet of ice-associated fauna may be of importance in the bioaccumulation of OCs instead (Norstrom *et al.*, 1998). As suggested by Pfirman *et al.* (1995), contaminated particles in the sea ice may be released in the melting areas of the Fram Strait and the Barents Sea, followed by bioaccumulation in ice-associated fauna.

#### *Northern Norway food web study*

Biomagnification of OCs was studied in the marine food web of Jarfjord, Norway (69°48'N, 30°25'E) (Ruus *et al.*, 1999). Samples included two species of fish (lesser sandeel (*Ammodytes marinus*) and cod) and seal (harbour/grey seals) collected in 1989 and 1990. Most OCs were found to biomagnify. The trophic level with the greatest biomagnification for  $\Sigma$ DDTs was identified from sandeel to harbour seal. The proportions of highly chlorinated PCBs, DDE and oxychlorane increased with trophic level, whereas PCBs that were mono-*ortho* substituted or *meta-para*-unsubstituted and DDD decreased from fish to seal, reflecting the influence of biotransformation. The study demonstrated the influence of physical-chemical properties and biotransformation on food web dynamics of OCs in Arctic marine food webs.

#### *Northwater Polynya food web study*

OCs and stable isotopes of nitrogen ( $\delta^{15}\text{N}$ ) were measured in zooplankton (six species), a benthic invertebrate (*Anonyx nugax*), Arctic cod, seabirds (six species), and ringed seals collected in 1998 in the Northwater Polynya. The purpose was to examine the effects of biological and chemical factors on trophic transfer of OCs in an Arctic marine food web (Fisk *et al.*, 2001c). The Northwater Polynya is located in northern Baffin Bay and is the largest and most productive polynya (area of open water in ice-covered sea) in the Canadian Arctic. The trophic relationships derived from stable isotope analysis for the Northwater Polynya food web fell into the range expected based on stable isotope results for another Arctic polynya food web (Hobson *et al.*, 1995), with seabirds and ringed seal at the top level, and zooplankton species occupying lower trophic levels. Strong positive relationships were found between recalcitrant



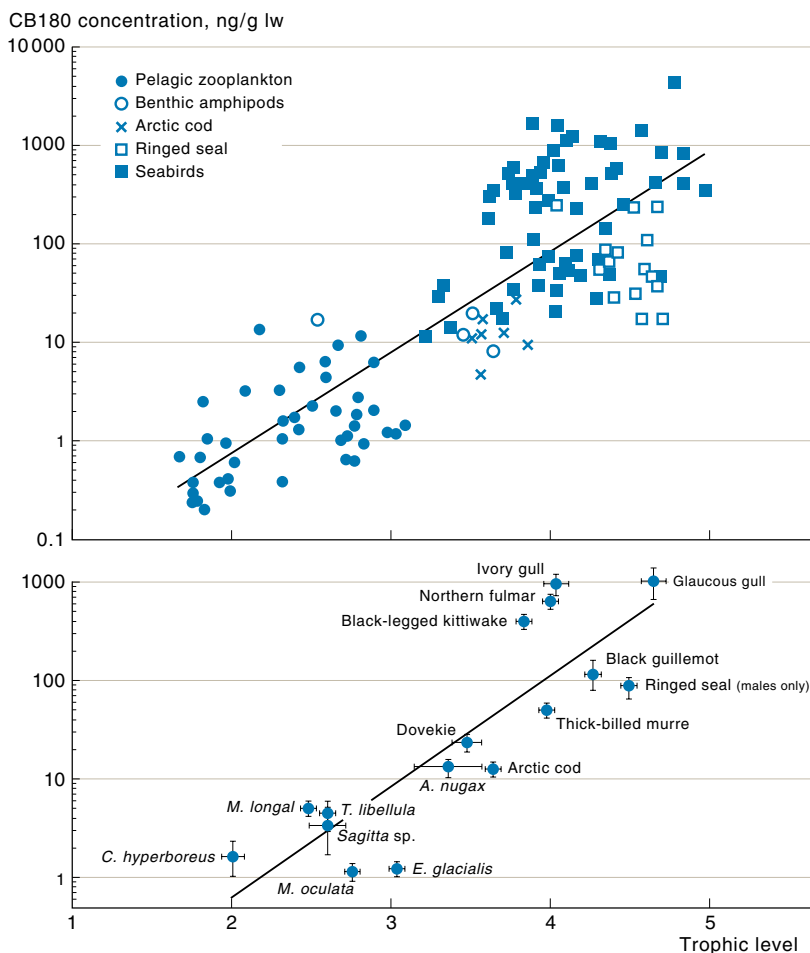


Figure 4.60. CB180 concentrations versus trophic level relationships for the Northwater Polynya marine food web (Fisk *et al.*, 2001c). The top graph shows all data points and the bottom graph shows mean ( $\pm 1$  SE) values for each species. Lines are log-linear regressions. Trophic level is based on  $\delta^{15}\text{N}$ .

OC concentrations (lipid corrected) and trophic level based on stable isotopes of nitrogen, providing clear evidence of OC biomagnification in Arctic marine food webs (Figure 4.60). Food web magnification factors (FWMF), determined from the slope of OC concentration/trophic level relationships in this work, are in good agreement with values obtained for food webs from temperate and Arctic ecosystems involving marine birds and mammals (Norstrom, 1994; Jarman *et al.*, 1996).

#### Biomagnification

Biomagnification factors (BMFs, the ratio of lipid-adjusted concentration in predator to lipid-adjusted concentration in prey) values for the Barents Sea and Northwater Polynya food webs are summarized in Table 4.10. BMFs determined for the Northwater Polynya food web were corrected for trophic level differences based on stable isotopes because many of these species have varied diets, and for many of these comparisons the predator was not a full trophic level above the prey based on  $\delta^{15}\text{N}$  values (Fisk *et al.*, 2001c). Since BMFs reflect concentrations throughout the food web, the lowest BMFs were found in trophic interactions with carnivorous zooplankton (*Parathemisto libellula*) as predators on copepods. The BMFs were higher with fish as predators, and much higher when seabirds were the predators on zooplankton and fish. In amphipods, Arctic cod, and cod, the low OC concentrations and biomagnification factors may be explained by their abilities to excrete contaminants through the respiratory surface into water. The seabirds' higher trophic position and energy requirements, and their lack of direct diffusion of contaminants can ex-

plain the high biomagnification factors in seabirds compared to fish. Generally, PCBs and *p,p'*-DDE had the highest BMFs, followed by HCB and chlordanes. Compared to other compound classes, the BMFs for HCHs were low, reflecting the low  $K_{ow}$  of HCHs. In general, there was good agreement between BMFs for the two studies, although there were some notable exceptions.

BMFs determined for amphipods, using copepods as the prey item, were one to two orders of magnitude higher in the Northwater Polynya food web versus the Barents Sea food web. Fisk *et al.* (2001c) suggested, however, that the Northwater Polynya BMFs for zooplankton were not realistic because concentrations of OCs in zooplankton may be controlled by OC concentrations in water and not prey (Fisk *et al.*, 2001a). In addition, the higher concentrations in the amphipods may be driven by their larger size (Fisk *et al.*, 2002a). Other variables, such as season, may influence invertebrate OC levels, and this requires more research. BMFs for Arctic cod/amphipods were around 1, but are in the range of BMFs reported for similar-sized fish in laboratory experiments (Fisk *et al.*, 1998) and field observations (Rasmussen *et al.*, 1990). BMFs calculated for Northwater Polynya ringed seals (Table 4.10) were slightly lower, but within the range of those reported for male ringed seals from the east central Canadian Arctic (Muir *et al.*, 1988).

The BMFs of seabirds, which used Arctic cod as the prey item, varied between species and food webs (Table 4.10) and appear to be related to scavenging of marine mammals and/or migration to more contaminated regions. The higher BMFs in the Barents Sea food web suggest that the diet of these seabirds includes a larger

Table 4-10. Biomagnification factors determined for key fauna in the marine food webs of the Barents Sea and northern Baffin Bay. Biomagnification factors are based on lipid corrected concentrations. Biomagnification factors for Baffin Bay were corrected to one full trophic level based on trophic levels derived from stable nitrogen isotopes.

Predator/prey	Site	HCB	$\Sigma$ HCHs	$\Sigma$ CHLs	$p',p'$ -DDE	$\Sigma$ PCBs
Amphipod/copepod	Barents Sea	1.5	1.3	1.9	0.4	1.1
	Baffin Bay	3.8	4.5	26.5	16	4.6
Arctic cod/amphipod	Barents Sea	2.0	1.2	3.5	1.7	2.2
	Baffin Bay	6.1	1.1	1.6	3.1	0.9
Thick-billed murre/Arctic cod	Barents Sea	15	1.3	2.6	62	17
	Baffin Bay	10.9	2.1	1.8	19	8.2
Black guillemot/Arctic cod	Barents Sea	8.6	1.1	3.9	40	19
	Baffin Bay	5.0	3.5	4.0	18.5	8.9
Black-legged kittiwake/Arctic cod	Barents Sea	22	1.2	5.9	70	164
	Baffin Bay	21.6	4.2	11.6	56	60.5
Glaucous gull/Arctic cod	Barents Sea	105	6.8	73	2299	1144
	Baffin Bay	6.7	5.2	80	49	28
Ringed seal/Arctic cod	Baffin Bay	0.5	2.0	2.4	7.0	5.5
Food web biomagnification factor (BMF)		4.1	2.7	7.0	14	4.6

percentage of higher trophic-level organisms than does the Northwater Polynya food web. BMFs were highest in black-legged kittiwakes and glaucous gulls, which reflect the migration of the kittiwakes to more contaminated regions and the scavenging of marine mammals and predation on other birds by glaucous gulls. The extremely high BMFs for the Barents Sea glaucous gulls suggest that scavenging and predation may be more prevalent in this population. BMFs reported for Arctic seabirds are in the range reported for other fish-eating birds (Braune and Norstrom, 1989; Hendriks, 1995). Braune and Norstrom (1989) reported whole-body BMFs for a range of OCs, including PCBs in Lake Ontario herring gulls that ranged from 18 to 59 for persistent POPs, and

from 1 to 9 for readily metabolized OCs. These are slightly lower than those calculated for glaucous gulls, which is likely due to scavenging highly contaminated marine mammals by the glaucous gulls and/or from having a higher burden of POPs due to accumulation from more polluted winter habitats, as discussed above.

Results of these studies provided insights into the fate and dynamics of OCs in marine food webs. The OC pattern in the Barents Sea food web changed from zooplankton and fish to seabirds, with decreasing relative contributions of HCHs, HCB, and chlordanes to total OC concentrations, and increasing relative contribution of DDTs, PCBs, persistent compounds, and metabolites (Figure 4-61). This corresponded to previous studies from

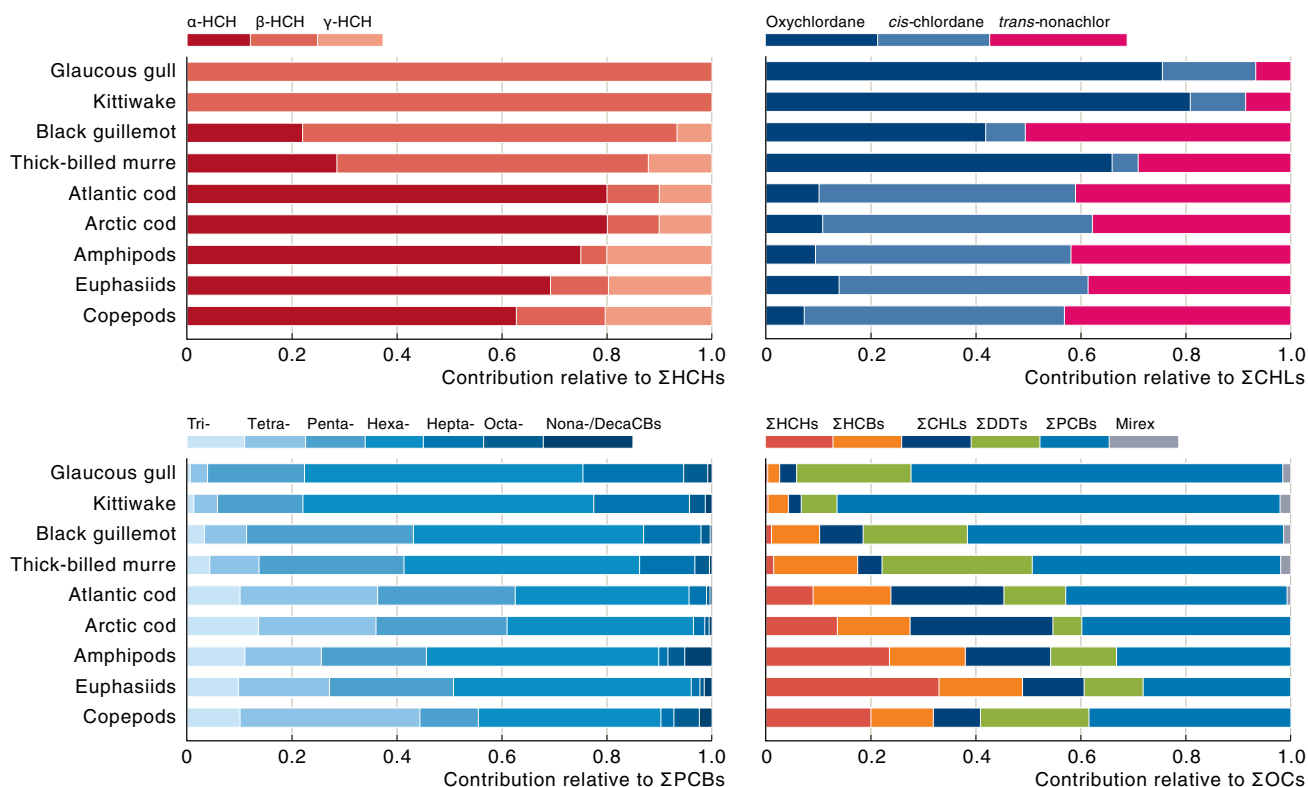


Figure 4-61. Relative proportions of individual OC compounds in the Barents Sea food web (Borgå *et al.*, 2001).

other areas (Tanabe *et al.*, 1984; Hargrave *et al.*, 1992; Ray *et al.*, 1999). The high contribution of compounds from the technical mixture of HCH ( $\alpha$ -HCH and  $\gamma$ -HCH) and chlordanes (*cis*-chlordane and *trans*-nonachlor) to  $\Sigma$ HCHs and  $\Sigma$ CHLs in zooplankton and fish is consistent with limited metabolic capacities of these organisms. In contrast, high relative contributions of the most persistent compounds and metabolites of HCHs ( $\beta$ -HCH) and chlordanes (oxychlordane) in seabirds are consistent with their higher ability to metabolize and excrete the other HCHs and chlordanes. The PCB congener pattern, showing increased influence of higher chlorinated congeners with trophic position of the organism (Figure 4.61), is consistent with the tendency of higher chlorinated congeners to biomagnify, since they are not readily metabolized and excreted (Oliver and Niimi, 1988).

One of the most striking differences in BMFs was between poikilotherms (fish) and homeotherms (seabirds and mammals) (Table 4.10). Large differences in BMFs between poikilotherms and homeotherms were first demonstrated in herring gulls and salmon for Lake Ontario (Braune and Norstrom, 1989). Greater BMFs, and hence, exposure to OCs in homeotherms, has been attributed to their greater energy requirements and feeding rates (Braune and Norstrom, 1989; Fisk *et al.*, 2001c). A similar relationship was also seen in the Jarfjord study, with BMFs for the cod-sandeel predator-prey relationship being lower (2-3 for recalcitrant POPs) than for the seal-cod relationship (8-10 for recalcitrant POPs) (Ruus *et al.*, 1999).

A strong relationship between FWMFs, determined from concentration-trophic level relationships, and  $\log K_{ow}$  was found for recalcitrant OCs (Figure 4.62). It is clear that increasing  $\log K_{ow}$  results in greater trophic transfer of recalcitrant POPs in Arctic marine food webs. This relationship provides insight into the behavior of a

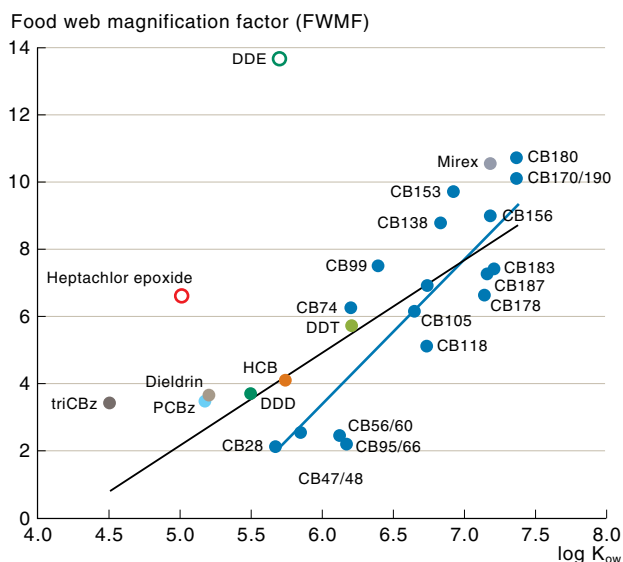


Figure 4.62. Relationship between food web magnification factors (FWMFs) and  $\log K_{ow}$  of recalcitrant POPs in the Northwater Polynya marine food web (Fisk *et al.*, 2001c). FWMFs were determined from the relationship between concentration (on a lipid weight basis) and trophic level.  $\log K_{ows}$  of PCBs are from Hawker and Connell (1988), and for all other POPs are from Mackay *et al.* (2000). Regression lines are shown for PCBs ( $BMF = -22.7 + 4.4 \log K_{ow}$ ,  $r^2 = 0.72$ ) and for 'all' compounds (including PCBs, but excluding DDE and heptachlor epoxide (HE)) ( $BMF = -11.8 + 2.8 \log K_{ow}$ ,  $r^2 = 0.64$ ).

number of POPs. For example, DDE and heptachlor epoxide have values of FWMFs that are much greater than predicted based on the FWMF- $\log K_{ow}$  relationships. DDE has been well established as a metabolite of DDT formed in animals. These results suggest that a large percentage of the high concentrations of DDE in upper trophic-level Arctic organisms are due to metabolic formation. Heptachlor epoxide, which is not in technical mixtures, is formed from heptachlor by photo-oxidation or in the liver of mammals (Buser and Müller, 1993). Results from this study suggest that heptachlor epoxide is formed in upper trophic-level Arctic organisms, and may account for a large percentage of concentrations in these animals.

#### 4.4.9.1. Trophic transfer of 'new' and chiral chemicals in marine food webs

Concentrations of four organohalogens, that are possibly naturally produced (collectively termed HDBPs), 1,1'-dimethyl-3,3',4-tribromo-4,5,5'-trichloro-2,2'-bipyrrrole (DBP-Br<sub>3</sub>Cl<sub>3</sub>), 1,1'-dimethyl-3,3',4,4'-tetrabromo-5,5'-dichloro-2,2'-bipyrrrole (DBP-Br<sub>4</sub>Cl<sub>2</sub>), 1,1'-dimethyl-3,3',4,4',5-pentabromo-5'-chloro-2,2'-bipyrrrole (DBP-Br<sub>5</sub>Cl), and 1,1'-dimethyl-3,3',4,4',5,5'-hexabromo-2,2'-bipyrrrole (DBP-Br<sub>6</sub>), were quantified in the Arctic marine food web of northern Baffin Bay (Tittlemier *et al.*, 2001). The extracts used for this analysis were also used for the OC analysis of the Northwater Polynya food web discussed in Section 4.4.9 but with fewer species. All HDBP congeners were found to significantly biomagnify, or increase in concentration with trophic level in the invertebrate → fish → seabird food web. None of the four HDBP congeners in ringed seals followed the general trend of increasing concentration with trophic level, which was likely due to an ability of the seals to metabolize HDBPs.

Metabolism of chiral contaminants can identify differences in species biotransformation ability and provide insights into the fate of OCs in food webs. Concentrations of HCH isomers ( $\alpha$ ,  $\beta$ , and  $\gamma$ ) and EFs of  $\alpha$ -HCH were determined in the northern Baffin Bay marine food web (Moisey *et al.*, 2001), also discussed in Section 4.4.9. For invertebrates and fish, the BMFs of the three isomers were >1, and the proportion of each isomer and the EFs of  $\alpha$ -HCH were similar to those found in water, suggesting minimal biotransformation. Seabirds appear to readily metabolize  $\gamma$ - and  $\alpha$ -HCH, based on low BMFs for these isomers, high proportions of  $\beta$ -HCH (62-96%), and high EFs (0.65 to 0.97) for  $\alpha$ -HCH. The  $\alpha$ - and  $\beta$ -HCH isomers appear to be recalcitrant in ringed seals based on BMFs >1, with this species having near racemic EFs for  $\alpha$ -HCH. The  $\beta$ -isomer appears to be recalcitrant in all species examined and had an overall FWMF of 3.9. EFs of  $\alpha$ -HCH provided conclusive evidence that biotransformation was accounting for much of the HCH isomer patterns observed in the northern Baffin Bay food web.

#### 4.4.9.2. Trophic transfer of metabolites in polar bear food web

Concentrations of MeSO<sub>2</sub>-PCBs and *p,p'*-DDEs and their precursor PCBs and *p,p'*-DDE were compared in the Arctic cod → ringed seal → polar bear food chain

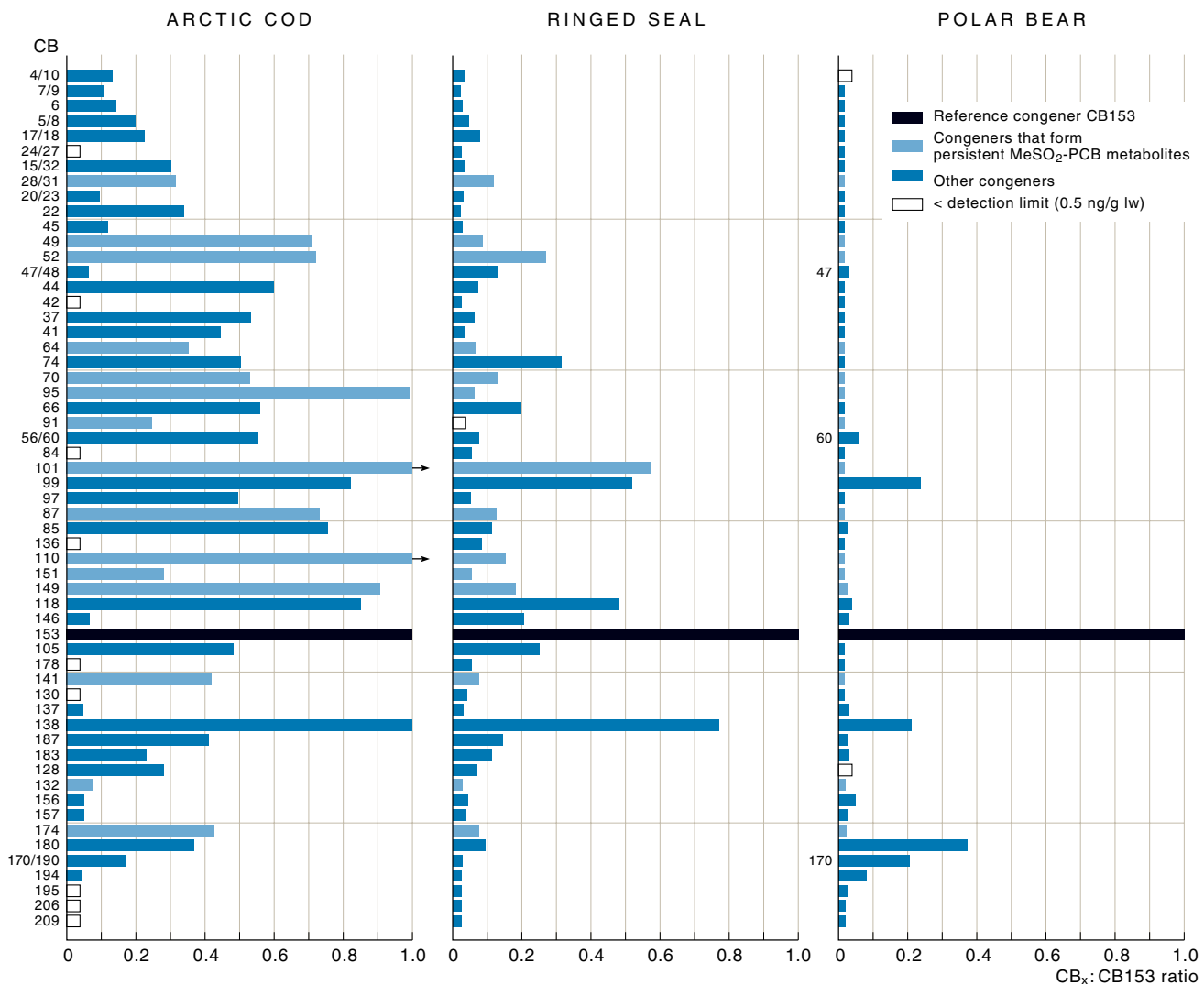


Figure 4-63. Ratios of CB congener concentration to CB153 concentration in whole Arctic cod, ringed seal blubber, and polar bear adipose tissue from the Resolute Bay area in the Canadian Arctic, in 1993 (Letcher *et al.*, 1998).

from Resolute Bay to determine the relative importance of bioaccumulation from the food chain and formation by metabolism of precursors in polar bear (Letcher *et al.*, 1996; Norstrom, 1997).

Overall, there was little difference in the PCB pattern in cod relative to the Aroclor standard (1242:1254:1260, 1:1:1). However, the PCB patterns changed noticeably in seal and bear, especially PCB congeners possessing hydrogens at the *meta-para* (3,4) position on one or more ring (Figure 4-63). *Meta-para* PCBs were present in seal blubber but notably absent in polar bear (<0.05 ng/g lw). These PCBs included CBs 31, 49, 64, 70, 91, 101, 110, 141, 132, and 174, all of which were also present in the form of their 3- and 4-MeSO<sub>2</sub>-PCB metabolites in seal and bear. Metabolites of CBs 52 and 95 were not found in the polar bear. These PCB congeners possess hydrogens at both *meta-para* positions, and may metabolize to *bis*-MeSO<sub>2</sub>-PCBs or to OH-PCBs.

#### 4.4.10. Summary and conclusions – marine environment

The greatest amount of research on OCs in the Arctic since the previous AMAP assessment, post-1996, has been in the marine environment. As might be expected,

circumpolar coverage of information on OCs in the Arctic is greatest in the marine environment. Research has focused on this environment because OC levels are elevated in top predators due to long food chains; the fact that the marine environment accounts for a large percentage of the area of the Arctic; and, the fact that many marine organisms are important components of the human diet.

A number of studies have examined OCs in marine zooplankton, filling a knowledge gap identified in the first AMAP POPs assessment. As expected, levels of OCs are low in marine zooplankton, reflecting their lower trophic levels. Data are available for similar species from Alaskan, Canadian, and European Arctic waters, providing almost circumpolar coverage; very limited data are however available for Russian seas. In addition, there is no information on ‘new’ chemicals in Arctic marine zooplankton, which is necessary to assess the trophic transfer of these chemicals to higher trophic levels. The circumpolar trends of OCs in zooplankton differ from those observed in higher trophic-level organisms, in that there are no large differences between the North American Arctic and the European Arctic. In calanoid copepods from the Barents Sea and northern Baffin Bay, OC levels, with the exception of HCH and CBz, were



also similar. OC concentrations observed in copepods from the Greenland Sea region were lower than those observed in the North American Arctic, which differs from the pattern of levels found in marine mammals from these areas. This could be due to a number of factors related to the season of collection, which varied with these studies. HCH levels were found to be greatest in Alaskan zooplankton, reflecting the proximity to Asian sources of these compounds. Ice-associated fauna from the marginal ice zone near Svalbard have low OC levels, comparable with levels in zooplankton at similar trophic positions from the Barents Sea.

OC levels in benthic invertebrates varied to a greater extent than in zooplankton. Benthic invertebrates that scavenge can accumulate OCs at levels similar to marine fish and lower trophic-level birds. There are currently insufficient data to examine temporal trends in these organisms. Toxaphene was measured in spider crabs collected north of Norway, demonstrating that this contaminant is present in invertebrates throughout the European as well as the North American Arctic. Data for benthic invertebrates are lacking from Russia.

OC data for a number of marine fish species have been generated since the previous AMAP assessment, mainly in Greenland and Canada but also for Arctic cod from a number of regions. Very little new data are available for Russian marine fish. OC concentrations are higher in marine than in freshwater fish, and for some species, are very high. PBDEs have been measured in a small number of species from Greenland and Norway at levels much lower than those of legacy OCs. Levels of OCs in Arctic cod in the Barents Sea were similar to those in northern Baffin Bay and Barrow, Alaska. OC concentrations were found to be very high in the Greenland shark, a large long-lived fish that feeds at a high trophic level.

A fair amount of new data has been produced for Arctic seabirds since the previous AMAP assessment. OC concentrations in seabirds vary dramatically between species, reflecting the large range in trophic levels at which they feed. Highest OC concentrations are observed in the scavenging seabirds, in particular, great skuas, great black-backed gulls, and glaucous gulls, which also prey on other seabirds, but migrating species such as black-legged kittiwakes can also accumulate high OC levels from southern habitats. Circumpolar trends of OCs in seabirds, particularly  $\Sigma$ PCBs and  $\Sigma$ DDTs, show highest levels in the Russian Barents Sea (Franz Josef Land, Novaya Zemlya) and lowest concentrations in the Canadian and Alaskan Arctic, with the exception of HCH. This is similar to what has previously been observed. Data are limited for the central and eastern Russian Arctic. Various combinations of non- and mono-*ortho* PCBs and PCDD/Fs have been determined in several seabird species from the Canadian and Norwegian Arctic, and the Faroe Islands. Highest TEQ levels are found for glaucous gulls on Svalbard. Data for 'new' chemicals, including PCNs, PBDEs, and butyltins, are also available.

There is wide coverage of POP concentration data in a diverse group of pinniped species, both those that are typical Arctic species (ringed, harp, and bearded seals, walrus), and those that are not typically Arctic (harbour, grey, and northern fur seals, Steller sea lions). There is good geographical coverage for ringed seals, but limited

spatial coverage for other Arctic species, some of which play an important role in Inuit diets. As was reported in the previous AMAP assessment, levels of OCs in ringed seals are greatest in the European and western Russian Arctic, in particular the Kara Sea, lower in the Canadian Arctic, and lowest in the Alaskan Arctic and the Chukotka Peninsula of eastern Russia. The exceptions are for chlorobenzene concentrations, which have a uniform circumpolar distribution, and HCH concentrations, which are higher in the Alaskan and western Canadian Arctic. OC concentrations in harp seal are much higher during their molt, when they live off their blubber reserves. Higher PCB concentrations are also indicated in Steller sea lions from the Aleutian Islands compared to the Gulf of Alaska and southeast Alaska. Non- and mono-*ortho* PCB and PCDD/Fs were determined in a few species at a few sites. PCDD/Fs were fairly insignificant contributors to the TEQs compared to the PCBs. Concentrations, as TEQs, were highest in ringed seals from the Kara Sea followed by the Canadian Arctic and Chukotka Peninsula. Northern fur seals had TEQs similar to Canadian ringed seals. Data for 'new' chemicals have also been produced for ringed seals, including PCNs, PBDEs, PFOS, and butyltins. From the limited data available, it appears that PBDE levels are higher in European ringed seals compared with Canadian ringed seals.

Coverage of mysticete whale species has been improved compared with the previous AMAP assessment. In particular, a large amount of data has been produced for minke whales, providing good spatial coverage in the North Atlantic region. Spatial trends of OCs in minke whale are similar to other marine mammals, with the highest OCs levels in the Kara Sea region of Russia and decreasing concentrations to the west and to the south (e.g., the North Sea). PCDD/Fs, non- and mono-*ortho* PCBs and PBDEs have also been analyzed in minke whales around Svalbard. In addition, comprehensive data have been produced for the bowhead whale in Alaskan waters. In general, OC concentrations are lower in the mysticetes (filter-feeding whales) compared to odontocetes (toothed whales). There are limited 'new' chemical data for these organisms.

New data on odontocetes are available, and the beluga is the most widely studied. Data for Russia continues to be limited. There has also been improved coverage of other odontocetes compared to the previous AMAP assessment, but further improvement is still warranted (e.g., for narwhal, which have POP levels as high as in beluga and are also important to native communities). Comparison of OC levels in odontocetes from different regions shows a similar pattern as that seen in ringed seals. Highest levels are found in Svalbard animals, and lowest levels are found in Alaskan animals. HCH levels are again higher in belugas from Alaska. Highest OC concentrations are found in transient killer whales, followed by resident killer whales in Alaska, long-finned pilot whales from the Faroe Islands, and harbour porpoises from northern Norway. PCDD/Fs, non- and mono-*ortho* PCBs have also been analyzed in several species of odontocetes including beluga, harbour porpoise, and killer whale. A reasonable amount of data are available on 'new' chemicals in odontocetes. PCNs, PBDEs and endosulfan sulfate were found in beluga. PBDE levels in these whales follow those found in ringed

seals, with levels higher in and around Svalbard. Highest levels of PBDEs were seen in long-finned pilot whales from the Faroe Islands region. Low concentrations of butyltins were found in Norwegian harbour porpoise and Faroe Islands long-finned pilot whale, but were not detected in Canadian belugas.

A large amount of OC data has been produced for polar bears, including studies on levels in plasma from Russian polar bears. This is an important dataset because it provides the most comprehensive spatial trends coverage that includes Russia. The results clearly indicate that the Russian Arctic around the Kara Sea is the most contaminated marine area of the Arctic, particularly for PCBs and DDTs. Levels decrease to the east and west of the Kara Sea, with the lowest levels seen in Alaska and the western Canadian Arctic. A number of 'new' chemicals have also been measured in polar bears, including PBDEs and PFOS. PFOS levels are high in the polar bear and require more study. There have also been a number of studies that have examined factors that influence OC levels in bears, including reproduction, sex, and fasting. Concentrations of MeSO<sub>2</sub>-PCB and *-p,p'*-DDE metabolites, OH-PCBs and a previously unidentified phenolic metabolite of OCS, 4-hydroxyheptachlorostyrene (4-OH-HpCS), have been determined in polar bears and found to be high.

New data for OCs have been generated for Arctic fox from sites in Alaska, Canada, and Iceland. Concentrations are related to food habits, with highest OC concentrations in foxes that feed in the marine food web. Mean PCB concentrations were lowest in Arctic fox from Canada, Barrow, Alaska, and inland Iceland, and higher in foxes from the Pribilof Islands, Alaska. The highest concentrations were found in foxes from coastal Iceland, and these are comparable to those found previously on Svalbard.

Within Alaska, OC concentrations (except for HCHs) were considerably higher in sea otters from the Aleutian Islands than from southeast Alaska, indicating possible local sources.

High levels of PCBs observed in blue mussels, sea otters, and bald eagles from the Aleutian Archipelago in Alaska appear to be due to local contamination. All of these species are non-migratory and would be exposed to contaminants from the local area. In the case of the bald eagles, high levels were observed in eagles nesting on islands that previously had military facilities. High *p,p'*-DDE levels in the bald eagles are correlated to reduced reproductive productivity. A similar situation has been found at Saglek Bay, Labrador, Canada, where local PCB contamination from a military radar site has led to higher PCB levels in marine sediments, invertebrates, fish, black guillemot, and ringed seals. Local sources of contamination in the Arctic warrant further consideration and monitoring. Steller sea lions from the Aleutian Islands also have higher PCB concentrations in their scat, indicating higher exposure there than in the Gulf of Alaska and southeast Alaska. Whether this is also due to exposure from local sources remains to be determined.

In addition to regional influences, age, sex, and trophic level all play an important role in the accumulation of OCs in marine mammals. Although there are exceptions, most commonly concentrations increase with age in males. In females, changing contaminant levels

with age, and the rate of that change, are more variable and probably depend not only on the female's level of exposure to contaminants, but also how often they successfully produce and how long they nurse their offspring. Lipid content of tissues also plays a significant role in contaminant accumulation, with OCs increasing with the lipid content of the tissue.

Although some new data have become available since the previous AMAP assessment, there is still a lack of contaminant data on marine mammals from the Russian Arctic. This is particularly significant given that available data indicate that the highest levels of many compounds in the marine environment occur in some of the Russian seas.

A number of comprehensive food web studies have been carried out since the previous AMAP POPs assessment. These studies have increased our understanding of how OCs move through marine food webs. The studies also show that OC levels, generally, are similar in lower trophic-level organisms (zooplankton, copepods, and Arctic cod) in northern Baffin Bay and the Barents Sea, with the exception of HCH, which is higher in the North American Arctic. However, in seabirds, ringed seal, and polar bear, there are clear spatial differences, with higher concentrations of DDT and PCB, in particular, in the Barents Sea region as compared to the northern Baffin Bay area. Thus, on its own, a diet of pelagic organisms does not appear to explain the elevated concentrations in top predators in the Barents Sea area. The full explanation for these spatial differences is, therefore, still unclear. All spatial trends that include the Russian Arctic clearly show, however, that PCB and DDT concentrations are highest in the eastern Barents Sea and Kara Sea area, indicating significant local sources of DDTs and PCBs to the Russian Arctic. This is also indicated by high OC inputs from the Ob, Taz, Nadym, Pur, and Yenisey Rivers, which have been shown to be major sources of OCs to the Kara Sea/Arctic Ocean via river water and sediments.

Toxaphene congeners are being measured more often, but there is still a need for a broader inclusion of these compounds in research programs, both as single congener data, and as sum of congeners. Existing data indicate that levels vary significantly between species, but recent data indicate that the highest levels are in the thousands of ng/g range in cetaceans. Toxaphenes have also been shown to be an important contaminant in the European Arctic.

Data for brominated flame retardants, such as PBDEs are not widely available, and are needed, both for individual compounds and sums. The highest reported levels are in the thousands of ng/g range in pilot whales, but otherwise, are in the low ng/g range. Although PBDE concentrations are much lower than for legacy OCs, their rapidly increasing concentrations over time warrant additional research and continued monitoring.

Problems remain with the number of individual components examined within a family of contaminants (e.g., PCBs, chlordanes, DDTs, and toxaphene) with laboratories continuing to report different numbers of compounds, especially in the case of PCBs and chlordanes. This continues to make it a challenge to compare results from studies within the Arctic as well as with those in the temperate and tropical marine environments.

## Temporal Variations in POP Levels

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A critical component of the assessment of POPs in the Arctic is knowledge of the temporal trends of these chemicals in the abiotic and biotic environment. The relationship between contaminant input into the Arctic and the levels and effects seen in wildlife and humans is ultimately tied to the use and restriction of these chemicals and any remedial action that is taken. It is, therefore, important to know if POPs are decreasing or increasing in the Arctic, and whether this varies throughout the Arctic and between various media. Temporal-trend datasets provide a tool to assess remedial actions, and also serve as an early warning system for potential changes in contaminant levels. Most of the Arctic nations have applied restrictions and bans on many of the major OCs for decades, resulting in significant declines of these chemicals in temperate and Arctic regions in the 1970s and 1980s. Continued use of chemicals such as PCBs, and emissions from areas where chemicals are still in use, however, provides a continuing source of contamination. The signing of the Stockholm Convention on Persistent Organic Pollutants in May 2001 (UNEP 2001), which calls for bans on a number of chlorinated pesticides, and reductions of use and emissions of POPs that are by-products, is likely to result in further declines. New concerns have arisen for current-use chemicals such as brominated flame retardants. The use of these chemicals is increasing and, consequently, levels in the environment have been increasing as well.

The first AMAP POPs assessment included a very limited number of good temporal-trend datasets with which to assess long-term trends of OCs in the Arctic. Many of the datasets were confounded by changes in analytical methodologies. For example, much of the older data were generated using packed column gas chromatography rather than current capillary column technology. The best temporal-trend datasets available include the monitoring of OCs in Arctic marine biota dating back to the early 1970s, and the database available on OCs in fish from northern Scandinavia dating back to the late 1960s. These datasets showed a decreasing trend in OCs in the 1980s but a reduction in the rate of decline during the early 1990s.

The assessment of temporal trends, however, is not a simple process and requires datasets from numerous locations and matrices. Regional differences in sources, changes in atmospheric and oceanic currents, and variability associated with biota all can influence OC levels and temporal trends. During the 1990s, weather patterns changed in the northern hemisphere, with an increase in the number of deep storms, and these storms penetrated deeper into the Arctic. The result has been a stronger connection between industrial areas of Europe and North America and the Arctic (Macdonald *et al.*, 2003). This may mean that there has been increased transport of contaminants to the Arctic during the past decade, which would affect temporal-trend monitoring.

The high degree of variability normally observed in OC data in both air and biotic matrices therefore, requires that temporal-trend datasets cover a large number of sampling years and have sufficient sample numbers for robust statistical analysis.

### 5.1. Air and precipitation

#### 5.1.1. Temporal trends in air

Data on sub-decadal (3-6 years) temporal trends of POPs in Arctic air are available from air monitoring stations at Alert (Nunavut, Canada), Stórhöfði (Iceland), Ny-Ålesund (Svalbard, Norway) and Pallas (Finland). Shorter-term (1-3 years) results are also available from Dunai (eastern Russian Arctic) and Amderma (western Russia). Comparisons of levels of major OCs and PAHs among these stations were made in Section 4.1. In this section, temporal trends in air concentrations of PCBs, PAH, and OC pesticides are evaluated and compared. Hung *et al.* (2001; 2002b) examined the temporal trends of PCBs and OC pesticides at Alert using both a temperature-normalization method (where concentrations are expressed as partial pressures and adjusted to a standard temperature (288°K)), and a digital filter method (where concentrations are expressed as partial pressures and short- and long-term variations are extracted using cut-off filters). The estimated half-lives from their work are included in the discussion of each chemical group. Similar time trend analyses are not available for the European Arctic stations with long-term data.

##### 5.1.1.1. OC pesticides

###### *DDT*

Time series for DDT-related compounds in Arctic air are available for Alert (1993-1998), Stórhöfði (1995-1999), Ny-Ålesund (1993-2000), and Pallas (1996-1999). DDT results are also available from Tagish (1993-94) and Dunai (1993) and these full datasets have been discussed previously (Halsall *et al.*, 1998). Since these stations did not operate beyond 1994, no data were generated in Phase II of AMAP. With the exception of Stórhöfði, where there was indication of local sources (Section 4.1.1), the DDT transformation products *p,p'*- and *o,p'*-DDE were mainly found. Figure 5-1 presents the concentration distribution of *p,p'*-DDE over the period 1993-2000. Concentrations at Ny-Ålesund and Stórhöfði were generally higher than at Alert and tended to be lower in 1999 than in earlier years. Indications of seasonal variation can be seen for all sites. The highest concentrations were always found during winter (January/February) and the lowest values were found in summer (August). The most extreme case is at Ny-Ålesund where *p,p'*-DDE concentrations increased 8 to 10 fold above prevailing concentrations in winter and early spring months, especially in 1996, 1997, and 1999. This reflects the association of DDE with parti-

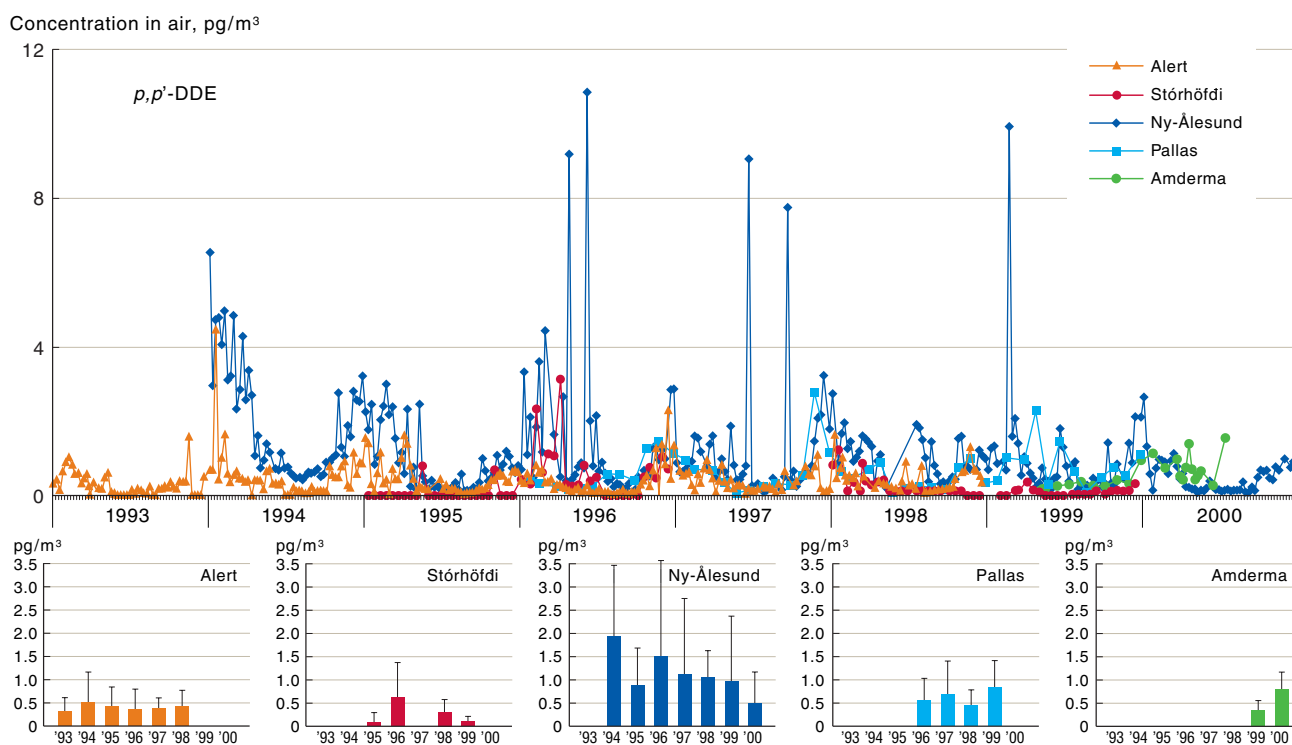


Figure 5-1. Temporal trends of  $p,p'$ -DDE in Arctic ambient air at five monitoring stations from 1993 to 2000.

cles under prevailing winter temperatures and higher particle concentrations during the winter associated with Arctic haze (Halsall *et al.*, 1998).  $p,p'$ -DDE is strongly associated with soils and may be transported with aeolian dusts that are advected into the Arctic (Welch *et al.*, 1991; Hung *et al.*, 2002b). Using temperature-adjusted data, Hung *et al.* (2002b) concluded that

$p,p'$ -DDE is decreasing slowly at Alert (estimated half-life of 19 years). Hung *et al.*, (2002b) also found that  $o,p'$ -DDT actually increased, though not significantly, in concentration at Alert over the period of 1993 to 1998 (Table 5-1). The presence of  $o,p'$ -DDT implies a continued source of technical DDT entering the Arctic atmosphere.

Table 5-1. Half-lives ( $t_{1/2}$ ) of selected OC pesticides and PCBs at Alert (developed using the digital filtration method; Hung *et al.*, 2001) and comparison with the Great Lakes region and western Europe.

OC	Alert, Nunavut		Great Lakes (Eagle Harbor, Lake Superior)		Great Lakes (Sleeping Bear Dunes, Lake Michigan)		Northern U.K. <sup>b</sup>
	$t_{1/2}$ <sup>a</sup> (yr)	$r^2$	$t_{1/2}$ <sup>a</sup> (yr)	$\pm$ SE	$t_{1/2}$ <sup>a</sup> (yr)	$\pm$ SE	
$\alpha$ -HCH	17	0.79	3.0	0.2	3.2	0.4	
$\gamma$ -HCH	4.9	0.88	4.4	0.6	3.4	0.6	
<i>trans</i> -chlordane	8.3	0.48	6.9	2.9	5.2	1.7	
<i>cis</i> -chlordane	4.1	0.71	23	25	9.7	4.9	
<i>trans</i> -nonachlor	6.2	0.71	33	53	6.0	2.1	
Dieldrin	incr.	<0.10	3.7	0.7	2.4	0.4	
$p,p'$ -DDE	19	<0.10	–	–	–	–	
$o,p'$ -DDT	incr.	0.29	–	–	–	–	
PeCA	3.9	0.71	–	–	–	–	
$\alpha$ -endosulfan	incr.	0.25	–	–	–	–	
Tetrachloroveratrole	incr.	<0.10	–	–	–	–	
CB28 (Cl <sub>3</sub> )	12	0.58	–	–	2.7	0.6	1.7-4.2
CB52 (Cl <sub>4</sub> )	3	0.86	–	–	4.3	1.3	1.6-2.3
CB101 (Cl <sub>5</sub> )	11	0.73	–	–	2.9	0.7	2.1-4.3
CB153 (Cl <sub>6</sub> )	17	0.18	–	–	1.8	0.4	2.3-6.6
CB180 (Cl <sub>7</sub> )	4	0.78	–	–	–	–	–

<sup>a</sup>  $t_{1/2}$  = half-life, calculated as  $\ln 2/s$  where  $s$  = slope of the linear regression. For Alert  $s$  = slope of the time trend developed using the digital filtration method. For other sites  $s$  = slope between  $\ln P_{288}$  and  $t$  (where  $t$  = time (years) and  $P_{288}$  = partial pressure of the chemical at 288 Kelvin). Data with increasing time trends are indicated as 'incr.'.  $r^2$  values give an indication of the statistical significance of the relationship of  $\ln P$  vs. time.

<sup>b</sup> Results from Sweetman and Jones (2000).



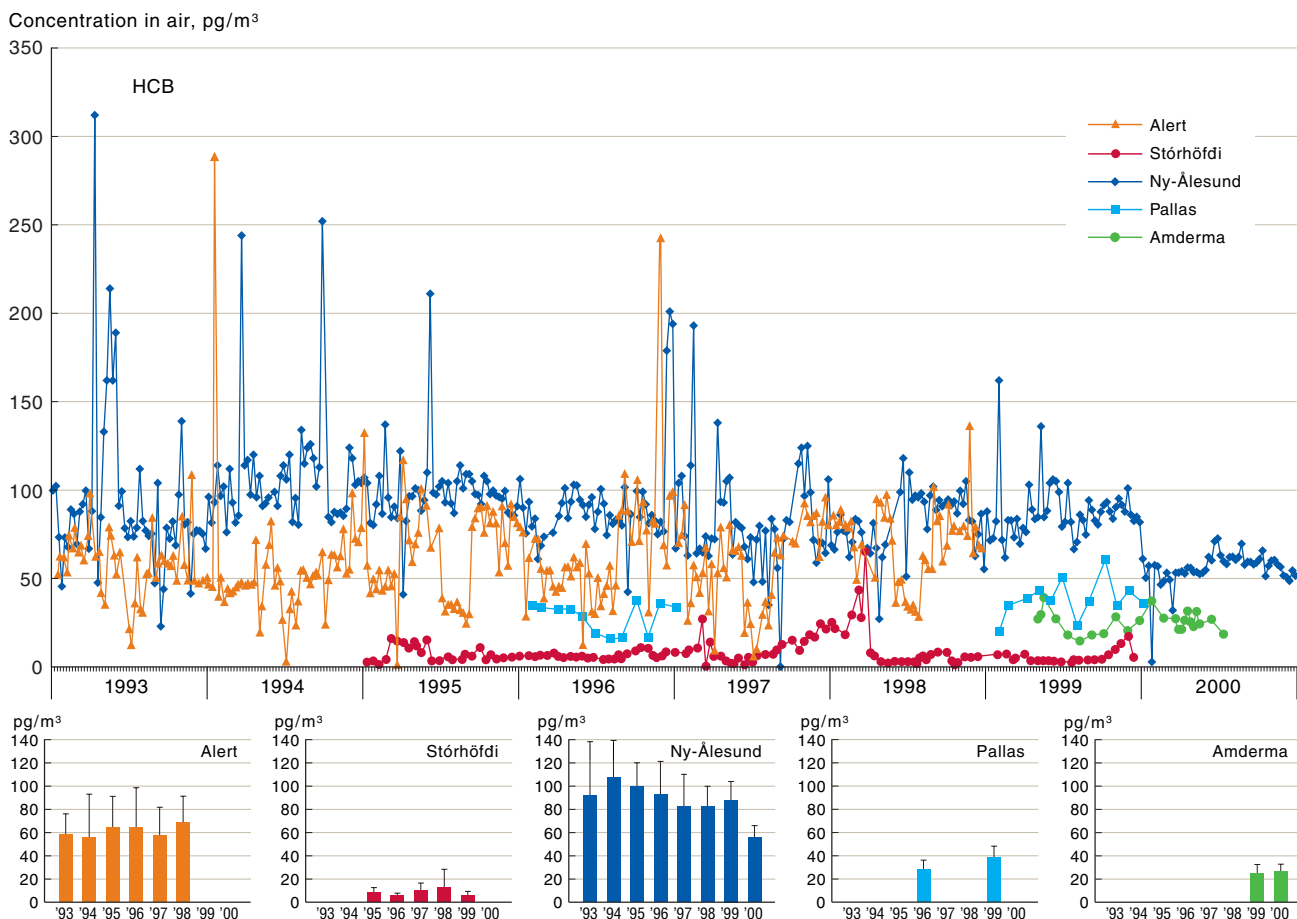


Figure 5-2. Temporal trends of HCB in Arctic ambient air at five monitoring stations from 1993 to 2000.

### HCB

No indication of major trends in concentrations of HCB in the period of 1993 to 1999 were found in the data reported from the three stations with the longest time series (Figure 5-2). HCB was relatively constant with occasionally elevated levels, especially at Ny-Ålesund and Alert. The sources of these elevated HCB concentrations have not been investigated. HCB concentrations show significant 'break-through' of the polyurethane foam collection material even at low temperatures (averaging 25.9%; P. Blanchard, pers. comm.), and thus, the concentrations might be underestimated. In 1997-1999, there were fewer short episodic increases in HCB concentrations at Ny-Ålesund and Alert than in earlier years.

### HCH

The time series for concentrations of  $\alpha$ - and  $\gamma$ -HCH are presented in Figure 5-3. A discernible decrease in concentration of  $\alpha$ -HCH from 1993 to 2000 at Ny-Ålesund and from 1995 to 1999 at Stórhöfði, is apparent from the raw data in Figure 5-3.  $\gamma$ -HCH concentrations show no general tendencies. In all Arctic air samples except those from Stórhöfði,  $\gamma$ -HCH represents about 15-20% of the total  $\alpha$ - and  $\gamma$ -HCH burden. Unlike several other semivolatile OCs (dieldrin, endosulfan, *cis*-chlordane),  $\alpha$ - and  $\gamma$ -HCH concentrations at Alert were not correlated with air temperature (Halsall *et al.*, 1998; Hung *et al.*, 2002b). This lack of correlation reflects the importance of long-range transport events that bring higher concentrations to each site. This is most

evident for  $\gamma$ -HCH in spring-time samples (weeks 14-27) at Ny-Ålesund between 1993 and 1997 and Stórhöfði in 1995-96 (Figure 5-3). A spring increase in  $\gamma$ -HCH was also observed at Alert; however, sharp increases, like those found in springtime at Ny-Ålesund and Stórhöfði were mainly observed in October-December samples at Alert. The reason for this is not clear, although it could be due to agricultural practices in Canada (a major lindane user during the 1990s) where autumn plowing may release residual lindane that was used for seed treatment in oilseed crops. Decreases in  $\gamma$ -HCH at Ny-Ålesund and Stórhöfði in 1999-2000 may reflect reductions and changing use patterns of lindane in Europe. In mid-1998, lindane use ceased in France, the main user in western Europe.

Linear regression analysis of the 'temperature-normalized' or 'digitally filtered' data revealed that  $\alpha$ -HCH and  $\gamma$ -HCH concentrations are declining at Alert (Table 5-1). The decline of  $\gamma$ -HCH was more rapid than that of  $\alpha$ -HCH over the five-year period (1993-1998) (Hung *et al.*, 2002b). Jantunen and Bidleman (1995) have shown  $\alpha$ -HCH to be outgassing from the Arctic Ocean, which will buffer the concentrations in the overlying atmosphere, and may help to explain the long half-life found in this study. On the other hand,  $\gamma$ -HCH in Arctic air has a half-life comparable to that found at temperate sites (Table 5-1). Haugen *et al.* (1998) reported no trend for  $\gamma$ -HCH in air from southern Norway (Lista air station) between 1991 and 1995. This probably reflects the effect of a strong European signal at that site.

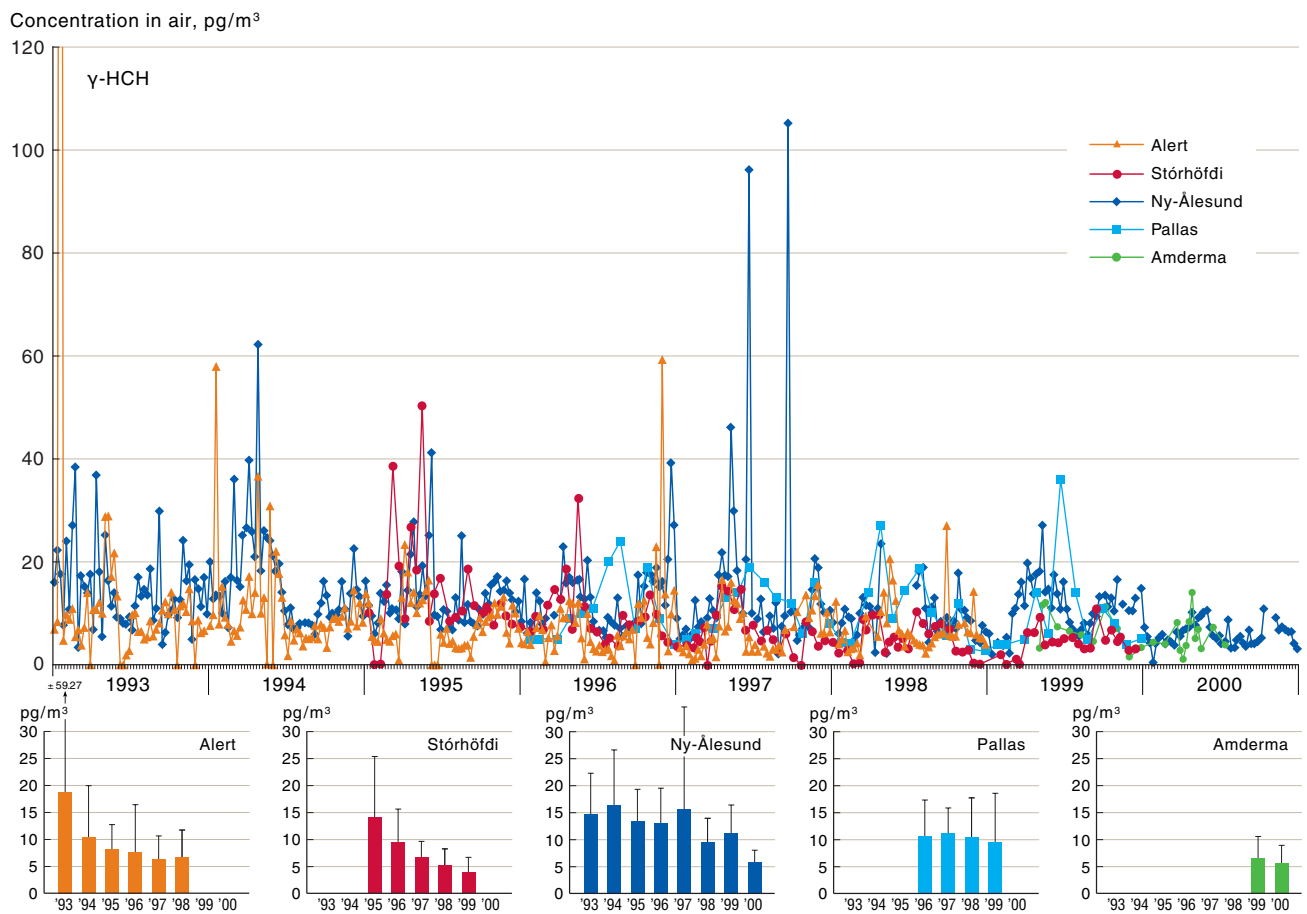
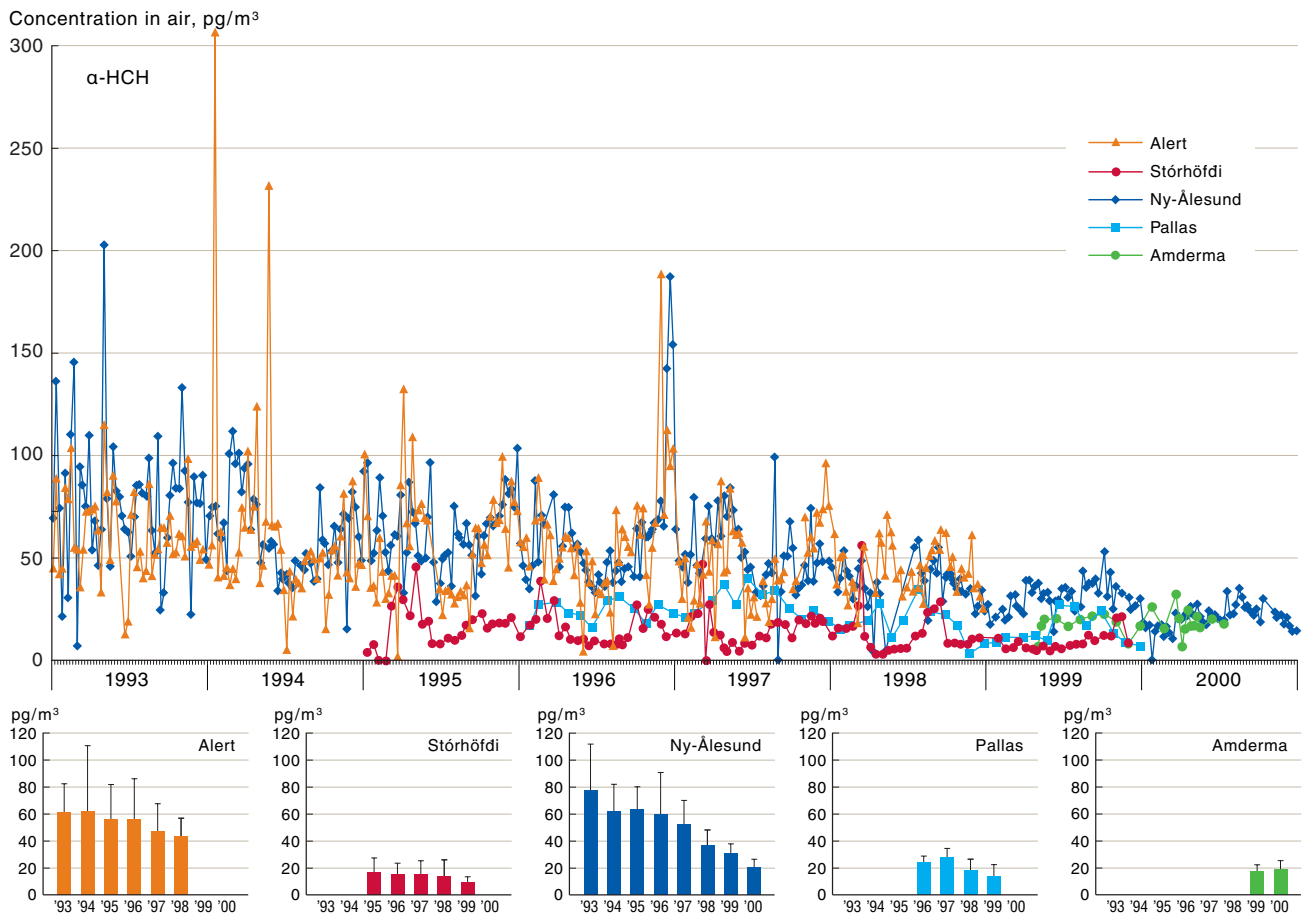


Figure 5-3. Temporal trends of  $\alpha$ - and  $\gamma$ -HCH in Arctic ambient air at five monitoring stations from 1993 to 2000.

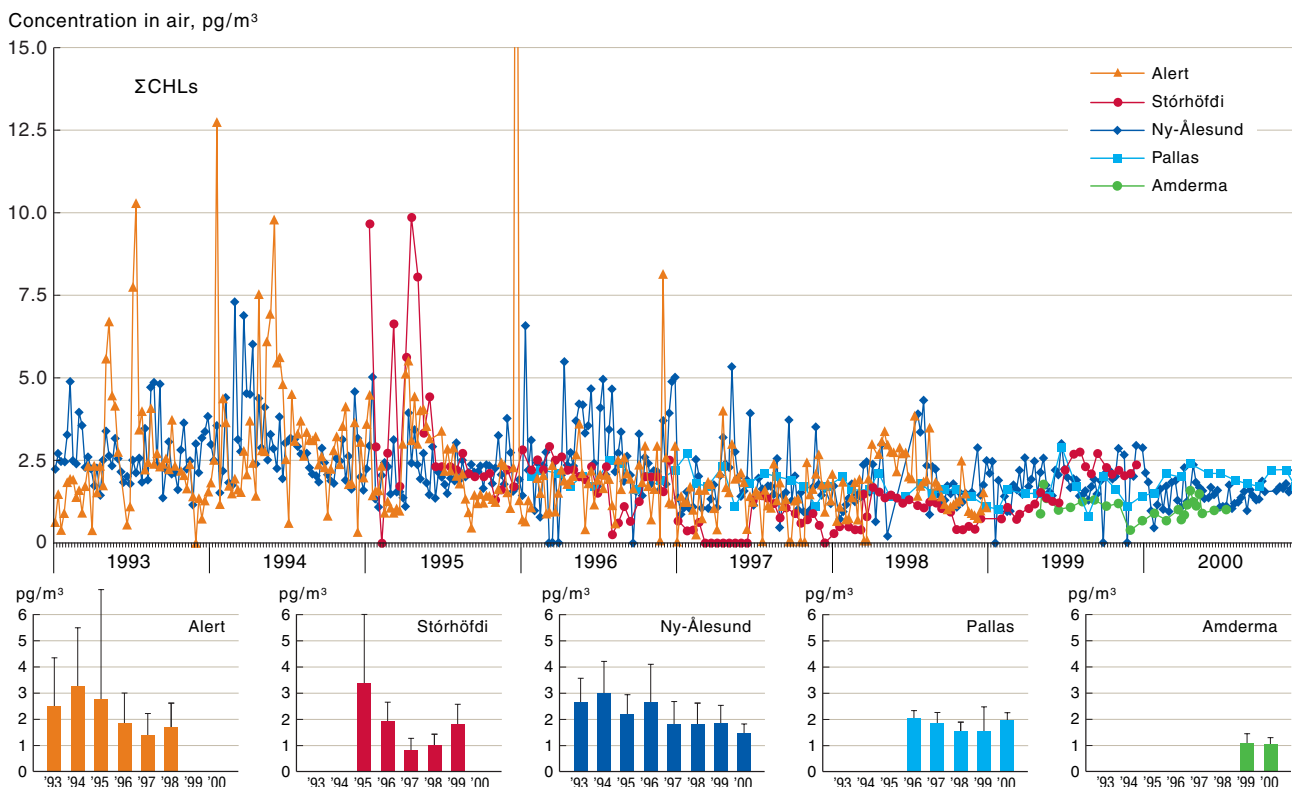


Figure 5-4. Temporal trends of total chlordanes (sum of *cis/trans*-chlordanes, *cis/trans*-nonachlor) in Arctic ambient air from 1993 to 2000.

#### *Cyclodiene pesticides including chlordanes*

Time trends for ΣCHLs and dieldrin are presented in Figures 5-4 and 5-5. For purposes of comparison, ΣCHLs was restricted to *cis/trans*-chlordanes and *cis/trans*-nonachlor. Alert shows relatively high average values of ΣCHLs from 1993-1994 and a subsequent continuous decrease in average concentrations until 1998. The time series of ΣCHLs at Alert, Ny-Ålesund, and Stórhöfði are characterized by sharp weekly episodes during the win-

ter months as well as a general rise in concentrations during summer. This trend was not apparent in northern Finland. These weekly episodes might be due to fresh use of chlordanes-based pesticides (Hung *et al.*, 2002b). Previous episodes of elevated *trans*-chlordanes at Ny-Ålesund were traced to North American sources (Oehme *et al.*, 1995b), which might explain why the seasonal trend is not as apparent at Pallas or Amderma. *Cis/trans*-nonachlor and *cis*-chlordanes were significantly correlated

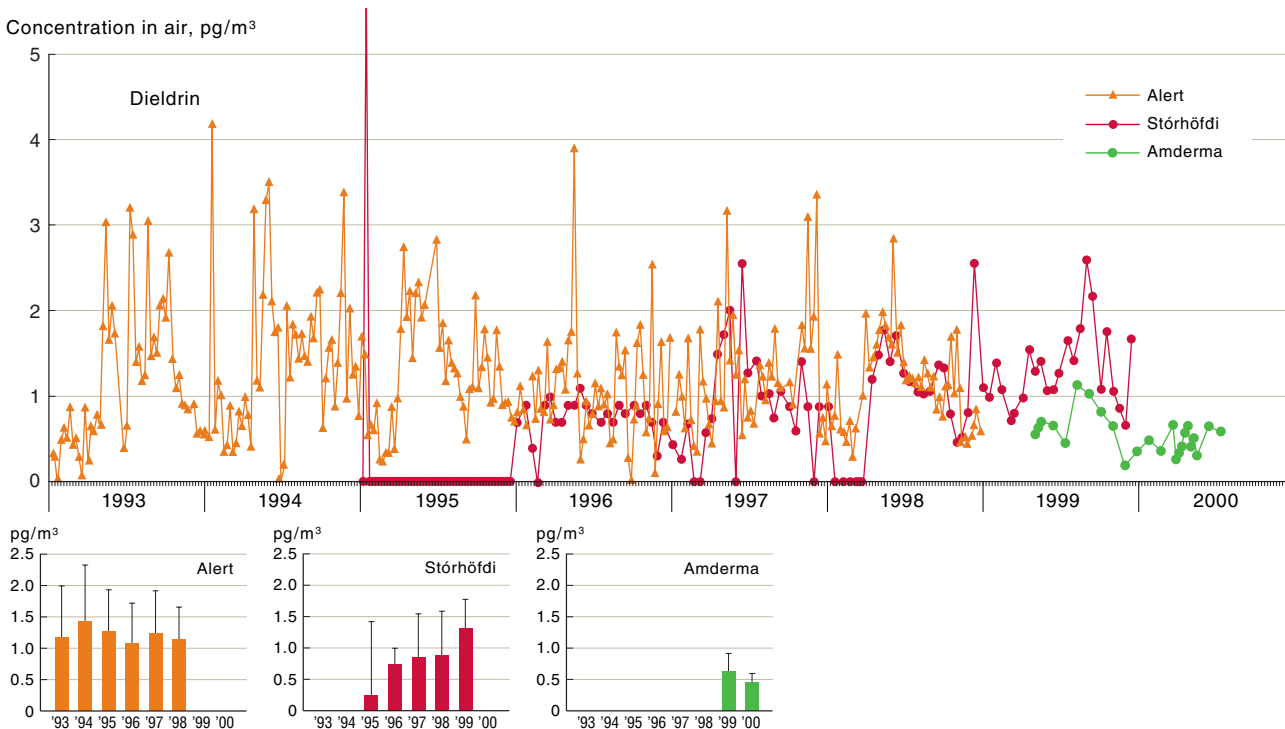


Figure 5-5. Temporal trends of dieldrin in Arctic ambient air from 1993 to 2000.

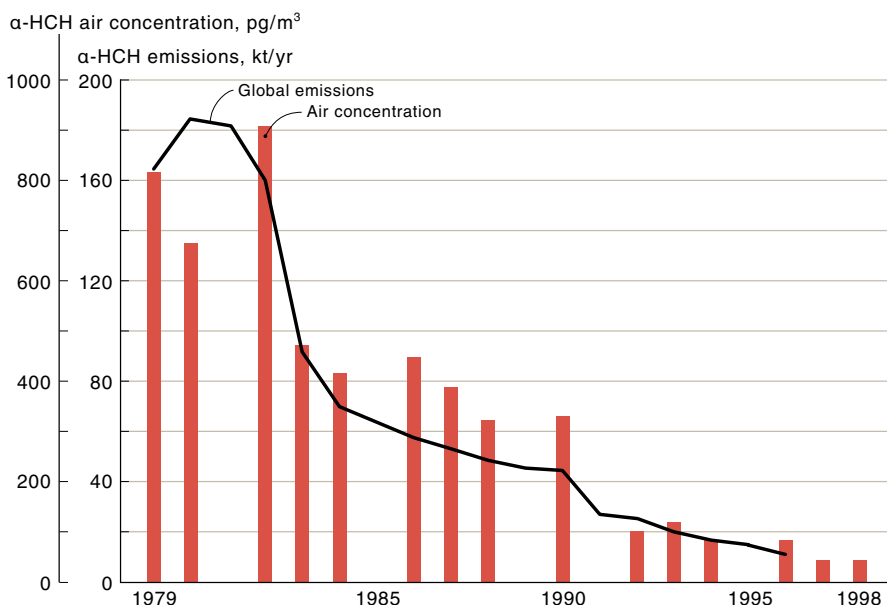


Figure 5-6. Global emissions of  $\alpha$ -HCH and mean concentrations of  $\alpha$ -HCH in Arctic air from 1979 to 1996. Air concentration data for  $\alpha$ -HCH in the Arctic have been measured at different stations by several research groups (i.e. Tanabe and Tatsukawa, 1980; Oehme and Ottar, 1984; Hargrave *et al.*, 1988; Patton *et al.*, 1989; 1991; Hinckley *et al.*, 1991; Oehme, 1991; Iwata *et al.*, 1993; Bidleman *et al.*, 1995; Falconer *et al.*, 1995; Jantunen and Bidleman, 1995; 1996; Oehme *et al.*, 1995c; Pacyna and Oehme, 1998).

with air temperature at Alert (Hung *et al.*, 2002b). Concentrations of  $\Sigma$ CHLs also declined at Stórhöfði until 1997 and then increased in 1999. Using digitally filtered data, Hung *et al.* (2002b) concluded that *cis-trans*-chlordane and *trans*-nonachlor concentrations declined significantly at Alert with half-lives of 4.1, 8.3 and 6.2 years, respectively (Table 5-1). These half-lives were comparable (within a factor of 2) to half-lives calculated for these pesticides at rural sites in the Great Lakes (Table 5-1) (Simcik *et al.*, 1999).

Hung *et al.* (2002b) concluded that dieldrin concentrations did not change significantly from 1993 to 1998 at Alert. Dieldrin concentrations appeared to increase at Stórhöfði from 1995 to 1999 (Figure 5-5). Dieldrin was not reported at the Ny-Ålesund or Pallas stations. Dieldrin concentrations were significantly correlated with air temperature at Alert and typically showed higher concentrations during summer at both Alert and Stórhöfði.

Concentrations of  $\alpha$ -endosulfan at Alert increased over the period of 1993 to 1998 (Hung *et al.*, 2002b). Unlike the more persistent, now banned, OC pesticides discussed above, this is explained by continued use of endosulfan in southern Canada and throughout the U.S. (NCFAP, 2001).

#### Other OC compounds

Tetrachloroveratrole, a by-product of wastewater and wood pulp chlorination also increased in concentration at Alert (Hung *et al.*, 2002b). PeCA declined in concentrations at Alert over the same time period. Time trends of other by-products and current-use pesticides measured at Alert (Section 4.1.1) have not been reported, mainly due to their low atmospheric concentrations, nor have they been studied at other monitoring stations.

#### Other time series for HCH, toxaphene, and chlordane

Li *et al.* (1998a) studied the relationship between the global trends in use of technical-HCH and air concentrations in the Arctic atmosphere by combining data mainly from short-term measurement campaigns in the Bering Sea, western Canadian Arctic and Svalbard from the 1980s and 1990s. Two significant drops in global technical HCH usage were identified, one in 1983 and

another one in 1990. Figure 5-6 shows the long-term trends in global emissions of  $\alpha$ -HCH and its mean air concentrations in the Arctic regions from 1979 to 1998. The trends shown here are similar to those shown by Li *et al.* (1998a), who reported technical HCH emissions. By comparing these two figures, it was found that concentrations of  $\alpha$ -HCH in the Arctic air are more highly correlated with global  $\alpha$ -HCH emissions than with global technical HCH usage (Li *et al.*, 2000; Li and Bidleman, 2003).

Li *et al.* (2001b) also examined the emissions of toxaphene in the U.S. and their influence on levels in Arctic air. Figure 5-7 shows the temporal trends in concentrations of toxaphene in Canadian Arctic air and the estimated emissions of toxaphene from the U.S. from 1985 to 1995. The estimated emissions of toxaphene are, in general, consistent with the concentrations of toxaphene in Canadian Arctic air in the summer. The consistency between toxaphene emissions from agricultural soils in

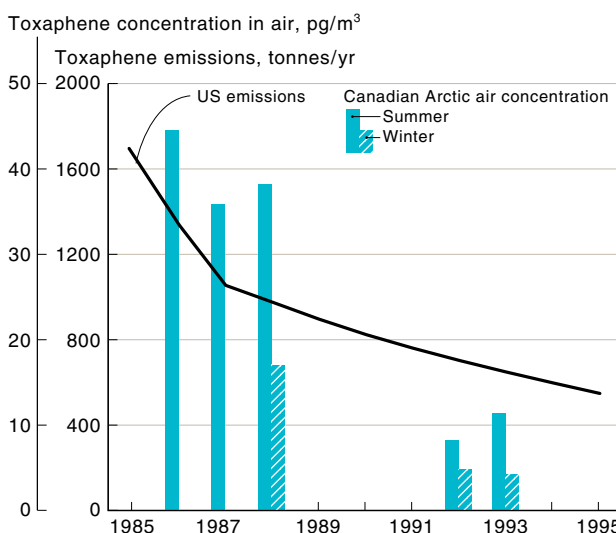


Figure 5-7. Temporal trends in concentrations of toxaphene in Canadian Arctic air and the calculated emissions of toxaphene from the United States from 1985 to 1995. (Sources of air concentration data: data for 1986 and 1987, Patton *et al.* (1989); data for 1988, Patton *et al.* (1991); Hinckley *et al.* (1991); data for 1992, Bidleman *et al.* (1995); Fellin *et al.* (1996); data for 1993, de March *et al.* (1998); Macdonald *et al.* (2000)).



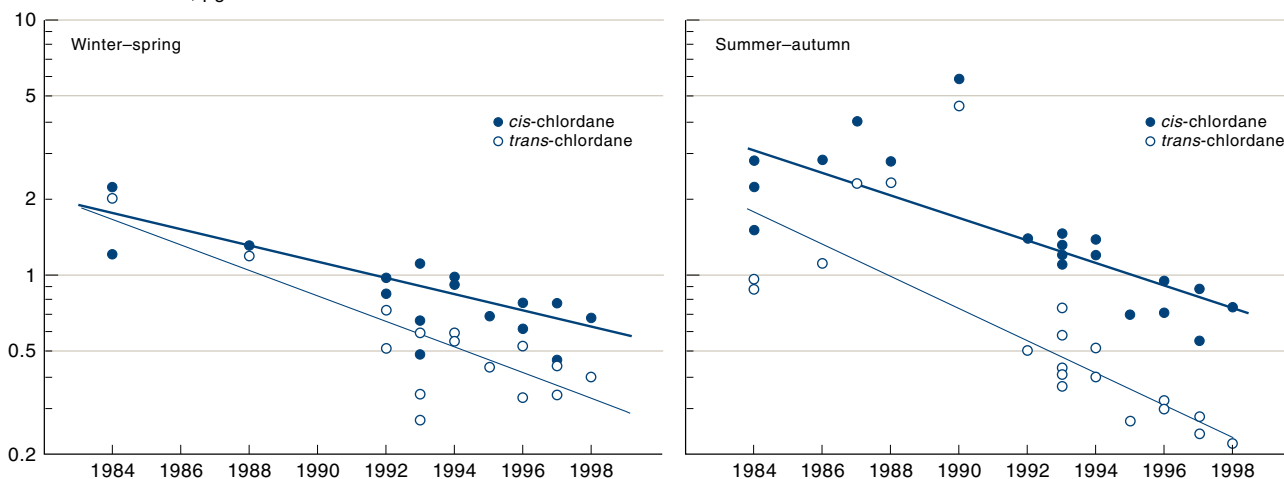
Concentration in air, pg/m<sup>3</sup>

Figure 5-8. Long-term time trends of *trans*- and *cis*-chlordane concentrations in Arctic air during winter-spring and summer-autumn (from Bidleman *et al.*, 2002a).

the U.S. and air concentrations of toxaphene in Canadian Arctic regions illustrates that toxaphene residues in the United States, the southeastern part in particular, could be the major sources of toxaphene in the Canadian Arctic atmosphere. Other sources of toxaphene could include countries in other regions, since toxaphene was still in use in Russia and eastern Europe during the 1980s (HELCOM, 2001). For example, Bidleman *et al.* (1987) found that toxaphene reached southern Sweden during 1984/1985 from eastern Europe and western Russia.

Bidleman *et al.*, (2002a) examined temporal trends of chlordane compounds in Arctic air over a 14-year period 1984-1998. Results for *cis*-/*trans*-chlordane and

*trans*-nonachlor were combined from literature reports from numerous monitoring programs beginning with measurements in Canada (Alert and Mould Bay) and Norway (Ny-Ålesund and Jergul) in 1984. Winter-spring and summer-autumn results were grouped and separated since previous reports had shown changing ratios of *trans*- to *cis*- compounds between these seasons. Significant declines for *cis*- and *trans*-chlordane were found (Figure 5-8) as well as for *trans*-nonachlor (not shown). Half-lives in Arctic air for *cis*- and *trans*-chlordane, and *trans*-nonachlor, were estimated to be 6.4, 9.7 and 6.3 years, respectively, in agreement with estimates of Hung *et al.* (2002b).

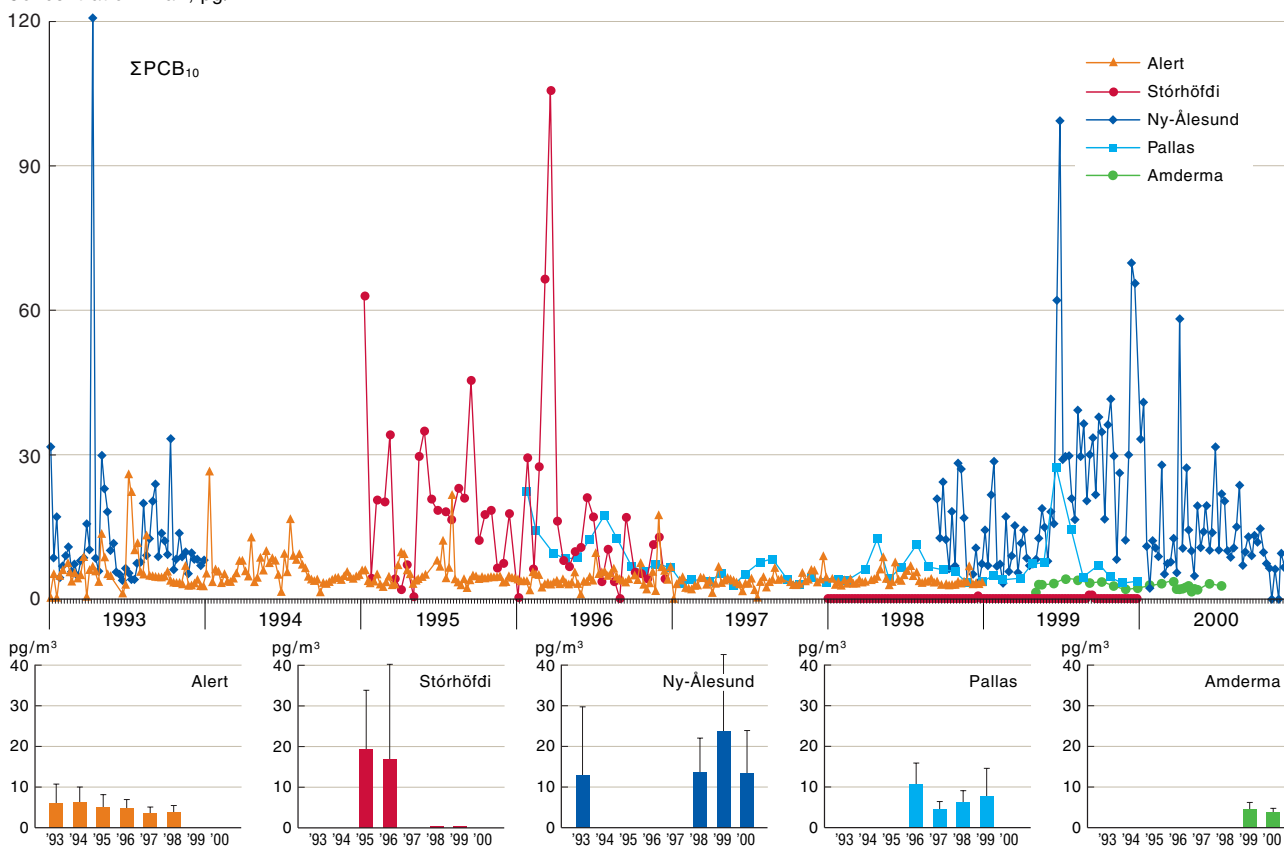
Concentration in air, pg/m<sup>3</sup>

Figure 5-9. Temporal trends of PCB (sum of ten congeners) in Arctic ambient air at five monitoring stations (1993-2000).

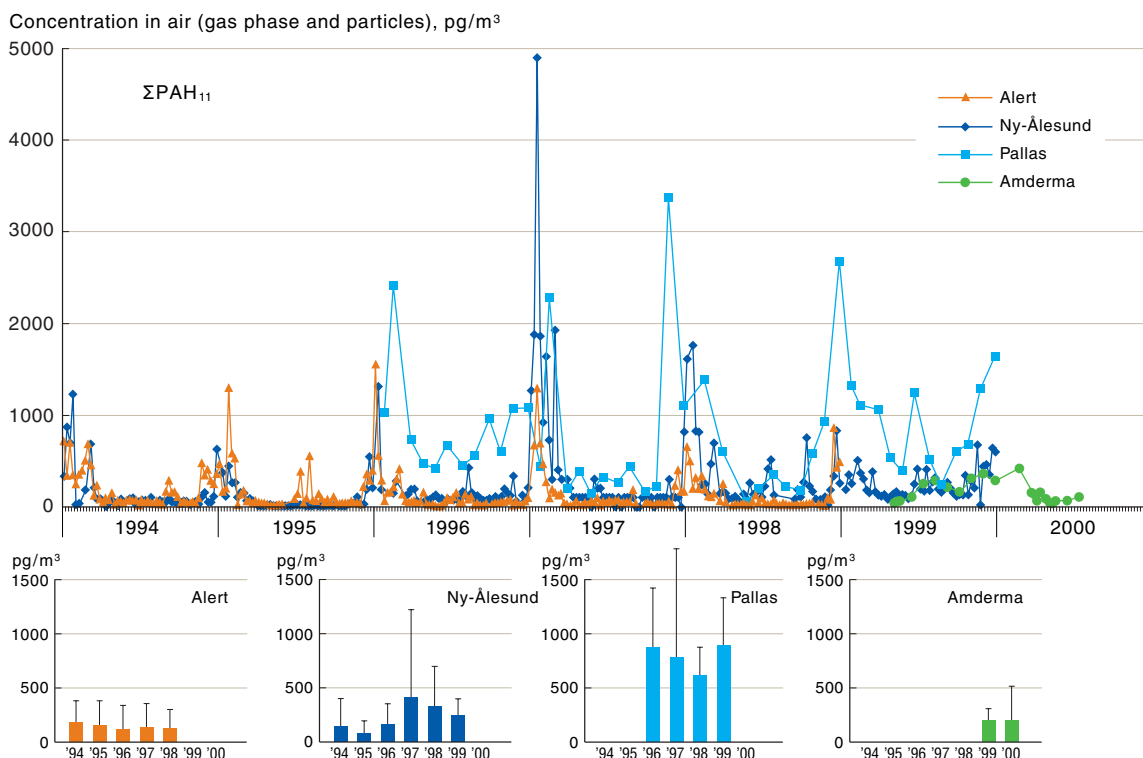


Figure 5-10. Temporal trends of  $\Sigma$ PAHs (11 compounds) in air (gas phase and particles) at Alert, Amderma, Ny-Ålesund, and Pallas.

#### 5.1.1.2. PCBs

The time series for PCBs at five air-monitoring stations is shown in Figure 5-9. A sum of ten PCB congeners was used to represent the time series because the total number of congeners measured at each station varied (see Section 4.1.1). The 1992 data from Alert, as well as data from Ny-Ålesund (1994-97), may reflect some on-site contamination and were therefore not used here or in previous studies of the datasets (Stern *et al.*, 1997; Hung *et al.*, 2001). PCB concentrations tend to be elevated in summer at all stations and, like OC pesticides, show sharp concentration ‘episodes’ in both winter and summer months. No clear downward trend is evident, except for Alert, where a distinct reduction in PCB levels from 1993 to 1998 is apparent. At Alert, penta- and hexachlorobiphenyls were significantly correlated with air temperature, while di- and trichlorobiphenyls were not. The lack of temperature dependence of the less chlorinated congeners suggests that regional/local revolatilization does not have a marked effect on atmospheric levels of the predominant PCBs in the Arctic atmosphere, and emphasizes the importance of long-range transport to the Arctic (Hung *et al.*, 2001).

Hung *et al.* (2001) found declining trends for several of the lower chlorinated congeners in the High Arctic atmosphere at Alert (Table 5-1). The half-lives observed for most of the PCBs at Alert were longer than those observed at a rural site in northern England (Sweetman and Jones, 2000) and at background/rural sites in the Great Lakes (Table 5-1) (Simcik *et al.*, 1999). Haugen *et al.* (1999) did not find an obvious decrease in atmospheric levels of PCBs (1992-1995) from Lista in southern Norway. However, longer time series combined with temperature adjustment would probably be needed to see any such trend. At Alert, CB180 (heptachlorinated)

showed a clear downward trend, with a calculated half-life of only four years (compared to 15 years for CB153). It is therefore possible that heavier congeners might be subjected to different removal processes than the lighter ones, including scavenging by precipitation.

#### 5.1.1.3. PAHs

Data for two or more years were available only for the Alert (1994-1998), Ny-Ålesund (1994-1999), and Pallas (1996-1999) stations and, with missing sampling times, from Amderma (1999-2000). The three longer-term stations determined from 11 to 25 PAH compounds of which anthracene, benzo[a]anthracene, benzo[a]pyrene, benzo[b,j,k]fluoranthenes, benzo[g,h,i]perylene, chrysene, dibenz[a,h]anthracene, fluoranthene, indeno[1,2,3-cd]pyrene, phenanthrene, and pyrene were common to all measurement programs. Trends for  $\Sigma$ PAH<sub>11</sub> at the four stations are shown in Figure 5-10. All sites show an annual cycle of higher concentrations in the winter months, especially during November to February, coinciding with the dark period at these latitudes and with the Arctic haze season. The air sampled at the Ny-Ålesund and Pallas stations is distinguished from Alert by significantly higher PAH concentrations in all seasons, particularly in winter months, over the period of 1996/7 to 1999. Unlike some OCs, no consistent trends can be derived from the PAH data at Ny-Ålesund or Alert. Maximum concentrations generally increased from 1994 to 1997, but were lower in 1998-1999 than in previous years. Ny-Ålesund also had consistently higher ratios of B[a]P:B[e]P, an indicator of reactive versus stable atmospheric PAH, indicating fresher sources than at Alert. B[a]P:B[e]P averaged 2.9 (range <0.1-10) compared with an average of 0.4 (range <0.1-1.7) at Alert. B[a]P concentrations at Ny-Ålesund were highest in 1994 and

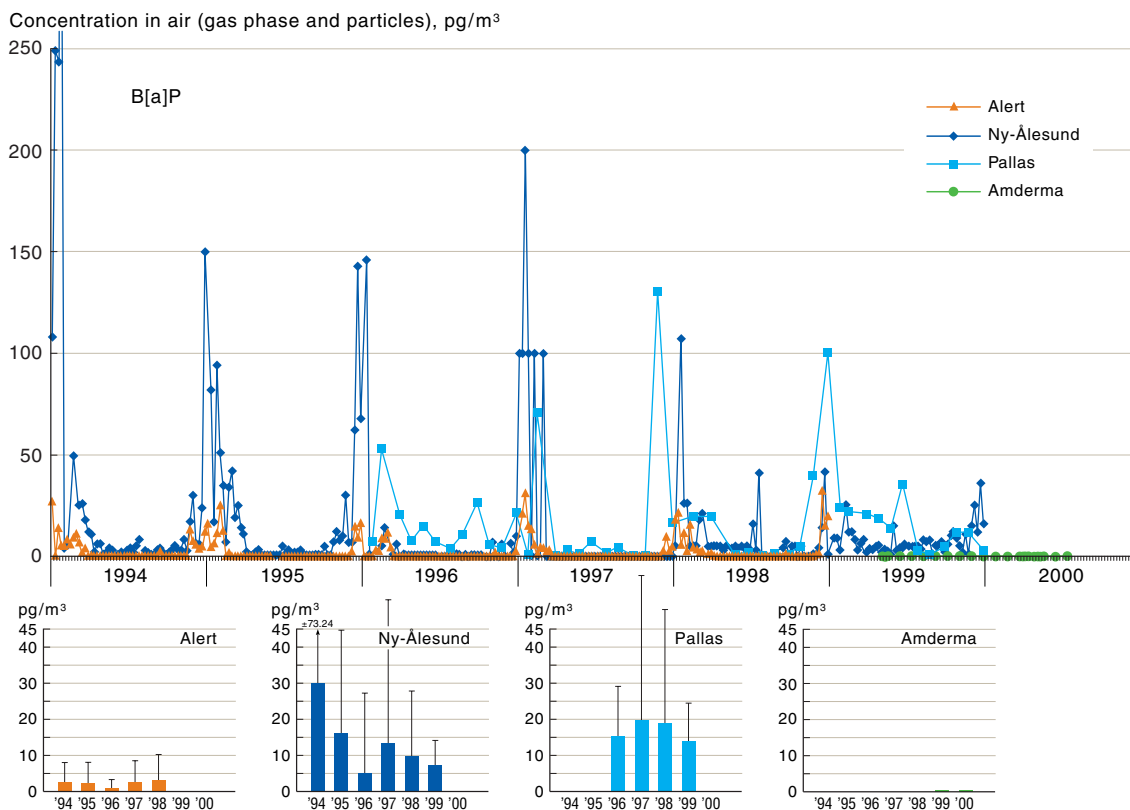


Figure 5-11. Temporal trends of B[a]P in air (gas phase and particles) at Alert (Nunavut), Ny-Ålesund (Svalbard) and Pallas (Northern Finland), and Amderma from 1994 to 2000. B[a]P is predominantly found on particles.

declined during the 1990s. During the summer months, B[a]P concentrations at Ny-Ålesund and Pallas were similar to those seen at Alert. However, much higher winter maxima were observed (Figure 5-11). No investigations of PAH trends similar to those carried out for PCB and OC pesticide data have been made to date.

### 5.1.2. Temporal trends in wet deposition

The time series of bulk deposition of PAHs, PCBs, and HCHs at Oulanka and Pallas in northern Finland during the summers (June to September/October each year) from 1993 to 2001 (Korhonen *et al.*, 1998; 2002) is shown in Figure 4-9. Highest deposition values for ΣPAHs were observed in 1993. Elevated PCB concentrations were found in mid-summer 1994, autumn 1995, and mid-summer 1998. Highest ΣHCH concentrations were observed in mid-summer 1998. Total PCBs in precipitation at Pallas appear to have declined significantly over the period of 1993 to 2001, while no trend was evident for PAHs or ΣHCHs.

### 5.1.3. Temporal trends in snow cores

#### 5.1.3.1. OC pesticides

Snow and ice samples were collected from the Lomonosovfonna Ice Cap, Svalbard, to identify the historic inputs of organic industrial contaminants and pesticides in the European Arctic (Matthews, 2001). The upper 38 m of an ice core, representing approximately the past 70-80 years, were analyzed for OC pesticides, while shorter cores and pit samples were also analyzed for PCBs. Dating results are not complete, so only depth is available for trend evaluation so far. The ice core results show that

the maximum concentrations of all measured contaminants occur below the snow surface, indicating that inputs have declined in recent years (Figure 5-12). The contaminant with the highest concentration at any depth is  $\alpha$ -HCH (12 600 pg/L) followed by  $\gamma$ -HCH (2780 pg/L), and *p,p'*-DDT (2710 pg/L) (Matthews, 2001). These results provide the first detailed picture of deposition of persistent chlorinated organics in the European Arctic. In the only other comparable study, from the Canadian Arctic, Franz *et al.* (1997) reported OC pesticides in snow layers dating from the late 1980s and early 1990s from the snow core on the Agassiz Ice Cap. The same layers were also analyzed for PCBs (Gregor *et al.*, 1995). Concentrations of all major OC pesticides in subsurface layers were about 10 fold lower than in the Lomonosovfonna core. Comparison of samples collected on the surface of the Agassiz Ice Cap with subsurface layers collected one to two years later and dating to the time of original collection at the surface, however, showed a 40% to more than 100% decrease following deposition. The greatest decrease was seen for the more volatile OCs, such as  $\alpha$ -HCH and  $\alpha$ -endosulfan. The loss is thought to be due to changes in surface area as the deposited snow undergoes metamorphosis, which revolatilizes the POPs (Wania, 1997).

The DDT results in the Svalbard study are unusual, both qualitatively and quantitatively. DDT, when manufactured, is 65%-85% *p,p'*-DDT, and the remainder is mainly *o,p'*-DDT with small amounts of other compounds, including *p,p'*-DDE and *p,p'*-DDD. In the environment, DDT normally degrades to either DDE (most common) or DDD. The amount of *p,p'*-DDT present relative to total DDT ranges from 62 to 90%, with an average of 76%. These numbers are very similar to those

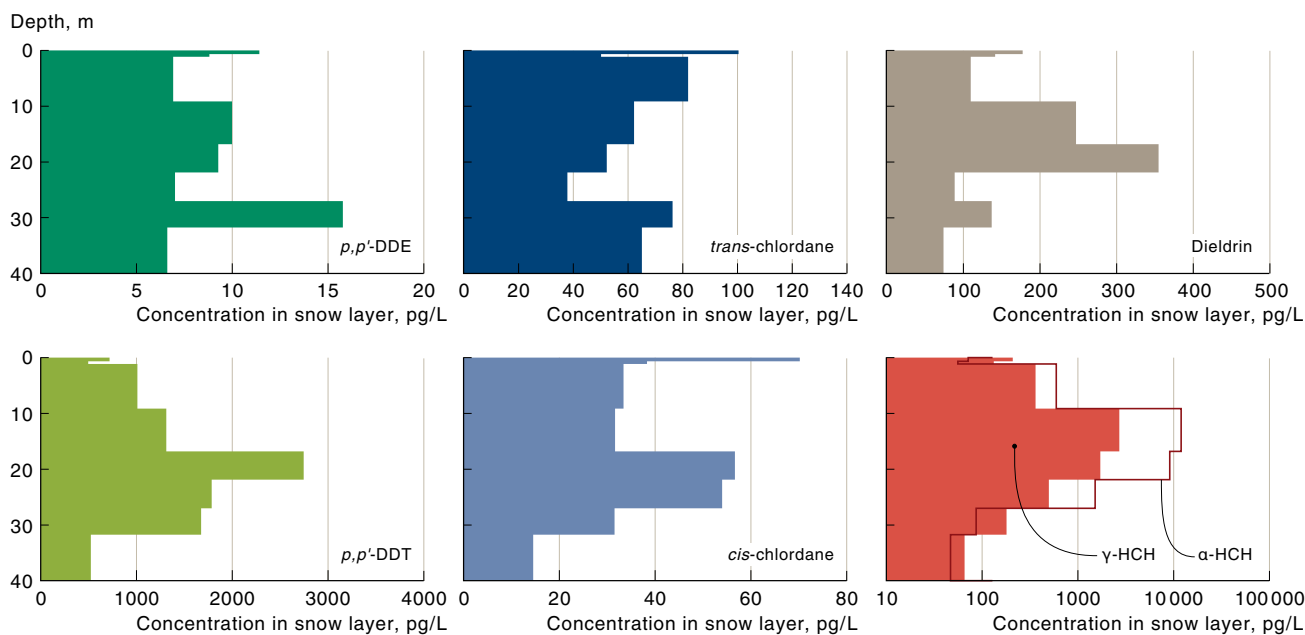


Figure 5-12. OC concentrations in snow cores collected on the Lomonosovfonna glacier in Svalbard (1999). Note: logarithmic scale on HCH graph.

found in the undecomposed technical product (Matthews, 2001). The high concentration and proportion of *p,p'*-DDT in the Lomonosovfonna ice core relative to both degradation products indicate that DDT was transported from its source to Svalbard without degrading, or that the much more volatile DDE and DDD evaporated from the snow surface following deposition. DDT use peaked before or during the 1970s in areas that are the closest source regions to Svalbard such as northwest Russia and Scandinavia (HELCOM, 2001). The proximity of Lomonosovfonna to population centers on Svalbard makes it more susceptible to accumulation of local contaminants. Evidence that sediments from an isolated lake near the coal-mining towns of Barentsburg and Longyearbyen had high PCBs and PAHs (Rose *et al.*, 2003; Section 4.3.2) lends support to this hypothesis.

Subsurface maxima of  $\alpha$ - and  $\gamma$ -HCH were found at Lomonosovfonna, although the maxima were in more recent deposits than maxima for DDT isomers, *cis*-chlordane, and dieldrin.  $\alpha$ - :  $\gamma$ -HCH ratios in deeper samples from Lomonosovfonna suggest some losses of  $\alpha$ -HCH, but in more recent samples, the pattern is characteristic of observations in other parts of the world. Samples covering the last 20 years show that the ratio has been decreasing, which corresponds to the phasing-out of  $\alpha$ -HCH-containing technical products and increasing use of lindane. Technical HCH, consisting of about 80%  $\alpha$ -HCH and about 15%  $\gamma$ -HCH was the major form of HCH released into the environment prior to 1980 (Li *et al.*, 1998a). The HCH isomers were the predominant OCs measured in snow at Svalbard, which is somewhat unusual because they are also the most volatile. Their high volatility has led other snow investigators to suspect high amounts of evaporation of HCH isomers following deposition to the snow surface (Wania *et al.*, 1999d). If true, the original concentrations of these compounds at Svalbard would have been even greater. Lomonosovfonna concentrations near the surface are similar to lake and river water concentrations throughout much of the Arctic. The highest concentrations are similar to the Yenisey River in Russia in 1993, one of the

most contaminated Arctic rivers (de March *et al.*, 1998). Overall, these HCH trends are consistent with what has been observed elsewhere in the Arctic in air, where higher concentrations of  $\alpha$ -HCH were seen in the 1980s (Li *et al.*, 1998a).

Dieldrin concentrations are much lower than  $\alpha$ - and  $\gamma$ -HCH or DDT (Figure 5-12), the subsurface maxima coinciding with that of *p,p'*-DDT. The results suggest a steady decline in deposition of dieldrin at Svalbard, probably since the mid-1970s.

Total chlordane was present at similar concentrations to dieldrin, but did not show clear subsurface maxima. Chlordane has two major isomers, *cis*-chlordane and *trans*-chlordane. The latter is less stable, being subject to photolysis in the atmosphere and decomposition in sunlight. The depth profile of the *cis*- : *trans*-chlordane ratio was consistently less than 1, and thus did not indicate large-scale degradation of *trans*-chlordane.

#### 5.1.3.2. PAHs

Masclat *et al.* (2000) reported a high-resolution historical profile of PAHs on particulate filtered from snow/ice cores collected in 1993 at Summit, Greenland. A strong seasonal variation was observed, with the highest PAH concentrations in winter, except for retene (Figure 5-13, next page). Retene deposits in snow dated to spring/summer 1991, corresponded to intense fires in the boreal forests of Canada and Siberia. PAH deposition correlated strongly with black carbon showing the same seasonal variation. Black carbon has the same source as PAHs, from anthropogenic and natural combustion, and its high correlation with PAH suggests a common source region. Total PAHs in the snow particulate were not strongly correlated with sulfate, another indicator of combustion (e.g., coal burning). However, maxima of fluoranthene and pyrene concentrations coincided with elevated sulfate in four strata indicating common sources. Since only the insoluble phase of the snow was analyzed for PAH, the lack of complete correspondence with sulfate is not surprising. Jaffrezo *et al.* (1994) found good



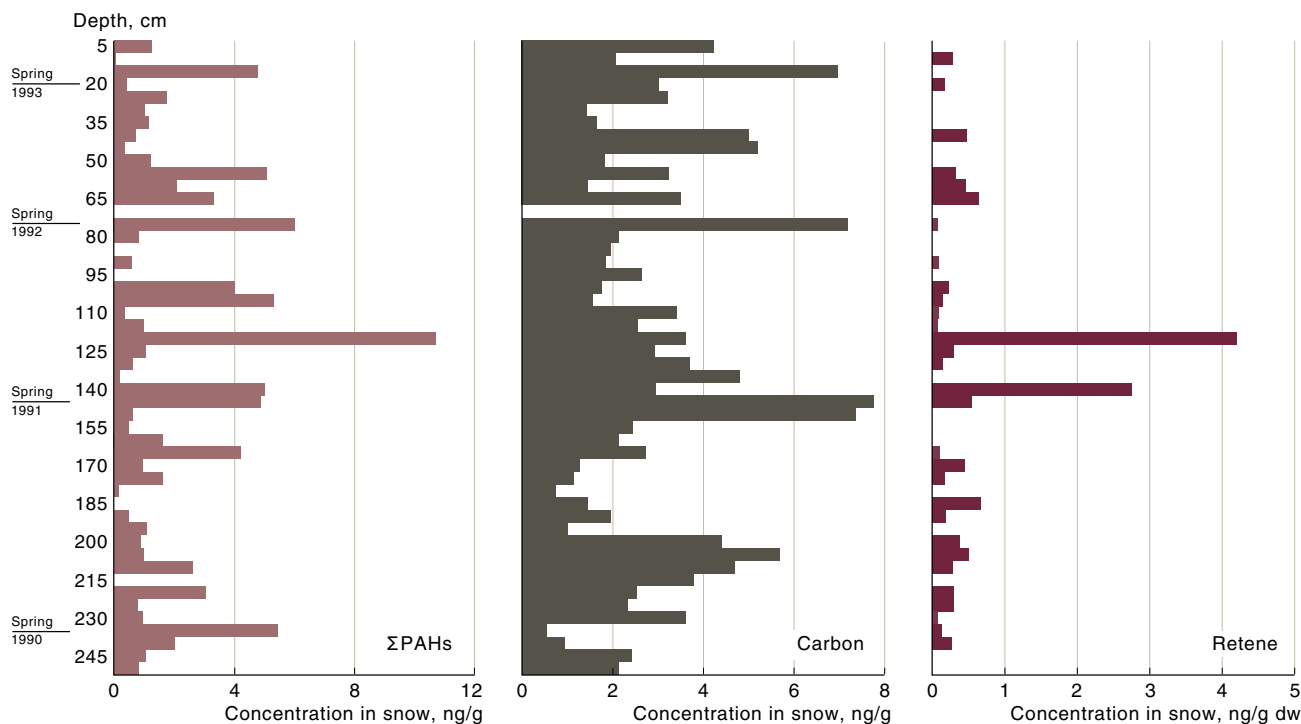


Figure 5-13. PAH concentrations in a snow core from Summit, Greenland, showing the relationship to black carbon and retene (an alkylated PAH generated in forest fires). Spring time each year is inferred from maximum deposits of calcium. Results adapted from Masclet *et al.* (2000).

correspondence between sulfate and  $\Sigma$ PAHs in a snow core from Summit that included the analysis of dissolved and particulate phases. These authors also noted post-depositional declines in concentrations of phenanthrene, fluoranthene, and B[a]P in the snow pack over a four-year period. In the case of B[a]P, possible degradation by OH radicals produced on the ice and snow surfaces was suggested as the main loss pathway. In the case of phenanthrene and fluoranthene, volatilization from snow as it undergoes metamorphosis may also occur (Jaffrezo *et al.*, 1994).

#### 5.1.4. Conclusions on temporal trends in air and precipitation

A sufficiently large dataset now exists to examine temporal trends for a number of POPs in Arctic air. Unfortunately, the detailed analysis of the data, which requires adjustment for temperature for comparison among sites and with temperate latitude, has been conducted only for data from Alert. The trends appear to be similar at Ny-Ålesund, but further study is required.

Comparisons of trends in the Arctic with temporal trends at rural sites in the Great Lakes show that, after temperature adjustment, half-lives of most OC pesticides and PCBs are similar overall, considering the associated margin of error, although interesting differences have been observed. For example, the half-life of  $\alpha$ -HCH at Alert is five times longer than at two sites in the Great Lakes. Outgassing of  $\alpha$ -HCH from the Arctic Ocean will buffer the concentrations in the overlying atmosphere at Alert. This process could also explain the apparent lack of decline of other OCs with similar air-water fugacity ratios (e.g., HCB, lower chlorinated PCBs, endosulfan).

The estimated half-lives of tri- to heptachlorobiphenyl congeners at Alert were typically longer than those

from rural Great Lakes locations. This might be due to lower temperatures encountered in the Arctic, coupled with winter darkness, which will slow both biotic and abiotic degradation relative to temperate regions. In theory, global fractionation (Wania and Mackay, 1995), a process whereby chemicals may be latitudinally fractionated according to ambient temperature and physical-chemical properties, will have the effect of maintaining Arctic air concentrations, while levels in temperate regions are showing apparent reductions. In support of this, in northwest England at a site much closer to major urban sources in the U.K., a suite of tri- to heptachlorobiphenyl congeners have half-lives of 2-6 years, which is shorter than those at Alert. The high week-to-week variation of air concentrations at Alert, Ny-Ålesund, and other sites, and the lack of a strong relationship with temperature indicates that, for the Arctic in general, long-range transport is having a marked influence on atmospheric levels, accounting for the large degree of scatter in the temporal database. This uncertainty will probably be reduced with longer-term datasets.

The unique, long-term datasets for  $\alpha$ -HCH, toxaphene, and chlordane demonstrate that the atmosphere plays a significant role in the global distribution of both compounds and responds rapidly to changes in emissions. Atmospheric long-range transport provides rapid dispersion of these OC pesticides from their areas of emission (primarily China in the case of HCH, and southern U.S. in the case of toxaphene and chlordane) into the Arctic. The results further imply that a reasonable estimate of historical air concentrations of HCH isomers and toxaphene in the western Canadian Arctic, and possibly in the Arctic as a whole, can be inferred from the global emission data, or emission data for the major-use area.

The historical trends of OC pesticides in a snow core from Svalbard provide the first insights into the histori-

cal inputs to the European Arctic. Unfortunately, direct comparisons with previous snow core work on the Agassiz Ice Cap on Ellesmere Island (de March *et al.*, 1998; Macdonald *et al.*, 2000) are not possible because that core was analyzed only for PCBs (Gregor *et al.*, 1995) and PAHs (Peters *et al.*, 1995). The results for DDT at Svalbard suggest a local source, possibly the mining towns within 50 km of the glacier. The dates of deposition are also unknown at present. However, maximum deposition of DDT around 1970 suggests that maximum deposition of HCH isomers and dieldrin was reached post-1970 (Matthews, 2001). For HCH, this is consistent with use in the northern hemisphere which peaked in the 1980s based on concentrations in Arctic air (Macdonald *et al.*, 2000) and global use patterns (Li *et al.*, 1998a; Li, 1999b). In the case of PAHs in snow cores at Summit, Greenland, and the Agassiz Ice Cap, there is a clear linkage to combustion sources in the mid-latitudes of North America and Eurasia.

Overall, the results for the depositional profiles of the OC pesticides and PAHs in glacial cores appear to have better temporal resolution than Arctic sediment cores, reflecting high annual deposition rates compared to most Arctic lakes or marine sediments. The post-depositional volatilization of semi-volatile organics such as HCH, HCB, and phenanthrene from snow as its density increases and its surface area decreases, may, however, confound the interpretation of temporal trends. Further study is needed to understand the basic processes governing the fate of semi-volatile organics in snow.

## 5.2. Terrestrial environment

A number of temporal-trend datasets for the terrestrial Arctic were reported in the previous AMAP POPs assessment (de March *et al.*, 1998), but none of these programs have been continued. This is likely due to the low levels found in this environment. Consistent with studies in the freshwater and marine systems, levels of OCs were found to be decreasing in the terrestrial environment prior to 1996.

One temporal-trend dataset that was not reported in the previous AMAP assessment examined OCs in Alaskan peregrine falcons from 1979 to 1995 (Ambrose *et al.*, 2000). Dieldrin, *p,p'*-DDE, heptachlor epoxide, oxychlorane, and total Aroclor PCBs were consistently detected and measured, and were tested statistically for relationships with time and productivity. Mirex was only measured from 1988 to 1995, but was detected in all samples. HCB, *p,p'*-DDD, *p,p'*-DDT,  $\beta$ -HCH, and *trans*-nonachlor were detected in >50% of samples, but were not tested for relationships with time or productivity because they were not consistently analyzed.  $\alpha$ -HCH,  $\gamma$ -HCH, *cis*-chlordane, *trans*-chlordane, *o,p'*-DDD, *o,p'*-DDE, *o,p'*-DDT, endosulfan II, and endrin were detected in <50% of samples, and because of the large proportion of data below detection limits, none were tested for time or productivity relationships. The five persistent OC contaminants that were consistently measured (dieldrin, *p,p'*-DDE, heptachlor epoxide, oxychlorane, total PCBs) declined significantly from 1979 to 1995, but the trend was weaker for total Aroclor PCBs than other contaminants.

## 5.3. Freshwater environment

### 5.3.1. Water and sediments

#### *Russian river water and sediment*

Monitoring data on OC pesticides in river water from the 1980s and early to mid-1990s have recently been published (Gordeev and Tsirkunov, 1998; Petrosyan *et al.*, 1998; Alexeeva *et al.*, 2001; Zhulidov *et al.*, 2002). This permits a historical perspective of pesticide loadings to Russian northern seas. Gordeev and Tsirkunov (1998) summarized the estimated fluxes of  $\Sigma$ HCHs and  $\Sigma$ DDTs for 32 rivers in Russia including 11 flowing to the Arctic Ocean. To obtain fluxes, they used annual arithmetic mean concentrations, summarized by Petrosyan *et al.* (1998), and river discharges taking into account seasonal variability in flows. Alexeeva *et al.* (2001) used a similar approach to estimate fluxes for the same pesticides for the period of 1990 to 1996. They used monitoring data from the regional laboratories of ROSHYDROMET and thus, used data from the same pesticides monitoring program that Gordeev and Tsirkunov's (1998) study used. The combined results (Figure 5-14) show a general decline in loadings of HCH and DDT isomers over the period of 1981 to 1996, coinciding with the reduction in use of HCH in Russia following conversion to use of lindane, and the complete cessation of agricultural use of DDT in the 1970s.

Zhulidov *et al.* (2002) reported temporal trends of  $\Sigma$ DDTs (*p,p'*-DDT, DDE, and DDD) and  $\Sigma$ HCHs ( $\gamma$ - and  $\alpha$ -isomers) in water and sediments from eight Russian Arctic rivers for the period of 1988 to 1994. OC levels in burbot liver were also determined from the same rivers (Section 5.3.4). DDT was not detected in any sediment, and DDE and DDD were only detected in sediments of three rivers. DDD was not detected in river water, and DDT and DDE were only present in North Dvina and Pechora River water. However, the detection limits for the methods used were relatively high in both

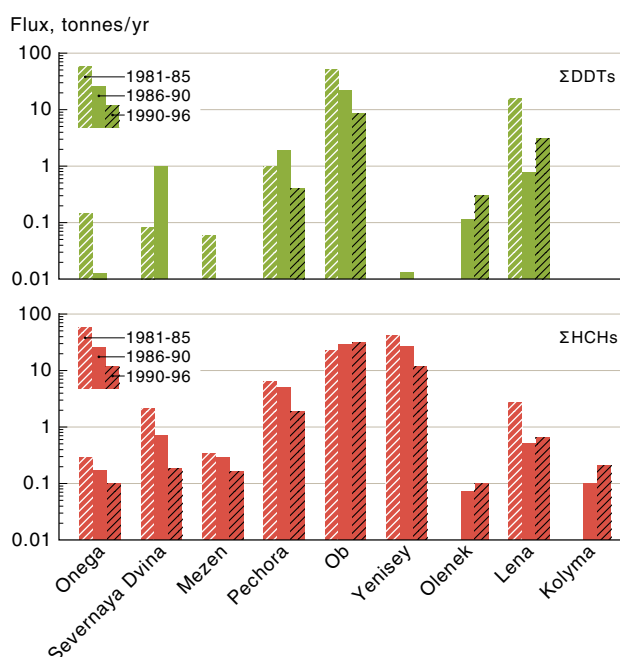


Figure 5-14. Temporal trends in fluxes of  $\Sigma$ DDTs and  $\Sigma$ HCHs in major rivers flowing in the Russian northern seas for the periods 1981-1985, 1986-1990, and 1990-1996 (from Gordeev and Tsirkunov (1998) and Alexeeva *et al.* (2001)).

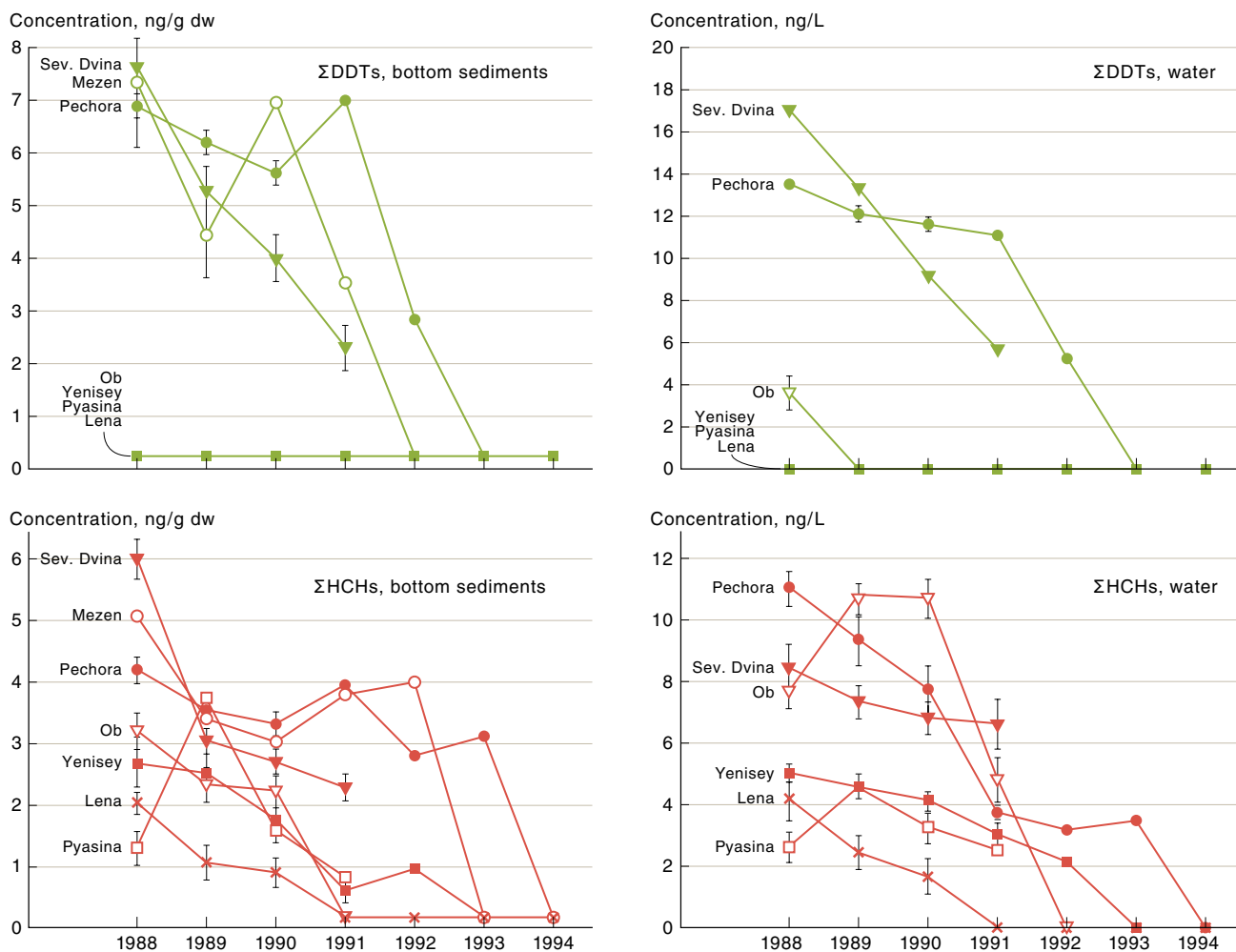


Figure 5-15. Temporal trends in concentrations of DDT and HCH isomers in water and sediments from major rivers flowing into the Russian northern seas for the periods 1988 to 1994 (Zhulidov *et al.*, 2002).

sediment and water. Both HCH isomers ( $\alpha$ - and  $\gamma$ -) were detected in all river sediments except Kolyma River and in water from all rivers. Concentrations of both DDT and HCH declined significantly over the period of 1988 to 1992 and were near detection limits for all chemicals in most rivers by 1992 (Figure 5-15).

#### Dated sediment cores from Canadian Arctic lakes

Outside of the Arctic, analysis of dated sediment cores has been used to infer the depositional history of POPs in the Great Lakes region (Jeremiason *et al.*, 1994; Wong *et al.*, 1995; Pearson *et al.*, 1998) and in European alpine lakes (Fernandez *et al.*, 1999; 2000; Grimalt *et al.*, 2001). The previous AMAP POPs assessment included results for PCBs and PCDD/Fs in Arctic sediment cores from Alaska, Canada, and Finland. These cores all showed a later onset of PCB deposition than that observed in most lakes closer to sources, as well as maximum inputs near or at the surface, indicating continuing inputs from atmospheric deposition. Further analysis of the data from Canadian lakes showed that lower chlorinated PCBs predominated in High Arctic cores, while sediments from temperate lakes had higher proportions of Hx-, Hp- and OcCBs (Muir *et al.*, 1996a). The sediment records from Arctic lakes, therefore, can provide information on temporal trends of deposition of these hydrophobic contaminants in the Arctic. However, interpretation of sediment records can be complicated by

'direct' input from non-atmospheric sources, and dynamic lake processes including bioturbation, sediment focusing, and re-suspension. Factors unique to Arctic lakes, such as long periods of ice cover and low sedimentation rates, may limit inputs to bottom sediments and make them a less significant reservoir for hydrophobic organics than temperate lakes (Macdonald *et al.*, 2000).

Additional sediment cores collected in the Canadian Arctic in the mid- to late 1990s have been analyzed for POPs since the previous AMAP assessment (Lockhart, 1997; Lockhart *et al.*, 1997; Macdonald *et al.*, 2000; Stern and Evans 2003; Muir *et al.*, 2002b; Rose *et al.*, 2000). Undated cores were also analyzed from Bjørnøya (Kallenborn, 2002b). However, there were no cores reported from other circumpolar countries.

A series of cores in the Yukon (western Canadian Arctic) were analyzed for PCBs and OC pesticides (Rawn *et al.*, 2001). DDT was found to be the most prominent OC in the sediment profiles of most of the lakes. Maximum  $\Sigma$ DDT levels (0.86 ng/g dw to 21.4 ng/g dw) were observed in sediment slices dated to the 1950s from lakes (Lake Laberge and Fox Lake) near populated areas, (Figure 5-16), as well as in Watson Lake (not shown). In contrast, in more remote lakes (Hanson and Lindeman Lakes), maximum  $\Sigma$ DDT concentrations were observed in sediments dated to the 1980s and 1990s, similar to other Arctic lakes (Muir *et al.*, 1995a). The sediment profiles from lakes situated near populated areas and

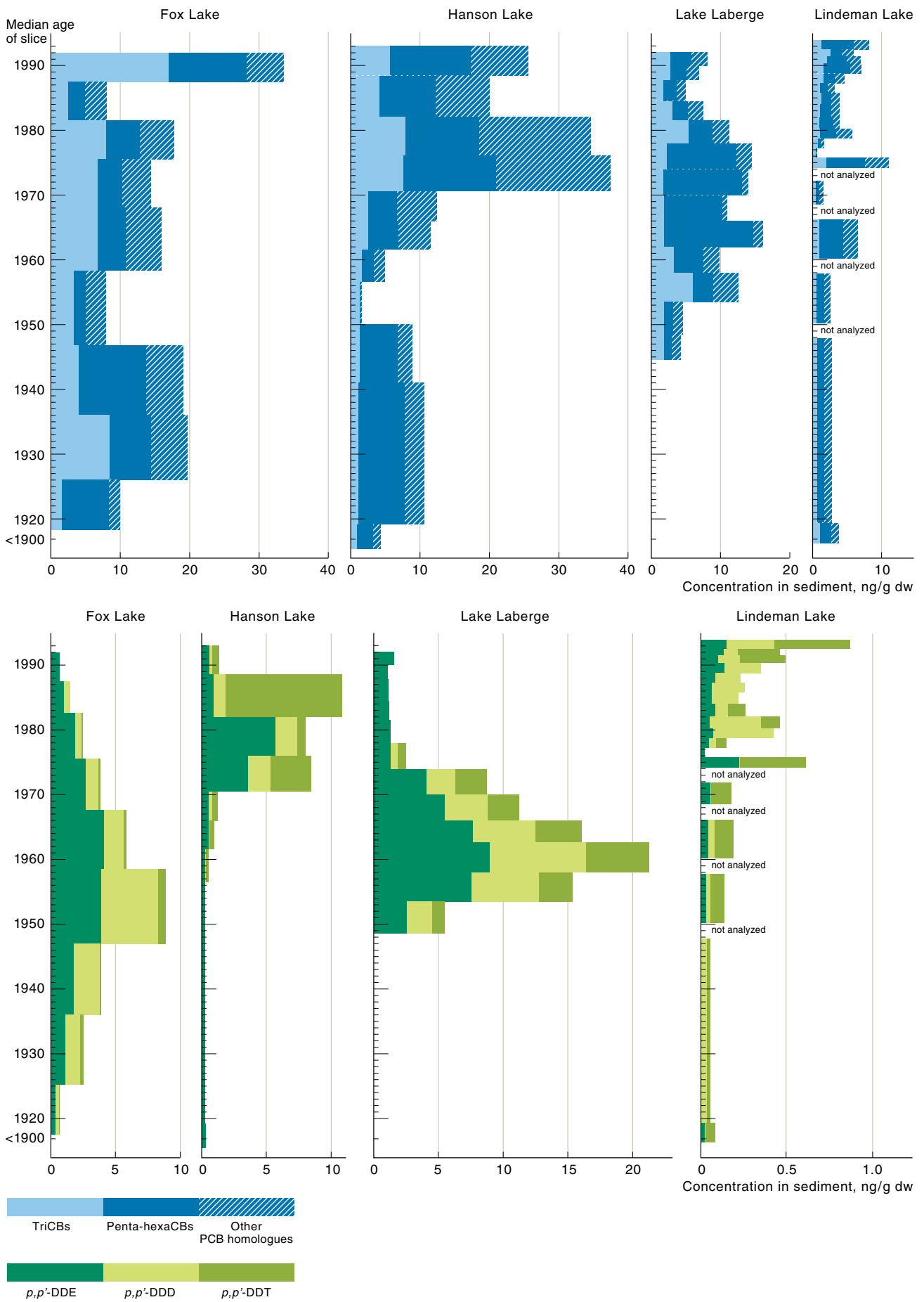


Figure 5-16. Historical profiles of PCBs and DDT-related compounds in four lakes within the Yukon River watershed (Rawn *et al.*, 2001), two remote lakes (Hanson, Lindeman) and two lakes nearer to populated areas (Fox, Laberge).



along the Alaska Highway in the Yukon indicate PCB and DDT contamination has occurred mainly from local usage and waste disposal, rather than long-range transport and deposition. The onset of appearance and maximum concentrations and fluxes of  $\Sigma$ PCBs and  $\Sigma$ DDTs are similar to cores from southern Canada, and coincide well with the use profile of these chemicals in North America in general, and with the known history of use in the Yukon as well. The historical profile of  $\Sigma$ PCBs and  $\Sigma$ DDTs in most of these Yukon sediments is quite different from sediments in the High Arctic, where delayed onset has been found (Muir *et al.*, 1996a). Even in the remote, glacier-fed Lindeman Lake, the lower chlorinated PCB congeners made relatively small contributions to  $\Sigma$ PCBs. This may be due to volatilization losses of the lower chlorinated congeners from melting snow and flowing glacial streams (Macdonald *et al.*, 1999; Blais *et al.*, 2001). The results suggest that glacial runoff is a significant source of OCs to small high-elevation lakes (Lindeman Lake) but not to larger lakes within the Yukon River drainage basin which are also affected by glacial sources (Lake Laberge, Kusawa Lake).

Historical deposition of toxaphene in five Yukon lakes is shown in Figure 5-17. Elevated toxaphene levels had been found in fish tissues from several of the large lakes in the Yukon River Basin system (Kidd *et al.*, 1993; 1995), and thus, the source of toxaphene was of interest. Sediment core results confirm that the source is mainly atmospheric rather than local. The exception is Hanson Lake which was treated with toxaphene in 1963 (Walker *et al.*, 1973). The toxaphene congeners in this lake consist almost entirely of a hexa- (B6-923) and heptachlorobornane (B7-1001) (these have no Parlax numbers). These same two congeners were also predominant in the sediment of two toxaphene-treated

lakes located in Alberta, Canada (Miskimmin *et al.*, 1995; Stern *et al.*, 1996), but not in remote lakes exposed only to atmospheric sources (Muir *et al.*, 1999a; Rose *et al.*, 2001). B6-923 and B7-1001 are formed via anaerobic reductive dechlorination of other, less stable, chlorobornanes present in the technical mixture but not generally in air (Vetter *et al.*, 1999). The toxaphene profile in Watson Lake (Figure 5-17) is not entirely consistent with what would be expected based solely on atmospheric deposition (i.e. peak levels occurring in the early to mid-1970s). The higher subsurface levels of toxaphene congeners B6-923 and B7-1001 (Stern and Evans, 2003) and the overlap of the toxaphene and  $\Sigma$ DDT historical concentration profiles, suggest that toxaphene usage in the Watson Lake area occurred prior to the banning of DDT, possibly for use as a fish toxin or as an additional active ingredient in the insecticidal DDT mixtures.

A laminated core from a lake on Devon Island (Nunavut) has been analyzed for a wide array of OC compounds (Figure 5-18). The laminations are due to annual layers of sediment, with differing color or texture. These form detectable laminations that can be counted. This is the first laminated core analyzed for POPs in the Arctic and provides an unusual degree of temporal resolution compared to most other cores from the High Arctic.  $\Sigma$ DDT concentrations peaked between the mid-1950s and early 1970s. An increase in toxaphene was first observed in the early 1970s, reaching a maximum in the 1980s. These results are consistent with the known historical usage of DDT and toxaphene in North America (Voldner and Li, 1995; Li *et al.*, 2001b).

Concentration profiles for total PCDD/Fs in a second sediment core from Lake DV09 on Devon Island, normalized to organic carbon, are also shown in Figure 5-18 (Stern and Evans, 2003). PCDF levels start to in-

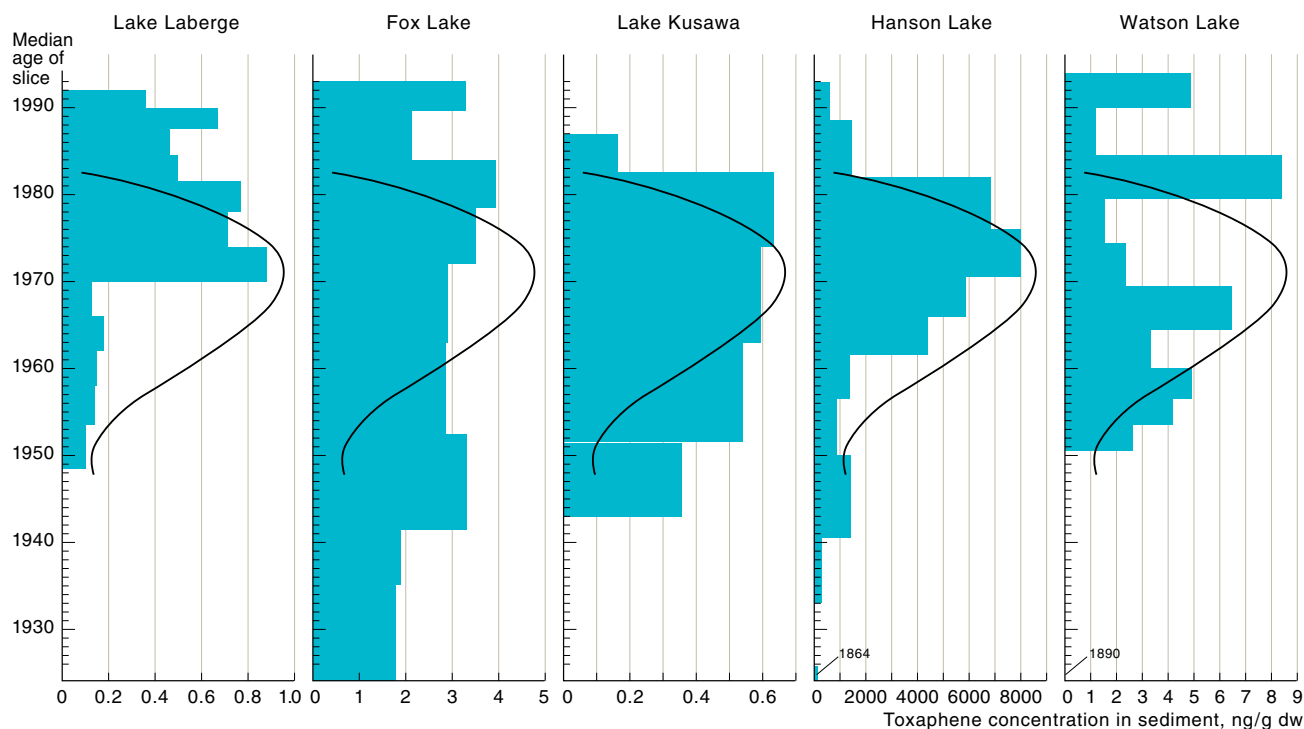


Figure 5-17. Concentration profiles of toxaphene in sediment cores from five Yukon lakes. Elevated concentrations in Hanson Lake are the result of application of toxaphene to this lake in 1963. The curves correspond to the atmospheric input functions for toxaphene derived by Rapaport and Eisenreich (1988) from peat cores.

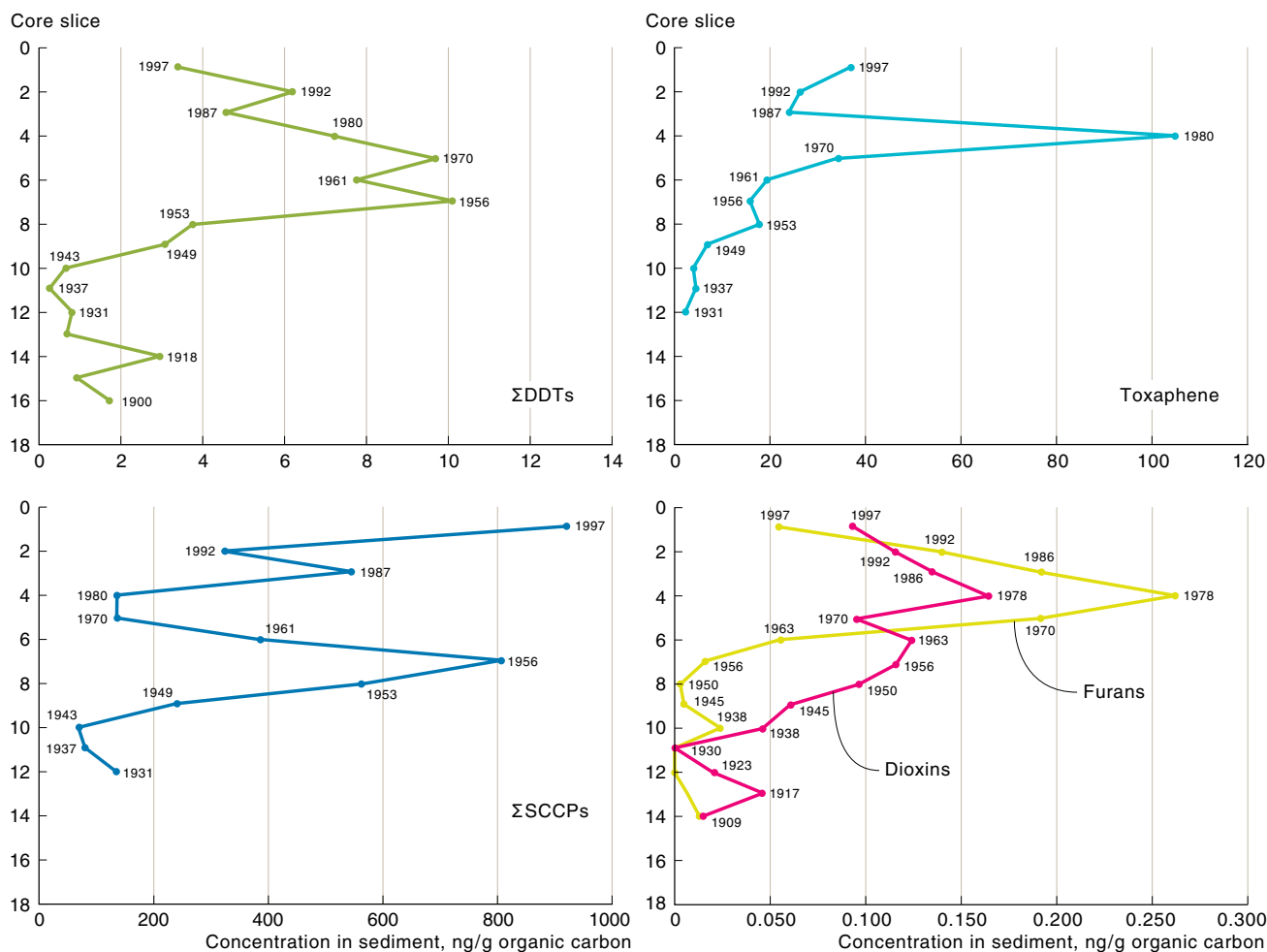


Figure 5-18. Concentration profiles of DDT, toxaphene, SCCPs, and PCDD/Fs in dated, laminated cores from Lake DV09 on Devon Island, Nunavut, Canada. Concentrations are in ng/g organic carbon.

crease in the early 1950s, peak at 0.27 ng/g organic carbon in 1978, and are dominated by the TCDF homologue group, in particular the 1,2,4,8-TCDF congener. PCDD levels start to increase about ten years earlier but also peak in 1978. Between 1938 and 1963, sediment core slices were dominated by octachlorinated dibenzo-p-dioxin (OCDD) ( $64 \pm 9\%$ ). This homologue profile is consistent with a signature resulting from usage of pentachlorophenol as a wood preservative. OCDD is the major impurity in the PCP technical mixtures and can also be formed by photolytic degradation of PCP (Crosby *et al.*, 1981). From 1970 to 1992, the PCDD/F profiles are indicative of signatures arising from combustion of coal and wood (Kjeller *et al.*, 1996).

The DV09 core was also analyzed for SCCPs (Figure 5-18). Maximum concentrations occur in the surface sediment and in the core slice dated to 1956. Maximum usage of SCCPs occurred much later, probably between 1978 and 1985 (Muir *et al.*, 2000a), and thus, the historical profile in this core is not consistent with historical usage or historical profiles in other lakes (Tomy *et al.*, 1999). Shorter carbon chain length and lower chlorinated  $C_{10}$  and  $C_{11}$  formula groups become more predominant downward in the core, suggesting microbial degradation of the longer chain, more highly chlorinated compounds, over time, and/or that the earlier emissions had higher proportions of more volatile SCCPs.

*Sediment cores from lakes on Bjørnøya*

Sediment cores were taken from two small lakes (Ellasjøen and Øyangen) on Bjørnøya, approximately half way between Norway and Svalbard, in 1996 for OC analysis as part of a larger study (see Section 4.3.5).  $\Sigma PCB_7$  concentrations decreased downward in the sediment to 0.5 and 0.4 ng/g dw at 13-15 cm in Ellasjøen and Øyangen, respectively (Figure 5-19). In Ellasjøen,  $\Sigma DDTs$  was 6.9 ng/g dw in surface sediment, of which *p,p'*-DDE comprised about 90%. In Øyangen, the corre-

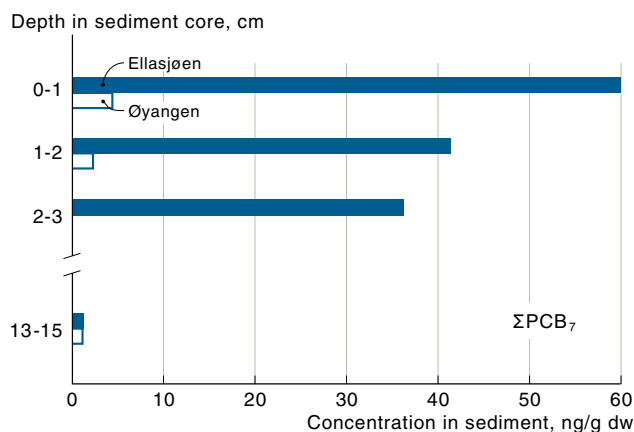


Figure 5-19.  $\Sigma PCB_7$  concentrations in sediment cores sampled from the lakes Ellasjøen and Øyangen on Bjørnøya in July, 1996 (Evenset *et al.*, 2002).

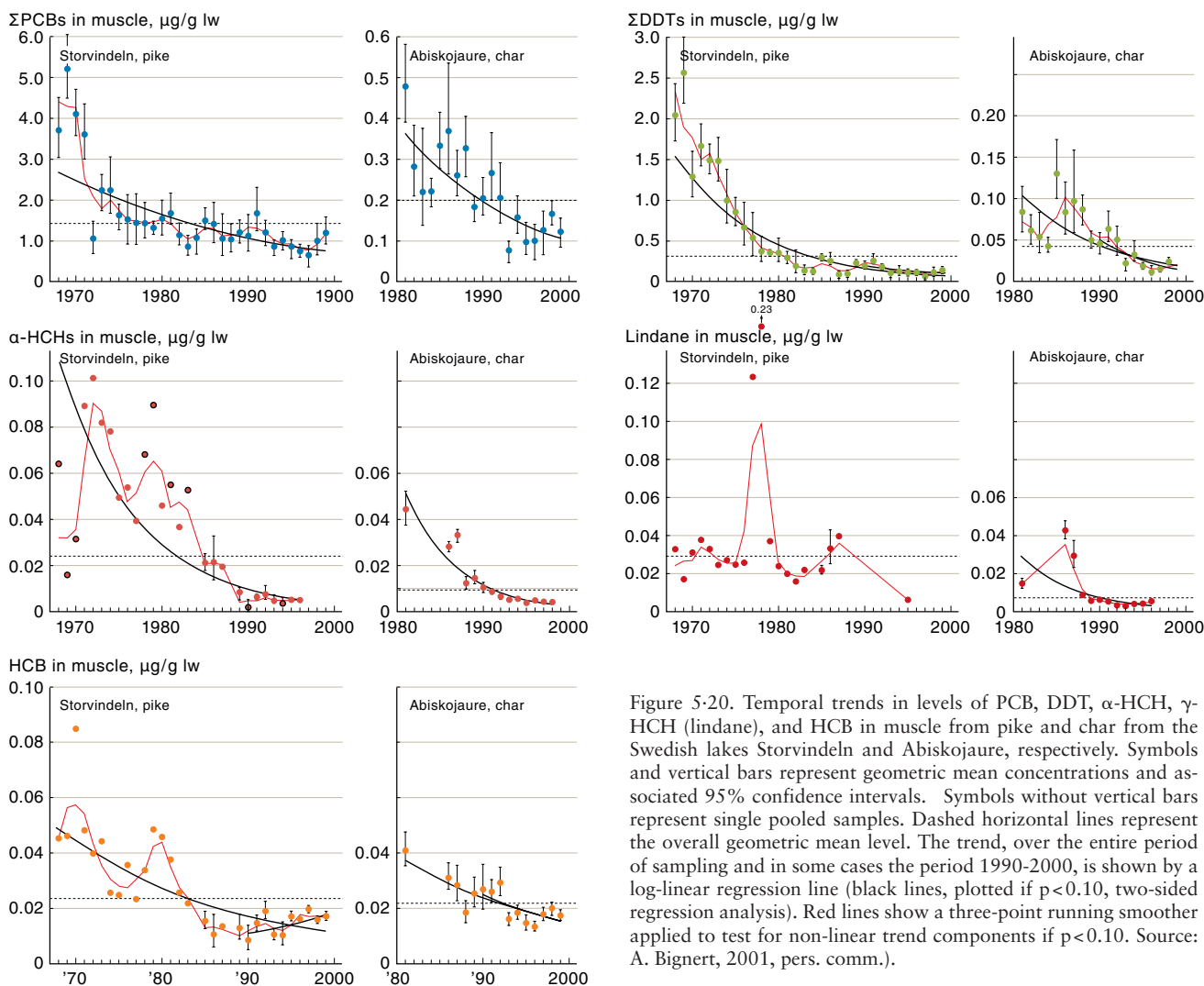


Figure 5-20. Temporal trends in levels of PCB, DDT,  $\alpha$ -HCH,  $\gamma$ -HCH (lindane), and HCB in muscle from pike and char from the Swedish lakes Storvindeln and Abiskojaure, respectively. Symbols and vertical bars represent geometric mean concentrations and associated 95% confidence intervals. Symbols without vertical bars represent single pooled samples. Dashed horizontal lines represent the overall geometric mean level. The trend, over the entire period of sampling and in some cases the period 1990-2000, is shown by a log-linear regression line (black lines, plotted if  $p < 0.10$ , two-sided regression analysis). Red lines show a three-point running smoother applied to test for non-linear trend components if  $p < 0.10$ . Source: A. Bignert, 2001, pers. comm.).

sponding numbers were 0.8 ng/g dw and 63%, respectively. The concentrations at 13-15 cm were 0.05 ng/g dw in Ellasjøen and 0.2 ng/g dw in Øyangen. These results imply that loadings of PCBs and DDT have been increasing in these systems, which is not consistent with other temporal trends in the Arctic. The sediment slices were not dated, however, and therefore, caution is needed regarding these trends.

### 5.3.2. Temporal trends in fish in northern Scandinavia

In 1967, a Swedish program was initiated to monitor PCB, DDT, HCB, and HCH levels in fish from areas that had little or no known point sources of pollution (Olsson and Reutergrårdh, 1986; Olsson and Bignert, 1997). Muscle samples from 20 northern pike have been collected every year since 1967 from Lake Storvindeln, a forest lake near the Swedish Alps occupying an area of 55 km<sup>2</sup>. Beginning in 1980, muscle samples from 20 Arctic char have been collected from the lake Abiskojaure (Åbeskojåvre), 200 km north of the Arctic Circle. Efforts have been made to collect specimens that are of similar sex, age, size, and in the same sampling season. This program has yielded an unparalleled temporal-trend dataset for OCs in the Arctic and subarctic.

As was reported in the previous AMAP POPs assessment (de March *et al.*, 1998), levels of OCs in Swedish

freshwater fish have declined significantly since the late 1960s into the 1990s (Figure 5-20 and Table 5-2). However, the rates of decline have slowed in the 1990s, and no significant decline has been observed for the past ten years. The concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs have been decreasing at a rate of approximately 3-7% and 10% per year, respectively, in all investigated time series (i.e. both southern and northern Sweden and in both species of fish). Since the monitoring began in 1967/68 for pike and 1981 for Arctic char, the temporal trend is long enough to be able to detect a change of this magnitude. For the last ten years, the decreasing trends in PCBs seem to have slowed in the northern parts of Sweden, as no significant decreasing trends are found for these years (Table 5-2). For DDT, over the past ten years, the decreasing trends have leveled off in the southern parts of Sweden, but continued at approximately the same annual rate (8-14%, significant decrease) in the northern parts.

HCH levels have decreased rapidly during the studied time period, more than 10% annually, following the bans and restricted use of technical HCH and lindane in countries neighboring Sweden. The concentrations in Swedish freshwater biota are now below or close to the detection limit (during recent years all samples have been below). A significant decreasing trend is also found for the last ten years in the northern part of Sweden. The high peak for lindane in pike from Lake Storvindeln dur-

ing 1977-1978 is, as yet, unexplained. HCB concentrations have decreased at a rate of approximately 5% per year, but northern pike have shown a nearly significant ( $p < 0.1$ ) increasing trend over the past ten years.

The comprehensive nature of this dataset, comprised of many samples and time points covering a long time period, provides an invaluable tool in assessing the time trends of OCs in the European Arctic. Olsson and Bignert (1997) found that the annual rate and the onset of the decline (1971-1972) in concentrations of DDT compounds did not differ between the Arctic and other remote areas of northern Sweden, or between the southern parts of the Baltic Sea and lakes in the southern part of the Swedish mainland. The results of this program have demonstrated that the banning and reduction of use of

Table 5-2. Mean annual rate of change (%) in concentrations in pike and Arctic char in two lakes in northern Sweden.

OC	Pike Location: Storvindeln		Arctic char Abiskojaure	
	Annual change (%) 1967-1999	Annual change (%) 1990-1999	Annual change (%) 1980-1999	Annual change (%) 1990-1999
ΣDDT	-10	-8.3	-9.9	-14
ΣPCB	-4.1	-4.1	-6.7	-6.0
α-HCH	-11	-10	-17	
γ-HCH	-3.4		-15	
HCB	-4.8	5.4	-5.0	-5.5

OCs in much of the world has resulted in significant declines in Arctic biota. However, in recent years this reduction has slowed, suggesting that residual sources of OCs are likely to continue to contaminate Arctic biota for some time to come.

### 5.3.3. Temporal trends in freshwater fish in the North American Arctic

A number of temporal-trend datasets for OC pesticides and PCBs are now available for North American Arctic freshwater fish. These datasets are of much shorter length, and comprise fewer sample collections compared to the monitoring studies currently being carried out in Sweden (Section 5.3.2). Therefore, they lack the statistical power to make conclusive statements about temporal trends of OCs, but they do provide some insights. Most of these studies do not include data prior to 1990. A number of studies can be combined to provide better insight into temporal trends. No definitive pattern emerges, but in general, OC concentrations appear to be decreasing in freshwater fish.

Concentration in burbot liver, ng/g ww

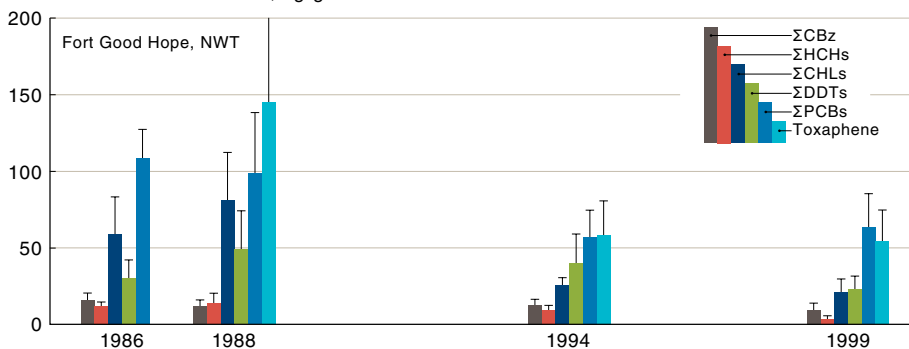


Figure 5-21. Trends in concentrations of major OC groups in liver of burbot at Fort Good Hope, NWT, Canada (1986-1999).

### Fort Good Hope burbot

POPs were measured in the livers of burbot collected at Fort Good Hope, NWT, in 1988 and 1999 to examine temporal trends of OCs (Stern *et al.*, 2001a). Significant declines, 2.0- and 3.1-fold, were observed for both α- and γ-HCH over this 11-year time period. β-HCH concentrations were below the detection limit in all samples. Interestingly, the α-:γ-HCH ratio has increased from 4.5 to 6.9, which is opposite to what one might have expected based on the decreased usage of the technical mixture (α-HCH (60-70%), β-HCH (5-12%), and γ-HCH (10-15%)) and the corresponding increase in the usage of lindane in western Canada since the early 1990s (Waite *et al.*, 2001; Li and Bidleman, 2003). This is likely due to the ability of the burbot to more efficiently degrade the γ- compared with the α-isomer. The γ-isomer is generally found to be the most easily degraded in biota (Moisey *et al.*, 2001). ΣDDT concentrations did not change over this 11-year time interval; however, a 1.8-fold decline and a 2.2-fold increase in the concentration of *p,p'*-DDT and its metabolite, *p,p'*-DDE (not age adjusted), respectively, was observed. These changes translated into a significant increase in the *p,p'*-DDE:ΣDDT ratio from 0.39 to 0.60 and suggests 'old' rather than recent DDT inputs. Overall, a 1.7-fold decline in the lipid-adjusted mean concentrations of ΣCHLs was observed. Oxychlordane, the principal metabolite of *cis*- and *trans*-chlordane, and second only to *trans*-nonachlor as the most abundant chlordane-related residue in the Fort Good Hope burbot liver, did not change significantly over this 11-year period. The decreasing *trans*-:*cis*-CHL ratio suggests 'old' rather than recent chlordane inputs. ΣTrCB concentrations have increased 2.9 fold, while all other PCB homologue groups have either declined in concentration or did not change significantly. ΣPCB levels declined 1.3 fold. No significant differences were observed in levels of nPCBs 77, 81 or 169. CB126 levels decreased 1.8 fold. Toxaphene and dieldrin concentrations decreased by 1.7 and 1.5 fold, respectively.

Combining the OC data generated for burbot by Stern *et al.* (2001a) with previously published data for burbot at Fort Good Hope (Muir *et al.*, 1990a; Muir and Lockhart, 1996), which includes the years 1986, 1988, and 1994, provides a longer-term dataset (Figure 5-21). The results from 1988 reported by Stern *et al.* (2001a) are much lower than those reported previously (Muir *et al.*, 1990a) and are excluded. Slow declines in all of the major OC groups and toxaphene are observed in the Fort Good Hope burbot, although the rate of change varies with the chemical and the period concerned.



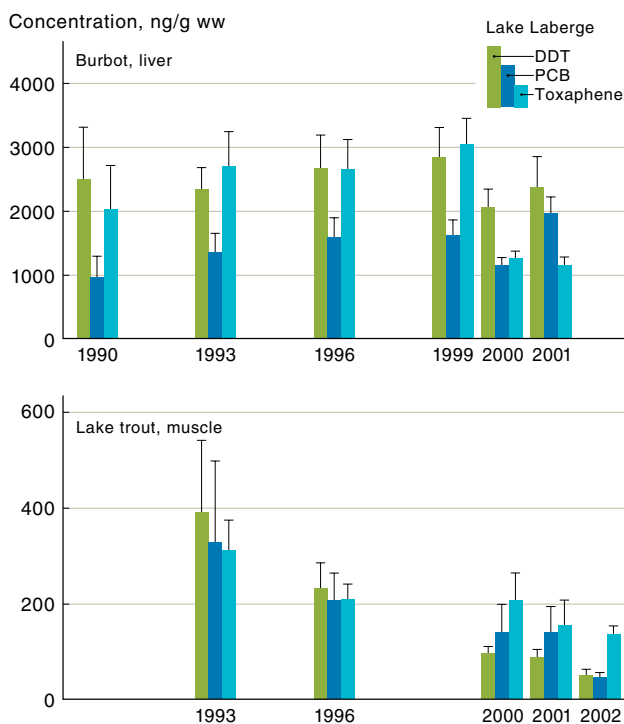


Figure 5-22. Concentrations (mean  $\pm$  1 SD) of DDT, PCBs, and toxaphene in liver of burbot and muscle of lake trout from Lake Laberge, Yukon, Canada (1990-1999).

#### Lake Laberge lake trout and burbot

Combining data from a number of studies on OCs for burbot and lake trout from Lake Laberge also allows an examination of temporal trends spanning almost twelve years (Muir and Lockhart, 1992; Kidd *et al.*, 1998; Stern *et al.*, 2000; Palmer and Roach, 2001; Ryan *et al.*, 2003). Levels of DDT and toxaphene in lake trout from Lake Laberge declined between 1993 and 2002 by approximately 86% and 55%, respectively (Figure 5-22). Although PCB levels also appear to have declined considerably (approx. 85%) since 1993, the high variability in the samples over this period has resulted in a marginally insignificant statistical result ( $p = 0.06$ ). Lipid levels in lake trout also show a significant decrease since 1993, which may account for some of the decrease in contaminants. This decline is in contrast to that observed in burbot collected in Lake Laberge, which showed no decline in DDT and PCB concentrations in liver between 1990 and 1999 (Figure 5-22). However, a significant decline in toxaphene concentrations in burbot (approx. 57%) was observed between 1993 and 2001 (Figure 5-22). Lake Laberge burbot liver displayed only a marginal decrease in fat content, which may be attributed to sampling variation (spring and summer fish). Studies on the Lake Laberge system continue (Stern *et al.*, 2001b).

#### Great Slave Lake burbot and lake trout

OC data spanning six years (1993-1999) in burbot and lake trout are also available for Great Slave Lake (Evans and Muir, 2000; 2001). No clear trend emerges for  $\Sigma$ PCBs in either species using lipid-normalized data, but  $\Sigma$ CHLs,  $\Sigma$ DDTs, and toxaphene do appear to be declining slowly (see Annex Table 7). These conclusions should be used with caution, because no effort has been made to correct for biological variables such as size or growth rate.

#### 5.3.4. Temporal trends in fish in the Russian Arctic

Zhulidov *et al.* (2002) studied temporal trends of  $\Sigma$ DDTs and  $\Sigma$ HCHs in burbot livers collected from eight Russian north-flowing rivers from 1988 to 1994. Three DDT isomers (*p,p'*-DDT, DDE, and DDD) and two HCH isomers ( $\gamma$  and  $\alpha$ ) were measured. Levels in sediment and water were also assessed from the same rivers (Section 5.3.1).  $\Sigma$ DDTs (up to 70 ng/g ww) and  $\Sigma$ HCHs (up to 18 ng/g ww) both declined significantly in burbot liver between 1988 and 1994 (Figure 5-23) in parallel with declines in river water. The magnitude of the declines for  $\Sigma$ DDTs in burbot was up to 10 fold depending on the river system, and much more rapid than observed in burbot from the Mackenzie River (Canada) over the same period. This may reflect the response to the relatively recent cessation of DDT use within some of the watersheds, and in Russia in general (see Section 2.3.1). A 3- to 4-fold decline was seen for  $\Sigma$ HCHs in burbot from the eight Russian rivers, which is similar to the decline in HCHs in burbot in the Canadian Arctic.

#### 5.4. Marine environment

##### 5.4.1. Temporal trends of TBT effects in invertebrates

There is only a limited amount of information concerning temporal trends in TBT and its effects on subarctic and Arctic biota. The only available information on effects within the AMAP area is for imposex (i.e. the development of a penis and vas deferens in female marine snails). This occurs only within neogastropods (Mollusca) and has been examined in dogwhelks.

Svavarsson (2000) studied imposex in dogwhelks in southwestern, western, and northwestern Iceland in 1998, and compared the imposex levels to those reported from Iceland in 1992-1993 by Svavarsson and Skarphéðinsdóttir (1995). There has been a substantial decline in the level of imposex in dogwhelks in Icelandic waters since 1992/1993. At 24 of the 31 studied localities in Iceland, the Vas Deferens Sequence Index (VDSI) was lower in 1998 than in 1992-1993 (Svavarsson, 2000). At one site, the species was extinct, at five sites the index was higher in 1998 than previously, and at one site, no change had occurred. The most pronounced changes occurred near large harbors, such as Reykjavik and Hafnarfjörður harbors, where effects had been previously most pronounced. Changes were also evident in the Relative Penis Size Index (RPSI) at both small and large harbors.

Similar imposex declines have been seen in dogwhelks in Norway just south of the AMAP area (Følsvik *et al.*, 1999). No evident improvements have, however, occurred in northern Norway (Green *et al.*, 2002) and in the Faroe Islands (FEA, 2002). Among eight studied sites in northern Norway, the VDSI levels were high (greater than 3) at six of these stations. Some improvement may have occurred at two stations, but the situation was worse at one station (Green *et al.*, 2002). In the Faroe Islands, no improvements were seen at the studied sites, apart from possibly one site (Kirkjutangi) (FEA, 2002). The study sites were few, and most had been observed previously to have both high VDSI and RPSI in dogwhelk. The latter is high only where tissue levels of TBT are very high.

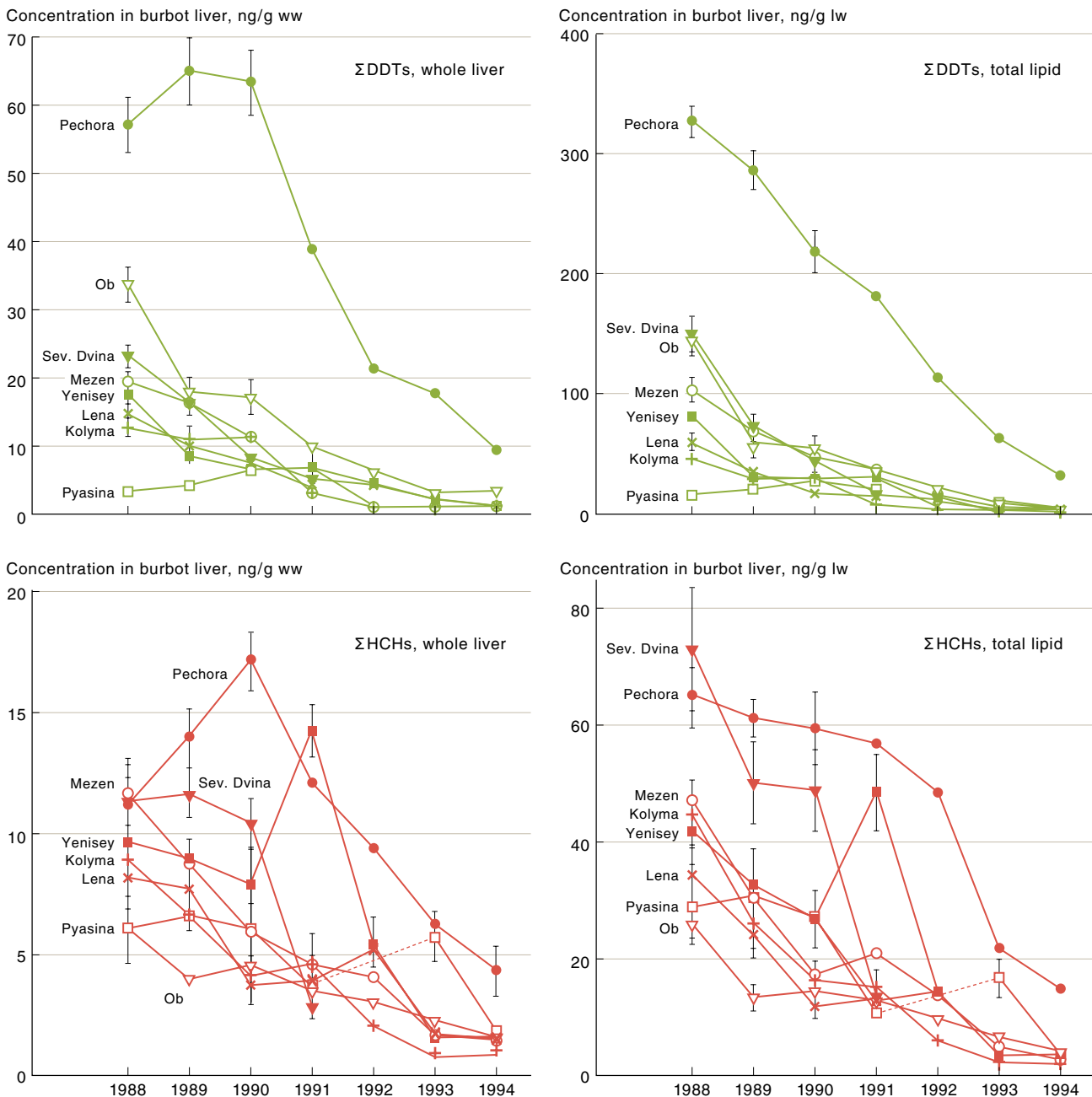


Figure 5-23. Temporal trends in concentrations of  $\Sigma$ DDTs and  $\Sigma$ HCHs in liver of burbot from major rivers flowing into the Russian northern seas for the periods 1988 to 1994 (Zhulidov *et al.*, 2002). Results are reported on a wet weight and lipid weight basis.

The declines in imposex seen in Iceland and southern Norway have been related to restrictions implemented in Iceland and Norway on the use of TBT on vessels smaller than 25 m (Følsvik *et al.*, 1999; Svavarsson, 2000). Svavarsson (2000) also related these changes to a change in marketing of paint in 1993 in Iceland. At that time, large Icelandic producers of anti-fouling paints started encouraging their customers to use non-TBT-based paints. Additionally, these improvements may partly be explained by developments in paint technology (see Bennett, 1996). The earlier paints were 'free association' paints, while later paints were copolymer paints with uniformly chemically bonded TBT with a constant, but minimal, release of TBT (Bennett, 1996; Svavarsson, 2000).

Studies of the occurrence of imposex in dogwhelks in the Faroe Islands were conducted in 1996 and in 2001

(FEA, 2002). Although the results do not indicate any major changes over the five years from the first to the second sampling period, and the occurrence of imposex in the Faroe Islands is still widespread, there are sites where the phenomenon is hardly seen.

#### 5.4.2. Temporal trends in marine fish

Temporal-trend data for marine fish were not available for the previous AMAP POPs assessment (de March *et al.*, 1998). Two studies have however been reported for the current AMAP assessment.

The first of these is a temporal-trend study on the OC levels in liver of Atlantic cod and dab in the coastal waters of Iceland (Yngvadóttir and Halldórsdóttir, 2002). This study covers the period 1991 through 2000, with data missing only from 1993, and includes the major

Concentration in Atlantic cod liver, ng/g ww

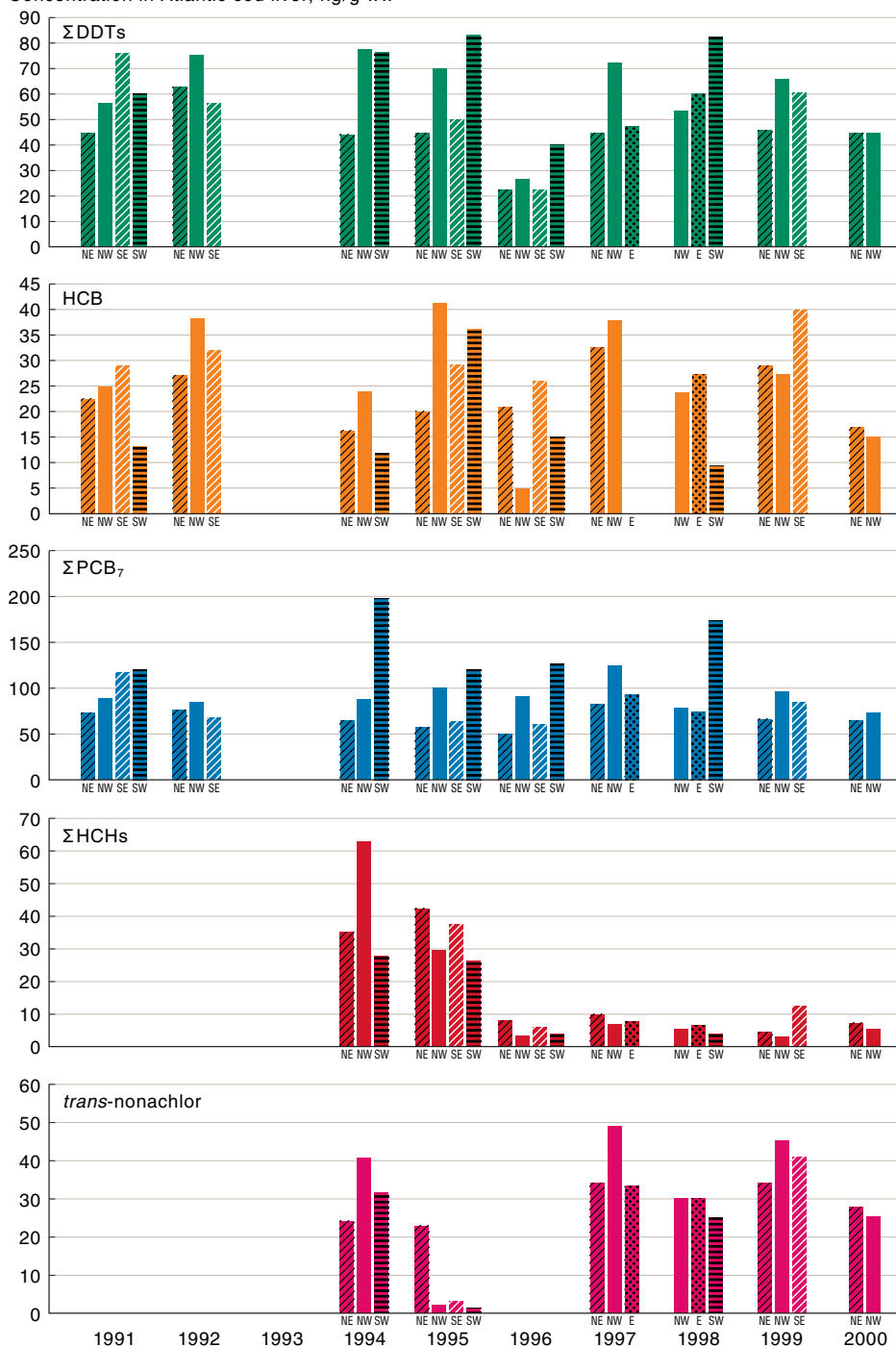


Figure 5-24. Average concentrations of OCs in the livers of Atlantic cod (length = 30-45 cm) in Icelandic waters, 1991-2000.

OC groups and toxaphene. The study involves collection of similar sized fish at a number of locations around the coast of Iceland. No significant temporal trends were found for any of the OC groups in either Atlantic cod or dab (Figures 5-24 and 5-25).

OCs have also been measured in cod (*Gadus morhua callarias*) liver from the Vestertana Fjord (N. Norway) from 1987 to 1998 (Sinkkonen and Paasivirta, 2000). Significant decreases in DDD,  $\alpha$ -HCH, and  $\gamma$ -HCH were observed, but no trends were found for DDE, TCDF, PCBs, chlordanes, PCNs, HCB or polychlorinated diphenyl ethers (PCDEs). Hepta- and octaCDFs were found to increase from 1987 to 1994, and to increase steeply from 1994 to 1998, probably due to the use of a chlorophenol product as a wood preservative.

#### 5.4.3. Temporal trends in seabirds

The previous AMAP POPs assessment detailed a number of studies on temporal trends of OCs in Arctic seabirds. These studies showed a downward trend in all 'legacy' OCs in seabirds around the circumpolar Arctic (de March *et al.*, 1998). There have been a number of new studies on OCs in Arctic seabirds, but few were designed to address temporal trends, and insufficient new data have been added to warrant reexamination of these time series in this report. Further, studies that were designed specifically to evaluate temporal trends however do provide results that yield firmer and more convincing conclusions about temporal trends.

One such study has monitored temporal trends of OCs in Arctic seabird eggs in the eastern Canadian Arc-

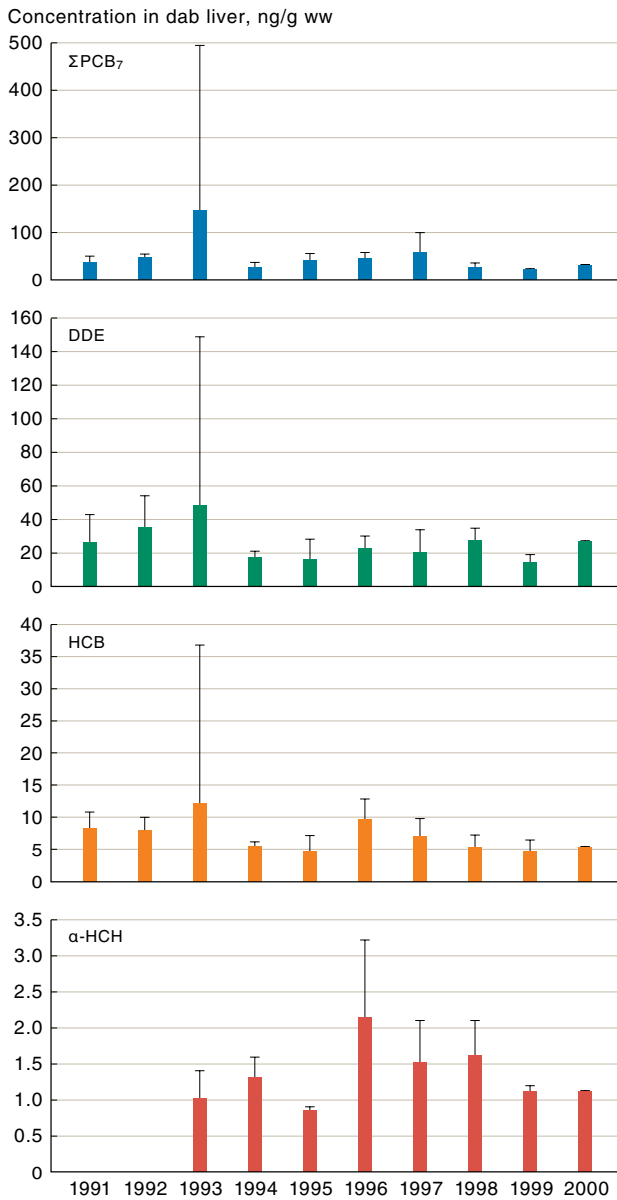


Figure 5-25. Average concentrations of OCs in the livers of dab in Icelandic waters, 1991-2000.

tic at Prince Leopold Island (Braune *et al.*, 2001a; 2001b). This study encompasses three species of seabirds and covers a long time period, 1975-1998 (Braune *et al.*, 2001a; 2001b). At the time of egg formation, OC compounds are transferred along with lipids to the eggs (Mineau *et al.*, 1984). Contaminant burden in the egg reflects residues assimilated over a long time period by the female and, particularly in migratory species, may integrate exposure from a number of different locations (Hebert, 1998; Monteiro *et al.*, 1999).

With the exception of ΣHCHs, OC compounds showed declines or, in some cases, no significant change in levels between 1975 and 1998. Levels of ΣPCBs and ΣDDTs (Figure 5-26) as well as ΣCBz decreased significantly in eggs of all three species, while ΣCHLs, dieldrin and mirex levels decreased in kittiwake eggs only. Kittiwakes, whose migration pattern would have historically brought them into closer contact with industrial sources of contaminants, such as PCBs, in the more temperate latitudes, showed the most dramatic declines through to 1998. The significant declines in

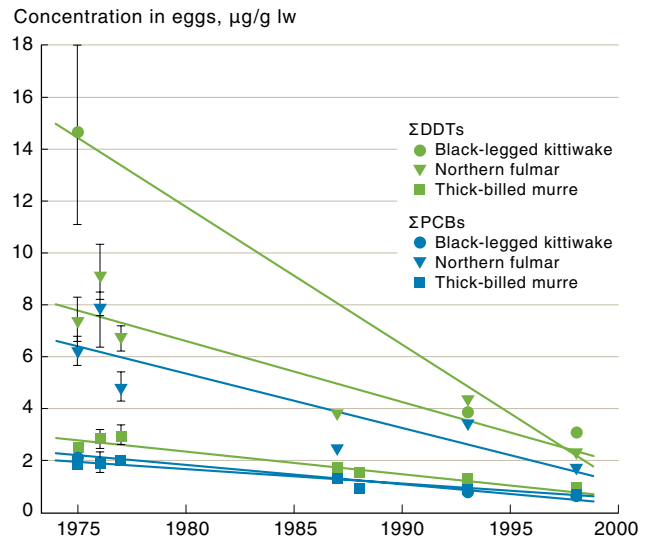


Figure 5-26. Concentrations of ΣDDTs and ΣPCBs in seabird eggs collected between 1975 and 1998 on Prince Leopold Island (Braune *et al.*, 2001a; 2001b).

concentrations of ΣPCBs and ΣDDTs in this study have also been observed in seabirds from other areas including the Baltic Sea (Olsson and Reutergårdh, 1986; Andersson *et al.*, 1988; Bignert *et al.*, 1995), the Barents Sea (Barrett *et al.*, 1996), and the Great Lakes (Hebert *et al.*, 1997). The only OC compound in this study for which a significant increase in concentrations was seen, was for ΣHCHs, particularly for β-HCH in murre (Figure 5-27) and fulmars. Stable-nitrogen isotope analyses ( $\delta^{15}\text{N}$ ) indicated that the temporal trends

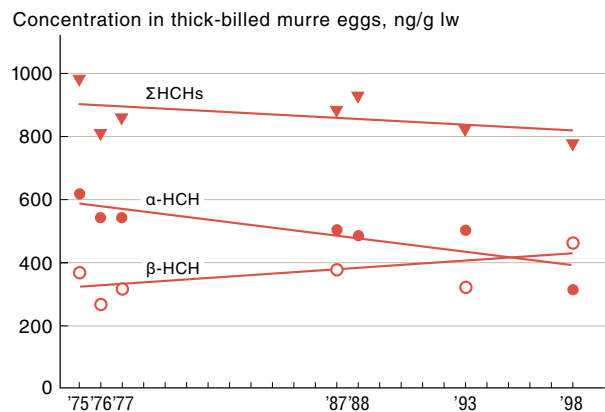
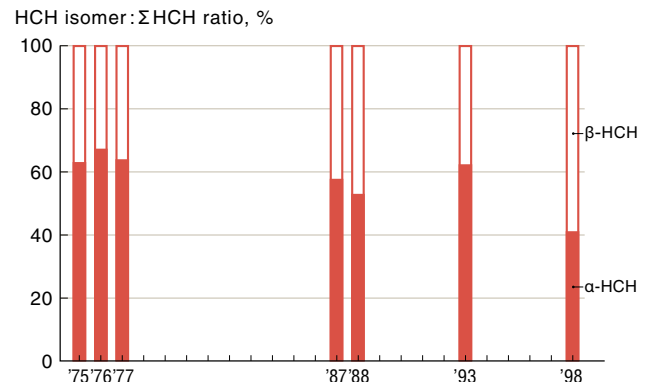


Figure 5-27. Temporal trends in relative proportions and concentrations of HCH isomers in thick-billed murre eggs. β-HCH shows a significant increase vs α- and γ-HCH (Braune *et al.*, 2001c). γ-HCH is <5% of ΣHCH.



observed in OC and trace metal concentrations in seabird eggs were not the result of shifts in trophic level over time (Braune *et al.*, 2001b). More likely, the trends reflect changes in contaminant deposition into the various marine environments that these birds occupy throughout the year, as well as the toxicokinetics of each contaminant as it is transported through the food chain.

Data from the late 1970s for two seabird colonies in the Bering Sea (common guillemot at St. George Island and thick-billed murres from Bogoslof Island) are available (Ohlendorf *et al.*, 1982) for comparison with data generated for 1999 and 2000 (Vander Pol *et al.*, 2002). There were significant ( $p < 0.05$ ) declines in mean concentrations of *p,p'*-DDE, oxychlorane and heptachlor epoxide in the common guillemot eggs, and *p,p'*-DDE and *cis*-nonachlor in the thick-billed murre eggs. HCB and dieldrin did not appear to have declined over the 20-year period. PCB concentrations were not compared due to analytical differences between the two studies. These temporal trends are consistent with what has been seen in guillemot eggs in the Baltic Sea (Bignert *et al.*, 1995) and in murres from Prince Leopold Island (Braune *et al.*, 2001b).

PCDD/Fs and toxaphene have also been measured in Canadian Arctic seabirds over a period of almost 20 years (Braune *et al.*, 2000), providing a time-series dataset. Liver samples of thick-billed murres, northern fulmars and black-legged kittiwakes collected in 1975 and 1993 from Prince Leopold Island, as well as egg samples from 1993, were analyzed for PCDDs, PCDFs and non-*ortho* PCBs (nPCBs) (Annex Table 16). In the kittiwake liver samples, concentrations of total PCDDs, PCDFs and nPCBs decreased from 1975 to 1993. In fulmar liver samples, concentrations of total PCDDs and PCDFs decreased, and nPCBs increased from 1975 to 1993. In murre liver samples, concentrations of total PCDDs and PCDFs increased and nPCBs decreased from 1975 to 1993. CB126 was the predominant nPCB congener in all years and in all Arctic seabird samples analyzed. The nPCB fraction of the TEQ decreased, and the PCDF fraction increased from 1975 to 1993 in kittiwake and murre livers. In fulmars, the PCDFs constituted the dominant fraction in both 1975 and 1993 livers. Calculated TEQ values were highest in northern fulmars both in 1975 and 1993 (Annex Table 16). The highest estimated level of toxaphene was found in the pool of kittiwake eggs from 1993 (64 ng/g ww), and the second highest level was found in the 1993 pool of fulmar eggs (53 ng/g ww), suggesting increasing concentrations.

PBDEs were also analyzed in kittiwake, northern fulmar, and thick-billed murre liver samples from 1975 and 1993 from Prince Leopold Island (Braune *et al.*, 2001a). PBDEs were below detection limits in samples from 1975 while low ng/g concentrations were found in those from 1993. These preliminary data indicate that PBDEs are increasing in concentration in seabirds as they are in other marine biota in the Canadian Arctic.

PCDDs and PCDFs, including the nPCBs (CBs 77, 126, and 169), were analyzed in guillemot eggs sampled in 1989 in Greenland, Iceland, and in the Faroe Islands (Cederberg *et al.*, 1991). Guillemot eggs were collected from the Faroe Islands in 2000 ( $n=10$ ) and

analyzed for PCDD/F (Mikkelsen, 2002). The results indicate a decrease in concentrations of PCDD/Fs, as TEQ decreased from 160 pg/g lw in 1989 to 66 pg/g lw in 2000.

#### 5.4.4. Temporal trends in pinnipeds and cetaceans

The previous AMAP POPs assessment noted that there were relatively few long-term (multi-decade) studies of POPs in Arctic marine mammals (de March *et al.* 1998). This is still the case. However, additional studies conducted using archived samples or analysis of new samples from previously studied sampling locations have significantly increased the information available.

##### 5.4.4.1. Pinnipeds

###### *Ringed seals*

Temporal trends of PCBs,  $\Sigma$ DDTs,  $\beta$ -HCH, and  $\gamma$ -HCH have been studied in ringed seals from the Canadian Arctic, sampled at the communities of Ausuittuq (Grise Fiord), Ikpiarjuk (Arctic Bay), and Holman (Addison and Smith, 1998; Muir *et al.*, 2001c). In the case of Holman, the results are part of a long-term study (Addison and Smith, 1998). Elsewhere, sampling locations were selected based on whether previous results were available (Muir *et al.*, 1988; Weis and Muir, 1997). At all three locations, results were available from the early 1970s to the late 1990s and 2000.

At all locations, only samples from female seals were selected for analysis in order to minimize age-related effects on OC levels (Addison and Smith, 1974). Mean concentrations ( $\pm 95\%$  confidence limits; ng/g lw) in ringed seals for  $\Sigma$ DDTs, PCBs,  $\Sigma$ HCHs and  $\Sigma$ CHLs, along with ratios of recalcitrant members of each class for each location, are shown in Figure 5-28 a-c.  $\Sigma$ PCB<sub>10</sub> (sum of CBs 28, 31, 52, 101, 105, 118, 138, 153, 156, and 180) in seals from Ikpiarjuk declined significantly (2.4 fold) from 1975 to 2000 (Figure 5-28 a) and 1.5 fold at Ausuittuq, based on the comparison of arithmetic means.  $\Sigma$ PCB<sub>10</sub> was used for comparison with earlier data rather than all congeners. All previous results, including the samples from 1972-1975, were based on capillary gas chromatography with quantitation using authentic standards (Muir *et al.*, 1988; 1999b; Muir, 1996; Weis and Muir, 1997; Muir *et al.*, 1999b).

At Holman,  $\Sigma$ PCBs (based on conversion of Aroclor 1254 to a  $\Sigma$ PCB value consisting of 20 major congeners) was significantly higher in 1972 than in 1981 and declined significantly over the ten-year period from 1991 ( $510 \pm 133$  ng/g lw) to 2001 ( $335 \pm 18$  ng/g lw). The overall decline for PCBs is 5.5 fold. Ratios of CB153:  $\Sigma$ PCBs increased over the 25-year period at Ikpiarjuk and Ausuittuq and between 1991 and 2001 at Holman, reflecting the slower elimination of CB153 over time by the seals, compared to other PCBs.

$\Sigma$ DDTs declined significantly in female ringed seals from all three sites, between the early/mid-1970s and late 1990s/2000.  $\Sigma$ DDTs exhibited the largest decline of any 'legacy' OC: 2.5 fold at Ausuittuq, 3.3 fold at Ikpiarjuk and 3.3 fold at Holman over the 25-30-year period. Significant increases in *p,p'*-DDE:  $\Sigma$ DDT ratios were also found at all three locations, reflecting the shift from fresh DDT to 'weathered' or degraded older sources.

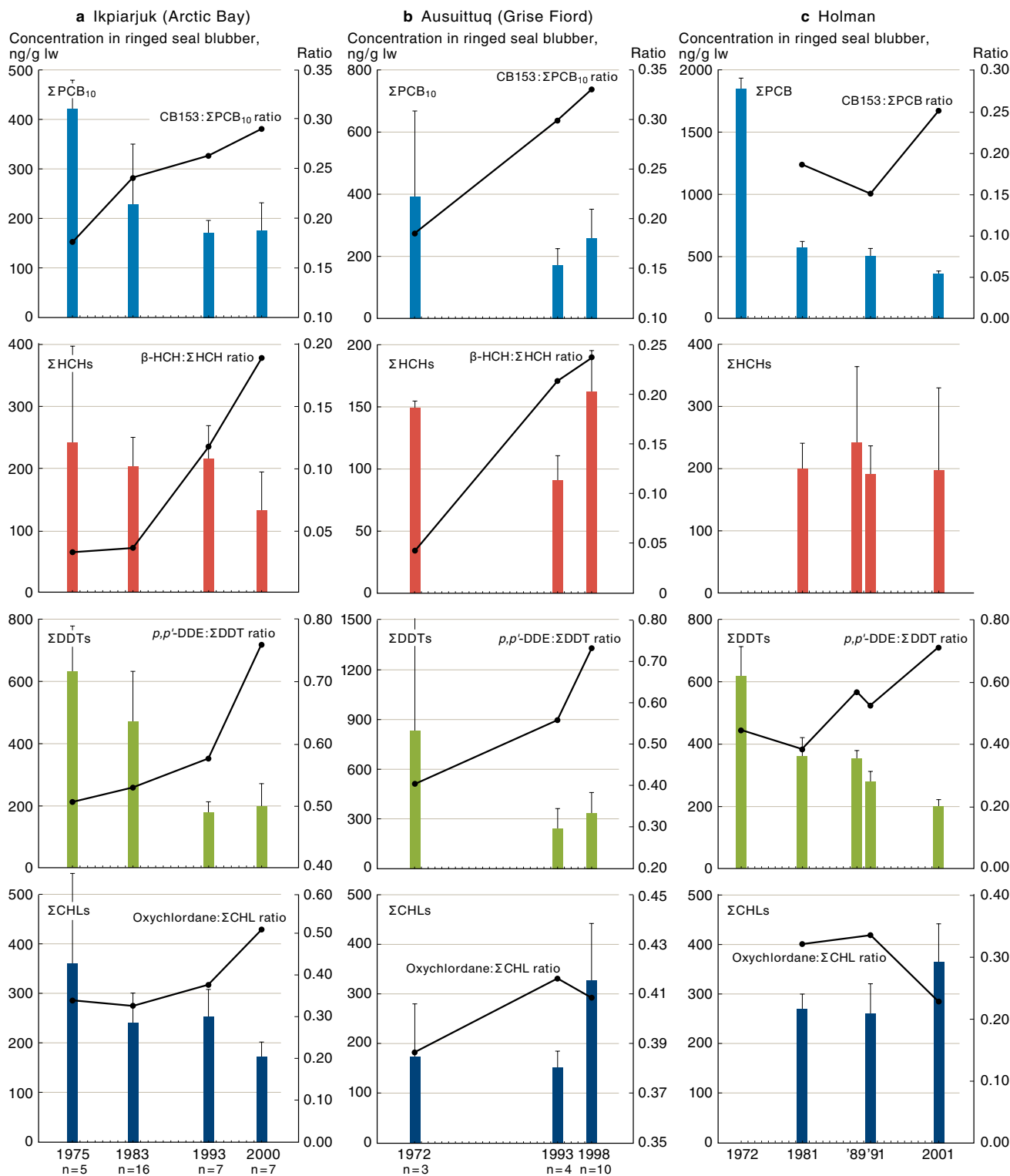


Figure 5-28. Temporal trends in concentrations and proportions of major OC components in blubber of female ringed seals from three sites in the Canadian Arctic Archipelago. Bars are arithmetic means and vertical lines are 95% confidence intervals. a) Ikpiarjuk (Arctic Bay); significant differences between 1975 and 1993 were found in the case of  $\Sigma\text{PCB}_{10}$  and  $\Sigma\text{DDTs}$ . b) Ausuittuq (Grise Fiord); no significant differences over time were found because of small sample sizes from 1972 and 1993. Nevertheless, the results suggest similar trends to those observed at Ikpiarjuk for concentrations and proportions of major components. c) Holman, NWT, in the western Canadian Arctic Archipelago. At Holman,  $\Sigma\text{PCB}$  includes all congeners analyzed.

$\Sigma\text{HCH}$  concentrations showed no significant changes in concentrations from the 1970s (1981 in the case of Holman) to 2001. However,  $\beta\text{-HCH}:\Sigma\text{HCH}$  ratios increased (3 fold at Ikpiarjuk). This shift in the composition of HCH, with higher proportions of  $\beta\text{-HCH}$ , has also been reported in seawater in the western Canadian Arctic during the 1980s and 1990s (Li *et al.*, 2002) (see Section 4.4.1.2).

$\Sigma\text{CHLs}$  showed quite a different trend from  $\Sigma\text{DDTs}$ , with increasing concentrations at Holman and Ausuittuq, and a slow decline (2.1 fold over 25 years) at Ikpiarjuk. Proportions of oxychlordane, a recalcitrant metabolite of chlordane in mammals, increased at all three locations.

Ikonomou *et al.* (2002) examined temporal trends of non- and mono-*ortho* PCB and PCDD/F concentrations

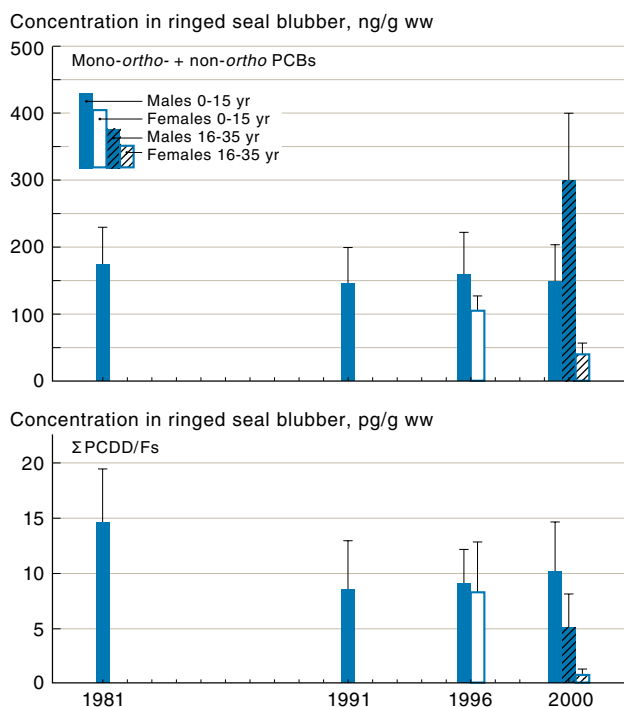


Figure 5-29. Concentrations of mono-ortho- and non-ortho-PCBs and PCDD/Fs in ringed seals from Holman, NWT, in the Canadian Arctic Archipelago (Ikononou *et al.*, 2002).

in male ringed seals from Holman. Concentrations of non- and mono-ortho PCBs (149-174 ng/g ww) and PCDD/Fs (8.6-14.6 pg/g ww) in ringed seals aged 0-15 years remained approximately constant from 1981 to 2000 (Ikononou *et al.*, 2002) (Figure 5-29). Total PCB concentrations did not decline significantly in males or females over the period of 1981 to 1991 (Addison and Smith, 1998). Older male seals (16-35 yr) from the 2000 sampling group have higher levels of non- and mono-ortho PCBs than their younger counterparts (0-15 yr; 302 vs. 150 ng/g ww). In female seals from 1996 and 2000, non- and mono-ortho PCB levels are much lower in the 16-35-year age group from 2000 (43 ng/g ww) than the 0-15-year age group from 1996 (105 ng/g ww).

No other temporal-trend studies of OCs in Arctic ringed seals have been reported. At Svalbard and northern Norway, where results are available since the mid-1990s, it is likely that this will be accomplished in the near future. In the White Sea, mean levels of PCBs, DDTs, chlordanes, HCHs, and HCB declined between 2 and 3 fold in blubber samples taken from harp seal pups between 1992 and 1998 (Muir *et al.*, 2002c). Mirex levels in 1998 samples were about one quarter of their 1992 levels, in the same study. These declines should be viewed with caution since there could be regional differences in exposure of adult harp seals to PCBs and DDT within the White Sea area depending on their proximity to urban areas.

No significant temporal trend in butyltin concentrations was observed in Steller sea lions sampled from Alaska between 1976 and 1985 (Kim *et al.*, 1996a). This was despite the fact that the annual consumption of organotin compounds doubled in the U.S. during the same period. The authors suggest that the butyltin compounds are degraded faster than the intake from diet in Steller sea lions.

Muir *et al.* (2000e) examined temporal trends in levels of a wide range of OC compounds in archived (1978) samples of northwestern Greenland walrus as well as more recently collected samples (1988) (Annex Table 12). They did not detect any significant differences in mean concentrations of any OCs in male walrus from the two time periods, but for females, they found significantly higher levels of di- and trichlorobiphenyls, dieldrin, toxaphene,  $\alpha$ -HCH, and  $\Sigma$ HCHs in the 1988 material, but no differences for PCBs or DDT compounds.

#### 5.4.4.2. Cetaceans

##### *Belugas*

Stern (1999) and Stern and Addison (1999) examined temporal trends of 'legacy' OCs in blubber of beluga from Cumberland Sound (southeast Baffin Island) between 1982 and 1997 (Annex Table 13). They reported a significant decline in  $\alpha$ -HCH concentrations over the 15-year interval from 1982 to 1997, while no significant differences were observed for the  $\beta$ - and  $\gamma$ -HCH isomers (Figure 5-30). In this regard, belugas differ from seabirds, ringed seals and polar bear, which all show increasing proportions of  $\beta$ -HCH.  $\Sigma$ DDT concentrations did not change over this 15-year interval. However, a 2.2-fold decline and a 1.3-fold increase in the (age-adjusted) concentration of  $p,p'$ -DDT and its metabolite  $p,p'$ -DDE, respectively, was observed (Figure 5-31). These changes translated into a significant increase in the  $p,p'$ -DDE: $\Sigma$ DDT ratio from 0.37 to 0.48 and suggests 'old' rather than recent DDT sources. Two of the most abundant congeners in technical chlordane, *cis*- and *trans*-nonachlor, increased in concentration by 1.4 and 1.7 fold, respectively, from 1982 to 1997, while only *cis*-chlordane showed any significant decline. Overall, a 1.2-fold increase in the concentrations of  $\Sigma$ CHLs

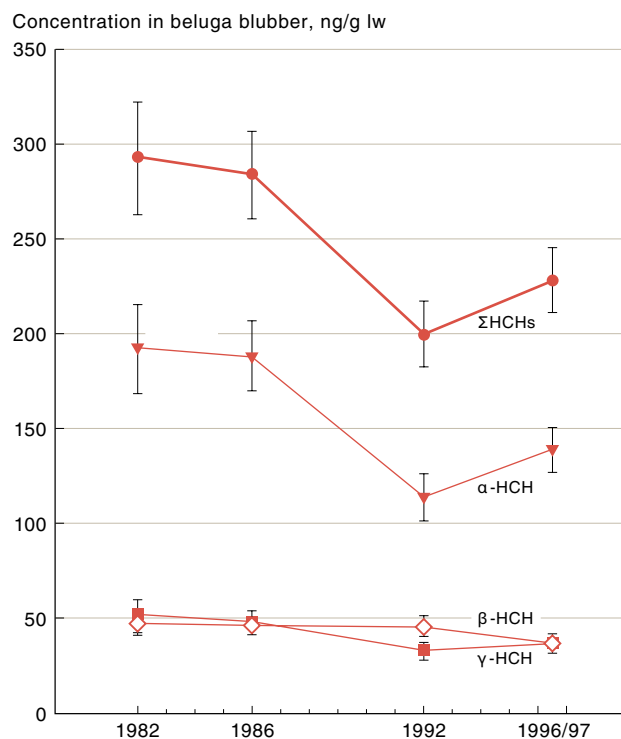


Figure 5-30. Temporal trends of age-adjusted concentrations of  $\Sigma$ HCHs,  $\alpha$ -HCH,  $\beta$ -HCH, and  $\gamma$ -HCH in male beluga blubber samples from Pangnirtung, Nunavut.

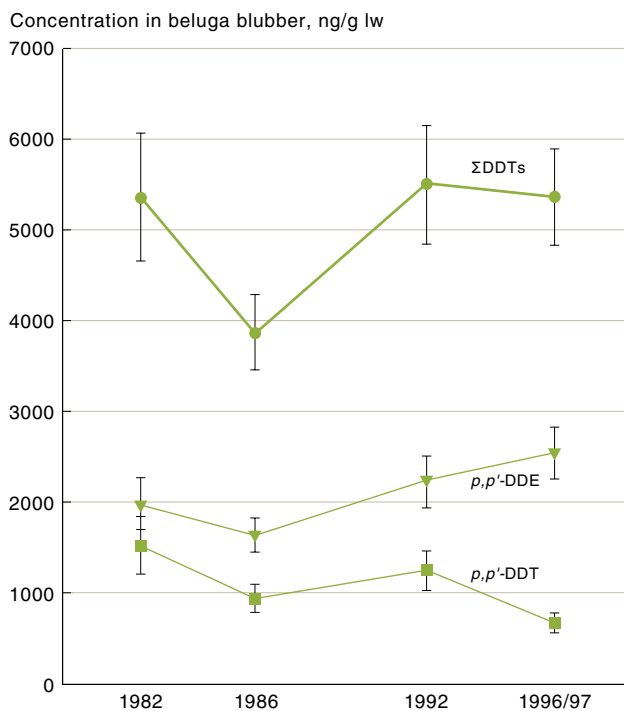


Figure 5-31. Temporal trends of age-adjusted concentrations of ΣDDTs, p,p'-DDE and p,p'-DDT in male beluga blubber samples from Pangnirtung, Nunavut.

was observed. Levels of oxychlordan, the principal metabolite of *cis*- and *trans*-chlordan, second only to *trans*-nonachlor as the most abundant chlordan-related residue in the southeast Baffin beluga blubber, did not change significantly over this 15-year period. For total toxaphene and Parlars 26 and 50, no clear trends were evident (Figure 5-32). For PCB congeners, significant

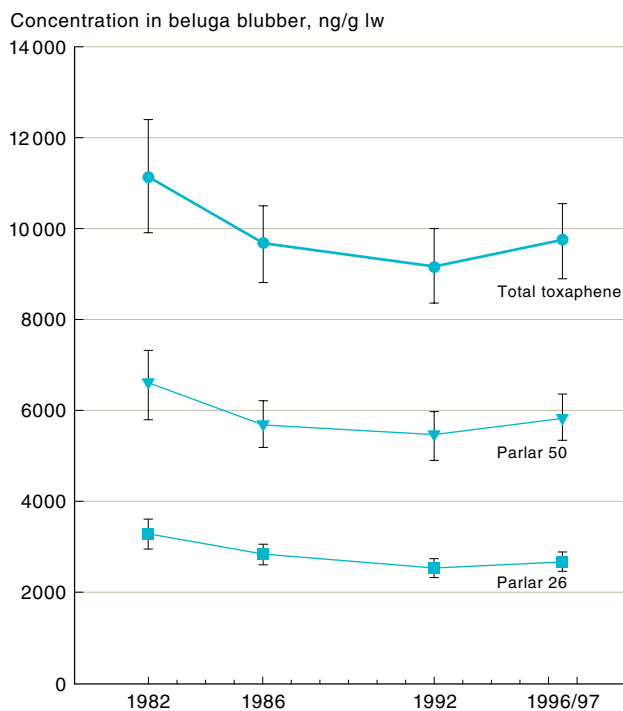


Figure 5-32. Temporal trends of age-adjusted concentrations of total toxaphene and Parlars 50 and 26 in male beluga blubber samples from Pangnirtung, Nunavut.

declines, ranging from 1.7 fold for CB81 to 2.8 fold for CB126, were observed (Figure 5-33). Non-ortho PCB TEQs (CBs 77, 126, and 169) declined from 16 to 6.1 pg/g lw (2.6 fold) from 1982 to 1997. Age-adjusted concentrations of major PCB homologue groups (hexa-, heptachlorobiphenyls) did not show a consistent decline over the 15-year period (Stern and Addison, 1999).

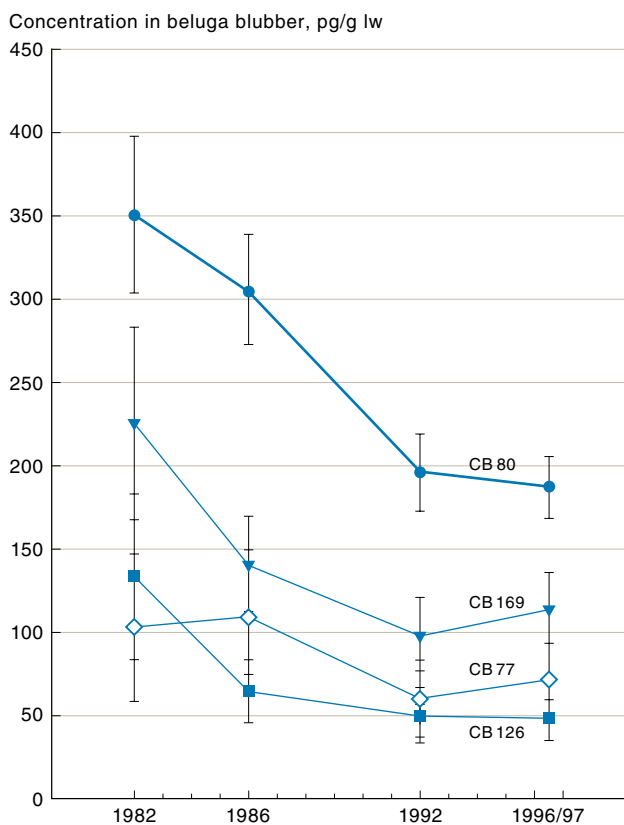
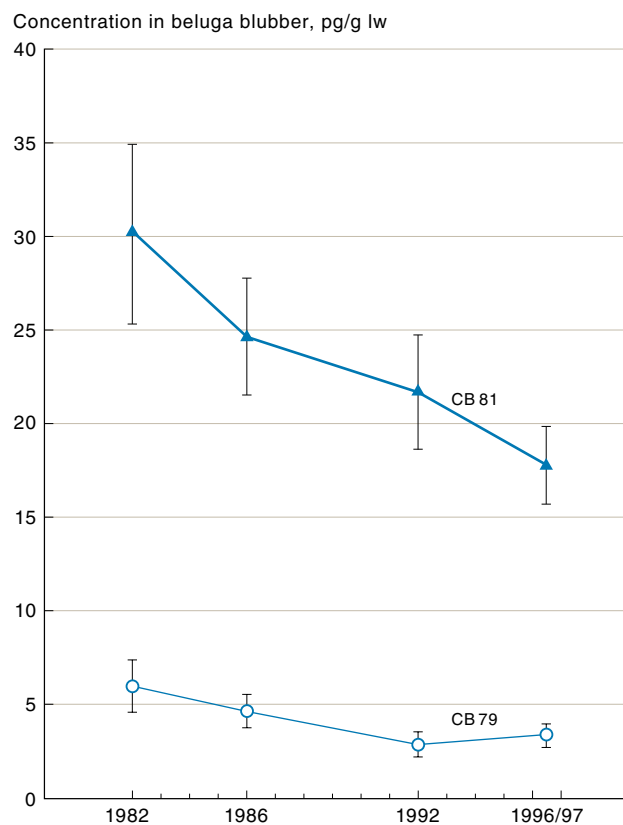


Figure 5-33. Temporal trends of age-adjusted concentrations of PCB congeners 77, 79, 80, 81, 126, and 169 in male beluga blubber samples from Pangnirtung, Nunavut.





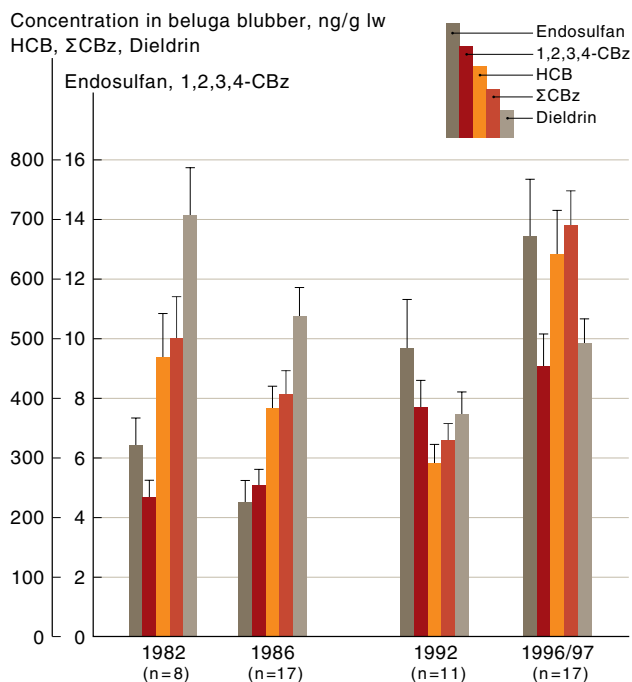


Figure 5-34. Temporal trends of age-adjusted concentrations of endosulfan, 1,2,3,4-chlorobenzenes, HCB, sum of tetra-, penta- and hexachlorobenzene ( $\Sigma$ CBz), and dieldrin in blubber of male belugas from Cumberland Sound, Nunavut, in the eastern Canadian Arctic (Stern and Addison, 1999).

A 2.1-fold increase in age-adjusted mean concentrations of endosulfan sulfate and a 1.4-fold decline in dieldrin were also observed over the 15-year period (Stern and Addison, 1999) (Figure 5-34).

#### Pilot whales

In 1997, a large number of pilot whale blubber samples were taken in the Faroe Islands to study levels of OCs (Dam and Bloch, 2000). Samples were analyzed in pools sorted according to sexual maturity and sex. Temporal trends were examined by comparing results from a subset of individual 1997 pilot whale samples (Dam, 2001) with a previous study of pilot whales collected in the Faroes in 1987 (Borrell, 1993). Statistical analysis of the data for the adult females and adult males from 1987 and 1997 showed a significant decrease during the ten-year interval for both PCB and DDT concentrations (Mikkelsen, 2002). There are some weaknesses in the statistical analysis since the total PCB concentration in the subset of adult females analyzed individually was slightly lower than in the entire 1997 bulk of adult females. The level was quite similar in the adult males, however, and this indicates that the observed decrease is real.

#### Narwhal

Results for OCs in blubber of narwhal from the Baffin Bay-Lancaster Sound region (Pond Inlet) of the Canadian Arctic were available from previous work (Muir *et al.*, 1992a), and more recent data for the same region are available (Stern, 2001). Using this information, it was possible to make a qualitative assessment of temporal changes in 'legacy' OCs in narwhal from this region over a 12-16-year period (1982-83 to 1999). Male narwhal were selected for temporal-trend comparisons because, unlike seals, they show little relationship between PCB concentrations and age (Muir *et al.*, 1992a,b). In agree-

ment with findings outlined in the previous AMAP assessment report (de March *et al.*, 1998), male narwhal from Pond Inlet showed no discernible changes in concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs or toxaphene.  $\Sigma$ HCHs and  $\Sigma$ CBz also showed no statistically significant trends over this time period. This lack of change in  $\Sigma$ DDTs,  $\Sigma$ CHLs,  $\Sigma$ PCBs, and toxaphene concentrations in narwhal is consistent with results for beluga in south-eastern Baffin Island. One difference however between these two odontocetes was the decline in  $\Sigma$ HCHs in beluga, while no such decline was found in narwhal.

### 5.4.5. Temporal trends in polar bear

#### 5.4.5.1. Canadian Arctic polar bears

Temporal trends of the major OC groups in polar bears from the Churchill area of western Hudson Bay (Canada) are presented in Figure 5-33. Biopsy samples from adult female polar bears were chosen to study temporal trends because there is no significant effect of age on OC levels, whereas there is a significant effect (increase with age) of highly chlorinated PCBs in male polar bears (Bernhoft *et al.*, 1997; Norstrom *et al.*, 1998). Bears less than five years old were also excluded because levels of some OCs are higher due to high exposure in milk the first two years of life. The trends observed in adult female polar bears are therefore more likely to represent the actual changes in OC levels in the polar bear food chain. Biopsy samples were collected opportunistically nearly every year throughout the 1990s (Norstrom, 2001). Archived adipose tissue samples from 1968, 1984, and 1989 were included to extend the time period for comparison. There were no consistent upward or downward trends over the period 1968 to 1989 for most OCs; therefore, only trends in the 1990s were analyzed statistically. There were no statistically significant ( $p < 0.05$ ) increasing trends in female bears over the entire nine-year period. There were however statistically significant ( $p < 0.05$ ) downward trends for total chlorobenzenes,  $\alpha$ -HCH, and  $\Sigma$ PCBs between 1991 and 1999. There were no significant ( $p < 0.05$ ) changes in chlordanes,  $\Sigma$ DDTs (mostly  $p,p'$ -DDE), dieldrin,  $\beta$ -HCH, and  $\Sigma$ HCHs.

There was a consistent (and significant) decrease in  $\Sigma$ DDTs throughout the entire 1968 to 1990s time period. Such strong trends are unusual in Arctic biota, suggesting that the phenomenon may be related to local conditions around Hudson Bay. Spraying of DDT for biting-insect control in communities and military bases in the Hudson Bay area in the 1950s and 1960s may have contributed a significant load to Hudson Bay during this period. This likely declined after the DDT ban and closing of the large military base at Churchill, and may not have contaminated other nearby areas. In support of this hypothesis, it can be seen in Annex Table 14 that  $\Sigma$ DDT levels were 2-3 times higher in polar bear fat from western Hudson Bay than in other areas of the Canadian Arctic in 1984.

The overall trend for  $\Sigma$ CBz appears to be an increase between 1968 and 1984, followed by a consistent downward trend since that time. Most of the decline in  $\Sigma$ CBz was due to HCB, which decreased with a half-life of approximately nine years during the 1990s. HCB and 1,2,4,5-TeCBz were each roughly half of the total, with a minor contribution from PeCBz. The pro-

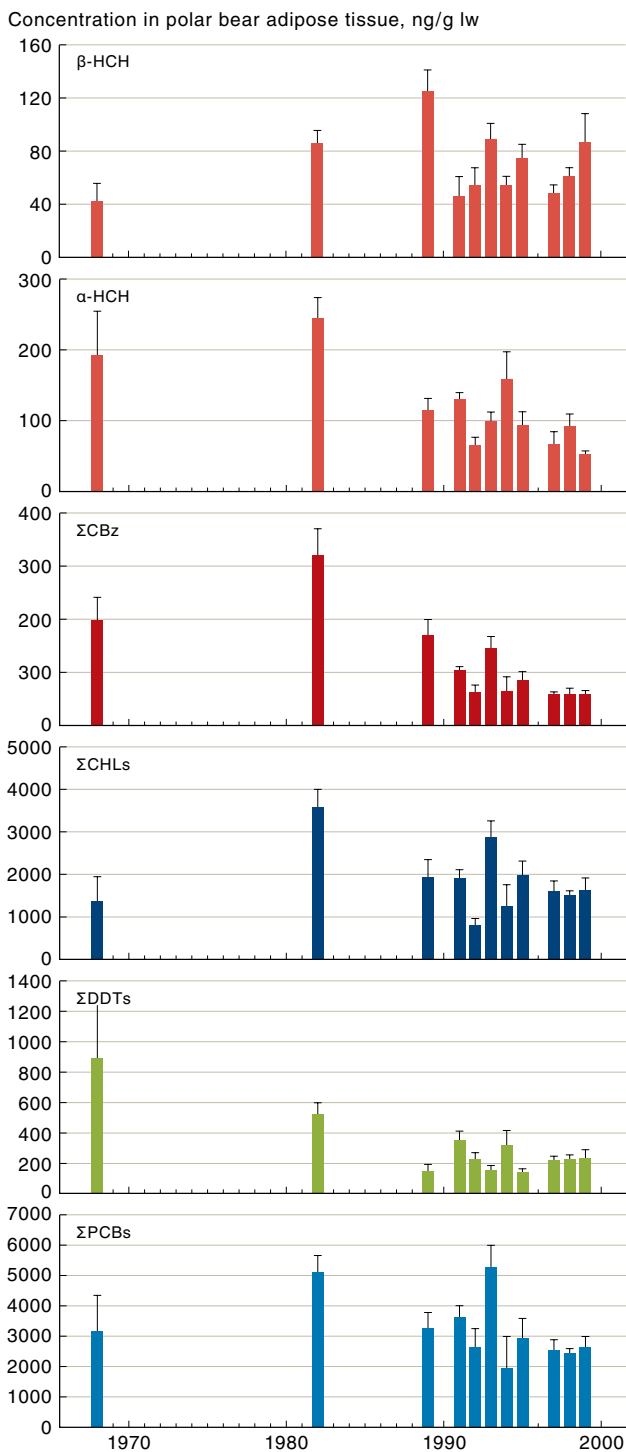


Figure 5-35. Trends of major OC compounds in female polar bear adipose tissue from the Churchill area of western Hudson Bay (Canada) from 1968 to 1999. Samples from 1991 to 1999 are fat biopsies, but earlier samples are adipose tissue.

portion of 1,2,4,5-TeCBz peaked at 53% between 1995 and 1997, and then decreased to values similar to the pre-1995 period of 40-45%.

The downward trend of  $\Sigma$ HCHs in Hudson Bay polar bears in the 1990s was not significant (Norstrom, 2001), but it was significant if 1984 and 1989 were considered (Figure 5-35). However, comparing  $\Sigma$ HCH trends is complicated by differences in temporal trends of  $\alpha$ -HCH and  $\beta$ -HCH ( $\alpha$ -HCH declines and  $\beta$ -HCH increases).  $\gamma$ -HCH was less than 2% of  $\Sigma$ HCHs and could not be reliably quantified in the majority of samples.

The apparent half-life of  $\alpha$ -HCH in the 1990s was approximately ten years, very similar to that of HCB. The decrease in the 1990s appears to be part of a general trend. Levels of  $\alpha$ -HCH were approximately two to three times higher in 1968 and 1984 than the average in the 1990s. By contrast,  $\beta$ -HCH concentrations were lower in 1968 than in any subsequent year, and the overall trend in the 1980s and 1990s is rather flat. As a consequence, a significantly higher proportion (approximately 50%) of present day  $\Sigma$ HCHs in polar bears from Hudson Bay is  $\beta$ -HCH compared to 1984 (25%) and 1968 (17%). A parallel trend is observed in ringed seals, the major prey of the polar bears (Section 5.4.4.1).

$\Sigma$ PCBs decreased fairly steadily throughout the 1990s, but with a long half-life of approximately 18 years. The shift in composition of the PCBs was subtle over the decade, but there was a clear tendency for the proportion of less chlorinated congeners to increase, and the highly chlorinated congeners to decrease. Thus, CB99 increased from approximately 10% to 12% of  $\Sigma$ PCBs, CB153 was relatively stable at approximately 35%, and CB180 decreased from 17% to 14%. The trends in these three congeners indicate that the half-life of CB153 (19 years) was similar to that of  $\Sigma$ PCBs. The half-life of CB180 (13 years) was shorter, and the half-life of CB99 (>50 years, not significant) was longer than  $\Sigma$ PCBs. Thus, the decreasing trend in  $\Sigma$ PCBs is driven by loss of the highly chlorinated congeners. There is less than a factor-2 difference in  $\Sigma$ PCB levels through the 1968-1999 period, and no long-term trend is apparent. Concentrations in the early 1990s in Hudson Bay were similar to those in the late 1960s, in sharp contrast to areas such as the Great Lakes or the North Atlantic, where PCB concentrations in herring gull eggs were on the order of ten times higher in the late 1960s and early 1970s than at present (Hebert *et al.*, 2000).

Shorter-term temporal trends in polar bears from Queen Maud Gulf (I) in the western Canadian Arctic Archipelago, Barrow Strait (II) in the central archipelago, and northern Baffin Bay (III) and Davis Strait (IV) in the eastern archipelago, and also one area in western Hudson Bay (V) were reported by Muir and Norstrom (2000) (Annex Table 14). In that comparison, samples were taken from eight to ten individual adult male polar bears per area in each of 1984 and 1989/1990. PCDD/PCDF concentration changes were also determined in pooled samples. There were significantly lower concentrations of  $\Sigma$ HCHs,  $\Sigma$ DDTs, and dieldrin in (I), a significant decrease in dieldrin in (II), no changes in (III), a significant increase in  $\Sigma$ CHLs in (IV), and a significant decrease in  $\Sigma$ CBz,  $\Sigma$ DDTs, and  $\Sigma$ PCBs in (V), between 1984 and 1989.

Results for  $\Sigma$ PCDD concentrations in pooled samples of polar bear adipose tissue from polar bear areas (I), (II), (III), and (IV) in the Canadian Arctic Archipelago in 1984 and 1990 (Norstrom, 1997) are given in Annex Table 16. PCDDs consisted almost entirely of 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD. Other PCDDs and all PCDFs were at sub-pg/g lw concentrations. Concentrations of TCDD were low and tended to be more evenly distributed in 1990 (0.8-3.1 pg/g lw) than in 1984 (2.0-15 g/g lw), as was the case for the other OCs. Because pooled samples were analyzed, the significance of the differences cannot be established. Nevertheless, con-

centrations of TCDD in (I) and (II) were a factor of 4-5 lower in 1990 than 1984. The results are consistent with the findings of Norstrom *et al.* (1990) in which ringed seal, beluga, and polar bears were analyzed. It is apparent that PCDD/Fs are at very low concentrations in the polar bear food chain and are unlikely to be of toxicological significance.

#### 5.4.5.2. East Greenland polar bears

Dietz *et al.* (2004) compared concentrations of PCBs and OC pesticides in polar bear from central East Greenland collected in 1999-2001 with results for 1990 reported by Norstrom *et al.* (1998). Overall a significant decrease was observed in OC contaminants in all age and sex groups studied over the period from 1990 to 1999-2001. Using the same congeners to calculate  $\Sigma$ PCB for both groups,  $\Sigma$ PCB showed a reduction of 78%. PCB congener proportion also changed over the ten-year period. A less chlorinated PCB congener like CB99 comprised 7.3% of the 1999-2001 bears from East Greenland, a slight decrease compared to the 8.3% that Norstrom *et al.* (1998) reported from the trans-Arctic survey of polar bears from 1989 to 1992. The decreases in  $\Sigma$ DDT and *p,p'*-DDE in East Greenland since 1990 (34% and 29%, respectively) were also significant, although less pronounced than for other OCs, and the estimated half-life for *p,p'*-DDE was the longest of all OCs measured (17.5 years). Total chlordane-related compounds and dieldrin were both found to decrease (between about 31% to 75% dependent of the sex and age group studied).  $\Sigma$ HCH concentrations declined about 60% over the ten-year period.  $\beta$ -HCH increased in western Hudson Bay polar bears during the 1990s (Norstrom, 2001) and a similar pattern was expected in East Greenland. However, this was not the case, as  $\alpha$ -HCH showed an increase from 18% to 25% and  $\beta$ -HCH showed a decrease from 82% to 75%. The high percentage of  $\beta$ -HCH is consistent with findings from the Svalbard region where Bernhoft *et al.* (1997) reported  $\beta$ -HCH to constitute 81% of  $\Sigma$ HCH. In general, the decrease in PCB and OC concentrations in East Greenland polar bear is faster than that observed over the same period in Canada, but similar to the decrease found in polar bears from Svalbard.

Relative change in concentration in polar bear plasma from 1967-1993/94

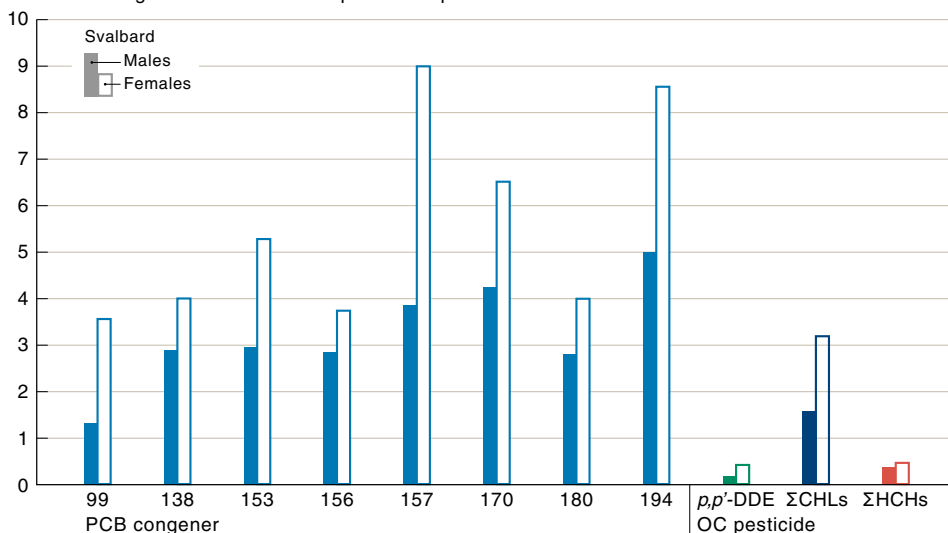


Figure 5-37. Relative change in major PCB congeners and OC pesticides from 1967 to 1993-94 for adult female and male polar bear samples at Svalbard, Norway (from Derocher *et al.*, 2003). Results for 1993-94 are from Skaare *et al.* (2001d) and Bernhoft *et al.* (1997). Values <1 imply a decrease in concentration.

Concentration in polar bear plasma, ng/g ww

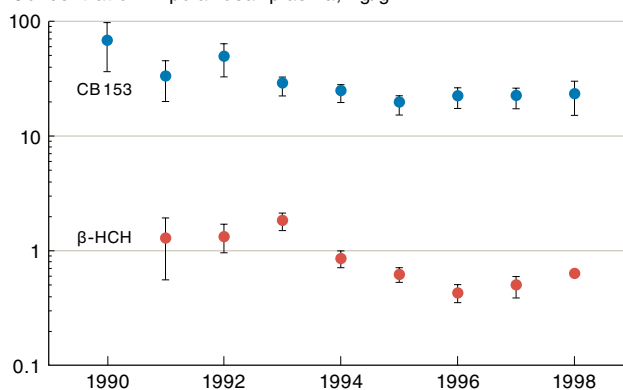


Figure 5-36. Temporal trends of two major OCs ( $\beta$ -HCH and CB153) in polar bear plasma from Svalbard (means  $\pm$  95% C.I.) for the period 1990-1998. CB153 concentrations are adjusted for covariates (extractable lipids, condition index and sampling site longitude).

#### 5.4.5.3. Svalbard polar bears

Henriksen *et al.* (2001) studied the trend in CB153 concentrations in blood from polar bears at Svalbard that was collected annually between 1990 and 1998. Decreases of approximately 40% occurred in the early 1990s, and stabilized thereafter (Figure 5-36). This is a somewhat steeper trend than was found in Hudson Bay during the same time period. However, Svalbard bears had significantly higher levels of PCBs in 1990 than those in Hudson Bay, probably due to the proximity of Svalbard to European sources. Svalbard PCB levels may have approached steady state with global distribution of PCBs later than in Hudson Bay because of this proximity. It is well established that trends of POPs in environments close to sources tend to follow production and use of the chemical rather closely, whereas those in environments remote from sources are significantly dampened. That is, peak concentrations are lower, took longer to be reached, and take longer to decrease than those near sources. Present results indicate that further decreases in PCB contamination of the Arctic may be slight.

$\beta$ -HCH also declined in plasma of polar bears from Svalbard between 1991 and 1999 (Lie and Skaare, 2002). Concentrations were similar from 1991 to 1993 and declined about 3 fold between 1993 and 1996 (Figure 5-36).

The trend for  $\beta$ -HCH in Svalbard polar bears differs from that in the Hudson Bay bears, where concentrations appeared to increase during the same time period.

Derocher *et al.*, (2003) compared PCBs and OC pesticides in blood plasma collected in 1967 from 32 polar bears in eastern Svalbard with results from 1993 and 1994. Most major, persistent, PCB congeners in polar bears (CBs 99, 138, 153, 180, 194) showed significant increases from 1967 to 1993-94 in both males and females (Figure 5-37). Chlordane compounds also increased while *p,p'*-DDE and HCH isomers declined. These increases for chlordanes, PCBs and decline in *p,p'*-DDE over the 26-27-year period were similar to trends in the eastern Canadian Arctic polar bears over the period 1969 to 1984 (Muir and Norstrom, 2000) (Figure 5-35).

#### 5.4.6. Temporal trends of 'new' POPs in marine mammals

##### 5.4.6.1. PBDEs

Ikonomou *et al.* (2002) reported exponential increases in  $\Sigma$ PBDEs (di- to heptabromodiphenyl ethers) in male ringed seals aged 0-15 years from Holman in the western Canadian Arctic over the period of 1981 to 2000 (Figure 5-38).  $\Sigma$ PBDEs increased 9 fold over this period. Pe- and HxBDEs were found to be increasing at approximately the same rate ( $t_2 = 4.7$  yr and 4.3 yr, respectively) and more rapidly than TeBDEs ( $t_2 = 8.6$  yr), while TrBDEs showed no increase in this age/sex group. The

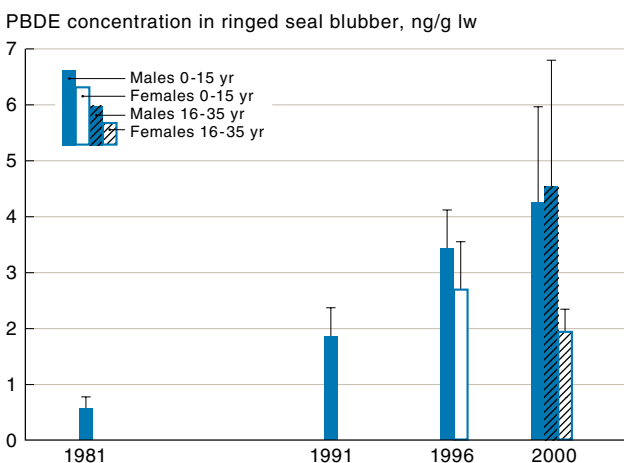


Figure 5-38. Temporal trends of PBDEs in blubber of ringed seals from Holman, NWT, in the western Canadian Arctic, 1981-2000 (Ikonomou *et al.*, 2002).

three most prevalent PBDE congeners: BDEs 47, 99, and 100, all increased over the 19-year period. However, only BDEs 47 and 100 increased in parallel with  $\Sigma$ PBDEs. BDE99 increased exponentially in a similar manner to  $\Sigma$ PBDEs and BDEs 47 and 100 from 1981 to 1996. However, the 2000 samples show that the levels of BDE99 are stabilizing. This suggests a shift in sources or change in composition of PBDE products. No difference in PBDE levels (both total and of individual congeners), were observed between younger (0-15 yr) and older (16-35 yr) male seals in 2000 ( $p = 0.98$  for  $\Sigma$ PBDEs), suggesting that, for the older seals, recent PBDE accumulation dominates potential historic accumulation.

Stern and Ikonomou (2000; 2001) studied temporal trends of PBDEs and polychlorinated diphenyl ethers

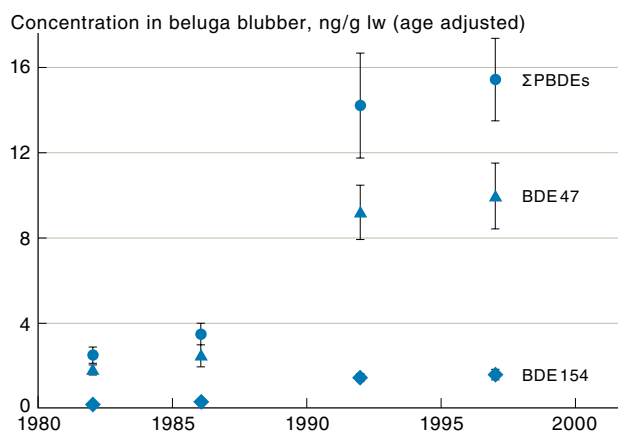


Figure 5-39. Temporal trends of PBDEs in blubber of beluga whales from southeast Baffin Island in the eastern Canadian Arctic, 1982-1997 (Stern and Ikonomou, 2000; 2001).

(PCDEs) in beluga blubber samples from southeast Baffin Island in the eastern Canadian Arctic. Levels of the total PBDEs (Di- to HpBDEs) and major congeners increased significantly in the southeast Baffin Bay beluga over the period of 1982 to 1997. Age-adjusted concentrations of BDE47, the most predominant PBDE congener, increased 6.5 fold over this 15-year period, while BDE154 increased 30 fold (Figure 5-39). Over the 15-year time span, contributions of the TrBDE homologue group and BDE47 to total PBDEs declined by 7% and 3%, respectively. Conversely, PeBDE and HxBDE contributions have increased by 20% and 80%, respectively. This change in the beluga could be related to the shift in composition of commercial PBDEs to more highly brominated mixtures (de Boer *et al.*, 2000).

##### 5.4.6.2. PCDEs

PCDE concentrations declined in the same beluga samples that were analyzed for PBDEs (Stern and Ikonomou, 2000). Maximum concentrations were found in samples from 1982 (Figure 5-40). The two most abundant congeners were CDE99 (2,2',4,4',5-CDE) and CDE154 (2,2',4,4',5,6'-CDE), which declined 2.5 fold and 1.8 fold, respectively, over the 15-year period. These congeners are prominent contaminants in pentachlorophenol-based wood preservatives, and the decline most likely reflects the ban on PCP use in Canada and use restrictions in the U.S.

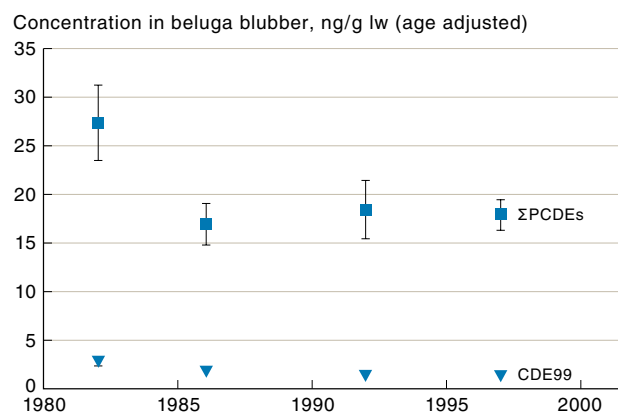


Figure 5-40. Temporal trends of PCDEs in blubber of beluga from southeast Baffin Island in the eastern Canadian Arctic, 1982-1997 (Stern and Ikonomou, 2000; 2001).



#### 5.4.7. Modeling temporal trends of PCBs and DDT in pinnipeds

Evaluating temporal trends of OCs in marine mammal populations, and linking them to trends in other biota or environmental media can be confounded by factors that affect tissue concentrations such as age, life span, sex, reproductive activity, and blubber thickness. OC concentrations in marine mammals also reflect their exposure history over many years, which could result in a significant lag in response to changes in their exposure levels. These factors can be accounted for quantitatively using species-specific dynamic bioaccumulation models as has been shown in an examination of the history of PCB contamination in the St. Lawrence beluga whale population (Hickie *et al.*, 2000). Similar individual- and population-based models developed for ringed seals (Kingsley and Hickie, 1993) are used here to reconstruct the history of accumulation of selected POPs ( $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs,  $\alpha$ -HCH, and  $\beta$ -HCH), and to predict their potential future trends in Arctic populations. In Figure 5-41, some results for males are compared to temporal-trend data for ringed seals from Holman Island, NWT from 1972 to 1991 (Addison and Smith, 1998) and from 2001 (Hoekstra *et al.*, 2003a).

For these simulations, the entire diet of ringed seals was assumed to consist of Arctic cod. Average POP concentrations reported for Arctic cod from several locations in the Canadian Arctic in the 1990s (Hargrave *et al.*, 1992; Muir *et al.*, 1999b) were used to calculate baseline average concentrations (ca. 1996) for use in simulations that extended over the period 1970 to 2010. Resulting baseline concentrations were 4.7 ng/g ww for  $\Sigma$ PCBs, 3.8 ng/g ww for  $\Sigma$ DDTs, 4.5 ng/g ww for  $\Sigma$ CHLs, 2.1 ng/g ww for  $\alpha$ -HCH, and 0.32 ng/g ww for  $\beta$ -HCH. Since temporal-trend data are lacking for Arctic cod in the Canadian Arctic, trends back to the early 1970s were estimated using temporal trends derived from the Lancaster Sound seabird egg monitoring program (Braune *et al.*, 2001a) using log<sub>e</sub>-linear regressions. Where discrepancies in temporal trends were noted between the three bird species examined, trends from thick-billed murres were used owing to their year-round Arctic residence. Significant declines were noted for  $\Sigma$ PCBs ( $-5.4\%/yr$ ) and  $\Sigma$ DDTs ( $-5.6\%/yr$ ) over the period of 1975 to 1998, while  $\beta$ -HCH levels increased over time ( $4.2\%/yr$ ). Although trends for  $\Sigma$ CHLs and  $\alpha$ -HCH were not statistically significant, resulting slopes ( $-1.1\%/yr$  and  $-0.4\%/yr$ , respectively) were used in model simulations. These trends were assumed to remain in effect in simulations to predict possible future concentrations up to the year 2010. Chemical elimination half-lives for seals used in these simulations were 4.1 years for  $\Sigma$ PCBs, 6.9 years for  $\Sigma$ DDTs, 3.3 years for  $\Sigma$ CHLs, 2.4 years for  $\alpha$ -HCH and 1.8 years for  $\beta$ -HCH. These were derived from model calibration exercises using independent datasets (Hickie, 2002). The half-life estimate of  $\Sigma$ PCBs was based on the weighted sum of half-life estimates for the 20 most abundant PCB congeners.

Overall, the simulations showed good agreement with observed temporal trends in the Holman ringed seal population for all five chemicals examined, and demonstrated that the temporal trends evident in ringed seals are consistent with those in seabirds, and likely reflect changes throughout lower trophic levels in the marine

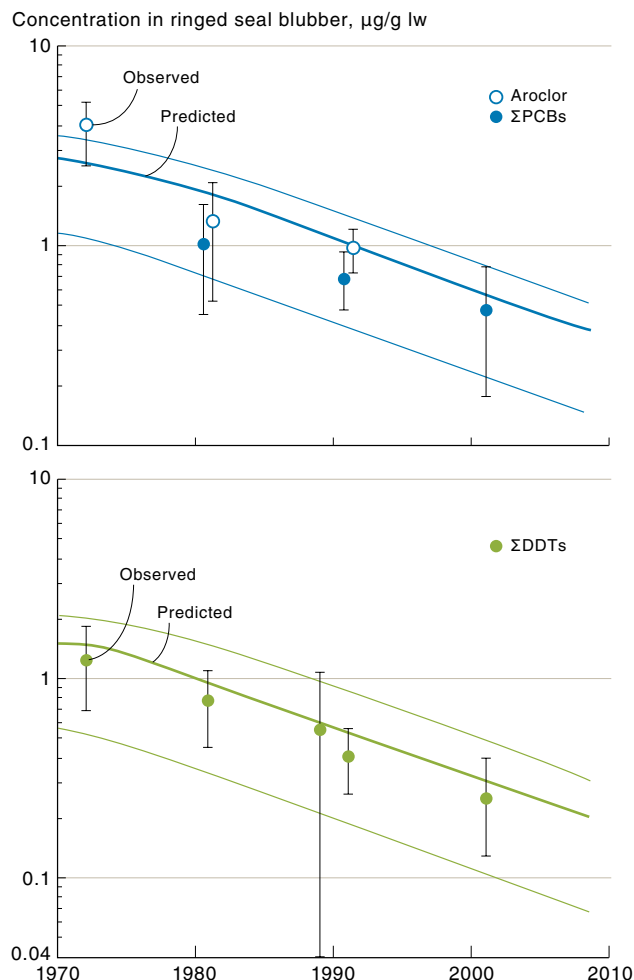


Figure 5-41. Modeled temporal trends of  $\Sigma$ PCBs and  $\Sigma$ DDTs in blubber of male ringed seals from 1972-2001 with projection into the near future.

ecosystem. The results also suggest that contaminant levels in ringed seal populations do not show any appreciable lag in response when contaminant exposure concentrations change gradually over time, as appears to be the case in the Arctic. The lack of a lag in response can be attributed to the relatively rapid elimination rates for the chemicals examined, combined with the effect of population turnover. The good agreement between the simulations and monitoring data serves to validate the model, and indicates that it can be used with some confidence in forecasting responses to potential future loading scenarios. Concentrations of  $\Sigma$ PCBs and  $\Sigma$ DDTs in blubber are predicted to decline by about 40% between 2000 and 2010, to ranges of 150-500 ng/g ww  $\Sigma$ PCBs and 60-300 ng/g ww  $\Sigma$ DDTs for the scenarios presented here.

#### 5.5. Summary and conclusions – temporal trends

A critical question in the assessment of POPs in the Arctic is whether concentrations are increasing or decreasing. The previous POPs assessment found a general lack of information on temporal trends of POPs in biota within the circumpolar Arctic, particularly for the High Arctic. While results for fish in northern Sweden demonstrated declines over the period of 1968 to 1996, there were only a few long-term datasets from other circumpolar countries. In the abiotic environment, temporal-

trend information was limited to sediment cores, which had relatively poor temporal resolution in most locations and measurements of HCH in seawater from the Bering Sea and the western Canadian Arctic. For the current assessment, considerably more data on temporal trends was available. Previous studies have been extended so that a 25 to 30-year perspective is now available for polar bears, seabirds, and ringed seals in the Canadian Arctic, as well as for fish in the Swedish Arctic. Studies covering a 10 to 15-year period are available for polar bear at Svalbard, peregrine falcons in Alaska, Atlantic cod in Iceland, glaucous gulls in Svalbard, burbot, lake trout and beluga whales in Canada and walrus in northwestern Greenland. Continuous air monitoring of POPs extends from 1992-93 to 2001 at Alert, Ny-Ålesund, and Pallas, although results were only available to 1998 for Alert and 1999 for Pallas. 'New' chemicals such as PBDEs have been added to the list of chemicals for which temporal trends are available in marine biota.

PCBs, the major OC contaminants in the Arctic, appear to be declining in most media. In air, half-lives of tri- to heptachlorobiphenyl congeners, based on digitally filtered results at Alert, ranged from 3 to 17 years. These are much longer than in temperate locations (Great Lakes and the U.K.), but they do indicate a slow, downward trend. Downward trends in air were also found for HCH and chlordane isomers; however, their half-lives were also generally longer at Alert than in the Great Lakes.

In the case of *p,p'*-DDE, *o,p'*-DDT, dieldrin, and endosulfan, slight increases in air at Alert were found during the 1990s. This is unexpected in the case of DDE and dieldrin because the use of their precursors (DDT and aldrin/dieldrin) has been restricted for more than ten years, and in some cases, up to 30 years in circumpolar and most northern-hemisphere countries. It does not parallel observations for marine or freshwater biota. The increases may be unique to the particular five-year dataset examined by Hung *et al.* (2002b) for Alert. At Ny-Ålesund, in the Norwegian Arctic, a pronounced decline in  $\Sigma$ DDTs in air was observed at least from 1997 to 2000. The great variation in concentrations of most OCs in air, particularly DDT at Ny-Ålesund, makes evaluation of temporal trends in air difficult. Long-range transport events appear to be very important for delivering DDT, PCBs and other OCs to these sampling sites and this may have been affected by changes in weather patterns during the 1990s (Macdonald *et al.*, 2003). Digital filtering and temperature normalization (Hung *et al.*, 2001; 2002b) have proved useful in interpretation of air data. Unfortunately, this detailed analysis has only been applied to data from Alert. The trends appear to be generally similar at Alert and Ny-Ålesund, but further study is required.

Temporal trends of POP concentrations in air and biota in the Arctic are not expected to parallel each other given the strong influence of the inventories of chemicals in the Arctic Ocean. This is illustrated in the case of  $\Sigma$ HCHs in ringed seals and polar bears. Air concentration data for  $\alpha$ -HCH, collected since 1979 by various groups at different Arctic locations, have been collated into a time series up to 1996 (Li and Bidleman, 2003; Li *et al.*, 2002). HCH levels decreased more than 20 fold in air from 1979 to 1996. However, there has been only a modest change in concentrations in ringed seals and polar bears in the eastern Canadian Arctic over

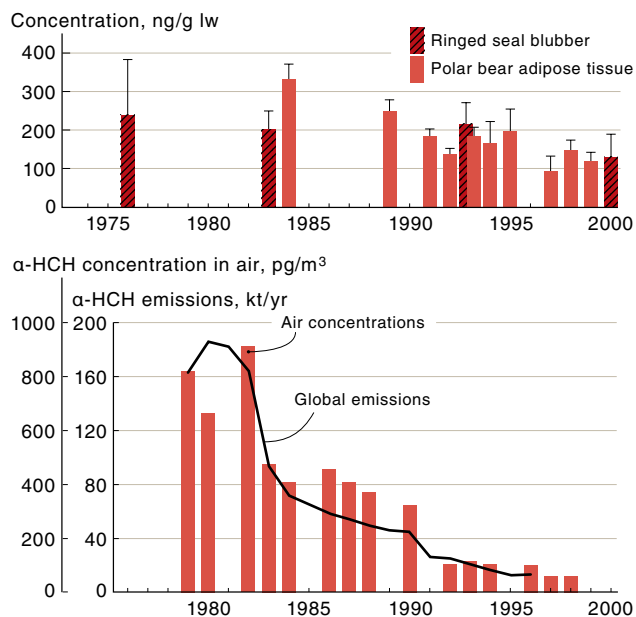


Figure 5-42. Comparison of temporal trends of  $\Sigma$ HCHs in ringed seals and polar bears and trends of  $\alpha$ -HCH in Arctic air (mainly from sites in the Bering/Chukchi/Beaufort Seas), as well as estimated emissions of  $\alpha$ -HCH (Li *et al.*, 2002) over the same time period.

the same time period (Figure 5-42). The observed trends in biota agree better with the much smaller estimated reduction (3%/yr) of  $\Sigma$ HCHs in surface seawater (Bidleman *et al.*, 1995; Jantunen and Bidleman, 1995). Unfortunately, no time-trend data are available for any other persistent OC compound concentrations for air (or seawater) over this long period. The comparison of air and biota indicates the long times needed to affect a change in concentration in long-lived marine biota following reductions in emissions of a chemical with multiple sources (e.g., a re-emitted chemical that is recycled globally) and long half-lives for some components (e.g.,  $\beta$ -HCH), which assume increasing importance in seals and polar bears over time.

While declines of major OC concentrations in biota are slow because of global cycling and long half-lives, the increasing concentrations of PBDEs in ringed seals and beluga (Figure 5-43) demonstrate that newly emit-

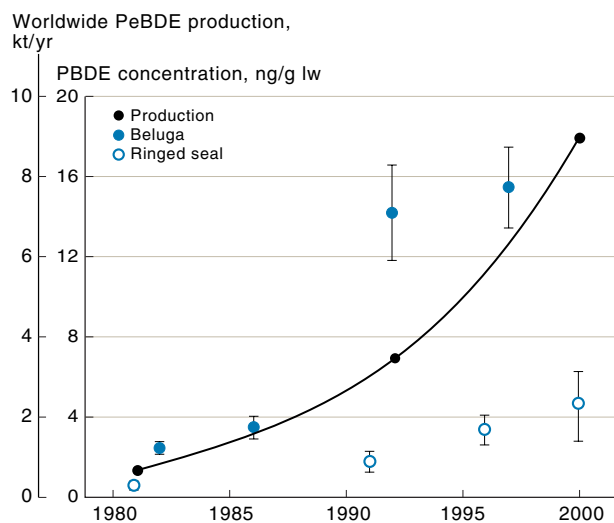


Figure 5-43. Comparison of temporal trends of  $\Sigma$ PBDE ( $Br_2$ - $Br_7$ ) in ringed seals and beluga in the Canadian Arctic to the estimated annual global production of PeBDE for the same time period.

ted persistent semi-volatile chemicals can increase in circumpolar countries in parallel with emissions. The doubling time for  $\Sigma$ PBDE concentrations in beluga during the early 1990s ( $t_2 = 3.0$  yr) was faster than in Holman ringed seals ( $t_2 = 4.5$  yr). By comparison, in the Great Lakes region, PBDE concentrations in herring gull eggs and lake trout are increasing at approximately the same rate as in beluga ( $t_2$  approximately 3 yr) (Norstrom *et al.*, 2002; Luross *et al.*, 2003). If the same rate of increase of PBDEs and declines in PCBs were to continue, PBDEs will reach parity with PCBs in Canadian Arctic ringed seals sometime between 2015 and 2025.

No temporal trends of PBDEs are available from the European Arctic to compare to the somewhat different temporal trends being seen in the Baltic Sea and northern Europe (exponential increases in the 1980s, with declines or no change occurring in some time series from the mid- or late 1990s) (Kierkegaard *et al.*, 1999; 2004; Sellström, 1999; Sellström *et al.*, 2003; Norén and Meironyté, 2000). The changing (and in some cases, declining) temporal trends of lower brominated congeners in Europe after voluntary withdrawal of the PeBDE product show the close relationship between discontinued use/production, reduced emissions, and decreased concentrations in the environment. The European Union (EU) will ban the PeBDE technical product in 2004, which should continue the reduction in emissions in Europe in the long term. However, it is not clear what effect this will have on PBDE concentrations in the Arctic, since no reductions in PeBDE use are presently in force in North America, and no temporal trends for PBDEs have been performed in the European Arctic.

A major unanswered question is the extent to which the temporal trends observed for various POPs to date apply throughout the circumpolar Arctic. This is partly addressed in Table 5-3 using the annual percent declines

of major OCs from the 1960s/early 1970s to late 1990s/early 2000s or from late 1980s/early 1990s to late 1990s/early 2000s, reported for selected species from remote locations. In general, declines are more rapid if calculated for the 25-30-year period than the more recent period, as seen in the pike and char from northern Sweden. The annual declines are best examined within species. A statistical comparison of temporal trends of HCB,  $\Sigma$ HCHs, and  $\Sigma$ PCBs among seabirds, ringed seal, and polar bear in the Canadian Arctic showed significant differences in rates of decline among species (Braune *et al.*, 2001c). This reflects different biological half-lives of the contaminants in each species as well as migratory behavior in the case of Arctic seabirds, and illustrates the need to have multi-species data when evaluating temporal trends.

One of the most rapid declines of POPs was observed by Zhulidov *et al.* (2002) who found that  $\Sigma$ DDTs and  $\Sigma$ HCHs declined 15 and 13% respectively in burbot over a six-year period in the Pechora River and at a similar rate in the Yenisey River. These rates are comparable to those for DDT and HCH isomers in char and pike in northern Sweden but much higher than in burbot in the Mackenzie River in Canada over the same period. This rapid decline may be related to reductions in the use of DDT and technical HCH during the 1980s in Russia, whereas this decline occurred earlier in Canada and Sweden due to usage bans in the early 1970s. Annual declines of PCB and  $\beta$ -HCH in polar bear from Svalbard and Hudson Bay during the 1990s differ. PCBs declined more rapidly, while  $\beta$ -HCH actually increased in Hudson Bay bears and declined in Svalbard. These temporal-trend results illustrate the need for information from multiple sites within the Arctic in assessing temporal trends of 'legacy' OCs, taking into account the history of POPs use within a given region.

Table 5-3. Annual percent declines for major OCs calculated from long-term temporal-trend data.

Species	Location	Time period	$\Sigma$ DDTs	$\Sigma$ PCBs	$\alpha$ -HCH <sup>a</sup>	$\gamma$ -HCH	HCB <sup>b</sup>
<i>Freshwater fish</i>							
Pike	Northern Sweden	1967-1999	-10	-4.1	-11	-3.4	-4.8
Pike	Northern Sweden	1990-1999	-8.3	-4.1	-10		5.4
Char	Northern Sweden	1980-1999	-9.9	-6.7	-17	-15	-5.0
Char	Northern Sweden	1990-1999	-14	-6.0			-5.5
Burbot	Mackenzie River	1986-1999	-1.8	-3.2	-5.2		-2.5
Burbot	Pechora River	1988-1994	-15		-13		
Burbot	Yenisey River	1988-1994	-16		-15		
<i>Marine mammals</i>							
Ringed seals	Eastern Canadian Arctic	1975-2000	-2.8	-1.8	-2.2	-0.8	-2.1
Ringed seals	Eastern Canadian Arctic	1983-2000	-2.6	0.5	-2.2	-0.2	-1.5
Ringed seals	Western Canadian Arctic	1972-2001	-2.4	-2.8	-3.3		
Ringed seals	Western Canadian Arctic	1991-2001	-3.7	-4.4			
Beluga	Eastern Canadian Arctic	1982-1997	0.3	0.5	-1.2		3.5
Polar bear	Eastern Canadian Arctic	1968-1999	-4.1	-0.3	-1.1		-1.4
Polar bear	Eastern Canadian Arctic	1989-1999	2.7	-2.7	-11.7		-18.3
Polar bear	Svalbard	1990-1998		-6.1	-7.2 <sup>a</sup>		
<i>Seabirds</i>							
Black-legged kittiwake	Central Canadian Arctic Archipelago	1975-1998	-3.6	-3.9	0.5		-3.0
Black-legged kittiwake	Central Canadian Arctic Archipelago	1975-1998	-7.3	-7.5	1.8		-2.2
Northern fulmar	Central Canadian Arctic Archipelago	1975-1998	-3.2	-3.1	0.9		-2.1
Northern fulmar	Central Canadian Arctic Archipelago	1975-1998	-0.4	-2.4	3.0		0.5
Thick-billed murre	Central Canadian Arctic Archipelago	1975-1998	-2.4	-2.5	2.8		-2.6
Thick-billed murre	Central Canadian Arctic Archipelago	1975-1998	-5.1	-5.6	-1.1		-5.5

<sup>a</sup>  $\Sigma$ HCH except for pike, char and ringed seals;  $\beta$ -HCH for polar bears at Svalbard.

<sup>b</sup> Total Te-HxCBz except for pike, char, and ringed seals.

## Chapter 6

# Biological Effects

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Biological effects can be measured at different levels of biological organization, from the molecular to the ecosystem level. Biomarkers measurable at a molecular level respond early but are not readily interpreted ecologically, while measures with established ecological relevance, such as population declines or reduced reproductive rates, respond too late to have diagnostic or preventive value.

In the previous AMAP assessment, there was relatively little knowledge of the biological effects of POPs in Arctic species, but concern was raised that the concentrations in some species put them at risk for effects on reproduction, the immune system, and on subtle neurobehavioral functions. Newer studies of biomarkers linked to POP exposures are beginning to show evidence that chemical contaminants may be present in sufficient quantities to have biologically significant effects on some species.

Two approaches have generally been taken in identifying and estimating the risk for possible effects. The first involves comparison and extrapolation. Researchers assess the risk for possible effects by comparing levels of POPs in Arctic species of interest to known detrimental levels, with this knowledge coming from laboratory studies, semi-field studies or from observations on affected animals in the wild. Extrapolation is routinely used in toxicology and is based on the conserved nature of many of the endpoints being measured across taxa. These include commonalities in endocrinology, immunology, and to a lesser extent, anatomy and reproductive biology. The extrapolation is based on similarities in mechanisms of action (e.g., mediation through the Ah-receptor, hormone receptors, CYP1A induction, OH-PCB disruption of vitamin A/thyroid, etc.). The difficulties in extrapolation relate generally to differences in sensitivity, where the same types of effects are seen but at different doses, or to differences in structure and function that are fairly obvious among species, especially those that are more distantly related to each other (i.e. fish vs. birds vs. mammals) (Kim and Hahn, 2002).

Comparison and extrapolation have some inherent weaknesses, however. Laboratory animals are most often exposed to single POPs or technical products at high doses for short periods of time, and it is difficult to extrapolate the toxic effects seen at high acute doses to possible adverse effects at lower but chronic exposures. Wild animals are generally exposed to lower concentrations of OCs than laboratory rodents in experimental studies, but they are exposed to mixtures of POPs and other stressors, and they are exposed over their entire lifetime.

They are also exposed to weathered mixtures due to the change in composition of many POP mixtures caused by abiotic degradation, metabolism and subsequent filtering up through the food web. For example, marine mammals at high trophic levels will be exposed

to very different PCB compositions, expressed as  $\Sigma$ PCBs, than is seen in a PCB technical product. As an illustration, a weathered mixture of PCB in contaminated fish was found to be more toxic to mink than a comparable  $\Sigma$ PCB concentration of a technical product (Aroclor 1254) (Giesy and Kannan, 1998).

Differences in species sensitivities to the effects of POPs make it difficult to know which of the tested species best represents those in the Arctic. For example, several Arctic species have delayed implantation (mink, otter, other mustelids, seal, walrus, and polar bear), which may make them more sensitive to the reproductive effects of POPs than tested laboratory animals without delayed implantation (Sandell, 1990). Arctic species also differ from laboratory animals because of their fat dynamics, differences in life styles and life strategies, and differences in toxicokinetics. For example, there are wide differences in metabolic capacity in different Arctic species compared to laboratory animals (see Section 3.2.2). Very little is known about the sensitivity of Arctic species, particularly marine mammals, to the effects POPs.

The second approach studies biological effects by examining subtle indicators of biological responses (biomarkers) to contaminants. Examination of the animals for responses known to be associated with the contaminants found in their tissues is perhaps the only way to make a convincing case either for or against the hypothesis that trace contaminants are acting biologically on the animals. Almost any biological change, from molecular to ecological, can serve as a biomarker; however, the term most often refers to changes at sub-cellular levels (McCarthy and Shugart, 1990; Huggett *et al.*, 1992; Peakall, 1992). Biomarkers, typically, are measures of normal processes that take on abnormal values as a result of exposure to chemicals of interest. Most of the biomarkers studied have established sensitivities (in laboratory animals) to some of the same contaminants measured in Arctic marine mammals and fish, notably several PCB congeners, PCDD/Fs, and PAHs.

The MFO cytochrome P450 system, a ubiquitous enzyme system common to mammals, birds, fish, and microorganisms, has probably been one of the most widely used biomarkers to date, with numerous laboratory and field cases of responses established (Payne *et al.*, 1987; Rattner *et al.*, 1989; Goksøyr and Förlin, 1992; Haasch *et al.*, 1993; Beyer *et al.*, 1996; Hylland *et al.*, 1996). The preferred field study design has been the comparison of an exposed group of individuals with similar groups not exposed to the same source. Another design, less commonly encountered, is the comparison of individuals within a group to search for linkages between biomarker values and exposure as indicated by chemical residues. This is a valuable approach for those contaminants stable enough to remain identifiable as residues.



There are also limitations with these types of studies, however. It is not possible to determine causality, only that a statistical association has been found between a biomarker and the contaminant in question. Most POPs covary, and thus, it is not possible to state unequivocally that the biomarker response has been caused by a particular contaminant. There may be other contaminants not analyzed that are just as important, or the response may be the result of synergistic, additive or antagonistic effects of contaminant mixtures. Biological variables such as age, sex, body condition, presence of disease or other stresses also may act as confounders, as they can cause similar biological effects as those seen from POPs. This means that for most reported biological effects in wildlife, the evidence for a causal link with a specific chemical contaminant is weak or non-existent. This is mainly due to the complexity of contaminant mixtures, the lack of chemical exposure data, lack of data on the sensitivity of the species concerned, and knowledge of mechanisms of action. Understanding the linkages between contaminants and health effects (e.g., on reproduction or immunosuppression) is most likely to come from studies in laboratory animals. Crucial in establishing causal evidence for chemical-induced wildlife effects are semi-field or laboratory studies using the wildlife species of concern. Semi-field studies represent a useful approach to bridge the gap between the controlled conditions of the laboratory experiments and the uncontrolled exposure conditions in the field. Based on these different types of studies, a weight-of-evidence argument can be established. For example, based on the combination of field, semi-field, and laboratory studies, it was concluded that dioxin-like PCBs that accumulated through the marine food chain aggravated the severity and extent of the 1988 morbillivirus-related epizootic in harbour seals in northwestern Europe (Ross *et al.*, 1996; Vos *et al.*, 2003).

In the following assessment, results from studies of biomarkers in Arctic biota are presented first if these have been performed. Where possible, levels of specific POPs in Arctic biota are also compared to no-observed-adverse-effect-levels (NOAEL) or no-observed-effect-levels (NOEL), and lowest-observed-adverse-effect-levels (LOAEL) or lowest-observed-effect-levels (LOEL) known to cause subtle effects in sensitive species. The purpose of these comparisons is to assess the likelihood that some Arctic species may be at risk for the effects of some POPs, and to identify these species, the types of effects they may be at risk for, and the contaminants that might be associated with these effects.

There are some problems in making such comparisons, particularly for mammals, and some caution is advised. Where possible, effects thresholds derived from wild species have been used, but the problems with comparing different species has already been discussed above. In the case of mammals, the  $\Sigma$ PCB thresholds are based on concentrations found in different types of tissues (blood, blubber, muscle) with very different lipid contents.

Some scientists consider it better to compare thresholds based on the same tissue on a wet weight basis in both the threshold species and the Arctic species,

whereas others prefer comparisons on a lipid weight basis. Wet weight comparisons are not always possible because different tissues with different lipid contents have been analyzed. Therefore, the values in this report have been normalized to lipid content to enable comparison of POP concentrations in different tissues. This conversion assumes that POP concentrations are evenly distributed in the lipid stores of all organs in an organism and are not fluctuating and, therefore, that the lipid-normalized concentrations in all organs will be the same. This is also known to not always be the case. In seals, however, lipid-normalized levels in blubber, liver and blood in the same individual were within a factor of 2 (Boon *et al.*, 1994) and in other marine mammals, they were usually within a factor of 2 and at most, a factor of 5 (Aguilar, 1985; Boon *et al.*, 1987; Boon *et al.*, 1992; Kannan *et al.*, 1993; Bernhoft and Skaare, 1994; Kannan *et al.*, 1994; Jenssen *et al.*, 1996; Nakata *et al.*, 1998b).

Lipid conversion also adds more uncertainty into the values since there are errors introduced from the lipid determination. In some cases, literature threshold values have been given on a lipid weight basis with no information available for converting back to wet weight. The comparisons for birds are somewhat less problematic, as thresholds have been determined in eggs and are expressed on a wet weight basis, and most data available for Arctic birds are for eggs on a wet weight basis as well.

Another weakness in this approach is that methods for quantifying  $\Sigma$ PCBs used in the thresholds studies and in the analyses of Arctic species are not identical, and use varying numbers of congeners or are based on quantification using a technical product (total PCB). Similarly, TEQs may have been calculated using PCDD/F, nPCB and/or mono-*ortho* PCB concentrations combined with toxic equivalency factors (TEF) from different schemes. Previously, TEF schemes were based on mammalian models, but fish- and bird-specific TEFs are now available (van den Berg *et al.*, 1998). Thus, the terms  $\Sigma$ PCBs and TEQ may not be completely comparable, and this may lead to under- or overestimations of exposure and risk. Mammalian TEFs are derived from studies in laboratory rodents and may not be appropriate for risk assessment in marine mammals. Similarly, bird TEFs are based on studies in domestic chickens, which are more sensitive to dioxin-like substances than fish-eating bird species (Sanderson *et al.*, 1998).

Many of the mammalian thresholds for  $\Sigma$ PCBs are based on studies in mink and otter. Mink, in particular, have been used as a surrogate for seals in many PCB studies, since mink also have delayed implantation. Mink and otter are extremely sensitive to the effects of PCBs and dioxin-like substances, and thresholds based on effects may overestimate the risk if Arctic species are less sensitive.

In a few cases, a threshold value is available only for laboratory species such as rats, mice, rabbits or dogs (e.g., toxaphene, PFOS, TBT and DBT). Because of the presence of these 'new' substances in the Arctic, these thresholds have been used to indicate whether any species is at potential risk for these substances, rather than not making any statement at all. For some

Table 6.1. Selected criteria, action levels or guidelines for critical pollutants in the Great Lakes. Parts of the table are modified from De Vault *et al.* (1995). All values for fish/aquatic organisms are based on wet weight, and for sediment on dry weight.

Contaminant	US FDA <sup>1</sup>	IJC <sup>2</sup>	GLI <sup>4</sup>	OMEE <sup>5</sup>	IJC <sup>3</sup>	TRG <sup>6</sup>	USEPA <sup>7</sup>	ERL <sup>8</sup>	ERM <sup>8</sup>	EQG <sup>6</sup>	EQG <sup>6</sup>
	Fish	Water	Water	Water	Fish tissue	Fish tissue	Fish tissue	Sedi-ment	Sedi-ment	Water	Sedi-ment
2,3,7,8 TCDD/ other PCDD/Fs as TEQs	25 pg/g		0.0096 pg/L			0.71 pg/g <sup>b,c</sup> 4.75 pg/g <sup>b,d</sup>	0.5 pg/g			0.02 pg/L	0.091 pg/g
DDT	5 µg/g	0.003 µg/L	0.00087 µg/L	0.003 µg/L	1.0 µg/g <sup>a</sup>	0.014 µg/g <sup>b</sup>	0.039 µg/g	1.6 ng/g	46 ng/g		
Total PCBs			17 pg/L	0.001 µg/L	0.1 µg/g <sup>a</sup>	0.015 µg/g <sup>b,c</sup> (0.79 pg/g TEQ) <sup>b,c</sup> 0.048 µg/g <sup>b,d</sup> (2.4 pg/g TEQ) <sup>b,d</sup>	0.16 µg/g	23 ng/g	180 ng/g		
Mirex	2 µg/g	< detection		0.001 µg/L							
Toxaphene	5 µg/g	0.008 µg/L		0.008 µg/L		0.0063 µg/g <sup>b</sup>					
Aldrin/dieldrin	0.3 µg/g	0.001 µg/L		0.001 µg/L	0.3 µg/g <sup>a</sup>						

<sup>a</sup> Whole fish.

<sup>b</sup> Aquatic organism.

<sup>c</sup> TEQ refers to dioxin toxic equivalents using toxic equivalency factors (TEFs) for mammals (van den Berg *et al.*, 1998).

<sup>d</sup> TEQ refers to dioxin toxic equivalents using toxic equivalency factors (TEFs) for birds (van den Berg *et al.*, 1998).

<sup>1</sup> USFDA (U.S. Food and Drug Administration) Action Levels in edible portions of fish for regulation of interstate commerce.

<sup>2,3</sup> International Joint Commission Annex 1 – objectives for protection of aquatic life and wildlife.

<sup>4</sup> USEPA (U.S. Environmental Protection Agency) Great Lakes Water Quality Guidance proposed criteria for protection of wildlife (USEPA, 1995).

<sup>5</sup> OMEE (Ontario Ministry of Environment and Energy) guideline for the protection of sediment quality (OMEE, 1993).

<sup>6</sup> Canadian Tissue Residue Guidelines for the protection of wildlife consumers of aquatic biota (Environment Canada, 2002). Total PCB values are calculated assuming that average PCB TEQs are equal to  $5 \times 10^{-5}$  of ΣPCB concentrations, which is the ratio in Arctic char (D. Muir, unpublished data).

<sup>7</sup> USEPA guideline values for assessment of hazards to fish-eating wildlife (USEPA, 1995).

<sup>8</sup> ERL = Effects range low and ERM = effects range median for sediments (Long *et al.*, 1995).

substances such as PBDEs, no threshold data are currently available, and the concentrations in Arctic biota can not be assessed for possible effects.

An attempt has also been made to compare POP levels in the diet of selected Arctic biota to known dietary no-observed-adverse-effect-concentrations (NOAEC) or no-observed-effect-concentrations (NOEC), and lowest-observed-adverse-effect-concentrations (LOAEC) or lowest-observed-effect-concentrations (LOEC), or to environmental quality criteria/guideline values for protecting aquatic biota/fish-eating wildlife that have been developed in various countries (Table 6·1). There are considerable limitations in this latter approach, as there is a general lack of knowledge on the diets of many Arctic organisms. It is also assumed that predators eat only one type of food, but even where food preferences are known, there may not be analytical data for these particular food items.

### Thresholds for effects in birds, mammals, and fish

#### Birds

In several review articles (Bosveld and van den Berg, 1994; Giesy *et al.*, 1994b; Barron *et al.*, 1995), the no-effect and low-effect levels for bird eggs and adults as well as dietary intakes associated with no or low effects have been compiled from the literature for ΣPCBs and dioxin-like compounds. For eggs of fish-eating and predatory birds (bald eagle, herring gull, Caspian tern, double-crested cormorant (*Phalacrocorax auritus*), common tern, Forster's tern (*Sterna forsteri*), great blue heron, black-crowned night heron (*Nycticorax nycticorax*)), the following ranges were found. For reproductive success, the NOEL range for ΣPCBs was 1.3-11 µg/g ww in eggs. The LOEL range for various endpoints of reproductive success (hatching success, egg mortality, deformities, and parental attentiveness) ranged from 3.5

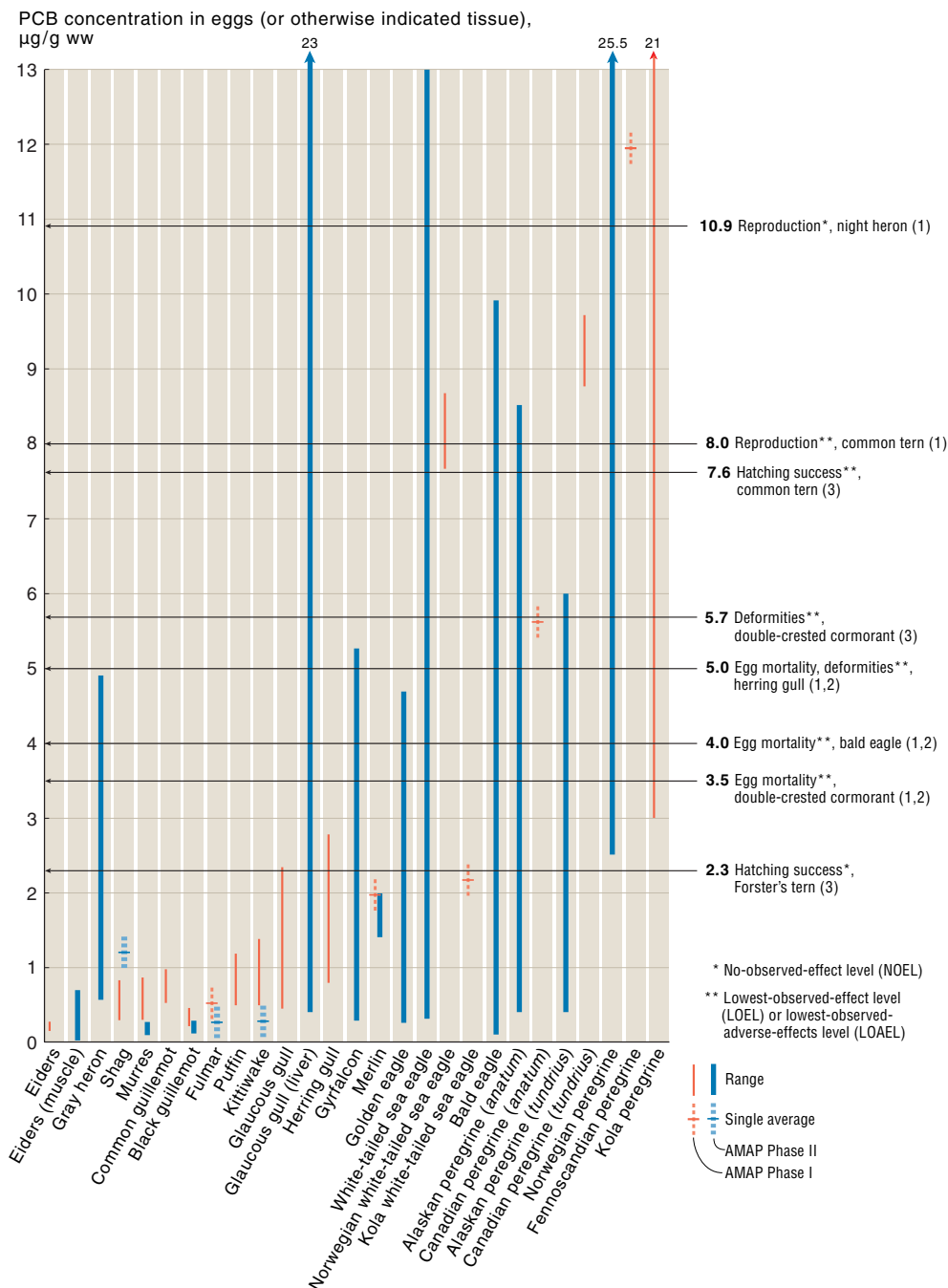


Figure 6-1. Concentrations of  $\Sigma\text{PCB}$  in Arctic bird eggs (or otherwise indicated tissue) compared to thresholds for avian effects (1. Barron *et al.*, 1995; 2. Giesy *et al.*, 1994b; 3. Bosveld and van den Berg, 1994). Due to numerous limitations in the thresholds data, in quantification methods for PCBs and problems with extrapolating such data across species, this comparison should be used with caution. See pages 163-165 for more details. AMAP Phase I data are from de March *et al.*, 1998.

to 22  $\mu\text{g}$   $\Sigma\text{PCBs/g ww}$  in eggs. For adults,  $\Sigma\text{PCB}$  concentrations in brain tissue higher than 300  $\mu\text{g/g ww}$  were associated with mortality. Relevant thresholds are presented in Figure 6-1.

PCBs with TCDD-like activity (nPCBs, mono-*ortho* PCBs) have been shown to adversely affect patterns of survival, reproduction, growth, and metabolism, with CB126 being among the most toxic of all PCB congeners to birds (Eisler and Belisle, 1996). For dioxin-like substances, the NOAEL range for reproductive effects was 1.5-200 pg TEQ/g ww in eggs, and the LOAEL range for various reproductive endpoints (deformities, hatching success, and mortality) ranged from 10 to 2200 pg TEQ/g ww in eggs. Elliott *et al.* (1996) have suggested a

NOEL of 100 pg/g and a LOEL of 210 pg/g TEQ on a whole egg (wet weight basis) for induction of cytochrome P450 (CYP) 1A in bald eagle chicks. Bosveld *et al.* (2000) have estimated the LOEL for induction of CYP1A in the common tern at approximately 25 000 pg TEQ/g lw in liver. These and other relevant thresholds are presented in Figure 6-2.

It has been concluded that peregrine falcon eggs with DDE residues of 15 to 20  $\mu\text{g/g ww}$  would experience reproductive failure (20% eggshell thinning) (Peakall *et al.*, 1990). The adverse effects threshold for dieldrin in peregrine falcon eggs is 1-4  $\mu\text{g/g ww}$ , and for heptachlor epoxide, the adverse effects threshold is 1.5  $\mu\text{g/g ww}$  (Peakall *et al.*, 1990). For HCB, results from Vos *et al.*

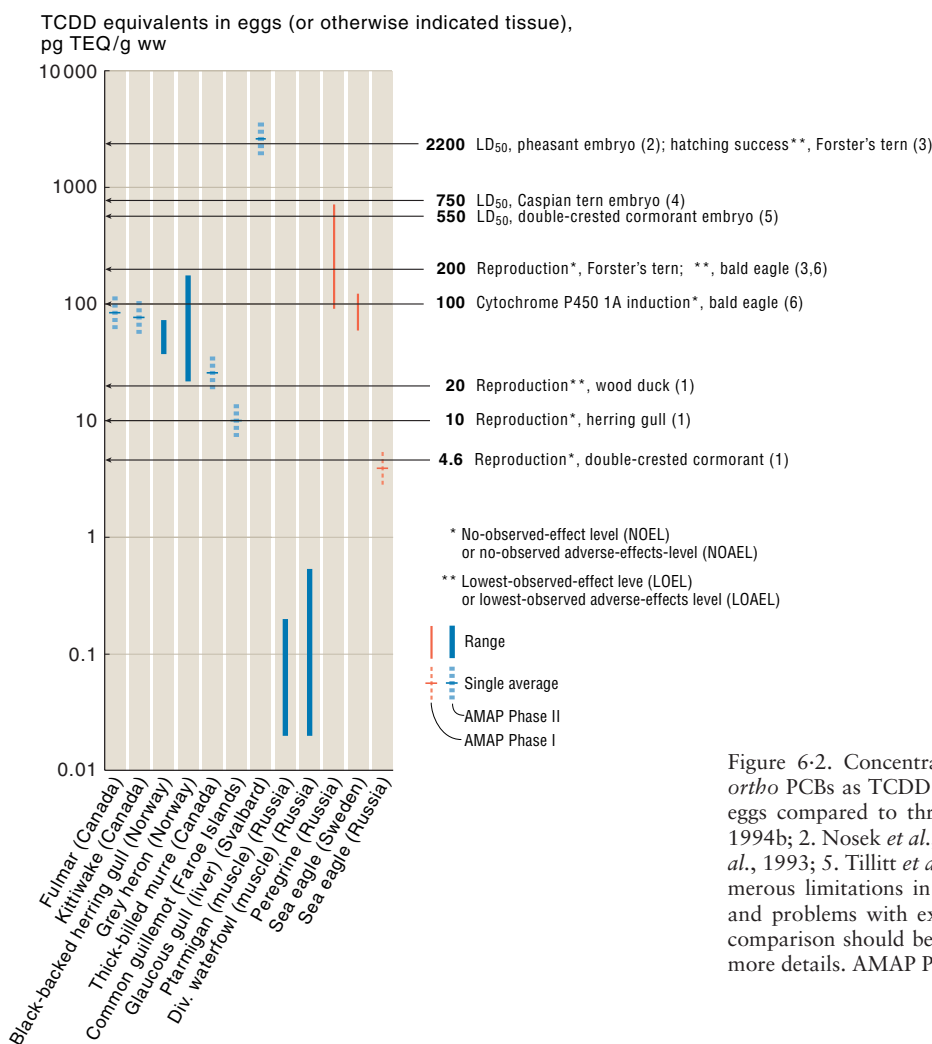


Figure 6-2. Concentrations of PCDD/Fs and non- and mono-ortho PCBs as TCDD equivalents (pg TEQ/g ww) in Arctic bird eggs compared to thresholds for avian effects (1. Giesy *et al.*, 1994b; 2. Nosek *et al.*, 1992; 3. Kubiak *et al.*, 1989; 4. Ludwig *et al.*, 1993; 5. Tillitt *et al.*, 1992; 6. Elliott *et al.*, 1996). Due to numerous limitations in the thresholds data, in calculating TEQs and problems with extrapolating such data across species, this comparison should be used with caution. See pages 163-165 for more details. AMAP Phase I data are from de March *et al.*, 1998.

(1972) indicate a NOEL in kestrels (*Falco tinnunculus*) of 40-49 µg/g ww in liver. Lindane concentrations of 100-200 µg/g ww in eggs of quail and chickens are associated with decreased egg production (Whitehead *et al.*, 1972a; 1972b; 1974). For bald eagle eggs, DDE concentrations below 3 µg/g ww are associated with near normal productivity, concentrations above 5.1 µg/g ww are associated with marked productivity declines, and concentrations above 15 µg/g ww are associated with complete reproductive failure (Wiemeyer *et al.*, 1984).

### Mammals

Platonow and Karstad (1973) reported that 1230 ng/g ww of Aroclor 1254 in mink liver tissue was associated with impaired reproductive success. Reduced growth and survival of mink kits were observed in female mink with 2000 ng/g ww Aroclor 1254 in liver tissue (Wren *et al.*, 1987a; 1987b). Olsson and Sandegren (1991a; 1991b) proposed an EC<sub>50</sub> of 50 000 ng total PCB/g lw, and Kihlström *et al.* (1992) proposed an EC<sub>50</sub> of 65 000 ng/g lw and a NOEL of 9000 ng/g lw for litter size in mink, based on muscle concentrations.

In a reassessment of all reproductive studies of PCBs on mink, the EC<sub>50</sub> in adult females for litter size was calculated to be 40 000-60 000 ng total PCB/g lw (approximately 1200 ng total PCB/g ww in muscle) and 2400 ng/g ww in muscle for kit survival (Leonards *et al.*, 1995). Brunström and Halldin (2000) found a LOAEL

of 12 000 ng PCB/g lw (muscle) in dams for decreased kit production and reduced kit body weight gain. The NOEL and LOEL for vitamin A reduction in otter were 4000 and 11 000 ng/g lw in liver for ΣPCB<sub>7</sub> (170 and 460 ng/g ww, respectively) (Murk *et al.*, 1998). Captive harbour seals from the Dutch Wadden Sea, exposed to PCBs via different fish diets, had reduced reproductive success at ΣPCB levels of 25000 ng/g lw in blood (16 ng/g ww) (Reijnders, 1986; Boon *et al.*, 1987).

Similarly, the EC<sub>50</sub> for dioxin-like compounds was calculated to be 160 pg TEQ/g ww (5300-8000 pg/g lw) in mink muscle for litter size and 200 pg TEQ/g ww (6600-10 000 pg/g lw) for kit survival (Leonards *et al.*, 1995). In experiments, Heaton (1992) found the LOAEL for mink kit survival to be 490 pg TEQ/g ww in liver. In free-living otter, the NOEL for vitamin A reduction was found to be 2000 pg TEQ/g lw in liver or blood (84 pg TEQ/g ww in liver) based on non- and mono-ortho PCBs (Murk *et al.*, 1998). The EC<sub>90</sub> level for vitamin A reduction was 5000 pg TEQ/g lw in liver and blood (210 pg TEQ/g ww in liver).

Experiments found reduced immune function and disruption of vitamin A physiology in captive harbour seals fed herring from the relatively contaminated Baltic Sea for 2.5 years. Total PCB concentrations of 16 500 ng/g lw, corresponding to total TEQ levels of approximately 210 pg/g lw, had accumulated in the blubber of these seals, suggesting a threshold for these effects below



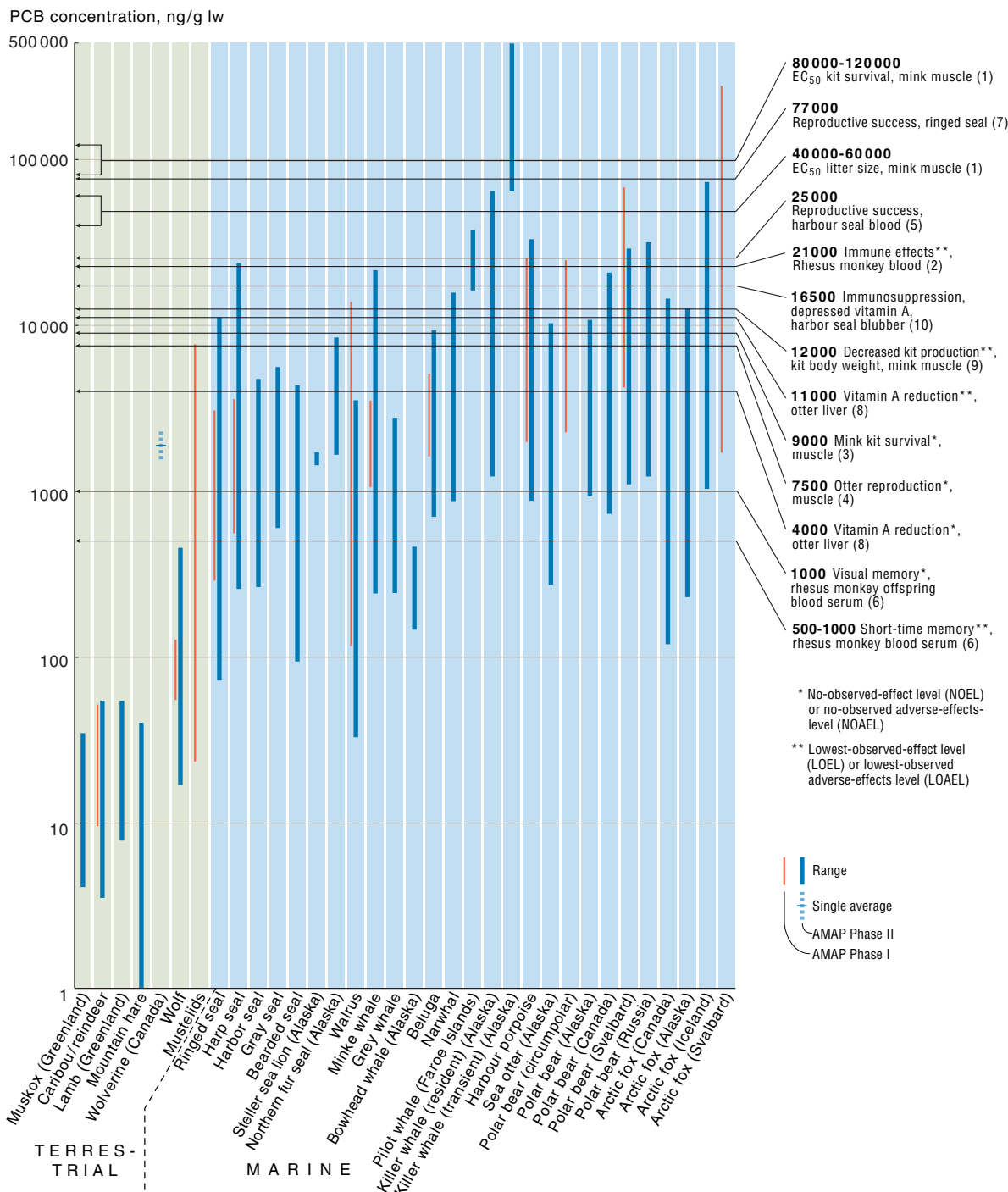


Figure 6-3. Concentrations of ΣPCBs in Arctic animals compared to thresholds for mammalian effects (1. Leonards *et al.*, 1995; 2. Tryphonas, 1994; 3. Kihlström *et al.*, 1992; 4. Roos *et al.*, 2001; 5. Boon *et al.*, 1987; 6. Ahlborg *et al.*, 1992; 7. Helle *et al.*, 1976; 8. Murk *et al.*, 1998; 9. Brunström and Halldin, 2000; 10. de Swart *et al.*, 1994, 1996). Due to numerous limitations in the thresholds data, in quantification of PCBs and problems with extrapolating such data across tissues and species, this comparison should be used with caution. See pages 163-165 for more details. AMAP Phase I data are from de March *et al.*, 1998.

these concentrations. Of the TEQs, PCDD/F accounted for 18 pg TEQ, nPCB for 51 pg TEQ, and mono- and di-ortho PCB for 140 pg TEQ (Ross *et al.*, 1995).

Kannan *et al.* (2000) have reviewed semi-field and field studies on seals, mink, and otter to establish mean threshold tissue concentrations of PCBs and TEQs linked to effects on hepatic vitamin A stores, thyroid hormone concentrations and the immune system. The purpose of this study was to derive threshold concentrations that could be used for estimating risk in marine mammals. For PCBs, the threshold concentrations in liver or blood for these effects were found to be 6600-

11 000 ng/g lw, and the authors suggest the geometric mean of 8700 ng/g lw as a threshold value. For TEQs, the threshold concentrations in liver and blood for these effects were 160-1400 pg/g lw (geometric mean of 520 pg/g lw).

Assessments based on subtle neurobehavioral effects in offspring of rhesus monkeys treated with PCBs and human mothers eating PCB-contaminated fish, have resulted in an estimated LOAEL for effects on short-term memory of 500-1000 ng/g lw, and a NOAEL for effects on visual memory of 1000 ng/g lw in offspring or cord blood serum (Ahlborg *et al.*, 1992). The LOAEL for im-

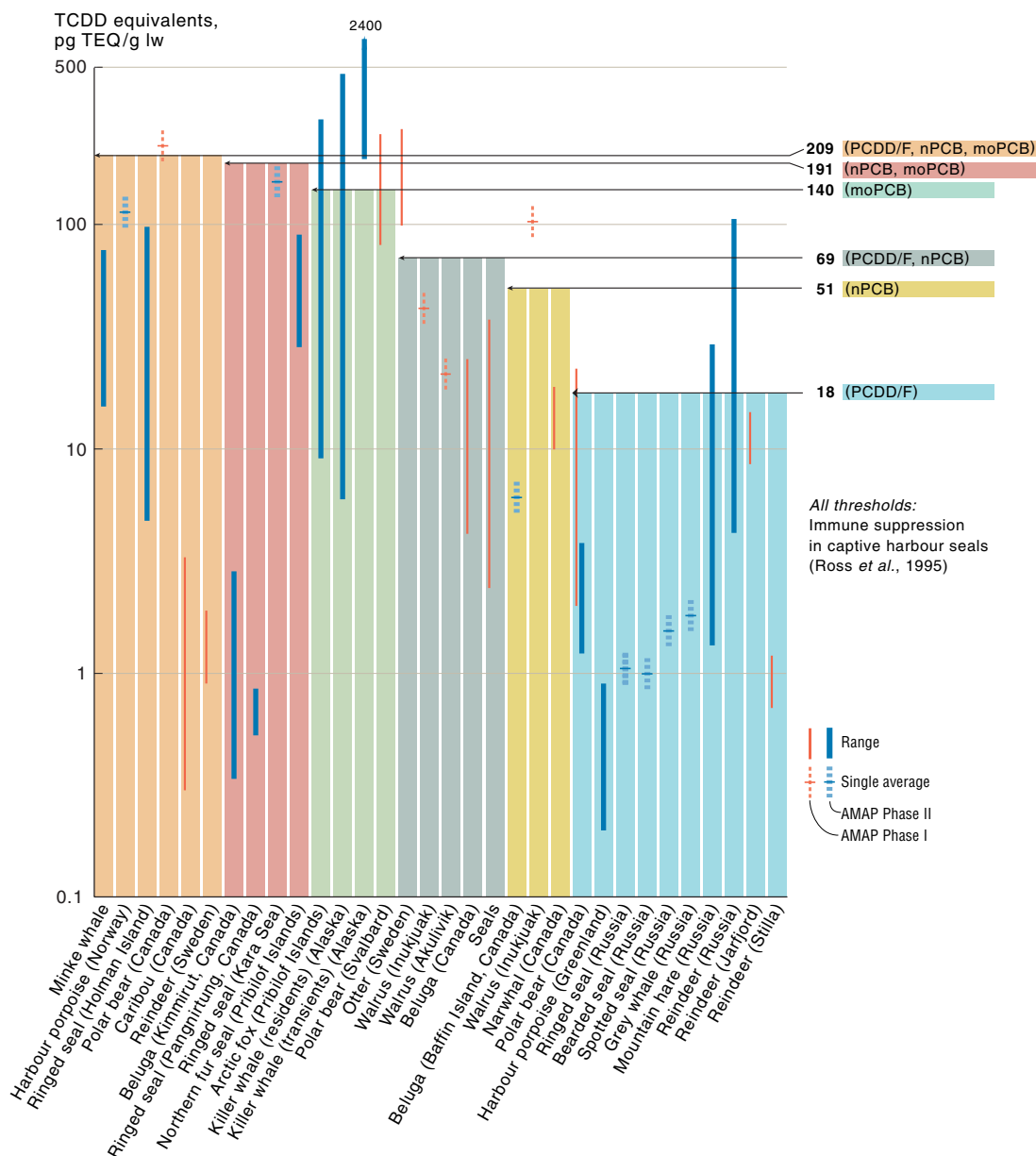


Figure 6-4. Body concentrations of PCDD/Fs and non- and mono-*ortho* PCBs as TCDD equivalents (pg/g lw) in Arctic mammals compared to thresholds for immunosuppression in harbour seal for the same combination of substances analyzed (Ross *et al.*, 1995). Due to numerous limitations in the thresholds data, in calculating TEQs and problems with extrapolating such data across tissues and species, this comparison should be used with caution. See pages 163-165 for more details. AMAP Phase I data are from de March *et al.*, 1998.

munosuppression is 21 000 ng  $\Sigma$ PCBs/g lw in rhesus monkeys (Tryphonas, 1994).  $\Sigma$ PCB concentrations of 2000-5000 ng/g ww in non-human primate brain tissue were associated with decreased dopamine concentrations (Seegal *et al.*, 1990). Relevant thresholds for  $\Sigma$ PCBs are presented in Figure 6-3 and for TEQs and effects on immunosuppression, in Figure 6-4.

Several effects thresholds have been determined for the effects of TBT or its metabolite, DBT. For *in vitro* inhibition of hepatic cytochrome P450 in Dall's porpoise and Steller sea lion liver, the threshold was found to be 100  $\mu$ M TBT (29000 ng/g ww) (Kim *et al.*, 1998b). For *in vivo* hepatotoxicity in mice, a threshold concentration of 2600 ng DBT/g ww in liver was found (Ueno *et al.*, 1994). For *in vitro* immunotoxicity to rat thymocytes (Snoei *et al.*, 1986) and rabbit polymorphonuclear neutrophils (PMNs) (Elferink *et al.*, 1986), the threshold concentration was found to be 1.0  $\mu$ M TBT (290 ng/g ww).

### Fish

The LOEL for EROD induction by  $\Sigma$ PCBs in Arctic char is 1000 ng/g ww in liver (Jørgensen *et al.*, 1999). Laboratory studies of Arctic char have found that single doses of PCB as low as 1000 ng/g body weight affect disease resistance and stress responses in starved specimens (Jørgensen, 2002). Mayer *et al.* (1977) found that adult fat-head minnows (*Pimephales promelas*) and channel catfish exposed to toxaphene had no decreases in hydroxyproline levels, but levels in exposed offspring were significantly decreased. Toxaphene concentrations of 3400 ng/g ww in channel catfish fry tissues were associated with decreased growth, and 600 ng/g led to altered bone development (Stickel and Hickey, 1977), identifying offspring as being more sensitive than adults are to toxic effects. Mayer *et al.* (1975) exposed brook trout to toxaphene in water and found higher mortality during spawning. A 50% mortality was associated with tox-



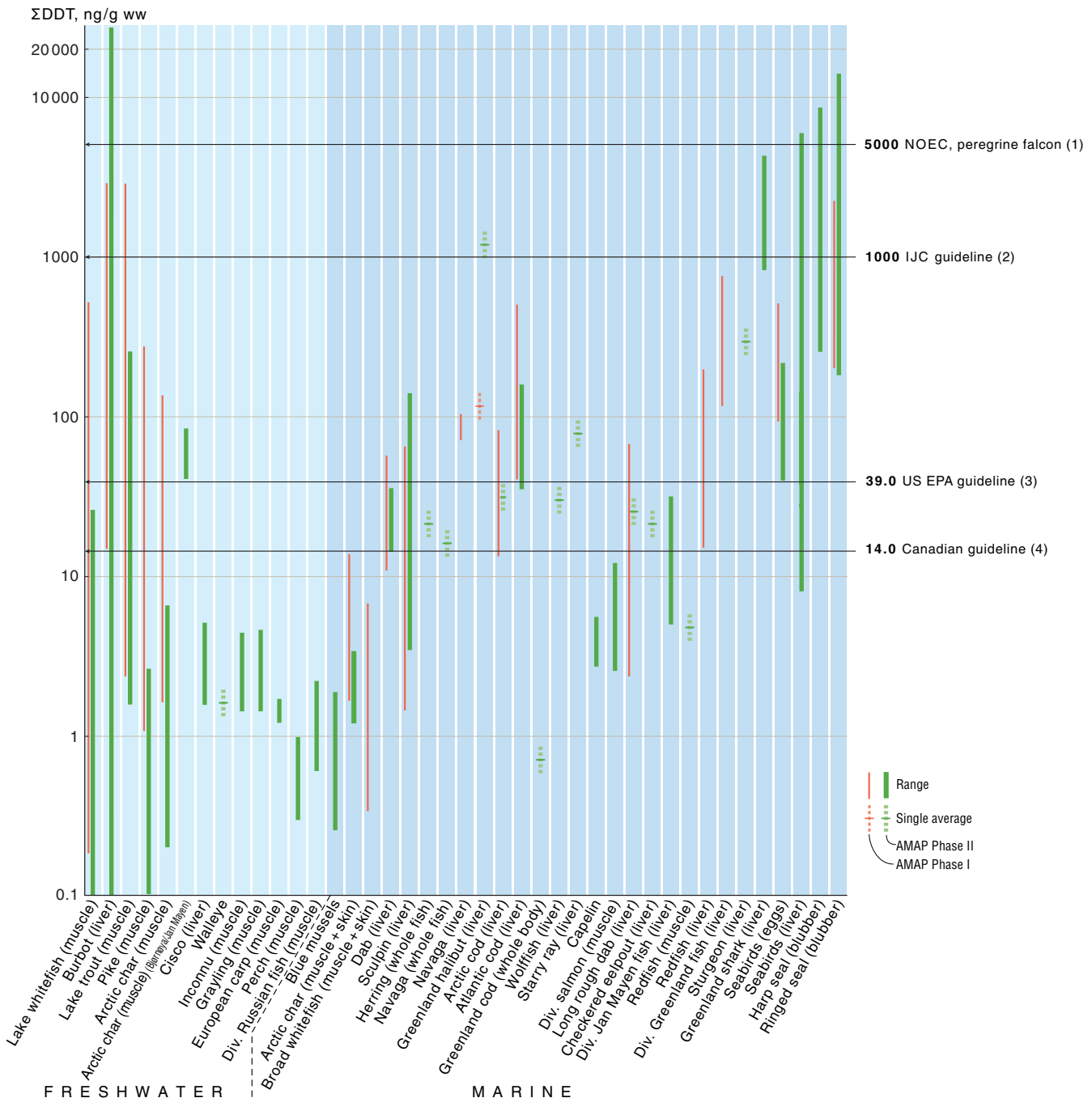


Figure 6-6. ΣDDTs in Arctic animals as food items compared to NOAECs and environmental quality guidelines for protecting fish-eating/aquatic wildlife (1. Baril *et al.*, 1990; 2. USEPA, 1995; 3. De Vault *et al.*, 1995; 4. Environment Canada, 2002). Due to problems with extrapolating data across tissues and species, this comparison should be used with caution. See pages 163-165 for more details. AMAP Phase I data are from de March *et al.*, 1998.

mono-ortho PCB TEQs was 11 pg TEQ/g lw in fish, and the LOEC was 29 pg TEQ/g lw in fish (Murdock *et al.*, 1998). For heptachlor epoxide, the dietary NOAEC for adult mink was 50 000 ng/g ww food (Aulerich *et al.*, 1990), and the LOAEC for kit growth was 6250 ng/g ww (Crum *et al.*, 1993). No information on the sensitivity of mink to toxaphene is available; however, a NOAEC of 4000 ng toxaphene/g ww food for thyroid effects has been estimated from studies in rats and dogs (Chu *et al.*, 1986b).

Relevant thresholds and guidelines for intakes of ΣPCBs are given in Figure 6-5, and for ΣDDTs in Figure 6-6. The environmental quality guidelines/objectives have been taken from Table 6-1. These have been developed by various organizations (International Joint Com-

mission, U.S. Environmental Protection Agency, Environment Canada) for the protection of aquatic life and wildlife that consume aquatic biota. The guidelines have been derived using contaminant concentrations in prey such as fish, known thresholds for effects in sensitive fish-eating wildlife species, and research results on bioaccumulation and biomagnification rates of particular substances. These data have then been used to back-calculate fish tissue concentrations that should be without effects in wildlife, and these have been designated as environmental quality guidelines/objectives.

In the discussion that follows, only new information (from 1996 or later) on biological effects studies, or where new concentration data warrant a discussion of the potential for biological effects, will be addressed.



## 6.1. Terrestrial environment

### 6.1.1. Terrestrial herbivores

#### 6.1.1.1. Arctic hare/mountain hare

No biological effects studies have been conducted on Arctic or mountain hares. Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs, and other POPs are available from West Greenland, several Russian sites and the Faroe Islands (Annex Table 5). The mean  $\Sigma$ PCB levels in different tissues (muscle, kidney, liver) range from 0.7 to 41 ng/g lw. These concentrations do not exceed any effects thresholds for  $\Sigma$ PCBs (Figure 6-3). TEQ values based on PCDD/F concentrations are also available for Russian mountain hares, and these varied from 1.4 to 30 pg TEQ/g lw (0.03 to 0.6 pg/g ww) in muscle (Annex Table 16). The highest TEQs were found in samples from hares from the Kola Peninsula (Lovozero) and are higher than levels associated with immunosuppressive effects in harbour seal (Figure 6-4).

#### 6.1.1.2. Caribou and reindeer

In the previous AMAP assessment (de March *et al.*, 1998), the concentrations of  $\alpha$ -HCH, HCB,  $\Sigma$ PCBs, chlor-danes,  $\Sigma$ DDTs, PCDD/F, and dieldrin in caribou and reindeer from across the Yukon, the Northwest Territories, Norway, Sweden, and Russia were very low. The Russian data were very limited, and concentrations from two consecutive years varied considerably. The POP levels were several orders of magnitude lower than those expected to lead to subtle biological effects. No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

A much better data set for  $\Sigma$ PCBs,  $\Sigma$ DDTs and other POPs is available from several sites across Russia (Annex Table 5). The mean  $\Sigma$ PCB levels in different tissues (muscle, kidney, liver) range from 0.2 to 2.4 ng/g ww. Based on an estimated lipid content of 4.5% in liver,  $\Sigma$ PCB concentrations are estimated to be 5.6-53 ng/g lw in the Russian reindeer.  $\Sigma$ PCB levels in reindeer from Finland were even lower (3.5 ng/g lw). These concentrations do not exceed any effects thresholds for  $\Sigma$ PCBs (Figure 6-3). TEQ values based on PCDD/F concentrations are also available for Russian reindeer, and these varied from 0.75 to 20 pg TEQ/g lw (0.05 to 0.98 pg/g ww) in muscle and 4.2 to 105 pg TEQ/g lw (0.24 to 6.5 pg/g ww) in liver (Annex Table 16). The highest TEQs were found in muscle and liver samples from reindeer from the Kola Peninsula (Lovozero) and are higher than levels associated with immunosuppressive effects in harbour seal (Figure 6-4). TEQs in liver from reindeer from Pechora and Taymir Dudinka were also above this threshold, but TEQs in muscle were not.

#### 6.1.1.3. Muskox

No biological effects studies have been conducted on muskox. Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs and other POPs are available from West Greenland (Annex Table 5). The mean  $\Sigma$ PCB levels in different tissues (muscle, kidney, liver, fat) range from 4.1 to 34 ng/g lw. These concentrations do not exceed any effects thresholds for  $\Sigma$ PCBs (Figure 6-3).

#### 6.1.1.4. Lamb

Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs and other POPs are available from West Greenland and Faroe Island lamb (Annex Table 5). The mean  $\Sigma$ PCB levels in different tissues (muscle, kidney, liver) range from 7.9 to 38 ng/g lw and are similar to the levels found in muskox. These concentrations do not exceed any effects thresholds for  $\Sigma$ PCBs (Figure 6-3).

### 6.1.2. Terrestrial birds

#### 6.1.2.1. Ptarmigan/willow grouse

No biological effects studies have been conducted on ptarmigan or willow grouse. Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs and other POPs are available from West Greenland and several Russian sites (Annex Table 5).  $\Sigma$ PCB concentrations ranged between 0.6 and 18.3 ng/g ww (24-270 ng/g lw). The levels are difficult to assess since the tissues analyzed are liver and muscle, but if the lipid levels in these tissues are assumed to be similar to that of eggs, then the liver and muscle concentrations do not exceed any thresholds for  $\Sigma$ PCBs (Figure 6-1). TEQ values based on PCDD/F concentrations for ptarmigan/willow grouse from several Russian sites varied from 0.02 to 0.2 pg TEQ/g ww (1.2-3.1 pg/g lw) in muscle (Annex Table 16). These are below concentrations expected to result in reproductive effects, assuming similar lipid levels in muscle compared to eggs (Figure 6-2).

### 6.1.3. Waterfowl

In the previous AMAP assessment (de March *et al.*, 1998), several waterfowl groups and species (molluscivores, piscivores, semipalmated plover, pintail, and oldsquaw) from Canada had levels of  $\Sigma$ PCBs above NOELs and LOELs for reproductive effects. No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

$\Sigma$ PCB and TEQ concentrations based on PCDD/Fs are available for several species of waterfowl from several Russian sites (Annex Tables 11 and 16). For  $\Sigma$ PCBs, results are available for liver and muscle from oldsquaw (*Clangula hyemalis*), goldeneye (*Bucephala clangula*), pintail (*Anas acuta*), wigeon (*Anas penelope*), green-winged teal (*Anas crecca*), greater white-footed goose (*Anas albifrons*), greater scaup (*Aythya marila*), scoters (*Melanitta* spp.), and bean goose (*Anser fabalis*).  $\Sigma$ PCB concentrations in liver range from 0.001 to 0.042  $\mu$ g/g ww, and for muscle from 0.001 to 0.017  $\mu$ g/g ww. The levels are difficult to assess since the tissues analyzed are liver and muscle, but if the lipid levels in these tissues are assumed to be similar to that of eggs, then the liver and muscle concentrations do not exceed any thresholds for  $\Sigma$ PCBs (Figure 6-1). For TEQs, results are available for oldsquaw, goldeneye, and pintail. TEQs range from 0.02 to 0.52 pg TEQ/g ww (0.5 to 6.4 pg/g lw) in muscle. These are below concentrations expected to result in reproductive effects, assuming similar lipid levels in muscle compared to eggs (Figure 6-2).

### 6.1.4. Birds of prey

#### 6.1.4.1. Peregrine falcon

In the previous AMAP assessment, it was concluded that Arctic populations of Canadian peregrine falcons were still at risk for reproductive effects from  $\Sigma$ DDTs and  $\Sigma$ PCBs in their eggs and in their food. For Fennoscandian peregrines, only high levels of  $\Sigma$ PCBs in eggs exceeded NOELs and LOELs for reproductive effects in other bird species. Peregrine falcons from the Kola Peninsula, Russia, had  $\Sigma$ PCB levels in eggs that exceeded most NOEL and LOEL levels for reproductive effects in other bird species. TEQs based on PCDD/F and dioxin-like PCB concentrations exceeded most NOAELs and LOAELs for reproductive effects in other bird species and several LD<sub>50</sub> values (the dose that causes the death of 50% of a group of test animals). No new effects or levels studies have been carried out in these populations since the previous assessment.

#### *Reproductive effects*

Peregrine falcon populations in North America and Europe declined drastically in most parts of the species' range in the 1960s and 1970s, primarily due to the effects of OC pesticides, especially DDT and dieldrin. Subspecies found in Alaska are the Arctic peregrine, which nests in northern tundra, the American peregrine, which nests in the forested interior and the Peale's peregrine, which nests along the southern coast from the Aleutian Islands to southeast Alaska.

Persistent OC contaminants were measured in American and Arctic peregrine falcon eggs from Alaska from 1979 to 1995 (Ambrose *et al.*, 2000). Dieldrin, *p,p'*-DDE, heptachlor epoxide, oxychlorodane, and total Arochlor PCBs were consistently measured and detected, and were tested multivariately for relationships with time and productivity. Eggshell thickness was significantly negatively correlated with *p,p'*-DDE concentrations, and mean eggshell thicknesses in 1991-1995 were 12.0 and 10.6% thinner in American and Arctic subspecies, respectively, than pre-DDT era peregrine falcon eggs from Alaska (Anderson and Hickey, 1972).

Significant multivariate analyses indicated that some POPs were associated with decreased reproduction. Differences were greatest early in the study, but over the entire time span, dieldrin, oxychlorodane and total PCBs were significantly greater in eggs from unsuccessful nests (no young at expected age of 1-3 wk) compared to successful nests ( $\geq 1$  young) for the American subspecies. There were no significant differences in *p,p'*-DDE or heptachlor epoxide, and no significant differences for any of the five contaminants in the Arctic subspecies. Eggs from unsuccessful nests also had higher mercury concentrations, which may also have affected reproduction (AMAP, 2004).

#### *Levels and intake assessment*

Concentrations of  $\Sigma$ PCBs in Alaskan peregrine eggs collected in 1991-1995 ranged between 0.4 and 8.5  $\mu\text{g/g}$  ww (geometric mean of 1.6  $\mu\text{g/g}$  ww) for the American subspecies and between 0.6 and 6.0  $\mu\text{g/g}$  ww (geometric mean of 1.3  $\mu\text{g/g}$  ww) for the Arctic subspecies (Ambrose *et al.*, 2000) (Annex Table 5). These mean  $\Sigma$ PCB levels are below thresholds for effects, but the maximum

$\Sigma$ PCB levels exceed most NOELs and LOELs found for hatching success, egg mortality and deformities in a number of wild bird species (Figure 6-1).

The *p,p'*-DDE concentrations in the American subspecies ranged between 0.48 and 14.1  $\mu\text{g/g}$  ww (geometric mean of 3.4  $\mu\text{g/g}$  ww), and in the Arctic subspecies between 1.2 and 13.3  $\mu\text{g/g}$  ww (geometric mean of 3.0  $\mu\text{g/g}$  ww), indicating that some individuals have concentrations just below the critical threshold for reproductive failure (15-20  $\mu\text{g/g}$  ww).

Geometric mean dieldrin concentrations were below the adverse effects threshold of 1-4  $\mu\text{g/g}$  ww for both subspecies and in 1991-1995, no individual eggs exceeded these levels. Geometric mean heptachlor epoxide concentrations never exceeded the adverse effects threshold of 1.5  $\mu\text{g/g}$  ww.

$\Sigma$ PCB concentrations in Norwegian peregrine falcon eggs collected in 1991-1997 ranged from 2.3 to 25.5  $\mu\text{g/g}$  ww (mean of 10.6  $\mu\text{g/g}$  ww) (Annex Table 5). These concentrations exceed most thresholds for reproductive effects in other bird species (Figure 6-1).

#### 6.1.4.2. Merlin

In the previous AMAP assessment, Fennoscandian merlin were suffering from 10% shell thinning, but the population was recovering. Merlin seemed to be less sensitive to the effects of DDT, since  $\Sigma$ DDT levels exceeded those expected to cause reproductive failure in peregrine falcons (20% eggshell thinning results in crushed shells).  $\Sigma$ PCB levels were in the range of some NOELs and LOELs for effects in white leghorn chicken, an extremely sensitive species. No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

$\Sigma$ PCB concentrations in Norwegian merlin eggs collected from 1991 to 1997 ranged between 1.4 and 2.0  $\mu\text{g/g}$  ww (Annex Table 5). These concentrations are below thresholds for reproductive effects in other bird species (Figure 6-1).

#### 6.1.4.3. White-tailed sea eagle

In the previous AMAP assessment, effects studies showed improvement in reproduction and population numbers for white-tailed sea eagles in Sweden and Norway, with only a small amount of eggshell thinning (2-5%). However, levels of TEQs in Swedish eagles, measured as PCDD/F and several dioxin-like PCBs, exceeded most NOAELs and LOAELs for reproductive effects in other bird species and approached the LD<sub>50</sub> for white leghorn chicken embryo.  $\Sigma$ PCB levels in the northerly Norwegian eagles exceeded or overlapped most NOELs and LOELs for subtle reproductive effects in other bird species.  $\Sigma$ PCB and TEQ levels in one Russian white-tailed sea eagle egg from the Kola Peninsula exceeded some thresholds for effects in other bird species. The intake assessment for Norwegian sea eagles, which prey mainly on deep-water marine fish species, concluded that dietary intakes of  $\Sigma$ DDTs and  $\Sigma$ PCBs may be high enough to lead to effects. No effects studies have been carried out since the previous assessment.

*Levels and intake assessment*

ΣPCB concentrations in Norwegian white-tailed sea eagle eggs collected in 1991-1997 ranged from 3.1 to 13.0 µg/g ww (mean of 9.0 µg/g ww) (Annex Table 5). These concentrations exceed most thresholds for reproductive effects in other bird species (Figure 6·1).

**6.1.4.4. Bald eagle***Reproductive effects*

The relationship between OC concentrations, diet, and productivity was studied in nesting bald eagles from Adak, Tanaga, Amchitka, and Kiska Islands in the Aleutian archipelago (Anthony *et al.*, 1999). Productivity on Kiska Island (the most westerly island) was depressed (lower breeding success and low clutch sizes), averaging 0.67 young per occupied site. This was associated with higher DDE and OC pesticide concentrations. Mercury concentrations were also higher. Productivity on the other three islands was comparable to healthy populations in other parts of the U.S. (0.88-1.24 young per occupied site), and DDE concentrations were lower. Eggshell thickness was not significantly different among the islands and was not correlated to DDE concentrations, but was significantly negatively correlated to PCB concentrations. PCB concentrations were not correlated to DDE concentrations, but instead, reflected previous military activity.

*Levels and intake assessment*

Mean ΣPCB concentrations in bald eagle eggs from the above four islands in the Aleutian chain were between 0.7 and 2.1 µg/g ww (range 0.1-9.9 µg/g ww) (Annex Table 11). The mean concentrations are just below the NOEL for hatching success in Forster's tern (Figure 6·1). However, the maximum ΣPCB concentrations at some sites exceed most of the thresholds for reproductive effects in other bird species including the LOAEL for egg mortality in bald eagles. The mean *p,p'*-DDE concentrations were between 0.7 and 2.75 µg/g ww (range 0.3 to 4.1 µg/g ww) with the highest concentrations on Kiska Island (range 1.5-4.1 µg/g ww). These concentrations are within the range known to cause reproductive impairment in bald eagles (Wiemeyer *et al.*, 1984).

**6.1.4.5. Golden eagle***Levels and intake assessment*

ΣPCB concentrations in Norwegian golden eagle eggs collected in 1991-1997 ranged from 0.26 to 4.7 µg/g ww (mean of 1.4 µg/g ww) (Annex Table 5). These concentrations exceed a few thresholds for reproductive effects in other bird species (Figure 6·1).

**6.1.4.6. Gyrfalcon**

In the previous AMAP assessment, DDE and ΣPCB concentrations in Canadian gyrfalcon eggs were several orders of magnitude lower than those considered to cause reproductive effects. Based on dietary LOECs, OC concentrations in gyrfalcon prey from Canada were also below those expected to cause effects. No effects studies have been carried out since the previous assessment.

*Levels and intake assessment*

ΣPCB concentrations in Norwegian gyrfalcon eggs collected in 1991-1997 ranged from 0.29 to 5.3 µg/g ww (mean of 2.0 µg/g ww) (Annex Table 5). These concentrations exceed a few thresholds for reproductive effects in other bird species (Figure 6·1).

**6.1.5. Carnivores****6.1.5.1. Mink**

In the previous AMAP assessment, Canadian mink from Quebec (Grand Baleine) and Fort Providence, NWT, had ΣPCB levels that exceeded the NOAEL and LOAEL for effects on subtle neurobehavioral effects in offspring of rhesus monkeys, and the Quebec mink levels were close to the NOEL for mink kit survival. The intake assessment for Canadian mink indicated that dietary intakes of dioxin-like compounds and ΣPCBs from some species of fish at some lakes may be high enough to lead to effects. No effects studies have been carried out since the previous assessment. No new information on current POP levels is available.

**6.1.5.2. Otter**

In the previous assessment, otter populations in northern Sweden were beginning to recover in the 1990s after having existed only as isolated groups (de March *et al.*, 1998). The ΣPCB levels were considered to be at the NOEL for otter reproduction. These levels exceeded the NOAEL and LOAEL for effects on subtle neurobehavioral effects in offspring of rhesus monkeys, however. The intake assessment for Swedish otter indicated that dietary intakes of dioxin-like compounds from some species of fish at some lakes may be high enough to lead to effects. No effects studies have been carried out since the previous assessment. No new information on current POP levels is available.

**6.1.5.3. Wolverine**

No biological effects studies have been conducted on wolverines. Concentrations of ΣPCBs, ΣDDTs, and other POPs are available from the Northwest Territories, Canada (Annex Table 5). The mean ΣPCB levels in liver are 1960 ng/g lw. These concentrations exceed the NOAEL and LOAEL for subtle neurobehavioral effects if wolverines are as sensitive as offspring of rhesus monkeys and humans (Figure 6·3).

**6.1.5.4. Wolf**

In the previous AMAP assessment, concentrations of ΣPCB levels in wolves from the Canadian Arctic were several orders of magnitude lower than those expected to result in effects on reproduction. The levels in caribou/reindeer were also much lower than the dietary concentrations expected to cause such effects. No biological effects studies have been conducted on wolf since the previous assessment.

*Levels and intake assessment*

Mean concentrations of ΣPCBs in wolves (liver) from the Canadian Yukon, and median concentrations from northwest Russia were between 17 ng/g lw and 450 ng/g



lw (Gamberg and Braune, 1999; Shore *et al.*, 2001). These concentrations are below those expected to result in effects (Figure 6-3). The dietary assessment is the same as for the previous AMAP assessment.

## 6.2. Freshwater environment

### 6.2.1. Fish

In the previous AMAP assessment, freshwater fish did not exceed effect levels for toxaphene, but fish from some lakes had levels close to the threshold for bone development effects and mortality during spawning, indicating that some individuals in Canada may exceed these levels. Besides Lake Laberge, possibly affected locations included several lakes in the Yukon (Bennett, Tagish, and Marsh Lakes), Atlin Lake in northern British Columbia, and Great Slave Lake in the Northwest Territories.

#### *Cytochrome P450 activities*

Arctic char from two lakes on Bjørnøya, Norway, have been studied for OC contamination and biomagnification: Ellasjøen, which is strongly affected by seabird guano from nearby colonies and has high  $\Sigma$ PCB levels and Øyangen, an oligotrophic lake unaffected by the bird colonies and with lower  $\Sigma$ PCB levels. Arctic char have also been studied for liver enzyme activity including testosterone hydroxylation enzymes. When comparing char from both lakes, correlations were seen between CYP enzyme activities and high  $\Sigma$ PCB levels. Levels of testosterone hydroxylation enzymes were similar at both lakes and not correlated to  $\Sigma$ PCB levels (Skotvold *et al.*, 1999).

#### *Levels and intake assessment*

Toxaphene levels in some burbot from the East Arm of Great Slave Lake, NWT and Lake Laberge and Kusawa Lake, Yukon (Canada), exceed levels associated with effects on bone development in channel catfish (600 ng/g ww) (Annex Table 7). Toxaphene levels in freshwater fish from all other sites measured in the Arctic are below threshold levels for effects.  $\Sigma$ PCB concentrations in burbot liver from Fairbanks and Yukon Flats (Alaska), Lake Laberge (Canada) and in some Arctic char from Bjørnøya (Ellasjøen) are close to or exceed the LOEL for induction of EROD in Arctic char (Annex Table 7).  $\Sigma$ PCB levels in other Arctic freshwater fish do not exceed this threshold.

## 6.3. Marine environment

### 6.3.1. Invertebrates

In the previous AMAP assessment, imposex, the induction of male sex characteristics in females, was found in females of the common whelk in Kongsfjorden, Svalbard, dogwhelks in Norway and Iceland, and a marine snail (*Nucella lima*) in Alaska exposed in harbors with significant boat mooring.

#### *Reproduction*

There have been only modest studies on TBT and its effects on gastropods recently in Arctic or subarctic waters. In Norway, TBT has been measured, and imposex has been evaluated in dogwhelks in 41 populations sam-

pled in 1993-1995 along the Norwegian coast (Følsvik *et al.*, 1999). Some degree of imposex occurred in almost all populations of dogwhelks studied, except in four from northern Norway. The concentration of organotin compounds in the gastropods from the unaffected populations was below the detection limit (7 ng Sn/g dw). The concentration of TBT in dogwhelks from affected populations was in the range of 48-1096 ng Sn/g dw. A positive relationship between the concentration of TBT in dogwhelks and the degree of imposex was found.

In Iceland, the status of imposex was evaluated in the dogwhelk in 1998 and compared to the levels of imposex evaluated in 1992/1993 (Svavarsson, 2000). The level of imposex has decreased considerably since 1992/1993, two years after implementation of restrictions on the use of TBT-based anti-fouling paint. VDSI and RPSI levels (see definition in Table 6-2) have declined considerably, both near large and small harbors. The impact area of a large harbor complex has decreased considerably, while lesser changes were seen in the impact areas near smaller harbors. This study shows that, at least in Icelandic waters, the situation has improved considerably.

Imposex has been observed in the subtidal common whelk near Svalbard and in Icelandic waters (Brick and Bolte, 1994; Svavarsson *et al.*, 2001). The levels observed in Breiðafjörður, southwestern Iceland, were low, and only 26.4% of the females observed had imposex, and the penises were small (0.7 mm  $\pm$  0.6 mm SD) (Svavarsson *et al.*, 2001). Near harbors (Reykjavík and Straumsvík harbors), the frequency was higher, and the female penises were considerably longer (mean 3.8 and 6.8 mm, respectively).

Studies of the occurrence of imposex in dogwhelks in the Faroe Islands were done in 1996 and in 2001 (FEA, 2002). The results do not indicate any major changes in the five-year period from the first to the second sampling period (Table 6-2). The occurrence of imposex in the Faroe Islands is still widespread, but there are sites where the phenomenon is hardly seen.

Table 6-2. Imposex in dogwhelks, *Nucella lapillus*, in the Faroe Islands.

Station	VDSI <sup>a</sup>	VDSI	RPSI <sup>b</sup>	RPSI
	Feb.-Mar. 1996	March 2001	Feb.-Mar. 1996	March 2001
<i>Tórshavn</i>				
Argir	4.4	4.0	32	48
Kirkjubour	0.1	0.6	0	0
Vestmanna	4.3	4.1	13	17
<i>Trongisvágsfjörður</i>				
Kolatoftrir	4.4		31	
Hvítanæs (April '01)		4.2		36
<i>Klaksvík</i>				
Kunoy	4.1	4.0	13	10
Skálafjörður	4.0		53	
<i>Nólsoy</i>				
Víkin <sup>c</sup>		4.1		20
Kirkjutangi	4.3	1.3 <sup>d</sup>	8	0 <sup>d</sup>

<sup>a</sup> VDSI = Vas deferens sequence index. The VDSI scale ranges from 0 (normal) to 6 (last stages of development of vas deferens and penis in females leading to sterility).

<sup>b</sup> RPSI = Relative penis size index (female mean penis length<sup>3</sup>/male mean penis length<sup>3</sup>)·100.

<sup>c</sup> Too few females in 1996 sample for analyses.

<sup>d</sup> Imposex in all stages were found, a sample with unusually large individual variation.

Source: FEA (2002).



In Greenland, TBT concentrations in blue mussels and the occurrence of imposex in several whelk species was studied in and around five harbors (Nuuk, Qaqortoq, Manitoq, Qeqertarsuak, and Uummanaq), and at an uninhabited reference site (Strand and Asmund, 2003). Some degree of imposex was found in several neogastropod species (*Buccinum* spp.) in all the harbors but not outside the harbors or at the reference site. Effects of contaminants on blue mussels have been evaluated at sites in southwestern Icelandic waters by studying Scope for Growth (SFG) (Widdows, 1998). This method allows evaluation of combined effects of contaminants on the physiology of the mussel. The studies show that SFG is low near Reykjavik harbor amongst mussels with high levels of TBT and PAHs, while among mussels living near small harbors (low TBT levels; intermediate PAH levels), the SFG is fairly high (Halldórsson, 2002).

### 6.3.2. Fish

In the previous AMAP assessment, only Greenland halibut (turbot) from the Canadian Arctic had toxaphene levels close to the effect levels for bone development and mortality during spawning. No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

Mean  $\Sigma$ PCB levels in marine and anadromous fish are below the LOEL for EROD induction in Arctic char except for Greenland shark (liver) from Cumberland Sound and for Greenland halibut (liver) from West Greenland (Annex Table 10). Based on mean toxaphene levels, no anadromous or marine fish exceed effects levels.

### 6.3.3. Seabirds

In the previous AMAP assessment,  $\Sigma$ PCB levels in seabirds from the Canadian and Norwegian Arctic approached or exceeded reproductive NOELs and LOELs at the low range of the scale compared to peregrine falcons. Mean  $\Sigma$ PCB levels in eider eggs were below those expected to result in reproductive effects, levels in shag (a species of cormorant) and fulmar were at or exceeded the LOAEL for embryo deformities and the lower NOEL for hatching success in white leghorn chicken, an extremely sensitive species. In addition, mean  $\Sigma$ PCB levels in puffin, murre, common guillemot, black guillemot, and kittiwake exceeded the upper NOEL for hatching success in white leghorn chicken.

Mean  $\Sigma$ PCB levels in glaucous gull and herring gull eggs were somewhat higher and approached or exceeded the LOEL for hatching success in white leghorn chicken and the NOEL for hatching success found for Forster's tern. None of the Arctic seabirds studied appear to have  $\Sigma$ PCB levels high enough to be associated with thresholds for egg mortality. Mean  $\Sigma$ DDT, HCH, dieldrin, and HCB levels in all seabirds studied were several orders of magnitude below those expected to cause effects.

Based on dietary LOECs for  $\Sigma$ PCBs, HCB, DDE, and dieldrin in peregrine falcon prey, levels in anadromous and marine fish were several orders of magnitude below those expected to cause effects in falcons. Mean dieldrin levels in Arctic anadromous and marine fish from Cana-

dian, Norwegian, Greenlandic, Icelandic, and Russian waters did not exceed any of the guidelines for protecting fish-eating wildlife. However,  $\Sigma$ PCB and  $\Sigma$ DDT levels in a range of fish species from many Arctic sites exceeded some, and in a few cases for  $\Sigma$ PCBs, all of the environmental quality guidelines, implying that dietary intakes of  $\Sigma$ DDTs and  $\Sigma$ PCBs in fish-eating seabirds might be high enough to lead to effects if seabirds prey on these fish species. A few fish species in Canada and Norway had PCDD/F or nPCB levels that exceeded the guidelines.

Based on POP levels found in eggs from guillemots, puffins, cormorants, and fulmars from the Canadian and Norwegian Arctic, dieldrin, HCB,  $\Sigma$ DDT, and  $\Sigma$ PCB levels were below dietary LOECs for reproductive effects in peregrine falcon. Mean dieldrin levels in these seabird species' eggs also did not exceed any environmental guidelines for protecting aquatic wildlife. However,  $\Sigma$ DDT levels in these species from these sites exceeded Canadian Environmental Quality Guidelines for protection of animals that consume aquatic biota and U.S. EPA guideline values for assessment of hazards to fish-eating wildlife.  $\Sigma$ PCB levels exceeded all guideline levels, implying that dietary intakes of  $\Sigma$ DDTs and  $\Sigma$ PCBs in some seabirds that prey on seabird eggs and chicks might be high enough to lead to effects.

#### 6.3.3.1. Eiders

In the previous AMAP assessment, common and Steller's eiders (*Polysticta stelleri*) from Canada, Russia, and Norway had  $\Sigma$ PCB levels below effects thresholds. A recent study in spectacled eiders (*Somateria fischeri*) from western Alaska found no biological effects that could be attributed to the low concentrations of POPs present (Trust *et al.*, 2000).

#### *Levels and intake assessment*

Mean  $\Sigma$ PCB levels in common eider muscle from Greenland, Iceland, and western Russia (Chukotka), and King eider (*Somateria spectabilis*) from Greenland, are below those expected to result in reproductive effects, assuming similar lipid levels in muscle compared to eggs (Figure 6-1).

Based on dietary LOECs for  $\Sigma$ PCBs (5000 ng/g ww), HCB (100 000 ng/g ww), DDE (5000 ng/g ww), and dieldrin (100 ng/g ww) in peregrine falcon prey, levels in blue mussels and anadromous and marine fish are several orders of magnitude below those expected to cause effects in animals that eat them (Annex Table 10). Mean dieldrin levels in blue mussels and Arctic anadromous and marine fish from Canadian, Norwegian, and Greenlandic waters (Annex Table 10) do not exceed any of the guidelines for protecting aquatic biota-eating wildlife (Table 6.1).  $\Sigma$ PCB and  $\Sigma$ DDT levels in a range of fish species from many Arctic sites exceed some, and in a few cases for  $\Sigma$ PCBs, all of the environmental quality guidelines for protecting aquatic biota consumers/fish-eating wildlife given in Table 6.1 (Figures 6.5 and 6.6). Toxaphene levels in several fish species also exceed the Canadian Tissue Residue Guideline for protecting wildlife consumers of aquatic biota (Table 6.1). This implies that dietary intakes of  $\Sigma$ DDTs,  $\Sigma$ PCBs, and toxaphene in some fish-eating seabirds may be high enough to lead to

effects if seabirds prey on these species. No data are available for levels of dioxin-like compounds in anadromous and marine fish so the dietary intake of TEQs cannot be assessed.

For ΣDDTs, the fish species that exceed both Canadian and U.S. EPA guidelines are spotted wolffish (liver) from southwest Greenland; Atlantic cod (liver) from Greenland, Iceland, and the Faroe Islands; short-horn sculpin (liver) from northeast and southwest Greenland and the Faroe Islands; starry ray (liver) from Greenland; Greenland shark (liver) from Canada; sturgeon (liver) from the Kara Sea; and, Greenland halibut (liver) from Greenland. The fish species that exceeded the International Joint Commission objectives for protection of aquatic life and wildlife of 1000 ng/g ww (Table 6.1) were Greenland shark (liver) from Canada and Greenland halibut (liver) from West Greenland (Figure 6.6).

For ΣPCBs, the species that exceed Canadian guidelines for avian predators are polar cod (liver) from Jan Mayen (Norway); Atlantic cod (liver) from Greenland and Iceland; shorthorn sculpin (liver) from Greenland and the Faroe Islands; starry ray (liver) from Greenland; Greenland shark (liver) from Canada; sturgeon (liver) from the Kara Sea; Greenland halibut (liver) from Greenland; Arctic char (muscle) from Nain, Canada, and a number of other marine species (liver) from around Jan Mayen (coalfish, long rough dab, checkered eelpout, and Atlantic poacher). The fish species that exceed International Joint Commission objectives are Atlantic cod (liver) from Greenland; shorthorn sculpin (liver) from southwest Greenland and the Faroe Islands; sturgeon (liver) from the Kara Sea; Greenland shark (liver) from Canada; Greenland halibut (liver) from Greenland; and, long rough dab (liver) and checkered eelpout from Jan Mayen. Only liver from sturgeon from the Kara Sea, Greenland halibut and Atlantic cod from Greenland, Greenland shark from Canada, and long rough dab and checkered eelpout from Jan Mayen exceed the U.S. EPA guideline values for assessment of hazards to fish-eating wildlife (Figure 6.5).

For toxaphene, the species that exceed the Canadian Tissue Residue Guideline are spotted wolffish (liver) from southwest Greenland; polar cod (liver) from Barrow, Alaska; Atlantic cod (liver) from southwest Greenland and Iceland; herring (whole fish) from the White Sea, Russia; shorthorn sculpin (liver) from southwest Greenland and the Faroe Islands; dab (liver) from Iceland; starry ray (liver) from southwest Greenland and Arctic char (fillet) from Barrow (Annex Table 10).

### 6.3.3.2. Grey heron, shag

#### *Reproductive effects*

A significant negative correlation was found between wet weight ΣPCB concentrations in yolk and egg volume, yolk mass, and hatchling mass in one-day-old shag hatchlings from the central Norwegian coast (Murvoll *et al.*, 1999).

#### *Retinol effects*

Grey heron hatchlings from two rookeries in Norway, one at Frøya on the central Norwegian coast and one at Finnjordøy on the north coast, were found to have sim-

ilar ΣPCB and TEQ (based on eight mono-*ortho* CBs) levels. No effects of either PCBs or TEQs on plasma retinol levels were found at either site (Jenssen *et al.*, 2001).

A borderline significant positive correlation was found between lipid weight ΣPCB concentrations in yolk and plasma retinol levels in one-day-old shag hatchlings from the central Norwegian coast (Murvoll *et al.*, 1999).

#### *Levels and intake assessment*

Mean ΣPCB concentrations in Norwegian shag hatchlings were 1.2 µg/g ww and for grey heron hatchling yolk sacs, 2.1-2.5 µg/g ww (range 0.57-4.9 µg/g ww) (Annex Table 11). Grey heron levels exceed the NOEL for hatching success in Forster's tern and several LOELs for egg mortality and embryo deformities in other bird species (Figure 6.1). For grey heron, the mean TEQ concentrations based on eight mono-*ortho* PCBs were 50-79 pg/g ww (range 21-179 pg TEQ/g ww) (1020-1170 pg/g lw), exceeding some NOAELs and LOAELs for reproductive effects in other bird species (Figure 6.2) (Annex Table 16).

The assessment for dietary intake of dieldrin, HCB, PCB, DDT, and toxaphene is the same as given for eiders (Section 6.3.3.1).

### 6.3.3.3. Alcids

#### *Cytochrome P450 activities/retinol effects*

Three groups of black guillemot nestlings in Saglek Bay, Canada, with low, medium, and high PCB exposures, were studied for several liver biomarkers (Kuzyk *et al.*, 2003). This bay has PCB-contaminated marine sediments due to a former military site. There was a significant dose-dependent increase in liver size, EROD activity up to a threshold of 100 ng/g ww liver, and reduced liver retinol and retinyl palmitate (females only) with increasing liver PCB concentrations. Many effects were more pronounced in female nestlings than in males.

#### *Levels and intake assessment*

In many cases, tissues other than eggs (muscle, liver and fat) have been analyzed in alcids (little auk, thick-billed murres, black guillemots and common guillemots) at various sites. This makes comparisons difficult because concentrations in other tissues may not be reflective of those found in the eggs (Braune and Norstrom, 1989; Drouillard and Norstrom, 2001). If only ΣPCB concentrations in eggs are used, values range from 0.087 to 0.27 µg/g ww for thick-billed murres from the Pribilof Islands, Alaska (Bering Sea), Lancaster Sound, Canada and Bjørnøya, and 0.11 to 0.29 µg/g ww in black guillemots from Qeqertarsuaq and Ittoqqortoormiit, Greenland and Bjørnøya (Annex Table 11). The ΣPCB concentrations based on eggs are below thresholds for effects in other fish-eating birds (Figure 6.1).

ΣPCB concentrations in liver of black guillemots from Baffin Bay and two sites at Saglek Bay (Canada), Qeqertarsuaq and Ittoqqortoormiit (Greenland), the Faroe Islands, and Svalbard are similar to or lower than levels seen in eggs from Greenland and Bjørnøya (eggs have lipid contents of approximately 10%, liver has a lipid content of 4-6%). However, ΣPCB concentrations in black guillemot livers from Jan Mayen (1.2 µg/g ww)

and at the more contaminated site at Saglek Bay (0.17–6.5 µg/g ww) are higher, and indicate that these populations probably have elevated levels in eggs as well. The levels at Jan Mayen are probably below thresholds for effects, but at Saglek Bay, they indicate that some individuals probably have levels that may be above thresholds for effects (Figure 6-1). ΣPCB concentrations in liver of thick-billed murres from Baffin Bay (Canada), Nuuk (Greenland), and Svalbard are lower than in eggs from Alaska and Lancaster Sound (Canada), indicating that eggs from these populations probably have levels below thresholds for effects.

TEQ concentrations based on PCDDs, PCDFs and non-*ortho* PCBs were 24 pg/g ww (719 pg/g lw) in liver and 26 pg/g ww in eggs for Canadian thick-billed murres collected in 1993 (Braune and Simon, 2002), 25 pg/g ww (640 pg/g lw) in black guillemot liver from Saglek Bay collected in 1999 and 9.8 pg/g ww (66 pg/g lw) for common guillemot eggs collected from the Faroe Islands in 2000 (Annex Table 16). The value for Canadian thick-billed murre eggs exceeds the LOAEL for reproductive effects in wood duck, a sensitive species, but for common guillemots, the levels are below thresholds (Figure 6-2). Lipid weight TEQ levels in liver in these species are well below the LOEL for induction of CYP1A in common terns (25000 pg TEQ/g lw).

The assessment for dietary intake of dieldrin, HCB, PCB, DDT, and toxaphene is the same as given for eiders (Section 6.3.3.1).

#### 6.3.3.4. Gulls

At Bjørnøya, Bourne and Bogan (1972) were the first to observe aberrant behaviors in glaucous gulls with high PCB levels. In the Svalbard archipelago, sick and dying glaucous gulls were found to have high PCB levels (Daelemans, 1994; Gabrielsen *et al.*, 1995).

##### *Reproductive effects*

Recently, a study was carried out to examine whether behavior at the nesting stage was negatively influenced by PCBs. Individual patterns of incubation and nest-site attentiveness were studied in relation to PCB concentrations in the blood of twenty-seven glaucous gulls in two breeding areas. PCB concentrations in the blood ranged from 52 ng/g ww to 1079 ng/g ww. There were significant differences between the two breeding areas, and females had significantly lower concentrations than males (Bustnes *et al.*, 2001).

Plasma PCB concentrations were positively related to the proportion of time the birds were absent from the nest, both overall and when not incubating, and to the number of absences, when controlling for possible confounding variables (area, sex, body condition, and others). Increased absence from the nest site in individual glaucous gulls with high blood concentrations of OCs suggests that they need more time to gather food because of either endocrine disruption or neurological disorders. This probably led to increased energetic costs during incubation and reduced reproductive output (Bustnes *et al.*, 2001).

To examine the effects of OCs on individual fitness, blood concentrations of various compounds were compared to reproductive parameters and adult survival in

individuals with different levels, controlling for a set of potential confounding variables (body condition, breeding areas, laying dates and others) (Bustnes *et al.*, 2003). Females with high circulating levels of OCs, including HCB, oxychlorane, DDE, and PCBs, were more likely to have non-viable eggs in the nest than females with low PCB levels. Moreover, the body condition at hatching was poorer for the first chick in the clutch in females with high concentrations of all persistent OCs. For the second chick in the clutch, only HCB, β-HCH, and CB28 showed such a negative relationship. Apart from a negative association between concentration of some OCs and laying date, no other reproductive parameters, such as clutch size, egg size, incubation time, nest predation or early chick survival, showed any association with OCs. Hence, reproduction seems moderately affected by OCs. Adult survival was significantly negatively related to four different OCs: HCB, DDE, CB153, and, in particular, oxychlorane. In long-lived birds, such as glaucous gulls, adult survival probability is the key parameter to which the population growth rate is most sensitive, suggesting that OCs may have considerable effect on growth in glaucous gull populations. However, the effects on specific populations will depend on the proportion of the population exposed to high intake of contaminants via the diet.

It was also demonstrated that glaucous gulls with high levels of various persistent OCs had asymmetry in wing feathers (i.e. feathers on the left and right wing were of different length). Feather asymmetry is a well-known indication of developmental stress, suggesting that OCs are an extra stress for the birds during molt. In this study, it was shown that the effect of HCB was much stronger than for the other OCs, such as PCBs and DDE (Bustnes *et al.*, 2002).

##### *Cytochrome P450 activities*

Only a weak association was found between ΣPCB (nine congeners) concentrations and CYP activity measured as EROD activity in Bjørnøya glaucous gulls (Henriksen *et al.*, 2000). No correlations were seen between OC levels and testosterone hydroxylation activity.

##### *Immunosuppression*

Numbers of intestinal macroparasites were compared to hepatic concentrations of several POPs in 40 glaucous gulls from Bjørnøya (Sagerup *et al.*, 2000). After controlling for nutritional condition, no single parasite species was significantly correlated with PCB or pesticide concentrations. However, the intensity of all nematodes grouped together was positively correlated with concentrations of *p,p'*-DDT, mirex, and ΣPCBs (nine congeners). This indicates that high POP concentrations may affect immune function in the glaucous gull.

##### *Thyroid and retinol effects*

No significant correlations were seen between ΣPCB concentrations and hepatic retinol and retinyl palmitate concentrations in forty glaucous gulls from Bjørnøya (Henriksen *et al.*, 2000). A significant negative correlation was seen between HCB, *p,p'*-DDE, and ΣPCB blood concentrations and plasma T4 levels in male glaucous gulls from Bjørnøya (Verreault *et al.*, 2002).



*Levels and intake assessment*

PCB concentrations in the brains of glaucous gulls found dead on Svalbard ranged from 0.9 to 29.5 µg/g ww (Gabrielsen *et al.*, 1995) and in living glaucous gulls, from 0.5 to 9.5 µg/g ww (Henriksen *et al.*, 1998b).

Tissues other than eggs (muscle, liver, and fat) have been analyzed in glaucous gulls at various sites (Annex Table 11). As stated previously, this makes comparisons difficult. Mean ΣPCB concentrations in liver range from 0.45 µg/g ww to 22.7 µg/g ww and indicate that several populations probably have elevated levels in eggs as well, some of which may be above thresholds for effects (Figure 6-1). The populations with high ΣPCB concentrations in liver that might be associated with egg levels above thresholds for effects are from Bjørnøya, Svalbard, Novaya Zemlya, Jan Mayen, and Franz Josef Land.

TEQ concentrations were calculated to be 2500 pg/g ww (approximately 60 000 pg/g lw) based on the nPCB and mono-*ortho* PCB concentrations in Svalbard glaucous gull liver (Daelemans *et al.*, 1992) (Annex Table 16). These TEQ levels exceed all NOAELs and LOAELs for reproductive effects and LD<sub>50</sub>s in a range of other bird species (Figure 6-2). Lipid weight TEQ levels in liver also exceed the LOEL for induction of CYP1A in common terns.

For ivory gull from north Baffin Bay, liver and fat tissues were analyzed (Annex Table 11). ΣPCB concentrations in liver are 0.3 µg/g ww, and indicate that levels in eggs are also low and probably below thresholds for effects. For great black-backed gulls from Jan Mayen, liver was analyzed. ΣPCB concentrations in liver are 9.6 µg/g ww, and indicate that levels in eggs may also be high and probably exceed some thresholds for effects.

TEQ concentrations based on PCDD/Fs, nPCBs and mono-*ortho* PCBs were determined in herring gulls and great black-backed gulls from four sites in northern Norway (Gabrielsen, 2002). These concentrations ranged from 37 to 72 pg TEQ/g ww. These TEQ levels exceed the LOAEL for reproductive effects in wood duck, a sensitive species (Figure 6-2).

Levels of DBT (n.d.-51 ng/g ww) and MBT (n.d.-14 ng/g ww) in glaucous gull livers from Bjørnøya (Berge *et al.*, 2002) are well below the thresholds associated with hepatic and immune effects. TBT levels were below the detection limits.

Ivory, herring, and great black-backed gulls feed primarily on fish but also scavenge. Glaucous gulls prey on eggs, chicks, and even adult seabirds as well as fish. The assessment for dietary intake of dieldrin, HCB, PCB, DDT, and toxaphene from fish is the same as given for eiders (Section 6.3.3.1). Based on contaminant levels found in eggs from thick-billed murres, black guillemots, black-legged kittiwakes, and fulmars from the Alaskan, Canadian, Greenlandic, and Norwegian Arctic (Annex Table 11), dieldrin, ΣDDT, and ΣPCB levels are below dietary LOECs for reproductive effects found in peregrine falcons. Mean dieldrin levels in these seabird species' eggs also do not exceed any environmental guidelines for protecting aquatic wildlife. However, ΣDDT levels in eggs and liver from these species from many of these sites, as well as kittiwakes from Russian sites in the Barents Sea, exceed Canadian and U.S. EPA guideline levels for protecting wildlife

that consume aquatic biota (Figure 6-6). Most ΣPCB levels exceed all guideline levels (Figure 6-5) and where measured, toxaphene levels exceed Canadian guidelines. PCDD/F and nPCB levels given as TEQs exceed Canadian (avian) and U.S. EPA guideline values (Table 6.1) in kittiwake, fulmar and murre eggs and liver, black guillemot liver and common guillemot eggs (Annex Table 16). Thus, seabirds that prey on seabird eggs, chicks and adults may have dietary intakes of TEQs, ΣDDTs, ΣPCBs and toxaphene high enough to lead to effects.

**6.3.3.5. Black-legged kittiwakes**

On a Swedish Arctic research expedition in 1996 in the Barents Sea, several juvenile kittiwakes with crossed bills and clump feet were observed (Kylin, 1997a; 1997b).

*Levels and intake assessment*

In many cases, tissues other than eggs (muscle, liver and fat) have been analyzed in black-legged kittiwakes at various sites. As stated previously, this makes comparisons difficult. If only values for ΣPCB concentrations in eggs are used, the mean is 0.28 µg/g ww at Lancaster Sound, Canada (Annex Table 11). The ΣPCB concentrations based on eggs do not exceed any thresholds for effects in fish-eating birds (Figure 6-1). ΣPCB concentrations in liver range from 0.12 to 1.4 µg/g ww in kittiwakes from sites in Baffin Bay, Nuuk (Greenland) Bjørnøya, Jan Mayen, Svalbard, Franz Josef Land, Novaya Zemlya, and along the Russian Barents Sea coast. This indicates that several populations probably have elevated levels in eggs as well, but these are probably below thresholds for effects.

TEQ concentrations based on PCDD/Fs and nPCBs were 47 pg/g ww (1117 pg TEQ/g lw) in liver and 78 pg/g ww in eggs for Canadian black-legged kittiwakes collected in 1993 (Annex Table 16) (Braune and Simon, 2002). The value for eggs exceeds the LOAEL for reproductive effects in wood duck, a sensitive species (Figure 6-2). Lipid weight levels in liver are well below the LOEL for induction of CYP1A in common terns.

The assessment for dietary intake of dieldrin, HCB, PCB, DDT, and toxaphene is the same as given for eiders (Section 6.3.3.1).

**6.3.3.6. Fulmar**

No effects studies have been carried out on fulmars in the Arctic.

*Levels and intake assessment*

Besides eggs, other tissues (liver, fat) have been analyzed in northern fulmars at various sites (Annex Table 11). As stated previously, this makes comparisons difficult. If only values for ΣPCB concentrations in eggs are used, the mean is 0.27 µg/g ww at Lancaster Sound, Canada (Annex Table 11). The mean ΣPCB concentrations in eggs from Lancaster Sound are below any effects thresholds (Figure 6-1). ΣPCB concentrations in liver range from 0.16 µg/g ww to 0.69 µg/g ww in fulmars from northern Baffin Bay and Jan Mayen. These ΣPCB concentrations indicate that levels in eggs are probably low and below thresholds for effects.



TEQ concentrations based on PCDD/Fs and non-ortho PCBs were 357 pg/g ww (8192 pg TEQ/g lw) in liver and 83 pg/g ww in eggs for Canadian Arctic fulmars collected in 1993 (Annex Table 16) (Braune and Simon, 2002). The value for eggs exceeds the LOAEL for reproductive effects in wood duck, a sensitive species (Figure 6-2). Lipid weight levels in liver are well below the LOEL for induction of CYP1A in common terns.

The assessment for dietary intake of dieldrin, HCB, PCB, DDT, and toxaphene is the same as given for eiders (Section 6.3.3.1).

#### 6.3.3.7. Great skua

No effects studies have been carried out on great skuas in the Arctic.

##### *Levels and intake assessment*

Only liver has been analyzed in great skuas from Jan Mayen. The mean  $\Sigma$ PCB concentration in liver was 15.9  $\mu$ g/g ww. This indicates that great skuas at Jan Mayen probably have elevated levels in eggs as well, and these are probably above thresholds for effects (Figure 6-1). The dietary assessment is the same as for glaucous gulls (Section 6.3.3.4).

#### 6.3.4. Pinnipeds

In the previous AMAP assessment, increased CYP1A enzyme activities were correlated to  $\Sigma$ PCB and dieldrin levels in Canadian ringed seals and to  $\Sigma$ PCB levels in West Ice (Jan Mayen area) hooded seals.  $\Sigma$ PCB levels in Arctic harp, ringed, harbour and grey seals from all sites studied exceeded the NOAEL and LOAEL for subtle neurobehavioral effects if they were as sensitive as the offspring of rhesus monkeys and humans, but were below the NOEL for otter reproduction and mink kit survival (Figure 6-3). PCDD/F and nPCB levels expressed as TEQs in ringed and harp seals from Svalbard, the Greenland Sea, and several sites in Canada were somewhat lower than levels associated with immunosuppressive effects in harbour seal (Figure 6-4).

Concentrations of dieldrin,  $\Sigma$ DDTs, and chlordanes in marine crustaceans and fish were several orders of magnitude below those expected to result in effects on seal reproduction, and toxaphene levels in fish were below those associated with thyroid effects.  $\Sigma$ PCB levels were also below effect levels in crustaceans, but levels exceeded the dietary NOAEC for reproduction in several fish species. However,  $\Sigma$ PCB and  $\Sigma$ DDT levels in a range of fish species from many Arctic sites exceeded some, and in a few cases for  $\Sigma$ PCBs, all of the environmental quality guidelines for protecting fish-eating wildlife. This indicates that dietary intakes of  $\Sigma$ DDTs and  $\Sigma$ PCBs in fish-eating marine mammals may be high enough to lead to effects.

##### 6.3.4.1. Seals and sea lions

###### *Reproductive effects*

Breeding rookeries for more than 72% of the world's population of northern fur seals are located on the two largest Pribilof Islands, St. Paul and St. George, Alaska in the Bering Sea (Loughlin *et al.*, 1994). The current

Pribilof stock abundance is less than half of historical levels and is listed as depleted under the *Marine Mammal Protection Act* (Loughlin *et al.*, 1994; York *et al.*, 1997). The St. George subpopulation underwent an unexplained decline of 4-6% per year for more than a decade prior to the mid-1990s study (York *et al.*, 1997). Long-term monitoring of population trends suggest that the decline was due, at least in part, to increased post-weaning mortality at sea (Trites and Larkin, 1989; Trites, 1992). Cause(s) of the increased mortality is unknown, but is thought to be associated with shifts in the abundance and composition of their primary prey species.

Steller sea lion populations have been declining in western Alaska including the Aleutian Islands, and the western stocks are considered endangered.

###### *Cytochrome P450 effects*

Studies have been done on liver enzyme induction in ringed and harp seals around Svalbard, correlated to levels of OCs such as PCB and toxaphene. Elevated hepatic EROD activities were found in subadult harp seals sampled from the northwest Barents Sea, east of Svalbard (Wolkers *et al.*, 2000). No correlation was found between these activities and the PCB concentrations in the seals. A highly positive correlation was, however, found between toxaphene levels and testosterone 6- $\beta$  hydroxylation activities (CYP3A). A positive relationship was found between CYP enzyme activity and  $\Sigma$ PCB levels in ringed seals from Svalbard (Wolkers *et al.*, 1998b). Ringed seals from Svalbard and grey seals from Sable Island (Canadian Arctic) had lower EROD and PROD activities than grey and ringed seals from the Baltic Sea (Nyman *et al.*, 2000).

###### *Thyroid and retinol effects*

Retinol levels in northern fur seal neonates were negatively correlated to two recalcitrant PCB congeners in whole blood: CB138 and CB153/87 (rank correlation coefficients of -0.403 and -0.452, respectively,  $p < 0.05$ ) (Beckmen, 1999). Serum retinol levels in neonates were also negatively correlated to the TEQs in the perinatal milk (correlation coefficient of -0.475,  $p = 0.029$ ,  $n = 21$ ). Total T4 was negatively correlated with CB101/99/149/196, CB118, CB138, and TEQs (Spearman rank correlation coefficients = -0.313, -0.519, -0.457, -0.354, respectively,  $p < 0.05$ ). The negative correlation of retinol and thyroid hormones with PCBs in northern fur seal blood suggests that blood levels of these may be reduced because of PCB exposure in young pups. Thus, PCB exposure in young pups has the potential to affect immune function, and therefore, health, both through direct immunosuppression and indirectly, by lowering circulating levels of retinol and thyroxine.

###### *Immune system effects*

The native Aleut populations of the villages on St. George and St. Paul are dependent on an annual subsistence harvest of subadult (2-4 years of age) male fur seals both culturally and as a major source of protein. Aleut concerns over the unexplained decline in the population of northern fur seals prompted a study to evaluate the potential effects of OC contaminant exposure on immune function in a cohort of free-ranging pups.

In 1996, 50 perinatal pups were captured for blood sample collection (during the ten-day perinatal period and referred to as neonates) and forty-three were re-sampled ('pups') 29 to 51 days later. Groups of pups were compared based on the relative age of the dam to study immunologic effects of exposure via milk to high (young dam, presumably primiparous) or low (old dam, multiparous) doses of OCs (Beckmen *et al.*, 1999). There were no significant differences in the mass or length (when adjusted for sex) of pups during the perinatal period between pups born to young and old dams. Likewise, growth rates during the study were similar, and the survival rates to the middle of the nursing period were not significantly different although the survival rates of pups of young dams were higher (88% vs. 84%,  $p=0.858$ ). The only significant difference was a slightly higher body condition index score in pups of old dams ( $p=0.047$ ). Mean blood OC levels were higher in neonates than at recapture, and neonates of young dams had higher mean blood OC levels than neonates of older dams (Beckmen, 1999; Beckmen *et al.*, 1999).

In the same pups, humoral immune function was assessed by antibody responses to tetanus toxoid vaccination and total immunoglobulin levels. Cellular immune function was assessed using mitogen-induced lymphocyte proliferation assays. Additional indicators of health status included complete blood cell counts and haptoglobin levels. A higher proportion of pups born to old dams developed a two-fold or greater increase in tetanus antibodies compared to the pups of young dams. Forty-one percent (9 of 22) of the pups of old dams responded with the expected 2-fold or greater increase, whereas only 5% of the pups of young dams responded to the antigen (1 of 21). The difference in the proportion of pups responding by the dam's age was significant ( $Z=2.443$ ,  $p=0.015$ ). When hemograms of neonates of young and old dams were compared, the mean hematocrit, total plasma protein and absolute eosinophil counts were significantly greater in the neonates of old dams. In neonates, elevated immature (band) neutrophil counts (indicative of inflammation) were correlated to higher TEQs ( $r=0.347$ ,  $p=0.016$ ,  $n=48$ ). In recaptured pups, the total leukocyte counts were positively correlated to higher total PCB concentrations in blood ( $r=0.359$ ,  $p=0.023$ ,  $n=41$ ).

Levels of serum haptoglobin were used as a further measure of subclinical inflammation. There were no differences between neonatal and recapture levels for the pups of old dams, but haptoglobin levels increased significantly from neonatal to recapture for pups of young dams (Mann-Whitney Rank Sum test,  $p<0.001$ ). No direct correlations of serum haptoglobin concentration with blood OC concentrations were detected. Neonates of young dams had significantly ( $p<0.001$ ) lower mean immunoglobulin levels than neonates of old dams,  $3.14 \pm 1.2$   $\mu\text{g/ml}$  versus  $4.6 \pm 1.1$   $\mu\text{g/ml}$ , respectively. Lymphoproliferative responses, when combined with the results of the previous year's cohort of neonatal fur seal pups, were negatively correlated with blood OC levels, indicating a possible suppression of cellular immune function (Beckman, 1999).

A recent study examining a cohort of 12, known-age, female pups born to young dams and sampled at ap-

proximately monthly intervals from birth to weaning, further substantiated the results of the original study. The pups were given both primary and secondary vaccinations with tetanus toxoid and examined for primary and secondary (memory) antibody response. When converted on a molecular weight basis, the effect of PCB blood levels at the time the initial and booster vaccinations with tetanus toxoid were given had a highly significant ( $p=0.008$  and  $0.026$ , respectively) negative effect on the increase in antibody titer. All congeners examined, except for *p,p'*-DDE, were significant individually. In these first-born pups, using set correlation analysis, it was determined that the total immunoglobulin concentrations were related to both TEQs (negative) and the age (in days) of the pup ( $p<0.0001$ ,  $p=0.0512$ , respectively). Additionally, total immunoglobulins were related to  $\Sigma\text{PCB}$  concentrations and the age of the pup. Perinatal TEQs had a significant negative correlation to the total immunoglobulin levels at that time and at the next recapture. TEQs at the first recapture were also significantly negatively correlated to total immunoglobulins at that time and nearly significantly negatively correlated subsequently ( $r=-0.65$  and  $-0.633$ ;  $p=0.0501$  and  $0.0583$ , respectively). Set correlation analysis confirms this effect is correlated to TEQs by taking into consideration the effect of age on immunoglobulin levels. The strong correlation with PCB exposure expressed as TEQs suggests that the effect on humoral immunity may be mediated through, or by a similar mechanism as, the Ah-receptor.  $\Sigma\text{PCBs}$ , TEQs, and *p,p'*-DDE were all negatively correlated (Spearman Rank Order Correlation) to lymphoproliferative assays. However, when a set correlation analysis was applied, age explained most of the effect. Thus, PCB exposure appears to have a negative impact on humoral immunity (immunoglobulin levels and tetanus antibody responses), but the effects on cellular immune function were not substantiated in this cohort (Beckmen, 2002).

Blood and blubber samples obtained from 24 free-ranging Steller sea lion pups and juveniles during the course of live-capture studies in the ranges of both eastern and western stocks, have recently been subjected to similar OC contaminant analysis and immune function assays as the northern fur seals. In this species as well, there were significant negative correlations between lymphocyte proliferative responses to both T cell and mixed B and T cell mitogens and increased blubber TEQs (concanavalin A:  $r=-0.440$ ; pokeweed mitogen:  $r=-0.448$ ,  $p<0.05$ ). In a subset of eight pups in the eastern, endangered stock, the total immunoglobulin serum concentrations were strongly negatively correlated with blubber concentrations of  $\Sigma\text{PCBs}$ , TCDD TEQs and *p,p'*-DDE ( $r=-0.933$ ,  $p=0.005$ ;  $r=-0.881$ ,  $p<0.001$ ;  $r=-0.893$ ,  $p<0.001$ , respectively) (Beckmen, 2002).

#### Levels and intake assessment

The mean  $\Sigma\text{PCB}$  levels in Arctic ringed, harbour, grey, spotted, and bearded seals, Steller sea lions (Gulf of Alaska and southeast Alaskan populations) and northern fur seals range from 72 to 8400 ng/g lw in blubber (Annex Table 12). These levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring

of rhesus monkeys and humans, the NOEL for vitamin A reduction in otter, and in some species, are above the NOEL for otter reproduction, but below that for mink kit survival (Figure 6-3). Highest  $\Sigma$ PCB levels were found for ringed seal from Saglek Bay, Labrador (9400 ng/g lw) and from the Kara Sea (1900-11000 ng/g lw). Both exceed the NOEL for mink kit survival, and the levels in the Kara Sea seals are just at the LOEL for vitamin A reduction in otter. These levels are considerably lower than those associated with poor reproductive success in harbour and ringed seal (Figure 6-3). No biopsy data are available for Steller sea lions from endangered populations in the Aleutian Islands. The  $\Sigma$ PCB levels are much higher in scat (up to 7000 ng/g lw) from these populations when compared to those from the other two areas, indicating higher body burdens, but no assessment of these endangered populations can be made. The  $\Sigma$ PCB levels in harp seal from Svalbard range from 247 to 20 400 ng/g lw in blubber. These levels exceed the thresholds for subtle neurobehavioral effects, the NOEL and LOEL for vitamin A reduction in otter, the NOEL for mink kit survival, the LOAEL for decreased kit production and kit body weight gain in mink (Figure 6-3). As well, these levels exceed the threshold associated with immunosuppression in harbour seals and are just below the threshold for immune effects in rhesus monkeys (Figure 6-3).

Levels of nPCBs and mono-*ortho* PCBs given as TEQs range from 27 to 90 pg/g lw in blubber and blood from various age groups of Alaskan northern fur seals (Annex Table 16). Highest TEQs are found in pups. The TEQ levels for pups are below the combined nPCB and mono-*ortho* TEQs (190 pg/g lw) associated with immunosuppressive effects in harbour seals (Figure 6-4). For ringed seals from Pangnirtung, nPCB and mono-*ortho* PCB concentrations in blubber given as TEQs were 0.51 to 0.85 pg/g lw. For ringed seals from Holman Island, TEQs based on PCDD/Fs, nPCBs and mono-*ortho* PCBs ranged from 4.8 to 97 pg/g lw and for ringed seals from the Kara Sea, nPCB and mono-*ortho* PCB concentrations in blubber given as TEQs were 160 pg/g lw. For ringed seals, bearded seals and spotted seals from eastern Russia (Chukotka), TEQs based on PCDD/Fs were 1.0-1.5 pg/g lw. These TEQ levels are all below the threshold for immunosuppression in harbour seals (Figure 6-4).

PFOS levels in grey, ringed, and northern fur seal from the Arctic as well as Steller sea lions from the Alaskan coast are lower than in marine mammals from more southerly latitudes, ranging from <3 to 120 ng/g ww in liver or plasma (Giesy and Kannan, 2001; Kannan *et al.*, 2001a). These levels are well below the NOAEL (15 000 ng/g ww in liver) and LOAEL (58 000 ng/g ww in liver) for second generation effects in rats.

Levels of TBT (1.9-5.6 ng/g ww) and DBT (n.d.-20 ng/g ww) in Alaskan Steller sea lions, in Svalbard ringed seals (DBT-3.1 ng/g ww, MBT-1.5 ng/g ww, TBT not detected), and Canadian ringed seals (below detection) are well below the thresholds associated with hepatic and immune effects in laboratory rodents.

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin,  $\Sigma$ DDTs (Figure 6-6), and chlordanes in marine fish are several orders of magnitude below those expected to result in effects on ma-

rine mammal reproduction. Based on the NOAEC for rats and dogs, toxaphene levels in fish are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of  $\Sigma$ PCBs in tissues from several fish species exceed the dietary NOAEC for reproduction of 72 ng/g ww (Figure 6-5). These include shorthorn sculpin (liver) from several sites on Greenland and the Faroe Islands; Greenland halibut (liver) and Atlantic cod (liver) from Greenland and Iceland; starry ray (liver) from Greenland; sturgeon (liver) from the Kara Sea; Greenland shark (liver) from Davis Strait and Cumberland Sound; polar cod (liver) from Jan Mayen; and, several species (liver) from around Jan Mayen (long rough dab, Atlantic poacher, checkered eelpout) (Annex Table 10).

Mean dieldrin levels in Arctic anadromous and marine fish from Canadian, Norwegian, and Greenlandic waters (Annex Table 10) do not exceed any of the guidelines for protecting fish-eating wildlife (Table 6.1). However,  $\Sigma$ PCB and  $\Sigma$ DDT levels in a range of fish species from many Arctic sites do exceed some, and in a few cases for  $\Sigma$ PCBs, all of the environmental quality guidelines for protecting fish-eating wildlife given in Table 6.1 (Figures 6.5 and 6.6). Toxaphene levels in several fish species also exceed the Canadian Tissue Residue Guideline for protecting wildlife consumers of aquatic biota. This implies that dietary intakes of  $\Sigma$ DDTs,  $\Sigma$ PCBs, and toxaphene in some fish-eating mammals may be high enough to lead to effects if mammals prey on these fish species. No data are available for levels of dioxin-like compounds in anadromous and marine fish so the dietary intake of TEQs cannot be assessed.

For  $\Sigma$ DDTs, the fish species that exceed both Canadian and U.S. EPA guidelines are spotted wolffish (liver) from southwest Greenland; Atlantic cod (liver) from Greenland, Iceland and the Faroe Islands; shorthorn sculpin (liver) from northeast and southwest Greenland and the Faroe Islands; starry ray (liver) from Greenland; Greenland shark (liver) from Canada; sturgeon (liver) from the Kara Sea; and, Greenland halibut (liver) from Greenland. The fish species that exceeded the International Joint Commission objectives for protection of aquatic life and wildlife of 1000 ng/g ww were Greenland shark (liver) from Canada and Greenland halibut (liver) from West Greenland (Figure 6-6).

For  $\Sigma$ PCBs, the species that exceed Canadian guidelines for mammalian predators are spotted wolffish (liver) from southwest Greenland; polar cod from Jan Mayen (Norway); Atlantic cod (liver) from Greenland, the Faroe Islands, Iceland and Svalbard; shorthorn sculpin (liver) from Greenland and the Faroe Islands; dab (liver) from Iceland; starry ray (liver) from Greenland; Greenland shark (liver) from Canada; sturgeon (liver) from the Kara Sea; Greenland halibut (liver and muscle) from Greenland; Arctic char (muscle) from several sites in Canada and liver from some Russian sites; Atlantic salmon (*Salmo salar*) (muscle) from West Greenland; and, a number of other marine species (liver) from around Jan Mayen (coalfish, long rough dab, checkered eelpout, daubed shanny, Atlantic poacher, grey gurnard) and the White Sea, Russia (*Gadus sp.*, herring, navaga (*Eliginius navaga*), sculpin). The fish species that exceed International Joint Commission objectives are Atlantic cod (liver) from Greenland; short-



horn sculpin (liver) from southwest Greenland and the Faroe Islands; sturgeon (liver) from the Kara Sea; Greenland shark (liver) from Canada; Greenland halibut (liver) from Greenland; and, long rough dab (liver) and checkered eelpout from Jan Mayen. Only liver from sturgeon from the Kara Sea, Greenland halibut, and Atlantic cod from Greenland, Greenland shark from Canada as well as long rough dab and checkered eelpout from Jan Mayen exceed the U.S. EPA guideline values for assessment of hazards to fish-eating wildlife (Figure 6-5).

For toxaphene, the species that exceed the Canadian Tissue Residue Guideline are spotted wolffish (liver) from southwest Greenland; polar cod (liver) from Barrow, Alaska; Atlantic cod (liver) from southwest Greenland and Iceland; herring (whole fish) from the White Sea, Russia; shorthorn sculpin (liver) from southwest Greenland, and the Faroe Islands; dab (liver) from Iceland; starry ray (liver) from southwest Greenland and Arctic char (fillet) from Barrow (Annex Table 10).

#### 6.3.4.2. Walrus

In the previous AMAP assessment,  $\Sigma$ PCB levels for the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans were exceeded in walrus from eastern Baffin Island, eastern Hudson Bay, northeastern Hudson Bay, and Svalbard.  $\Sigma$ PCB levels in walrus from eastern Hudson Bay (Inukjuak) and Svalbard also exceeded the NOEL for otter reproduction and mink kit survival (Figure 6-3).

Walrus from Inukjuak in eastern Hudson Bay had PCDD/F and/or nPCB TEQ levels that exceeded TEQ levels associated with immunosuppression in harbour seals (Figure 6-4).

For walrus feeding on marine invertebrates, no risks for reproductive effects were indicated based on levels of dieldrin,  $\Sigma$ DDTs, chlordanes,  $\Sigma$ PCBs, and PCDD/Fs, or based on environmental guidelines.

Some walrus prey on ringed seal, and  $\Sigma$ PCBs,  $\Sigma$ DDTs, and TEQs in ringed seal blubber exceeded dietary NOAECs and a range of guidelines for protecting aquatic wildlife, indicating exposure to these substances at levels that could be expected to lead to effects.

No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

Mean  $\Sigma$ PCB levels in walrus from Alaska (Bering Sea), Canada, and northeast and northwest Greenland ranged from 33 ng/g lw to 3412 ng/g lw (Annex Table 12).  $\Sigma$ PCB levels in walrus from Alaska, Canada, and northwest Greenland (Avanersuaq) are below all thresholds for effects (Figure 6-3).  $\Sigma$ PCB levels in northeast Greenland walrus from Ittoqqortoormiit exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, and are close to the NOEL for vitamin A reduction in otter (Figure 6-3).

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin,  $\Sigma$ DDTs, and chlordanes in marine invertebrates are below those expected to result in effects on walrus reproduction.  $\Sigma$ PCB levels in the diet are also below effect levels in invertebrates.

Levels of  $\Sigma$ DDTs,  $\Sigma$ PCBs, and PCDD/Fs in invertebrates are also below the various environmental guidelines (Table 6.1). Mean dieldrin levels in blue mussels from Canadian and Greenlandic waters (Annex Table 10) do not exceed any of the guidelines for protecting fish-eating wildlife (Table 6.1). Toxaphene levels in blue mussels from one Greenland site and from several sites around Iceland are below the Canadian guideline. However,  $\Sigma$ PCB levels in blue mussels from some sites in northern Quebec, Canada, exceed the Canadian Tissue Residue Guideline for protecting wildlife consumers of aquatic biota (Figure 6-5).

#### 6.3.5. Cetaceans

In the previous AMAP assessment, increased CYP1A enzyme activities were correlated to mono-*ortho* and nPCB concentrations in beluga from the Canadian Arctic. Minke whale, beluga, and narwhal had  $\Sigma$ PCB levels which exceeded the NOAEL and LOAEL for subtle neurobehavioral effects if they are as sensitive as offspring of rhesus monkeys and humans, but were below the NOEL for otter reproduction and mink kit survival (Figure 6-3). For harbour porpoise from the southern Barents Sea,  $\Sigma$ PCB levels also exceeded the NOEL for otter reproduction, mink kit survival, and the levels associated with immunosuppression, and approached the levels associated with poor reproductive success in harbour seal (Figure 6-3). Canadian beluga and narwhal had TEQ levels that were considerably lower than those associated with immunosuppressive effects (Figure 6-4).

Concentrations of dieldrin,  $\Sigma$ DDTs, and chlordanes in cetacean food items, such as marine crustaceans and fish, were several orders of magnitude below those expected to result in effects on reproduction, and dietary toxaphene levels in fish were below those associated with thyroid effects.  $\Sigma$ PCB levels were also below dietary effect levels in crustaceans, but levels exceeded the dietary NOAEC for reproduction in several fish species. However,  $\Sigma$ PCB and  $\Sigma$ DDT levels in a range of fish species from many Arctic sites exceeded some, and in a few cases for  $\Sigma$ PCBs, all of the environmental quality guidelines for protecting fish-eating wildlife, indicating that dietary intakes of  $\Sigma$ DDTs and  $\Sigma$ PCBs in fish-eating mammals could be high enough to lead to effects.

No biological effects studies have been carried out on cetaceans since the previous assessment.

#### 6.3.5.1. Mysticetes

##### 6.3.5.1.1. Minke whales

#### *Levels and intake assessment*

$\Sigma$ PCB levels in minke whales ranged from 230 ng/g lw to 20760 ng/g lw. Mean  $\Sigma$ PCB levels in all minke whales exceed the NOAEL and LOAEL for subtle neurobehavioral effects if they are as sensitive as the offspring of rhesus monkeys and humans. Some minke whales from several sites (Jan Mayen, North Sea, Svalbard, northwest Kola Peninsula, northern Norway/northwest Russia) have  $\Sigma$ PCB levels that exceed the NOEL and LOEL for vitamin A reduction in otter. Some minke whales from northern Norway/northwest Russia also exceed



the NOEL for otter reproduction and mink kit survival, the threshold for decreased kit production and kit body weight gain in mink, the threshold associated with immunosuppression and vitamin A disruption in harbour seals, but are just below the LOAEL for immune effects in rhesus monkeys (Figure 6-3).

TEQ concentrations based on PCDD/Fs, nPCB, and mono-*ortho* PCBs were 15-74 pg TEQ/g lw in Svalbard (Spitsbergen) minke whales (Annex Table 16). These concentrations are below the threshold associated with immunosuppression in harbour seals (Figure 6-4).

The dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene is the same as for seals and sea lions (Section 6.3.4.1).

#### 6.3.5.1.2. Grey whales

##### *Levels and intake assessment*

$\Sigma$ PCB levels in Bering Sea and Chukotka grey whales ranged from 230 to 2700 ng/g lw, with means of 460-1400 ng/g lw. The levels in the Bering Sea whales exceed the NOAEL and LOAEL for subtle neurobehavioral effects if they are as sensitive as the offspring of rhesus monkeys and humans, but are below the NOELs for otter reproduction and mink kit survival (Figure 6-3). The TEQ concentration based on PCDD/F levels in the Chukotka whales was 1.8 pg/g lw, which is below thresholds for effects (Figure 6-4).

#### 6.3.5.1.3. Bowhead whales

##### *Levels and intake assessment*

Mean  $\Sigma$ PCB levels in Alaskan bowhead whales are below thresholds for effects (Figure 6-3).

#### 6.3.5.2. Odontocetes

##### 6.3.5.2.1. Beluga

##### *Levels and intake assessment*

The mean  $\Sigma$ PCB levels in beluga from Alaska, Canada, Greenland, and Svalbard range from 700 to 9000 ng/g lw with the highest concentrations at Point Lay (Alaska), Hendriksen Island (western Canada) and Kimmirut (eastern Canada) (Annex Table 13). These  $\Sigma$ PCB levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL and LOEL for vitamin A reduction in otter, and the NOEL for otter reproduction and mink kit survival (Figure 6-3). The  $\Sigma$ PCB levels in beluga brain from Alaska are below those associated with decreased dopamine concentrations in non-human primate brain tissue.

TEQ concentrations based on nPCBs and mono-*ortho* PCBs in beluga from Kimmirut were 0.3-2.4 pg/g lw (Annex Table 16). If PCNs are included, the TEQs were 0.33-2.9 pg/g lw. TEQ concentrations based on nPCBs in beluga from Cumberland Sound (Baffin Island) were 6.1 pg/g lw. The TEQ levels in beluga are well below the threshold associated with immunosuppression in harbour seals (Figure 6-4).

Levels of TBT and DBT in Canadian beluga were below detection limits. The dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene is the same as for seals and sea lions (Section 6.3.4.1).

##### 6.3.5.2.2. Killer whales

##### *Levels and intake assessment*

The mean  $\Sigma$ PCB levels in killer whales from Alaska (Prince William Sound) are 14 400 ng/g lw in residents (range: 1100-65 000 ng/g lw) and 240 000 ng/g lw in transients (range: 66 000-550 000 ng/g lw) (Annex Table 13). The mean  $\Sigma$ PCB levels in resident killer whales exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL and LOEL for vitamin A reduction in otter, the NOEL for otter reproduction and mink kit survival, and the threshold for decreased kit production and kit body weight gain in mink. Based on the maximum levels,  $\Sigma$ PCB concentrations exceed the threshold associated with immunosuppression and vitamin A disruption in harbour seals, the LOAEL for immune effects in rhesus monkeys, the threshold for poor reproductive success in harbour seal, and the EC<sub>50</sub> for decreased litter size in mink (Figure 6-3). For the transient killer whales, mean and maximum  $\Sigma$ PCB levels are above all thresholds (Figure 6-3).

Mean TEQ concentrations based on mono-*ortho* PCBs were 100 pg/g lw (range: 5.9-470 pg/g lw) for residents and 860 pg/g lw (range: 190-2400 pg/g lw) for transients (Annex Table 16). The mean TEQ for residents is below the threshold associated with immunosuppression in harbour seals, but individuals with maximum values exceed this threshold (Figure 6-4). All TEQs for transients exceed this threshold.

The resident killer whales feed primarily on fish. Therefore, the dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene for resident killer whales is the same as for seals and sea lions (Section 6.3.4.1). The transient killer whales feed on other marine mammals. Based on mean POP levels in ringed, bearded and northern fur seals as well as Steller sea lion blubber from various sites in Alaska (Annex Table 12), dieldrin, chlordanes (heptachlor epoxide), and  $\Sigma$ DDT levels are below the dietary NOAECs for reproductive effects in mink. Assuming that marine mammal-eating transient killer whales are as sensitive as mink, mean levels of  $\Sigma$ PCBs in these seal species exceed the dietary NOAEC for reproduction of 72 ng/g ww. As prey items, mean  $\Sigma$ DDT levels in all Alaskan seal species exceed Canadian and U.S. EPA guidelines for protecting aquatic wildlife, and for northern fur seal, International Joint Commission objectives for protection of aquatic life and wildlife are also exceeded (Figure 6-6). For  $\Sigma$ PCBs, all Alaskan seal species exceed all environmental guidelines for protecting aquatic wildlife (Figure 6-5) (Table 6.1). For nPCB TEQs, northern fur seal blubber exceeds both the Canadian mammalian tissue residue guideline and the U.S. EPA guideline for protecting wildlife. No data for toxaphene are available in Alaskan seal species and the dietary intake cannot be assessed.

##### 6.3.5.2.3. Long-finned pilot whales

##### *Levels and intake assessment*

The mean  $\Sigma$ PCB concentrations in long-finned pilot whales from the Faroe Islands ranged from 16 000 ng/g lw to 38 000 ng/g lw (Annex Table 13). The mean  $\Sigma$ PCB

concentrations exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL and LOEL for vitamin A reduction in otter, the NOEL for otter reproduction and mink kit survival, and the threshold for decreased kit production and kit body weight gain in mink (Figure 6-3). Based on the maximum levels,  $\Sigma$ PCB concentrations exceed the threshold associated with immunosuppression and vitamin A disruption in harbour seals, the LOAEL for immune effects in rhesus monkeys, and the threshold for poor reproductive success in harbour seal (Figure 6-3). Low concentrations of DBT and TBT were found in a few adults and one fetus, but levels were well below thresholds for effects.

The dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene is the same as for seals and sea lions (Section 6.3.4.1).

#### 6.3.5.2.4. Narwhal

##### *Levels and intake assessment*

Many measurements of  $\Sigma$ PCB concentrations in narwhal blubber are based on only ten congeners ( $\Sigma$ PCB<sub>10</sub>). The mean  $\Sigma$ PCB<sub>10</sub> concentrations in narwhal range from 260 to 4750 ng/g lw (Annex Table 13). In general, the  $\Sigma$ PCB<sub>10</sub> levels are approximately 30% of  $\Sigma$ PCB levels in beluga, and in one set of narwhal data where both values were calculated. Assuming this is the case for the other narwhal data, then the range of  $\Sigma$ PCB concentrations in narwhal from Canada, Greenland, and Svalbard is estimated to be 900 to 16 000 ng/g lw. These  $\Sigma$ PCB levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL and LOEL for vitamin A reduction in otter, the NOEL for otter reproduction and mink kit survival, and the threshold for decreased kit production and kit body weight gain in mink, but are just below the threshold associated with immunosuppression and vitamin A disruption in harbour seals (Figure 6-3).

Narwhal eat squid, Arctic cod, shrimp, and Greenland halibut. Based on POP concentrations in Arctic marine fish, the dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene is the same as for seals and sea lions (Section 6.3.4.1).

#### 6.3.5.2.5. Harbour porpoise

##### *Levels and intake assessment*

Mean  $\Sigma$ PCB levels in harbour porpoise from northern Norway are 15 000 ng/g lw (range: 7200-33 000 ng/g lw) and from southwest Greenland, 1300 ng/g lw (range: 880-1530 ng/g lw) (Annex Table 13). The mean  $\Sigma$ PCB levels from both sites exceed the NOAEL and LOAEL for subtle neurobehavioral effects if harbour porpoise are as sensitive as offspring of rhesus monkeys and humans. The mean  $\Sigma$ PCB levels for the porpoises from Norway also exceed the NOEL and LOEL for vitamin A reduction in otter, the NOEL for otter reproduction and mink kit survival, and the threshold for decreased kit production and kit body weight gain in mink, but are below the threshold associated with immunosuppression and decreased vitamin A in harbour seals (Figure 6-3). The maximum  $\Sigma$ PCB levels, however, exceed the threshold associated with immunosuppression and vitamin A

disruption in harbour seals, the LOAEL for immune effects in rhesus monkeys, and the threshold for poor reproductive success in harbour seals. The dietary intake assessment for  $\Sigma$ PCBs,  $\Sigma$ DDTs, dieldrin, chlordanes, and toxaphene is the same as for seals and sea lions (Section 6.3.4.1).

Levels of PCDD/Fs given as TEQs in Greenland harbour porpoise range from 0.2 to 0.9 pg/g lw, and the mean TEQ concentration for PCDD/Fs, nPCBs, and mono-*ortho* PCBs in the Norwegian harbour porpoises was 111 pg/g lw (Annex Table 16). These TEQ levels do not exceed the threshold associated with immunosuppression in harbour seals (Figure 6-4).

Levels of TBT (55-151 ng/g ww), DBT (127-490 ng/g ww), and MBT (11-58 ng/g ww) in Norwegian harbour porpoise from the coast of the Barents Sea are below the thresholds associated with hepatic and immune effects in laboratory rodents.

#### 6.3.6. Polar bear

In the previous AMAP assessment, high cub mortality was found in Svalbard polar bears. A significant negative correlation was found between retinol and  $\Sigma$ PCB concentrations in Svalbard polar bears. Hepatic CYP1A1 and 1A2 content in Canadian male polar bears were found to be correlated with levels of mono-*ortho*- and nPCBs, and CYP2B content was correlated with concentrations of total chlordane (mainly oxychlordane and nonachlor), and total *ortho*-substituted PCBs. A spatial study of POP concentrations in fat was carried out which included polar bear populations from 16 areas across the Arctic; from Canada, Greenland, and Svalbard. All  $\Sigma$ PCB levels exceeded the NOAEL and LOAEL levels found for subtle neurobehavioral effects in offspring, if polar bear are as sensitive as offspring of rhesus monkeys and humans. The  $\Sigma$ PCB levels also exceeded the NOEL for kit survival in mink in four of the areas: Svalbard, East Greenland, M'Clure Strait, and eastern Hudson Bay.  $\Sigma$ PCB levels were close to the kit survival NOEL in several other areas.  $\Sigma$ PCB levels in polar bear from three areas were at or above the LOAEL for immunosuppression in rhesus monkeys: Svalbard, East Greenland, and M'Clure Strait.

Highest  $\Sigma$ PCB levels were found in the population at Svalbard. The  $\Sigma$ PCB levels for different groups of Svalbard polar bears exceeded the NOAEL and LOAEL for offspring neurobehavioral effects as well as the NOEL for mink kit survival. Some individuals in all groups over three years of age exceeded the LOAEL for immunosuppression. Some individuals also exceeded the levels known to be correlated with poor reproductive success in harbour seals, as well as those correlated with poor reproductive success in ringed seal and the EC<sub>50</sub> for reduced litter size in mink.

Some polar bear from the Canadian and Svalbard populations had TEQ levels based on PCDD/F and/or nPCB levels that exceeded levels associated with immunosuppressive effects in harbour seal.

Concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDT, and TEQs in ringed seal blubber exceeded dietary NOAEC levels and a range of guidelines for protecting aquatic wildlife, indicating exposure to these substances at levels that could be expected to lead to effects.

The elevated levels of PCBs in adipose tissue of polar bears from the Svalbard area (ranging from 4790 to 80 300 ng/g) (Bernhoft *et al.*, 1997) reported in the previous POPs assessment, have prompted further biological effects studies linked to contaminant analyses.

#### Reproductive and developmental effects

Some adult female polar bears at Svalbard were equipped with satellite transmitters programmed to send information every six days for two to three years. Information on the location of the transmitter as well as sensor data on internal transmitter temperature and short- and long-term bear activity were recorded. Female polar bears normally have a three-year reproductive cycle (Ramsay and Stirling, 1988). Only pregnant bears den over winter. Reproductive rates were estimated from satellite data (Wiig, 1995; 1998).

From these studies of adult female polar bears at Svalbard, the reproductive rate was found to be approximately 0.75. This is similar to corresponding values found in other polar bear populations (Wiig, 1998). No difference in the PCB levels between females available for mating that became pregnant, and those that did not become pregnant, was found. The sample sizes for this comparison were small, however (Bernhoft *et al.*, 1997). Relatively low cub survival was found at Svalbard, and there are indications that the reproduction cycle was less than three years (Wiig *et al.*, 1992; Wiig, 1998). Thus, epizootological studies suggest that reproduction and cub survival in polar bears at Svalbard may be impaired. The high intake of PCBs at a crucial period could adversely influence the early development of cubs and lead to higher mortality. However, other factors such as population density may play a role, and a causal link with PCBs cannot be established.

POP concentrations were determined in adipose tissue, plasma, and milk samples from seven female polar bears and their cubs near Cape Churchill, Hudson Bay, between 1992-1996 (Norstrom, 1999b; Polischuk, 1999). Pregnant females were captured from August 7 to October 7, and the same females with cubs were captured from March 2 to March 17 of the following year, after emerging from dens but before they had moved onto the ice to begin hunting seals. Mothers that were recaptured a third time in the following autumn without cubs were found to have had high OC concentrations in their milk when they emerged from the den in the previous spring (Figure 4-55). By comparison, mothers recaptured in autumn and still accompanied by cubs, had low OC concentrations in their milk the previous spring. The differences in concentrations were significant ( $p < 0.05$ ) for all residue classes. For example, PCB concentrations were approximately three times higher (5780 ng/g lw) in females that lost their cubs than in females that kept their cubs (1830 ng/g lw). It is not known how much significance can be attached to this finding in terms of reproductive performance, but it is suggestive, at least, that cub survival may be dependent on degree of exposure to OCs in milk (Norstrom, 1999b).

The age structure of polar bears on Svalbard was compared to that of several other populations from less contaminated areas (Derocher *et al.*, 2003) as a low frequency of older female polar bears on Svalbard has been

reported previously (Wiig, 1998). The proportion of females with cubs-of-the-year that were older than 16 years old was found to be significantly lower on Svalbard (12.7%) when compared to western Hudson Bay (40.3%).

A significant negative relationship was found between  $\Sigma$ PCBs and testosterone, as well as total pesticides and testosterone in polar bear plasma, after correcting for possible confounders such as age and condition, indicating that PCBs and/or pesticides may decrease circulating testosterone levels in male polar bears (Oskam *et al.*, 2001) (Figure 6-7). Testosterone is the major androgenic steroid hormone playing a crucial role in male sexual development.

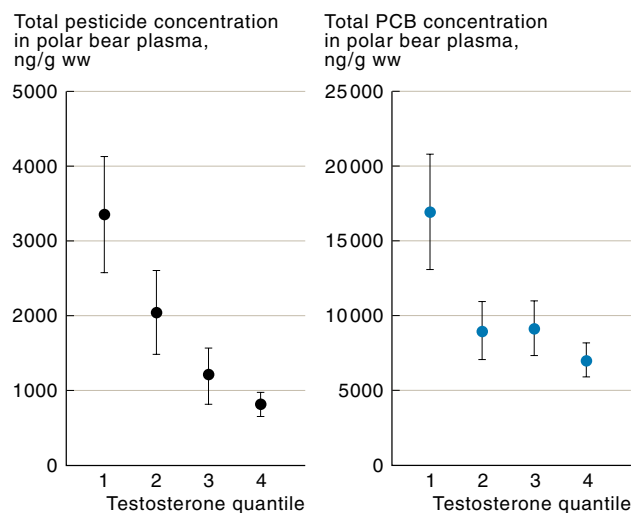


Figure 6-7. Total pesticide and PCB concentrations (mean and 95% C.I.) by testosterone quantile (Oskam *et al.*, 2001).

In 1999, 52 experienced polar bear hunters living in East Greenland were interviewed about their observations of aberrant bears including pathological changes/anomalies in internal organs (Dietz *et al.*, 2001; Sandell *et al.*, 2001). Information on approximately 1110 bears that had been shot between 1945 and 1999 was obtained. Thirteen anomalous polar bears were reported. The anomalies included supernumerary nipples or claws, unilateral collapse of lung, abnormal and missing claws, partial melanism, missing limbs, and a malformed newborn.

A study of contaminant-induced changes in polar bear skulls from East Greenland was initiated in 1999. The results are still very preliminary as both the POP analyses and the age determination of the bears are still in progress. Approximately 180 skulls (1892-1987) from the National Zoological Museum in Copenhagen were compared with 100 recent (1999-2001) skulls to detect possible macroscopical pathological changes, asymmetrical changes as well as osteopenia (osteoporosis and osteomalacia). Both directional and fluctuating asymmetry were found in several bones of the skulls. Surprisingly, the amount of fluctuating asymmetry seemed to have decreased from the period of 1892-1960 to the period 1960-2001. The results are only preliminary, however, and awaiting the inclusion of age data in the analysis.

The frequency of paradontitis (loose teeth) among subadult polar bears was higher, but not statistically significant, in the period of 1960-2001 than in the period



1892-1960. A clear sex difference was observed in the bone mineral density measured as calcium-phosphate content. There was also a tendency for the males from the period of 1960-2001 to have lower bone mineral density as compared to males from the period of 1900-1960.

Several female polar bears from Svalbard have been found to be pseudohermaphrodites, and high PCB levels have been hypothesized as one possible causative factor (Wiig *et al.*, 1998). The link to POP exposure; however, is not strong because of the occurrence of the same syndrome in black bears (Norstrom, 2002). One pseudohermaphroditic female was also found among more than 100 sampled polar bears in 1999 on East Greenland (Sonne-Hansen *et al.*, 2002) and the occurrence of this one case is lower than the estimated frequency found at Svalbard of approximately 3% (Derocher, 2002).

#### Cytochrome P450 activities

CYP1A1 has been determined in white blood cells of polar bears and the results ( $n=13$ ) show a significant positive correlation between  $\Sigma$ PCBs (the sum of PCBs 99, 118, 153, 156, 180, and 194) and CYP1A1 area in western blots ( $p=0.026$ ). The strongest correlation was found with CB156 ( $p=0.011$ ). These results are promising with regard to the potential for CYP1A1 in blood cells as a biomarker for PCB exposure in polar bears (Skaare *et al.*, 2000).

#### Thyroid and retinol effects

Normal regulation of vitamin A and thyroid hormones is important for a wide range of biological functions, such as growth, cell differentiation, reproduction, behavior, and the immune system. In Svalbard polar bears, retinol and thyroid hormones (T3 and T4), and concentrations of several OC contaminants have been determined in blood plasma of 71 individuals collected from 1991 to 1994. The determination of multivariate associations between retinol, thyroid hormones, and the ratio of total and free thyroid hormones, respectively, and the concentrations of various OC components (PCBs, DDE, HCB and HCHs) revealed significant OC associations for retinol and the ratio of total T4:free T4, after correcting for age and sex (Skaare *et al.*, 2001a). Significant negative correlations were found between retinol and  $\Sigma$ PCBs (Figure 6:8), as well as retinol and HCB and HCHs. Significant negative correlations were also found between total T4:free T4 and  $\Sigma$ PCBs (Figure 6:9) as well as HCB.

Concentrations of the thyroid hormones, T3 and T4, and retinol were determined in plasma of polar bears from Resolute Bay in Canada and Svalbard, which have among the lowest and highest POP concentrations, respectively, in polar bears (Norstrom, 2000). Free T3 (FT3) and free T4 (FT4) indices were also determined. Resolute bears had significantly higher total T4 and FT4 index, and lower total T3 and FT3 index than Svalbard bears ( $p<0.001$ ). Retinol concentrations were not significantly different between regions. None of the biological measures were significantly related to age, even when separated into region and sex categories. This is in contrast to the above study, which found that total T4, FT4, total T3, and FT3 are associated with age in male polar bears (Skaare *et al.*, 2001a). Total T3 was higher

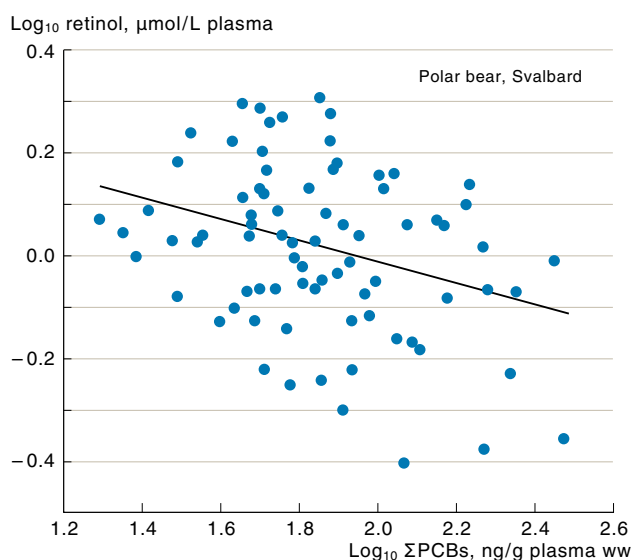


Figure 6-8. The association between retinol (corrected for age and sex) and  $\Sigma$ PCBs in plasma of 79 polar bears at Svalbard by regression analysis. The regression line is shown ( $\log$  residual retinol =  $0.40 - 0.21 \log \Sigma$ PCBs). The Pearson correlation coefficient is  $r = -0.33$  ( $p = 0.003$ ) (Skaare *et al.*, 2001a).

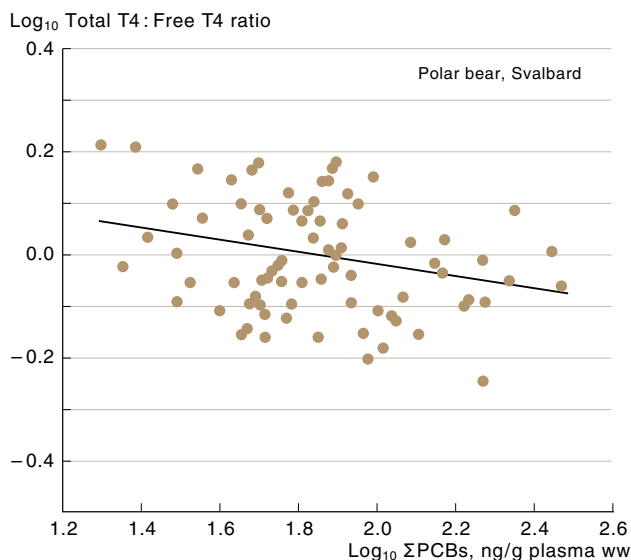


Figure 6-9. The association between the ratio of total T4 and free T4 (TT4:FT4) (corrected for age and sex) and  $\Sigma$ PCBs in plasma of 78 polar bears at Svalbard by regression analysis. The regression line is shown ( $\log$  residual total T4:free T4 =  $0.21 - 0.12 \log \Sigma$ PCBs). The Pearson correlation coefficient is  $r = -0.28$  ( $p = 0.013$ ) (Skaare *et al.*, 2001a).

in females from the Svalbard population, and FT3 index was lowest in Resolute females.

Correlations among thyroid hormone concentrations, FT3 and FT4 indices, retinol concentrations and the complete suite of POP concentrations in adult polar bear plasma ( $n=60$ ) from Resolute Bay, Canada, and Svalbard were examined by principal component analysis. Because hydroxy compounds may affect circulating levels of free and bound T4 due to competitive binding of hydroxy metabolites and T4 to TTR, all chemical concentrations were converted from mass to molar concentrations prior to statistical analysis.

Using the entire data set ( $n=60$ ), retinol concentrations were negatively correlated ( $r=-0.465$ ,  $p<0.001$ ) with persistent PCBs (PC1) and positively correlated



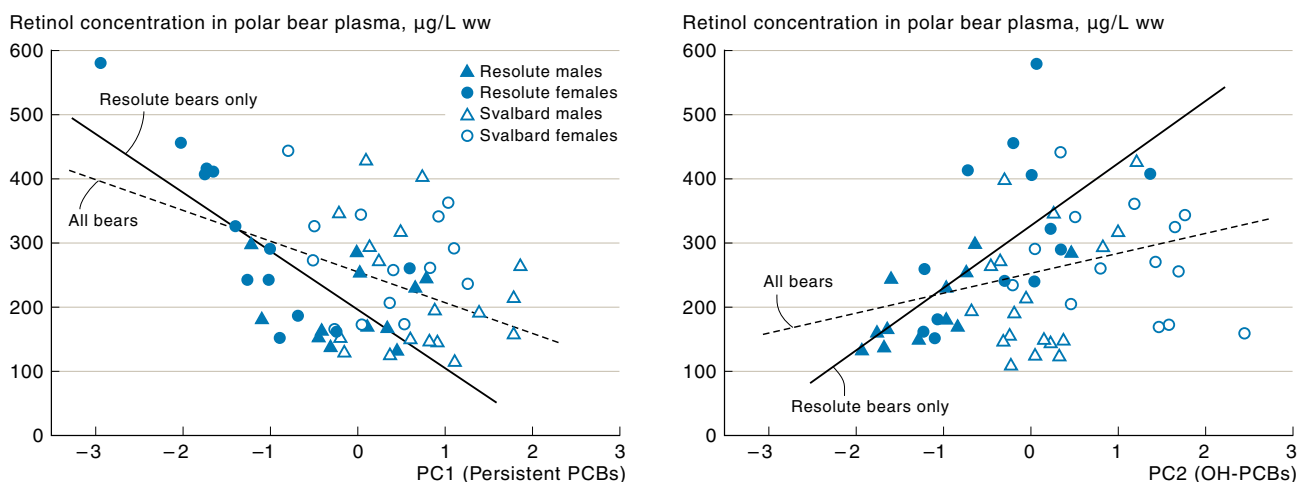


Figure 6-10. Correlation between retinol concentrations ( $\mu\text{g/L}$ ) and the first two principal components from analysis of POP concentrations in polar bear plasma from Resolute Bay in the Canadian Arctic in April-May 1997, and from Svalbard in April-May 1998.

( $r = 0.309$ ,  $p = 0.02$ ) with OH-PCBs (PC2) (Figure 6-10). If only Resolute bears ( $n = 25$ ) were included in the analysis, retinol was more highly correlated with PC1 ( $r = -0.744$ ,  $p < 0.001$ ) and PC2 ( $r = 0.692$ ,  $p < 0.001$ ). These results suggest that plasma retinol concentrations are more likely to be affected by the influence of persistent PCBs on retinol metabolism and storage in liver than by the interference of OH-PCBs with the transport of retinol via RBP:TTR dimer formation.

Total T4 plasma concentrations were negatively associated with both persistent PCBs (PC1),  $r = -0.337$ ,  $p = 0.01$ ,  $n = 56$ ) and non-persistent PCBs (PC3) ( $r = -0.293$ ,  $p = 0.03$ ,  $n = 56$ ), but not with any other contaminant group, including OH-PCBs. Since both PC1 and PC3 are PCB-related, this correlation suggests a common mechanism of action of all PCB congeners in reduction of plasma T4 concentrations. Total T4 concentrations were also negatively correlated with concentrations of  $\Sigma\text{PCBs}$  ( $r = -0.29$ ,  $p = 0.04$ ,  $n = 56$ ). Thirty-three female polar bears without cubs and with two-year old cubs were sampled at Svalbard and in the Barents Sea region during the period 1995-1998, and blood samples were analyzed for a range of POPs, thyroid hormones, and progesterone (Braathen *et al.*, 2000). Significant differences were seen in progesterone levels between the females from the two areas, with higher progesterone levels in the Barents Sea bears. However, no relationship could be shown between progesterone levels and the  $\Sigma\text{PCB}$  concentrations. For thyroid hormones, significant differences were found for the ratio of total T3: free T3 with higher ratios in the Svalbard bears, but no relationship could be seen between these and  $\Sigma\text{PCB}$  levels.

#### Immune effects

Production of antibodies plays an important role in protection against infections. Antibodies are divided into different immunoglobulin classes, where IgG is the major one in blood. IgG and OC concentrations were determined in blood sampled from 56 free-living polar bears of different ages and both sexes between 1991 and 1994 (Bernhoft *et al.*, 2000). Total IgG concentration increased with age and was significantly higher in males than in females. A significant decrease of IgG with increased  $\Sigma\text{PCB}$  level was found ( $r = -0.29$ ,  $p = 0.03$ ). IgG was standardized for sex and age, since both sex and age

may influence the levels. Three individual PCB congeners showed significant inverse correlations to IgG: CBs 99, 194 and 206 (Figure 6-11). In addition, HCB was also inversely correlated to IgG. OCs were found to account for 11% of the variation of IgG levels.

In a study of mothers and cubs on Svalbard sampled in 1995-1998, a negative correlation was found between IgG levels in cub plasma and plasma  $\Sigma\text{PCB}$  concentrations (Lie *et al.*, 2002). The results of these studies demonstrate a possible contaminant-associated suppression of antibody-mediated immunity in polar bears at Svalbard.

The effects of high PCB exposure on the immune system have been further studied by comparing the immune system functions in polar bears with high (Svalbard) and low (Canada) PCB exposure (Skaare *et al.*, 2001b; 2002; Larsen *et al.*, 2002c). This field study comprised a vaccination model with recapture after immunization. Thirty bears at Svalbard and thirty in Canada were immunized with different herpes-, reo-, and influenza viruses and tetanus toxoid to stimulate the production of protective antibodies such as virus-neutralizing antibodies, virus hemagglutination inhibition antibodies and toxin-neutralizing antibodies. The immunization also included keyhole limpet hemocyanin that, together with tetanus toxoid, would stimulate cell-mediated immune response. Blood was sampled at immunization, and four to six

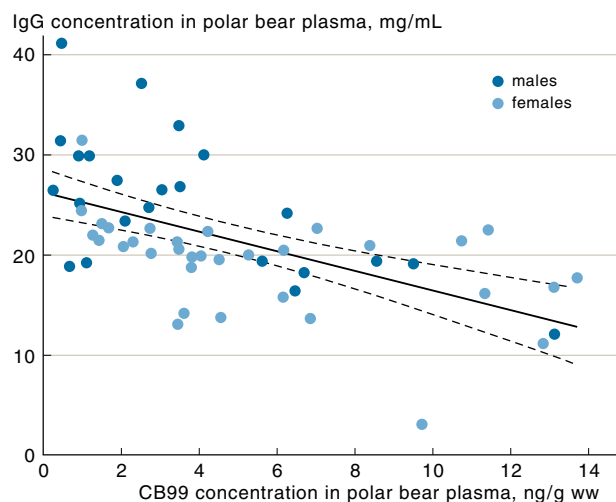


Figure 6-11. Linear regression of IgG level and CB99 in blood plasma of polar bears of both sexes at Svalbard (Bernhoft *et al.*, 2000).

weeks later, for detection of antibodies. In addition, *in vitro* lymphocyte stimulation was performed with mitogens and specific antigens (phytohemagglutinin, concanavalin A, pokeweed mitogen, *Mycobacterium* antigen, and lipopolysaccharide) (Larsen *et al.*, 2002a; 2002b). The resistance factor was measured directly as neutralization of virus infections in cell cultures, inhibition of virus hemagglutination, and toxin neutralization. Therefore, the effect of PCBs on infection resistance was measured without dependence on infection experiments or by conducting registration of disease outbreaks.

Preliminary results from this study demonstrated significantly higher PCB plasma levels in polar bears at Svalbard as compared to Canada. In the Canadian bears, significantly higher antibody titers against influenza, reo- and herpes viruses were found following immunization than at Svalbard. Furthermore, significantly lower antibody titers against influenza virus following immunization, were associated with higher PCB levels. Significantly lower lymphocyte responses were found to pokeweed mitogen, lipopolysaccharide from *E. coli* and *Mycobacterium* antigen with high PCB exposure levels, and significant negative correlations between PCBs and IgG were also found.

From the present preliminary results on effects of PCBs on the immune system of polar bears, it is reasonable to assume that PCBs are associated with decreased resistance to infections. This is supported by the finding that polar bears with high plasma PCB have higher incidence of *Pasteurella* bacteria, one of the most common microbes in the environment (Larsen, 2002).

#### Levels and intake assessment

$\Sigma$ PCB concentrations (22-40 congeners) in polar bear fat collected between 1996 and 1997 at several sites around Alaska (Beaufort Sea, Bering Sea, Chukchi Sea) ranged between 910 and 11 000 ng/g lw with means in the range of 2600-9640 ng/g lw (Annex Table 14). Mean  $\Sigma$ PCB concentrations in polar bear fat collected in 1999-2000 from East Greenland ranged from 4800 to 7700 ng/g lw. In polar bear fat from one individual from the pack ice near Iceland and collected in 1993,  $\Sigma$ PCB concentration (7 congeners) was 7860 ng/g ww (approximately 10 000 ng/g lw) (Annex Table 14).  $\Sigma$ PCB concentrations (1990) from a recent temporal-trend study of Canadian polar bear from several sites ranged from 720 to 20 200 ng/g lw with geometric means in the range of 2600-7500 ng/g lw (Annex Table 14). Russian polar bears collected from four areas (Franz Josef Land, Kara Sea, Siberian Sea, Chukchi Sea) have plasma  $\Sigma$ PCB concentrations (5 congeners) ranging from 1100 to 31 000 ng/g lw, and Svalbard polar bears have plasma  $\Sigma$ PCB concentrations ranging from 2100-14 000 ng/g lw in the same study (Andersen *et al.*, 2001b). For Svalbard polar bears of different ages collected in 1995-1998, plasma  $\Sigma$ PCB levels range from 1060 to 29 000 ng/g lw, with highest concentrations found in cubs-of-the-year and yearlings (Annex Table 14).

Most  $\Sigma$ PCB levels in these polar bears exceed the NOAELs and LOAELs found for subtle neurobehavioral effects in offspring if polar bears are as sensitive as offspring of rhesus monkeys and humans, and also, the NOEL for vitamin A reduction in otter (Figure 6-3). The  $\Sigma$ PCB levels in polar bears from several sites in Alaska and Canada, on the Iceland pack ice, at Svalbard and

three sites in Russia (Franz Josef Land, Kara Sea, Siberian Sea) overlap the NOEL for kit survival in mink, and several of these also exceed the LOEL for vitamin A reduction in otter (Figure 6-3). The threshold associated with immunosuppression and vitamin A disruption in harbour seal blubber is 16 500 ng  $\Sigma$ PCB/g lw, and the LOAEL for immunosuppression is 21 000 ng  $\Sigma$ PCBs/g lw (blood) in rhesus monkeys.  $\Sigma$ PCB levels in polar bears from three areas are at or above these two thresholds: Franz Josef Land, Kara Sea, and Svalbard (cubs and yearlings). Since Svalbard polar bears with high  $\Sigma$ PCB levels have been shown to exhibit signs of immunosuppression, it is probable that such effects are also occurring in Russian polar bears from these sites.  $\Sigma$ PCB levels in some polar bears from Davis Strait, Canada are above the threshold for immunosuppression in harbour seals and close to the LOAEL for immunosuppression in rhesus monkeys. Temporal-trend studies for  $\Sigma$ PCBs in Canadian and Svalbard polar bears indicate that there have been no declines during the 1990s. This implies that the assessment of contaminant levels compared to thresholds for other polar bear populations, given in the previous AMAP assessment, is still valid.

Levels of 2,3,7,8-TeCDD and 1,2,3,7,8-PeCDD in Canadian polar bears from four sites sampled in 1990 range from 1.2 to 3.8 pg TEQ/g lw in adipose tissue (Annex Table 16). These are well below the threshold for immunosuppression in harbour seals (Figure 6-4). This may, however, be an underestimation since mono-ortho PCBs usually contribute most to the total TEQs compared to nPCBs and PCDD/Fs (Letcher *et al.*, 1996) and these were not included in the study.

PFOS levels in polar bears range from 26 to 52 ng/g ml in plasma in Beaufort Sea individuals and 180-680 ng/g ww in liver from bears from Barrow, Alaska (Kannan *et al.*, 2001a; Giesy and Kannan, 2001). These levels are well below the NOAEL (15000 ng/g ww in liver) and LOAEL (58000 ng/g ww in liver) for second-generation effects in rats.

Based on mean POP levels in ringed seal blubber from various sites in the Arctic (Annex Table 12), dieldrin, chlordanes (heptachlor epoxide), and  $\Sigma$ DDT levels are below the dietary NOAECs found for reproductive effects in mink. Some harp seals from east of Svalbard have chlordane levels that exceed the LOAEC found for kit growth in mink. Based on the NOAEC (4000 ng/g ww) for rats and dogs, toxaphene levels in ringed and harp seals are below those associated with thyroid effects. Assuming that polar bears are as sensitive as mink, mean levels of  $\Sigma$ PCBs in ringed and harp seal blubber from all sites exceed the dietary NOAEC for reproduction of 72 ng/g ww. Mean  $\Sigma$ DDT levels in ringed and harp seals exceed Canadian and U.S. EPA guidelines for protecting aquatic wildlife, and in some cases (i.e. Svalbard, Russia), International Joint Commission objectives are also exceeded (Table 6.1) (Figure 6-6). For  $\Sigma$ PCB levels, ringed and harp seal blubber from all sites exceed all environmental guidelines for protecting aquatic wildlife (Figure 6-5). Toxaphene levels in ringed and harp seal blubber, where measured, exceed Canadian guidelines for protecting wildlife consumers. Data for dioxin-like compounds are very limited but TEQs in ringed seal blubber from Canada and Russia exceed Canadian (mammalian) and U.S. EPA guidelines (Annex Table 16).

### 6.3.7. Arctic fox

In the previous AMAP assessment, mean  $\Sigma$ PCB levels in Svalbard Arctic fox from 1993 and 1994 were found to exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL for kit survival in mink, levels known to cause poor reproductive success in harbour seals, and the  $EC_{50}$  for reduced litter size in mink. Some individuals also exceeded the levels associated with poor reproductive success in ringed seals and the  $EC_{50}$  for kit survival in mink (80 000-120 000 ng/g lw). The mean  $\Sigma$ PCB levels in Arctic fox were also above the LOAEL for immunosuppression in rhesus monkeys. No effects studies have been carried out since the previous assessment.

#### *Levels and intake assessment*

Mean concentrations of  $\Sigma$ PCBs in Arctic fox collected in 1999-2000 from Holman Island, NWT, were 860 ng/g lw in muscle (range: 76-8050 ng/g lw) and 1350 ng/g lw in liver (range: 110-14 600 ng/g lw). Mean  $\Sigma$ PCB concentrations in Arctic fox from Barrow, Alaska and from inland Iceland were 1000-1600 ng/g lw in liver. Mean concentrations of  $\Sigma$ PCBs in Arctic fox fat collected in 1996-1997 in the Pribilof Islands, Alaska, were somewhat higher, between 2100 ng/g lw and 5000 ng/g lw (range: 240-12 000 ng/g lw). Highest mean  $\Sigma$ PCB concentrations were seen in Arctic fox from the coast of Iceland at 72 500 ng/g lw (Annex Table 14). The  $\Sigma$ PCB levels for Arctic fox from all sites exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans. The  $\Sigma$ PCB levels in Canadian, Pribilof Islands, and coastal Iceland foxes also exceed the NOELs for vitamin A reduction in otter, effects on otter reproduction and mink kit survival, the LOAELs for vitamin A reduction in otter, and decreased kit production and kit body weight gain in mink (Figure 6-3). The  $\Sigma$ PCB levels in the coastal Iceland foxes also exceed the threshold for immunosuppression and vitamin A disruption in harbour seals, the LOAEL for immune effects in rhesus monkeys, the threshold for poor reproduction in harbour seals, and the  $EC_{50}$  for litter size in mink. Levels of mono-*ortho* PCBs given as TEQs in Pribilof Islands Arctic foxes ranged between 9 and 290 pg/g lw (means: 48 and 151 ng/g lw for females and males, respectively) and these exceed the mono-*ortho* TEQs associated with immunosuppressive effects in harbour seals (Figure 6-4).

Besides feeding on terrestrial mammals and birds, Arctic foxes may also eat marine birds and eggs, seal pups, and placentas, as well as scavenge on seals, mostly from polar bear kills, depending on what is available at the particular site they are located. Based on contaminant levels found in eggs and liver from thick-billed murre, black guillemots, black-legged kittiwakes, glaucous gulls, herring gulls, great black-backed gulls, and fulmars from the Alaskan, Canadian, Greenlandic, Norwegian, and western Russian Arctic (Annex Table 11), dieldrin,  $\Sigma$ DDT, and heptachlor levels are below the dietary NOECs and LOECs for reproductive effects in mink.  $\Sigma$ PCB levels are above the dietary NOEC for reproductive effects and TEQs, based on PCDD/Fs, nPCBs, and/or mono-*ortho* PCBs are above the NOECs and LOAECs for effects on reproduction and vitamin A in mink. Dietary toxaphene levels are below those asso-

ciated with thyroid effects. Mean dieldrin levels in these seabird species' eggs also do not exceed any environmental guidelines for protecting aquatic wildlife. However,  $\Sigma$ DDT levels in eggs and liver from many of these species from many of these sites exceed Canadian and U.S. EPA guideline levels for protecting wildlife that consume aquatic biota (Figure 6-6). Most  $\Sigma$ PCB levels exceed all guideline levels (Figure 6-5) and where measured, toxaphene levels exceed Canadian guidelines. PCDD/F and nPCB levels given as TEQs exceed Canadian (mammalian) and U.S. EPA guideline values (Table 6.1) in kittiwake, fulmar, murre, black guillemot, common guillemot, glaucous gull, herring gull, and great black-backed gull eggs and/or liver (Annex Table 16). Thus, Arctic fox that prey on seabird eggs, chicks, and adults may have dietary intakes of TEQs,  $\Sigma$ DDTs,  $\Sigma$ PCBs, and toxaphene high enough to lead to effects. The dietary assessment for Arctic fox that feed on seals is the same as for polar bears (Section 6.3.6).

### 6.3.8. Sea otter

No biological effects studies have been carried out in sea otter.

#### *Levels and intake assessment*

Mean concentrations of  $\Sigma$ PCBs in sea otter from southeast Alaska were 270 ng/g lw and for sea otter from the Aleutian Islands, 10 300 ng/g lw (Annex Table 14). The  $\Sigma$ PCB levels in sea otter from southeast Alaska do not exceed any threshold levels. The  $\Sigma$ PCB levels in sea otter from the Aleutian Islands exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOELs for vitamin A reduction in otter, and effects on otter reproduction and mink kit survival (Figure 6-3).

## 6.4. Summary and conclusions – biological effects

It is very difficult to link contaminant levels or biochemical indicators of effects to effects on Arctic animals at the individual or population level. Such assessments are also complicated by the fact that the thresholds for effects of many contaminants are not well known, and very little is known about effects of contaminant mixtures. It is also possible that there may be other causes of the effects seen that are unrelated to POP exposure.

As far as OCs are concerned, Arctic marine mammals are often regarded as controls for much more contaminated members of their populations or related species in temperate regions. Biological effects studies on Arctic animals do, however, show some subtle responses, and there are now stronger links between these effects and current levels of some POP contaminants. Based on the results of the biological effects studies that have been carried out, the following conclusions can be drawn.

### 6.4.1. Observed effects

#### 6.4.1.1. Field studies

##### *Reproduction*

- Alaskan peregrine falcons (*tundrius* and *anatum* subspecies) still suffer from eggshell thinning of 10.6%



and 12%, respectively, when compared to eggs from the pre-DDT era.

- Bald eagles showed reduced productivity on Adak Island in the Aleutians, which was associated with the higher DDE concentrations found there than at three other Aleutian islands. Eggshell thickness was significantly negatively correlated with PCB concentrations on the four islands.
- Positive correlations were found between TBT concentrations and degree of imposex in dogwhelks from the Norwegian coast, and imposex is also found along the Faroe Islands and Greenlandic coasts. Imposex has also been seen in dogwhelk from Icelandic harbors but has decreased after restrictions on the use of TBT as an anti-fouling paint on boat hulls were implemented in 1990.
- In glaucous gulls from Bjørnøya, Norway, increased absence from nests was correlated with high OC levels. Female gulls with high OC levels were more likely to have non-viable eggs and chick body condition was poorer. Adult survival was significantly negatively correlated to OC levels. Glaucous gulls with high OC levels have wing feather asymmetry, an indication of developmental stress.
- Egg volume, yolk mass, and hatchling mass in one-day-old shag were negatively correlated to PCB concentrations.
- A significant negative correlation was found between testosterone levels and PCB levels, as well as total pesticide levels in polar bear plasma, indicating that these POPs may decrease circulating testosterone levels. Females from southwest Hudson Bay that had lost their cubs between emerging from the den and the following summer/autumn had significantly higher OC concentrations when emerging from the den than females that kept their cubs.

Other observations have been made where POPs are suspected of playing a role, but where there is as yet no certain link between the effects seen and POP concentrations. These are the following.

- Black-legged kittiwakes with crossed bills and clump feet have been observed during an expedition in the Barents Sea.
- Steller sea lion populations have been declining in western Alaska including the Aleutian Islands and the western stock is considered endangered. These populations excrete higher concentrations of POPs than less-affected populations with lower POP concentrations.
- Reproductive rates in Svalbard polar bears are similar to those of other polar bear populations, but cub survival is lower. Female pseudohermaphroditic polar bears have been found on Svalbard and Greenland. The proportion of females with cubs-of-the-year that were older than 16 years was significantly lower on Svalbard than in western Hudson Bay.

#### *Cytochrome P450 activity*

- CYP enzyme activities are correlated to PCB levels in Arctic char from two freshwater lakes (one with high POP levels, one with low POP levels) on Bjørnøya.
- A weak association between PCB levels and EROD activity was seen in glaucous gulls at Bjørnøya.
- A positive correlation was found in black guillemot from Saglek Bay, Canada, between PCB concentrations

and EROD activity up to a threshold of 100 ng/g ww in liver. After this, EROD activity leveled off.

- High correlations have been found between testosterone 6- $\beta$  hydroxylation (CYP3A) and toxaphene levels in harp seals around Svalbard. Positive correlations have been found between EROD levels and  $\Sigma$ PCBs in Svalbard ringed seals.
- A significant correlation was found between CYP1A1 in polar bear white blood cells and PCB levels.

#### *Thyroid and retinol effects*

- A borderline significant correlation was found between PCB concentrations in yolk and plasma retinol levels in one-day-old shag from Norway.
- A significant negative correlation was found between liver PCB concentrations and liver retinol and retinyl palmitate levels in black guillemots at Saglek Bay.
- A significant negative correlation was seen between HCB, *p,p'*-DDE, and  $\Sigma$ PCB blood concentrations and plasma T4 levels in male glaucous gulls from Bjørnøya.
- Retinol levels and total T4 levels in northern fur seal pups were found to be negatively correlated to PCB congeners and to TEQs.
- In Svalbard polar bears, significant negative correlations were found between retinol and PCB levels, as well as retinol and HCB and HCHs. Significant negative correlations were also found between the ratio of total T4:free T4 and PCB as well as HCB. In a comparison of polar bears from Svalbard (high PCB levels) and Resolute, Canada (low PCB levels), a significant negative correlation was found between total T4 levels and  $\Sigma$ PCBs but not with OH-PCBs. Retinol levels were negatively correlated with persistent PCBs and positively correlated with OH-PCBs, suggesting that PCB affects retinol metabolism and storage in the liver.

#### *Immune effects*

- In glaucous gulls from Bjørnøya, nematode density was positively correlated with concentrations of *p,p'*-DDT, mirex, and  $\Sigma$ PCBs.
- In northern fur seal and Steller sea lion pups, various measures of normal immune function were negatively correlated to PCB levels, indicating that high PCB exposure may be causing immunodysfunction.
- In Svalbard polar bears, a significant decrease in antibodies (IgG) with increased PCB levels was found. In mothers and cubs, a similar negative correlation was found for IgG levels in cubs and plasma PCB levels as well as with a number of specific congeners (CBs 99, 137, 153, 157, 170, 180, and 194). In a vaccination study using two polar bear populations, one with higher PCB levels (Svalbard) and one with lower PCB levels (Canada), polar bears with high PCB levels were found to exhibit immunosuppression expressed as reduced IgG production and lowered lymphocyte responses. This may indicate decreased resistance to infections.

#### 6.4.1.2. Laboratory studies using Arctic species

##### *Cytochrome P450 activity*

- Increased EROD activity was found in Arctic char fed a single oral dose of Aroclor 1260 and then starved, compared to PCB-exposed and fed char, non-exposed and fed char, and non-exposed and starved char.



*Immune effects*

- Increased cortisol levels were found in Arctic char fed a single oral dose of Aroclor 1260 and then given food, compared to PCB-exposed and starved char, non-exposed and fed char, and non-exposed and starved char. In another study, basal cortisol levels were suppressed by PCBs in starved fish but were elevated in fed fish after handling, indicating that stress responses are compromised by PCBs, and the effect of fasting makes char sensitive to the effects of PCBs. Fasting and PCB exposure were also studied in relation to disease susceptibility. Disease susceptibility was highest in the fed char with no difference due to PCB exposure, while disease susceptibility increased with PCB exposure in the starved group. The results indicate that PCBs reduce immunocompetence in starved Arctic char, but that starved fish are more disease resistant than fed fish.
- Glaucous gull chicks with high dietary PCB exposure show an impaired ability to produce antibodies when challenged with an antigen.
- Two juvenile harp seals treated with increasing doses of selected PCB congeners for 40 days and then fasted for 30 days had increased serum cortisol and aldosterone levels as well as tumor necrosis factor alpha compared to the controls.

*Mutagenic effects*

- Higher frequencies of chromosome aberrations and DNA adducts were found in glaucous gull chicks fed a POPs contaminated diet.

**6.4.2. Assessment of current levels in biota**

Current concentrations of some POPs in several Arctic species are at or above the known thresholds associated with effects that have been seen in other species studied either in the laboratory or in the field.

Canadian wolverines have  $\Sigma$ PCB levels that exceed those associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans. Mountain hare and reindeer from the Kola Peninsula (Lovozero) have PCDD/F concentrations, expressed as TEQs, which exceed levels associated with immunosuppression in harbour seals. TEQs in reindeer from Pechora and Taymir Dudinka also exceed this threshold.

Eggshell thinning in Alaskan peregrine falcon eggs has improved, but eggshells are still thinner than pre-DDT era eggs, indicating that present DDE levels are still causing effects. The *p,p'*-DDE concentrations in some Alaskan peregrine falcon eggs are just below the critical threshold for reproductive failure.  $\Sigma$ PCB levels in Alaskan peregrines exceed some NOELs and LOELs for reproductive endpoints in a wide range of wild bird species.  $\Sigma$ PCB concentrations in Norwegian peregrine falcons and white-tailed sea eagle exceed most or all thresholds for reproductive effects in other bird species. Norwegian gyrfalcon and golden eagles have lower  $\Sigma$ PCB concentrations, but these still exceed some thresholds for reproductive effects. Bald eagles from one of four studied Aleutian Islands have *p,p'*-DDE concentrations in the range known to cause reproductive impairment. Maximum  $\Sigma$ PCB concentrations in some Aleutian bald eagles exceed most of the thresholds for reproductive effects in other bird species including the LOAEL for egg mortality in bald eagles.

$\Sigma$ PCB levels in Alaskan and Canadian burbot liver from some sites and in some Arctic char from Bjørnøya are close to or exceed the LOEL for induction of liver enzymes found in Arctic char. Toxaphene levels in some Canadian burbot exceed levels associated with effects on bone development found in channel catfish. In marine and anadromous fish,  $\Sigma$ PCB levels in Greenland shark from Cumberland Sound and Greenland halibut from West Greenland exceed the LOEL for induction of liver enzymes found in Arctic char.

Piscivorous seabirds such as grey herons, alcids, and kittiwakes have lower  $\Sigma$ PCB levels than predatory seabirds, and only grey herons had  $\Sigma$ PCB levels that exceeded some reproductive thresholds for effects. Black guillemots from the most contaminated site at Saglek Bay, Canada and predatory seabirds, such as glaucous and great black-backed gulls and great skuas, have  $\Sigma$ PCB levels in liver that indicate that egg levels might exceed some threshold levels for reproductive effects. For grey heron, mono-*ortho* PCB levels, expressed as TEQs, exceeded some reproductive thresholds in other bird species. TEQs based on PCDD/F and nPCB levels for Canadian thick-billed murres, black-legged kittiwakes, and northern fulmars, and TEQs based on PCDD/Fs, nPCBs and mono-*ortho* PCBs for herring gull and great black-backed gull eggs from northern Norway exceed the LOAEL for reproductive effects in wood duck, a sensitive species. Glaucous gulls from Svalbard have nPCB and mono-*ortho* PCB levels expressed as TEQs that exceed all thresholds for reproductive effects in other bird species and the threshold for induction of liver enzymes found in common terns.

Ringed seals from Alaska, Canada, and Greenland, harbour seals from Alaska, Steller sea lions from Alaskan eastern stocks, harbour porpoises and walrus from Greenland, and grey whales from the Bering Sea, have  $\Sigma$ PCB levels that are low but that still exceed levels associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans. Higher  $\Sigma$ PCB levels are found in ringed seal from the Barents Sea/Kara Sea area and Saglek Bay, Labrador, harbour, grey, bearded and northern fur seals and some Steller sea lions. These  $\Sigma$ PCB levels also exceed the NOEL associated with decreased vitamin A in otter. Svalbard, Kara Sea, and Saglek Bay ringed seals and northern fur seals also exceed the threshold for decreased reproduction in otter, and sea otter from the Aleutian Islands. Some ringed seal (Kara Sea) and some beluga exceed the NOEL for mink kit survival. Harbour porpoises from Norway, long-finned pilot whales, narwhal, killer whales (residents and transients), harp seals, and some minke whales have even higher  $\Sigma$ PCB levels that are associated with decreased vitamin A in otter, and decreased otter and mink reproduction. Harp seals and some minke whales exceed  $\Sigma$ PCB levels associated with immunosuppression and vitamin A disruption in harbour seals but are below the threshold for immunosuppression in rhesus monkeys. In addition, some harbour porpoises from northern Norway, some resident killer whales and all transient killer whales from Alaska, and some long-finned pilot whales from the Faroe Islands have  $\Sigma$ PCB levels that exceed thresholds for immunosuppression in rhesus monkeys and poor reproductive success in harbour seal. Some resident and many transient killer whales exceed the EC<sub>50</sub>

for litter size in mink, and many transient killer whales also exceed the threshold for poor reproductive success in ringed seal and the  $EC_{50}$  for kit survival in mink.

Levels of mono-*ortho* PCBs, given as TEQs, in Arctic fox from the Pribilof Islands, and in resident and transient killer whales, exceed the mono-*ortho* TEQs associated with immunosuppressive effects in harbour seals.

Current  $\Sigma$ PCB levels in polar bear from Alaska, Canada, Greenland, the Iceland ice pack, Svalbard, and Russia indicate that these populations have levels that exceed those associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans, decreased vitamin A in otter, and NOELs for otter and mink reproduction.  $\Sigma$ PCB levels are highest in the Svalbard and Russian populations, and these levels exceed those associated with immunosuppression in harbour seals and rhesus monkeys and poor reproductive success in harbour seals. The results from biological marker studies, particularly immune system effects in Svalbard polar bears and observations of decreased cub survival, support this assessment.

Arctic fox from Barrow, Alaska, and inland Iceland have  $\Sigma$ PCB levels associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans. Arctic fox from the Pribilof Islands, Alaska, Canada, and coastal Iceland have  $\Sigma$ PCB levels associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans, decreased vitamin A in otter and reproductive effects in otter and mink.  $\Sigma$ PCB levels in Arctic fox from coastal Iceland also exceed those associated with immunosuppression in harbour seals and rhesus monkeys, poor reproductive success in harbour seals and the  $EC_{50}$  for litter size in mink. Levels of mono-*ortho* PCBs, given as TEQs, in Pribilof Islands Arctic foxes exceed the mono-*ortho* TEQs associated with immunosuppressive effects in harbour seals.

Consequently, based on known thresholds for effects, several Arctic species appear to be at risk for, primarily, reproductive and/or immunosuppressive effects from current levels of  $\Sigma$ DDTs,  $\Sigma$ PCBs, and/or dioxin-like substances. Those at greatest risk include peregrine falcons, bald eagles, white-tailed sea eagles, glaucous and great black-backed gulls, great skuas, some alcids, harbour porpoises, seals, northern fur seals, Steller sea lions, belugas, long-finned pilot whales, narwhal, minke whales, killer whales, sea otters, polar bears, and Arctic fox. If the risk for subtle neurobehavioral effects from  $\Sigma$ PCBs in exposed offspring of mammals is included, then some reindeer and mountain hares, wolverines, walrus, and grey whales are also potentially at risk. Burbot from some sites have toxaphene levels high enough to affect bone development in fry. Burbot, land-locked char (Bjørnøya), Greenland shark, and Greenland halibut have  $\Sigma$ PCB levels associated with increased liver enzyme production in Arctic char. Some invertebrates are at risk for the reproductive effects of TBT, particularly those that are exposed in harbors.

An assessment of risks from dietary intake has been attempted based on results from laboratory feeding experiments, POP levels in prey items, and information on different species' food preferences. Reproduction in piscivorous marine mammals, such as minke whale, beluga, narwhal, long-finned pilot whale, harbour porpoise, and seals, may be affected by dietary levels of  $\Sigma$ PCBs in ma-

rine fish. Killer whales, walrus, polar bear, and Arctic fox that prey on seals have dietary intakes of  $\Sigma$ PCBs that may cause reproductive effects. When environmental quality guidelines for protecting aquatic wildlife are used, dietary intakes of  $\Sigma$ DDTs,  $\Sigma$ PCBs, and dioxin-like substances are problematic for many marine and freshwater piscivorous species and, in some cases, for molluscivores. The same is true for predatory seabirds feeding on seabird eggs and chicks, and for killer whale, walrus, polar bear, and Arctic fox that consume marine mammals.

### 6.4.3. Conclusions

Assessment of contaminant levels in the previous AMAP assessment suggested that several species were at risk for neurobehavioral, reproductive, and immune system effects, particularly polar bears, predatory birds, seabirds, and seals. This has been borne out in biological effects studies done on polar bears, northern fur seals, glaucous gulls and possibly also Steller sea lions. The implications of these findings are that there are other populations of these species, and other highly contaminated species, that are being affected by current levels of some POPs.

Effects that are biologically significant are those that affect resistance to infection, reproduction, and behavior. Anything that affects these negatively reduces the margin of safety for the affected species, putting them at higher risk. Biomarkers for POP effects measure changes at the cellular or individual level, and are warning signals. The results from biomarker studies in the Arctic have shown that there are associations between several biomarkers and concentrations of some POPs. Results from field experiments and laboratory studies give added weight to the possible link between some POPs and specific effects. Therefore, based on the present evidence, it is believed that effects of biological significance are occurring in some Arctic species related to POPs exposure. These effects of biological significance are:

- polar bears are at higher risk for infections due to immune effects of POPs;
- glaucous gulls with high POP levels are at higher risk of immune, behavioral and reproductive effects, and effects on adult survival;
- northern fur seals are at higher risk for infections due to immune dysfunction correlated to POP exposure;
- peregrine falcons continue to exhibit egg shell thinning and reproductive effects of POPs;
- Arctic char exhibit immune effects of PCB; and
- dogwhelks exhibit the reproductive effects of TBT.

Knowledge gaps, such as understanding the influence of confounding factors, indicate that other biomarkers studied in Arctic biota (thyroid hormones, vitamin A and cytochrome P450 activity) should be considered indicators of increased exposure. It is not yet possible to conclude that any documented changes in these biomarkers imply increased risk. Studies on the effects of other stressors, such as long periods of fasting, on PCB effects in Arctic char show that PCB exposure increases susceptibility.

The lack of experimental dosage/response data continues to limit the ability to interpret residues in Arctic animals.

## Conclusions and Recommendations

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The previous AMAP POPs assessment (de March *et al.*, 1998) made recommendations and identified information gaps on the basis of the state of knowledge that existed in 1997. The research and monitoring of OCs and other halogenated contaminants (i.e., brominated and fluorinated compounds) in the Arctic that has occurred since that time has been influenced by that document. Although the state of knowledge on OCs in the Arctic has clearly advanced, and many knowledge gaps have been filled, others still remain and new questions and concerns have arisen.

One of the most important accomplishments of Arctic research concerning OCs, and the previous AMAP POPs assessment was the role it played in the negotiations of a global agreement to ban the 'dirty dozen' OCs (PCBs, DDT, etc.).

The measurement of 'new' chemicals, in particular brominated and fluorinated compounds, in the Arctic environment and the evidence of biological effects of OCs in polar bears, glaucous gulls, and northern fur seals are highlights of the recent research carried out on POPs in the Arctic.

Despite these advances, the general recommendations made by the authors of the previous AMAP POPs assessment continue to apply. With slight changes, these recommendations are:

- to continue monitoring levels of POPs in the abiotic environment and in biota, with emphasis on temporal and spatial trends and 'new' chemicals;
- to increase the research and understanding of OC transport and fate processes. This is of paramount importance in light of concerns about fundamental changes to Arctic ecosystems (e.g., climate change);
- to continue to refine and develop methods for determining subtle biological effects, relating OC levels to these effects, and integrating these data with information on population level effects and health, with due consideration to confounding factors such as age, sex, condition etc.; and,
- to continue to promote measures to reduce levels of OCs, in particular 'new' and current-use chemicals, in the environment.

### 7.1. Levels and effects

#### 7.1.1. Air and precipitation

Measurements of POPs in Arctic air have continued on a weekly basis at locations in Canada, Iceland, Norway, Finland, and Russia. A large temporal-trend dataset is now available. Comprehensive interpretation of the data is limited to results from Alert. These results show that half-lives of several PCB congeners, HCH, and chlor-dane-related compounds are much longer than at temperate air monitoring sites (e.g., Great Lakes and the U.K.). Spatial trends are difficult to evaluate because of the large week-to-week variation in concentrations, due

to temperature changes and long-range transport events, as discussed in Section 5.1. The different number and types of chemicals analyzed at each site also makes inter-comparisons difficult. Nevertheless, it is clear that concentrations of most OCs are higher at Ny-Ålesund and in eastern (Dunai) and western (Amderma) Russia than in the Canadian Arctic. 'New' chemicals such as PBDEs and PCNs appear to be important contaminants in Arctic air, however, little is known about their long-term trends. There exist problems with determination of PBDEs and potentially also with other widely used products such as flame retardants, surfactants, and plasticizers (including chlorinated paraffins, perfluorinated alkyl sulfonates and carboxylates and phthalates). The sampling media, emissions from laboratory building materials, or use near the site could inadvertently contaminate the samples.

Long-term precipitation measurements are limited to one site in northern Finland (Pallas) and to studies of glacial snow cores. These studies provide a valuable record of ongoing deposition, which complements air monitoring. Despite the importance of precipitation in delivering POPs to terrestrial and aquatic environments, the geographic coverage of deposition studies does not include sites in the North American Arctic and in Russia. Glaciers in the Russian, Canadian and Alaskan Arctic offer potential sites for examining temporal trends of POPs in snow/ice.

*Further work is urgently needed to monitor the levels of PBDEs in air given their increasing presence in Arctic biota (see Sections 4.1.2.5.3 and 5.4.6.1).*

*Air monitoring at selected locations should continue to study long-term temporal trends of POPs. Efforts should be made to standardize the analytes measured at each site, so that data are more easily inter-compared. The continuation of air and precipitation monitoring is particularly relevant to agreements such as the Stockholm Convention and the UN ECE POPs Protocol, as the resulting data provide a valuable baseline from which to measure future progress. This work should be expanded to new classes of chemicals. Continued studies of dry and wet deposition mechanisms, and scavenging by snow, are needed in order to understand and better model transport pathways as well as to determine fate and sources to the Arctic. Deposition monitoring should be encouraged at sites in North America and Russia. New passive air sampling techniques such as SPMDs should be considered to improve the geographical coverage of air measurements.*

#### 7.1.2. Sea- and freshwater

Recent work on HCH isomers has revealed that ocean currents may now be a driving force in OC transport to the Arctic, and ultimately in influencing levels observed in biota. Originally,  $\alpha$ -HCH was transported to the Arc-



tic via the atmosphere where it partitioned strongly into cold Arctic water. The reduction in emissions of many 'legacy' OCs has now resulted in their fugacities being greater in the ocean compared with air. For many chemicals, the ocean is therefore now the largest reservoir in the Arctic. Due to a combination of ice cover and circulation of older water from the European Arctic, the highest levels of  $\alpha$ -HCH in the world's oceans are found in the Canada Basin and the Canadian Arctic Archipelago. Furthermore, due to its higher water solubility,  $\beta$ -HCH seems to have more efficiently rained out of the atmosphere into the north Pacific surface water, and has subsequently entered the Arctic in ocean currents passing through the Bering Strait, resulting in a time-lag in delivery to the Arctic in comparison with  $\alpha$ -HCH. Ultimately therefore, ocean transport may be more important than atmospheric transport in influencing observed levels of some chemicals in Arctic marine biota. Ocean currents, ice cover, and water-air exchange of OCs are all subject to alteration as a result of climate change.

With the exception of HCH isomers, measurements of most other POPs in seawater are still too limited, especially in the European Arctic, to support comprehensive assessment of spatial trends. Such information would be valuable, for example for better understanding the higher concentrations of PCBs and DDT in biota in the European Arctic. Ultimately, it is the inventory of contaminants in the water column that is likely to drive the temporal trends of POPs in marine biota. PCB measurements in seawater are very challenging because of low levels and the potential for shipboard as well as laboratory contamination. Ultraclean techniques used by German, Swedish, and Norwegian researchers have yielded far lower PCB concentrations than measurements by Canadian scientists using *in situ* samplers or large-volume solvent extraction.

*It is recommended that monitoring of OCs in seawater continue and encompass a greater geographical range, with particular emphasis on the European Arctic for which few measurements are presently available. 'New' chemicals should be incorporated in this monitoring, especially those that are considered persistent but non-volatile (e.g., as demonstrated by recent observations with  $\beta$ -HCH). New technologies, such as SPMDs, should be investigated for obtaining information on long-term, prevailing concentrations in ocean waters and seasonal changes, especially of PCBs. Sites for long-term monitoring of temporal trends should be selected. The difference between various sampling methods for low-level PCBs in Arctic Ocean waters need to be resolved, possibly using side-by-side comparison of sampling methods.*

High HCH and DDT levels in Russian river waters first reported in AMAP Phase I have been verified by further data analysis and some additional measurements. Ratios of  $\alpha$ -HCH to  $\gamma$ -HCH indicate use of lindane. High proportions of DDT in  $\Sigma$ DDT measurements in Russian waters suggest recent use of DDT. Current levels of POPs in Russian Arctic rivers are, however, largely unknown with few new results available since the mid-1990s. A limited number of samples from the recent

RAIPON/AMAP/GEF study in Russia suggest continuing, relatively high, PCB contamination in surface Arctic freshwaters when compared to measurements from the mid-1990s in Canada.

The general lack of knowledge concerning OC concentrations in Arctic lake waters identified in the previous AMAP assessment report has not been addressed. There is almost no information on 'new' chemicals in Arctic freshwaters. Concentrations of  $\Sigma$ PCBs in the waters of some Arctic lakes in Canada and Russia exceed some guidelines for protection of freshwater aquatic life. Lack of spatial trends of OCs in freshwater makes the assessment of geographic variability in OC levels observed in biota difficult.

*Additional studies are needed on geographical trends of OC levels in Arctic lake waters. Determination and monitoring of 'new' chemicals in Arctic freshwaters are needed. The high OC levels found in Russian lake and river waters need further detailed study, including information on sources and spatial and temporal trends.*

### 7.1.3. Sediments

The previous AMAP POPs assessment concluded that "PCB levels in both freshwater and marine sediments generally do not exceed thresholds associated with biological effects". New data on levels of PCBs and OC pesticides in marine sediments of the Canadian Arctic have verified that levels are low in non-harbor areas. PAHs in sediments were not considered in this assessment. Based on recent studies in Norway and the Kola Peninsula, there is a need to assess the role of harbors as a source of OCs to oceans and the local environment. This is particularly true for 'new' and current-use chemicals.

Limited new data on POPs levels in freshwater sediments have been produced since the previous AMAP assessment. Recent OC measurements in Russian freshwater sediments found levels that are similar to other regions of the Arctic, although a single sample from one location had exceptionally high levels of DDT, which appear to be fresh based on high proportions of DDT to its metabolites. Newly available information on OCs in sediments from Russian Arctic rivers showed declining concentrations over the period 1988-1994.

*Research on the extent of the influence of harbors on ocean contamination is needed, particularly for 'new' chemicals. Further verification of the extent of DDT contamination in Russian rivers and lakes is desirable.*

### 7.1.4. Soils and vegetation

New data on levels of OCs in Russian soil and vegetation suggest little geographical variation in OC levels in these matrices across the Russian-European Arctic. Levels are at or near detection limits and approach levels found in blanks, which may obscure any trends that may exist. There is a general lack of recent data, making it difficult to assess the global reservoir of OCs and to model global cycling of OCs. Soils and vegetation can represent a significant component of the global environmental reservoir of OCs. A recent global study on the levels and loadings of OCs in soil included only a few Arctic sites.



*Extensive monitoring of OCs in Arctic soils and vegetation is not required, although a survey of soil and plant loading of OCs in the Arctic would contribute significantly to global modeling efforts to understand the ultimate fate and time to virtual elimination of these compounds.*

#### 7.1.5. Biota

Levels of OCs in Arctic species and environments are generally lower than in temperate areas, except where impacted by a local source. 'New' POPs, such as brominated and fluorinated compounds have been measured in Arctic biota. PBDE levels have increased significantly in the past ten years but are currently still at levels that are orders of magnitude lower than legacy OCs. Species monitored in the circumpolar Arctic during AMAP Phase II are essentially the same as those monitored during AMAP Phase I, with the emphasis on the most heavily contaminant species (polar bear, ringed seals, beluga, glaucous gulls). New data on polar bears, ringed seals, seabirds, and terrestrial mammals and birds from Russia have provided additional insights into circumpolar trends of OCs.

New studies of OC levels at different trophic levels in different marine ecosystems (Alaskan, Canadian, and European) have significantly increased understanding of the factors that influence OC levels and trophic transfer, confirming that biomagnification is a dominant factor in explaining high OC levels in Arctic biota. However, migration is also an important factor in several species and their prey. Physical-chemical properties of the contaminants, and the biological characteristics of the organisms play important roles in observed concentrations throughout the Arctic. Warm-blooded animals (birds and mammals) have been shown to accumulate OCs at a much higher rate than cold-blooded animals (fish and zooplankton). Size of zooplankton and their feeding habits are important variables influencing OC levels, but the understanding of factors influencing OC levels at the base of food webs is limited. Detailed models of bioaccumulation and fate of OCs have been developed for ringed seals and beluga but not for other species, and they have not been linked to food web models.

*The role of abiotic-biotic interfaces at the base of food webs needs to be better understood in both marine and freshwater environments. This is particularly important if changes in climate and ocean currents alter food webs and distribution or delivery of contaminants. There is a need to study the trophic transfer of 'new' chemicals in marine and freshwater food webs.*

Few new data on OCs have been produced for terrestrial biota in Canada, the U.S. or Norway. Terrestrial mammals and birds were, however, included in a recent extensive survey of contaminants in Russian terrestrial herbivores used as food items in traditional diets. In addition, mammals were included in studies of contaminants in Greenland and Faroe Island biota, and in Finland, reindeer were studied. In general, concentrations of persistent OCs in terrestrial mammals were low in the Russian Arctic, the Faroe Islands, Finland, and in Greenland. The exception is high PCDD/F concentrations in

Russian reindeer and mountain hare from the Kola Peninsula, due to the presence there of non-ferrous metal smelters. Concentrations of PCBs in Russian reindeer samples were slightly higher than previous reports for Canadian caribou (mid-1990s). However, no recent data are available from Canada for comparison.

In the previous AMAP assessment, there was some concern for carnivorous and piscivorous mammals (e.g., wolf and mink) which have elevated OC levels, however these levels are orders of magnitude lower than those found in some marine mammals. Few new data are available for these species. For migratory birds of prey, however, levels of OCs are still high and remain a cause for concern, while PBDE levels measured to date are higher than in marine mammals.

*While extensive monitoring of OCs in Arctic terrestrial biota is not required, further studies are needed to develop baseline levels for new, emerging POPs such as PBDEs and perfluorinated acids. Monitoring OCs and 'new' chemicals in birds of prey is still warranted, however.*

Freshwater biota contain higher levels of POPs than those in terrestrial environments, mainly due to more complex food webs. Lipid-weight PCB levels in freshwater fish are similar or slightly lower than levels in anadromous and marine fish with some major exceptions. Studies on the factors affecting levels in top trophic-level fish in different lake systems are ongoing.

Levels of POPs in the biota of Lake Ellasjøen on the island of Bjørnøya, north of Norway, are the highest measured in a freshwater system of the Arctic. These high levels are believed to be caused by the input of contaminants in guano from seabirds. Nearby lakes that do not receive guano, have much lower POP levels. A similar situation is indicated in fish from a freshwater lake on Jan Mayen.

*Ongoing surveys and studies of contaminants in the freshwater food web and the variables influencing levels should continue. Locations where there are elevated OC levels due to local contamination or food web effects, such as Lake Ellasjøen on Bjørnøya, are the most appropriate sites for further work.*

A wider range of marine species has been studied compared to the previous AMAP assessment, and considerably more data were available for Alaska and Russia in the current assessment. This wider range of marine species includes plankton, invertebrates, more species of fish, seabirds, pinnipeds, and cetaceans, as well as sea otters, Arctic fox, and polar bears. Levels of OCs in Arctic biota are generally highest in the top trophic-level marine organisms (e.g., great skuas, glaucous gulls, great black-backed gulls, killer whales, pilot whales, Arctic fox, and polar bears). This is particularly true for biomagnifying OCs such as PCBs and DDT. Within species, dietary intake at different trophic levels affects OC levels, as shown for polar bear, walrus, killer whales, Arctic fox, and glaucous gulls; with predation at higher trophic levels leading to higher OC concentrations. New data for OCs have been generated for Arctic fox from sites in Alaska, Canada, and Iceland, filling an important data

gap in the previous assessment. In Arctic fox on Svalbard, concentrations were comparable to, or higher than, in polar bears. The PCB concentrations found in Arctic fox from Alaska, Canada, and inland Iceland were lower, but in foxes from coastal populations in Iceland that feed in the marine food web, concentrations were comparable to those found previously on Svalbard.

Concentrations of MeSO<sub>2</sub>-PCB and -*p,p'*-DDE metabolites, OH-PCBs, and a previously unidentified phenolic metabolite of OCS, 4-hydroxyheptachlorostyrene (4-OH-HpCS) have been determined in polar bear and found to be high. Therefore, for Arctic organisms with efficient biotransformation capabilities, metabolite formation may be substantial and analysis of only parent compounds may give a skewed picture of actual contaminant levels. There is also evidence of bioaccumulation of some MeSO<sub>2</sub>-PCB metabolites in the Arctic cod – ringed seal – polar bear food web.

*Ongoing surveys and studies of contaminants in marine organisms, and the variables influencing levels, should continue.*

#### 7.1.6. 'New' chemicals

A number of 'new' chemicals that may be potential POPs have been found in the Arctic. The most surprising finding is the presence of PFOS, which has never been reported previously. This is of particular note because, based on its physical-chemical properties, it was not expected that PFOS would be found in the Arctic. PFOS levels are high in polar bear liver and it is one of the most prominent individual organohalogen chemicals in polar bear, when levels of PCBs, chlordane, and HCH-related chemicals are considered. The presence of PBDEs, chlorinated paraffins, and PCNs was anticipated based on very limited monitoring reported in AMAP Phase I. These newly identified organohalogen compounds are generally at much lower concentrations in Arctic air and biota compared with temperate regions. Nevertheless, temporal-trend studies have proved valuable in demonstrating increasing concentrations in Arctic biota, particularly the PBDEs.

Until recently, measurements of persistent organic chemicals in the Arctic have been confined to those which can be easily measured using gas chromatography, representing only a fraction of the possible persistent chemicals in commerce. However, the presence of chemicals like PFOS illustrates the need for a very broad analytical approach (PFOS can only be analyzed by Liquid Chromatography-Mass Spectrometry (LC-MS)). Natural halogenated compounds such as Q1 (a heptachlorobipyrrole), MHC-1 (a chloro-bromo-monoterpene), and halogenated dimethyl bipyrroles that have been detected in Arctic biota are another class of chemicals that needs to be considered. Temporal trends and body burdens of these chemicals, which are produced by marine algae, need to be considered when assessing impacts of some closely related anthropogenic chemicals like the PBDEs.

*Additional research and monitoring of 'new' chemicals is needed, in particular for brominated compounds that are increasing in concentration. An understanding of circumpolar trends for these 'new' chemicals is needed, in*

*particular for the Russian Arctic. Work on persistent organic compounds in current industrial, consumer, and agricultural uses should be encouraged, even for chemicals that are not considered to have potential for atmospheric transport to the Arctic based on their physical properties. The influence of point sources for chemicals, such as harbors and waste disposal sites, including waste burning, should be assessed.*

In the previous AMAP assessment, only limited data were available on toxaphene concentrations in Arctic biota. A great deal more data are now available, showing that toxaphene is widespread throughout the Arctic. In some seal species, toxaphene concentrations are comparable to PCB concentrations. Some whale species, such as beluga and narwhal, also have particularly high toxaphene levels. Only very limited temporal trend data are available for toxaphene.

*Monitoring of toxaphene should be extended and temporal trend monitoring studies of toxaphene established. Biomarkers for toxaphene exposure need to be developed and biological effects monitoring undertaken in those species with high levels.*

TBT is found extensively in invertebrates of Arctic harbors, but information on levels in organisms in regions away from harbors remains limited. Butyltin compounds (MBT, DBT, TBT) were present in very low or non-detectable levels in Canadian Arctic and Faroe Island marine mammals, but in Norwegian marine mammals and seabirds are at levels that warrant further study.

*Future measurement of TBT should continue to focus on the invertebrate community. Additional work should be carried out to determine levels of mono- and dibutyltin in abiotic and biotic samples in the Arctic because of their ongoing use, which is unrelated to TBT uses.*

#### 7.1.7. Biological effects

There has been limited advancement in understanding the effects of low tissue levels or body burdens of OCs or low-level intakes in Arctic biota, which was an identified knowledge gap in AMAP Phase I. Most of the information has been developed for PCDD/Fs, non-*ortho* PCBs, PCB, and DDT using non-Arctic animals. Threshold levels have mainly been established in laboratory animals for effects on reproduction, neurobehavioral development, and immunosuppression. There are major species differences in susceptibility to the toxic effects of POPs. A major gap in knowledge is the sensitivity of Arctic species for effects compared to other species where more knowledge is available. This, in turn, makes it difficult to know if the threshold values determined in other species are valid for comparison with the contaminant levels found in Arctic species.

Another major knowledge gap is the fact that the toxicity mechanisms for many POPs are still not known, and this is an urgent research priority, especially for those substances found at high concentrations in Arctic biota.

There are no, or insufficient thresholds data for many 'new' POPs such as PBDEs and PFOS, as well as for biologically active metabolites of some POPs, such as MeSO<sub>2</sub>-PCBs, MeSO<sub>2</sub>-*p,p'*-DDE, and OH-PCBs. Thus, it is not currently possible to assess the effects of current levels of these contaminants in Arctic biota.

*A future priority area for effects studies and risk assessment is, therefore, determining effects thresholds of POPs for Arctic species, so that more relevant and reliable comparisons can be made. More research is also needed on toxicity mechanisms of many POPs including establishment of effects thresholds for new substances and metabolites.*

The most biologically significant effects are those that affect resistance to infection, reproduction and behavior. Anything that negatively affects resistance to infection, reproduction or behavior reduces the margin of safety for the affected species, putting them at higher risk. Assessment of contaminant levels from the previous AMAP assessment suggested that several species were potentially at risk for neurobehavioral, reproductive, and immune system effects. This has now been borne out in biological effects studies carried out on polar bears, northern fur seals, glaucous gulls, and possibly also Steller sea lions. Results from field experiments and laboratory studies give added weight to the possible link between some POPs and specific effects. The implications of these findings are thus, that there are both populations of these species in other areas, and other highly contaminated species, that are being affected by current levels of some OCs. Based on the present evidence, it is believed that effects of biological significance related to OCs exposure are occurring in some Arctic species. These are as follows:

- polar bears may be at higher risk for infections due to immune effects of OCs;
- glaucous gulls with high OC levels may be at risk due to immune, behavioral and reproductive effects, and effects on adult survival;
- northern fur seals may be at higher risk for infections due to immune dysfunction correlated to POP exposure;
- peregrine falcons continue to exhibit eggshell thinning and reproductive effects of OCs;
- Arctic char exhibit immune effects of PCBs; and,
- dogwhelks exhibit the reproductive effects of TBT.

The biological effects studies on polar bears in the Norwegian Arctic show that there is some evidence of reduced cub survival, suppressed immune function, and disturbance of thyroid hormone and retinol homeostasis. Similarly, there are also indications that high OC burdens may affect cub survival in Hudson Bay polar bears. Biological effects studies comparing Canadian and Svalbard polar bears indicate that some serious disturbances in immune responses, as well as in thyroid and retinol systems, may be related to current levels of PCBs. Assessment of these results indicates that population status and health of polar bears with very high PCB levels may be at risk. The significance of these findings on the individual and population levels has therefore to be further investigated.

Although the study of northern fur seals discussed in Section 6.3.4.1 was not designed to elucidate fully the role of OC contaminant exposure-induced immunosuppression in the decline of St. George Island northern fur seals, it did identify a cohort (pups born to young dams) that is at higher risk. OC contaminant exposure at an extremely sensitive and critical period of development must be considered as a potential contributing factor to reduced post-weaning survival based on evidence of a compromised immune system.

The potential effects of environmental contaminant exposure during critical developmental life stages emphasizes the need for further monitoring and research. There is an ongoing need for better biomarkers, and for biomarkers to cover more effects, such as other types of hormone disruption.

Biomarkers for OC effects measure changes at the cellular or individual level and provide warning signals. The results from biomarker studies in the Arctic have shown that there are associations between several biomarkers and OC levels based on measuring biomarker responses and correlating these to levels of OCs. Such results show association but not causation. Many OCs covary so that it is not possible to state equivocally that a certain OC is the cause of the effect seen. These factors make risk assessment very difficult.

There is a general lack of knowledge of the physiology of most Arctic species, particularly those with high OC exposures. This includes knowledge concerning baseline levels of hormones, vitamins, blood variables, immune factors, etc., and other factors that affect these (e.g., time of day, time of year, reproductive state, health status, fasting, etc.) Because of these knowledge gaps, and the influence of confounding factors, other biomarkers studied in Arctic biota (such as thyroid hormones, vitamin A, and cytochrome P450 activity) should be considered indicators of increased exposure. It is however not yet possible to conclude that changes in these imply increased risk.

*All the species at risk should be monitored directly for contaminant levels and possible biological effects. Biological monitoring of the most heavily contaminated species should be encouraged, together with studies including the entire food web, in order to clarify and understand the transfer and biomagnification of contaminants. In the Russian Arctic, high-level predators such as polar bear should be studied since OC levels seem to be higher than at Svalbard.*

*There is a need for refinement and development of sensitive biochemical- or physiological-level assays for use in Arctic biota. For example, development of in vitro methods based on tissues from Arctic species. Practical problems that must be overcome include: the difficulty in collecting biopsy samples from live animals or fresh tissue from hunted animals in remote regions, and related problems in sample preservation and storage. Linkage of the results for planar OCs to bioassays of cytochrome P450 1A1 and 2B activity on sample extracts, a technique widely used in contaminant studies in the Great Lakes, should be considered in order to confirm that the biological activity associated with the measured contaminants is accounted for.*

*Well-designed studies combining laboratory, semi-field and field studies are needed to determine causation,*



particularly for the most highly exposed species in the Arctic. Arctic fox could be a possible model for Svalbard polar bear as they have similar feeding habits on Svalbard. Svalbard foxes have high OC levels. They also go through fasting periods, and immunological tools are available for foxes. Arctic fox can be studied in the field, in semi-field conditions and in the laboratory if blue fox (a color variant of Arctic fox raised on fur farms) are used. A coordinated international project should be developed along these lines so that as many methods, and the knowledge of as many pertinent research groups as possible, can be used. Similar studies are feasible using glaucous gulls.

Studies on the influence of other stressors, such as long periods of fasting in Arctic char, show modulation of these OC-related effects.

*More knowledge concerning starvation effects in birds and mammals is needed. Interpretation of correlative hormone studies is hampered by lack of information on other variables, such as long-term fasting/starvation, that may affect hormone levels in wild populations. This makes drawing conclusions from some biomarker studies tenuous.*

Other stressors, including the effects of parasites, light, noise, and higher temperatures in the wake of global warming, may make organisms more or less susceptible to the effects of OCs; however, little is known about such influences. For example, there is evidence of immunosuppression in marine mammals due to thermal stress related to a small increase in ocean water temperature. Noise pollution also appears to be immunosuppressive.

*More research is needed into the influence of other stressors on organisms, including modulation of OC-related effects.*

Outside the Arctic, tumors have been found in wildlife highly exposed to OCs, such as beluga from the St. Lawrence Estuary. An increase in chromosome aberrations and DNA adducts were found in glaucous gull chicks exposed to OCs via food. No other studies of possible mutagenic effects or tumor occurrence have been carried out in any Arctic biota.

*Where possible, surveys of tumor incidence in highly exposed marine mammals and seabirds should be carried out, possibly as collaborative efforts with native hunters. Surveys of mutagenic effects in some highly exposed species should be carried out to determine the prevalence of these effects.*

Imposex in invertebrates such as dogwhelk has been found to be more widespread in the Arctic than previously thought and is correlated to TBT levels. Imposex has now been reported along the Arctic coast of Alaska, Iceland, Norway, Greenland, and the Faroe Islands. Improvement has been seen in countries where TBT has been banned for use on boat hulls.

*Further monitoring of imposex and TBT concentrations along Arctic coasts should continue so as to follow current trends as well as to establish occurrence in areas not previously studied. Monitoring is also needed in order to follow temporal trends when TBT is banned for use as an antifoulant paint on ships. The potential effects of new antifoulant paints that are beginning to replace TBT should be addressed.*

## 7.2. Spatial trends

Extensive spatial surveys of OCs in marine mammals (polar bears, ringed seals) and seabirds now give better circumpolar coverage than was available in the previous AMAP assessment. All spatial trends that include the Russian Arctic clearly show that PCB and DDT concentrations are highest in the eastern Barents Sea and Kara Sea area, with concentrations decreasing to the east and west of these, indicating significant local sources of DDTs and PCBs in Russia. This is also indicated by high OC inputs to the Arctic Ocean from the Ob, Taz, Nadym, Pur, and Yenisey Rivers. These rivers have been shown to be significant sources of OCs to the Kara Sea/Arctic Ocean via river water and sediments. Levels of OCs in birds and marine mammals are generally higher in the European Arctic compared to the Alaskan and Canadian Arctic. The exception is HCH, where levels are highest in the Alaskan and western Canadian Arctic due to the recent use of this chemical in Asia. There are also local spatial trends in biota in the Aleutian Islands (Alaska) and Saglek Bay (Canada), particularly in PCB concentrations. These seem to be related to local sources, primarily military sites.

Studies of ringed and harp seals and their food web in the White Sea have shown that levels of PCBs and DDT-related compounds are higher there than in the North American Arctic. Harp seals in the White Sea and eastern Svalbard (the same migrating population) have elevated levels of toxaphene compared to ringed seals in the Canadian Arctic. Toxaphene levels were also relatively high in minke whales from the Barents Sea and eastern Svalbard area compared to the West Greenland stock.

There are numerous gaps in knowledge of spatial trends of OCs in marine biota. Little is known about levels of OCs in biota from the Kara, Laptev, and East Siberian Seas with the exception of a study of PCB/OC pesticides in polar bear blood, and another on ringed seals in the Kara Sea.

*Additional surveys of 'legacy' and 'new' POPs are needed in the Russian marine system, particularly in the Kara, Laptev, and East Siberian Seas. Further research is needed to determine and quantify PCB and DDT sources to the Barents/Kara Sea area. In particular, more studies are needed on the influence of military sites (active and inactive) as local sources of OC contamination to the marine environment.*

PBDEs (mainly BDE47 and BDE99) have been detected in numerous Arctic animals, including birds of prey, freshwater fish, ringed seals, minke whales, long-finned pilot whales, beluga, polar bears, and seabirds. The



highest levels are in the thousands of ng/g range in pilot whales and birds of prey but otherwise, PBDEs are in the low ng/g range. PBDE levels appear to be higher in glaucous gulls, ringed seals, and beluga at Svalbard than in the same species in the Canadian Arctic. Highest levels are found in some Scandinavian migratory birds of prey such as peregrine falcons and white-tailed sea eagle. Spatial trends of PBDEs are not yet well documented, however. Studies of PBDEs in eggs of birds of prey in Norway and Sweden have found no clear south to north trends.

*Circumpolar trends of 'new' chemicals in the Arctic are a very significant data gap and need to be addressed in both the abiotic and biotic environment.*

An emerging issue is the possible effect of shifts in trophic structure due to ongoing 'regime shifts' or warming trends, such as the one being observed in the southern Beaufort Sea. This could lead to differences in diet for top predators, which could affect trends in persistent organohalogen contaminants. An illustration of this dietary effect was discussed at the AMAP conference in Tromsø, Norway (January 2002). Most OCs are lower in polar bears in the Chukchi and southern Beaufort Sea coasts than elsewhere. It was pointed out that this may be related to the availability of bowhead whale carcasses in the region, the only area of the Arctic where bowheads are hunted extensively. If, because of climatic shifts, polar bears from this area had more ringed seal in their diet, they would have higher PCB levels since seals are higher in the food web than bowheads and have a different pattern of PCB congeners.

The example of feeding on whales also underlines the need to consider dietary sources and food web pathways of contaminants before inferring regional/geographical trends in levels, and sources in trends. Monitoring programs have begun to make greater use of N and C stable isotope data to gain insight into such dietary effects as well as to study food webs together with top predator sentinel organisms. However, such work requires a multidisciplinary team and substantially more funding for ship time and fieldwork than is usually available for Arctic contaminants work.

*Use of stable isotopes of both N and C and newer methods such as fatty acid patterns to elucidate food web effects should be incorporated into future studies.*

With the exception of HCH isomers, spatial trends of OCs in marine zooplankton and fish do not match those observed in seabirds and marine mammals. For example, levels of PCBs in lower trophic-level organisms, such as zooplankton and Arctic cod, do not differ between the eastern Canadian Arctic and the European Arctic, although differences are clear for top predators such as seabirds, ringed seals, and polar bears. Toxaphene and PCB levels in seawater in the White Sea were similar or lower than observed in the southern Beaufort Sea. Concentrations of OCs in calanoid copepods and other zooplankton are similar or higher, depending on the OC group, in the Alaskan and Canadian Arctic than in regions around Svalbard. No data are available for marine zooplankton in the Russian sector of the Arctic Ocean. Food web differences may account for more

of the observed differences than has been assumed previously.

The data on which these conclusions are based are however limited, especially in the European Arctic. A comparative survey of OCs in seawater and zooplankton, using similar collection (e.g., time of year) and analytical methods, is therefore suggested. Additional studies on factors influencing OC levels in lower trophic-level biota are also encouraged.

*More research is needed to explain the large differences in POP levels at high trophic levels in the Barents Sea ecosystem, as compared to the North American Arctic.*

### 7.3. Temporal trends

Long-term datasets for OCs in air at Alert and Ny-Ålesund have proven to be very valuable records of the current status of OCs in the Arctic and are of particular relevance in light of new global bans on OCs. An assessment of trends in air shows that PCBs at the Alert station are declining very slowly.

Key species such as polar bear, ringed seals, glaucous gulls, and guillemots continue to be monitored for PCBs and OC pesticides. Temporal-trend data are now available over a 25-30-year period for ringed seals and for eggs of several seabird species in the Canadian Arctic, as well as for Arctic char and pike from Swedish lakes. Trends in PCDD/Fs and non- and mono-ortho PCBs have been studied in seabird eggs over an 18-year period in the Canadian Arctic. In addition, new temporal-trend studies have been conducted using archived samples (polar bears in Hudson Bay; beluga whales in east Baffin Island; guillemots and Atlantic cod in Iceland; birds of prey in northern mainland Norway; polar bears at Svalbard; and, glaucous gulls at Svalbard). Temporal trends during the 1990s are available for PCBs, DDT, and toxaphene in ringed seals from Nunavik, Hudson Bay, and Greenland, and in harp seals from the White Sea. Temporal trends of OCs in seabirds from Alaska are available, and sample archives now exist which would allow further assessment of trends during the 1990s.

The general observation in all studies and species is that concentrations of total PCBs and total DDT are declining slowly. This reflects the slow decline in inputs from circumpolar countries and the northern hemisphere as these chemicals are phased-out. Major reservoirs, especially of PCBs and DDT, remain in soils in the northern temperate zone. Chlordane-related chemicals as well as dieldrin are also declining in the same species. Less is known about temporal trends of other chemicals that are potential POPs. ΣHCH concentrations have remained stable in ringed seals in the Canadian Arctic, but the proportion of recalcitrant β-HCH, an endocrine active chemical, has increased over the past 25 years. β-HCH has declined in concentration in polar bears in Svalbard (1991-2000) but not in polar bears from western Hudson Bay over the same period. However, β-HCH concentrations have increased while α- and γ-HCH concentrations have declined in seabirds in the Canadian Arctic. This is probably due to the time lag in delivery of β-HCH, which was washed out from the atmosphere via precipitation into the northern Pacific/Bering Sea and has only recently entered the Arctic via

the Bering Strait. Generally, the trends in rates of decline for DDT and  $\alpha$ -HCH indicate more rapid declines in the European and Russian Arctic and slower declines in the North American Arctic, particularly during the past decade. PBDE concentrations have increased in beluga and ringed seals in the Canadian Arctic between the early 1980s and the late 1990s. Endosulfan sulfate also appears to have increased in beluga. Chlorinated diphenyl ethers, by-products from the production of pentachlorophenol, have declined in beluga from Cumberland Sound in the eastern Canadian Arctic during the 1990s. Toxaphene levels show increasing temporal trends in seabirds, decreases in burbot and no change in beluga and narwhal in the Canadian Arctic.

There are numerous gaps in knowledge of temporal trends of OCs in marine biota. Temporal-trend information is limited to a relatively narrow range of OC compounds (e.g., PCBs and OC pesticides) with the exception of new studies in the Canadian Arctic on PBDEs and chlorinated paraffins in beluga and ringed seals. No temporal trend information on PBDEs or chlorinated paraffins is available yet for the European Arctic. The temporal-trend studies, especially those involving retrospective analyses of 'new' OCs, are dependent on tissue archives. Maintaining these archives is crucial for continued trend monitoring.

*Specimen banking remains a cornerstone of work on temporal trends of OCs in the Arctic and will be critical for future retrospective work on other persistent organics in biota. Only a few Arctic countries (Canada, U.S., and Sweden) have well-established specimen banks and even those are not designed for all major Arctic species. For work on more polar compounds, such as hydroxy-PCBs, halogenated phenolics and perfluorinated acids, which are found mainly in blood plasma, it is clear that a wider range of tissues will need to be archived.*

*Temporal trend monitoring of a broader range of contaminants in biota, including potential 'new' POPs, should be established. A circumpolar expert group is needed to design guidelines for archiving Arctic samples for future chemical analyses and biological effects studies.*

#### 7.4. Sources

High OC levels in Russian rivers reported in the previous AMAP assessment have been verified, although evidence also shows declining trends during the early 1990s. Extremely high levels of DDT were found in the sediment of the Pechora River in Russia in the recent RAIPON/AMAP/GEF study. High levels of OCs in Russian terrestrial and marine biota have also been found. Air, water, and biota monitoring suggest higher proportions of *p,p'*-DDT and lindane compared with other Arctic regions and point to recent or ongoing use. High PCDD/F levels in Russian reindeer, mountain hare, and freshwater fish from the Kola Peninsula are probably the result of proximity to local sources.

*Additional studies, including surveys and inventories, are needed to understand the sources of POPs in Russia.*

Based on studies in northern Norway and the Kola Peninsula, harbors may be a source of PCBs and other

OCs to oceans. The potential for northern harbors to be continuing sources of contamination to the Arctic Ocean, or to contaminate biota that migrate to other more pristine areas, needs further study.

Results from OC measurements in biota from the Aleutian Islands, and abiotic and biotic samples from Saglek Bay, Labrador, indicate military sites to be a source of PCB input to the local marine environment. Numerous local sources of other OCs may also exist, but very few have been studied. These local sources include burning of wastes in Arctic communities, smelter emissions of PCDD/Fs, industrial and community effluents, dumps, waste sites, etc. Some 'new' chemicals, in particular PBDE, are found in current-use products and pose a local contaminant concern. The role these sources play in loadings to the Arctic are not known.

*Surveys of local sources of contamination by OCs, in particular 'new' chemicals, within the Arctic are needed to quantify the emissions and leakage. These include both harbors and military sites. Where emissions are high, actions should be taken to reduce or remediate them.*

*All eight Arctic countries have now signed both the UN ECE Protocol on POPs and the global Stockholm Convention on Persistent Organic Pollutants. The Arctic countries are encouraged to rapidly ratify and implement these conventions.*

#### 7.5. General monitoring and assessment

Variability between analytical methods continues to be a problem (e.g., number of PCB congeners analyzed continues to vary between laboratories), although the situation has improved since the previous AMAP assessment. The analysis of toxaphene and 'new' chemicals needs to be addressed, especially chemicals such as perfluorinated compounds that have very different analytical methodologies.

Virtually all laboratories submitting analytical results for this assessment have taken part satisfactorily in international interlaboratory comparisons. The AMAP POPs program has been a major stimulus for this. However, contamination during sampling and laboratory analysis remains an issue, especially for PCBs and brominated flame retardants. Detection limits used by various studies vary widely. Levels of some POPs approach blank levels especially in the case of terrestrial herbivores.

*Future AMAP monitoring programs for POPs should continue to use a quality assurance program with mandatory participation of laboratories in interlaboratory comparisons for key matrices. Quality assurance programs for sampling strategy and sample collection should also be developed.*

Despite AMAP recommendations, PCB results continue to be given as sums of anywhere from seven to 90 congeners, making comparisons difficult. Most laboratories determined at least seven CB congeners (CBs 31/28, 52, 101, 118, 138, 153, and 180). However, the sum of the above seven represented between 11 and 65% of  $\Sigma$ PCBs in sediments, 50% in mosses, 50 to 66% in biota, and

only 10 to 30% of  $\Sigma$ PCBs in air. The lack of quantification of more than 7-10 congeners in air samples has made it difficult to use the results in source and pathway studies.

*The number of CB congeners should be standardized, and future analyses should require, as a minimum, a somewhat larger number of congeners to be determined, including toxicologically important mono-ortho PCBs such as CB105 and 118. Determination of 30 to 40 selected congeners (the exact number would have to be assessed by careful consideration of congener patterns in each matrix) should give results close to those for the sum of all possible congeners, with the other congeners contributing relatively little to  $\Sigma$ PCBs.*

Current methods for quantifying toxaphene may overestimate levels in some samples such as marine mammals and underestimate it in others such as air. The use of different quantification methods has limited the assessment of current atmospheric loadings of toxaphene to the Arctic Ocean and is a matter that needs to be resolved. Some laboratories are now reporting only concentrations of specific toxaphene congeners, which makes comparisons to older data difficult.

*Future monitoring should include determination of total toxaphene (by negative ion MS) for comparison with past work as well as selected chlorobornane congeners. This work should include at least ten major congeners found in air and biota, not just Parlars 26, 50, and 62. Lack of commercial standards hampers this, and there is an urgent need for more standards to be made available.*

While broad ranges of 'new' chemicals are being investigated in Arctic biota, there is also a need for models to predict the kinds of compounds that are in commercial use, and that may have 'Arctic accumulation potential'. Much progress in modeling has been made in this regard, but a strategy for developing a list of possible chemical contaminants needs to be considered in order to anticipate new chemical threats rather than focusing most resources on the list of 'legacy' OCs, as is being done at present.

Future studies of OCs in Arctic marine biota could benefit from more of a 'campaign' approach to coordinate monitoring and measurements (sites, sampling timing, and analytical methodology) similar to the approach taken for some human tissue sampling under the AMAP program. There is presently some degree of coordination for air measurements of OCs, although lists of analytes vary between laboratories, but there is almost no coordination for measurements in biota. Most groups are using the OSPAR Joint Assessment and Monitoring Program (JAMP) protocols for monitoring biota recom-

mended by AMAP. Extension of mussel watch type programs from more southerly regions is possible in some locations and has been implemented on an experimental basis in northern Labrador/Nunavik, northern Norway, and Greenland.

For air and water sampling, future programs should consider use of passive sampling devices, for example, plastic/triolein or XAD resin SPMDs for air and water, which are showing promising results in trials in Norway and Canada.

OC levels in biota can vary dramatically between individuals of the same species and population due to differences in age, size, growth rate, and sex. These differences are even more of an issue when OC concentrations are compared between populations from different regions or sites, and different times. Robust statistical analysis that incorporates these biological characteristics is of paramount importance when assessing spatial and temporal trends. Failure to carry out such statistical analysis can result in faulty conclusions and recommendations.

Also critical for future evaluation of temporal and spatial trends of OCs, is the availability of data for the assessment, including results for individual samples and associated biological data. The AMAP data centers can theoretically perform this function. Unfortunately, large amounts of data that have now been published in peer-reviewed journals and used in the previous AMAP assessment are still not yet in the AMAP data centers, especially data from Canada, the U.S., and Russia. This situation is bound to get worse as scientists involved in the previous and current AMAP assessments retire or change jobs. A renewed effort by AMAP to archive data in data centers with full access after a reasonable period is needed.

*It is strongly suggested that statistical analysis of all OC data incorporate all relevant and available biological data. New monitoring programs should include a suite of biological measurements. Results from monitoring programs should be published in peer-reviewed journals, and efforts should be made to have government reports peer-reviewed. Data should be compiled in (AMAP) data centers to ensure their future accessibility.*

Many current analytical reference materials have relatively high levels of contaminants, which are not always appropriate for use in analysis of low-level samples from the Arctic, and can result in cross-contamination during sample analysis. There are currently no standard analytical reference materials for 'new' contaminants.

*There is a need for development of 'made for the Arctic' analytical reference materials based on common matrices used for analysis (blubber, blood, other tissues, sediments). These should include legacy OCs, their metabolites, toxaphene, and other new chemicals of concern.*

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Annex Table 1. Physical-chemical properties of persistent organic pollutants discussed in the text.

Common name	Isomer/congener/abbreviation or chemical name	Formula	CAS number	Molecular weight	Water solubility, Cs*(mmol/m <sup>3</sup> ) (at 25°C)	Log K <sub>ow</sub> (at 25°C)	Henry's Law Constant, H (Pa m <sup>3</sup> /mol) (at 25°C)	Henry's Law Constant (Pa m <sup>3</sup> /mol) (at 0°C)	Vapor pressure, Ps** Pa (at 25°C)	Atmospheric half-life (hrs)	Reference
<i>Organochlorine pesticides</i>											
Chlordane	<i>cis</i> -CHL	C <sub>10</sub> H <sub>6</sub> Cl <sub>8</sub>	5103-71-9	409.8	0.137	6.0	0.342	0.87	0.0004	55 (~2 d)	1
	<i>trans</i> -CHL	C <sub>10</sub> H <sub>6</sub> Cl <sub>8</sub>	5103-74-2	409.8	0.137	6.0	0.262	0.87	0.00052	55 (~2 d)	1
DDT	<i>o,p'</i> -DDE	C <sub>14</sub> H <sub>8</sub> Cl <sub>4</sub>	3424-82-6	318	0.126	5.8	7.95	–	0.000866	170 (~1 wk)	1
	<i>o,p'</i> -DDT	C <sub>14</sub> H <sub>9</sub> Cl <sub>5</sub>	789-02-6	354.5	0.0733	6.0	0.347	0.16	2.5 × 10 <sup>-5</sup>	–	1
	<i>p,p'</i> -DDT	C <sub>14</sub> H <sub>9</sub> Cl <sub>5</sub>	50-29-3	354.5	0.0155	6.19	2.36	0.16	2.0 × 10 <sup>-5</sup>	170 (~1 wk)	1
Dieldrin	dieldrin	C <sub>12</sub> H <sub>8</sub> Cl <sub>6</sub> O	60-57-1	380.9	0.446	5.20	1.12	–	0.0005	55 (~2 d)	1
Endosulfan	α-endosulfan	C <sub>9</sub> H <sub>6</sub> Cl <sub>6</sub> O <sub>3</sub> S	959-98-8	406.9	1.23	3.62	0.718	–	0.0013	–	1
Hexachlorocyclohexane	α-HCH	C <sub>6</sub> H <sub>6</sub> Cl <sub>6</sub>	319-84-6	290.8	3.44	3.81	0.872	0.215 (8°C)	0.003	–	1
	β-HCH	C <sub>6</sub> H <sub>6</sub> Cl <sub>6</sub>	319-85-7	290.8	0.344	3.8	0.116	–	4.0 × 10 <sup>-5</sup>	–	1
	γ-HCH	C <sub>6</sub> H <sub>6</sub> Cl <sub>6</sub>	58-89-9	290.8	25.1	3.7	0.149	–	0.00374	170 (~1 wk)	1
Mirex	mirex	C <sub>10</sub> Cl <sub>12</sub>	2385-85-5	545.5	0.000119	6.9	839.4	–	0.0001	170 (~1 wk)	1
Toxaphene	technical	C <sub>10</sub> H <sub>10</sub> Cl <sub>8</sub>	8001-35-2	413.8	1.21	5.50	0.745	0.067	0.0009	170 (~1 wk)	1
	P26	C <sub>10</sub> H <sub>10</sub> Cl <sub>8</sub>	–	414	–	5.52	–	–	–	–	2
	P50	C <sub>10</sub> H <sub>9</sub> Cl <sub>9</sub>	–	448	–	5.84	–	–	–	–	2
<i>Byproducts</i>											
Hexachlorobenzene	HCB	C <sub>6</sub> Cl <sub>6</sub>	118-74-1	284.8	0.0176	5.50	131	–	0.0023	17000 (~2 yr)	3
Pentachlorobenzene	PeCBz	C <sub>6</sub> HCl <sub>5</sub>	608-93-5	250.3	2.6	5.00	85	–	0.22	5500 (~8 mo)	3
Octachlorostyrene	OCS	C <sub>8</sub> Cl <sub>8</sub>	29082-74-4	379.7	0.00458	7.46	23.3	–	0.00176	–	4
Polychlorinated dibenzo- <i>p</i> -dioxins	TCDD	C <sub>12</sub> H <sub>4</sub> O <sub>2</sub> Cl <sub>4</sub>	30756-58-8, 67028-18-6, 30746-58-8, 1746-01-6	322	0.000993-0.0013	6.60-7.10	0.704-3.747	0.60 (10°C)	2.00 × 10 <sup>-7</sup> -1.00 × 10 <sup>-6</sup>	170 (~1 wk)	5
	PeCDD	C <sub>12</sub> H <sub>3</sub> O <sub>2</sub> Cl <sub>5</sub>	39227-61-7	356.4	0.000331	7.40	0.266	–	8.80 × 10 <sup>-8</sup>	550 (~3 wk)	5
	HxCDD	C <sub>12</sub> H <sub>2</sub> O <sub>2</sub> Cl <sub>6</sub>	39227-26-8	391.0	1.13 × 10 <sup>-5</sup>	7.80	1.084	–	5.10 × 10 <sup>-9</sup>	550 (~3 wk)	5
	HpCDD	C <sub>12</sub> H <sub>2</sub> O <sub>2</sub> Cl <sub>7</sub>	35822-46-9	425.3	5.64 × 10 <sup>-6</sup>	8.00	1.273	–	7.50 × 10 <sup>-10</sup>	550 (~3 wk)	5
	OCDD	C <sub>12</sub> O <sub>2</sub> Cl <sub>8</sub>	3268-87-9	460	1.61 × 10 <sup>-7</sup>	8.20	0.684	–	1.10 × 10 <sup>-10</sup>	550 (~3 wk)	5
Polychlorinated dibenzofurans	TCDF	C <sub>12</sub> H <sub>4</sub> OCl <sub>4</sub>	51207-31-9	306	1.37 × 10 <sup>-3</sup>	6.1	1.461	–	2.00 × 10 <sup>-6</sup>	170 (~1 wk)	5
	PnCDF	C <sub>12</sub> H <sub>3</sub> OCl <sub>5</sub>	57117-31-4	340.42	6.93 × 10 <sup>-4</sup>	6.5	0.505	–	3.50 × 10 <sup>-7</sup>	550 (~3 wk)	5
	HxCDF	C <sub>12</sub> H <sub>2</sub> OCl <sub>6</sub>	70658-26-9, 57117-44-9	374.87	2.2 × 10 <sup>-5</sup> -4.72 × 10 <sup>-5</sup>	7.0	0.741-1.454	–	3.20 × 10 <sup>-8</sup> -3.50 × 10 <sup>-8</sup>	–	5
	HpCDF	C <sub>12</sub> HOCl <sub>7</sub>	67462-39-4, 55673-89-7	409.31	3.30 × 10 <sup>-6</sup>	7.4	1.425	–	4.70 × 10 <sup>-9</sup> -6.20 × 10 <sup>-9</sup>	550 (~3 wk)	5
	OCDF	C <sub>12</sub> OCl <sub>8</sub>	39001-02-0	443.76	2.61 × 10 <sup>-6</sup>	8.0	0.191	–	5.0 × 10 <sup>-10</sup>	550 (~3 wk)	5
<i>Polycyclic aromatic hydrocarbons</i>											
Acenaphthene	1,8-hydroacenaphthylene	C <sub>12</sub> H <sub>10</sub>	83-32-9	154.2	24.642	3.92	12.17	–	0.3	–	5
Fluorene	diphenylenemethane	C <sub>13</sub> H <sub>10</sub>	86-73-7	166.2	11.43	4.18	7.87	–	0.09	55 (~2 d)	5
Phenanthrene	phenanthrene	C <sub>14</sub> H <sub>10</sub>	85-01-8	178.2	6.173	4.57	3.24	0.691 (15°C)	0.02	55 (~2 d)	5
Fluoranthene	1,2-benzacenaphthene	C <sub>16</sub> H <sub>10</sub>	206-44-0	202.3	1.186	5.22	1.037	0.134 (15°C)	0.00123	170 (~1 wk)	5
Pyrene	pyrene	C <sub>16</sub> H <sub>10</sub>	129-00-0	202.3	0.652	5.18	0.92	0.125 (15°C)	0.0006	170 (~1 wk)	5
Benzo[a]pyrene	benzo[a]pyrene	C <sub>20</sub> H <sub>12</sub>	50-32-8	252.3	0.0151	6.04	0.046	0.0079 (10°C)	7.00 × 10 <sup>-7</sup>	170 (~1 wk)	5
Benzo[k]fluoranthene	8,9-benzofluoranthene	C <sub>20</sub> H <sub>12</sub>	207-08-9	252.3	0.00317	6.00	0.016	0.111 (15°C)	5.20 × 10 <sup>-8</sup>	170 (~1 wk)	5
Perylene	perylene	C <sub>20</sub> H <sub>12</sub>	198-55-0	252.32	0.00159	6.25	0.003	–	1.40 × 10 <sup>-8</sup>	170 (~1 wk)	5
<i>Halogenated flame retardants and surfactants</i>											
Polychlorinated biphenyls	dichlorobiphenyls	C <sub>12</sub> H <sub>8</sub> Cl <sub>2</sub>	25512-42-9	223.1	0.269-8.96	4.9-5.30	17.0-92.21	–	0.0048-0.279	170 (~1 wk)	3
	trichlorobiphenyls	C <sub>12</sub> H <sub>7</sub> Cl <sub>3</sub>	25323-68-6	257.5	0.0582-1.55	5.5-5.90	24.29-92.21	–	0.0136-0.143	550 (~3 wk)	3
	tetrachlorobiphenyls	C <sub>12</sub> H <sub>6</sub> Cl <sub>4</sub>	26914-33-0	292	0.0147-0.342	5.6-6.50	1.72-47.59	–	5.9 × 10 <sup>-5</sup> -0.0054	1700 (~2 mo)	3
	pentachlorobiphenyls	C <sub>12</sub> H <sub>5</sub> Cl <sub>5</sub>	25429-29-2	326.4	0.0123-0.0613	6.2-6.50	24.8-151.4	–	0.0003-0.0093	1700 (~2 mo)	3
	hexachlorobiphenyls	C <sub>12</sub> H <sub>4</sub> Cl <sub>6</sub>	26601-64-9	360.9	0.0011-0.002	6.7-7.30	11.9-818	–	2.0 × 10 <sup>-5</sup> -0.0015	5500 (~8 mo)	3
	heptachlorobiphenyls	C <sub>12</sub> H <sub>3</sub> Cl <sub>7</sub>	28655-71-2	395.3	0.00114-0.0051	6.7-7.0	5.4	–	2.73 × 10 <sup>-5</sup>	5500 (~8 mo)	3
	octachlorobiphenyls	C <sub>12</sub> H <sub>2</sub> Cl <sub>8</sub>	31472-83-0	429.8	0.00047-0.0007	7.1	38.08	–	2.66 × 10 <sup>-5</sup>	17000 (2 yr)	3
	nonachlorobiphenyls	C <sub>12</sub> HCl <sub>9</sub>	53742-07-7	464.2	3.8 × 10 <sup>-5</sup> -2.4 × 10 <sup>-4</sup>	7.2-8.16	–	–	–	17000 (2 yr)	3
Polybrominated diphenyl ethers	BDE47	C <sub>12</sub> H <sub>6</sub> Br <sub>4</sub> O	5436-43-1	485.2	0.0225	6	14	–	3.19 × 10 <sup>-3</sup>	–	6, 7
	BDE99	C <sub>12</sub> H <sub>5</sub> Br <sub>5</sub> O	60348-60-9	564.75	0.0042	6.5	159	–	6.82 × 10 <sup>-5</sup>	–	6, 7
	BDE209	C <sub>12</sub> Br <sub>10</sub> O	1163-19-5	959.22	2.09E-08	~10	–	–	<1 × 10 <sup>-6</sup>	–	6, 7
Short-chain chlorinated paraffins	SCCP	C <sub>10-13</sub> H <sub>16-20</sub> Cl <sub>5-9</sub> ***	85535-84-8	211.17-774.97	0.0045-6.32 × 10 <sup>-7</sup>	5.85-7.3	0.34-648	–	2.8 × 10 <sup>-7</sup> -0.5	–	8
Perfluoro-octane sulfonate	PFOS	C <sub>8</sub> F <sub>17</sub> SO <sub>3</sub>	1763-23-1	499	1363	~1	<1 × 10 <sup>-6</sup>	–	3 × 10 <sup>-4</sup>	–	9

\* Cs = water solubility of the chemical in the solid state;

\*\* Ps = vapor pressure of the pure chemical in the solid state.

\*\*\* Includes molecular formulas such as C<sub>10</sub>H<sub>20</sub>Cl<sub>2</sub>, C<sub>10</sub>H<sub>18</sub>Cl<sub>4</sub>, C<sub>10</sub>H<sub>17</sub>Cl<sub>5</sub>, C<sub>11</sub>H<sub>20</sub>Cl<sub>4</sub>, C<sub>11</sub>H<sub>19</sub>Cl<sub>5</sub>, C<sub>12</sub>H<sub>24</sub>Cl<sub>2</sub>, C<sub>12</sub>H<sub>20</sub>Cl<sub>6</sub>, C<sub>13</sub>H<sub>23</sub>Cl<sub>5</sub>, C<sub>13</sub>H<sub>16</sub>Cl<sub>12</sub>.*References*

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Annex Table 2. List of POPs measured in air at each Arctic monitoring station.

PAH	Pesticides	PCB
<i>Alert, Amderma, Dunai, and Tagish</i>		
Acenaphthylene, acenaphthalene, fluorine, phenanthrene, anthracene, fluoranthene, pyrene, retene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno[123-cd]perylene, dibenz[ah]anthracene, benzo[ghi]perylene, dibenzothiophene, perylene	1,2,3,4-tetrachlorobenzene, 1,2,4,5-tetrachlorobenzene, $\alpha$ -HCH, HCB, $\beta$ -HCH, <i>cis</i> -nonachlor, dieldrin, endrin, $\gamma$ -HCH, heptachlor, heptachloro-epoxide, methoxychlor, mirex, octachlorostyrene, <i>o,p'</i> -DDD, <i>o,p'</i> -DDE, <i>o,p'</i> -DDT, oxychlorodane, pentachloroanisole, pentachlorobenzene, photo-omirex, <i>p,p'</i> -DDD, <i>p,p'</i> -DDE, <i>p,p'</i> -DDT, tetrachloroveratrole, <i>trans</i> -chlordane, <i>trans</i> -nonachlor, trichloroveratrole, trifluralin, <i>cis</i> -chlordane, endosulfan	CB 1, CB 3, CB 4/10, CB 7, CB 6, CB 8/5, CB 19, CB 18, CB 17, CB 24/27, CB 16/32, CB 26, CB 25, CB 31, CB 28, CB 33, CB 22, CB 45, CB 46, CB 52, CB 49, CB 47, CB 48, CB 44, CB 42, CB 41/71, CB 64, CB 40, CB 74, CB 70/76, CB 66, CB 95, CB 56/60, CB 91, CB 84/89, CB 101, CB 99, CB 83, CB 97, CB 87, CB 85, CB 136, CB 110, CB 82, CB 151, CB 144/135, CB 149, CB 118, CB 134, CB 114, CB 131, CB 146, CB 153, CB 132, CB 105, CB 141, CB 179, CB 137, CB 130/176, CB 138, CB 158, CB 178/129, CB 175, CB 187, CB 183, CB 128, CB 185, CB 174, CB 177, CB 171, CB 156, CB 201/157, CB 172/197, CB 180, CB 193, CB 191, CB 200, CB 170, CB 198, CB 199, CB 196/203, CB 189, CB 195, CB 207, CB 194, CB 205, CB 206, CB 209
<i>Pallas</i>		
Anthracene, benzo[a]pyrene, benzo[ghi]perylene, phenanthrene, pyrene, chrysene-triphenylene, fluoranthene, benz[a]anthracene, indeno[123cd]pyrene, benzo[k]fluoranthene, benzo[b]fluoranthene	$\alpha$ -HCH, $\gamma$ -HCH	CB 101, CB 138, CB 153, CB 180, CB 28, CB 52, CB 118
<i>Stórhöfði</i>		
—	$\alpha$ -HCH, $\beta$ -HCH, <i>cis</i> -chlordane, dieldrin, $\gamma$ -HCH, HCB, <i>p,p'</i> -DDD, <i>p,p'</i> -DDE, <i>p,p'</i> -DDT, <i>trans</i> -chlordane, <i>trans</i> -nonachlor, <i>o,p'</i> -DDE, <i>o,p'</i> -DDT	CB 101, CB 105, CB 118, CB 138, CB 153, CB 156, CB 180, CB 28, CB 31, CB 52
<i>Zeppelin</i>		
Naphthalene, dibenzothiophene, fluorine, dibenzofuran, acenaphthene, biphenyl, phenanthrene, acenaphthylene, N1methyl-naphthalene, N2methyl-naphthalene, N2methyl-anthracene, N2methylphenanthrene, anthracene, benzo[a]fluoranthene, benzo[a]pyrene, chrysene-triphenylene, benzo[e]pyrene, benzo[b]k]fluoranthenes, benz[a]anthracene, perylene, benzo[ghi]perylene, coronene, dibenzo[ac-ah]anthracene, anthanthrene, indeno[123cd]pyrene, pyrene, retene, benzo[a]fluorine, benzo[b]fluorine, benzo[ghi]fluoranthene, cyclopenta[cd]pyrene, dibenzo[ae]pyrene, dibenzo[ai]pyrene, fluoranthene, N1methylphenanthrene, N9methylphenanthrene, dibenzo[ah]pyrene, N3methylphenanthrene	$\alpha$ -HCH, $\gamma$ -HCH, <i>trans</i> -chlordane, <i>cis</i> -chlordane, <i>trans</i> -nonachlor, <i>cis</i> -nonachlor, <i>o,p'</i> -DDE, <i>p,p'</i> -DDE, <i>o,p'</i> -DDD, <i>p,p'</i> -DDD, HCB	CB 18, CB 28, CB 31, CB 33, CB 37, CB 47, CB 52, CB 60, CB 66, CB 74, CB 99, CB 101, CB 105, CB 114, CB 118, CB 122, CB 123, CB 128, CB 138, CB 141, CB 149, CB 153, CB 156, CB 157, CB 167, CB 170, CB 180, CB 183, CB 187, CB 189, CB 194, CB 206, CB 209

Annex Table 3. Concentrations of organochlorines in air, from Ny-Ålesund (Svalbard) and Bjørnøya (Bear Island), and fog water, snow and rainwater on Bjørnøya, Norway.

	Site	Year	n	Statistic	HCB	$\Sigma$ HCHs	$\Sigma$ CHLs	$\Sigma$ PCBs	Reference
Air (pg/m <sup>3</sup> )	Zeppelin mountain (Ny-Ålesund)	1999 & 2000	2	mean range	97.9 95.2-101	39.9 35.5-44.3	2.32 1.99-2.66	4.39 1.75-7.03	1
	Meteorological station (Bjørnøya)	1999 & 2000	2	mean range	93.4 88.0-98.8	50.6 46.0-55.3	5.10 4.5-5.7	33.5 16.1-51.0	1
Fog water (pg/L)	Meteorological station (Bjørnøya)	1999 & 2000	3	mean range	27.1 10.6-56.4	459 n.d.-2090	—	5783 2930-10900	2
	Lake Ellasjøen (Bjørnøya)	1999 & 2000	2	mean range	5.25 n.d.-10.5	502 n.d.-1004	—	7350 1605-13105	2
Snow (pg/L)	Meteorological station (Bjørnøya)	2000	1	mean	2	78	—	7220	2
	Lake Ellasjøen (Bjørnøya)	2000	1	mean	1.1	32	—	3200	2
Meltwater (pg/L)	Meteorological station (Bjørnøya)	2000	2	mean range	-	1145 491-1798	—	1010 845-1180	2
	Lake Ellasjøen (Bjørnøya)	1999 & 2000	2	mean range	14.4* 14.4*	830 750-920	—	2064 391-3737	2

n.d. = not detected.

\* &lt; limit of quantification.

*References*

1. Evenset *et al.*, 2002;  $\Sigma$ HCHs = sum of  $\alpha$ - and  $\gamma$ -HCH;  $\Sigma$ CHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, chlordene, *cis*-heptachlor epoxide, and oxychlorodane;  $\Sigma$ PCBs = sum of 7 congeners (101, 105, 118, 138, 153, 156, 180).
2. Evenset *et al.*, 2002 as in footnote 1, except  $\Sigma$ PCBs = sum of 17 congeners (28/31, 52, 99, 101, 105, 118, 128, 138/163, 149, 153, 156, 169, 170, 180, 183, 187, 194).

Annex Table 4. Mean ± standard deviation (SD) concentrations of organochlorines (ng/g dw, except pg/g for PCDD/Fs) in terrestrial soil and vegetation.

Soil/species	Location	Year	Statistic	n	% org C	ΣCBz	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Mirex	ΣPBDEs	PCDD/Fs (pg/g)	Reference
<b>Soil</b>																
Surface soil	Alaska, USA	1991-93	mean±SE	16	–	0.25±0.08	0.18±0.08	0.19±0.16	19.5±19.5	13.2±12.4	–	–	–	–	–	1
”	Kola Peninsula, Russia	1991-93	mean±SE	12	–	1.03±0.48	1.54±0.93	0.22±0.14	3.82±2.67	2.21±1.34	–	–	–	–	–	1
”	Taymir Peninsula, Russia	1991-93	mean±SE	7	–	n.d.	n.d.	n.d.	n.d.	0.02±0.02	–	–	–	–	–	1
Peat, peat litter, litter, loamy sand & sand	Lovozero, Kola Peninsula	2000-01	mean±SE	5	–	0.74±0.30	1.19±0.70	0.18±0.05	1.72±0.76	4.07±0.59	–	<0.01	0.20±0.18	<0.05	–	2
Soil	Nelmin Nos, Pechora Basin	2000-01	–	1	–	0.67	0.22	0.14	0.62	3.17	–	<0.01	0.07	<0.05	–	2
”	Dudinka, Taymir Peninsula	2000-01	–	1	–	0.18	0.19	<0.05	0.34	2.91	–	<0.01	0.10	<0.05	–	2
”	Khatanga, Taymir Peninsula	2000-01	mean	2	–	0.58	0.18	<0.05	0.74	2.43	–	<0.05	0.08	<0.02	–	2
”	Kanchalan, Chukotka Peninsula	2000-01	–	1	–	0.75	0.77	0.17	1.06	5.2	–	<0.01	0.41	0.23	–	2
”	Lavrentiya, Chukotka Peninsula	2000-01	–	1	–	0.9	0.46	0.08	0.92	12.9	–	<0.05	0.22	0.16	–	2
”	Norway, 67°00'N,15°98'E	1998-2000	–	1	15.1	–	–	–	–	406.1	253.3	–	–	–	–	3
”	Norway, 67°00'N,15°98'E	1998-2001	–	1	94.4	–	–	–	–	1544.8	774.3	–	–	–	–	3
”	Norway, 67°38'N,14°67'E	1998-2002	–	1	5.9	–	–	–	–	207.1	120.5	–	–	–	–	3
”	Norway, 67°38'N,14°67'E	1998-2003	–	1	20.3	–	–	–	–	1286.0	715.0	–	–	–	–	3
”	Russia, 67°44'N,63°76'E	1998-2004	–	1	82.4	–	–	–	–	10357.1	5605.5	–	–	–	–	3
”	Russia, 67°45'N,63°72'E	1998-2005	–	1	84.4	–	–	–	–	6188.4	3324.3	–	–	–	–	3
”	Russia, 67°61'N,53°16'E	1998-2006	–	1	41.5	–	–	–	–	46.5	0.0	–	–	–	–	3
”	Russia, 67°83'N,32°79'E	1998-2007	–	1	48.3	–	–	–	–	10184.0	5567.2	–	–	–	–	3
”	Russia, 67°86'N,32°79'E	1998-2008	–	1	59.5	–	–	–	–	48277.0	25065.1	–	–	–	–	3
”	Norway, 67°97'N,15°95'E	1998-2009	–	1	80.7	–	–	–	–	5415.9	3085.1	–	–	–	–	3
”	Greenland, 68°00'N,32°00'W	1998-2010	–	1	0.2	–	–	–	–	4.2	2.7	–	–	–	–	3
”	Norway, 69°12'N,19°75'E	1998-2011	–	1	12.7	–	–	–	–	242.0	163.1	–	–	–	–	3
”	Norway, 69°12'N,19°75'E	1998-2012	–	1	82.7	–	–	–	–	1723.2	998.4	–	–	–	–	3
”	Norway, 69°78'N,18°63'E	1998-2013	–	1	89.6	–	–	–	–	5146.6	3026.6	–	–	–	–	3
”	Norway, 69°83'N,25°17'E	1998-2014	–	1	94.7	–	–	–	–	3074.4	1683.7	–	–	–	–	3
”	Norway, 70°47'N,27°97'E	1998-2015	–	1	96.3	–	–	–	–	2540.7	1325.2	–	–	–	–	3
”	Norway (Bjørnøya), 75°00'N,19°00'E	1998-2016	–	1	3.6	–	–	–	–	17.2	16.2	–	–	–	–	3
”	Norway (Bjørnøya), 75°00'N,19°00'E	1998-2017	–	1	4.3	–	–	–	–	18.1	15.8	–	–	–	–	3
”	Norway (Bjørnøya), 75°00'N,19°00'E	1998-2018	–	1	7.3	–	–	–	–	10.3	10.3	–	–	–	–	3
”	Norway (Bjørnøya), 75°00'N,19°00'E	1998-2019	–	1	4.3	–	–	–	–	4.4	1.8	–	–	–	–	3
”	Canada, 78°88'N,76°00'W	1998-2020	–	1	31.5	–	–	–	–	3397.4	1855.0	–	–	–	–	3
”	Canada, 81°75'N,64°92'W	1998-2021	–	1	5.8	–	–	–	–	62.6	38.4	–	–	–	–	3
”	Alaska, 60°5'N,161°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	316	4
”	Yukon, 65°3'N,138°2'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	6.21	4
”	Yukon, 66°3'N,136°8'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	13	4
”	Yukon, 66°3'N,136°8'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	28	4
”	Yukon, 62°9'N,136°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	246	4
”	Yukon, 66°9'N,136°4'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	21	4
”	Yukon, 60°2'N,132°9'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	249	4
”	NWT, 62°5'N,114°2'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	39	4
”	W Greenland, 69°4'N,53°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	229	4
”	W Greenland, 69°4'N,53°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	126	4
”	Norway, 69°0'N,19°0'E	1998	–	1	–	–	–	–	–	–	–	–	–	–	134	4
<b>Vegetation</b>																
Tree bark	Alaska, 60°5'N,161°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	9.2	4
”	Yukon, 65°3'N,138°2'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	2.1	4
”	Yukon, 66°3'N,136°8'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	3.7	4
”	Yukon, 62°9'N,136°5'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	2.4	4
”	Yukon, 66°9'N,136°4'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	4.7	4
”	Yukon, 60°2'N,132°9'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	17	4
”	Yukon, 60°4'N,129°1'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	<0.1	4
”	NWT, 62°5'N,114°2'W	1998	–	1	–	–	–	–	–	–	–	–	–	–	10	4
”	Norway, 69°0'N,19°0'E	1998	–	1	–	–	–	–	–	–	–	–	–	–	5	4
<i>Cassiope tetragona</i>	Alaska	1991-93	–	5	–	n.d.	n.d.	n.d.	n.d.	n.d.	–	–	–	–	–	1
<i>Carex</i> spp.	Alaska	1991-93	–	1	–	n.d.	n.d.	n.d.	n.d.	n.d.	–	–	–	–	–	1
<i>Dryas octopetala achenes</i>	Alaska	1991-93	mean	2	–	n.d.	n.d.	n.d.	n.d.	n.d.	–	–	–	–	–	1
<i>Epilobium latifolium</i>	Alaska	1991-93	–	1	–	0.10	1.48	0.15	0.12	1.96	–	–	–	–	–	1
Mushroom	Alaska	1991-93	–	1	–	n.d.	n.d.	n.d.	n.d.	n.d.	–	–	–	–	–	1
<i>Oxytropis</i> spp.	Alaska	1991-93	–	1	–	n.d.	0.12	n.d.	n.d.	n.d.	–	–	–	–	–	1
<b>Berries</b>																
<i>Vaccinium uliginosum</i> (blueberry)	Alaska	1991-93	mean±SE	6	–	0.07±0.04	0.43±0.11	0.77±0.24	0.11±0.08	2.81±1.29	–	–	–	–	–	1
<i>Vaccinium uliginosum</i>	Dudinka, Taymir Peninsula	2000-01	–	1 pool (n=20)	–	0.72	n.d.	0.14	1.04	4.25	–	<0.02	<0.10	<0.05	–	2
<i>Vaccinium vitis-idaea</i> , <i>V. uliginosum</i>	Kanchalan, Chukotka Peninsula	2000-01	mean	2 pools (n=8,10)	–	0.54	0.19	n.d.	0.13	1.15	–	<0.02	<0.10	<0.05	–	2
<i>Vaccinium vitis-idaea</i> , <i>V. uliginosum</i>	Khatanga, Taymir Peninsula	2000-01	mean	2 pools (n=8,10)	–	0.32	0.18	n.d.	0.79	1.55	–	<0.02	0.20	<0.05	–	2
<i>Rubus chamaemorus</i> , <i>Empetrum nigrum</i>	Lavrentiya, Chukotka Peninsula	2000-01	mean	2 pools (n=8,12)	–	0.16	n.d.	n.d.	0.05	1.24	–	<0.02	<0.10	<0.05	–	2
<i>Vaccinium vitis-idaea</i> , <i>V. myrtilis</i>	Lovozero, Kola Peninsula	2000-01	mean	2 pools (n=4,16)	–	0.20	0.06	0.08	1.50	1.54	–	<0.02	<0.10	<0.05	–	2
<i>Vaccinium vitis-idaea</i> , <i>V. myrtilis</i>	Nelmin Nos, Pechora Basin	2000-01	mean	2 pools (n=10)	–	0.26	0.16	<0.1	0.21	1.59	–	<0.02	<0.10	<0.05	–	2

continued next page

Annex Table 4 continued.

Soil/species	Location	Year	Statistic	n	% org C	ΣCBz	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Mirex	ΣPBDEs	PCDD/Fs (pg/g)	Reference
<b>Lichens</b>																
<i>Masonhalea richardsonii</i>	Alaska	1991-93	mean±SE	36	–	0.16±0.05	0.58±0.08	0.05±0.01	0.14±0.05	1.47±1.14	–	–	–	–	–	1
<i>Cetraria cucullata</i>	Alaska, USA	1991-93	mean±SE	32	–	1.20±0.13	4.23±0.32	0.08±0.02	0.03±0.01	4.09±0.71	–	–	–	–	–	1
	Kola Peninsula	1991-93	mean±SE	8	–	1.19±0.18	6.26±0.78	0.19±0.04	0.34±0.09	2.56±0.26	–	–	–	–	–	1
	Taymir Peninsula	1991-93	mean±SE	9	–	3.05±0.30	7.98±0.83	0.07±0.01	0.18±0.02	8.41±1.27	–	–	–	–	–	1
<i>Cetraria islandica</i>	Alaska	1991-93	mean	2	–	0.15	0.43	0.05	0.04	0.29	–	–	–	–	–	1
	Kola Peninsula	1991-93	mean±SE	8	–	0.26±0.10	1.87±0.29	0.15±0.03	0.32±0.07	0.71±0.11	–	–	–	–	–	1
<i>Cetraria islandica, Cladonia rangiferina</i>	Dudinka, Taymir Peninsula	2000-01	mean	2 pools (n=10)	–	1.28	1.37	0.18	3.04	3.36	–	<0.02	0.18	<0.05	–	2
<i>Cetraria cucullata, Cladonia rangiferina</i>	Kanchalan, Chukotka Peninsula	2000-01	mean	2 pools (n=10)	–	0.95	0.88	n.d.	1.29	3.33	–	<0.02	<0.10	<0.05	–	2
<i>Cladonia rangiferina, Cetraria islandica</i>	Khatanga, Taymir Peninsula	2000-01	mean	2 pools (n=10)	–	2.09	1.63	0.47	2.83	3.38	–	<0.02	0.29	<0.05	–	2
<i>Cetraria cucullata</i>	Lavrentiya, Chukotka Peninsula	2000-01	–	1 pool (n=20)	–	0.70	0.76	n.d.	1.13	3.81	–	<0.02	<0.10	<0.05	–	2
<i>Cladonia rangiferina, C. alpica, C. islandica, C. stellaris, C. mitis</i>	Lovozero, Kola Peninsula	2000-01	mean±SE	4 pools (n=3-6)	–	1.15±0.23	0.44±0.11	0.05±0.05	0.89±0.14	3.76±0.16	–	<0.02	<0.10	<0.05	–	2
	Nelmin Nos, Pechora Basin	2000-01	–	1 pool (n=20)	–	2.12	0.64	0.29	2.07	3.06	–	<0.02	0.52	<0.05	–	2
<b>Mosses</b>																
<i>Racomitrium lanuginosum</i>	Alaska	1991-93	mean±SE	25	–	0.07±0.02	1.19±0.28	0.19±0.09	0.04±0.01	0.61±0.21	–	–	–	–	–	1
	Kola Peninsula	1991-93	mean	2	–	n.d.	8.59±2.61	0.52±0.25	1.92±0.36	4.75±2.33	–	–	–	–	–	1
	Taymir Peninsula	1991-93	mean±SE	3	–	0.32±0.16	2.20±0.75	0.35±0.05	0.10±0.01	1.07±0.68	–	–	–	–	–	1
<i>Hylocomium splendens</i>	Alaska	1991-93	mean±SE	33	–	0.24±0.02	1.24±0.16	0.15±0.03	0.06±0.02	1.57±0.39	–	–	–	–	–	1
	Taymir Peninsula	1991-93	mean±SE	6	–	0.35±0.11	1.59±0.48	0.14±0.05	0.06±0.03	10.6±2.98	–	–	–	–	–	1
<i>Hylocomium splendens</i>	Nelmin Nos, Pechora Basin	2000-01	–	1 pool (n=20)	–	1.15	1.38	0.33	2.24	7.45	–	<0.02	0.44	<0.05	–	2
<i>Hylcomium splendens, Sphagnum balticum</i>	Dudinka, Taymir Peninsula	2000-01	mean	2 pools (n=6,14)	–	1.68	2.32	0.69	2.47	11.7	–	<0.02	0.47	<0.05	–	2
<i>Hylocomium splendens, Sphagnum balticum</i>	Khatanga, Taymir Peninsula	2000-01	mean	2 pools (n=10)	–	1.50	1.94	0.46	2.94	10.2	–	<0.02	0.19	<0.05	–	2
<i>Dicranum sp., Sphagnum balticum</i>	Kanchalan, Chukotka Peninsula	2000-01	mean	2 pools (n=10)	–	1.17	0.69	n.d.	1.32	13.0	–	<0.02	<0.10	<0.05	–	2
<i>Dicranum sp.</i>	Lavrentiya, Chukotka Peninsula	2000-01	–	1 pool (n=20)	–	0.60	0.36	n.d.	0.92	13.8	–	<0.02	<0.10	<0.05	–	2
<i>Sphagnum magellanicum, Pleurozium schreberi &amp; Politrichum commune</i>	Lovozero, Kola Peninsula	2000-01	mean	3 pools (n=3-7)	–	0.37	0.71	n.d.	1.21	12.5	–	<0.02	0.15	<0.05	–	2

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

## References

- Ford *et al.*, 2000; CBz = HCB; ΣHCHs = α-, β-, and γ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide; ΣDDTs = *o,p'*- and *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT; ΣPCBs = sum of 22 congeners (8, 18, 28, 29, 44, 50, 52, 66, 87, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 200, 206, 209).
- RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62; ΣPBDEs = sum of 2,2',4,4'-tetrabromodiphenyl, 2,2',4,4', 5-pentabromodiphenyl and 2,2',4,4',5'-pentabromodiphenyl ethers.
- Ockenden *et al.*, 2002. ΣPCBs = sum of 27 congeners (18, 31, 28, 52, 49, 44, 70, 95, 90/101, 99, 87, 110, 123, 118, 151, 149, 153/132, 141, 138, 158, 187, 183, 180, 170, 199, 203, 194), ΣPCB<sub>10</sub> = sum of 31, 28, 52, 101, 105, 118, 153/132, 138, 156, 180).
- Wagrowski and Hites, 2000.



Annex Table 5. Mean ± standard deviation (SD) concentrations of organochlorines in terrestrial mammals and birds.

Species/Location	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Heptachlor epoxide	Oxy-chlordane	ΣPBDEs	Reference
<b>Terrestrial mammals (ng/g ww)</b>																			
<i>Arctic hare (Lepus timidus)</i>																			
Lovozero, Kola Peninsula	kidney	2000-01	m,f	–	2 pools (n=4,6)	mean	0.06	0.23	0.05	0.10	0.61	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=4,6)	mean	0.43	0.43	0.14	0.38	0.93	–	<0.02	–	≤0.06	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=4,6)	mean	0.29	0.32	0.05	0.07	0.84	–	<0.02	–	<0.05	–	–	<0.05	1
Nelmin Nos, Pechora Basin	kidney	2000-01	m,f	–	2 pools (n=7,8)	mean	0.05	0.25	0.06	0.10	0.70	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=7,8)	mean	0.48	0.55	0.17	0.38	1.12	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=7,8)	mean	0.31	0.42	0.07	0.05	0.77	–	<0.02	–	<0.05	–	–	<0.05	1
Dudinka, Taymir Peninsula	kidney	2000-01	m,f	–	2 pools (n=5,10)	mean	0.17	0.18	n.d.	0.12	0.27	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=5,10)	mean	0.40	0.46	0.18	0.40	0.47	–	<0.02	–	≤0.12	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=5,10)	mean	0.31	0.17	0.05	0.08	0.48	–	<0.02	–	≤0.06	–	–	<0.05	1
Khatanga, Taymir Peninsula	kidney	2000-01	m,f	–	2 pools (n=6,8)	mean	0.05	0.26	0.05	0.09	0.21	–	<0.02	–	0.07	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=6,8)	mean	0.55	0.47	0.31	0.30	0.35	–	<0.02	–	0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=6,8)	mean	0.23	0.38	0.07	0.06	0.43	–	<0.02	–	0.07	–	–	<0.05	1
Kanchalan, Chukotka Peninsula	kidney	2000-01	m,f	–	2 pools (n=4,6)	mean	0.15	0.27	0.08	0.13	0.38	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=4,6)	mean	0.62	0.66	0.31	0.47	0.60	–	<0.02	–	0.09	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=4,6)	mean	0.39	0.24	0.08	0.06	0.54	–	<0.02	–	<0.05	–	–	<0.05	1
Qeqertarsuaq, SW Greenland	kidney	1999	m,f	32.5±18.8	5	mean±SD	2.46±1.65	0.46±0.27	0.92±0.67	0.18±0.18	6.09±3.78	0.80±0.76	2.63±0.08	–	–	–	–	–	2
„	liver	1999	m,f	4.1±0.5	5	mean±SD	0.22±0.05	0.05±0.02	3.41±2.19	0.05±0.06	0.44±0.14	0.10±0.04	0.48±0.30	–	–	–	–	–	2
„	muscle	1999	m,f	3.8±2.7	5	mean±SD	0.36±0.20	0.05±0.03	0.16±0.09	0.01±0.02	1.55±1.08	0.11±0.08	0.04±0.04	–	–	–	–	–	2
Faroe Islands, Denmark	liver	1999	juveniles	2.12±0.87	1 pool (n=4)	mean±SD	0.93±0.94	–	0.76±0.21	0.14±0.05	0.09±0.01	–	n.d.	–	0.07±0.02	–	0.76±0.21	–	3
„	„	„	adults f	1.94±0.57	1 pool (n=4)	mean±SD	0.50±0.38	–	0.40±0.18	0.14±0.04	0.06±0.03	–	n.d.	–	n.d.	–	0.40±0.18	–	3
„	„	„	adults m	1.77±0.41	1 pool (n=5)	mean±SD	0.45±0.22	–	0.46±0.14	0.10±0.05	0.08±0.03	–	n.d.	–	0.05±0.02	–	0.46±0.14	–	3
<i>Sheep/lamb (Ovis spp.)</i>																			
Narsaq, SW Greenland	fat	1999	m,f	90.2±3.1	5	mean±SD	1.21±0.29	0.22±0.09	0.28±0.07	1.20±0.65	7.13±10.9	1.85±0.75	<0.1	–	–	–	–	–	2
„	kidney	1999	m,f	4.2±0.4	5	mean±SD	0.21±0.05	0.13±0.04	0.03±0.02	0.11±0.06	2.31±1.82	0.51±0.33	<0.1	–	–	–	–	–	2
„	liver	1999	m,f	9.6±2.0	5	mean±SD	0.51±0.13	0.15±0.08	0.08±0.04	0.39±0.26	5.33±7.45	2.04±1.93	<0.1	–	–	–	–	–	2
„	muscle	1999	m,f	12.3±9.8	5	mean±SD	0.59±0.32	0.08±0.04	0.11±0.05	0.33±0.12	1.19±0.81	0.71±0.39	<0.1	–	–	–	–	–	2
Faroe Islands, Denmark	liver	1997	–(lambs)	7.50±0.15	17	mean	0.75	–	n.d.	0.225	0.375	–	n.d.	–	–	–	–	–	4
„	tallow near kidney	1997	–(lambs)	89.0±0.04	17	mean	3.56	–	n.d.	n.d.	0.445-1.78	–	n.d.	–	–	–	–	–	4
„	liver	1997	–(sheep)	7.13±0.65	8	mean	0.713	–	n.d.	0.428	0.143	–	n.d.	–	–	–	–	–	4
„	tallow near kidney	1997	–(sheep)	88.3±0.07	8	mean	3.53	–	n.d.	n.d.	0.441-1.77	–	n.d.	–	–	–	–	–	4
<i>Muskox (Ovibos moschatus)</i>																			
Kangerlussuaq, SW Greenland	fat	1999	m,f	72.1±39.8	5	mean±SD	3.43±1.94	0.53±0.27	0.36±0.22	0.32±0.25	2.99±1.40	1.54±0.82	<0.1	–	–	–	–	–	2
„	kidney	1999	m,f	3.2±0.6	5	mean±SD	0.43±0.10	0.17±0.03	0.1±0.02	0.06±0.04	1.08±0.96	0.40±0.41	<0.1	–	–	–	–	–	2
„	liver	1999	m,f	10.3±1.8	5	mean±SD	1.24±0.20	0.59±0.07	0.68±0.30	0.11±0.02	2.22±0.44	0.83±0.21	<0.1	–	–	–	–	–	2
„	muscle	1999	m,f	15.3±29.8	5	mean±SD	2.72±5.49	0.31±0.51	0.19±0.37	0.11±0.22	3.23±5.83	1.46±2.77	<0.1	–	–	–	–	–	2
<i>Reindeer (Rangifer tarandus)</i>																			
Finland	fat	2000	–	–	10	mean±SD	14.3±4.44	n.d.	5.0	6.14±1.92	3.5±8.8	–	–	–	–	–	–	–	5
Lovozero, Kola Peninsula	kidney	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.19±0.03	0.19±0.07	0.09±0.03	0.41±0.05	0.67±0.06	–	<0.02	–	0.12±0.03	–	–	<0.05	1
„	liver	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.67±0.06	1.07±0.20	0.22±0.05	0.85±0.14	1.47±0.17	–	<0.02	–	0.11±0.01	–	–	<0.05	1
„	muscle	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.54±0.06	0.27±0.07	0.08±0.03	0.43±0.07	1.28±0.24	–	<0.02	–	<0.05	–	–	<0.05	1
Nelmin Nos, Pechora Basin	kidney	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.18±0.03	0.35±0.09	0.06±0.03	0.38±0.04	0.76±0.07	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.69±0.09	1.05±0.11	0.16±0.05	0.68±0.13	1.78±0.16	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	6 pools (n=1-5)	mean±SE	0.43±0.04	0.28±0.03	0.04±0.02	0.37±0.05	1.78±0.27	–	<0.02	–	<0.05	–	–	<0.05	1
Dudinka, Taymir Peninsula	kidney	2000-01	m,f	–	5 pools (n=1-3)	mean±SE	0.18±0.03	0.18±0.04	0.07±0.03	0.31±0.08	0.53±0.06	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	5 pools (n=1-3)	mean±SE	0.84±0.04	0.79±0.13	0.10±0.07	0.51±0.11	1.31±0.23	–	<0.02	–	0.09	–	–	<0.05	1
„	muscle	2000-01	m,f	–	5 pools (n=1-3)	mean±SE	0.32±0.04	0.18±0.01	0.03±0.02	0.34±0.09	1.35±0.19	–	<0.02	–	<0.05	–	–	<0.05	1
Khatanga, Taymir Peninsula	kidney	2000-01	m,f	–	4 pools (n=1-3)	mean±SE	0.14±0.02	0.38±0.09	0.08±0.06	0.38±0.06	0.34±0.07	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	4 pools (n=1-3)	mean±SE	0.67±0.10	0.73±0.12	0.20±0.07	0.42±0.07	0.70±0.15	–	<0.02	–	0.13	–	–	<0.05	1
„	muscle	2000-01	m,f	–	4 pools (n=1-3)	mean±SE	0.32±0.04	0.31±0.05	0.09±0.01	0.45±0.04	0.67±0.18	–	<0.02	–	<0.05	–	–	<0.05	1
Kanchalan, Chukotka Peninsula	kidney	2000-01	m,f	–	2 pools (n=4,6)	mean	0.47	0.93	n.d.	0.46	0.37	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=4,6)	mean	1.78	1.75	0.23	1.34	0.89	–	<0.02	–	£0.06	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=4,6)	mean	0.66	0.89	0.20	0.41	0.62	–	<0.02	–	0.18	–	–	<0.05	1
Lavrentiya, Chukotka Peninsula	kidney	2000-01	m,f	–	2 pools (n=4,6)	mean	0.60	0.87	<0.05	0.55	0.42	–	<0.02	–	<0.05	–	–	<0.05	1
„	liver	2000-01	m,f	–	2 pools (n=4,6)	mean	2.05	1.56	0.18	1.32	0.88	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=4,6)	mean	0.76	0.87	0.17	0.42	0.77	–	<0.02	–	<0.05	–	–	<0.05	1
<i>Wolf (Canis lupus)</i>																			
Tver & Smoliensk, Russia	liver	–	m,f	4.89	58	median range	–	–	–	–	25	–	–	13.1	–	–	–	–	6
„	„	„	„	1.22-63.4	„	„	„	„	„	„	n.d.-559	–	–	(n.d.-79.8)	–	–	–	–	6
Watson Lake & Haines Junction, Yukon	liver	1993-94	m, <18mo	4.1	1 pool (n=3)	–	4.2	n.d.	0.50	n.d.	0.7	–	–	n.d.	n.d.	–	–	–	7
„	liver	1993-94	m, 19-36 mo	5.3	1 pool (n=2)	–	2.0	n.d.	4.9	n.d.	8.1	–	–	0.3	n.d.	–	–	–	7
„	liver	1993-94	f, 19-36 mo	3.7	1 pool (n=7)	–	2.6	n.d.	1.5	n.d.	9.5	–	–	1.5	n.d.	–	–	–	7
„	liver	1993-94	m, >36 mo	4.9	1 pool (n=5)	–	7.2	n.d.	2.4	n.d.	5.2	–	–	0.4	n.d.	–	–	–	7
„	liver	1993-94	f, >36 mo	4.4	1 pool (n=3)	–	3.2	n.d.	2.8	n.d.	15.1	–	–	0.8	n.d.	–	–	–	7
<i>W</i>																			

Annex Table 5 continued.

Species/Location	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Heptachlor epoxide	Oxy-chlordane	ΣPBDEs	Reference
<b>Birds (ng/g ww)</b>																			
American peregrine falcon ( <i>Falco peregrinus anatum</i> )																			
Alaska	eggs	1979-84			31	geo.mean range	–	–	–	10700 4300-30700	2700 800-28000	–	–	200 n.d.-700	–	300 n.d.-3300	100 n.d.-1000	–	9
„	eggs	1988-90			26	geo.mean range	**	**	–	5030 1690-17000	2000.0 700-15000	–	–	80 13-1187	**	130 n.d.-730	80 30-330	–	9
„	eggs	1991-95			32	geo.mean range	30** n.d.-1020	30** n.d.-390	–	3410 480-14100	1600 400-8500	–	–	70 10-360	130** 20-540	50 10-570	50 20-130	–	9
Arctic peregrine falcon ( <i>Falco peregrinus tundrius</i> )																			
Alaska	eggs	1979-84			19	geo.mean range	–	–	–	9400 1500-46400	2100 600-6300	–	–	300 n.d.-1700	–	300 n.d.-1900	100 30-300	–	9
„	eggs	1988-90			29	geo.mean range	**	**	–	3170 0610-10300	2100 700-14800	–	–	110 20-570	**	150 n.d.-880	90 40-200	–	9
„	eggs	1991-95			20	geo.mean range	20** n.d.-1280	30** n.d.-500	–	3040 1230-13300	1300 600-6000	–	–	100.00 20-320	130** 30-530	60 20-160	60 20-140	–	9
Peregrine falcon ( <i>Falco peregrinus</i> )																			
Norway	eggs	1991-97	–	–	5	mean range	*	–	3120 1530-5470	* 2270-25600	10600 2270-25600	–	8.0 n.d.-32	*	–	*	*	–	10
Merlin ( <i>Falco columbarius</i> )																			
Norway	eggs	1991-97	–	–	3	mean range	*	–	3050 2360-3520	* 1400-1970	1650 1400-1970	–	n.d.	*	–	*	*	–	10
Gyrfalcon ( <i>Falco rusticolus</i> )																			
Norway	eggs	1991-97	–	–	3	mean range	*	–	353 102-766	* 285-5310	2020 285-5310	–	7.0 n.d.-21	*	–	*	*	–	10
Golden eagle ( <i>Aquila chrysaetos</i> )																			
Norway	eggs	1991-97	–	–	12	mean range	*	–	602 36-1900	* 264-4720	1450 264-4720	–	30 n.d.-131	*	–	*	*	–	10
Willow grouse ( <i>Lagopus lagopus</i> )																			
Lovozero, Kola Peninsula	liver	2001	m,f	–	2 pools (n=9,12)	mean	0.43	0.85	0.20	1.64	1.62	–	<0.02	–	0.06	–	–	<0.05	1
„	muscle	2001	m,f	–	2 pools (n=9,12)	mean	0.25	0.26	n.d.	0.61	1.70	–	<0.02	–	<0.05	–	–	<0.05	1
Khatanga, Taymir Peninsula	liver	2000-01	m,f	–	2 pools (n=2,18)	mean	0.58	0.57	0.24	1.18	0.94	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=2,18)	mean	0.23	0.24	n.d.	0.54	0.90	–	<0.02	–	<0.05	–	–	<0.05	1
Kanchalan, Chukotka Peninsula	liver	2000-01	m,f	–	2 pools (n=9,11)	mean	0.92	1.60	0.38	1.82	1.28	–	<0.02	–	0.09	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=9,11)	mean	0.56	0.34	n.d.	0.76	1.13	–	<0.02	–	<0.05	–	–	<0.05	1
Lavrentiya, Chukotka Peninsula	liver	2000-01	m,f	–	2 pools (n=6,14)	mean	0.80	0.82	0.40	1.65	1.24	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=6,14)	mean	0.46	0.34	n.d.	0.85	1.12	–	<0.02	–	<0.05	–	–	<0.05	1
Ptarmigan ( <i>Lagopus mutus</i> )																			
Nuuk, SW Greenland	liver	1999	m,f	6.8±0.5	5	mean±SD	0.79±0.70	0.16±0.03	0.68±0.43	0.69±0.46	18.3±13.6	1.55±1.12	0.73±0.45	–	–	–	–	–	2
„	muscle	1999	m,f	3.8±0.7	5	mean±SD	0.22±0.08	0.22±0.12	1.66±0.06	0.04±0.02	9.02±4.16	1.76±0.62	0.69±0.49	–	–	–	–	–	2
Nelmin Nos, Pechora Basin	liver	2001	m,f	–	2 pools (n=8,12)	mean	0.48	0.60	0.18	1.55	1.77	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2001	m,f	–	2 pools (n=8,12)	mean	0.11	0.24	n.d.	0.61	1.47	–	<0.02	–	<0.05	–	–	<0.05	1
Dudinka, Taymir Peninsula	liver	2000-01	m,f	–	2 pools (n=8,12)	mean	0.50	0.76	0.24	1.25	0.86	–	<0.02	–	<0.05	–	–	<0.05	1
„	muscle	2000-01	m,f	–	2 pools (n=8,12)	mean	0.22	0.23	n.d.	0.05	0.80	–	<0.02	–	<0.05	–	–	<0.05	1

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.\* Data for sum of pesticides; sum of HCB, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor epoxide, *p*, *p'*-DDE, and dieldrin are summed and listed together under ΣCHLs.

\*\* 1988-90 and 1991-95 data were combined for ΣHCHs, ΣCBz, and mirex.

## References

- RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- & *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62; ΣPBDEs = sum of 2,2',4,4'-tetrabromodiphenyl, 2,2',4,4',5-pentabromodiphenyl and 2,2',4,4',5'-pentabromodiphenyl ethers.
- Muir and Johansen, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 106 congeners peaks (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 3, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Hoydal *et al.*, 2001; ΣCBz = HCB; ΣCHLs = oxychlordane, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = PCB 153.
- Larsen and Dam, 1999; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 153 and 138.
- Hirvi and Hentonen, 2002; ΣCBz = HCB; ΣHCHs = γ-HCH; ΣCHLs = oxychlordane, *cis*- and *trans*-chlordane, *trans*-nonachlor, heptachlor, and heptachlor epoxide; ΣDDTs = *p,p'*-DDT, *o,p'*- and *p,p'*-DDT, -DDE, and -DDD; ΣPCBs = sum of 15 congeners (8, 15, 28, 52, 101, 114, 118, 128, 138, 141, 149, 151, 153, 170, 180).
- Shore *et al.*, 2001; ΣPCBs = as Aroclor 1254.
- Gamberg and Braune, 1999; ΣCBz = sum of di-, tri-, tetra-, penta-, and hexa-chlorobenzene; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 101 congeners (8/5, 15, 16/32, 17, 18, 19, 22, 24/27, 25, 26, 31/28, 33, 40, 41/71/64, 42, 44, 45, 46, 47/48, 49, 52, 56/60, 66, 70/76, 74, 83, 84/89, 85, 87, 90/101, 91, 95, 97, 99, 105, 107, 110, 114, 118, 128, 129, 130, 131, 134, 136, 137, 138/163/164, 141, 144/135, 146, 149, 151, 153, 156, 157, 158, 170/190, 171, 172, 174, 175, 176, 177, 178, 179, 180, 183, 185, 187/182, 189, 191, 193, 194, 195, 196/203, 197, 198, 199, 201, 205, 206, 207, 208, 209).
- Braune *et al.*, 2001d; ΣCBz = 1,3-,1,4-,1,2-diCBz, 1,3,5-,1,2,4-,1,2,3-triCBz, 1,2,3,4-tetraCBz, and HCBz; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor, and heptachlor epoxide; ΣDDTs = *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 106 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Ambrose *et al.*, 2000; ΣCBz = HCB; ΣHCHs = α-, β-, and γ-HCH; ΣDDTs = *p*, *p'*-DDE; ΣPCBs = Aroclor sum.
- Herzke *et al.*, 2002; ΣPCBs = sum of 15 congeners (99, 101, 105, 118, 126, 128, 138, 149, 153, 169, 170, 180, 183, 187, 194); Toxaphene = Parlars 26, 32, 38, 50, 51, 58, 62, 69.

Annex Table 6. Mean concentrations of organochlorines in freshwater, and lake and river sediments.

Species/Location	Region	Approx. Lat/long	Year	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Reference
<b>Water (ng/L)</b>														
Lake Lovozero	Lovozero, Kola Peninsula	68°00'N, 34°30'E	2000-01	4 pools (n=3-9)	mean±SE	0.65±0.17	0.23±0.15	0.41±0.02	0.17±0.02	1.85±0.36	3.00±0.49	–	<0.01	1
Pechora River	Nelmin Nos, Pechora Basin	68°00'N, 54°00'E	2000-01	3 pools (n=4)	mean±SE	0.30±0.01	0.23±0.04	0.21±0.05	0.14±0.03	0.85±0.10	1.93±0.20	–	<0.01	1
Yenisey River	Dudinka, Taymir Peninsula	69°00'N, 86°00'E	2000-01	2 pools (n=4)	–	0.44	0.24	0.28	0.08	0.31	1.24	–	<0.01	1
Khatanga River	Khatanga, Taymir Peninsula	72°00'N, 102°00'E	2000-01	2 pools (n=4)	–	0.66	0.27	0.33	0.03	0.91	2.07	–	<0.01	1
Kanchalan River	Kanchalan, Chukotka	65°20'N, 176°30'E	2000-01	1 pool (n=8)	–	0.30	0.30	1.01	<0.05	0.44	1.72	–	<0.05	1
<b>River sediments (ng/g dw)</b>														
Pechora River	Nelmin Nos, Pechora Basin	68°00'N, 54°00'E	2000-01	1 pool (n=10)	–	0.56	0.21	0.96	0.26	69.4	2.38	–	<0.01	1
Yenisey River	Dudinka, Taymir Peninsula	69°00'N, 86°00'E	2000-01	1 pool (n=10)	–	0.44	0.18	0.11	0.13	0.85	2.48	–	<0.01	1
Khatanga River	Khatanga, Taymir Peninsula	72°00'N, 102°00'E	2000-01	1 pool (n=10)	–	0.48	0.14	0.13	0.13	1.02	2.39	–	<0.01	1
Kanchalan River	Kanchalan, Chukotka	65°20'N, 176°30'E	2000-01	1 pool (n=10)	–	0.53	0.21	0.09	<0.05	0.16	1.58	–	<0.05	1
<b>Lake sediments</b>														
Bjørnøya, Norway	Lake Ellasjøen (0-1 cm)	74°30'N, 19°00'E	1996	1 pool (n=4)	–	–	<0.8	<0.2	0.69	6.9	–	60 *	–	2
"	Lake Ellasjøen (13-15 cm)	74°30'N, 19°00'E	1996	1 pool (n=4)	–	–	n.d.	n.d.	n.d.	0.05	–	0.5 *	–	2
"	Lake Øyangen (0-1 cm)	74°30'N, 19°00'E	1996	1 pool (n=4)	–	–	<0.8	<0.4	0.17	0.8	–	4.4 *	–	2
"	Lake Øyangen (13-15 cm)	74°30'N, 19°00'E	1996	1 pool (n=4)	–	–	n.d.	n.d.	n.d.	0.2	–	0.4 *	–	2
Lake Lovozero	Lovozero, Kola Peninsula	68°00'N, 34°30'E	2000-01	1 pool (n=10)	–	0.15	0.15	0.02	0.07	0.87	2.52	–	<0.01	1
Chandler	Alaska north slope	68°18'N, 152°40'W	1994	1	–	–	–	–	–	–	0.06	–	–	3
Lindeman Lake	Northern BC	59°47'N, 135°03'W	1994	1	–	0.5	–	0.1	0.3	0.9	8.3	1.8	–	4
Fox Lake	Southern Yukon	61°15'N, 135°29'W	1993	1	–	0.7	–	0.8	0.0	0.7	33.5	12.7	1.3	4,5
Lake Laberge	"	61°12'N, 135°11'W	1992	2	–	0.4	–	1.2	0.4	0.8	12.7	3.7	0.18	4,5
Little Atlin Lake	"	60°16'N, 134°00'W	1993	1	–	0.3	–	1.1	3.9	3.5	28.3	7.4	–	4
Yaya	Mackenzie River Delta	69°10'N, 134°39'W	1994	1	–	1.57	–	0.06	0.23	0.43	6.23	2.1	–	6
Lake 6	"	68°41'N, 134°29'W	1993	1	–	–	–	0.15	–	1.30	2.60	–	–	7
Lake 7	"	68°41'N, 134°28'W	1993	1	–	–	–	1.40	–	0.10	10.80	–	–	7
Hanson Lake	Central Yukon	64°00'N, 135°21'W	1995	1	–	1.4	–	0.6	0.6	1.3	25.9	5.9	600	4,5
Watson Lake	Southern Yukon	60°06'N, 128°48'W	1995	2	–	1.3	–	0.7	0.9	122.6	38.6	5.2	–	4
Great Bear	Central NWT	65°05'N, 120°47'W	1994	1	–	1.87	–	0.20	0.18	0.50	27.8	7.1	–	8
Great Slave	Southern NWT	61°40'N, 114°00'W	1995	1	–	1.41	–	0.31	0.26	0.41	8.75	3.0	1.3	9
Char	Cornwallis Island	74°40'N, 94°50'W	1998	1	–	0.01	–	0.01	0.52	0.14	0.96	0.4	1.00	10
Peter Lake	Western Nunavut	63°00'N, 92°00'W	1994	1	–	0.28	–	0.09	0.17	0.11	4.44	1.0	–	11
DV09	Devon Island	75°34'N, 89°19'W	1999	1	–	–	–	0.23	0.18	0.07	2.97	–	0.72	12
Ax-Aj	Axel Heiberg Island	80°00'N, 87°00'W	1999	1	–	0.10	–	0.14	0.51	0.23	2.66	0.2	–	10
Romulus	Ellesmere Island	79°54'N, 85°06'W	2000	1	–	0.27	–	0.69	0.10	0.06	3.65	1.2	–	10
Baird Inlet	"	79°20'N, 75°00'W	1999	1	–	0.07	–	0.08	0.05	0.03	0.17	0.0	–	10
Bjørnvatnet	NW Spitsbergen	79°45'N, 11°30'E	1995	1	–	–	–	–	–	–	–	6.4 **	–	13
Ytertjørna	"	79°30'N, 12°00'E	1995	1	–	–	–	–	–	–	–	6.44 **	–	13
Ossian	"	79°00'N, 13°00'E	1995	1	–	–	–	–	–	–	–	2.74 **	–	13
Daltjørna	SW Spitsbergen	77°30'N, 14°00'E	1995	1	–	–	–	–	–	–	–	2.58 **	–	13
Tenndammen	"	78°15'N, 15°00'E	1995	1	–	–	–	–	–	–	–	13.52 **	–	13
Latnjajaure	Swedish Arctic	68°30'N, 18°20'E	1998-2000	1	–	–	0.16	0.06	–	0.3	–	0.6 ***	–	14
Njalakjaure	"	67°00'N, 16°45'E	1998-2000	1	–	–	0.15	0.08	–	0.4	–	0.6 ***	–	14
Louvvaure	"	66°45'N, 18°00'E	1998-2000	1	–	–	0.16	0.14	–	1.7	–	1.4 ***	–	14
Jutsajaure	"	67°00'N, 20°10'E	1998-2000	1	–	–	0.07	0.13	–	0.6	–	1 ***	–	14
Pahajärvi	"	66°55'N, 23°30'E	1998-2000	1	–	–	0.12	0.15	–	2.6	–	1.4 ***	–	14
Abiskojaure	"	68°30'N, 18°25'E	1998-2000	1	–	–	0.13	0.02	–	0.2	–	0.3 ***	–	14

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

\* sum of 7 congeners (101, 105, 118, 138, 153, 156, 180).

\*\* sum of 7 congeners (28, 52, 101, 118, 138, 153, 180).

\*\*\* sum of 7 congeners (28, 52, 101, 105, 138, 153, 180).

## References

1. RAIPON/AMAP/GEF Project, 2001. ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCH = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62.
2. Evenset *et al.*, 2002; ΣHCHs = sum of α- and γ-HCH.
3. Cleverly *et al.*, 1996.
4. Rawn *et al.*, 2001.
5. Lockhart *et al.*, 1997.
6. Lockhart, 1997.
7. Graf Pannatier, 1997.
8. Muir and Lockhart 1997, unpublished.
9. Evans *et al.*, 1996. ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners; Toxaphene = total toxaphene using single response factor.
10. Muir *et al.*, 2002b. Toxaphene determined by GC-ECNIMS.
11. Kidd *et al.*, 1998.
12. Stern and Lockhart, 2001. Toxaphene determined by GC-ECNIMS.
13. Rose *et al.*, 2003.
14. Söderström *et al.*, 2002.

Annex Table 7. Mean concentrations of organochlorines in freshwater invertebrates and fish. All Arctic char data are for landlocked fish; values for anadromous fish are in Annex Table 10 (marine and anadromous fish).

Species/Region	Location	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference	
<b>Invertebrates (ng/g ww)</b>																					
Plankton																					
NWT	Great Slave Lake, W Basin	whole body	1993-95	-	1.4±0.6	?	mean±SD	-	-	-	0.2±0.1	0.3±0.3	3.4±2.4	-	1.2±0.2	0.1±0.1	-	-	-	3	
"	Great Slave Lake, E Arm	whole body	1993-95	-	26.2±12.6	?	mean±SD	-	-	-	2.2±3.6	0.5±0.2	3.9±1.9	-	9.2±2.2	0.7±1.1	-	-	-	3	
Mysids																					
NWT	Great Slave Lake, W Basin	whole body	1993-95	-	33.6±4.6	?	mean±SD	-	-	-	0.89±0.6	0.3±0.1	2.2±0.7	-	6.5±2.2	0.2±0.2	-	-	-	3	
Amphipods																					
NWT	Great Slave Lake, W Basin	whole body	1993-95	-	2.2±0.6	?	mean±SD	-	-	-	0.7±0.4	0.4±0.2	2.4±0.8	-	6.1±1.1	0.2±0.1	-	-	-	3	
"	Great Slave Lake, E Arm	whole body	1993-95	-	9.9±8.2	?	mean±SD	-	-	-	1.4±1.2	0.8±0.6	2.6±1.2	-	7.1±3.4	0.3±0.2	-	-	-	3	
Unidentified zooplankton																					
Bjørnøya	Lake Ellasjøen	whole body	1996	-	2.02	-	-	-	0.56	-	-	3.48	-	32.0**	-	-	-	-	-	2	
"	"	whole body	1999	-	1.48	-	-	-	0.76	-	-	1.71	27.0	17.6**	-	-	-	-	-	2	
"	Lake Øyangen	whole body	1996	-	1.54	-	-	-	0.31	-	-	0.47	-	4.29**	-	-	-	-	-	2	
"	"	whole body	1999	-	0.62	-	-	-	0.08	-	-	0.16	2.15	1.13**	-	-	-	-	-	2	
"	Lake Skutilen	whole body	1999	-	1.31	-	-	-	0.16	-	-	0.25	7.18	2.57**	-	-	-	-	-	2	
Chironomidae sp.																					
Bjørnøya	Lake Ellasjøen	whole body	1996	-	2.81	2	-	-	0.88	-	-	10.2	-	56.6**	-	-	-	-	-	2	
"	"	whole body	1999	-	3.68	-	-	-	1.92	-	-	5.42	44.2	46.1**	-	-	-	-	-	2	
"	Lake Øyangen	whole body	1996	-	2.15	-	-	-	0.37	-	-	2.10	-	20.9**	-	-	-	-	-	2	
Lepidurus arcticus																					
Bjørnøya	Lake Øyangen	whole body	1998	-	2.37	-	-	-	0.62	-	-	0.54	7.09	4.12**	-	-	-	-	-	2	
"	Lake Skutilen	whole body	1999	-	0.39	-	-	-	0.06	-	-	0.11	1.93	0.93**	-	-	-	-	-	2	
<b>Freshwater fish (ng/g ww)</b>																					
Lota lota (burbot)																					
Alaska	Fairbanks	liver	1998	-	40	9	median	-	15	-	2.65	48	540	-	140	3	-	-	-	-	
"	"	"	"	"	6-57	"	range	-	4-19	-	<0.32-15	0.79-470	270-1400	-	24-200	1.4-5.6	-	-	-	-	
"	Kanuti	liver	1998	-	12	6	median	-	1.4	-	-	-	-	-	-	-	-	-	-	4	
"	"	"	"	"	4-32	"	range	-	0.9-3.7	-	<0.32-0.46	0.08-44	<16-110	-	<2.9-18	<0.26-2.8	-	-	-	-	
"	Tetlin	liver	1998	-	32	11	median	-	2.4	-	0.0435	-	-	-	12	0.6	-	-	-	4	
"	"	"	"	"	4-48	"	range	-	1.1-7.1	-	<0.32-1	<0.40-28	<1.6-170.0	-	<2.9-27.0	<0.26-1.2	-	-	-	-	
"	Yukon Flats	liver	1998	-	57	3	median	-	31	-	4.75	7.2	420	-	290	8	-	-	-	4	
"	"	"	"	"	53-61	"	range	-	23-45	-	1.8-21	0.45-290	160-950	-	87-420	3-14	-	-	-	-	
"	Nuiqsut (Colville R.)	liver	2000	-	41.4±2.92	11	mean±SE	10.2±0.67	8.76±0.58	4.8±0.45	30.24±4.10	18.4±1.35	51.8±4.50	-	-	4.79±0.31	0.35±0.09	0.64±0.09	-	5	
NWT	Great Slave Lake, W Basin	liver	1993	-	22.9±8.9	?	mean±SD	-	-	-	63.1±16.0	26.9±5.1	76.7±16.9	-	263±100	-	-	-	-	3	
"	"	liver	1995	-	21.2±13.7	?	mean±SD	-	-	-	92.3±29.0	50.0±16.9	158±21.6	-	424±199	-	-	-	-	3	
"	"	liver	1996	-	43.3±9.2	?	mean±SD	-	-	-	74.7±16.7	27.7±2.2	96.4±9.5	-	348±114	-	-	-	-	3	
"	"	liver	1999	-	-	?	mean±SD	-	-	-	71.7±19.3	32.0±9.1	114±47.3	-	277±48.4	-	-	-	-	3	
"	Great Slave Lake, E Arm	liver	1993	-	30.0±7.9	?	mean±SD	-	-	-	93.5±34.7	51.2±25.7	138±52.2	-	762±298	-	-	-	-	3	
"	"	liver	1999	-	-	?	mean±SD	-	-	-	45.7±23.0	23.0±12.1	79.3±50.4	-	190±60.6	-	-	-	-	3	
"	Fort Good Hope (Mackenzie R.)	liver	1988	m,f	30.2±13.5	10	mean±SD	13.6±4.21	13.7±4.06	5.53±1.71	23.8±7.37	16.2±5.25	58.1±18.4	-	60.8±19.3	2.38±0.74	0.57±0.17	-	-	6	
"	"	liver	1994	m,f	30.6±11.6	9	mean±SD	8.63±2.63	8.17±2.48	4.31±1.27	17.3±6.14	19.0±8.28	50.0±17.5	-	46.8±14.5	2.02±0.62	0.41±0.14	-	-	6	
"	"	liver	1999	m,f	42.1±13.3	21	mean±SD	10.0±3.81	5.43±2.17	3.78±1.38	21.0±8.04	22.8±8.59	62.8±22.3	-	54.0±20.4	2.38±0.93	0.59±0.24	-	1.59±1.15	6	
"	"	liver	2000	m,f	36.2±15.2	20	mean±SD	8.72±5.24	4.78±2.89	3.29±1.98	19.0±12.5	21.2±14.9	54.6±36.2	-	47.0±29.0	2.21±1.57	0.58±0.55	-	-	6	
Yukon	Fox Lake	liver	1998	m,f	29.1±7.57	9	mean±SD	0.49±0.18	4.45±1.61	9.26±3.42	16.8±6.17	41.3±13.4	32.2±10.3	-	40.8±13.9	3.11±1.34	0.35±0.14	-	-	7	
"	Lake Laberge	liver	1997	f	54.5	1	-	5.20	-	19.4	5.20	1620	915	-	1950	-	-	-	-	8	
"	"	liver	1999	m,f	53.8±6.56	11	mean±SD	41.6±17.6	-	50.3±23.7	248±122	2850±1200	1630±727	-	3050±1210	-	-	-	-	-	7
"	"	liver	1999	m,f	47.9±2.47	7	mean±SE	3.45±0.27	-	17.6±1.80	3.45±0.27	2380±497	985±148	-	2450±463	-	-	-	-	-	8
"	Kusawa Lake	liver	1999	m,f	23.8±8.46	11	mean±SD	5.28±2.10	-	5.55±2.15	35.6±16.8	56.2±31.1	130±51.6	-	208±69.0	-	-	-	-	-	7
"	"	liver	1999	m,f	25.6±2.67	9	mean±SE	0.90±0.13	-	5.11±0.62	0.90±0.13	46.1±7.45	116±15.3	-	618±85.5	-	-	-	-	-	8
"	Quite Lake	liver	1997	-	30.0±4.66	7	mean±SE	1.06±0.14	-	14.7±1.77	1.06±0.14	92.8±13.9	48.3±7.16	-	74.4±11.7	-	-	-	-	8	
"	"	liver	1997-99	m,f	25.6±12.3	9	mean±SD	7.26±1.72	-	13.3±4.97	27.8±11.6	87.9±32.3	121±114	-	99.6±46.1	-	-	-	-	-	7
"	Klukshu Lake	liver	1996	-	36.0	1	-	1.94	-	3.07	1.94	45.6	54.3	-	53.0	-	-	-	-	8	
Norway	Lake Grensefoss (Pasvikelva)	muscle	1999-2000	-	11.6	1	-	-	-	-	-	-	205	-	-	-	-	-	20.3	16	
Russia	Kola Peninsula	liver	2001	f	-	2 pools (n=6,7)	mean±SD	-	-	3.36±0.13	22.9±5.36	48.6±0.14	62.3±5.36	-	-	-	0.75±0.33	-	-	1	
"	"	liver	2001	m	-	2 pools (n=6,7)	mean±SD	-	-	0.52±0.23	4.27±0.51	17.0±12.4	33.1±1.08	-	-	-	0.10	-	-	1	
"	"	muscle	2001	f	-	2 pools (n=6,7)	mean±SD	-	-	0.05±0.06	<0.05	0.84±1.08	2.34±0.04	-	-	-	0.06	-	-	1	
"	"	muscle	2001	m	-	2 pools (n=6,7)	mean±SD	-	-	0.19±0.13	<0.05	1.03±0.17	5.36±0.52	-	-	-	<0.05	-	-	1	
"	Dudinka, Taymir Peninsula	liver	2001	f	-	3 pools (n=2-6)	mean±SD	-	-	1.13±0.30	30.7±9.00	472±44.6	516±55.7	-	-	-	0.68±0.12	-	-	1	
"	"	muscle	2001	f	-	3 pools (n=2-6)	mean±SD	-	-	0.06	<0.05	1.08±0.18	5.16±0.41	-	-	-	0.13±0.03	-	-	1	
"	"	liver	2001	m	-	3 pools (n=1-5)	mean±SD	-	-	2.07±0.14	21.9±3.72	366±60.2	390±45.7	-	-	-	0.65±0.05	-	-	1	
"	"	muscle	2001	m	-	3 pools (n=1-5)	mean±SD	-	-	0.06	0.12	0.69±0.07	4.35±1.16	-	-	-	0.12	-	-	1	
"	Khatanga, Taymir Peninsula	liver	2001	f	-	2 pools (n=2,5)	mean±SD	-	-	3.67±0.31	40.1±6.47	90.6±11.9	108±18.4	-	-	-	1.65	-	-	1	
"	"	liver	2001	m	-	2 pools (n=3,10)	mean±SD	-	-	2.9±0.03	26.0±2.33	84.4±11.5	105±6.02	-	-	-	2.33	-	-	1	
"	"	muscle	2001	f	-	2 pools (n=2,5)	mean±SD	-	-	0.12±0.04	<0.05	0.23±0.13	2.16±0.10	-	-	-	<0.05	-	-	1	
"	"	muscle	2001	m	-	2 pools (n=3,10)	mean±SD	-	-	0.02±0.03	<0.05	0.57	1.83±0.18	-	-	-	<0.05	-	-	1	

Continued next page.



Annex Table 7 continued.

Species/Region	Location	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference
<i>Salvelinus alpinus</i> (Arctic char)																				
Cornwallis Isl., Nunavut	Resolute Lake	muscle+skin	1997	–	5.17±2.79	10	mean±SD	1.70±1.64	1.55±1.52	0.67±0.58	6.80±8.89	9.23±13.0	160±208	–	–	1.63±1.60	0.09±0.13	–	–	9
“	“	muscle+skin	1999	–	3.85±1.28	10	mean±SD	1.47±0.41	1.34±0.37	0.70±0.28	4.67±2.26	6.69±3.34	96.5±56.7	–	–	1.47±0.47	0.08±0.06	–	–	9
“	Resolute Lake	muscle+skin	2000	–	5.45±1.52	8	mean±SD	2.08±0.56	1.90±0.52	0.70±0.18	1.73±0.66	8.79±4.83	133±68.7	–	–	1.95±0.56	0.10±0.08	–	–	9
Cornwallis Isl., Nunavut	Char Lake	muscle+skin	1999	–	2.86±1.77	4	mean±SD	1.14±0.55	1.06±0.50	0.33±0.17	1.03±0.40	32.3±8.29	125±48.1	–	–	1.85±1.32	0.48±0.26	–	–	9
“	“	muscle+skin	2000	–	2.76±0.40	3	mean±SD	1.00±0.05	0.87±0.04	0.46±0.07	2.13±0.75	14.8±9.27	64.9±31.4	–	–	1.24±0.14	0.14±0.06	–	–	9
SW Greenland	Isortoq	muscle	1999	m	1.1±1.1	9	mean±SD	0.33±0.31	0.31±0.28	0.13±0.13	1.5±0.8	1.6±0.8	14±6	1.6±0.5	–	0.23±0.2	0.16±0.07	–	–	10
“	“	muscle	1999	f	1.4±0.8	9	mean±SD	0.45±0.20	0.42±0.18	0.14±0.08	2.1±0.9	2.1±1.2	16±7	2.7±2.5	–	0.31±0.18	0.18±0.08	–	–	10
NE Greenland	Zackenber	muscle	1999	m	2.5±0.7	2	mean±SD	1.29±0.06	1.22±0.04	0.2±0.05	2.3±0.4	1.7±0.4	11±2	4.5±0.9	–	0.47±0.13	0.1±0.02	–	–	10
“	“	muscle	1999	f	2.2±1.4	11	mean±SD	1.13±0.62	1.06±0.58	0.20±0.12	2.0±1.8	1.5±1.1	6±4	2.5±1.3	–	0.71±0.74	0.05±0.04	–	–	10
“	“	muscle	1999	both	1.0±0.4	8	mean±SD	0.51±0.18	0.49±0.17	0.06±0.05	2.1±1.0	2.6±1.7	14±9	4.7±3.2	–	0.36±0.17	0.1±0.07	–	–	10
“	Ittoqqortoormiit	muscle	2000	m	2.9±1.4	6	mean±SD	0.74±0.25	0.65±0.20	0.65±0.50	4.9±1.8	5.3±2.6	23±12	7.7±3.9	–	1.3±0.29	0.05±0.03	–	–	10
“	“	muscle	2000	f	2.4±1.1	14	mean±SD	0.69±0.26	0.62±0.23	0.43±0.39	3.6±1.7	3.8±2.4	16±8	5.4±2.8	–	1.16±0.44	0.05±0.03	–	–	10
Faroe Islands	mountain dam	muscle	2000-2001	–	–	65	mean	–	0.9	–	–	1	0.9	–	–	–	–	–	–	11
Jan Mayen	–	liver	1995/96	–	2.5	1 pool	mean	–	0.9	1.50	8.8	70.7	155	–	–	–	–	–	–	12
Bjørnøya	Lake Øyangen	muscle	1998	–	2.18 1.57-2.97	3 pools (n=3)	mean range	–	–	–	–	3.85 1.62-6.46	64.8 32.2-97.5	37.1* 18.2-55.9	–	–	–	–	–	2
“	Lake Ellasjøen	muscle	1999	–	3.14 6.65-4.03?	3 pools (n=1-2)	mean range	–	–	–	–	62.7 40.9-84.7	1140 596-1630	643* 374-907	–	–	–	–	–	2
Finland	Lake Pahtajärvi	muscle	1999	f	1.34±0.49	5	mean±SD	–	0.10±0.49	0.02±0.02	0.04±0.02	0.18±0.02	0.15±0.06	–	–	–	–	–	–	13
“	“	muscle	1999	m	1.09±0.42	5	mean±SD	–	0.09±0.03	0.01±0.02	0.02±0.03	0.24±0.21	0.28±0.46	–	–	–	–	–	–	13
“	Lake Kilpisjärvi	muscle	2001	–	0.97	1 pool (n=5)	–	–	0.26	0.07	0.37	0.93	3.33	–	–	–	–	–	–	13
<i>Esox locius</i> (northern pike)																				
NWT	Great Slave Lake, W Basin	muscle	1996	–	2.2±0.6	?	mean±SD	–	–	–	2.3±0.7	2.7±0.8	5.3±1.5	–	22.2±6.6	–	–	–	–	3
“	“	muscle	1999	–	0.9±0.3	?	mean±SD	–	–	–	1.9±1.1	2.3±1.1	4.8±2.5	–	21.5±7.3	–	–	–	–	3
“	Great Slave Lake, E Arm	muscle	1999	–	1.1±0.8	?	mean±SD	–	–	–	0.8±0.2	1.3±0.4	2.5±0.9	–	12.1±2.5	–	–	–	–	3
Finland	Lake Inari	muscle	1997,99	f	0.43±0.09	2	mean±SD	–	0.16±0.04	n.d.	0.16±0.03	0.89±0.78	1.62±0.83	–	–	–	–	–	–	13
“	River Tornionjoki	muscle	1997	m	0.35	1	–	–	0.09	0.03	n.d.	0.10	0.33	–	–	–	–	–	–	13
“	“	muscle	1997	f	0.46	1	–	–	0.22	n.d.	0.35	2.36	8.78	–	–	–	–	–	–	13
“	Reservoir Lokka	muscle	1997-2001	f	0.35±0.08	4	mean±SD	–	0.08±0.02	0.01±0.02	0.01±0.02	0.15±0.18	n.d.	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.28±0.03	3	mean±SD	–	0.07±0.01	n.d.	n.d.	0.10±0.02	0.13±0.15	–	–	–	–	–	–	13
“	Lake Kemijärvi	muscle	2001	f	0.26±0.04	3	mean±SD	–	0.26±0.04	n.d.	n.d.	0.22±0.08	0.90±0.28	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.30±0.01	2	mean±SD	–	0.07±0.01	n.d.	0.06±0.03	0.78±0.64	2.47±1.44	–	–	–	–	–	–	13
Russia	Kola Peninsula	liver	2001	f	–	2 pools (n=6,7)	mean±SD	–	–	0.64±0.03	1.47±0.82	4.15±2.10	11.9±2.38	–	–	–	0.12	–	–	1
“	“	liver	2001	m	–	2 pools (n=3,5)	mean±SD	–	–	0.73±0.04	2.05±1.39	4.31±0.64	10.5±2.45	–	–	–	0.14	–	–	1
“	“	muscle	2001	f	–	2 pools (n=6,7)	mean±SD	–	–	0.18±0.03	0.11±0.03	0.51±0.19	1.33±0.32	–	–	–	<0.05	–	–	1
“	“	muscle	2001	m	–	2 pools (n=5,3)	mean±SD	–	–	0.10±0.06	<0.05	0.90±0.21	1.18±0.45	–	–	–	<0.05	–	–	1
“	Kanchalan, Chukotka Peninsula	liver	2001	f	–	3 pools (n=2-6)	mean±SD	–	–	1.2±0.40	4.83±1.36	10.6±2.46	10.2±3.13	–	–	–	<0.05	–	–	1
“	“	liver	2001	m	–	3 pools (n=1-5)	mean±SD	–	–	0.97±0.28	1.52±0.53	5.87±1.58	7.97±1.59	–	–	–	<0.05	–	–	1
“	“	muscle	2001	f	–	3 pools (n=2-6)	mean±SD	–	–	<0.10	<0.05	0.15±0.25	1.73±0.65	–	–	–	<0.05	–	–	1
“	“	muscle	2001	m	–	3 pools (n=1-5)	mean±SD	–	–	0.07±0.06	<0.05	<0.05	2.91±0.96	–	–	–	<0.05	–	–	1
<i>Perca fluviatilis</i> (European perch)																				
Finland	Lake Inari	muscle	1997	m	0.69	1	–	–	0.13	0.03	0.07	0.32	0.42	–	–	–	–	–	–	13
“	Reservoir Lokka	muscle	1997	f	0.44	1	–	–	0.14	0.03	<0.06	n.d.	n.d.	–	–	–	–	–	–	13
Russia	Pechora Basin	liver	2001	f	–	3 pools (n=5-9)	mean±SD	–	–	1.95±0.92	0.33±0.18	2.32±0.34	3.50±0.41	–	–	–	<0.05	–	–	1
“	“	muscle	2001	f	–	3 pools (n=5-9)	mean±SD	–	–	0.25±0.24	0.17±0.08	0.99±0.40	1.03±0.40	–	–	–	<0.05	–	–	1
<i>Coregonus</i> sp. (whitefish)																				
Yukon	Lake Laberge	muscle	1998	–	1.39±0.14	5	mean±SE	0.004±0.002	–	0.32±0.06	2.32±0.25	24.6±2.36	4.58±1.00	–	42.7±7.68	–	–	–	–	8
“	Watson Lake	muscle	1997	f	2.99±1.55	2	mean	0.02	–	2.66	9.02	6.43	24.7	–	20.3±3.13	–	–	–	–	8
“	“	muscle	1998	m,f	1.71±0.12	12	mean±SE	0.26±0.04	–	0.89±0.15	1.66±0.23	6.54±0.79	7.92±1.03	–	7.26±0.98	–	–	–	–	8
NWT	Great Slave Lake, W Basin	?	1993-95	–	9.9±7.0	?	mean±SD	–	–	–	4.2±3.9	1.9±1.2	4.7±4.2	–	21.7±16.6	0.7±0.3	–	–	–	3
“	Great Slave Lake, E Arm	?	1993-95	–	5.9±4.1	?	mean±SD	–	–	–	2.9±2.3	2.8±1.1	6.6±2.8	–	25.5±14.3	0.5±0.2	–	–	–	3
Finland	Lake Inari	muscle	2001	f	0.78±0.31	2	mean±SD	–	0.12±0.05	0.02±0.03	0.05±0.02	0.29±0.22	0.64±0.63	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.71±0.27	3	mean±SD	–	0.13±0.04	0.01±0.02	0.06±0.03	0.37±0.17	0.80±0.19	–	–	–	–	–	–	13
“	River Tornionjoki	muscle	2001	f	0.70	1	mean±SD	–	0.62	n.d.	0.63	10.3	19.5	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.78±0.36	4	mean±SD	–	0.51±0.37	n.d.	0.57±0.43	5.32±4.48	14.7±10.1	–	–	–	–	–	–	13
“	Lake Nitsijärvi	muscle	2001	f	0.49	1	mean±SD	–	0.06	0.02	n.d.	0.22	0.47	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.77±0.36	4	mean±SD	–	0.10±0.04	0.03±0.04	0.09±0.09	0.41±0.21	1.09±0.57	–	–	–	–	–	–	13
“	Reservoir Lokka	muscle	2001	f	0.75±0.67	5	mean±SD	–	0.08±0.06	0.02±0.04	0.01±0.02	0.06±0.06	0.02±0.04	–	–	–	–	–	–	13
“	“	muscle	2001	m	0.68±0.35	5	mean±SD	–	0.07±0.02	0.02±0.03	0.01±0.01	0.04±0.04	0.01±0.02	–	–	–	–	–	–	13
<i>C. lavaretus</i> (common whitefish)																				
Russia	Kola Peninsula	liver	2001	f	–	2 pools (n=4)	mean±SD	–	–	0.93±0.33	1.78±0.16	6.56±2.02	5.42±1.10	–	–	–	≤0.05	–	–	1
“	“	liver	2001	m	–	2 pools (n=6)	mean±SD	–	–	0.47±0.17	1.12±0.24	4.29±1.48	5.43±0.91	–	–	–	0.11±0.06	–	–	1
“	“	muscle	2001	f	–	2 pools (n=4)	mean±SD	–	–	0.39±0.06	0.19	1.94±0.04	2.22±0.42	–	–	–	<0.05	–	–	1
“	“	muscle	2001	m	–	2 pools (n=6)	mean±SD	–												

Annex Table 7 continued.

Species/Location	Region	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference
Russia	Pechora Basin	muscle	2001	f	–	2 pools (n=3,4)	mean±SD	–	–	0.15	0.07	1.53±0.32	1.81±0.35	–	–	–	<0.05	–	–	1
”	”	muscle	2001	m	–	3 pools (n=1-7)	mean±SD	–	–	0.17±0.15	0.41±0.13	1.75±0.26	2.03±0.47	–	–	–	<0.05	–	–	1
”	Dudinka, Taymir Peninsula	liver	2001	f	–	2 pools (n=4,6)	mean±SD	–	–	0.33±0.26	0.67±0.16	3.19±0.25	3.35±1.24	–	–	–	<0.05	–	–	1
”	”	muscle	2001	f	–	2 pools (n=4,6)	mean±SD	–	–	0.13±0.18	0.49±0.24	2.22±0.41	4.13±2.98	–	–	–	≤0.05	–	–	1
”	”	liver	2001	m	–	2 pools (n=3,7)	mean±SD	–	–	0.37±0.03	0.67±0.18	3.64±0.57	4.13±2.00	–	–	–	<0.05	–	–	1
”	”	muscle	2001	m	–	2 pools (n=3,7)	mean±SD	–	–	0.18±0.13	0.29±0.09	2.15±0.10	2.81±0.84	–	–	–	<0.05	–	–	1
”	Khatanga, Taymir Peninsula	liver	2001	f	–	3 pools (n=1-3)	mean±SD	–	–	0.36±0.13	3.49±1.32	3.35±0.68	4.48±2.26	–	–	–	0.19±0.01	–	–	1
”	”	liver	2001	m	–	3 pools (n=3-6)	mean±SD	–	–	0.53±0.25	3.57±0.57	4.11±1.28	5.12±1.92	–	–	–	0.17±0.11	–	–	1
”	”	muscle	2001	f	–	3 pools (n=1-3)	mean±SD	–	–	0.22±0.09	1.41±0.39	2.74±0.54	3.08±0.90	–	–	–	<0.05	–	–	1
”	”	muscle	2001	m	–	3 pools (n=3-6)	mean±SD	–	–	0.27±0.05	1.56±0.08	2.22±0.55	3.15±0.64	–	–	–	<0.05	–	–	1
<i>C. nasus</i> (broad whitefish)																				
Russia	Khatanga, Taymir Peninsula	liver	2001	f	–	3 pools (n=2-5)	mean±SD	–	–	0.84±0.62	0.97±0.46	0.80±0.37	2.32±0.30	–	–	–	<0.05	–	–	1
”	”	liver	2001	m	–	3 pools (n=1-6)	mean±SD	–	–	0.78±0.53	1.06±0.47	0.76±0.34	2.18±1.02	–	–	–	≤0.05	–	–	1
”	”	muscle	2001	f	–	3 pools (n=2-5)	mean±SD	–	–	0.08±0.14	1.48±0.69	1.41±0.65	3.21±0.52	–	–	–	≤0.05	–	–	1
”	”	muscle	2001	m	–	3 pools (n=1-6)	mean±SD	–	–	0.16±0.15	0.69±0.38	1.11±0.30	2.35±0.79	–	–	–	0.06	–	–	1
”	Kanchalan, Chukotka Peninsula	liver	2001	f	–	1 pool (n=13)	–	–	–	0.42	0.37	1.01	1.72	–	–	–	0.10	–	–	1
”	”	liver	2001	m	–	1 pool (n=7)	–	–	–	0.54	0.09	0.98	1.78	–	–	–	0.14	–	–	1
”	”	muscle	2001	f	–	1 pool (n=13)	–	–	–	0.22	0.13	0.63	1.15	–	–	–	<0.05	–	–	1
”	”	muscle	2001	m	–	1 pool (n=7)	–	–	–	0.16	<0.05	0.71	1.31	–	–	–	<0.05	–	–	1
<i>Salvelinus namaycush</i> (lake trout)																				
Yukon	Atlin Lake	muscle	1998	m,f	1.76±0.58	11	mean±SD	2.24±0.84	–	1.73±0.60	4.71±2.76	4.39±1.58	17.2±7.16	–	12.7±4.97	–	–	–	–	7
”	Fox Lake	muscle	1998	m,f	1.07	2	mean	0.05	–	0.31	0.52	1.46	2.37	–	5.19	–	–	–	–	8
”	Kusawa Lake	muscle	1999	m,f	4.21±0.62	3	mean±SE	0.32±0.03	–	2.22±0.65	20.8±2.72	197±41.3	125±21.2	–	307±58.0	–	–	–	–	8
”	”	muscle	1999	m,f	4.61±2.98	14	mean±SD	1.52±0.73	–	1.68±0.84	27.4±10.2	139±73.8	91.1±44.3	–	149±110	–	–	–	–	7
”	Lake Laberge	muscle	1993	m,f	8.22±4.49	15	mean±SD	4.09±2.77	–	5.10±3.53	42.9±27.4	250±98.4	183±91.1	–	229±66.9	–	–	–	–	7
”	”	muscle	1996	m,f	8.07±4.17	5	mean±SD	2.24±0.90	–	2.16±0.82	22.3±13.2	102±51.1	74.7±35.1	–	125±35.1	–	–	–	–	7
”	”	muscle	1998	m,f	8.98±1.27	7	mean±SD	2.06±0.56	–	1.92±0.51	13.1±3.62	52.7±14.6	42.6±10.5	–	70.9±38.2	–	–	–	–	7
”	”	muscle	1999	m	3.45	1	–	0.15	–	0.20	2.73	19.6	22.8	–	8.06	–	–	–	–	8
”	Quiet Lake	muscle	1999	m,f	2.40±0.90	8	mean±SD	0.93±0.30	–	0.25±0.12	3.01±0.72	1.62±0.79	7.24±2.27	–	5.99±1.88	–	–	–	–	7
”	Coal Lake	muscle	1999	m,f	3.97±2.79	12	mean±SD	0.59±0.44	–	0.73±0.53	4.67±5.43	11.9±9.70	25.2±20.1	–	6.78±4.85	–	–	–	–	7
”	Mandanna Lake	muscle	2000	f	1.83	2	mean	0.15	–	0.45	0.65	3.40	8.18	–	15.2	–	–	–	–	8
NWT	Great Slave Lake, W Basin	?	1993-95	–	12.8±3.1	?	mean±SD	–	–	–	9.0±5.6	5.8±3.4	13.9±7.2	–	48.5±23.4	0.7±0.4	–	–	–	3
”	Great Slave Lake, E Arm	?	1993-95	–	10.8±6.2	?	mean±SD	–	–	–	14.6±8.5	8.9±5.8	23.2±5.3	–	122±88.1	1.1±0.5	–	–	–	3
”	Great Slave Lake Lutsel K'e Region	muscle	1995	–	16.0±5.8	?	mean±SD	–	–	–	–	8.0±3.0	20.5±9.0	–	77.1±28.1	–	–	–	–	3
”	”	liver	1995	–	9.6±6.2	?	mean±SD	–	–	–	–	13.9±11.4	89.4±65.5	–	164±121	–	–	–	–	3
”	”	stomach	1995	–	17.9±12.8	?	mean±SD	–	–	–	–	6.1±3.5	42.7±27.8	–	331±234	–	–	–	–	3
”	Great Slave Lake, W Basin	muscle	1993	–	12.8±3.1	?	mean±SD	–	–	–	9.0±5.6	5.1±3.7	12.3±8.2	–	43.5±27.2	–	–	–	–	3
”	”	muscle	1999	–	12.6±6.8	?	mean±SD	–	–	–	12.6±6.8	6.7±3.4	30.2±16.7	–	80.3±28.9	–	–	–	–	3
”	Great Slave Lake, E Arm	muscle	1993	–	7.6±3.9	?	mean±SD	–	–	–	16.8±9.9	9.6±7.2	24.9±18.5	–	151±102	–	–	–	–	3
”	”	muscle	1995	–	16.0±5.8	?	mean±SD	–	–	–	11.2±4.5	8.0±3.0	20.6±9.0	–	77.1±28.1	–	–	–	–	3
”	”	muscle	1999	–	4.9±3.7	?	mean±SD	–	–	–	6.3±2.7	6.4±2.5	14.4±6.8	–	52.4±21.1	–	–	–	–	3
<i>Salmo trutta</i> (brown trout), landlocked																				
Faroe Islands	Fjallavatn Lake	liver	1997	–	–	9(28.6 cm)	mean	–	0.5	–	–	1-2	3-4	–	–	–	–	–	–	14
”	”	liver	1997	–	–	19(23.4 cm)	mean	–	0.5	–	–	1-2	3-4	–	–	–	–	–	–	14
”	Leitisvatn Lake	liver	1997	–	–	23(23.4 & 28.6 cm)	mean	–	0.5	–	–	1-2	3-4	–	–	–	–	–	–	14
Norway	Lake Takvatn	muscle	1999-2000	–	1.8	1?	–	–	–	–	–	–	–	2.04*	–	–	–	–	–	0.15
”	Lake Fjellfrøsvatnet	muscle	1999-2000	–	1.1	1?	–	–	–	–	–	–	–	0.87*	–	–	–	–	–	0.14
”	Lake Grunnvatnet	muscle	1999-2000	–	1.3	1?	–	–	–	–	–	–	–	0.23*	–	–	–	–	–	0.10
”	Lake Store Raudvannet	muscle	1999-2000	–	2.5	1?	–	–	–	–	–	–	–	3.20*	–	–	–	–	–	0.36
<i>Stizostedion vitreum</i> (walleye)																				
NWT	Great Slave Lake, W Basin	?	1996	?	3.4±1.3	?	mean±SD	–	–	–	1.3±0.6	1.6±1.0	4.5±1.5	–	14.7±9.8	0.1±0.02	–	–	–	3
<i>Stenodus leucichthys nelma</i> (inconnu)																				
Yukon	Peel River	muscle	1999	m,f	5.70±1.22	10	mean±SE	0.22±0.04	–	0.09±0.02	1.19±0.24	1.28±0.17	3.44±0.41	–	1.46±0.33	–	–	–	–	8
NWT	Great Slave Lake, W Basin	muscle	1996	–	20.5±6.2	–	mean±SD	–	–	–	6.8±1.9	4.4±1.1	11.5±2.0	–	31.3±5.5	0.9±0.3	–	–	–	3
<i>Stenodus nelma</i> (nelma)																				
Russia	Kanchalan, Chukotka Peninsula	liver	2001	m	–	2 pools (n=3,6)	mean±SD	–	–	0.30±0.15	0.70±0.21	2.55±1.11	5.07±0.71	–	–	–	0.31±0.02	–	–	1
”	”	muscle	2001	m	–	2 pools (n=3,6)	mean±SD	–	–	0.21±0.30	0.12±0.16	0.96±0.13	1.30±0.13	–	–	–	<0.05	–	–	1
<i>Coregonus autumnalis</i> (Arctic cisco)																				
NWT	Great Slave Lake, W Basin	–	1995	–	8.1±6.3	–	mean±SD	–	–	–	0.6±0.4	1.5±0.4	3.4±3.3	–	1.7±0.8	0.1±0.1	–	–	–	3
Russia	Dudinka, Taymir Peninsula	liver	2001	f	–	2 pools (n=4,6)	mean±SD	–	–	0.54±0.28	1.48±0.78	5.15±0.26	3.65±1.10	–	–	–	0.08	–	–	1
”	”	muscle	2001	f	–	2 pools (n=2,8)	mean±SD	–	–	0.27±0.25	0.62±0.42	1.80±1.08	3.21±0.54	–	–	–	<0.05	–	–	1
”	”	liver	2001	m	–	2 pools (n=4,6)	mean±SD	–	–	0.37±0.06	1.14±0.29	4.48±0.37	3.78±0.64	–	–	–	<0.05	–	–	1
”	”	muscle	2001	m	–	2 pools (n=2,8)	mean±SD	–	–	0.34±0.05	0.99±0.35	2.16±1.38	4.17±0.52	–	–	–	0.07	–	–	1
<i>Thymallus arcticus</i> (Arctic grayling)																				
Alaska	Nuiqsut (Colville R.)	muscle	2000	m	4.45±1.91	2	mean±SE	4.83±1.83	3.41±2.24	0.26±0.20	1.28±0.63	1.32±0.51	2.13±1.21	–	–	0.08±0.07	0.04±0.03	0.04±0.02	–	5

Continued next page.

Annex Table 7 continued.

Species/Location	Region	Tissue	Year	Sex	% lipid	n	Statistic	ΣCBz	HCb	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference
<i>Thymallus thymallus</i> (grayling)																				
Russia	Lavrentiya, Chukotka Peninsula	liver	2001	f	-	1	-	-	-	0.18	1.12	14.1	5.28	-	-	-	<0.05	-	-	1
"	"	liver	2001	m	-	1 (6)	-	-	-	0.24	1.13	12.5	5.19	-	-	-	<0.05	-	-	1
"	"	muscle	2001	f	-	1	-	-	-	0.19	0.66	3.81	1.89	-	-	-	<0.05	-	-	1
"	"	muscle	2001	m	-	1 (6)	-	-	-	0.12	0.43	4.50	2.04	-	-	-	<0.05	-	-	1
<i>Leuciscus idus</i> (European carp)																				
Russia	Pechora Basin	liver	2001	f	-	2 pools (n=2,5)	mean±SD	-	-	1.45±0.25	0.27±0.17	2.46±0.09	2.75±0.36	-	-	-	<0.05	-	-	1
"	"	liver	2001	m	-	3 pools (n=3-5)	mean±SD	-	-	1.39±0.41	0.49±0.24	2.01±0.55	2.84±0.81	-	-	-	<0.05	-	-	1
"	"	muscle	2001	f	-	2 pools (n=2,5)	mean±SD	-	-	0.47±0.02	0.14±0.07	1.73±0.27	1.83±0.24	-	-	-	≤0.05	-	-	1
"	"	muscle	2001	m	-	3 pools (n=3-5)	mean±SD	-	-	0.59±0.10	0.33±0.17	1.05±0.34	1.58±0.36	-	-	-	≤0.07	-	-	1
<i>Acipenser</i> spp. (sturgeon)																				
Russia	Kara Sea	liver	1993	-	-	-	mean±SD	-	-	23±10	18±10	640±390	290±200	-	-	-	-	-	-	15

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

\* sum of 7 congeners (101, 105, 118, 138, 153, 156, 180).

## References

- RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5-, and 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62; ΣPBDEs = sum of 2,2',4,4'-tetrabromodiphenyl, 2,2',4,4',5'-pentabromodiphenyl and 2,2',4,4',5'-pentabromodiphenyl ethers.
- Evenset *et al.*, 2002; ΣHCHs = sum of α- and γ-HCH; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 33 congeners (18, 28, 31, 33, 37, 47, 52, 60, 66, 74, 99, 101, 105, 114, 118, 122, 123, 128, 138, 141, 149, 153, 156, 157, 167, 170, 180, 183, 187, 189, 194, 206, 209)
- Evans, 2001; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners; Toxaphene = total toxaphene.
- Mueller and Matz, 2000; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣCHLs = sum of *cis*-chlordane and heptachlor epoxide; ΣPCBs = total Aroclors 1242, 1254, 1260.
- Hoekstra, 2002a; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, and γ-, δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209).
- Stern *et al.*, 2001a; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners; Toxaphene = total toxaphene.
- Stern *et al.*, 2000; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners.
- Palmer and Roach, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5- & 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners.
- Muir *et al.*, 2001a; ΣCBz = sum of 1,2,3,4-tetra, penta- and hexachlorobenzene; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, and *trans*- and *cis*-nonachlor; ΣDDTs = *o,p'*-DDT, *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 103 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Denmark, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣHCHs = sum of α-, β-, and γ-HCH; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners.
- Hoydal *et al.*, 2001; ΣDDTs = *p,p'*-DDE; ΣPCBs = CB153.
- Gabrielsen *et al.*, 1997; ΣPCBs = sum of 33 congeners.
- Mannio, 2002; ΣHCHs = sum α-, β-, and γ-HCH; ΣCHLs = sum *cis*-chlordane and *trans*-nonachlor; ΣDDTs = *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum 15 congeners (28, 31, 52, 66, 101, 149, 118, 153, 105, 138, 187, 156, 180, 170).
- Larsen and Dam, 1999; ΣCHLs = *trans*-nonachlor; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 3 congeners (153, 138/163, 180).
- Sericano *et al.*, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and heptachlor epoxide; ΣDDTs = *o,p'*- and *p,p'*-DDE, -DDD, -DDT; ΣPCBs = sum of all measurable congeners.
- Schlabach *et al.*, 2001.

Annex Table 8. Concentrations (mean ±SE) of major OC groups in seawater (ng/L) from the Arctic.

Region	Location/year/depth	n	Method	ΣCBz	ΣHCHs	ΣCHLs	ΣDDTs	Endosulfan	Toxaphene	ΣPCBs	ΣPCB <sub>10</sub>	Reference/ cruise
Central Archipelago, Canada	Resolute, 1999, surface	1	SPE	–	3.9	–	–	–	–	–	–	1
„	Hudson Strait, 1999, surface	9	SPE	–	3.1	0.013	–	0.0091	–	–	–	1
„	Foxe Basin/Channel, 1999, surface	4	SPE	–	3	0.005	–	0.0049	–	–	–	1
„	Melville Sound/McClure Strait, 1997, surface	10	Go-Flo & XAD	0.04 ±0.03	3.09±0.136	0.007±0.001	0.024±0.032	0.002±0.002	–	0.740±0.731	0.208±0.209	2
„	Hudson Str., Foxe Basin, Pr. Regent In., 1999, surface	6	Go-Flo & XAD	0.017±0.003	1.33±0.081	0.011±0.007	0.017±0.014	<0.001	–	0.277±0.126	0.065±0.004	3
Chukchi Sea, Chukchi Plateau	SHEBA 1998 (July and Sept, 10-30 m)	4	Infiltrax, <i>in situ</i>	0.013± 0.007	1.10 ±0.80	0.005±0.003	0.001±0.001	0.0007±0.0004	0.040 ±0.016	0.042 ±0.022	–	4
North Barents Sea	Oden cruise, 1996, surface	20	Go-Flo & XAD	0.004 ±0.002	0.967±0.499	0.009±0.014	0.003±0.003	0.001±0.002	–	0.161±0.074	0.057±0.027	5
„	Oden cruise, 1996, >100 m	21	Go-Flo & XAD	0.086	0.549	0.007	0.008	0.002	–	0.053	–	5
„	Oden cruise, 1996–50–300 m	17	Go-Flo & XAD	–	0.971±0.45	–	–	–	–	–	–	6
„	Oden cruise, 1996, 500–1000 m	10	Go-Flo & XAD	–	0.465±0.23	–	–	–	–	–	–	6
SW Beaufort Sea	Barrow, AK 1999–2000, surface	7	Infiltrax, <i>in situ</i>	0.120±0.006	2.11±0.46	0.010±0.004	0.019±0.006	0.001±0.001	0.188±0.046	0.231±0.090	0.058±0.058	7
SE Beaufort Sea	Holman, NWT 1999, surface	3	Infiltrax, <i>in situ</i>	0.043±0.004	2.28±0.15	0.008±0.002	0.005±0.001	0.002±0.001	0.150±0.040	0.135±0.018	0.039±0.013	7
North Baffin Bay	Northwater Polynya, 1998, >100 m	9	Go-Flo & XAD	0.094	0.959	0.004	0.015	0.002	–	0.405	–	8
„	Northwater Polynya, 1998, surface	8	Go-Flo & XAD	0.018±0.005	1.42±0.273	0.010±0.004	0.008±0.006	0.003±0.002	–	0.187±0.098	0.058±0.046	8
NW Russia White Sea	West Onega Bay; Central Basin, 1999–00, surface	10	Infiltrax, <i>in situ</i>	0.009± 0.004	0.84 ±0.273	0.007±0.004	0.003±0.001	0.003±0.002	0.041±0.008	0.105±0.057	0.028±0.014	9
Greenland Sea, E	74.6°N, 12.0°E, surface	5	SPE	–	0.572±0.203	–	–	–	–	–	–	10
„	74.9°N, 4.2°E, surface	2	SPE	–	0.371	–	–	–	–	–	–	10
„	76.0°N, 1.2°E, surface	1	SPE	–	0.487	–	–	–	–	–	–	10
„	62.1°N, 5.9°W, surface	2	SPE	–	0.698	–	–	–	–	–	–	10
Greenland Sea, W	74.6°N, 6.0°W, surface	1	SPE	–	0.131	–	–	–	–	–	–	10
„	68.2°N, 10.1°W, surface	1	SPE	–	0.438	–	–	–	–	–	–	10
„	77.2°N, 11.1°W, surface	1	SPE	–	0.26	–	–	–	–	–	–	10
NW Atlantic	63.67°N, 33.0°W, 50 m	1	Kiel <i>in situ</i>	–	–	–	–	–	–	–	0.00195	11
„	63.67°N, 33.0°W, 100 m	1	Kiel <i>in situ</i>	–	–	–	–	–	–	–	0.00066	11
Greenland Sea	68.2°N, 22.67°W, 300 m	1	Kiel <i>in situ</i>	–	–	–	–	–	–	–	0.00209	11
„	68.2°N, 22.67°W, 870 m	1	Kiel <i>in situ</i>	–	–	–	–	–	–	–	0.00054	11
Norwegian Sea	62.11°N, 4.57°E–66.43°N, 10.32°E	2	On-lineGFF/XAD	–	–	–	–	–	–	–	0.0014±0.00023	12
Barents Sea	77.50°N, 29.54°E–88.57°N, 1.13°W	4	On-lineGFF/XAD	–	–	–	–	–	–	–	0.00055±0.00043	12
„	76°58'6''N, 32°59'6''W	1	Large volume & XAD	–	–	–	–	–	–	0.00164	0.00086	13
„	76°07'N, 32°56'W	1	Large volume & XAD	–	–	–	–	–	–	0.00211	0.00125	13
Laptev Sea	72–75°N, 115–135°W (1993–96)	16	Kiel <i>in situ</i> ; dissolved phase	–	0.018 (0.0008–0.060)	–	0.00093 (0.0001–0.00178)	–	–	0.0034 (0.0001–0.011)	–	14
Laptev Sea	72–75°N, 115–135°W (1993–96)	15	Kiel <i>in situ</i> , particle phase	–	0.0002 (<0.0001–0.0003)	–	0.00065 (0.00036–0.00087)	–	–	0.005 (0.0006–0.022)	–	14
Chukotka Peninsula, Russia	Lavrentiya (0–29 m)	3 pools (n=3)	SPE	1.04±0.70	0.2	0.12	0.56	–	<0.05	2.31±0.54	–	15

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

## References

- Jantunen and Bidleman, 2002, Tundra 1999; ΣHCHs = α-HCH and γ-HCH; ΣCHLs = sum of *cis*-chlordan, *trans*-chlordan, oxychlordan, *cis*-nonachlor, *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide.
- Strachan, 2002, Strachan *et al.*, 2000, JOIST cruise, 1997; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = α-HCH and γ-HCH; ΣCHLs = sum of *cis*-chlordan, *trans*-chlordan, oxychlordan, *cis*-nonachlor, *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209).
- Strachan, 2002; Strachan *et al.*, 2000; Tundra 99; see footnote 2.
- Macdonald *et al.*, 2001.
- Strachan *et al.*, 2000; ODEN cruise; see footnote 2.
- Harner *et al.*, 1999; ΣHCHs = α- and γ-HCH.
- Hoekstra *et al.*, 2002b; see footnote 4.
- Strachan *et al.*, 2000; Northwater; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = α-HCH and γ-HCH; ΣCHLs = sum of *cis*-chlordan, *trans*-chlordan, oxychlordan, *cis*-nonachlor,

- trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209).
- Muir *et al.*, 2002a; see footnote 4, except ΣHCHs = γ-HCH only.
- Lakaschus *et al.*, 2002; ΣHCHs = α-HCH and γ-HCH.
- Schultz-Bull *et al.*, 1998. Sum of 23 PCB congeners including 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.
- Sobek and Gustafsson, 2002. Sum of CB8, 28, 52, 70, 90/101, 110, 123, 149, 118, 153, 132, 105, 138, 158/160, 180, 199, 194.
- Olsson, 2002. Sum of CB8, 28, 52, 70, 90/101, 110, 123, 149, 118, 153, 132, 105, 138, 158/160, 180, 199, 194.
- Utschakowski, 1998. ΣPCBs = sum of 40 congeners. Two high PCB values, 1428 pg/L and 97.3 pg/L omitted from the mean. Sample numbers refer to number analyzed for PCBs.
- RAIPON/AMAP/GEF Project, 2001; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordan, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62; ΣCBz = 1,2,3,4-, 1,2,3,5-, and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene.



Annex Table 9. Concentrations of major OC groups in marine surficial sediments (ng/g dw) from the Arctic.

Region	Location	Year	Water depth	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	Toxaphene	OCS	Reference
Canada	Beaufort	Sep-1998	370 m	1	–	–	–	0.04	0.13	0.08	0.78	–	–	1
„	M'Clure Strait	Sep-1998	499 m	1	–	–	–	0.01	0.04	0.05	0.29	–	–	1
„	V. Melville Sound	Sep-1997	203 m	1	–	–	–	0.02	0.08	0.04	0.28	–	–	1
„	Barrow Strait	Sep-1998	221 m	1	–	–	–	0.07	0.11	0.28	1.42	–	–	1
„	Peary Channel	Aug-1998	635 m	1	–	–	–	0.04	0.06	0.08	0.68	–	–	1
„	Penny Strait	Aug-1998	307 m	1	–	–	–	0.01	0.01	0.01	0.09	–	–	1
„	Barrow Strait	Sep-1997	264 m	1	–	–	–	0.35	0.19	0.23	2.70	–	–	1
„	Massey Sound	Aug-1998	292 m	1	–	–	–	0.06	0.12	0.13	0.45	–	–	1
„	Wellington Channel	Aug-1998	252 m	1	–	–	–	0.15	0.08	0.07	0.36	–	–	1
„	Norwegian Bay	Aug-1998	492 m	1	–	–	–	0.06	0.20	0.12	2.76	–	–	1
„	Lancaster Sound	Aug-1998	550 m	1	–	–	–	0.01	0.06	0.09	0.75	–	–	1
„	Strathcona Sound	Sep-1998	229 m	1	–	–	–	0.04	0.12	0.09	2.57	–	–	1
„	Jones Sound	Sep-1998	844 m	1	–	–	–	0.18	0.20	0.44	2.87	–	–	1
„	E Hudson Bay	Sep-1993	133 m	1	–	–	–	0.14	0.30	0.51	8.37	–	–	2
„	N Baffin Bay	Jul-1998	265-560 m	14	–	–	–	0.16	0.17	0.27	1.80	–	–	3
N Norway	Tromsø	1997	–	18	range	–	0.20-3.1	0.04-0.79	–	0.34-9.8	3.3-49	–	0.02-0.20	4
„	Harstad	1997	–	14	range	–	0.05-6.7	n.d.-3.8	–	0.23-67	1.8-308	–	0.01-0.38	4
„	Honningsvåg	1997	–	7	range	–	0.39-3.6	0.05-2.1	–	1.1-8.6	14-50	–	0.03-0.13	4
„	Hammerfest	1997	–	9	range	–	0.12-1.7	0.06-0.31	–	0.61-17	8.6-438	–	0.01-0.17	4
Kola Peninsula	Kola Bay	1993	–	5	range	–	0.10-1.5	0.10-0.40	–	2.30-10	11-253	–	<0.1	4
„	Penchenga	1997	–	7	range	–	0.28-1.8	0.08-0.68	–	0.27-37	0.70-32	–	n.d.-0.10	4
„	Guba Zapadnaya Litsa	1997	–	3	range	–	0.15-2.8	0.65	0.04-0.16	0.44-33	112-371	19-85	–	5
„	Kola Bay	1997	–	10	range	–	0.09-1350	5.5-42	0.11-0.47	24.5-2040	31-8740	3.5-681	0.16-0.18	5
„	Guba Penchenga (Liinakhamari)	1997	–	7	range	–	0.28-1.76	0.08 - 0.68	–	0.27 - 36.7	1.11 - 37.9	–	–	6
S Barents Sea	Guba Penchenga (outer areas)	1997	–	3	range	–	0.74-1.33	0.05 - 0.54	–	0.27 - 3.72	1.06 - 4.1	–	–	6
White Sea	Kandalashka Bay	1999	–	1	–	–	0.2	0.80	–	4.4	26.4	–	–	7
Chukota Peninsula	Kanchalan	2000-2001	–	1	–	0.44	–	0.09	n.d.	0.17	1.56	–	–	8
„	Lavrentiya	2000-2001	–	1	–	0.21	0.49	0.06	0.07	0.61	2.79	–	–	8
Kara Sea	–	1993	<10 m	39	mean±SD	–	–	0.36±0.30	<0.10	0.44±0.30	0.41±0.34	–	–	9
Svalbard	Kongsfjorden	1997	363 m	1	–	0.416	–	0.230	0.12	0.377	0.503	<0.1	–	10
„	Kongsfjorden	1997	290 m	1	–	0.121	–	0.098	0.03	0.086	0.139	<0.1	–	10
„	Kongsfjorden	1997	80 m	1	–	–	–	–	–	–	–	–	–	10
„	Questrenna	1997	2224 m	1	–	0.252	–	–	0.03	0.618	0.611	<0.1	–	10
„	Lomfjorden	1997	56 m	1	–	0.0286	–	0.026	0.03	0.016	0.025	<0.1	–	10
„	Linhopen	1997	160 m	1	–	0.166	–	0.191	0.04	0.084	0.124	<0.1	–	10
„	Erik Eriksen Strait	1997	107 m	1	–	0.163	–	0.253	0.05	0.100	0.107	<0.1	–	10
„	Erik Eriksen Strait	1997	80 m	1	–	0.32	–	<1550	0.02	0.0186	0.0466	<0.1	–	10
„	Erik Eriksen Strait	1997	219 m	1	–	0.174	–	0.068	0.07	0.186	0.153	<0.1	–	10
„	Kong Karls Land	1997	117 m	1	–	0.184	–	0.222	0.08	0.134	0.141	<0.1	–	10
„	Storfjorden	1997	109 m	1	–	0.362	–	0.661	0.10	0.131	0.148	<0.1	–	10
Laptev Sea	Offshore Lena River Delta	1993	<10 m	19	mean±SD	–	–	–	–	–	0.525±419	–	–	11

n.d. = not detected.

## References

- Stern and Lockhart, 2001; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners.
- Lockhart, 1997; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners.
- Fisk *et al.*, 2002c; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDTs = *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = 102 congeners.
- Dahle *et al.*, 2000; ΣHCHs = α- and γ-HCH; ΣDDTs = *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 7 congeners (28, 52, 101, 118, 153, 138, 180).
- Savinova *et al.*, 2000a; see Reference 4; Toxaphene = sum of Parlars 26, 32, 50, 62.
- Savinov *et al.*, 2003. See Reference 4.
- Muir *et al.*, 2002a; see Reference 4, except ΣHCHs = γ-HCH only.
- RAIPON/AMAP/GEF Project, 2001; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62, ΣCBz, = 1,2,3,4-, 1,2,3,5- & 1,2,4,5-tetrachlorobenzene, hexachlorobenzene.
- Sericano *et al.*, 2001; ΣHCHs = α-, β-, γ-HCH; ΣPCBs = sum of all measurable congeners.
- Evans *et al.*, 2002; ΣCBz = HCB; ΣHCHs = α-, γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor, heptachlor epoxide; ΣDDTs = *o,p'*- and *p,p'*-DDE, -DDD and -DDT; ΣPCBs = sum of 7 congeners (101, 105, 118, 138, 153, 156, 180); Toxaphene = sum of Parlars 26, 32, 50, 62.
- Utschakowski, 1998.

Annex Table 10. Concentrations of organochlorines in marine invertebrates and marine and anadromous fish.

Species	Region	Location	Tissue	Year	Sex	Length, cm	% lipid	n	Statistic	ΣCBz	HCb	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference
<b>Marine invertebrates (ng/g ww)</b>																						
Phytoplankton	Barents Sea	Bjørnøya	-	2000	-	-	-	-	-	-	0.03	<0.1	<0.1	0.09	0.65	0.34*	0.30	<0.1	-	<0.1	-	1
<b>Zooplankton</b>																						
<b>Copepods</b>																						
<i>Calanus</i> spp. <sup>a</sup>	Beaufort Sea	Holman, NWT	whole body	1999	-	-	48.90±6.60	5	mean±SE	3.83±0.58	-	6.21±1.08	3.32±0.85	5.33±0.94	33.8±6.1	-	38.4±3.3	-	-	-	-	2
"	"	Kaktovik, AK	whole body	1998	-	-	43.3±4.2	5	mean±SE	2.16±0.28	-	5.61±1.66	1.13±0.19	3.22±0.62	29.2 ±7.26	-	32.4±9.26	-	-	-	-	2
"	Chukchi Sea	Barrow, AK	whole body	2000	-	-	27.3±0.9	3	mean±SE	2.89±0.35	-	5.08±0.64	0.79±0.45	1.18±0.95	12.6±0.74	-	26.0±7.50	-	-	-	-	2
<i>Calanus hyperboreus</i>	Baffin Bay	-	whole body	1998	-	-	6.3±0.70	20	mean±SE	0.43±0.05	-	2.5±0.79	0.85±0.19	0.84±0.15	5.1±0.63	-	-	-	-	-	-	3
"	N of Svalbard	-	whole body	1999	-	-	9.5±0.8	6	mean±SD	-	0.11±0.01	1.03±0.19	1.10±0.14	0.66±0.11	1.25±0.15	-	-	-	-	-	-	4
"	Greenland Sea	-	whole body	1999	-	-	-	5	mean±SD	-	0.06±0.01	2.26±0.30	2.41±0.16	0.86±0.31	1.54±0.22	-	-	-	-	-	-	4
<i>Metridia longa</i>	Baffin Bay	-	whole body	1998	-	-	2.1±0.12	3	mean±SE	0.32±0.09	-	0.78±0.11	1.3±0.12	1.8±0.27	7.6±1.9	-	-	-	-	-	-	3
<i>Euchaeta glacialis</i>	"	-	whole body	1998	-	-	5.4±0.33	3	mean±SE	0.59±0.03	-	1.3±0.11	2.8±0.11	2.5±0.12	6.0±0.04	-	-	-	-	-	-	3
Copepods (90% <i>C. glacialis</i> )	Barents Sea	Bjørnøya	whole body	1995	-	-	1.45	2	mean	-	0.20	0.34	0.17	0.35	0.65	-	-	-	-	-	-	5
Mainly copepods	"	Bjørnøya	whole body	2000	-	<1 mm	3.26	-	-	-	0.17	0.36	0.17	0.53	2.50	1.43*	0.29	0.38	-	0.12	-	1
<b>Euphausiids</b>																						
<i>Mysis oculata</i>	Baffin Bay	-	whole body	1998	-	-	5.0±0.53	7	mean±SE	0.35±0.05	-	0.58±0.18	0.73±0.18	1.4±0.11	5.8±0.46	-	-	-	-	-	-	3
<i>Pandalus</i> sp.	"	-	whole body	1998	-	-	4.9	2	mean	0.65	-	1.5	1.7	2	29.9	-	-	-	-	-	-	3
<i>Pandalus borealis</i>	Barents Sea	S & SE Svalbard	whole body	1991	-	-	1.9	3	median	-	-	-	-	-	130	-	-	-	-	-	-	6
<i>Thysanoessa inermis</i> (90%)	Barents Sea	Bjørnøya	whole body	1995	-	-	6.35	2	mean	-	1.05	2.16	0.79	0.76	18.1	-	-	-	-	-	-	5
<i>Thysanoessa inermis</i>	Greenland Sea	-	whole body	1999	-	-	10.7±0.4	4	mean±SD	-	2.11±0.12	2.78±0.10	1.99±0.21	1.17±0.20	2.12±0.42	-	-	-	-	-	-	4
<b>Amphipods</b>																						
<i>Themisto libellula</i> (adults)	Baffin Bay	-	whole body	1998	-	-	2.2±0.40	4	mean±SE	0.43±0.04	-	1.7±0.25	3.8±0.45	3.1±0.53	8.8±1.3	-	-	-	-	-	-	3
<i>Themisto libellula</i> (juvs)	"	-	whole body	1998	-	-	3.1	1	mean	0.29	-	0.81	0.9	0.55	3.2	-	-	-	-	-	-	3
<i>Anonyx mugax</i>	"	-	whole body	1998	-	-	2.5±0.31	5	mean±SE	2.2±0.57	-	10.0±4.7	5.0±0.32	5.0±0.29	16.5±4.0	-	-	-	-	-	-	3
"	Cumberland Sound	Pangnirtung	whole body	1999	-	-	4.2	1	mean	5.7	-	26.2	20.5	19.8	54.6	-	-	-	-	-	-	3
<i>Themisto libellula</i>	Barents Sea	Bjørnøya	whole body	1995	-	-	3.35	2	mean	-	0.68	1.10	0.75	0.63	1.54	-	-	-	-	-	-	5
"	N of Svalbard	-	whole body	1999	-	-	5.8±0.8	1	mean±SD	-	0.76	0.93	1.82	1.32	2.18	-	-	-	-	-	-	4
"	Greenland Sea	-	whole body	1999	-	-	-	3	mean±SD	-	1.72±0.42	1.65±0.50	5.04±0.51	4.74±0.16	4.56±0.62	-	-	-	-	-	-	4
<b>Miscellaneous</b>																						
<i>Eukrobia hamata</i> & <i>Sagitta elegans</i>	N of Svalbard	-	whole body	1999	-	-	1.4±0.5	1	mean±SD	-	0.19	0.14	0.76	0.59	0.86	-	-	-	-	-	-	4
<i>Apherusa glacialis</i>	Greenland Sea	-	whole body	1999	-	-	-	1	mean±SD	-	0.25	0.36	0.65	0.44	0.68	-	-	-	-	-	-	4
"	N of Svalbard	-	whole body	1999	-	-	7.7±0.8	3	mean±SD	-	1.56±0.20	1.49±1.15	2.30±0.57	1.40±0.48	2.48±0.61	-	-	-	-	-	-	4
"	Greenland Sea	-	whole body	1999	-	-	-	2	mean±SD	-	1.47±0.06	2.65±0.09	1.83±0.15	1.02±0.12	1.90±0.08	-	-	-	-	-	-	4
<i>Gammarus wilkitzkii</i>	N of Svalbard	-	whole body	1999	-	-	3.9±0.3	8	mean±SD	-	1.18±0.25	1.53±1.47	1.22±0.39	0.73±0.37	1.49±0.34	-	-	-	-	-	-	4
"	Greenland Sea	-	whole body	1999	-	-	-	6	mean±SD	-	1.62±0.40	3.44±2.51	1.09±0.59	0.37±0.25	1.62±0.59	-	-	-	-	-	-	4
<i>Onisimus glacialis</i> & <i>Onisimus nansenii</i>	N of Svalbard	-	whole body	1999	-	-	10.1±0.4	1	mean±SD	-	3.34	2.19	5.17	2.51	5.1	-	-	-	-	-	-	4
<i>Sagitta</i> spp.	Greenland Sea	-	whole body	1999	-	-	-	2	mean±SD	-	2.70±0.78	6.03±0.38	5.81±5.43	4.19±4.66	8.24±6.25	-	-	-	-	-	-	4
Mainly comb jellies, arrow worms & medusa	N Baffin Bay	-	whole body	1999	-	-	2.1±0.30	6	mean±SE	0.31±0.16	-	0.23±0.06	0.33±0.09	0.36±0.04	2.2±0.25	-	-	-	-	-	-	3
Crustaceans from the gullet of little auks	Barents Sea	Bjørnøya	whole body	2000	-	>1mm	4.08	-	-	-	0.33	0.32	0.39	0.98	11.5	7.40*	0.67	0.64	-	0.42	-	1
<i>Calanus</i> spp, <i>Th. libellula</i> , ctenophores, gasteropods <sup>a</sup>	"	"	whole body	2000	-	-	7.03	-	-	-	0.94	1.23	0.48	1.52	4.69	2.11*	2.16	3.28	-	1.33	-	1
<i>Calanus</i> spp, <i>Th. libellula</i> , ctenophores, gasteropods <sup>a</sup>	Barents Sea	S & SE Svalbard	whole body	1991	-	-	0.26	6	median	-	-	-	-	-	230	-	-	-	-	-	-	6
<b>Benthic Invertebrates</b>																						
<b>Echinodermata</b>																						
<i>Gorgonocephalus arcticus</i>	Baffin Bay	-	soft tissue	1998	-	-	8.5±1.2	3	mean±SE	0.64±0.12	-	3.7±1.4	26.3±6.9	25.5±7.5	28.1±9.6	-	-	-	-	-	-	3
"	Cumberland Sound	Pangnirtung	soft tissue	1999	-	-	11.4±1.3	3	mean±SE	2.0±0.30	-	13.5±0.94	48.5±5.8	27.1±4.8	31.6±4.1	-	-	-	-	-	-	3
<i>Ctenodiscus crispatus</i>	Baffin Bay	-	soft tissue	1998	-	-	1.4±1.8	2	mean±SE	0.21±0.12	-	0.04±0.01	0.03±0.02	0.08±0.08	2.0±2.2	-	-	-	-	-	-	3
<b>Bivalves</b>																						
<i>Y. thraciaeformis</i>	Baffin Bay	-	soft tissue	1998	-	-	2.3	1	mean±SE	0.22	-	0.04	0.07	0.33	3.4	-	-	-	-	-	-	3
<i>Mytilus edulis</i> (Blue mussel)	N Quebec	Quaqtaq	soft tissue, 1kg pooled	1998	-	-	4.95	2	mean	0.57	-	2.79	1.30	1.29	29.0	6.7	-	1.20	0.0	-	-	3
"	"	Kuujuaq	soft tissue, 1kg pooled	1998	-	-	2.95	2	mean	1.11	-	1.37	0.97	0.86	45.8	12.8	-	0.77	0.03	-	-	3
"	"	Kangipsujuaq (Wakeham)	soft tissue, 1kg pooled	1998	-	-	4.75	2	mean	0.63	-	2.94	1.83	1.48	13.8	4.38	-	1.25	0.01	-	-	3
"	"	Kangisqualjuuaq (George River)	soft tissue, 1kg pooled	1998	-	-	2.00	2	mean	0.29	-	1.12	0.79	0.49	7.01	20.9	-	0.55	0.02	-	-	3
"	Labrador	Makkovik	soft tissue, 1kg pooled	1998	-	-	2.16	3	mean	2.52	-	1.85	9.29	1.89	5.83	1.27	-	1.25	0.24	-	-	3
"	"	Nain	soft tissue, 1kg pooled	1998	-	-	2.49	2	mean	2.06	-	1.69	7.10	1.42	7.09	1.49	-	1.36	0.01	-	-	3
"	N Quebec	Deception Bay	soft tissue, 1kg pooled	1998	-	-	1.65	3	mean	1.20	-	0.61	2.22	0.24	3.68	0.81	-	0.39	0.07	-	-	3
"	SW Greenland	Usuk	soft tissue	2000	-	-	2.0	1 pool	mean	-	-	0.28	1.1	0.44	0.78	0.64	0.21	-	-	-	-	7
"	Iceland	9 sites around	soft tissue	1998	-	4-6	0.41±0.06	10	mean±SE	-	0.21±0.04	0.46±0.10	0.90±0.53	1.82±0.74	5.56±2.04	-	0.94±0.38	-	-	-	-	8
"	"	9 sites around	soft tissue	1999	-	4-6	0.28±0.04	9	mean±SE	-	0.13±0.01	0.38±0.04	0.50±0.05	0.75±0.11	2.69±0.57	-	0.77±0.12	-	-	-	-	8
"	Faroe Islands	-	soft tissue	1997	-	-	2.0	1 pool (n=57)	maximum	n.d.	-	0.1	-	0.7	0.7	-	-	-	-	-	-	9
<i>M. truncata</i>	Frobisher Bay	Iqaluit	soft tissue	1993	-	-	1.5±0.26	6	mean±SE	0.27±0.04	-	0.49±0.03	0.94±0.19	0.64±0.06	7.8±0.89	-	-	-	-	-	-	3
<b>Isopods</b>																						
<i>S. entamon</i> <sup>a</sup>	Chukchi/Beaufort Sea	Barrow, AK	whole body	1998	-	-	2.1±0.4	3	mean±SE	0.36±0.04	-	1.7±0.3	0.61±0.05	0.50±0.07	3.1±0.16	-	0.72±0.34	-	-	-	-	10

Continued next page.

Annex Table 10 continued.

Species	Region	Location	Tissue	Year	Sex	Length, cm	% lipid	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference	
Decapods																							
Deep sea shrimp	SW Greenland	Nuuk	muscle	2000	-	-	0.94±0.12	11	mean±SD	0.69±0.19	-	0.06±0.04	0.51±0.11	0.15±0.07	1.70±2.93	0.69±0.87	-	-	-	-	-	11	
<i>Chionoecetes opilio</i> (Snow crab)	"	"	muscle	2000	-	-	0.74±0.17	5	mean±SD	0.48±0.18	-	0.36±0.16	3.52±0.87	2.15±0.82	3.83±2.50	1.97±1.34	-	-	-	-	-	11	
"	"	"	hepatopancreas	2000	-	-	5.97±5.62	5	mean±SD	3.03±2.83	-	2.36±1.64	5.25±4.47	37.9±46.2	69.1±75.7	41.0±44.8	-	-	-	-	-	11	
<i>Hyas araneus</i> (Spider crab)	Barents Sea	N Bjørnøya	muscle	1999	-	<5	6.76	1 pool (n=12)	-	-	1.21	1.75	0.91	3.77	13.5	8.55	1.54	1.55	-	0.90	-	1	
"	"	"	muscle	1999	-	>5	9.00	1 pool (n=11)	-	-	2.32	2.22	1.09	6.84	26.4	15.8	2.15	1.98	-	1.24	-	1	
"	"	SW Bjørnøya	muscle	2000	-	<5	9.47	1 pool (n=16)	-	-	1.70	1.57	1.27	4.55	19.1	12.0	1.30	1.19	-	0.74	-	1	
"	"	"	muscle	2000	-	>5	9.79	1 pool (n=12)	-	-	2.03	1.53	0.42	4.24	16.5	10.0	1.46	1.11	-	0.72	-	1	
"	"	Svalbard	hepatopancreas	1998-99	-	-	-	38	mean	-	-	-	-	-	16.59	-	-	-	-	-	-	12	
Gastropods (snails)																							
<i>Buccinum undatum</i>	Barents Sea	Svalbard	upper soft parts	1998-99	-	-	-	33	mean±SD	-	0.250±0.127-	-	-	-	5.72±6.52	-	-	-	-	-	-	12	
<i>Buccinum glaciale</i>	"	"	upper soft parts	1998-99	-	-	-	6	mean±SD	-	0.339±0.318-	-	-	-	5.69±5.59	-	-	-	-	-	-	12	
Worms ( <i>Nephtys</i> sp., Sipunculid, wormtube) <sup>a</sup>	Kara Sea	-	whole body	1993	-	-	-	-	mean±SD	-	-	1.0±0.55	0.46±0.38	1.6±1.9	2.6±0.69	-	-	-	-	-	-	13	
Crustaceans (isopods, amphipods) <sup>a</sup>	"	-	whole body	1993	-	-	-	-	mean±SD	-	-	3.8±2.0	5.6±3.9	18±24	14±6.1	-	-	-	-	-	-	13	
Bivalves (clams) <sup>a</sup>	"	-	whole body	1993	-	-	-	-	mean±SD	-	-	8.4±4.0	7.3±4.1	26±35	21±20	-	-	-	-	-	-	13	
Marine fish (ng/g ww)																							
<i>Anarchichas minor</i> (Spotted wolffish)	SW Greenland	Usuk	liver	2000	m	-	15.5	1 pool	mean	-	4.0	4.4	44.4	30	29	26	8.5	-	-	-	-	7	
<i>Boreogadus saida</i> (Arctic or polar cod)	Alaska	Barrow	whole body	1998	-	10.5±0.2	4.43±0.42	3 pools	mean±SE	1.05±0.34	0.83±0.08	1.62±0.20	1.10±0.13	0.57±0.05	2.38±0.60	-	-	0.41±0.07	-	0.31±0.07	-	2	
"	"	"	whole body	2000	-	15.4±0.5	3.45±0.45	9	mean±SE	3.63±0.32	1.79±0.18	1.65±0.26	3.41±0.56	2.76±0.28	11.9±2.18	-	8.04±1.45	0.51±0.08	0.05±0.01	0.18±0.11	-	2	
"	Baffin B. (Greenland)	Qaanaq	muscle, with skin	1998	f,m	-	1.21±0.71	8	mean±SD	1.17±0.32	-	0.93±0.53	3.13±0.97	2.61±0.62	3.73±1.46	-	-	-	-	-	-	14	
"	Jan Mayen	-	liver	1995-96	-	-	55.1±3.7	2	mean±SD	-	9.4±0.5	10.8±1.1	25.5±5.1	30.7±3.7	74.7±4.4	-	-	-	-	-	-	15	
"	Barents Sea	Svalbard	pooled extracts	1995	-	-	7.1±0.7	3	mean	-	2.77	2.63	5.40	1.21	7.67	-	-	-	-	-	-	5	
<i>Gadus ogac</i> (Greenland cod)	Beaufort Sea	Holman, NWT	whole body	1999	-	47.0±2.2	2.61±0.75	10	mean±SD	1.11±0.48	1.03±0.45	2.40±0.97	2.44±1.32	0.71±0.42	0.89±0.51	-	-	-	-	-	-	2	
<i>Gadus morhua</i> (Atlantic cod)	SW Greenland	Nuuk	muscle	2000	m,f	-	0.67±0.05	9	mean±SD	0.27±0.10	-	0.13±0.07	0.43±0.09	0.60±0.30	3.96±3.27	1.44 ±1.86	-	-	-	-	-	3.35	11
"	"	"	liver	2000	m,f	-	57.6±3.81	3	mean±SD	17.5±2.29	-	15.1±0.99	65.2±19.9	82.8±18.6	162±24.4	53.0±8.61	-	-	-	-	-	5.13±0.85	11
"	"	Usuk	liver	2000	m	-	34.9	1 pool	-	-	18.2	8.3	244	156	178	161	41.4	-	-	-	-	7	
"	"	"	liver	2000	f	-	43.9	1 pool	-	-	20.9	11.1	138	102	120	105	28.4	-	-	-	-	7	
"	Iceland	E, NE, NW of Iceland	liver	1999	m,f	30-45	47.4±2.2	23	mean±SE	-	32.6±1.8	10.5±0.6	82.8±2.5	92.1±3.11	80.1±3.1	-	81.7±2.7	-	-	-	-	8	
"	"	NE, NW, W of Iceland	liver	2000	m,f	30-45	41.0±3.4	21	mean±SE	-	15.6±1.5	6.38±0.6	51.7±3.4	72.8±3.7	68.0±2.1	-	69.0±5.2	-	-	-	-	8	
"	Faroe Islands shelf	-	liver	1997	-	-	56.9	45	mean	-	-	-	20	40	25	-	-	-	-	-	-	9	
"	Barents Sea	Svalbard	pooled extracts	1995	-	-	3.8±0.2	3	mean±SE	-	2.47	1.56	3.80	1.60	7.79	-	-	-	-	-	-	5	
<i>Gadus</i> sp. (all)	White Sea	-	muscle	1999-2000	-	-	1.01±0.30	8	mean±SD	0.25±0.06	0.17±0.05	0.26±0.14	0.52±0.12	5.10±2.80	15.6±7.57	6.16±3.46	1.47±0.27	-	-	-	-	16	
<i>Clupea pallazi</i> (Herring)	"	-	whole fish	1999-2000	-	-	7.80±0.24	3 pools (n=10)	mean±SD	6.31±2.89	2.36±0.43	3.40±1.10	3.69±0.18	21.3±2.34	41.2±5.57	17.7±3.10	28.5±1.65	-	-	-	-	16	
<i>Eliginus navaga</i> (Navaga)	"	-	whole fish	1999-2000	-	-	1.54±0.11	3 pools (n=10)	mean±SD	5.95±1.52	0.62±0.05	1.71±0.26	4.30±2.98	15.8±4.30	40.6±5.92	20.7±5.81	1.41±0.39	-	-	-	-	16	
<i>Myoxocephalus scorpius</i> (Shorthorn sculpin)	Centr. W Greenland	Qeqertarsuaq	liver	1999	m	26.4±7.2	19.6±7.0	5	mean±SD	2.9±1.1	2.5±0.9	5.7±2.0	6.7±2.8	6.1±3.7	10.7±5.4	6.2±3.0	-	2.8±1.1	0.07±0.04	-	-	17	
"	"	"	liver	1999	f	31.1±4.9	14.6±4.5	15	mean±SD	2.8±2.0	2.4±1.7	4.6±2.6	7.9±6.4	6.0±4.4	14.9±11.3	7.4±5.3	-	2.7±2.2	0.12±0.12	-	-	17	
"	"	"	liver	2000	m	21.6±2.8	9.4±1.3	4	mean±SD	1.9±0.6	1.7±0.5	2.8±0.3	6.3±5.6	7.1±4.2	52.7±40.5	14.7±8.9	-	2.5±0.9	0.19±0.17	-	-	17	
"	"	"	liver	2000	m	29.6±4.7	11.1±3.0	11	mean±SD	3.8±1.2	1.7±0.6	2.9±0.9	3.8±1.2	4.4±1.9	29±13.4	8.4±2.9	-	2.5±1.0	0.06±0.02	-	-	17	
"	NE Greenland	Ittoqqortoormiit	liver	1999	-	-	19.3±7.5	20	mean±SD	12.9±5.5	11.2±4.9	14.2±5.2	49.7±25.9	44.3±23.0	101±41	47±19	-	13.3±6.7	0.96±0.38	-	-	17	
"	"	"	liver	2000	m	20.9±1.9	23.3±10.5	6	mean±SD	13.0±5.9	11.0±5.0	14.4±4.8	40.3±17.3	37.9±18.8	137±37	59±15	-	11.9±4.9	0.92±0.30	-	-	17	
"	"	"	liver	2000	f	24.8±2.5	22.0±6.7	13	mean±SD	13.1±3.2	10.9±2.8	13.6±3.8	38.1±10.6	40.6±14.2	111±27	46±16	-	11.4±2.7	0.78±0.30	-	-	17	
"	SW Greenland	Usuk	liver	2000	m	-	19.1	1 pool	-	-	6.1	4.2	84.8	34.3	42.7	39.1	36.3	-	-	-	-	7	
"	"	Usuk,Igaliko, Qaqortoq	liver	2000	f	-	17.7	3 pools	mean±SD	-	9.8±5.7	4.6±1.2	48.9±4.2	41.8±9.5	140±160	128±146	37.7±2.3	-	-	-	-	7	
"	Faroe Islands	-	liver	1999	-	18.97±5.44	7.77±3.3	13	mean	-	2.19±1.3	-	97.87	76.46±105.854.1±53.36	-	8.43	-	1.58±1.56	-	-	-	18	
"	"	-	liver	2000	-	23.3±4.3	6.22±4.1	15	mean	-	1.1±0.5	-	24.8	98.0±55.0	38.1±39.1	-	12.62	-	0.75±0.81	-	-	18	
"	"	-	liver	2001	-	25.33±3.7	6.22±4.2	5 pools	mean	-	2.05±1.4	-	11.8	38.74±49.0	66.5±25.10	-	2.67	-	1.95±0.81	-	-	18	
<i>Myoxocephalus quadricornis</i> (Fourhorn sculpin)	Alaska	Barrow	whole body	1998	-	-	1.44±0.48	7	mean±SE	2.67±0.79	0.24±0.06	0.35±0.12	0.44±0.06	0.38±0.07	6.46±1.07	-	-	0.12±0.04	0.06±0.02	0.09±0.01	-	2	
<i>Myoxocephalus</i> spp. (sculpin)	White Sea	-	muscle & skin	-	-	-	1.11±0.09	-	mean±SD	0.13±0.05	0.10±0.04	0.24±0.14	1.52±0.81	14.4±7.49	26.3±7.12	12.6±4.95	-	-	-	-	-	16	
<i>Limanda limanda</i> (dab)	Iceland	NW, SW, E, W of Iceland	liver	1999	m,f	20-35	11.4±0.48	6	mean±SE	-	4.08±0.44	1.53±0.05	8.87±1.48	14.3±2.49	18.8±1.35	-	8.33±1.55	-	-	-	-	8	
"	"	SW, NW, W of Iceland	liver	2000	m,f	20-35	15.2	1	-	-	5.00	1.50	16.5	34.8	30.1	-	28.6	-	-	-	-	8	
<i>Raja radiata</i> (Starry ray)	SW Greenland	Usuk	liver	2000	m	-	42.0	1 pool	-	-	17.2	12.6	101	78	88	80	19.3	-	-	-	-	7	
<i>Reinhardtius hippoglossoides</i> (Turbot/Greenl. halibut)	Cumberland Sound	Pangnirtung	muscle, with skin	1999	-	60.8±6.0	18.6±2.44	4	mean±SE	10.2±1.56	-	14.8±1.38	31.1±7.25	15.0±3.1	11.1±3.97	-	-	-	-	-	-	3	
"	SW Greenland	Nuuk	muscle	2000	m,f	-	9.87±2.96	6	mean±SD	4.71±1.96	-	0.88±0.54	14.5±8.21	26.7±17.1	33.9±19.7	11.5±7.04	-	-	-	-	-	11	
<i>Sebastes marinus, mentella</i> (Redfish)	"	"	muscle	2000	m,f	-	2.48±1.56	5	mean±SD	0.92±0.69	-	0.66±0.57	3.45±2.77	4.75±3.96	9.31±6.06	3.17±2.05	-	-	-	-	-	11	

Continued next page.

Annex Table 10 continued.

Species	Region	Location	Tissue	Year	Sex	Length, cm	% lipid	n	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	Endrin	ΣPBDEs	Reference
<i>Reinhardtius hippoglossoides</i> (Turbot/Greenland halibut)	Cumberland Sound SW Greenland	Pangnirtung Nuuk	muscle, with skin	1999	–	60.8±6.0	18.6±2.44	4	mean±SE	10.2±1.56	–	14.8±1.38	31.1±7.25	15.0±3.1	11.1±3.97	–	–	–	–	–	–	3
<i>Sebastes marinus, mentella</i> (Redfish)	“	“	muscle	2000	m,f	–	9.87±2.96	6	mean±SD	4.71±1.96	–	0.88±0.54	14.5±8.21	26.7±17.1	33.9±19.7	11.5±7.04	–	–	–	–	–	11
<i>Mallotus villosus</i> (Capelin)	“	“	muscle	2000	m	–	1.78±0.69	10	mean±SD	0.92±0.25	–	0.45±0.20	3.38±0.68	5.62±1.01	6.12±1.45	1.84±0.41	–	–	–	–	–	11
<i>Hippoglossoides platessoides</i> (Long rough dab)	Jan Mayen	–	liver	1995-96	–	–	1.7±0.7	2, pooled	mean±SD	–	0.4±0.2	0.4±0.2	0.8±0.1	2.8±1.4	12.5±5.5	–	–	–	–	–	–	15
<i>Eutrigla gurnardus</i> (Grey gurnard)	“	–	liver	1995-96	–	–	14.4±8.7	3	mean±SD	–	4.9±2.7	3.2±1.6	10.6±5.9	24.5±14.7	170.4±88.7	–	–	–	–	–	–	15
<i>Pollachius virens</i> (Coalfish)	“	–	liver	1995-96	–	–	7.7±4.4	2, pooled	mean±SD	–	2.2±1.7	1.3±1.1	6.9±6.3	9.4±9.4	27.1±22.7	–	–	–	–	–	–	15
<i>Leptagonus decagonus</i> (Atlantic poacher)	“	–	liver	1995-96	–	–	51.5±5.9	5	mean±SD	–	8.4±1.1	10.5±1.1	25.2±4.7	24.1±2.2	62.6±6.2	–	–	–	–	–	–	15
<i>Lumpenus maculatus</i> (Daubed shanny)	“	–	liver	1995-96	–	–	11.2±4.5	2, pooled	mean±SD	–	3.8±1.6	2.3±1.1	18.0±8.9	30.9±15.0	91.6±21.8	–	–	–	–	–	–	15
<i>Lycodes vahli</i> (Checkered eelpout)	“	–	liver	1995-96	–	–	2.6±4.5	2, pooled	mean±SD	–	0.7±0.4	0.3±0.4	1.8±0.2	4.6±0.2	17.2±0.7	–	–	–	–	–	–	15
<i>Somniosus microcephalus</i> (Greenland shark)	Cumberland Sound Davis Strait	Pangnirtung	liver	1999	–	283.6±5.7	39.7±3.6	15	mean±SE	192±30	–	29.1±3.5	1050±185	4158±830	2000±426	–	–	–	–	–	–	3
			liver	1997	–	135±0	55.9±2.1	2	mean±SE	53.5±4.74	–	27.1±10.5	262±28.4	817±39.5	590±176	–	–	–	–	–	–	3
<b>Anadromous fish</b>																						
<i>Salvelinus alpinus</i> (Arctic char)	Alaska	Barrow	fillet	1998	–	–	8.54±3.45	2	mean±SE	0.83±0.79	–	2.46±1.88	–	1.64±1.08	12.0±5.06	–	11.4±9.06	1.08±0.82	–	0.45±0.11	–	2
	E Canada	Kangirsuk	muscle, with skin	1998	–	–	8.53±2.21	9	mean±SD	1.11±0.64	0.90±0.56	1.14±0.75	1.19±0.63	1.66±1.25	17.6±14.9	4.49±4.15	–	0.94±0.67	0.02±0.05	–	–	19
	“	Makkovik	muscle, with skin	1998	–	–	4.88±1.23	2	mean±SD	9.26±23.3	9.13±23.4	1.66±0.68	1.22±0.57	2.61±2.7	29.8±54.8	3.43±1.53	–	1.07±0.79	0.06±0.14	–	–	19
	“	Nain	muscle, with skin	1998	–	–	6.86±2.00	7	mean±SD	1.22±0.8	1.22±0.63	2.64±1.30	1.71±0.63	3.38±2.98	63.3±77.4	7.99±7.72	–	1.47±0.70	0.12±0.22	–	–	19
	“	Quaqtaq	muscle, with skin	1998	–	–	6.06±6.12	7	mean±SD	1.13±0.96	1.09±0.81	1.01±1.02	1.13±0.81	1.16±0.96	14.2±16.1	4.53±4.69	–	1.06±0.86	0.02±0.04	–	–	19
	“	Wakeham	muscle, with skin	1998	–	–	3.9±3.58	7	mean±SD	1.03±0.54	0.91±0.45	0.47±0.27	1.29±0.69	1.25±0.73	12.7±8.80	2.97±1.86	–	0.99±0.70	0.04±0.03	–	–	19
	Finland	Lake Pahtajärvi	muscle	1999	f	304±11.9	1.34±0.49	5	mean±SD	–	0.10±0.49	0.02±0.02	0.04±0.02	0.18±0.02	0.15±0.06	–	–	–	–	–	–	20
	“	“	muscle	1999	m	346±17.1	1.09±0.42	5	mean±SD	–	0.09±0.03	0.01±0.02	0.02±0.03	0.24±0.21	0.28±0.46	–	–	–	–	–	–	20
	“	Lake Kilpisjärvi	muscle	1999	?	210	0.97	5, pooled	–	–	0.26	0.07	0.37	0.93	3.33	–	–	–	–	–	–	20
	Russia	Lavrentiya, Chukotka Peninsula	liver	2001	f	–	–	8, pooled	–	–	–	0.81	2.18	6.20	31.3	–	–	–	2.62	–	–	21
	“	“	liver	2001	m	–	–	12, pooled	–	–	–	0.53	1.84	5.47	10.6	–	–	–	2.69	–	–	21
	“	“	muscle	2001	f	–	–	8, pooled	–	–	–	0.21	2.28	6.53	26.1	–	–	–	0.33	–	–	21
	“	“	muscle	2001	m	–	–	12, pooled	–	–	–	0.24	0.67	1.62	6.14	–	–	–	0.23	–	–	21
<i>Salmo salar</i> (Atlantic salmon)	SW Greenland	Nuuk	muscle	2000	m,f	–	11.2±5.59	7	mean±SD	1.84±0.72	–	3.77±1.90	5.10±2.04	12.0±3.97	17.9±5.26	5.96±1.71	–	–	–	–	–	11
<i>Oncorhynchus gorbuscha</i> (Pink salmon)	Alaska	Barrow	muscle	1998	–	–	1.25±0.05	7	mean±SE	0.23±0.08	–	0.24±0.05	–	2.58±0.46	6.64±1.08	–	2.07±0.44	0.21±0.04	–	0.23±0.03	–	2
<i>Oncorhynchus keta</i> (Chum salmon)	Russia	Lavrentiya, Chukotka Peninsula	liver	2001	f	–	–	11, pooled	–	–	–	0.65	2.74	4.15	3.59	–	–	–	<0.05	–	–	21
	“	“	liver	2001	m	–	–	9, pooled	–	–	–	0.72	2.80	5.97	4.66	–	–	–	0.07	–	–	21
	“	“	muscle	2001	f	–	–	11, pooled	–	–	–	0.56	0.75	2.21	1.34	–	–	–	<0.05	–	–	21
	“	“	muscle	2001	m	–	–	9, pooled	–	–	–	0.58	0.53	1.52	1.13	–	–	–	<0.05	–	–	21
<i>Coregonus sardinella</i> (Least cisco)	Alaska	Nuiqsut (Colville R.)	fillet	2000	f	–	3.10±1.07	2	mean±SE	4.89±2.93	4.14±2.82	0.85±0.01	1.32±0.90	1.47±0.62	2.32±0.92	–	–	0.12±0.10	0.02±0.01	0.14±0.10	–	2

<sup>a</sup> concentrations are dry weight.

PCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

\* sum of 7 congeners (101, 105, 118, 138, 153, 156, 180).

#### References

1. Evenset *et al.*, 2002; ΣHCHs = α-HCH; ΣCHLs = oxychlordanes; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 33 congeners (18, 28, 31, 33, 37, 47, 52, 60, 66, 74, 99, 101, 105, 114, 118, 122, 123, 128, 138, 141, 149, 153, 156, 157, 167, 170, 180, 183, 187, 189, 194, 206, 209); Toxaphene = sum of Parlars 26, 50, 62.
2. Hoekstra *et al.*, 2002b; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209); Toxaphene = total toxaphene.
3. Fisk *et al.*, 2002a; ΣCBz = sum of 1,2,4,5-tetraCBz, 1,2,3,4-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*-chlordanes, *trans*-chlordanes, *cis*-nonachlor, *trans*-nonachlor, and oxychlordanes; ΣDDTs = sum of *p,p'*- and *o,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 86 congeners (1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 95/66, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 194, 196/203, 189, 208, 195, 207, 194, 205, 206, 209).
4. Borgå *et al.*, 2002a; ΣCHLs = sum of oxychlordanes, *cis*- and *trans*-chlordanes, and *trans*-nonachlor; ΣDDTs = sum of *p,p'*-DDE, -DDD and -DDT; ΣPCBs = sum of 9 congeners (31, 28, 52, 99, 118, 153, 105, 138, 180).
5. Borgå *et al.*, 2001; see footnote 4; data converted from lipid weight to wet weight using mean % lipid content.
6. Joiris *et al.*, 1997; ΣPCBs = Aroclor 1254.
7. Denmark, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*-DDT and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 13 congeners; Toxaphene = sum of Parlars 26, 40, 41, 44, 50, and 62.
8. Yngvadóttir and Halldórsdóttir, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *trans*-nonachlor, *cis*- and *trans*-chlordanes, oxychlordanes; ΣDDTs = sum of *p,p'*-DDE, -DDD and -DDT, and *o,p'*-DDT; ΣPCBs = sum of 7 congeners (28, 52, 101, 118, 138, 153, 180); Toxaphene = sum of Parlars 26, 50, and 62.
9. Larsen and Dam, 1999; ΣCBz = HCB; ΣHCHs = γ-HCH; ΣDDTs = *p,p'*-DDE; ΣPCBs = congener 153.
10. Fisk *et al.*, 2002a; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz,



- and hexaCBz;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -,  $\gamma$ -, and  $\delta$ -HCH;  $\Sigma$ CHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide;  $\Sigma$ DDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT;  $\Sigma$ PCBs = sum of 101 congeners.
11. Muir and Johansen, 2001;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor;  $\Sigma$ DDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT;  $\Sigma$ PCBs = sum of 104 congeners peaks (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
12. Hop *et al.*, 2001;  $\Sigma$ PCBs = sum of 33 congeners (18, 28, 31, 33, 37, 47, 52, 60, 66, 74, 99, 101, 105, 114, 118, 122, 123, 128, 138, 141, 149, 153, 156, 157, 167, 170, 180, 183, 187, 189, 194, 206, 209).
13. Sericano *et al.*, 2001;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHLs = *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, and heptachlor epoxide,  $\Sigma$ DDTs = *o,p'*- and *p,p'*-DDE, -DDD, and -DDT;  $\Sigma$ PCBs = sum of all measurable congeners.
14. Fisk, 2002c; see footnote 10.
15. Gabrielsen *et al.*, 1997;  $\Sigma$ PCBs = sum of 33 congeners.
16. Muir *et al.*, 2003;  $\Sigma$ CBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -,  $\gamma$ -, and  $\delta$ -HCH;  $\Sigma$ CHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide;  $\Sigma$ DDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE and -DDT;  $\Sigma$ PCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197,

- 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209).
17. Denmark, 2002;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHLs = sum of *cis*- and *trans*-chlordanes, oxychlordanes, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor;  $\Sigma$ DDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT;  $\Sigma$ PCBs = sum of 104 congeners peaks (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
18. Hoydal *et al.*, 2001;  $\Sigma$ DDTs = *p,p'*-DDE;  $\Sigma$ PCBs = congener 153.
19. Muir *et al.*, 2001a;  $\Sigma$ CBz = sum of 1,2,3,4-tetra-, penta-, and hexachlorobenzene;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHLs = sum of heptachlor, heptachlor epoxide, *cis*-chlordanes, *trans*-chlordanes, *cis*-nonachlor, and *trans*-nonachlor;  $\Sigma$ DDTs = sum of *p,p'*-DDE, -DDD and -DDT, and *o,p'*-DDT;  $\Sigma$ PCBs = sum of 103 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
20. Mannio, 2002;  $\Sigma$ HCHs = sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHLs = sum *cis*-chlordanes, *trans*-nonachlor;  $\Sigma$ DDTs = *p,p'*-DDE, -DDD, and -DDT;  $\Sigma$ PCBs = sum 15 congeners (28, 31, 52, 66, 101, 149, 118, 153, 105, 138, 187, 156, 180, 170).
21. RAIPON/AMAP/GEF Project, 2001;  $\Sigma$ HCHs =  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH;  $\Sigma$ CHL = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordanes, and *cis*- and *trans*-nonachlor;  $\Sigma$ DDTs = sum of *o,p'* and *p,p'*-DDE, -DDD, and -DDT;  $\Sigma$ PCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187).

Annex Table 11. Recent mean concentrations (ng/g ww) of organochlorines in Arctic seabirds.

Species/Location	Tissue	Year	Sex	Age	% lipid	n(#pools)	Statistic	$\Sigma$ CBz	HCb	$\Sigma$ HCHs	$\Sigma$ CHLs	$\Sigma$ DDTs	$\Sigma$ PCBs	$\Sigma$ PCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	References	
<i>Somateria mollissima</i> (common eider)																			
Nuuk, SW Greenland	liver	1999	m,f	-	5.1±0.3	5	mean±SD	3.63±0.95	-	0.81±0.23	12.8±5.55	5.78±4.14	25.5±17.2	13.2±11.0	12.4±4.92	-	-	-	1
"	muscle	1999	m,f	-	3.9±0.8	10	mean±SD	2.50±0.82	-	0.58±0.20	4.57±0.69	3.84±1.40	23.0±31.2	12.9±20.0	14.3±7.43	-	-	-	1
W Iceland	liver	1993, Feb	-	-	3.5	3	mean(range)	-	0.66(0.52-0.75)	-	-	17.4 (8.1-26.8)	90.1 (78.0-99.3)	-	-	-	-	-	2
"	muscle	1993, Feb	-	-	3.4	3	mean(range)	-	0.58(0.25-0.83)	-	-	11.0 (10.0-12.2)	97.5 (85.4-111)	-	-	-	-	-	2
"	muscle	1993, Feb	f	-	2.67	10	mean	-	0.85	-	-	10.3	77	-	-	-	-	-	2
"	muscle	1993, Feb	m	-	2.97	7	mean	-	0.77	-	-	13.1	90.7	-	-	-	-	-	2
"	liver	1993, May	-	-	4.1	4	mean(range)	-	1.96(1.39-2.48)	-	-	23.1 (14.0-35.6)	66.6 (58.4-85.7)	-	-	-	-	-	2
"	muscle	1993, May	-	-	2.8	4	mean(range)	-	2.02(0.53-3.97)	-	-	17.9 (2.99-36.4)	58.9 (13.6-109)	-	-	-	-	-	2
"	muscle	1993, May	f	-	4.26	7	mean	-	2.36	-	-	17.1	80.7	-	-	-	-	-	2
"	muscle	1993, May	m	-	3.14	7	mean	-	3.43	-	-	24.7	100	-	-	-	-	-	2
"	liver	1993, June	-	-	3.9	4	mean(range)	-	6.9(4.4-11.8)	-	-	47.4 (31.8-72.4)	207 (164-253)	-	-	-	-	-	2
"	muscle	1993, June	-	-	2.8	4	mean(range)	-	6.5(0.59-14.9)	-	-	50.9 (7.74-144)	252 (77.0-688)	-	-	-	-	-	2
"	muscle	1993, June	f	-	1.42	5	mean	-	4.07	-	-	44.4	247	-	-	-	-	-	2
"	muscle	1993, June	m	-	2.36	8	mean	-	4.46	-	-	36.2	182	-	-	-	-	-	2
"	muscle	1993, Nov	-	-	5.2	7	mean(range)	-	1.93(0.34-3.85)	-	-	21.2 (5.4-49.0)	169 (21.2-404)	-	-	-	-	-	2
"	muscle	1993, Nov	f	-	3.09	3	mean	-	1.93	-	-	19.6	146	-	-	-	-	-	2
"	muscle	1993, Nov	m	-	3.94	4	mean	-	1.66	-	-	21.8	181	-	-	-	-	-	2
Lavrentiya, Chukotka Peninsula, Russia	liver	2001	f	1-3	-	11(1)	-	-	-	0.81	0.55	0.70	2.45	-	-	-	-	0.15	3
"	muscle	2001	f	1-3	-	11(1)	-	-	-	<0.10	<0.05	0.26	3.38	-	-	-	-	<0.05	3
<i>Somateria spectabilis</i> (king eider)																			
Nuuk, SW Greenland	liver	1999	-	-	5.2±0.3	5	mean±SD	4.13±1.79	-	1.28±0.72	16.0±9.69	5.70±2.66	27.3±10.7	8.71±2.77	13.5±10.2	-	-	-	1
"	muscle	1999	-	-	3.9±1.2	10	mean±SD	3.14±1.40	-	1.03±0.38	4.98±1.78	3.88±2.15	14.5±5.55	6.46±2.33	8.67±11.2	-	-	-	1
<i>Bucephala clangula</i> (goldeneye)																			
Kola Peninsula, Russia	liver	2001	f	1-3	-	5(1)	-	-	-	0.36	1.43	6.9	21.5	-	-	-	-	0.09	3
"	liver	2001	m	1-3	-	5(1)	-	-	-	0.68	2.2	13.9	26.2	-	-	-	-	0.08	3
"	muscle	2001	f	1-3	-	5(1)	-	-	-	0.50	0.23	1.29	2.47	-	-	-	-	<0.05	3
"	muscle	2001	m	1-3	-	5(1)	-	-	-	0.47	0.37	1.84	5.76	-	-	-	-	<0.05	3
<i>Aythya marila</i> (greater scaup)																			
"	liver	2001	f	1-3	-	2(1)	-	-	-	0.36	0.12	15.4	14.3	-	-	-	-	0.14	3
"	liver	2001	m	2-3	-	2(1)	-	-	-	0.29	0.13	13.2	18.1	-	-	-	-	0.06	3
"	muscle	2001	f	1-3	-	2(1)	-	-	-	<0.10	0.13	1.25	3.48	-	-	-	-	0.11	3
"	muscle	2001	m	2-3	-	2(1)	-	-	-	<0.10	0.15	1.51	4.41	-	-	-	-	0.10	3
<i>Anas acuta</i> (northern pintail)																			
"	liver	2001	f	1-3	-	4(1)	-	-	-	0.07	0.24	0.47	2.41	-	-	-	-	<0.05	3
"	liver	2001	m	1-3	-	6(1)	-	-	-	0.10	0.16	0.34	2.23	-	-	-	-	-	3
"	muscle	2001	f	1-3	-	4(1)	-	-	-	<0.10	0.06	0.41	1.61	-	-	-	-	0.13	3
"	muscle	2001	m	1-3	-	6(1)	-	-	-	<0.10	0.06	0.23	1.44	-	-	-	-	0.16	3

Continued next page.

Annex Table 11 continued.

Species/Location	Tissue	Year	Sex	Age	% lipid	n(#pools)	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	References
Dudinka, Taymir Peninsula, Russia	liver	2001	f	1-3	–	4(1)	–	–	–	0.11	0.20	1.51	2.20	–	–	–	–	3
„	liver	2001	m	1-3	–	1	–	–	–	0.10	0.32	2.21	2.13	–	–	–	–	3
„	muscle	2001	f	1-3	–	4(1)	–	–	–	0.34	0.08	10.8	2.87	–	–	–	0.09	3
„	muscle	2001	m	1-3	–	1	–	–	–	0.06	0.14	2.06	2.19	–	–	–	<0.05	3
„	liver	2001	f	2-3	–	1	–	–	–	0.27	0.32	1.49	1.30	–	–	–	<0.05	3
„	muscle	2001	f	2-3	–	1	–	–	–	0.19	0.47	1.67	2.05	–	–	–	<0.05	3
<i>Anas penelope</i> (wigeon)																		
Pechora Basin, Russia	liver	2001	f	1-3	–	3(1)	–	–	–	0.2	1.77	2.81	4.51	–	–	–	–	3
„	liver	2001	m	1-3	–	2(1)	–	–	–	0.79	2.01	2.53	–	–	–	–	–	3
„	muscle	2001	f	1-3	–	3(1)	–	–	–	0.33	0.42	0.34	2.75	–	–	–	<0.05	3
„	muscle	2001	m	1-3	–	2(1)	–	–	–	0.74	0.17	0.47	–	–	–	–	<0.05	3
<i>Anas crecca</i> (green-winged teal)																		
Dudinka, Taymir Peninsula, Russia	liver	2001	m	1-2	–	2(1)	–	–	–	0.07	1.27	5.75	4.34	–	–	–	0.10	3
„	muscle	2001	m	1-2	–	2(1)	–	–	–	0.40	0.36	3.67	3.37	–	–	–	<0.05	3
Khatanga, Taymir Peninsula, Russia	liver	2001	m	1-2	–	2(1)	–	–	–	1.15	1.54	6.91	5.34	–	–	–	<0.05	3
„	muscle	2001	m	1-2	–	2(1)	–	–	–	0.97	0.92	4.87	2.39	–	–	–	<0.05	3
<i>Clangula hyernalis</i> (oldsquaw)																		
Khatanga, Taymir Peninsula, Russia	liver	2001	f	1-2	–	4(1)	–	–	–	9.01	8.06	31.7	42.3	–	–	–	1.28	3
„	liver	2001	m	1-2	–	2(1)	–	–	–	6.49	7.80	23.7	38.3	–	–	–	0.90	3
„	muscle	2001	f	1-2	–	4(1)	–	–	–	1.78	2.06	9.63	17.2	–	–	–	0.09	3
„	muscle	2001	m	1-2	–	2(1)	–	–	–	2.64	3.70	10.6	10.7	–	–	–	0.08	3
<i>Melanitta</i> sp. (scoter)																		
Kanchalan, Chukotka Peninsula, Russia	liver	2001	f	2-3	–	1	–	–	–	<0.10	0.11	0.94	2.16	–	–	–	0.10	3
„	liver	2001	m	2-3	–	2(1)	–	–	–	<0.10	0.10	0.64	1.82	–	–	–	<0.05	3
„	muscle	2001	f	2-3	–	1	–	–	–	<0.10	<0.05	0.31	1.64	–	–	–	0.05	3
„	muscle	2001	m	2-3	–	2(1)	–	–	–	<0.10	<0.05	0.54	1.58	–	–	–	0.08	3
<i>Anser albifrons</i> (greater white-footed goose)																		
Dudinka, Taymir Peninsula, Russia	liver	2001	f	1-3	–	9(1)	–	–	–	0.13	0.17	8.62	2.31	–	–	–	<0.05	3
„	muscle	2001	f	1-3	–	9(1)	–	–	–	0.18	<0.05	3.45	1.92	–	–	–	0.06	3
„	liver	2001	m	1-3	–	4(1)	–	–	–	0.21	0.23	8.01	2.72	–	–	–	<0.05	3
„	muscle	2001	m	1-3	–	4(1)	–	–	–	0.18	0.07	4.89	3.72	–	–	–	0.15	3
Khatanga, Taymir Peninsula, Russia	liver	2001	f	1-4	–	5(1)	–	–	–	0.11	0.45	8.75	2.13	–	–	–	0.28	3
„	muscle	2001	f	1-4	–	5(1)	–	–	–	0.12	0.15	4.67	2.45	–	–	–	0.21	3
„	liver	2001	m	1-4	–	5(1)	–	–	–	0.15	0.58	11.0	2.97	–	–	–	0.23	3
„	muscle	2001	m	1-4	–	5(1)	–	–	–	0.18	0.12	6.04	3.01	–	–	–	0.07	3
<i>Anser fabalis</i> (bean goose)																		
Kanchalan, Chukotka Peninsula, Russia	liver	2001	f	1-2	–	2(1)	–	–	–	<0.10	0.25	2.01	2.05	–	–	–	0.31	3
„	muscle	2001	f	1-2	–	2(1)	–	–	–	<0.10	0.06	0.48	0.94	–	–	–	<0.05	3
<i>Phalacrocorax aristolelis</i> (shag)																		
Sklinna Island, central Norway	egg, yolk sac	1995	–	–	6.8±1.8	10	mean±SD	–	–	–	–	–	1220±5	–	–	–	–	4
<i>Ardea cinerea</i> (grey heron)																		
Froya, central Norway	egg, yolk sac	?	–	–	5.8±3.0	4	mean±SD	–	–	–	–	–	2070±1820	–	–	–	–	5
Finnfjordoy, N Norway	egg, yolk sac	?	–	–	11.0±4.5	7	mean±SD	–	–	–	–	–	2450 ±1410	–	–	–	–	5
<i>Alle alle</i> (dovekie or little auk)																		
N Baffin Bay	liver	1998	–	–	4.0±0.2	7(2)	mean±SE	2.9±0.47	2.0±0.33	3.5±0.82	10.2±1.9	9.4±1.5	15.7±3.2	–	–	–	–	6,7
„	fat	1998	–	–	63.7±2.1	10	mean±SE	102±8.3	63.5±5.4	149±12.4	459±32.8	364±63.5	635±63.5	–	–	175±14.8	10.1±1.1	6,7
Jan Mayen	liver	1995-96	–	–	6.7±1.8	5	mean±SD	–	11.3±3.9	2.6±0.9	21.6±9.7	60.4±25.2	105.7±34	–	–	–	2.0±0.6	8
Bjørnøya	muscle	1999	m	–	2.31 (2.24-2.43)	3	mean (range)	–	3.56 (2.01-5.57)	1.26 (0.82-1.71)	4.94 (3.35-6.50)	38.9 (27.4-46.9)	208 (83.3-313)	59.3** (12.4-80.5)	8.12 (5.38-12.4)	10.8 (4.21-22.3)	–	9
„	guano	–	–	–	0.43	–	–	–	0.54	0.11	0.01	–	6.42	3.09**	–	<0.04	–	9
„	posterior colon content	1999	–	–	2.21	15(1)	–	–	5.98	0.25	2.68	15.6	60.6	29.1**	4.47	3.51	–	9
<i>Uria lomvia</i> (thick-billed murre or Brünnich's guillemot)																		
St. George Isl., Bering Sea	eggs	2000	–	–	8.4±3.6	10	mean±SD	–	52.5±10	–	9.48±2.55	97.4±14.7	86.7±21	–	–	2.31±1.7	0.625±0.23	10
Bogoslof Isl., Bering Sea	eggs	2000	–	–	11.1±1.4	7	mean±SD	–	51.7±12	–	10.6±3.43	122±34.3	87.5±29	–	–	10.3±13	0.975±0.60	10
Prince Leopold Isl., Lancaster Sound	eggs	1998	–	–	12.9±0.4	15(5)	mean±SE	53±2.0	–	17±1.0	30±0.4	100±7.0	130±9.0	–	–	15±1.0	3.0±0.5	11
N Baffin Bay	liver	1998	–	–	4.1±0.5	10	mean±SE	11.7±1.3	9.8±1.2	2.6±0.35	6.7±0.72	33.4±3.8	37.3±5.1	–	–	–	–	6,7
„	fat	1998	–	–	60.0±1.9	10	mean±SE	178±15.0	149±13.6	50.6±4.1	158±14.9	658±57.2	772±62.2	–	–	76.4±9.6	11.4±1.1	6,7
Nuuk, SW Greenland	liver	1999	–	–	5.4±0.7	5	mean±SD	6.4±2.1	–	0.63±0.17	4.57±1.31	8.11±3.41	20.2±6.10	8.81±2.95	53.0±15.1	–	–	1
„	muscle	1999	–	–	3.5±0.4	19	mean±SD	3.9±2.4	–	0.73±0.24	4.97±5.17	6.97±5.22	19.6±11.1	8.43±5.03	27.3±9.19	–	–	1
Barents Sea, Svalbard	liver	1995	–	–	2.5±0.5	10	mean	–	14.4	1.12	4.8	26.9	44.5	–	–	–	–	12
Bjørnøya	eggs	2000	–	–	12.5 (11.8-13.7)	3	mean (range)	–	51.4 (43.9-58.9)	3.69 (1.66-5.79)	10.4 (5.59-13.6)	136 (117-166)	194 (123-274)	92.0** (87.4-133)	31.8 (17.8-40.8)	10.6 (4.29-14.2)	–	9

Continued next page.

Annex Table 11 continued.

Species/Location	Tissue	Year	Sex	Age	% lipid	n(#pools)	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	References
<i>Cephus grylle</i> (black guillemot)																		
Saglek Bay (reference site), Labrador	liver	1999	m,f	22d	–	10	mean (range)	–	2.3	–	0.39	1.8	25 (18-34)	–	–	1.2	–	13
Saglek Bay (island sites), Labrador	liver	1999	m,f	22d	–	10	mean (range)	–	3.4	–	0.64	4.1	73 (46-117)	–	–	1.2	–	13
Saglek Bay (beach site), Labrador	liver	1999	m,f	22d	–	11	mean (range)	–	8.1	–	n.d.	8.9	830 (340-2030)	–	–	n.d.	–	13
N Baffin Bay	liver	1998	–	–	3.4±0.3	9	mean±SE	8.6±1.4	7.5±1.2	5.7±0.71	24.9±2.5	54.5±11.0	59.6±11.3	–	–	–	–	6,7
”	fat	1998	–	–	60.0±6.0	7	mean±SE	277±32.4	222±25.4	161±20.6	681±75.9	602±74.6	1263±177	–	–	135±15	16.7±2.3	6,7
Qeqertarsuaq, central W Greenland	egg	2001	–	–	10.3±0.6	7	mean±SD	–	22.9±4.7	5.6±0.9	–	44.5±9.8	109±23	82.2±17.7	57.9±14.8	–	–	14
”	liver	1999	m	juveniles	4.3±0.1	2	mean±SD	10.6±5.9	9.6±5.4	2.2±1.5	15.8±9.3	22.3±2.7	53.6±11.5	22.7±2.6	–	6.8±5.6	0.5±0.04	15
”	liver	1999	f	juveniles	3.9	1	mean	13.6	12.2	2.3	17.4	26.1	16.3	44.7	–	3.8	1.36	15
”	liver	1999	m	adults	4.9±1.2	6	mean±SD	17.1±6.4	15.7±6.0	3.3±1.4	25.9±7.6	37.3±14.3	103±34	50.8±18.4	–	8.6±3.7	1.3±0.5	15
”	liver	1999	f	adults	5.5±0.9	9	mean±SD	19.7±3.0	17.6±2.8	3.5±0.6	26.8±3.9	39.9±10.5	135±62	69.5±36.3	–	7.5±2.5	1.6±0.7	15
”	liver	2000	m	adults	6.6±1.8	9	mean±SD	–	20.6±7.0	3.7±1.3	22.5±6.9	28.3±7.2	45.2±16.4	34.6±12.8	28.2±11.1	–	–	14
”	liver	2000	f	adults	6.0±1.5	10	mean±SD	–	14.6±6.3	2.8±1.2	16.8±7.2	24.4±9.9	34.3±16.6	27.1±14.3	21.6±10.9	–	–	14
Ittoqqortoormiit, NE Greenland	egg	1999	–	–	9.3±0.6	10	mean±SD	34.1±5.3	30.1±4.8	17.3±2.4	60±8.9	110±17.0	293±55	153±31	–	12±1.7	3.4±0.8	15
”	liver	2000	m	juveniles	5.0	1	mean	–	9.4	4.1	9.7	12.4	20.9±	14.6	13.5	–	–	14
”	liver	2000	f	juveniles	5.4±3.1	2	mean±SD	–	10.4±1.9	4.7±1.0	12.7	14.5±0.9	27.1±5.0	17.7±2.0	13.1	–	–	14
”	liver	2001	m	adults	3.6±1.8	8	mean±SD	–	20.1±13.1	4.0±3.7	26.9±9.7	47.8±33.3	102±63	64.4±48.0	32.0±15.7	–	–	14
”	liver	2002	f	adults	8.7±4.1	4	mean±SD	–	31.7±19.9	6.9±5.1	71.9±40.3	79.9±47.9	164±106	113±74	64.1±47.9	–	–	14
W Iceland	muscle	1991, '92, '96	–	–	2.5	10	mean	–	6.4	0.36	–	22.5	114	–	–	–	–	22
Faroe Islands	liver	1995-1996	–	adults	–	56	mean	–	3.85	–	–	15.8	51.0	–	–	–	–	16
Jan Mayen	liver	1995-1996	–	–	7.8	1	mean	–	13.7	3.7	31.5	456	1197	–	–	–	12.8	8
Barents Sea, Svalbard	liver	1995	–	–	4.4±0.8	10	mean	–	14.7	1.58	12.8	31.5	96.3	–	–	–	–	12
Bjørnøya	eggs	2000	–	–	12.1	3	mean	–	38.1	1.39	10.5	137	251	148**	14.7	5.06	–	9
					11.1-12.8		range	–	33.7-42.8	1.09-1.65	10.3-10.6	125-143	221-290	105-196	13.0-16.3	3.59-7.23	–	–
<i>Rissa tridactyla</i> (black-legged kittiwakes)																		
Prince Leopold Isl., Lancaster Sound	eggs	1998	–	–	9.0±0.2	15(5)	mean±SE	29±4.0	–	5.0±0.8	58.0±9.0	60.0±8.0	281±22.0	–	–	9.0±0.2	10.0±7.0	11
N Baffin Bay	liver	1998	–	–	4.1±0.5	10	mean±SE	13.2±1.2	11.6±1.1	2.5±0.3	25.8±3.0	58.9±6.5	143±13.4	–	–	–	–	6,7
”	fat	1998	–	–	72.4±3.9	8	mean±SE	223±84.7	186±19.1	32.8±3.0	385±33.9	921±147	3343±480	–	–	103±10.7	64.2±10.0	6,7
Nuuk, SW Greenland	liver	1999	m,f	–	6.9±2.3	5	mean±SD	9.10±4.62	–	1.09±0.67	4.68±4.65	8.31±5.65	126±125	73.8±73.7	47.9±23.8	–	–	1
”	muscle	1999	m,f	–	14.3±5.4	9	mean±SD	18.8±3.37	–	4.08±0.73	23.9±6.46	30.9±12.1	191±140	98.1±77.0	96.4±32.7	–	–	1
Jan Mayen	liver	1995-1996	–	–	4.1±0.8	6	mean±SD	–	9.6±3.2	0.8±0.3	13.3±4.4	19.6±11.9	378.5±150.6	–	8.6±4.0	–	–	8
Bjørnøya	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	20	520	–	–	–	–	8
Svalbard	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	60	480	–	–	–	–	8
Chornaya Bay, Novaya Zemlya	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	48	400	–	–	–	–	8
Kharlov, Kola Peninsula	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	36	560	–	–	–	–	8
Franz Josef Land	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	76	760	–	–	–	–	8
Chornaya Bay, Novaya Zemlya	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	60	840	–	–	–	–	8
Vajgach Island, southeast of Novaya Zemlya	liver	1995-1996	–	–	est. 4	–	mean	–	–	–	–	32	440	–	–	–	–	8
Barents Sea, Svalbard	liver	1995	–	–	3.6±0.7	10	mean	–	31.2	1.55	16.1	53.0	647	–	–	–	–	12
Bjørnøya	muscle	1999	m	–	9.24	3	mean	–	11.7	1.95	31.7	40.2	736	507**	18.5	10.3	–	9
”	posterior colon content	1999	–	–	(5.54-7.64)	10	range	–	(6.83-17.4)	(1.32-2.95)	(15.0-40.4)	(25.1-68.4)	(332-938)	(188-672)	(17.2-19.8)	(8.08-14.1)	–	9
					2.87		–	–	5.59	0.14	9.39	–	178	107**	–	4.57	–	–
<i>Pagophila eburnea</i> (ivory gull)																		
N Baffin Bay	liver	1998	–	–	3.4±0.6	5	mean±SE	20.1±2.8	18.3±2.6	8.5±2.3	89.7±25.9	260±50.6	325±71.3	–	–	–	–	6,7
”	fat	1998	–	–	81.1±5.0	4	mean±SE	459±129	396±108	112±21.1	1618±297	5717±463	11664±796	–	–	292±41.9	203±15.9	6,7
<i>Larus hyperboreus</i> (glaucous gull)																		
N Baffin Bay	liver	1998	–	–	5.6±0.4	11	mean±SE	30.3±2.1	26.1±1.8	23.9±3.6	120±12.0	382±52	453±75.2	–	–	–	–	6,7
N Baffin Bay	fat	1998	–	–	72.8±4.2	11	mean±SE	508±35.8	427±31.9	327±39.0	1985±196	4336±355	11719±1891	–	–	389±53.9	226±54.5	6,7
Jan Mayen	liver	1995-1996	–	–	4.8±1.0	7	mean±SD	–	228.2±278.7	88.1±213.9	1659±3263	6983±11894	22727±40088	–	–	–	317.5±488.7	8
Jan Mayen	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	2500	7600	–	–	–	–	8
Bjørnøya	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	250	1300	–	–	–	–	8
Svalbard	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	550	1650	–	–	–	–	8
Kolguyev Island, SW of Novaya Zemlya	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	1050	3300	–	–	–	–	8
Franz Josef Land	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	3500	11700	–	–	–	–	8
Chornaya Bay, Novaya Zemlya	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	1200	3600	–	–	–	–	8
Vajgach Island, SE of Novaya Zemlya	liver	1995-1996	–	–	est. 5	–	mean	–	–	–	–	1500	3500	–	–	–	–	8
Barents Sea, Svalbard	liver	1995	–	–	4.1±0.5	10	mean	–	46.0	10.4	227	1720	5350	–	–	–	–	12
Bjørnøya	liver	1996	–	–	5.97	40	mean(range)	–	133(40-428)	–	–	1439	4413	–	–	–	–	17
					(3.6-10.0)		–	–	–	–	–	(226-1294)	(782-17381)	–	–	–	–	–
SE Bjørnøya	liver	1996	m,f	>3 yrs	–	40	mean	–	112	–	321	1250	3760	–	–	–	58	18
Bjørnøya	liver	1999	m,f	–	5.9	15	mean (range)	–	29.7	–	113	570	1261(f), 4206(m)	–	29	–	112	19
”	intestines	1999	–	–	(3.4-11.5)	1	–	–	96.4	–	153	764	4500	–	85	–	–	1
”	muscle	1999	m	–	8.72	3	mean (range)	–	99.1	11.7	390	1610	14400	9550**	175	9.9	–	9
					(6.64-11.0)		–	–	(43.8-159)	(3.5-18.1)	(97.1-831)	(819-2660)	(4270-32300)	(2580-22100)	(51.2-346)	(11.5-253)	–	–
”	guano	–	–	–	0.11	–	–	–	2.91	0.06	0.01	–	85.3	51.9**	–	<0.04	–	9
”	posterior colon content	1999	–	–	5.94	15	–	–	94.6	0.31	153	764	5710	3370**	53.1	35.1	–	9

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Annex Table 11 continued.

Species/Location	Tissue	Year	Sex	Age	% lipid	n(#pools)	Statistic	ΣCBz	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	References
<i>Larus marinus</i> (great black-backed gull)																		
Jan Mayen	liver	1995-1996	-	-	6.9	1	mean	-	195	26.3	543	3140	9565	-	-	-	175	8
<i>Fulmarus glacialis</i> (northern fulmars)																		
Prince Leopold Isl., Lancaster Sound	eggs	1998	-	-	11.3±0.4	15(5)	mean±SE	35±4.0	-	2.0±0.2	96±10	209±22	268±20	-	-	14±1.0	13±2.0	11
N Baffin Bay	liver	1998	-	-	3.8±0.6	10	mean±SE	18.7±1.7	17.4±1.6	1.8±1.7	95.7±6.9	174±22.9	158±18.5	-	-	-	-	6,7
"	fat	1998	-	-	71.9±4.0	10	mean±SE	460±31.2	410±29.9	45.2±3.0	1195±92.1	3502±422	6832±894	-	-	133±17.0	162±24.0	6,7
Jan Mayen	liver	1995-1996	-	-	4.8±0.7	6	mean±SD	-	36.4±19.4	0.6±0.3	172.0±75.7	467.8±488.4	686±735	-	-	-	10.5±8.5	8
<i>Haliaeetus leucocephalus</i> (bald eagle)																		
Kiska Isl., Aleutian Archipelago	eggs	1993-1994	-	-	-	25	geo. mean	-	40	50	-	2750	2000	-	-	40	-	20
Adak Isl., Aleutian Archipelago	eggs	1993-1994	-	-	-	25	geo. mean	-	20	20	-	750	2100	-	-	10	-	20
Tanaga Isl., Aleutian Archipelago	eggs	1993-1994	-	-	-	25	geo. mean	-	20	30	-	800	700	-	-	20	-	20
Amchitka Isl., Aleutian Archipelago	eggs	1993-1994	-	-	-	25	geo. mean	-	20	20	-	950	1700	-	-	20	-	20
<i>Haliaeetus albicilla</i> (white-tailed sea eagle)																		
Norway	eggs	1991-1997	-	-	-	7	mean(range)	-	*	-	1690(617-4320)	*	8960(3080-13000)	-	90(n.d.-243)	*	-	21
<i>Stercorarius skua</i> (great skua)																		
Jan Mayen	liver	1995-1996	-	-	4.7±0.2	3	mean±SD	-	237.7±101.6	17.3±6.0	911.5±911.9	5415±2229	15887±6674	-	-	-	262±94.7	8

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

\* Data for sum of pesticides; sum of HCB, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor epoxide, *p*, *p'*-DDE, and dieldrin are summed and listed together under ΣCHLs.

\*\* sum of 7 congeners (101, 105, 118, 138, 153, 156, 180).

#### References

- Muir and Johansen, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Olafsdottir *et al.*, 1998.
- RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5- and 1,2,4,5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28,31,52,99,101,105,118,128,138,153,156,170,180,183,187); Toxaphene = sum of Parlars 26, 50, 62.
- Murvoll *et al.*, 1999.
- Jenssen *et al.*, 2001.
- Fisk *et al.*, 2001b; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners.
- Fisk, 2002c; see footnote 6.
- Gabrielsen *et al.*, 1997; ΣPCBs = sum of 33 congeners. Concentrations converted to wet weight assuming 4% lipid for black-legged kittiwakes and 5% lipid for glaucous gulls.
- Evenset *et al.*, 2002; ΣHCHs = α-HCH; ΣCHLs = oxychlordane; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 33 congeners (18, 28, 31, 33, 37, 47, 52, 60, 66, 74, 99, 101, 105, 114, 118, 122, 123, 128, 138, 141, 149, 153, 156, 157, 167, 170, 180, 183, 187, 189, 194, 206, 209); Toxaphene = sum of Parlars 26, 50, 62.
- Vander Pol *et al.*, 2002; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 53 congeners.
- Braune *et al.*, 2001b
- Borgå *et al.*, 2001; ΣCHLs = sum of oxychlordane, *cis*- and *trans*-chlordane, *trans*-nonachlor; ΣDDTs = sum of *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 9 congeners (31, 28, 52, 99, 118, 153, 105, 138, 180); converted to lipid weight from wet weight using mean % lipid content.
- Kuzyk *et al.*, 2002; ΣCHLs = *cis*-nonachlor; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 45 congeners; n = 1, 2, 1 for pesticide concentrations.
- Denmark, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*-DDT and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 22 congeners peaks; Toxaphene = sum of Parlars 26, 40, 41, 44, 50, 62.
- Denmark, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Hoydal *et al.*, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣDDTs = *p,p'*-DDE; ΣHCHs = sum of α-, β-, and γ-HCH; ΣPCBs = congener 7; converted to wet weight from lipid weight assuming 5.5% lipid content.
- Henriksen *et al.*, 2000; ΣHCHs = sum of α-, β-, and γ-HCH; ΣPCBs = sum of 9 congeners (28, 52, 99, 101, 118, 138, 153, 170, 180), ΣHCH = sum of α-, β-, and γ-HCH; ΣDDTs = *p,p'*-DDE only.
- Sagerup *et al.*, 2002; ΣCHLs = oxychlordane; ΣDDTs = sum of *p,p'*-DDE and -DDT; ΣPCBs = sum of 9 congeners (28, 52, 101, 99, 118, 153, 138, 180, 170). Converted from lipid weight to wet weight assuming a 5% lipid content.
- Herzke *et al.*, 2003; ΣCHLs = oxychlordane; ΣDDTs = *p,p'*-DDE, ΣPCBs = sum of 15 congeners.
- Anthony *et al.*, 1999; ΣHCHs = β-HCH; ΣPCBs = Aroclor 1260; ΣDDTs = *p,p'*-DDE.
- Herzke *et al.*, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣPCBs = sum of 15 congeners (99, 101, 105, 118, 126, 128, 138, 149, 153, 169, 170, 180, 183, 187, 194); Toxaphene = sum of 8 Parlars (26, 32, 38, 50, 51, 58, 62, 69).
- Olafsdottir *et al.*, 2001; ΣDDTs = sum of *p,p'*-DDT, -DDD and -DDE, and *o,p'*-DDT; ΣHCHs = sum of α-, β-, and γ-HCH; ΣPCBs = sum of 21 congeners.



Annex Table 12. Concentrations (ng/g ww) of organochlorines in pinnipeds inhabiting Arctic waters.

Species/Region/Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Ref.
<b>Ringed seal</b>																		
<b>Alaska</b>																		
Barrow	blubber	1998-1999	f	1-7	87.0±6.47	6	mean±95% CI	127±25.1	115±18.5	391±135	178±43.5	465±140	–	734±511	11.9±2.53	18.4±6.92	–	1
”	blubber	1998-1999	m	1-20	82.2±4.45	14	mean±95% CI	102±3.53	167±44.6	390±75.8	237±47.6	566±89.6	–	479±94.1	18.4±4.56	19.9±3.40	–	1
”	blubber	1988-98	f	3.3±1.9 (n=9)	81.8±1.6	10	mean±95% CI	29.4±9.8	233±97 (n=7)	452±182	429±100	632±78	–	–	38.5±13	9.3±3.4	–	2
”	blubber	1988-98	m	5.6±1.6	86.0±1.5	7	mean±95% CI	12.2±2.8	318±167 (n=5)	855±484	575±2.5	782±254	–	–	41.9±20	14.3±10 (n=5)	–	2
Nome	blubber	1988-98	f	2.9±2.4 (n=5)	86.2±3.7	8	mean±95% CI	18.3±4.3	197±66	269±96	214±65	342±77	–	–	20.0±5.5	7.6±2.8	–	2
”	blubber	1988-98	m	1.0±0.4 (n=5)	87.3±1.6	8	mean±95% CI	23.3±9.2	236±127 (n=3)	195±66	193±52	301±8 (n=3)	–	–	23.4±17 (n=7)	3.3±1.0 (n=7)	–	2
Norton Sound	blubber	1989-95	f	2,?	87±1.0	2	mean±SD	26.0±1.0	–	255±40	194±27	346±17	–	–	26.0±5.0	–	–	3
”	blubber	1989-95	m	1-2,?	89±4.0	6	mean±SD	20.3±13.9	–	157±67	188±63	249±75	–	–	14.8±5.2	–	–	3
<b>Canada</b>																		
Holman, NWT	blubber	2001	f	8-25	85.7±10.7	5	mean±SD	64.0±15.3	234±137	367±87.0	188±37.6	335±19.9	–	–	50.4±4.49	11.1±2.83	–	1
”	blubber	2001	m	2-14	83.0±6.38	5	mean±SD	64.6±29.4	225±131	514±218	263±134	487±306	–	–	44.2±6.93	7.59±2.20	–	1
Hudson Strait (Salluit+Quaqtaq)	blubber	1998	f	–	80.4±10.4	10	mean±SD	5.9±2.8	117±38.4	190±162	405±403	608±500	329±285	266±250	43.7±16.5	5.3±4.9	–	4
”	blubber	1998	m	–	77.0±9.8	11	mean±SD	9.0±13.3	140±91.7	303±493	884±2130	1010±1860	559±1030	309±218	70.6±97.3	10.8±27.4	–	4
Ungava Bay	blubber	1998	f	–	101±16.5	16	mean±SD	8.2±2.8	155±63.3	161±78.6	259±142	513±225	278±127	87.7±55.1	58.3±39.0	4.1±3.1	–	4
(George R, Kangnirsuk, Kuujuaq)	blubber	1998	m	–	92.4±15.7	5	mean±SD	11.4±4.4	299±171	81.1±24.6	728±526	1040±670	542±324	117±82.4	78.4±24.6	6.5±6.5	–	4
”	blubber	1998	f	–	87.3±5.6	6	mean±SD	5.1±3.7	103±41.8	109±62.0	187±107	730±450	397±246	n.d.	32.1±19.8	3.4±3.0	–	4
”	blubber	1998	m	–	88.2±5.5	14	mean±SD	5.2±0.2	90.8±47.9	108±45.1	198±86.0	572±290	321±161	n.d.	31.6±21.2	2.3±1.9	–	4
Nain, Labrador	blubber	1998	m	1-24	–	4	mean/range	–	–	–	228	344-625	–	–	–	–	–	5
Saglek Bay, Labrador	blubber	1997-1998	m	1-11	–	6	range	–	–	–	–	500-9400	–	–	–	–	–	5
Arctic Bay, Nunavut	blubber	2000	f	6.9±3.5	94.1±4.7	7	mean±SD	10.4±4.5	133±87.0	172±43.3	200±108	358±145	174±78	–	57.4±17.0	4.0±4.2	–	6
Pangnirtung, Baffin Island	blubber	1998	f	–	–	3	mean±SD	–	–	–	510±190	585±180	–	207±33	–	–	0.50±0.18	7
”	blubber	1998	m	–	–	3	mean±SD	–	–	–	1200±1660	976±1090	–	185±121	–	–	0.97 (n=2)	7
Grise Fiord, Nunavut	blubber	1998	f	19±3.9	90.4±1.8	15	mean±SE	42±4	148±12	338±38	394±89	483±86	–	–	–	–	–	8
”	blubber	1998	m	14.8±3.0	91.2±1.4	12	mean±SE	47±10	179±38	854±168	963±271	1000±206	–	–	–	–	–	8
<b>NW Greenland</b>																		
Qaanaaq	blubber	1998	f	15.3±3.6	85.6±3.6	14	mean±SE	25±3	68±6	187±27	390±63	476±78	–	–	–	–	–	8
”	blubber	1998	m	14.8±3.3	87.8±4.7	13	mean±SE	38±6	136±31	546±156	944±229	1480±348	–	–	–	–	–	8
Avanersuaq	blubber	1998	m	15.3±13.2	85.8±13.0	14	mean±SD	11.0±3.5	68.1±21.3	187±98	390±225	476±282	269±165	–	33.0±11.0	5.3±4.1	–	9
”	blubber	1998	f	14.9±11.5	87.7±16.2	13	mean±SD	14.4±9.2	136±107	546±539	944±793	1480±1210	894±738	–	74.0±59.0	10.4±10.3	–	9
<b>Central W Greenland</b>																		
Qeqertarsuaq	blubber	1998	m,f	–	92.2±4.9	10	mean±SD	14.8±5.58	74.1±26.9	241±114	439±205	544±231	286±132	219±81.0	–	–	–	10
”	liver	1998	m,f	–	5.5±0.6	5	mean±SD	0.60±0.31	2.2±0.81	9.21±2.61	8.18±3.00	46.7±51.3	7.51±1.44	0.81±0.08	–	–	–	10
”	kidney	1998	m,f	–	3.8±0.5	5	mean±SD	0.33±0.17	0.75±0.20	1.71±0.72	2.07±0.48	6.1±2.5	1.48±0.15	39.6±12.4	–	–	–	10
”	muscle	1998	m,f	–	5.4±6.4	20	mean±SD	0.71±1.0	3.0±5.1	7.5±11.1	13.5±20.3	22.9±27.8	10.9±15.3	8.58±14.5	–	–	–	10
”	blubber	1999	f	3.5±2.9	92.3±3.0	4	mean±SD	8.3±3.4	59.8±19.3	148±49	305±100	403±73	31±5.0	–	42±21	2.3±1.5	–	11
”	blubber	1999	m	1.6±1.9	92.1±6.1	6	mean±SD	6.9±0.8	83.7±28.4	285±102	528±213	645±254	43±11	–	52±29	3.0±1.8	–	11
”	muscle	1999	f	3.4±3.1	5.5±8.1	9	mean±SD	0.4±0.6	2.5±3.9	5.9±9.4	9.8±17.1	23±29	1.7±2.2	–	1.7±2.7	0.09±0.10	–	11
”	muscle	1999	m	1.3±1.4	5.3±5.3	10	mean±SD	0.4±0.5	3.8±6.3	9.3±12.8	17.6±23.9	31±30	2.0±2.1	–	2.2±3.2	0.11±0.12	–	11
”	liver	1999	f	4.3±2.9	5.4±0.6	3	mean±SD	0.4±0.4	2.3±1.0	8.4±3.0	6.8±2.5	69±64	2.1±0.9	–	8.1±2.3	0.11±0.07	–	11
”	liver	1999	m	1	5.6±0.9	2	mean±SD	0.3±0.1	2.0±0.8	10.7±1.4	10.4±2.9	24±0.4	1.8±0.2	–	6.5±3.0	0.14±0.05	–	11
”	kidney	1999	f	4.3±2.9	3.9±0.5	3	mean±SD	0.3±0.1	0.8±0.2	1.5±0.5	2.2±0.7	12±2.0	0.6±0.0	–	1.2±0.7	0.04±0.01	–	11
”	kidney	1999	m	1	3.6±0.5	2	mean±SD	0.2±0.1	0.8±0.2	2.0±1.1	2.1±0.3	8.0±2.0	0.7±0.2	–	0.9±0.2	0.03±0.01	–	11
”	blubber	2000	f	1.0	92.5±3.0	6	mean±SD	6.3±2.1	59.9±19.0	264±107	301±102	235±79	198±63	49.0±73.3	–	–	–	12
”	blubber	2000	m	1.3±1.2	91.2±2.8	14	mean±SD	8.0±2.1	69.5±21.3	268±101	298±148	237±89	203±79	13.2±11.4	–	–	–	12
<b>NE Greenland</b>																		
Ittoqqortoormiit	blubber	1999	f	4.7±2.6	101±16.3	8	mean±SD	12.4±2.3	97.8±63.3	287±140	598±244	921±470	339±470	–	61±14	7.2±4.9	–	13
”	blubber	1999	m	4.7±2.9	102±14.0	11	mean±SD	9.1±5.4	108±64.1	395±368	671±381	969±828	312±227	–	106±87	8.3±10.5	–	13
”	liver	1999	f	4	5.3±0.8	3	mean±SD	0.61±0.10	2.8±1.1	10.7±1.6	12.6±2.7	35.9±18.3	2.67±1.84	–	7.6±1.8	0.62±0.47	–	13
”	liver	1999	m	6.5±4.9	6.6±1.6	2	mean±SD	0.94±0.49	5.1±4.2	34.0±24.2	47.0±38.1	111±86	8.4±6.0	–	8.3±2.2	1.72±1.36	–	13
”	muscle	1999	f	4.7±2.6	12.2±6.9	9	mean±SD	1.99±1.74	11.3±9.0	30.0±22.6	95.0±95.8	181±182	19.2±23.1	–	6.8±5.3	1.67±1.95	–	13
”	muscle	1999	m	4.7±2.9	11.9±10.3	11	mean±SD	1.44±1.57	11.6±19.2	30.9±42.7	42.8±25.7	111±160	8.7±12.7	–	10.6±16.0	0.89±1.24	–	13
”	blubber	2000	f	4.4±2.4	90.0±3.4	11	mean±SD	14.9±6.1	93.1±59.1	461±301	892±304	924±171	719±158	41.4±60.1	–	–	–	12
”	blubber	2000	m	4.5±2.3	88.1±5.9	8	mean±SD	15.1±7.7	83.6±30.3	558±487	1050±1220	1450±1750	1190±1520	26.9±20.5	–	–	–	12
<b>Barents Sea</b>																		
Kongsfjorden, Svalbard	blubber	–	f	4	–	7	mean, 95%CI	–	–	–	–	910, 795-1050	–	35.4	–	–	–	14
”	blubber	–	m	4	–	7	mean, 95%CI	–	–	–	–	1800, 1330-2410	–	43.3	–	–	–	14
”	blubber	–	juv	≤4	–	14	mean, 95%CI	–	–	–	–	444, 398-494	–	23.4	–	–	–	14
”	blubber	1992	f	6.0±1.4	84.6-98.0	4	range	–	–	–	639-1332	1010-2650	–	–	–	–	–	15
”	blubber	1992	m	7.3±3.9	84.9-89.8	4	range	–	–	–	783-5300	696-6500	–	–	–	–	–	15
”	blubber	1992	m,f	1.7±0.6	75.4-95.4	3	range	–	–	–	272-1850	382-1680	–	–	–	–	–	15
”	blubber	1995	m	12 (5-16)	95(91-97)	6	mean (range)	13(10-24)	74(45-126)	804(368-1460)	2580(861-5510)	2660(946-5440)	–	–	–	–	–	16
Svalbard	blubber	1996	f	8.5±6.6	–	11	mean	–	–	–	375	963	702	–	–	–	–	1
”	blubber	1996	m	7.6±5.1	–	8	mean	–	–	–	643	1520	1120	–	–	–	–	17
Kongsfjorden, Svalbard	blood	1994-1996	f	–	0.65±0.13	6	mean	0.11	0.35	0.74	1.07	2.19	–	–	–	0.07	–	18
”	blood	1994-1996	m	–	0.75±0.15</													

Annex Table 12 continued.

Species/Region/Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Ref.	
Russia																			
Sorocskaya Guba Bay, White Sea	blubber	1993	m,f	(1- >15)	-	12	mean (range)	17(14-30)	-	-	3200(2100-6500)	2600(1800-4400)	-	-	-	-	-	-	19
Dvina Bay, White Sea	blubber	1998	m,f	1.1±0.6	86.4±6.0	17	mean±SD	9.9±7.1	42.8±15.0	171±132	1530±1250	1500±931	1070±677	166±233	-	-	8.8±5.5	-	20
Gorlo Basin, White Sea	blubber	2001	f	6.6±3.1	92.2±2.1	9	mean±SD	24.5±15.1	19.2±7.7	39.9±24.9	517±123	1170±388	759±238	-	-	-	-	-	20
"	blubber	2001	m	6.8±3.1	91.0±2.5	15	mean±SD	17.7±3.5	20.8±10.1	40.1±23.9	587±256	1120±445	705±310	-	-	-	-	-	20
Lavrentiya, Chukotka Peninsula	blubber	2001	f	1-3	-	3	mean±SD	-	49.1±11.1	27.6±6.72	48.1±23.9	97.0±9.45	-	-	-	-	1.95±0.31	-	21
"	blubber	2001	m	1-3	-	3	mean±SD	-	63.8±10.9	72.8±24.4	82.4±27.5	110±42.3	-	-	-	-	3.74±0.85	-	21
"	liver	2001	f	1-3	-	3	mean±SD	-	1.29±0.23	0.38±0.06	1.74±0.17	2.58±0.46	-	-	-	-	<0.05	-	21
"	liver	2001	m	1-3	-	3	mean±SD	-	1.10±0.30	0.65±0.47	1.78±0.62	2.44±0.93	-	-	-	-	<0.05	-	21
"	kidney	2001	f	1-3	-	3	mean±SD	-	0.94±0.12	0.69±0.30	1.26±0.50	2.92±0.47	-	-	-	-	<0.05	-	21
"	kidney	2001	m	1-3	-	3	mean±SD	-	0.74±0.33	0.65±0.22	1.28±0.61	1.26±0.35	-	-	-	-	<0.05	-	21
"	muscle	2001	f	1-3	-	3	mean±SD	-	1.49±0.26	0.94±0.12	1.91±0.40	2.65±0.20	-	-	-	-	<0.05	-	21
"	muscle	2001	m	1-3	-	3	mean±SD	-	0.61±0.14	0.60±0.19	1.19±0.32	1.73±0.40	-	-	-	-	<0.05	-	21
Yenisey River estuary, Kara Sea	blubber	1995	m	4.8±2.2	89±3.6	29	mean	25	160	420	3200	3700	-	-	-	-	-	-	33
"	blubber	1995	f	6.3±5.6	92±1.4	9	mean	23	110	360	2100	3100	-	-	-	-	-	-	33
Harp seal																			
Barents Sea																			
E of Svalbard	blubber	1997	-	≤4	-	13	geo. mean (95% CI)	49.1(43.5-76.7)	53.1(52.7-74.0)	249(221-389)	297(278-437)	(247-381)	261	422(418-587)	45.1	-	-	-	22, 23
"	blubber	1995	m	19(6-27)	85(71-96)	7	mean (range)	230(44.2-813)	91.8(35.7-150)	4290(329-7560)	5820(1240-8840)	9440(1780-17300)	-	-	-	-	-	-	16
N of White Sea	blubber	1993	m	10(7-18)	94(92-97)	9	mean (range)	77.1(34.8-183)	84(61.1-112)	1020(324-2240)	1590(514-3730)	2300(704-4860)	-	-	-	-	-	-	16
White Sea																			
Gorlo Basin	blubber	1998	f	14.2±4.1	85.1±3.7	20	mean±SD	43.7±15.1	29.4±8.7	282±134	525±276	904±429	596±319	548±148	-	-	65.2±40.2	-	20
"	blubber	1998	m,f	<0.1	68.3±3.1	10	mean±SD	49.3±29.5	29.8±8.3	157±55.0	340±104	403±123	236±74.9	658±234	-	-	2.9±1.1	-	20
"	blubber	1992	m,f	<0.1	80.4±9.7	11	mean±SD	145±101	69.3±22.0	526±240	710±437	1140±547	795±388	-	-	-	12.0±7.5	-	20
Harbour seal																			
Gulf of Alaska																			
Prince William Sound	blubber	1992-1993	f	adults	89	2	mean±SD	9.0±1.0	-	91±11	139±9.0	233±7.0	-	-	3.5±0.5	-	-	-	3
"	blubber	1992-1993	m	pup,adults	66	3	mean±SD	13.7±2.6	-	281±38	430±67	599±143	-	-	6.3±2.1	-	-	-	3
Barents Sea																			
Jarfjord, N. Norway	blubber	1989-1990	m,f	7 (1-17)	92±2	7	geo. mean	10.1	73.6	647	1900	4490	-	-	-	-	-	-	24
Grey seal																			
Faroe Islands																			
-	blubber	1993-1995	m	>8yr (adults)	-	4	mean	-	-	-	-	4900	-	-	-	-	-	-	25,26
-	blubber	1993-1995	f	adults, pregnant	-	20	mean	-	-	-	-	540	-	-	-	-	-	-	25,26
-	blubber	1993-1995	-	2-4 yrs (juveniles)	-	21	mean	-	-	-	-	1200	-	-	-	-	-	-	25,26
Barents Sea																			
Jarfjord, N Norway	blubber	1989-90	m,f	10 (0.2-26)	93±3	23	geo. mean	40	26	684	1840	5220	-	-	-	-	-	-	24
Bearded seal																			
Alaska																			
Barrow	blubber	2000	f	4-18	76.8±6.30	4	mean±SE	32.0±13.2	98.7±67.5	155±2.12	132±12.7	284±43.0	-	-	9.54±4.04	27.2±1.79	-	-	1
"	blubber	2000	m	2-4	83.1±4.40	3	mean±SE	19.8±2.17	76.6±6.56	232±7.62	188±44.3	305±53.5	-	-	4.95±1.05	10.8±1.44	-	-	1
Norton Sound	blubber	1993-1995	f	-	78	1	-	6.6	-	168	96	199	-	-	4.5	-	-	-	3
"	blubber	1993-1995	m	mature	80±5	5	mean±SD	4.0±2.7	-	152±159	104±133	153±110	-	-	3.5±3.0	-	-	-	3
Barents Sea																			
Kongsfjorden, Svalbard	blood	1994-1996	f	-	0.39±0.13	6	mean±SD	0.018±0.010	0.26±0.18	0.22±0.16	0.18±0.16	0.62±0.51	-	-	-	-	0.050±0.015	-	18
"	blood	1994-1996	m	-	0.39±0.12	6	mean±SD	0.036±0.005	0.310±0.072	0.52±0.11	0.63±0.28	0.97±0.36	-	-	-	-	0.041±0.013	-	18
Russia																			
Dvina Bay, White Sea	blubber	1998	m	2.5	84.8	2	mean	6.7	14.3	396	3360	3540	2860	518	-	-	25.3	-	20
Lavrentiya, Chukotka Peninsula	blubber	2001	m	2-3	-	2	mean±SD	-	8.92±2.72	34.4±3.20	50.5±15.4	87.2±11.8	-	-	-	-	1.22	-	21
"	liver	2001	m	2-3	-	2	mean±SD	-	0.33±0.08	0.64±0.35	1.99±0.42	3.48±0.32	-	-	-	-	0.12	-	21
"	kidney	2001	m	2-3	-	2	mean±SD	-	0.59±0.12	1.63±0.18	2.63±0.76	4.72±0.39	-	-	-	-	<0.05	-	21
"	muscle	2001	m	2-3	-	2	mean±SD	-	<0.10	0.18±0.18	1.31±0.97	2.07±0.11	-	-	-	-	<0.05	-	21
Spotted seal																			
Russia																			
Lavrentiya, Chukotka Peninsula	blubber	2001	f	1-2	-	5	mean±SD	-	60.1±25.5	39.7±16.2	68.9±32.0	127±23.0	-	-	-	-	1.73±0.92	-	21
"	blubber	2001	m	1-2	-	5	mean±SD	-	51.4±13.4	34.2±7.00	48.8±7.03	121±15.6	-	-	-	-	2.44±1.98	-	21
"	liver	2001	f	1-2	-	5	mean±SD	-	1.39±0.53	0.63±0.22	2.47±0.58	3.15±1.12	-	-	-	-	<0.05	-	21
"	liver	2001	m	1-2	-	5	mean±SD	-	1.35±0.31	0.66±0.33	2.47±1.31	2.66±0.91	-	-	-	-	<0.05	-	21
"	kidney	2001	f	1-2	-	5	mean±SD	-	0.79±0.36	0.46±0.22	1.05±0.23	1.77±0.35	-	-	-	-	£0.10	-	21
"	kidney	2001	m	1-2	-	5	mean±SD	-	0.69±0.22	0.40±0.21	0.95±0.38	1.58±0.33	-	-	-	-	<0.05	-	21
"	muscle	2001	f	1-2	-	5	mean±SD	-	3.32±0.98	1.92±1.23	5.64±1.73	8.02±1.56	-	-	-	-	0.15±0.01	-	21
"	muscle	2001	m	1-2	-	5	mean±SD	-	2.26±0.76	1.73±0.28	3.73±0.70	7.27±1.62	-	-	-	-	0.16±0.04	-	21

Continued next page.

Annex Table 12 continued.

Species/Region/Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	HCB	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Ref.
<b>Steller sea lion</b>																		
Gulf of Alaska																		
SE Alaska	blubber	1998-2000	-	7<12mo, 3-23mo.	38.0±10.4	10	mean	-	-	-	616	631	-	-	-	-	-	27
Prince William Sound	blubber	1998-2000	-	14<12mo, 5-23mo.	32.0±80.3	19	mean	-	-	-	435	467	-	-	-	-	-	27
<b>Northern fur seal</b>																		
Alaska																		
Pribilof Islands	blubber	1997-2000	m,f	subadults	58.6±11.0	10	mean	-	-	-	1710	1190	-	-	-	-	-	27
"	blubber	1997-2000	-	pups	35.3±20.6	3	mean	-	-	-	4170	2130	-	-	-	-	-	27
"	blubber	1997-2000	f	adults	45.7±4.27	4	mean	-	-	-	3050	1350	-	-	-	-	-	27
"	blubber	1997-2000	-	fetuses	38.5±21.9	2	mean	-	-	-	1810	624	-	-	-	-	-	27
St. Paul Isl., Pribilof Islands	blubber	1990	m	2-3, subadults	54±14	7	mean±SD	0.6±0.7	-	792±361	2710±1470	1340±522	-	-	52.3±85.5	-	-	3
St. George Isl., Pribilof Islands	blood	1996	-	neonates of young dams	0.30±0.39	23	mean±SD	-	-	-	13.2±11.7	22.8±7.62	-	-	-	-	-	28
"	blood	1996	-	neonates of old dams	0.22±0.15	25	mean±SD	-	-	-	2.94±2.16	18.5±5.53	-	-	-	-	-	28
"	blood	1996	-	pups of young dams	0.28±0.48	21	mean±SD	-	-	-	6.64±3.42	16.2±4.54	-	-	-	-	-	28
"	blood	1996	-	pups of old dams	0.31±0.32	21	mean±SD	-	-	-	6.05±4.72	19.1±7.26	-	-	-	-	-	28
<b>Walrus</b>																		
Bering Sea																		
Russia & Alaska	blubber	1991	m	13.1±9.5	78.6±11.8	8	mean±SD	n.d.	120±50	140±90	n.d.	450±410	-	-	80±30	n.d.	-	29
"	blubber	1991	f	15.2±9.7	82.7±3.7	19	mean±SD	n.d.	90±50	60±60	n.d.	160±160	-	-	50±50	n.d.	-	29
Alaska	blubber	1993-1996	m	8-25	67.2± 12.7	9	mean±SD	<0.6-2.06	96.0±55.3	111±56.8	6.1±3.7	107±58.3	-	-	51.2±23.5	-	-	30
"	blubber	1993-1996	f	5-10	81.4± 2.15	5	mean±SD	<0.6	138±79.3	71.4±33.9	5.5±1.22	63.0±30.7	-	-	62.3±34.4	6.35± 3.02	-	30
"	blubber	1996	f	calf	64.3	1	-	<0.6	90.8	22.8	4.3	21.2	-	-	28.3	<1	-	30
Canada																		
E Hudson Bay	blubber	1999	-	-	84.3±3.96	6	mean±SD	0.33±0.06	104±47.8	291±167	53.9±34.5	300±157	129±81.7	-	-	-	-	31
NW Greenland																		
Avanersuaq	blubber	1978	m	15±3	79.6±14.8	8	mean±SD	0.97±0.68	74.9±52.8	89.1±62.2	60.7±43.2	246±138	-	321±95.0	46.5±35.4	7.91±6.23	-	32
"	blubber	1978	f	15±4	85.8±5.2	9	mean±SD	0.74±0.48	60.1±51.2	71.0±60.6	48.2±50.4	189±131	-	212±89.0	25.2±18.4	4.71±3.83	-	32
"	blubber	1988	m	10±0	86.9±4.7	5	mean±SD	0.92±0.35	109±29	116±45.0	79.3±48.2	301±99.0	-	319±39.0	100±59.0	8.86±1.92	-	32
"	blubber	1988	f	10±1	86.3±3.4	11	mean±SD	1.04±0.54	107±20	98.6±38.9	70.4±39.0	244±67.0	-	314±41.0	85.9±43.5	7.36±2.72	-	32
NE Greenland																		
Ittoqqortoormiit	blubber	1989	m	18±6	83.7±2.2	8	mean±SD	1.52±0.37	66.7±34.1	744±226	3410±1350	2860±1010	-	1610±305	761±215	19.4±7.0	-	32

ΣPCB<sub>10</sub> = sum of 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.

n.d. = not detected;

## References

- Hoekstra, 2003a; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209).
- Kucklick and Krahn, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of oxychlordane, heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of 2,4'- and 4,4'-DDD, DDE, and DDT; ΣPCBs = sum of 29 congeners.
- Krahn *et al.*, 1997; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-nonachlor, and oxychlordane; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 17 congeners (18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, 209).
- Muir *et al.*, 1999c; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 103 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209); Toxaphene = total toxaphene.
- Environmental Sciences Group (ESG), 2002.
- Muir *et al.*, 2001c; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, and *trans*- and *cis*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 86 peaks (103 congeners: 1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209); Toxaphene = total toxaphene.
- Muir *et al.*, 1999d; ΣDDTs = *o,p'* and *p,p'*-DDD, -DDT, and -DDE; ΣPCBs = 102 congeners; Toxaphene = determined by GC-ECD; ΣPBDEs = sum of mono-, di-, tri-, tetra-, penta-, hexa-, and hepta-brominated diphenyl ethers.
- Fisk *et al.*, 2002c; HCB = sum of 1,2,4,5-TeCBz, 1,2,3,4-TeCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane. ΣDDTs = sum of *p,p'*- and *o,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 77 congeners (1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 95/66, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 194, 196/203, 189, 208, 195, 207, 194, 205, 206, 209).
- Denmark, 2002; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, and heptachlor epoxide; ΣDDTs = sum of *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 59 congeners.
- Muir and Johansen, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Denmark, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Denmark, 2002; ΣHCHs and ΣDDTs see footnote 11, but ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, and *cis*- and *trans*-nonachlor; ΣPCBs = sum of 22 congeners; Toxaphene = sum of Parlars 26, 40, 41, 44, 50, and 62.
- Denmark, 2002; see footnote 11 except ΣPCBs = sum of 102 congeners.
- Wolkers *et al.*, 1998b; ΣPCBs = sum of 15 congeners (28, 52, 74, 99, 101, 105, 118, 128, 138, 149, 153, 156, 170, 180, 187); Toxaphene = 3 times sum of Parlars 26 and 50. Wet weight values estimated assuming 85% lipid content.
- Severinsen *et al.*, 2000; ΣDDTs = *p,p'*-DDE only; ΣPCBs = sum of 12 congeners (99, 101, 105, 118, 128, 138, 149, 153, 156, 170, 180, 187); used data from animals back, ranges include inner and outer layers of blubber. Wet weight values estimated assuming 90% lipid content.
- Kleivane *et al.*, 2000; ΣHCHs = γ-, α-, and β-HCH; ΣCHLs = oxychlordane and *trans*-nonachlor; ΣDDTs = *p,p'*-DDT and -DDE; ΣPCBs = sum of 16 congeners (66, 101, 99, 118, 153, 105, 138, 187, 128, 156, 157, 180, 170, 194, 206, 209). Wet weight values estimated assuming mean % lipid level.

17. Nyman *et al.*, 2002; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 33 congeners (18, 33, 51, 52, 49, 47, 74, 80, 66, 60, 101, 99, 110, 123, 118, 114, 122, 105, 153, 141, 138, 167, 128, 156, 157, 180, 170, 187, 183, 189, 194, 206, 209). Wet weight values estimated assuming 90% lipid content.
18. Bang *et al.*, 2001; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = oxychlorodane, *cis*- and *trans*-chlorodane, and *cis*- and *trans*-nonachlor; ΣDDTs = *o,p'*-DDD and -DDT, *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 33 congeners (31, 28, 52, 47, 74, 66, 56, 101, 99, 87, 136, 110, 151, 149, 118, 114, 153, 105, 141, 137, 138, 187, 128, 156, 157, 180, 170, 199, 196, 189, 194, 206, 209). Wet weight values estimated assuming mean % lipid level.
19. Kostamo *et al.*, 2000; ΣDDTs = sum of *p,p'*-DDT, -DDE, and -DDD; ΣPCBs = sum of 7 congeners (28, 52, 101, 118, 138, 153, 180).
20. Muir *et al.*, 2000c; see footnote 5
21. RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1, 2, 3, 4-, 1, 2, 3, 5-, and 1, 2, 4, 5-tetrachlorobenzene, and hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlorodane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of Parlars 26, 50, 62; ΣPBDEs = sum of 2,2',4,4'-tetrabromodiphenyl, 2,2',4,4',5-pentabromodiphenyl and 2,2',4,4',5'-pentabromodiphenyl ethers.
22. Wolkers *et al.*, 1999, Wet weight values estimated assuming 85% lipid content, to revert to actual lipid weight values divide all values by 0.85.
23. Wolkers *et al.*, 2000; ΣHCHs = sum α-, β-, and γ-HCH; ΣCHLs = sum *cis*- and *trans*-chlorodane, *cis*- and *trans*-nonachlor, oxychlorodane, heptachlor epoxide, U82, MC5, and MC7; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; Toxaphene = 3 times sum of Parlars 26 and 50. Wet weight values estimated assuming 85% lipid content.
24. Ruus *et al.*, 1999; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of oxychlorodane and *trans*-nonachlor; ΣDDTs = sum of *p,p'*-DDT, -DDD and -DDE, and *o,p'*-DDD; ΣPCBs = sum of 17 congeners (28, 99, 101, 105, 118, 128, 138, 141, 153, 156, 157, 170, 180, 187, 194, 206, 209). Wet weight values estimated assuming mean % lipid level.
25. Larsen and Dam, 1999; ΣPCBs = congener 153.
26. Dam, 2001; ΣPCB = congener 153.
27. Beckmen, 2002; ΣDDTs = *p,p'*-DDE only; ΣPCBs = sum of 11 congeners (101/99/149/196, 105, 110, 118, 128, 138, 153/87, 156, 157, 170/194, 180). Wet weight values estimated assuming mean % lipid level.

28. Beckmen *et al.*, 1999; ΣDDTs = sum of *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 11 congeners (101/99/149/196, 105, 110, 118, 128, 138, 153/87, 156, 157, 170/194, 180).
29. Seagars and Garlich-Miller, 2001; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of heptachlor epoxide, oxychlorodane, *cis*- and *trans*-chlorodane, and *cis*-nonachlor; ΣDDTs = sum of *o,p'* and *p,p'*-DDD, -DDE and DDT;
30. Kucklick and Struntz, 2002; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlorodane, oxychlorodane, *cis*- and *trans*-nonachlor, heptachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 35 congeners (8, 18, 29, 31/50, 28, 52, 49, 104, 44, 95, 66, 101, 99, 87, 154, 110, 151, 154, 149, 118, 188, 153, 105, 138, 187, 183, 128, 201, 156, 180, 170, 195, 194, 206, 209).
31. Muir and Kwan, 2000; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of oxychlorodane, *cis*- and *trans*-chlorodane, *cis*- and *trans*-nonachlor, heptachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCB = sum of 92 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
32. Muir *et al.*, 2000e; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of 'C', heptachlor, MC1, MC3, MC4, MC5, MC6, photoheptachlor, oxychlorodane, *cis*- and *trans*-chlorodane, *cis*- and *trans*-nonachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCB = sum of 85 congeners (8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138/163, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209).
33. Nakata *et al.*, 1998a; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of oxychlorodane, *cis*- and *trans*-chlorodane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of 4,4'-DDE, -DDD, and -DDT; ΣPCBs = sum of 50 congeners (31/28, 41/64, 47, 49/69, 52, 58/74, 60, 66, 70, 83, 84/90/92, 85, 87/117, 91/95, 97/113, 99, 101, 102, 105, 118, 132, 137, 138, 141, 144/149, 151, 153, 156, 170, 172, 173, 177, 178, 180, 187, 194, 196/201, 198, 202 and 8 unidentified congeners).

Annex Table 13. Mean concentrations (ng/g ww) of organochlorines in cetaceans inhabiting Arctic waters.

Species/Region/Location	Tissue	Year	Sex	Age, years	Length, cm	% lipid	n	Statistic	HCb <sup>a</sup>	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Reference
<b>MYSTICETES</b>																			
<b>Minke whales</b>																			
<b>W Greenland</b>																			
Davis Strait	blubber	1998	f	juveniles, adults	683±124	72	34	mean±SD	81.8±73.9	67.9±58.7	311±261	650±561	2290±2900	–	595±732	275±203	6.49±10.0	–	1,2
Davis Strait	blubber	1998	m	juveniles, adults	663±117	73.3±14.8	8	mean±SD	83.3±61.8	73.3±25.6	356±213	732±383	2560±2130	–	671±627	262±75.5	5.69±4.69	–	1,2
<b>SW Greenland</b>																			
Ammassalik	blubber	1998	f	juveniles, adults	795±125	73.4±12.4	4	mean±SD	41.8±32.5	70.0±45.9	408±396	242±173	855±320	–	152±101	129±88.9	2.06±1.47	–	1,2
<b>Greenland Sea</b>																			
Jan Mayen	blubber	1998	f	juveniles, adults	775±53.2	73.7±8.6	19	mean±SD	72.3±36.3	36.6±13.7	279±99.1	809±653	1660±749	–	1630±1220	210±80.2	6.51±3.37	–	1,2
Jan Mayen	blubber	1998	m	juveniles, adults	645±48.0	75.1±5.6	4	mean±SD	127±24.5	41.3±4.36	491±230	1320±425	3430±2000	–	2740±4250	326±149	11.9±4.56	–	1,2
<b>North Sea</b>																			
North Sea	blubber	1998	f	juveniles, adults	786±94.9	66.3±12.5	14	mean±SD	44.1±47.5	45.7±18.1	254±134	621±380	1220±816	–	1240±928	326±160	4.55±3.53	–	1,2
North Sea	blubber	1998	m	juveniles, adults	755±65.2	66.9±12.9	9	mean±SD	241±412	76.4±40.2	572±299	2130±1630	4100±2930	–	3470±1890	593±492	36.5±81.3	–	1,2
North Sea	blubber	2001	f	adults	783(639-881)	72(43-88)	5	mean (range)	–	–	–	–	2190(876-3650)	–	–	–	–	–	3
<b>Norwegian Sea</b>																			
Vestfjorden/Lofoten	blubber	1998	f	juveniles, adults	719±128	82.4±8.0	7	mean±SD	72.0±42.9	40.5±22.3	318±194	549±289	1120±816	–	185±106	250±149	6.35±9.54	–	1,2
Vestfjorden/Lofoten	blubber	1998	m	juveniles, adults	624±151	71.3±10.4	7	mean±SD	93.6±32.9	56.2±14.9	484±284	1750±1550	2570±1860	–	262±131	509±324	7.88±8.13	–	1,2
<b>Barents Sea</b>																			
West Svalbard	blubber	1998	f	juveniles, adults	768±41.1	57.3±12.0	15	mean±SD	67.3±43.0	38.1±15.1	209±83.6	510±238	1420±610	–	1090±644	226±87.6	5.75±3.44	–	1,2
West Svalbard	blubber	1998	m	adult	700	79.2	1	mean	27.4	39.2	126	541	1410	–	785	225	6.28	–	1,2
Spitsbergen	blubber	2001	f	adult	775(698-832)	63(35-75)	5	mean (range)	–	–	–	–	461(162-595)	–	–	–	–	–	3
NW Kola Peninsula	blubber	1998	f	juveniles, adults	757±19.3	65±8.1	30	mean±SD	89.8±44.1	37.4±20.2	343±271	693±282	2360±1330	–	589±547	238±113	6.36±3.86	–	1,2
NW Kola Peninsula	blubber	1998	m	adults	764±42.9	67.5±3.4	3	mean±SD	176±45.5	84.1±35.9	475±127	1830±1240	4820±2280	–	1090±902	398±143	8.08±3.91	–	1,2
N Norway, NW Russia	blubber	1992	m	adults	797(720-878)	89(82-97)	22	mean (range)	231.4(107-338)	133(53.4-347)	1450(427-4510)	3440(1130-13140)	5140(2030-18500)	–	–	–	–	–	4
N Norway, NW Russia	blubber	1992	f	adults	773(670-882)	87(71-96)	22	mean (range)	165(69.6-374)	69.6(34.8-157)	652(244-1640)	1310(609-2930)	1970	–	–	–	–	–	4
N Norway, NW Russia	blubber	1992	m	juveniles	634(500-712)	90(85-90)	15	mean (range)	207(126-423)	81(36-162)	819(270-1760)	1750(468-4068)	2680(540-5240)	–	–	–	–	–	4
N Norway, NW Russia	blubber	1992	f	juveniles	625(485-742)	86(72-96)	13	mean (range)	310(146-929)	129(51.6-499)	1100(396-2740)	2380(808-5640)	3240(1190-8920)	–	–	–	–	–	4
<b>Gray whales</b>																			
<b>Russia</b>																			
Chirikov Basin, Bering Sea	blubber	1994	f	juveniles	884±28	48±6.1	13	mean±SE	240±41	–	–	180±40	680±98	–	–	–	–	–	5,6
Chirikov Basin, Bering Sea	blubber	1994	m	juveniles	833±26	48±11	4	mean±SE	200±16	–	–	87±10	480±130	–	–	–	–	–	5,6
Lavrentiya, Chukotka Peninsula	blubber	2001	m	2-3	–	–	1	–	–	135	101	93.0	195	–	–	–	1.58	–	7
Lavrentiya, Chukotka Peninsula	liver	2001	m	2-3	–	–	1	–	–	5.86	1.27	2.25	2.98	–	–	–	<0.05	–	7
Lavrentiya, Chukotka Peninsula	kidney	2001	m	2-3	–	–	1	–	–	1.44	0.42	0.73	1.87	–	–	–	<0.05	–	7
Lavrentiya, Chukotka Peninsula	muscle	2001	m	2-3	–	–	1	–	–	1.23	0.53	0.92	1.41	–	–	–	<0.05	–	7

Continued next page.



Annex Table 13 continued.

Species/Region/Location	Tissue	Year	Sex	Age, years	Length, cm	% lipid	n	Statistic	HCb <sup>a</sup>	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Reference
<b>Bowhead whales</b>																			
Bering-Chukchi-Beaufort Sea																			
Barrow & Kaktovik, Alaska	blubber	1997-2000	m,f	–	–	75.8±1.6	71	mean±SE	106±7.5*	230±13	183±11	331±55	359±28	–	455±45	–	–	–	8
''	liver	1997-2000	m,f	–	–	6.6±0.4	23	mean±SE	4.2±0.4*	9.5±0.5	5.5±0.5	3.7±0.3	9.1±0.9	–	8.8±5	–	–	–	8
<b>ODONTOCETES</b>																			
<b>Beluga whales</b>																			
Gulf of Alaska																			
Cook Inlet, Alaska	blubber	1992-97	f	9.9±5.6 (n=4)	–	89±4.9	10	mean±SD	150±130	–	300±220	590±450	790±560	–	–	57±50	9.1±3.7	–	9
''	blubber	1992-97	m	9.3±0.9 (n=4)	–	90±1.4	10	mean±SD	220±93	–	560±250	1350±730	1490±700	–	–	92±47	13±5.6	–	9
Bering-Chukchi Sea																			
Point Lay, Alaska	blubber	1998-1999	m	–	–	84.8±1.82	20	mean±SE	238±20	212±36	1091±100	1979±213	2734±229	–	975±173	215±19	59±6.2	–	10
''	blubber	1992	f	fetus	–	–	1	–	–	–	621	522	1215	–	3340	–	–	–	11
''	blubber	1992	f	16 (mother)	–	–	1	–	–	–	558	468	1130	–	2880	–	–	–	11
Point Lay, Alaska	blubber	1992	f	12(11-19)	–	–	3	mean(range)	–	–	560(288-891)	631(288-1050)	1260(630-1940)	–	2810(2140-3190)	–	–	–	11
''	blubber	1992	m	9.5(6-13)	–	–	2	mean(range)	–	–	2860(2220-3490)	4500(2860-6150)	6600(4720-8480)	–	9000(3650-14300)	–	–	–	11
''	blubber	1990-1996	f	16.4±7.5	–	89±4.1	8	mean±SD	230±280	–	790±610	930±850	1500±1120	–	–	120±96	22±6.6	–	9
''	blubber	1990-1996	m	12.3±4.5	–	86±6.3	11	mean±SD	810±120	–	2420±460	3630±900	5200±900	–	–	390±86	63±21	–	9
Beaufort Sea																			
Point Hope, Alaska	blubber	1989	f	6.3(4.5-8)	–	92(89-94)	2	mean(range)	570(230-900)	–	1300(1080-1530)	1240(1140-1330)	2220(1880-2560)	–	–	210(140-280)	17(14-20)	–	9
W Canada																			
Hendrikson Island	brain	1992	m	–	441(427-457)	8.88(2.16-13.1)	3	mean(range)	45.3(32.0-68.3)	24.9(19.4-30.4)	13.1(9.90-13.5)	46(21.9-86.5)	130(107-173)	–	–	11.8	1.56	–	12
''	liver	1992	m	–	441(427-457)	2.82(2.01-3.71)	3	mean(range)	37.9(17.0-70.4)	2.7(2.1-3.3)	19.8(17.2-23)	73.6(39.0-100)	132(85.8-172)	–	–	13.6	2.05	–	12
''	muscle	1992	m	–	441(427-457)	0.98(0.61-1.31)	3	mean(range)	10.7(9.0-12.8)	1.0(0.5-1.4)	9(8.1-10.4)	28.2(16.5-43.9)	56.1(32.9-81.5)	–	–	4.71	0.70	–	12
''	blubber	2001	m	–	–	92.2±4.21	10	mean±SD	418±265*	172±97.4	929±61	1120±846	2790±1060	–	2330±925	–	–	–	13
E Canada																			
Naskapoka R., E Hudson Bay	blubber	1999	f	–	–	–	2	mean(range)	203**(88.4-318)	120(97.1-142)	904(784-1020)	1300(1240-1360)	2220(2140-2300)	755(734-777)	–	–	–	–	14
''	blubber	1999	m	–	–	–	8	mean±SD	253±97**	119±50.9	1460±768	2630±1930	3550±1950	1270±720	–	–	–	–	14
''	blubber	1999	m	–	–	–	8	(range)	(97.2-414)	(62.2-205)	(578-2860)	(732-6480)	(1260-7010)	(453-2540)	–	–	–	–	14
W Hudson Bay	blubber	1993	f	–	–	35.3±16.9	4	mean±SD	256±127***	171±59.2	1240±232	1070±172	1490±225	–	–	318±124	15.4±5.04	–	15
''	blubber	1993	m	–	–	40.7±19.0	6	mean±SD	340±76.7***	216±57.2	1590±293	2100±857	2200±363	–	–	477±134	20.2±4.47	–	15
Cumberland Sound	blubber	1982	m	9.7±5.4	–	90.8±2.4	8	mean±SD	455±66*	267±37	1650±372	5340±2550	4230±1360	–	9650±2390	616±108	–	–	16
''	blubber	1986	m	5.6±1.7	–	91.2±2.3	17	mean±SD	379±93*	261±69	1420±313	3280±1050	2570±609	–	7760±1790	445±102	–	–	16
''	blubber	1992	m	12.9±4.9	–	90.9±2.4	11	mean±SD	343±182*	189±48	2030±519	5650±1870	3990±1140	–	9430±2540	387±119	–	–	16
''	blubber	1996-1997	m	13.5±5.4	–	89.8±4.1	17	mean±SD	694±226*	217±57	2110±552	5570±1740	4570±942	–	10600±3850	568±215	–	–	17
Kimmirut, NT	blubber	1994	f	21± 18	–	92.9±1.7	3	mean±SD	–	–	–	3540±2620	4390±3120	–	7350±4460	–	–	1.44±1.01	18
''	blubber	1994	m	6±6	–	93.5±1.9	3	mean±SD	–	–	–	7110±3360	7650±2870	–	16300±8890	–	–	2.84±0.90	18
Central W Greenland																			
Saqqaq	blubber	2000	m,f	–	–	87.9±2.4	10	mean±SD	260±158*	136±72.2	1200±624	1560±765	2450±1130	1020±483	–	–	–	–	19
''	kidney	2000	m,f	–	–	4.4±2.2	5	mean±SD	10.9±9.28*	2.48±1.99	21.2±19.0	35.2±36.5	76.7±72.9	26.1±24.3	–	–	–	–	19
''	liver	2000	m,f	–	–	6.8±3.0	5	mean±SD	13.3±7.96*	2.60±1.22	17.4±8.59	24.1±12.1	55.3±25.2	20.4±8.93	–	–	–	–	19
''	muscle	2000	m,f	–	–	2.3±1.0	20	mean±SD	8.20±5.55*	2.64±1.60	23.6±20.1	29.2±25.2	59.5±46.0	21.7±17.9	–	–	–	–	19
''	skin	2000	m,f	–	–	3.6±0.8	5	mean±SD	6.76±4.97*	3.10±1.45	44.7±24.4	57.1±26.5	78.5±42.6	35.4±18.8	–	–	–	–	19
Barents Sea																			
E Spitsbergen (Svalbard)	blubber	1998	m,f	sub-adults	–	–	5	geo. mean	2.97	–	848	3440	2970	–	1420	–	–	–	20
Svalbard	blubber	1995-97	m	adults	–	28±30	10	mean	142	43.4	804	1430	1430	–	3190	319	–	–	21,22
''	blubber	1998	m,f	juveniles, adults	205-440	4-90	9	range	–	–	–	–	–	–	–	–	–	17-170	23
<b>Narwhals</b>																			
Canada																			
Broughton Isl., E. Baffin Island	blubber	1993-1996	f	–	398±62	84.4±4.0	18	mean±SD	440±177*	112±41	1470±482	3610±1340	3890±1330	–	5150±1410	–	–	–	13
''	blubber	1993-1997	m	–	443±45	86.6±4.5	32	mean±SD	512±191*	126±48	1750±610	4830±2270	4820±1830	–	7440±4030	–	–	–	13
Grise Fiord, Jones Sound	blubber	1993-1995	f	–	394	82.5±5.4	2	mean±SD	332±115*	81±3.4	1420±96	3800±417	3500 ±440	–	8090±210	–	–	–	13
''	blubber	1995-1999	m	–	407±93	87.6±4.6	13	mean±SD	369±130*	102±32	1500±450	4020±1750	3440±955	–	7950±1960	–	–	–	13
Pangnirtung, SE Baffin Island	blubber	1996	m	–	358±54	91.2±3.9	5	mean±SD	458 ±70*	117±31	1850±348	4950±1990	4600±1090	–	8760±3670	–	–	–	13
Pond Inlet, NE Baffin Island	blubber	1994-1999	f	–	393±33	92.1±4.8	10	mean±SD	344±84*	114±32	1610±525	4470±945	5200±1470	–	–	–	–	–	13
''	blubber	1992-1999	m	–	387±52	92.6±3.7	14	mean±SD	672±362*	165±74	2600±1060	7220±2380	5820±2020	–	11800±4440	–	–	–	13
NW Greenland																			
Avanersuaq	blubber	1984	f	5.4±4.0	350	97.7±10.9	4	mean±SD	350±316	119±53	559±383	1900±1230	–	766±476	4450±2950	–	–	–	24
''	blubber	1985	m	5.1±3.3	351±103	89.1±9.2	15	mean±SD	618±293	203±72	919±312	3620±1350	–	1620±578	4970±1720	–	–	–	24
''	blubber	1985	f	5.2±2.8	350±85	90.6±5.1	7	mean±SD	404±489	139±95	611±564	2170±2170	–	972±893	4760±4580	–	–	–	24
''	blubber	1993	m	13.0	454	96.6	1	–	1300	227	2000	3650	–	2114	6700	–	–	–	24
''	blubber	1993	f	8.7±1.5	390±22	96.0±2.8	3	mean±SD	80±20	68±16	233±101	436±147	–	246±71	2030±321	–	–	–	24
Balgoni Island	blubber	1993	m	3.4±3.9	298±97	95.7±2.5	4	mean±SD	1450±1270	156±36	3250±2780	9720±11700	–	4541±5190	8820±3920	–	–	–	24
''	blubber	1993	f	4.6±4.9	345±121	96.6±0.6	2	mean±SD	865±1040	150±133	2360±2320	6920±6620	–	2980±2980	7250±5300	–	–	–	24

Continued next page.

Annex Table 13 continued

Species/Region/Location	Tissue	Year	Sex	Age, years	Length, cm	% lipid	n	Statistic	HCb <sup>a</sup>	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	ΣPCB <sub>10</sub>	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Reference	
Central W Greenland																				
Kitsissuarsuit	blubber	1990	m	16.0	441±23	96.8±0.5	2	mean±SD	830±170	204±11	1950±354	6600±2500	–	3210±590	7500±1270	–	–	–	24	
”	blubber	1990	m	2.9±0.7	299±19	96.2±4.6	2	mean±SD	835±106	178±88	1010±127	1710±587	–	1190±383	6800±1560	–	–	–	24	
Uummannaq	blubber	1993	m	9.7±7.6	385±94	93.1±5.7	33	mean±SD	662±293	179±64	1220±292	3460±1350	–	1540±468	5190±1150	–	–	–	24	
”	blubber	1993	f	6.1±4.5	349±72	93.8±4.0	17	mean±SD	522±537	158±107	1010±942	2260±2440	–	1140±1200	4020±2330	–	–	–	24	
Saqqaq	blubber	2000	–	–	–	78.6±16.0	3	mean±SD	462±253*	128±86	1190±736	1910±1330	3280±1870	991±557	–	–	–	–	19	
Barents Sea																				
E Spitsbergen (Svalbard)	blubber	1998	m,f	sub-adults	–	–	3	geo. mean	522	110	9180	11400	8910	–	4780	–	–	–	20	
Killer whales																				
Alaska, USA (residents)																				
Kenai Fiords/Prince William S.	blubber	1994-1999	m,f	immature & mature	–	27±9.9	66 (30m, 31f, 5?)	mean±SD	220±230	–	–	3800±4700	3900±4400	–	–	–	–	–	25	
Alaska, USA (transients)																				
Kenai Fiords/Prince William S.	blubber	1994-1999	m,f	immature & mature	–	23±11	14 (6m, 6f, 2?)	mean±SD	1300±1200	–	–	78000±63000	55000±43000	–	–	–	–	–	25	
Long-finned pilot whales																				
Faroe Islands																				
Hvannasund	blubber	1994	f	adult	395-512	82	1 pool (n=9)	–	–	–	–	–	–	–	–	–	–	691	23	
Vestmanna	blubber	1996	m,f	adults, juveniles	244-533	66-79	4 pools (n=19,8,13,4)	range	–	–	–	–	–	–	–	–	–	–	828-2400	23,26
Tórshavn	blubber	1997	m,f	adults, juveniles	–	52-85	12	range	–	–	–	–	–	–	–	–	–	–	133-1470	23,26
Leynar	blubber	1997	m,f	adults, juveniles	–	83-86	4 pools (n=3,28,9,10)	range	–	–	–	–	–	–	–	–	–	–	231-686	23,26
Sandavágur	blubber	1997	m,f	adults, juveniles	–	64-80	4 pools (n=8,18,19,7)	range	–	–	–	–	–	–	–	–	–	–	313-987	23,26
Hvalvik	blubber	1998	m,f	adults, juveniles	–	66-83	4 pools (n=6,20,9,16)	range	–	–	–	–	–	–	–	–	–	–	339-655	23,26
”	blubber	2000	f,?	3 adults, 3 fetuses	–	2-83	6	range	–	–	–	–	–	–	–	–	–	–	7.7-277	23
Harbour porpoises																				
SW Greenland																				
Davis Strait	blubber	1995	m,f	1-2	1250(1220-1320)	–	4	median(range)	117(63-162)	36(18-126)	–	702(486-810)	1180(792-1380)	–	–	–	–	–	27	
Norwegian Sea																				
W coast of Norway	blubber	1988-1990	m	5.7±0.89(5-7)	–	–	8	mean(range)	–	–	–	8190(2790-19800)	13500(6480-29700)	–	–	–	–	–	28	

<sup>a</sup>Hexachlorobenzene unless otherwise indicated.

\* Sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz.

\*\* Sum of 1,2,3,4-tetra, penta-, and hexachlorobenzene.

\*\*\* Sum of 1,2,4,5-tetraCBz, 1,2,3,4-tetraCBz, penta-CBz, and HCBz;

#### References

- Hobbs *et al.*, 2003; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 102 congeners (4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 50/31, 28, 20/53/33, 51, 22, 45, 46, 52, 43, 49, 48, 47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70, 76/98, 66/95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147/107, 149, 118, 133, 114/134, 131, 146, 153, 132, 105, 141, 179, 137, 176/130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171/156, 157/200/173, 204, 172, 197, 180, 193, 191, 199, 170, 190, 198, 201, 203/196, 189, 208, 195, 207, 194, 205, 206, 209).
- Hobbs *et al.*, 2002a; Toxaphene = total toxaphene.
- Skaare *et al.*, 2001c; ΣPCBs = sum of 35 congeners (28, 31, 52, 47, 74, 66, 56, 101, 99, 87, 136, 110, 151, 149, 118, 114, 153, 105, 141, 132, 137, 138, 187, 183, 128, 156, 157, 180, 170, 199, 196, 189, 194, 206, 209).
- Kleivane and Skaare, 1998; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = oxychlordane, *trans*- and *cis*-chlordane, and *trans*- and *cis*-nonachlor; ΣDDTs = *p,p'*-DDT, -DDE and -DDD, and *o,p'*-DDT and -DDD; ΣPCBs = sum of 18 congeners (66, 101, 99, 110, 149, 118, 153, 105, 138, 187, 128, 156, 157, 180, 170, 194, 206, 209). Concentrations converted from lipid weight to wet weight using mean % lipid content values.
- Krahn *et al.*, 2001; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 17 congeners (18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, 209) multiplied times two to estimate total PCB concentrations.
- Tilbury *et al.*, 2002; see reference 5 for details of contaminants analyzed.
- RAIPON/AMAP/GEF Project, 2001; ΣCBz = 1,2,3,4-, 1,2,3,5-, and 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 15 congeners (28, 31, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, 187); Toxaphene = sum of congeners 26, 50, 62; ΣPBDEs = sum of 2,2',4,4'-tetrabromodiphenyl, 2,2',4,4',5-pentabromodiphenyl and 2,2',4,4',5'-pentabromodiphenyl ethers.
- Hoekstra *et al.*, 2002c; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, and *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, 209); Toxaphene = sum of 24 chloroborane congeners.
- Krahn *et al.*, 2000; ΣCHLs = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, and nonachlor III; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 17 congeners (18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, 209) multiplied by 2 to estimate total PCBs.
- Hoekstra, 2002d; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208 & 209).
- Wade *et al.*, 1997; Concentrations converted from lipid weight to wet weight assuming 90% lipid content.
- Metcalfe *et al.*, 1999; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *trans*- and *cis*-chlordane, and *trans*- and *cis*-nonachlor; ΣDDTs = sum of *p,p'*-DDT, -DDD, and -DDE; ΣPCBs = sum of 27 congeners (18, 28, 31, 52, 49, 47, 44, 66, 87, 99, 101, 110, 105, 118, 119, 151, 149, 153, 156, 138, 180, 170, 199, 195, 196, 194, 209).
- Stern, 2001; ΣPCBs = sum of 86 peaks (103 congeners); ΣDDTs = sum *o,p'*- and *o,p'*-DDE, -DDD, and -DDT; Toxaphene = determined by GC-ECD.
- Sang *et al.*, 2000; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, and *trans*- and *cis*-nonachlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 103 congeners (1, 3, 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33, 20, 53, 51, 22, 45, 46, 52, 49, 43, 48/47, 44, 59, 42, 64/41/71, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 87/81, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 138/163, 158, 129, 178, 175, 187/182, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/201, 172, 197, 180, 193, 191, 200, 170/190, 198, 199, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
- Hobbs *et al.*, 2002b; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, nonachlor, C, C3, C5, heptachlor, C1B/U6, C2/U5, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 85 congeners (1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47/48, 44, 42, 41/71/64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134/131, 114, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/157, 172/197, 180, 193, 191, 200, 170/190, 198, 199, 196/203, 189, 208/195, 207, 194, 205, 206, 209).

16. Stern, 1999; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor, heptachlor epoxide, and nonachlor III; ΣDDTs = sum of *o,p'* and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 90 PCB congeners; Toxaphene = quantified using a single response factor based on 27 peaks in a technical mixture (primarily Parlars 26, 50).
17. Stern and Addison, 1999; see reference 16 for details of contaminants analyzed.
18. Muir *et al.*, 1999d; ΣPCBs = sum of 86 peaks (103 congeners), ΣDDTs = sum *o,p'*- and *o,p'*-DDE, -DDD, and -DDT; Toxaphene = determined by GC-ECD, ΣPBDEs = sum of mono-, di-, tri-, tetra-, penta-, hexa-, and hepta-brominated diphenyl ethers.
19. Muir and Johansen, 2001; ΣHCHs = sum of α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide, and methoxychlor; ΣDDTs = sum of *o,p'*- and *p,p'*-DDE, -DDD, and -DDT; ΣPCBs = sum of 104 congeners peaks (1, 3, 4/10, 7/9, 6, 8/5, 19, 30, 12/13, 18, 15/17, 24/27, 16/32, 54/29, 26, 25, 31/28, 50, 33/20, 53, 51, 22, 45, 46, 52, 43, 49, 47/48, 44, 59, 42, 71/41/64, 40, 100, 63, 74, 70/76/98, 66, 95, 91, 55, 56/60, 92, 84, 101, 99, 119, 83, 97, 81/87, 85, 136, 110, 82, 151, 135/144, 147, 107, 149, 118, 133, 114, 134/131, 146, 153, 132, 105, 141, 179, 137, 176, 130, 163/138, 158, 129, 178, 175, 182/187, 183, 128, 167, 185, 174, 177, 202/171, 156, 173, 157/200, 204, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 203/196, 189, 208/195, 207, 194, 205, 206, 209).
20. Wolkers, 2002; ΣHCHs = α-, β-, and γ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor epoxide; ΣDDTs = sum of *o,p'*-DDD, *o,p'*- and *p,p'*-DDE, and -DDT; ΣPCBs = sum of 23 congeners (28, 52, 70, 76, 95, 99, 101, 105, 110, 118, 128, 138, 141, 146, 149, 153, 156, 169, 170, 174, 180, 183, 187); Toxaphene = sum of Parlars 26, 32, 40, 44, 50; Concentrations converted from lipid weight to wet weight assuming 90% lipid content.

21. Andersen *et al.*, 2001a; ΣCHLs = sum of heptachlor epoxide, oxychlordane, *cis*- and *trans*-nonachlor, and *cis*-nonachlor; ΣDDTs = *p,p'*-DDT, -DDE, and -DDD; ΣPCBs = sum of 27 congeners (47, 52, 66, 74, 87, 99, 101, 105, 110, 118, 128, 136, 137, 138, 141, 149, 151, 153, 156, 157, 170, 180, 183, 187, 194, 196, 206); Concentrations converted from lipid weight to wet weight basis using mean % lipid value.
22. Andersen *et al.*, 2002; Toxaphene = sum of congeners 26, 50, 62; Concentrations converted from lipid weight to wet weight basis using mean % lipid value.
23. van Bavel *et al.*, 2001; ΣPBDEs = sum of 21 tetra-, penta- and hexa-BDEs (several unnamed, but includes 47, 99, 85, 153, 138).
24. Denmark, 2002; ΣHCHs = sum of α-, β-, γ-HCH; ΣCHLs = only *trans*-nonachlor; ΣDDTs = sum of *p,p'*-DDE, -DDD, -DDT; Toxaphene = total toxaphene.
25. Ylitalo *et al.*, 2001; ΣDDTs = sum of *o,p'*-DDD and -DDT, *p,p'*-DDD, -DDE and -DDT; ΣPCBs = sum of 17 congeners (77, 105, 110, 118, 126, 156, 157, 169, 189, 101/99/149/183/196, 128, 138, 156, 157, 153/87, 170/194, 180), determined by a different method (HPLC, not ECD).
26. Lindström *et al.*, 1999; ΣPBDEs = sum of 21 tetra-, penta- and hexa-BDEs (several unnamed, but includes 47, 99, 85, 153, 138).
27. Bruhn *et al.*, 1999; ΣHCHs = α-HCH; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 49 congeners (44, 52, 66, 74, 91, 92, 95, 97, 99, 101, 105, 107, 110, 118, 128, 130, 132, 136, 138, 141, 146, 149, 151, 153, 156, 158, 170, 171, 172, 174, 175, 137/176, 177, 178, 179, 180, 183, 185, 187, 189, 191, 193, 194, 197, 199, 201, 202, 196/203, 195/208); Concentrations converted from lipid weight to wet weight assuming 90% lipid content.
28. Berggren *et al.*, 1999; ΣDDTs = sum of *o,p'* and *p,p'*-DDD, and -DDT; ΣPCBs = sum of 6 congeners (52, 101, 118, 138, 153, 180); Concentrations converted from lipid weight to wet weight assuming 90% lipid content.

Annex Table 14. Mean concentrations of organochlorines in Arctic fox, polar bears and sea otters.

Species/ Region/Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	ΣCBz	HCb	α-HCH	β-HCH	γ-HCH (lindane)	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Ref- erence
<b>Arctic fox (<i>Alopex lagopus</i>) (ng/g ww)</b>																					
Alaska																					
Pribilof Islands	fat	1996-97	f	~2	60	2	mean(range)	-	24.5(10-39)	-	141(43-240)	-	-	853(148-1560)	107(4-210)	1320(140-2500)	-	38.4(4.9-72)	7.5(2-13)	-	1
"	fat	1996-97	m	1.7	66	5	mean±SD	-	23.7±26.4	-	810±632	-	-	2850±2150	254±233	3330±3090	-	157±138	36.9±37.6	-	1
Barrow	muscle	1999-2000	m,f	<1	7.1±1.2	18	mean±SE	9.9±2.6	6.1±1.3	1.7±0.23	11±4.8	-	14±5.0	42±17	5.8±0.7	81±22	10±2.6	-	-	-	2
"	liver	1999-2000	m,f	<1	7.7±1.6	18	mean±SE	12±1.7	9.9±2.6	2.0±0.23	18±5.5	-	22±5.1	236±82	8.7±0.9	124±32	28±8.1	-	-	-	2
Canada																					
Holman, NWT	muscle	1999-2000	m,f	0-2	5.6±0.6	20	mean±SD	4.43±0.63	1.4±0.25	1.5±0.39	3.9±0.64	-	5.40±0.91	34.0±8.86	1.91±0.47	48.0±9.15	4.5±1.2	-	-	-	2
"	liver	1999-2000	m,f	0-2	9.2±1.3	20	mean±SD	4.16±0.95	2.8±0.85	3.1±0.94	7.7±1.6	-	10.9±2.12	219±56	3.99±0.82	124±39.4	17±4.5	-	-	-	2
Iceland																					
Árnessysla (inland)	liver	1993-1994	m,f	7 mo-3 yr	6	6	mean±SD	-	3.6±2.79	-	6.83±3.92	6.5±3.51	13.3±7.23	30.8±19.6	2.00±1.26	60.8±38.3	-	-	-	-	3
Borgarfjörður E (coast)	liver	1993-1994	m,f	5 mo-4 yr	6	4	mean±SD	-	59.7±39.7	-	11.5±4.04	3.75±1.5	15.2±4.35	3680±3550	46.0±57.4	4350±4100	-	-	-	-	3
<b>Polar bear (<i>Ursus maritimus</i>) (ng/g ww, except for reference 5 which are ng/g lw)</b>																					
Beaufort Sea																					
Barrow, Alaska	fat	1996-1997	f	adults	74.7±12.5	3	mean±SD	-	57±12.0	-	-	<1	-	2020±493	137±97	4870±2080	-	89±20	-	-	1
"	fat	1996-1997	f	-	80.2±4.6	3	mean±SD	-	201±111	-	-	1.5±0.8	-	2010±247	167±103	5130±874	-	147±21	-	-	1
Barrow & St. Lawrence Island	fat	1996-1998	m	adults	83.3±9.3	7	mean±SD	-	106	59±25	768±479	4.1±2.4	829±460	573±383	80±71	3330±1570	-	89±20	-	-	1
Barrow, Alaska	fat	1996-1997	m	cubs, unknown	78.2±7.4	2,4	mean±SD	-	137±121	-	-	1.6±0.5	-	2080±1280	71±40	7520±2400	-	151±74	-	-	1
Southern Beaufort Sea																					
N Alaska	fat	1996-2002	m	adults	79.5 (57.3-100)	11	geo.mean(range)	180	(96.9-462)	-	-	-	766	932(355-2210)	46.9(210-1940)	3450(18.3-117)	(1760-6950)	90.6	(36.6-302)	-	4
Chukchi/Bering Sea																					
W Alaska	fat	1996-2002	m	adults	79.4 (36.8-100)	16	geo.mean(range)	131	(63.5-649)	-	-	-	759	588(1530-2830)	47.6(224-1260)	2090(11.1-240)	(720-8640)	75.4	(34.9-305)	-	4
Canadian Arctic																					
Queen Maud Gulf,	fat	1984	-	-	-	8-10	geo.mean(range)	244(136-423)	-	-	-	-	358*(167-475)	1459(861-2547)	98*(42-225)	3004(963-6992)	-	172*(98-274)	-	-	5
Larsen Sound (I)	fat	1990	-	-	-	8-10	geo.mean(range)	175(66-374)	-	-	-	-	228*(89-478)	1284(333-4194)	29.7*(13-151)	2616(718-6514)	-	88*(25-167)	-	-	5
Barrow Strait,	fat	1984	-	-	-	8-10	geo.mean(range)	197(111-394)	-	-	-	-	206*(137-281)	1201(492-2449)	109(55-246)	3377(1637-8261)	-	148*(76-229)	-	-	5
Lancaster Sound (II)	fat	1990	-	-	-	8-10	geo.mean(range)	129(75-39)	-	-	-	-	294*(147-485)	928(378-1372)	96(31-233)	3018(1884-4622)	-	89*(40-139)	-	-	5
N Baffin Bay (III)	fat	1984	-	-	-	8-10	geo.mean(range)	188(23-345)	-	-	-	-	184(80-361)	1048(301-2483)	131(68-204)	3725(910-8233)	-	148(40-251)	-	-	5
"	fat	1990	-	-	-	8-10	geo.mean(range)	225(130-384)	-	-	-	-	1746(691-4501)	140(89-218)	4887(1096-8493)	-	169	-	-	-	5
W Hudson Bay (V)	fat	1968	-	-	-	8-10	geo.mean(range)	196*(135-378)	-	-	-	-	235(130-616)	1086(453-4527)	884*(275-1851)	3824*(1926-9467)	-	191(114-482)	-	-	5
"	fat	1984	-	-	-	8-10	geo.mean(range)	320*(107-839)	-	-	-	-	335(161-648)	2525(1244-5255)	525*(214-1142)	7132*(4252-14944)	-	215(47-750)	-	-	5
"	fat	1989	-	-	-	8-10	geo.mean(range)	169*(39-432)	-	-	-	-	252(125-461)	1383(457-5839)	148*(21-587)	4602*(1849-11912)	-	234(82-566)	-	-	5
Davis Strait (IV)	fat	1984	-	-	-	8-10	geo.mean(range)	221(148-322)	-	-	-	-	172(111-263)	1293*(880-1637)	288(105-1147)	4268(2201-13683.4)	-	149(96-250)	-	-	5
"	fat	1990	-	-	-	8-10	geo.mean(range)	186(138-301)	-	-	-	-	181(113-494)	1838*(838-4999)	152(78-390)	7541(2140-20215)	-	209(114-536)	-	-	5
Resolute	whole plasma	1997	-	subadults	-	6	mean±SD	7.5±5.2	-	-	-	-	5.0±2.7	56±46	1.5±1.1	79±54	-	-	-	-	6
"	whole plasma	1997	m	adults	-	13	mean±SD	3.0±0.8	-	-	-	-	3.6±1.2	14±5.8	0.7±0.4	42±16	-	-	-	-	6
"	whole plasma	1997	f	adults	-	14	mean±SD	3.2±1.7	-	-	-	-	2.9±1.2	29±16	1.7±3.2	33±21	-	-	-	-	6

Continued next page.

Annex Table 14 continued.

Species/ Region/Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	ΣCBz	HCB	α-HCH	β-HCH	γ-HCH (lindane)	ΣHCHs	ΣCHLs	ΣDDTs	ΣPCBs	Toxaphene	Dieldrin	Mirex	ΣPBDEs	Ref- erence	
E Greenland	fat	1999-2000	?	<3	65.9	2	mean	26.0	-	-	-	-	95.1	459	299	3190	-	-	-	-	7	
"	fat	1999-2000	f	2-10	82.6±4.86	8	mean±SD	37.4±34.1	-	-	-	-	132±52.0	678±292	282±52.8	5980±2310	-	-	-	-	7	
"	fat	1999-2000	m	2-10	79.7±8.62	9	mean±SD	19.5±10.3	-	-	-	-	231±171	487±226	371±231	6120±2730	-	-	-	-	7	
NW Iceland																						
pack ice, near Horn	fat	1993	?	3.5	-	1	-	-	102	98	84	8	190	1570	678	7860	-	-	-	-	3	
"	liver	1993		3.5	-	1	-	-	15	12	16	8	36	1760	270	1660	-	-	-	-	3	
Barents Sea																						
Svalbard	fat	1998	m,f	5-26 yrs	5-67	20	range	-	-	-	-	-	-	-	-	-	-	-	-	-	(3.6-49.0)	8
"	blood plasma	1995-1998	-	cubs (<1 yr)	1.3	33	geo.mean(range)	-	-	-	-	-	-	-	-	-	160(45.2-377)	-	-	-	-	9
"	blood plasma	1995-1998	f	adults w. cub	1.2	25	geo.mean(range)	-	-	-	-	-	-	-	-	-	69.8(19.1-139)	-	-	-	-	9
"	blood plasma	1995-1998	-	yearling	1.6	12	geo.mean(range)	-	-	-	-	-	-	-	-	-	109(24.5-376)	-	-	-	-	9
"	blood plasma	1995-1998	f	adults w. yearl.	1.3	10	geo.mean(range)	-	-	-	-	-	-	-	-	-	38.3(13.8-97.8)	-	-	-	-	9
"	whole plasma	1998	-	subadults	-	4	mean±SD	4.5±1.6	-	-	-	-	1.4±0.8	55±16	0.8±0.2	228±36	-	-	-	-	6	
"	whole plasma	1998	m	adults	-	18	mean±SD	3.0±2.6	-	-	-	-	2.7±1.5	19±11	1.1±0.6	95±50	-	-	-	-	6	
"	whole plasma	1998	f	adults	-	16	mean±SD	2.1±1.0	-	-	-	-	1.9±1.9	26±8.7	1.0±0.6	92±43	-	-	-	-	6	
"	blood	1991-1994	f	adults	0.7	32	geo.mean (range)	- (0.34-1.04)	1.08 (0.22-3.98)	0.57 (0.15-1.52)	1.05 (0.10-2.76)	-	-	11.3 (3.87-23.2)	0.43 (0.08-1.69)	35.3 (1.50-93.8)	-	-	-	-	10,11	
Franz Josef Land	blood	1995	f	adults	0.52 (0.35-0.77)	17	geo.mean (range)	- (0.49-10.2)	1.16 (0.16-0.55)	0.30 (0.83-2.69)	0.83 (0.33-2.69)	-	-	15.4 (9.36-26.7)	0.44 (0.11-2.96)	58.2 (31.0-162)	-	-	-	-	10,11	
Kara Sea	blood	1991-1994	f	adults	0.83 (0.76-1.13)	12	geo.mean (range)	- (7.14-80.8)	21.7 (0.83-5.64)	2.90 (0.83-5.64)	21.6 (5.06-44.9)	-	-	2540 (665-4930)	7.80 (2.99-31.2)	781 (228-2040)	-	-	-	-	10,11	
E-Siberian Sea	blood	1992-1993	f	adults	0.75 (0.55-0.97)	8	geo.mean (range)	- (0.89-2.59)	1.41 (0.60-1.44)	0.82 (0.60-1.44)	2.22 (1.02-4.42)	-	-	20.5 (9.75-58.8)	0.28 (0.075-3.64)	26.7 (11.8-81.7)	-	-	-	-	10,11	
Chukchi Sea	blood	1987-1992	f	adults	0.55 (0.39-0.76)	21	geo.mean (range)	- (0.94-6.33)	1.78 (0.81-2.65)	14.9 (0.81-2.65)	2.95 (1.45-6.22)	-	-	15.8 (6.64-31.6)	0.14 (0.03-0.38)	18.4 (8.25-37.2)	-	-	-	-	10,11	
Sea Otter ( <i>Enhydra lutris</i> ) (ng/g ww)																						
Alaska																						
SE Alaska	liver	1991	m	adults	3±1	7	mean±SD	-	1	-	6±3	-	-	1±1	1±3	8±14	-	2±4	-	-	12	
Aleutian Islands	liver	1991-1992	m,f	adults/young	3±2	7	mean±SD	-	2±4	-	5±16	-	-	15±97	36±500	310±480	-	3±5	-	-	12	

\* Significant differences (p&lt;0.05) between 1984 and 1990.

## References

- Krahn *et al.*, 2002; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, nonachlor III, heptachlor, and heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*- DDE, -DDD, and -DDT; ΣPCBs = sum of 40 congeners (17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 87, 82, 95, 99, 101, 105, 110, 118, 128, 138, 149, 151, 153, 156, 158, 170, 171, 177, 180, 183, 187, 191, 194, 195, 199, 205, 206, 208, 209).
- Hoekstra *et al.*, 2003b; ΣCBz = sum of 1,2-diCBz, 1,4-diCBz, 1,2,3-triCBz, 1,2,4-triCBz, 1,3,5-triCBz, 1,2,3,4-tetraCBz, 1,2,3,5-tetraCBz, pentaCBz, and hexaCBz; ΣHCHs = sum of α-, β-, γ-, and δ-HCH; ΣCHLs = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDTs = sum of *o,p'*- and *p,p'*-DDD, -DDE, and -DDT; ΣPCBs = sum of 101 congeners (4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 31/28, 33/21/53, 51, 22, 45, 46, 52/49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95, 66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180/193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208 & 209); Toxaphene = sum of 24 chloroborane congeners.
- Klobes *et al.*, 1998b; ΣHCHs = sum of β- and γ-HCH; ΣCHLs = sum of *trans*-nonachlor and oxychlordane; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 7 congeners (118, 153, 138, 163, 180, 170, 194).
- Evans, 2001; ΣCBz = sum of 3 compounds; ΣHCHs = α-, β-, γ-, δ-HCH; ΣDDTs = *o,p'*- and *p,p'*-DDE, -DDD and -DDT; ΣCHLs = sum of 13 compounds, including oxychlordane, nonachlor III, *trans*-nonachlor, and heptachlor epoxide; ΣPCBs = sum of 22 congeners. Concentrations converted to wet weight using mean % lipid content.
- Norstrom, 1999a; 1999b; Muir and Norstrom, 2000; ΣCBz = sum of 1,2,4,5-TeCBz, PeCBz, and HCB; ΣHCHs = α-, β-, and γ-HCH.
- Sandau, 2000.
- Sonne-Hansen, 2002.
- van Bavel *et al.*, 2001; ΣPBDEs = sum of 21 tetra-, penta-, and hexa-BDEs (several unnamed, but includes 47, 99, 85, 153, 138).
- Lie *et al.*, 2000; ΣPCBs = sum of 34 congeners (28, 31, 47, 52, 56, 66, 74, 87, 99, 101, 105, 110, 114, 118, 128, 136, 137, 138, 141, 149, 151, 153, 156, 157, 170, 180, 183, 187, 189, 194, 196, 199, 206, 209). Concentrations converted to wet weight using mean % lipid content.
- Andersen *et al.*, 2001b; ΣPCBs = sum of 6 congeners (99,118,153,156,180,194). Concentrations converted to wet weight using mean % lipid content.
- Lie *et al.*, 2003; ΣCHLs = oxychlordane; ΣDDTs = *p,p'*-DDE; ΣPCBs = sum of 6 congeners (99, 118, 153, 156, 180, 194). Concentrations converted to wet weight using mean % lipid content.
- Bacon *et al.*, 1999.



Annex Table 15. Congener specific concentrations of toxaphenes in marine biota.

Species/Region	Location	Tissue	Year	Sex	Age, years	% lipid	n	Statistic	Parlar 26	Parlar 32	Parlar 40	Parlar 44	Parlar 50	Parlar 62	Total toxaphene	Reference
<b>Freshwater fish and invertebrates (ng/g ww)</b>																
Unidentified zooplankton																
Bjørnøya	Lake Øyangen	whole body	1998	-	-	-	-	-	-	-	-	-	-	-	n.d.	1
"	Lake Ellasjøen	whole body	1999	-	-	-	-	-	-	-	-	-	-	-	1.2	1
Arctic char																
Bjørnøya	Lake Øyangen	muscle	1998	-	-	-	1 pool (n=3)	-	-	-	-	-	-	-	4.5	1
"	"	muscle	1998	-	-	-	1 pool (n=3)	-	-	-	-	-	-	-	n.d.	1
"	"	muscle	1998	-	-	-	1 pool (n=3)	-	-	-	-	-	-	-	n.d.	1
"	Lake Ellasjøen	muscle	1999	-	-	-	?	-	-	-	-	-	-	-	8.3	1
"	"	muscle	1999	-	-	-	?	-	-	-	-	-	-	-	7.8	1
"	"	muscle	1999	-	-	-	?	-	-	-	-	-	-	-	9.2	1
<b>Marine fish (ng/g ww)</b>																
Cod																
Norway	Jarfjord	liver	-	m	-	60±5	5	mean±SD	23±12	-	-	-	27±16	4.7±4.7	-	2
"	"	liver	-	f	-	61±3	5	mean±SD	21±11	-	-	-	26±15	4.2±1.6	-	2
Russia	White Sea	muscle & skin	-	-	-	1.01±0.30	-	mean±SD	0.1±0.10	-	-	-	0.08±0.06	n.d.	1.47±0.27	3
Redfish																
Barents Sea	N Norway	muscle	-	-	-	2.80(1.3-6.2)	24	mean (range)	-	-	-	-	-	-	76.2(37.0-181)	4
Navaga																
Russia	White Sea	muscle & skin	-	-	-	1.54±0.11	-	mean±SD	0.21±0.08	-	-	-	0.06±0.05	n.d.	1.41±0.39	3
Herring																
Russia	White Sea	muscle & skin	-	-	-	7.80±0.24	-	mean±SD	1.03±0.32	-	-	-	1.31±0.30	n.d.	28.5±1.65	3
<b>Pinnipeds (ng/g ww)</b>																
Ringed seal																
Alaska	Barrow	blubber	1998-1999	f	1-7	87.0±6.47	5	mean±SD	24.1±31.7	0.50±0.30	3.6±3.2	29.1±31.1	4.7±5.0	-	272±194	5
"	"	blubber	1998-2000	m	1-20	82.2±4.45	14	mean±SD	13.2±17.10	0.47±0.52	9.66±12.7	80.3±83.8	12.4±26.2	-	479±339	5
Canada	Hudson Strait	blubber	1998	f	-	80.4±10.4	10	mean±SD	42.4±33.4	-	-	-	6.58±5.14	1.28±2.15	266±250	6
"	"	blubber	1998	m	-	77±9.8	11	mean±SD	49.4±26.9	-	-	-	7.36±4.21	1.78±1.97	309±218	6
"	Ungava Bay	blubber	1998	f	-	101±16.5	16	mean±SD	16.7±10.4	-	-	-	2.28±1.68	0.32±0.54	87.7±55.1	6
"	"	blubber	1998	m	-	92.4±15.7	5	mean±SD	24.3±26.5	-	-	-	3.00±3.00	0.60±1.20	117±82.4	6
"	Labrador	blubber	1998	f	-	87.3±5.6	6	mean±SD	n.d.	-	-	-	n.d.	n.d.	n.d.	6
"	"	blubber	1998	m	-	88.2±5.5	14	mean±SD	n.d.	-	-	-	n.d.	n.d.	n.d.	6
Barents Sea	Kongsfjorden, NW Svalbard	blubber	-	f	≥4	-	7	mean	5.67	-	-	-	6.75	-	37.4	7
"	"	blubber	-	m	≥4	-	7	mean	6.93	-	-	-	8.10	-	45.9	7
"	"	blubber	-	juv	≤4	-	14	mean	3.6	-	-	-	4.68	-	24.7	7
"	"	blubber	1992	f	6	98±1	5	mean±SD (range)	1.8±0.4(1.3-2.3)	-	-	-	3.1±0.5(2.6-3.9)	1.6±0.3(1.1-2.0)	-	2
"	"	blubber	1992	m	-	96±3	5	mean±SD (range)	2.6±1.1(1.4-3.9)	-	-	-	4.9±7.7(2.4-7.7)	1.8±1.1(0.8-3.4)	-	2
White Sea	Dvina Bay	blubber	1998	m,f	1.1±0.6	86.4±6.0	17	mean±SD	8.2±19.2	-	-	-	5.6±14.3	0.1±0.6	166±233	4
Harp seal																
Barents Sea	E of Svalbard	blubber	1997	-	≤4	-	13	geo. mean	67.5	-	-	-	81.0	14.7	446	8
White Sea	-	blubber	1998	f	14.2±4.1	85.1±3.7	20	mean±SD	64.9±17.7	-	-	-	50.3±13.4	6.9±2.3	548±148	4
"	-	blubber	1998	m,f	<0.1	68.3±3.1	10	mean±SD	74.1±65.0	-	-	-	32.2±11.0	2.7±1.4	658±234	4
Bearded seal																
White Sea	-	blubber	1998	m	2.5	84.8	2	mean	34.0	-	-	-	32.2	n.d.	518	4
<b>Cetaceans (ng/g ww)</b>																
Beluga																
Barents Sea	E Spitsbergen (Svalbard)	blubber	-	-	subadults	-	-	geo. mean (95% CI)	398(189-838)	-	165(78.2-347)	-	863(409-1820)	-	1420(704-2890)	9
Narwhal																
Barents Sea	E Spitsbergen (Svalbard)	blubber	-	-	subadults	-	-	geo. mean (95% CI)	1520(1090-2140)	63.4(50.6-79.6)	634(452-891)	319(227-447)	2240(1590-3140)	-	4780(3400-6710)	9
<b>Polar bears (ng/g ww)</b>																
Polar bear																
Barents Sea	Svalbard	blubber	1990-94	f	>5	92±6(85-96)	3	mean±SD (range)	5.7±1.9(3.8-7.5)	-	-	-	3.3±0.9(2.2-3.8)	0.4±0.1(0.3-0.4)	-	2
"	"	blubber	1990-94	m	>5	72±33(7-91)	6	mean±SD (range)	17±26(0.8-68)	-	-	-	27±46(0.4-119)	4.1±6.2(0.2-11)	-	2
<b>Seabirds (ng/g ww)</b>																
Glaucous gull																
Barents Sea	Bjørnøya	liver	1995	m	-	8.1 (7.0-9.2)	3	mean±SD	44.4±23.6	n.d.	-	-	74.0±38.3	-	-	10
"	"	liver	1999	m,f	-	5.9 (3.4-11.5)	15	mean±SD	10.1±6.6	5.0±6.3	6.6±4.7	-	19.2±14.0	-	-	10
"	"	gut	1999	-	-	-	1 pool (n=15)	-	-	-	-	-	-	-	84.2	1
"	Longyearbyen, Norway	liver	1995	m,f	-	7.0 (6.0-8.4)	4	mean±SD	17.7±5.2	n.d.	-	-	26.0±9.5	-	-	10
Great black-backed & herring gulls																
Norway	Alta	eggs	2001	-	-	-	20	mean	44.4	<0.2	-	-	111	44.3	200	11
"	Kongsfjord (Finmark)	eggs	2001	-	-	-	20	mean	35.7	<0.24	-	-	102	26.6	165	11
"	Sommarøy	eggs	2001	-	-	-	20	mean	55.7	<0.27	-	-	147	83.5	286	11
"	Vardø	eggs	2001	-	-	-	20	mean	34.5	<0.33	-	-	94.6	41.1	170	11

## References

- Kallenborn, 2002c; Total toxaphene = sum 10 congeners (Parlars 26, 32, 38, 40, 44, 50, 51, 58, 62, 69).
- Føreid *et al.*, 2000.
- Muir *et al.*, 2003.
- McHugh *et al.*, 2000; Total toxaphene = sum of Parlars 26, 50, 62.
- Hoekstra, 2003a.
- Muir *et al.*, 1999c.
- Wolkers *et al.*, 1998b; Concentrations converted to wet weight assuming 90% lipid content.
- Wolkers *et al.*, 2000; Concentrations converted to wet weight assuming 90% lipid content.
- Wolkers, 2002; Concentrations converted to wet weight basis assuming 90% lipid content.
- Herzke *et al.*, 2003.
- Gabrielsen, 2002; Total toxaphene = sum of Parlars 26, 50, 62.

Annex Table 16. Concentrations and TEQs for PCDDs, PCDFs and non-ortho PCBs (nPCBs) in biota in pg/g dw, ww or lw.

Species/Region	Location	Tissue	Year	Age/Sex	% Lipid	n	Units	Concentration			TEQ			ΣTEQs		Reference
								ΣPCDDs	ΣPCDFs	ΣnPCBs	ΣPCDDs	ΣPCDFs	ΣnPCBs	pg/g lw	pg/g ww	
<b>Freshwater invertebrates</b>																
<b>Plankton</b>																
Great Slave Lake, NWT	W Basin	whole body	1994-95	–	–	–	pg/g ww	45.3	22.8	–	–	–	–	–	–	1
„	E Arm	whole body	1994-95	–	–	–	pg/g ww	–	–	–	–	–	–	–	–	1
<b>Freshwater fish</b>																
<b>Whitefish</b>																
Great Slave Lake, NWT	W Basin	muscle	1994-95	–	–	–	pg/g ww	0.05	0.9	–	–	–	–	–	–	1
„	E Arm	muscle	1994-95	–	–	–	pg/g ww	n.d.	0.6	–	–	–	–	–	–	1
Kola Peninsula	–	muscle	2000-01	7-11, m	0.9	10	pg/g ww	0.26	0.77	–	–	–	11.23	0.1	–	2
Pechora Basin	–	muscle	2000-01	7-14, m	4.0	12	pg/g ww	0.37	0.38	–	–	–	1.93	0.88	–	2
Taymir Peninsula	Dudinka	muscle	2000-01	8-14, m	1.7	10	pg/g ww	0.17	0.23	–	–	–	1.67	0.03	–	2
„	Khatanga	muscle	2000-01	8-14, m	2.0	10	pg/g ww	0.12	0.27	–	–	–	1.98	0.04	–	2
<b>Broad whitefish</b>																
Chukotka Peninsula	Kanchalan	muscle	2000-01	7-11, m	1.1	10	pg/g ww	0.22	0.13	–	–	–	2.19	0.02	–	2
Taymir Peninsula	Khatanga	muscle	2000-01	6-14, m	1.6	10	pg/g ww	0.29	0.18	–	–	–	2.46	0.04	–	2
<b>Lake trout</b>																
Great Slave Lake, NWT	W Basin	muscle	1994-95	–	–	–	pg/g ww	0.15	1.0	–	–	–	–	–	–	1
„	E Arm	muscle	1994-95	–	–	–	pg/g ww	n.d.	0.7	–	–	–	–	–	–	1
Norway	Lake Takvatn	muscle	1999/2000	–	1.8	1	pg/g ww	–	–	–	0.11	–	0.20	17.2	0.31	18
„	Lake Fjellfrøsvatnet	muscle	1999/2000	–	1.1	1	pg/g ww	–	–	–	0.09	–	0.12	19.1	0.21	18
„	Lake Grunnvatnet	muscle	1999/2000	–	1.3	1	pg/g ww	–	–	–	0.11	–	0.06	13.1	0.17	18
„	Lake Store Raudvannet	muscle	1999/2000	–	2.5	1	pg/g ww	–	–	–	0.31	–	0.55	34.4	0.86	18
<b>Burbot</b>																
Great Slave Lake, NWT	W Basin	liver	1994-95	–	–	–	pg/g ww	1.05	2.7	–	–	–	–	–	–	1
„	E Arm	liver	1994-95	–	–	–	pg/g ww	n.d.	5.7	–	–	–	–	–	–	1
Norway	Lake Grensefoss (Paskvikelva)	muscle	1999/2000	–	11.6	1	pg/g ww	–	–	–	3.25	–	9.50	120	12.8	18
Omul	Dudinka	muscle	2000-01	8-12, m	2.9	10	pg/g ww	0.42	0.84	–	–	–	–	5.9	0.17	2
<b>Arctic char</b>																
Norway	Lake Ellasjøen	muscle	1999/2000	–	1.3	1	pg/g ww	–	–	–	0.2	–	7.16	566	7.4	18
Chukotka Peninsula	Lavrentiya	muscle	2000-01	3-7, m	3.1	10	pg/g ww	2.76	0.99	–	–	–	–	1.41	0.14	2
Ide	Pechora Basin	muscle	2000-01	6-11, m	3.3	12	pg/g ww	0.42	0.23	–	–	–	–	1.31	0.04	2
Inconnu	Chukotka Peninsula	muscle	2000-01	7-14, m	0.76	10	pg/g ww	0.26	0.16	–	–	–	–	3.09	0.02	2
Northern Pike	Kola Peninsula	muscle	2000-01	7-13, m	0.64	12	pg/g ww	0.26	0.50	–	–	–	–	11.15	0.07	2
<b>Marine invertebrates</b>																
<b>Worms (Nephtys, Sipanclid, wormtube)</b>																
Kara Sea	–	whole body	1993	–	–	–	pg/g dw	29	1.25	17	–	–	–	–	–	3
<b>Crustaceans (isopods, amphipods)</b>																
Kara Sea	–	whole body	1993	–	–	–	pg/g dw	7.8	20.0	84	–	–	–	–	–	3
<b>Bivalves (clams)</b>																
Kara Sea	–	whole body	1993	–	–	–	pg/g dw	23	6.8	33	–	–	–	–	–	3
<b>Marine fish</b>																
<b>Chum salmon</b>																
Chukotka Peninsula	Lavrentiya	muscle	2000-01	4-7, m	0.52	10	pg/g ww	0.32	0.40	–	–	–	–	14	0.07	2
<b>Sturgeon</b>																
Kara Sea	–	liver	1993	–	–	–	pg/g dw	3.4	33	77	–	–	–	–	–	3
<b>Seabirds</b>																
<b>Black-legged kittiwake</b>																
Lancaster Sound, NT	Prince Leopold Isl.	liver	1975	–	9.0	4 pools (n=3)	pg/g ww	59.4	58.50	1850	34	44	13.6	2390	214	4
„	„	liver	1993	–	4.3	5 pools (n=3)	pg/g ww	8.6	8.60	28	5.2	21.5	21.5	1120	47	4
„	„	egg	1993	–	5.1	1 pool (n=3)	pg/g ww	9.7	44.00	554	6.6	34	37	1530	78	4
<b>Northern fulmar</b>																
Lancaster Sound, NT	Prince Leopold Isl.	liver	1975	–	2.6	5 pools (n=3)	pg/g ww	222	712.0	140	70.5	621.0	10.1	27000	69	4
„	„	liver	1993	–	4.4	5 pools (n=3)	pg/g ww	108	318.00	1060	27.7	273	60	8190	357	4
„	„	egg	1993	–	8.9	1 pool (n=3)	pg/g ww	21.4	58.00	487	10.7	40	33	940	83	4
<b>Thick-billed murres or Brünnich's guillemot</b>																
Lancaster Sound, NT	Prince Leopold Isl.	liver	1975	–	8.6	3 pools (n=3)	pg/g ww	2.6	4.30	2760	1.7	3.4	161	1920	166	4
„	„	liver	1993	–	3.3	3 pools (n=3)	pg/g ww	5.6	14.20	167	3.0	9.6	11.2	720	24	4
„	„	egg	1993	–	6.8	1 pool (n=3)	pg/g ww	4.8	14.30	224	2.7	8.2	15.0	380	26	4
<b>Black guillemots</b>																
Labrador	Saglek Bay (beach site)	liver	1999	22 d	3.8	11	pg/g ww	8.30	4.50	63600	4.5	–	20.0	640	25	5

Continued next page.

Annex Table 16 continued.

Species/Region	Location	Tissue	Year	Age/Sex	% Lipid	n	Units	Concentration			TEQ			ΣTEQs		Reference
								ΣPCDDs	ΣPCDFs	ΣnPCBs	ΣPCDDs	ΣPCDFs	ΣnPCBs	pg/g lw	pg/g ww	
Common guillemot																
Faroe Islands	–	eggs	2000		15.0	10	pg/g ww	–	–	–	–	–	–	66	10	19
Great black-backed & herring gulls																
Norway	Alta	eggs	2001	–	–	1 pool (n=20)	pg/g ww	–	–	–	3.7	–	32.8	–	54	6
	Kongsfjord (Finmark)	eggs	2001	–	–	1 pool (n=20)	pg/g ww	–	–	–	4.2	–	43.0	–	72	6
	Sommarøy	eggs	2001	–	–	1 pool (n=20)	pg/g ww	–	–	–	3.7	–	32.5	–	59	6
	Vardø	eggs	2001	–	–	1 pool (n=20)	pg/g ww	–	–	–	5.2	–	0.20	–	36	6
Glaucous gull																
Svalbard	Longyearbyen	liver	1990	adults	4.2±0.2	13	pg/g ww	–	–	2700000	–	–	2500	60000	2500	20
Goldeneye																
Kola Peninsula	–	muscle	2001	1-3	3.3	1 pool (n=12)	pg/g ww	0.57	0.45	–	–	–	–	5	0.17	2
Northern pintail																
Pechora Basin	–	muscle	2001	1-3	2.6	1 pool (n=10)	pg/g ww	0.17	0.11	–	–	–	–	1	0.3	2
Taymir Peninsula	Dudinka	muscle	2001	1-3	2.4	1 pool (n=5)	pg/g ww	0.42	0.36	–	–	–	–	4.6	0.11	2
Oldsquaw																
Taymir Peninsula	Khatanga	muscle	2001	1-2	2.2	1 pool (n=6)	pg/g ww	0.10	0.22	–	–	–	–	6.4	0.14	2
Chukotka Peninsula	Kanchalan	muscle	2001	1-2	2.7	1 pool (n=2)	pg/g ww	0.12	0.09	–	–	–	–	1.9	0.52	2
”	Lavrentiya	muscle	2001	1-3	3.6	1 pool (n=9)	pg/g ww	0.13	0.11	–	–	–	–	0.5	0.02	2
Grey heron																
mid-Norway	Frøya	yolk sac	–	1 d	–	4	pg/g ww	–	–	–	–	–	–	1170	79.4	7
N Norway	Finnfjordøy	yolk sac	–	1 d	–	7	pg/g ww	–	–	–	–	–	–	1020	93.2	7
Marine mammals																
Ringed seal																
NT, Canada	Pangnirtung, Cumberland Sound	blubber	1993	7-9, m		3	pg/g lw	–	–	31000±11200	–	–	0.72±0.20	0.72	–	8
”	Pangnirtung, Cumberland Sound	blubber	1993	8-13, f		3	pg/g lw	–	–	17800±1590	–	–	0.59±0.12	0.59	–	8
NWT, Canada	Holman Island	blubber	2000	0-35 year old m,f	91.3±3.1	20	pg/g ww	–	–	–	2.6	–	8.6±7.3	25	22	9,24
Russia	Kara Sea	blubber	1995	adults, f	92	3	pg/g ww	–	–	6530	–	–	147	160	147	23
Chukotka Peninsula	Lavrentiya	muscle	2001	1-3	5.4	1 pool (n=6)	pg/g ww	0.64	0.44	–	–	–	–	1.9	0.10	2
”	”	blubber	2001	1-3	98.9	1 pool (n=6)	pg/g ww	4.86	4.84	–	–	–	–	1.1	1.10	2
Bearded seal																
Chukotka Peninsula	Lavrentiya	muscle	2001	2-3	2.3	1 pool (n=2)	pg/g ww	0.65	0.38	–	–	–	–	4.4	0.10	2
”	”	blubber	2001	2-3	86.2	1 pool (n=2)	pg/g ww	10.60	4.30	–	–	–	–	1.0	0.97	2
Spotted seal																
Chukotka Peninsula	Lavrentiya	muscle	2001	1-2	4.5	1 pool (n=10)	pg/g ww	0.79	1.16	–	–	–	–	4.3	0.19	2
”	”	blubber	2001	1-2	83.0	1 pool (n=10)	pg/g ww	4.52	3.84	–	–	–	–	1.5	1.25	2
Northern fur seal																
Alaska	Pribilof Islands	blubber	1997-2000	subadults m,f	58.6±11.0	10	pg/g lw	–	–	–	–	–	28.7±17.9	30	17	10
”	”	blubber	1997-2000	pups	35.3±20.6	3	pg/g lw	–	–	–	–	–	89.8±94.4	90	32	10
”	”	blubber	1997-2000	adults f	45.7±4.27	4	pg/g lw	–	–	–	–	–	46.2±25.7	46	21	10
”	”	blubber	1997-2000	fetuses	38.5±21.9	2	pg/g lw	–	–	–	–	–	28.4±1.43	28	11	10
”	St. George Isl., Pribilof Islands	blood	1996	neonates of young dams	0.30±0.39	23	pg/g ww	–	–	–	–	–	0.13±0.10	43	0.13	11
”	”	blood	1996	neonates of old dams	0.22±0.15	25	pg/g ww	–	–	–	–	–	0.06±0.06	27	0.06	11
”	”	blood	1996	pups of young dams	0.28±0.48	21	pg/g ww	–	–	–	–	–	0.08±0.04	29	0.08	11
”	”	blood	1996	pups of old dams	0.31±0.32	21	pg/g ww	–	–	–	–	–	0.08±0.32	26	0.08	11
Minke whale																
North Sea	–	blubber	2001	adults f	72	5	pg/g ww	–	–	–	1.9	–	27.5	93	66.9	12
					43-88						0.6-2.3	–	8.2-48.4		25.1-103	
Barents Sea	Spitsbergen	blubber	2001	adults f	63	5	pg/g ww	–	–	–	1.2	–	11.8	38	24.1	12
					35-75						0.5-1.8	–	5.3-19.7		10.3-36.6	
Gray whale																
Chukotka Peninsula	Lavrentiya	muscle	2001	2-3		1	pg/g ww	0.61	0.22	–	–	–	–	8.1	0.06	2
”	”	blubber	2001	2-3		1	pg/g ww	8.60	2.25	–	–	–	–	1.76	0.75	2
Beluga																
NT, Canada	Kimmirut	blubber	1994	1-6, m		3	pg/g lw	–	–	185000±116000	–	–	1.84±0.66	1.84	–	13
”	”	blubber	1994	6-20, f		3	pg/g lw	–	–	68300±47100	–	–	1.49±1.05	1.49	–	13
Baffin Island	Cumberland Sound	blubber	1997											6.1	–	22
Killer whales																
Alaska (resident)	Kenai Fiords/ Prince William Sound	blubber	1994-99	immature and mature, m,f	28±9.8	64	pg/g ww	–	–	–	–	–	29±33	100±98	29±33	14
Alaska (transient)	Kenai Fiords/ Prince William Sound	blubber	1994-99	immature and mature, m,f	24±9.5	13	pg/g ww	–	–	–	–	–	220±190	860±640	220±190	14
Harbour porpoise																
Norwegian Sea	W coast of Norway	blubber	1988-90	m	–	8	pg/g lw	12±4.8		1560±2740	3.1	–	108	111	–	15
Greenland	SW coast	blubber	1995	immature and mature, m,f	–	4	pg/g lw	3.6			0.4	–	–	0.4	–	21
Arctic fox																
Alaska	Pribilof Islands	fat	1996-97	f	44.0	3	pg/g ww	–	–	–	–	–	22.7	48	22.7	16
”	”	fat	1996-97	m	49.0	5	pg/g ww	–	–	–	–	–	69.5	151	69.5	16

Continued next page.

Annex Table 16 continued.

Species/Region	Location	Tissue	Year	Age/Sex	% Lipid	n	Units	Concentration			TEQ			ΣTEQs		Reference
								ΣPCDDs	ΣPCDFs	ΣnPCBs	ΣPCDDs	ΣPCDFs	ΣnPCBs	pg/g lw	pg/g ww	
<b>Polar bear</b>																
Canada	Queen Maud Gulf (I)	adipose	1984	–	–	1 pool (n=9)	pg/g lw	13.6	–	–	–	–	–	–	–	17
”	”	adipose	1990	–	–	1 pool (n=10)	pg/g lw	3.78	–	–	–	–	–	–	–	17
”	Barrow Strait (II)	adipose	1984	–	–	1 pool (n=7)	pg/g lw	15.8	–	–	–	–	–	–	–	17
”	”	adipose	1990	–	–	1 pool (n=9)	pg/g lw	3.40	–	–	–	–	–	–	–	17
”	North Baffin Bay (III)	adipose	1984	–	–	1 pool (n=10)	pg/g lw	4.00	–	–	–	–	–	–	–	17
”	”	adipose	1990	–	–	1 pool (n=10)	pg/g lw	2.90	–	–	–	–	–	–	–	17
”	Davis Strait (IV)	adipose	1984	–	–	1 pool (n=7)	pg/g lw	2.20	–	–	–	–	–	–	–	17
”	”	adipose	1990	–	–	1 pool (n=10)	pg/g lw	1.20	–	–	–	–	–	–	–	17
<b>Terrestrial mammals &amp; birds</b>																
<b>Reindeer</b>																
Russia	Kola Peninsula	liver	2001	2-4	6.3	10	pg/g ww	6.49	23.6	–	–	–	–	105	6.52	2
”	”	muscle	2001	2-4	4.9	10	pg/g ww	0.78	4.24	–	–	–	–	20	0.98	2
”	Pechora Basin	liver	2001	2-4	6.4	10	pg/g ww	1.52	10.5	–	–	–	–	38	2.45	2
”	”	muscle	2001	2-4	4.5	10	pg/g ww	0.37	0.38	–	–	–	–	2.2	0.10	2
”	Dudinka, Taymir Peninsula	liver	2001	1-8	3.9	10	pg/g ww	0.59	2.40	–	–	–	–	18.2	0.71	2
”	”	muscle	2001	1-8	2.4	10	pg/g ww	0.16	0.13	–	–	–	–	1.3	0.03	2
”	Khatanga, Taymir Peninsula	liver	2001	1-8	6.1	10	pg/g ww	0.61	1.51	–	–	–	–	8.0	0.49	2
”	”	muscle	2001	1-8	11.1	10	pg/g ww	0.29	0.19	–	–	–	–	0.75	0.08	2
”	Kanchalan, Chukotka Peninsula	liver	2001	3-4	4.3	10	pg/g ww	1.03	0.80	–	–	–	–	5.2	0.22	2
”	”	muscle	2001	3-4	1.5	10	pg/g ww	0.32	0.34	–	–	–	–	4.4	0.07	2
”	Lavrentiya, Chukotka Peninsula	liver	2001	2-5	5.7	10	pg/g ww	0.48	0.65	–	–	–	–	4.2	0.24	2
”	”	muscle	2001	2-5	2.1	10	pg/g ww	0.57	0.33	–	–	–	–	2.6	0.05	2
<b>Hare</b>																
Russia	Kola Peninsula	muscle	2001	0-2	2.0	10	pg/g ww	0.46	2.86	–	–	–	–	29.5	0.60	2
”	Pechora Basin	muscle	2001	0-3	1.6	10	pg/g ww	0.09	0.20	–	–	–	–	2.7	0.04	2
”	Dudinka, Taymir Peninsula	muscle	2001	0-3	2.3	15	pg/g ww	0.30	0.10	–	–	–	–	1.4	0.03	2
”	Khatanga, Taymir Peninsula	muscle	2001	1-3	1.2	10	pg/g ww	0.14	0.09	–	–	–	–	3.4	0.04	2
”	Lavrentiya, Chukotka Peninsula	liver	2001	1-3	3.9	10	pg/g ww	0.85	0.35	–	–	–	–	2.5	0.10	2
”	”	muscle	2001	1-3	2.1	10	pg/g ww	0.13	0.14	–	–	–	–	1.8	0.04	2
<b>Ptarmigan</b>																
Russia	Pechora Basin	muscle	2001	0-3	1.7	1 pool (n=10)	pg/g ww	0.12	0.25	–	–	–	–	2.8	0.05	2
”	Dudinka, Taymir Peninsula	muscle	2001	1-3	2.1	1 pool (n=20)	pg/g ww	0.22	0.16	–	–	–	–	1.8	0.04	2
<b>Willow grouse</b>																
Russia	Kola Peninsula	muscle	2001	1-3	1.9	1 pool (n=20)	pg/g ww	0.40	0.24	–	–	–	–	3.1	0.06	2
”	Khatanga, Taymir Peninsula	muscle	2001	1-2	2.2	1 pool (n=10)	pg/g ww	0.47	0.14	–	–	–	–	2.2	0.05	2
”	Kanchalan, Chukotka Peninsula	muscle	2001	1-3	2.3	1 pool (n=10)	pg/g ww	0.07	0.08	–	–	–	–	0.9	0.20	2
”	Lavrentiya, Chukotka Peninsula	muscle	2001	1-3	1.5	1 pool (n=10)	pg/g ww	0.03	0.08	–	–	–	–	1.2	0.02	2

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- Norstrom, 1994; ΣPCDDs = sum of 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD.
- Schlabach *et al.*, 2001.
- Mikkelsen *et al.*, in prep.
- Daelemans *et al.*, 1992; ΣnPCBs = PCB77, 126, 169, 114, 118, 123, 156, 157, 167, 189.
- Bruhn *et al.*, 1999.
- Stern and Addison, 1999.
- Nakata *et al.*, 1998a; ΣnPCBs = PCB77, 126, 169, 105, 118, 156, 180.
- Ikonomou, 2002.



Annex Table 17. Congener specific concentrations of PBDEs in marine, freshwater and terrestrial biota.

Species/Region	Tissue	Year	Sex	% lipid	n	Statistic	BDE15	BDE52	BDE47	BDE49	BDE85	BDE99	BDE100	BDE138	BDE153	BDE154	BDE183	BDE209	ΣPBDEs	Reference
<b>Terrestrial biota (ng/g ww)</b>																				
Peregrine falcon																				
N Sweden	egg	1991-1999	-	5.57 4.05-6.73	17	mean range	-	-	16 1.3-150	-	-	40 7.6-130	25 4.3-210	-	90 17-410	19 3.2-180	15 3-68	5.5 (n=8) 1.3-14	260 43-1580	1
<b>Freshwater fish (ng/g ww)</b>																				
Burbot																				
Fort Good Hope, NWT	liver	1988	m,f	30.2±13.5	10	mean±SD	-	-	226±280	-	-	84.5±131	35.2±46.7	-	29.4±44.7	20.5±28.9	-	-	-	2
"	liver	1999	m,f	35.0±9.59	4	mean±SD	-	-	583±522	-	-	370±270	208±155	-	161±125	157±116	-	-	-	2
"	liver	2000	m,f	33.3±13.1	11	mean±SD	-	-	620±629	-	-	320±274	180±183	-	135±134	81.3±84.2	-	-	-	2
Lake Hurdal, Norway	liver	-	-	45.7	1	-	<0.01	<0.02	149	-	-	43.0	-	-	26.8	-	-	-	-	3
Mjøsa/Furnes, Norway	liver	-	-	49.7	1	-	<0.01	<0.02	1040	-	-	910	-	-	188	-	-	-	-	3
Møsa/Lillehammer	liver	-	-	42.7	1	-	<0.01	<0.02	324	-	-	332	-	-	32.1	-	-	-	-	3
Lake Grensefoss (Pasvikelva), Norway	muscle	1999/2000	-	11.6	1?	-	-	-	9.69	-	-	10.6	-	-	-	-	-	-	20.3	4
Trout																				
Lake Takvatn, Norway	muscle	1999/2000	-	1.8	1?	-	-	-	0.10	-	-	0.06	-	-	-	-	-	-	0.15	4
Lake Fjellfrøsvatnet 99, Norway	muscle	1999/2000	-	1.1	1?	-	-	-	0.08	-	-	0.07	-	-	-	-	-	-	0.14	4
Lake Grunnvatnet, Norway	muscle	1999/2000	-	1.3	1?	-	-	-	0.06	-	-	0.04	-	-	-	-	-	-	0.10	4
Lake Store Raudvannet, Norway	muscle	1999/2000	-	2.5	1?	-	-	-	0.21	-	-	0.16	-	-	-	-	-	-	0.36	4
Char																				
Lake Ellasjøen, Svalbard	muscle	1999/2000	-	1.3	1?	-	-	-	8.27	-	-	8.02	-	-	-	-	-	-	16.3	4
<b>Marine fish (ng/g ww)</b>																				
Atlantic cod																				
Lofoten, Norway	liver	-	-	67.7	1	-	<0.01	<0.01	15.0	-	-	0.77	-	-	0.60	-	-	-	-	5
W coast, Norway	liver	-	-	75.2	1	-	<0.01	<0.03	19.5	-	-	0.83	-	-	1.00	-	-	-	-	5
S coast, Norway	liver	-	-	45.2	1	-	<0.02	<0.04	48.8	-	-	0.52	-	-	0.30	-	-	-	-	5
Oslofjord, Norway	liver	-	-	40.0	1	-	0.10	0.02	32.3	-	-	0.78	-	-	0.85	-	-	-	-	5
SW Greenland	liver	2000	-	57.6±3.8	3	mean±SD	-	-	1.79±0.20	-	-	0.32±0.07	-	-	-	-	-	-	5.13±0.85	6
"	muscle	2000	-	0.7	2	mean	-	-	0.21	-	-	3.35	-	-	-	-	-	-	3.35	6
Greenland halibut																				
SW Greenland	liver	2000	-	36.3±6.3	3	mean±SD	-	-	1.04±0.58	-	-	0.18±0.07	-	-	-	-	-	-	2.71±0.99	6
Tusk																				
Norwegian Sea	liver	-	-	60.4	1	-	<0.02	<0.03	36.5	-	-	1.48	-	-	1.55	-	-	-	-	5
Nordfjord	liver	-	-	46.8	1	-	<0.02	<0.04	142	-	-	1.98	-	-	8.70	-	-	-	-	5
<b>Seabirds (ng/g ww)</b>																				
Glaucous gull																				
Bjørnøya	liver	1995	m,f	3.4-11.5	18	range	n.d.	n.d.	0.14-1.8	-	-	-	-	-	n.d.	-	-	-	-	7
Great black-backed and herring gulls																				
Alta, Norway	eggs	2001	-	-	20	-	-	-	38.3	-	-	2.8	-	-	-	-	-	-	-	9
Kongsfjord (Finmark), Norway	eggs	2001	-	-	20	-	-	-	40.9	-	-	3.7	-	-	-	-	-	-	-	9
Sommarøy, Norway	eggs	2001	-	-	20	-	-	-	51.6	-	-	1.7	-	-	-	-	-	-	-	9
Vardø, Norway	eggs	2001	-	-	20	-	-	-	50.2	-	-	4.5	-	-	-	-	-	-	-	9
Thick-billed murre																				
SW Greenland	liver	1999	-	5.1±0.6	4	mean±SD	-	-	0.36±0.11	-	-	0.26±0.10	-	-	-	-	-	-	1.71±1.55	6
Black guillemot																				
Greenland	liver	1999	-	4.9±1.2	5	mean±SD	-	-	1.12±0.56	-	-	0.22±0.09	-	-	-	-	-	-	2.97±2.80	6
<b>Marine mammals (ng/g ww)</b>																				
Ringed seal																				
Holman	blubber	1996	m	-	8	mean	-	-	2.8	-	-	-	-	-	-	-	-	-	3.44	10
"	blubber	2000	m	-	8	mean	-	-	3.7	-	-	-	-	-	-	-	-	-	4.62	10
W Greenland	muscle	2000	-	10.5	2	mean	-	-	4.63	-	-	0.37	-	-	-	-	-	-	6.28	6
"	blubber	2000	-	88.1±7.1	9	mean±SD	-	-	2.4±0.7	-	-	0.4±0.2	-	-	-	-	-	-	3.6±1.1	6
NE Greenland	blubber	2000	-	106±18.7	8	mean±SD	-	-	40.6±15.5	-	-	4.6±1.2	-	-	-	-	-	-	58.0±22.6	6
Pangnirtung, Baffin Island	blubber	1993	f	94±1.5	3	mean±SD	-	-	0.33±0.15	-	-	0.11±0.01	-	-	-	-	-	-	0.05±0.18	11
"	blubber	1993	m	95	2	mean	-	-	0.76	-	-	0.09	-	-	-	-	-	-	0.97	11
Minke whale																				
Barents Sea	muscle	-	-	0.47	1	-	<0.01	<0.01	0.12	-	-	0.03	-	-	<0.05	-	-	-	-	3
"	muscle	-	-	1.03	1	-	<0.01	<0.01	0.35	-	-	0.10	-	-	<0.03	-	-	-	-	3
Norwegian Sea	muscle	-	-	0.72	1	-	<0.01	<0.01	2.12	-	-	0.94	-	-	0.07	-	-	-	-	3
"	muscle	-	-	3.62	1	-	<0.01	<0.01	11.0	-	-	3.48	-	-	<0.05	-	-	-	-	
Beluga																				
Kimmirut, Baffin Island	blubber	1994	f	92.9±1.7	3	mean±SD	-	-	1.09±0.82	-	-	0.19±0.06	-	-	-	-	-	-	1.44±1.01	11
"	blubber	1994	m	93.5±1.9	3	mean±SD	-	-	2.14±0.66	-	-	0.32±0.07	-	-	-	-	-	-	2.84±0.90	11
Hendrickson Island, W Canada	blubber	1989	m	90.1±3.7	12	mean±SD	-	-	5.81±2.26	3.45±1.53	-	0.59±0.27	0.60±0.23	-	0.03±0.02	0.11±0.04	-	-	11.7±4.43	12
"	blubber	1995	m	92.6±1.8	9	mean±SD	-	-	8.76±2.45	3.12±1.61	-	1.03±0.28	1.13±0.30	-	0.12±0.04	0.38±0.11	-	-	15.5±4.40	12
"	blubber	2001	m	93.1±3.5	11	mean±SD	-	-	8.77±4.78	1.29±0.88	-	1.71±0.99	1.48±0.90	-	0.36±0.19	0.63±0.40	-	-	15.5±8.52	12
Svalbard	blubber	1998	m,f	4-90	9	mean±SD	-	-	77.8±46.4	-	n.d.	5.15±2.78	-	n.d.	n.d.	-	-	-	92.9±56.5	13

Continued next page.

Annex Table 17 continued.

Species/Region	Tissue	Year	Sex	% lipid	n	Statistic	BDE15	BDE52	BDE47	BDE49	BDE85	BDE99	BDE100	BDE138	BDE153	BDE154	BDE183	BDE209	ΣPBDEs	Reference
Long-finned pilot whales																				
Faroe Islands, Denmark																				
Hvannasund	blubber	1994	f	82	9, pooled	–	–	–	412	–	–	164	–	–	32.0	–	–	–	691	13
Vestmanna	blubber	1996	m,f	66-79	4 pools (n=19,8,13,4)	range	–	–	896±471	–	–	305±148	–	–	46.8±18.6	–	–	–	1620±790	13,14
Torshavn	blubber	1997	m,f	52-85	12	range	–	–	289±192	–	n.d.	61.9±36.9	–	n.d.	5.50±3.05	–	–	–	647±410	13,14
Leynar	blubber	1997	m,f	83-86	4 pools (n=3,28,9,10)	range	–	–	232±80.1	–	n.d.	46.6±9.46	–	n.d.	3.91±0.72	–	–	–	522±210	13,14
Sandavagur	blubber	1997	m,f	64-80	4 pools (n=8,18,19,7)	range	–	–	358±142	–	n.d.	64.1±26.0	–	n.d.	3.12±1.20	–	–	–	770±310	13,14
Hvalvik	blubber	1998	m,f	66-83	4 pools (n=6,20,9,16)	range	–	–	188±48.2	–	n.d.	34.8±7.92	–	n.d.	4.58±1.49	–	–	–	554±147	13,14
Faroe Islands	blubber	2000	f,?	2-83	6	range	–	–	76.6±53.5	–	n.d.	13.2±10.7	–	n.d.	2.1±0.3	–	–	–	144±107	13
Polar bear																				
Svalbard	adipose	1998	m,f	5-67	20	range	–	–	17.5±9.75	–	n.d.	n.d.	–	n.d.	n.d.	–	–	–	17.5±9.75	13

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- Stern *et al.*, 2001a.
- Herzke *et al.*, 2001b.
- Schlabach *et al.*, 2001. ΣPBDEs = sum of BDE47 and 99.
- Herzke, 2002a; Concentrations converted from lipid weight to wet weight using mean % lipid content.
- Muir and Johansen, 2001; ΣPBDEs = sum of 42 congeners, including 209.
- Herzke, 2003.
- Braune *et al.* 2001a.
- Gabrielsen, 2002.
- Ikonomou *et al.* 2002.
- Muir *et al.*, 1999d; ΣPBDEs = sum of mono-, di-, tri- tetra-, penta-, hexa-, and hepta-brominated diphenyl ethers.
- Stern, 2001.
- van Bavel *et al.*, 2001; ΣPBDEs = sum of 21 tetra-, penta- and hexa-brominated diphenyl ethers (several unnamed, but includes 47, 99, 85, 153, 138).
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## Abbreviations and definitions of some terms

- 4-OH-HpCS . . . . . 4-Hydroxy-heptachlorostyrene.
- $\delta^{13}\text{C}$  . . . . .  $^{13}\text{C}/^{12}\text{C}$  stable isotope ratio.
- $\delta^{15}\text{N}$  . . . . .  $^{15}\text{N}/^{14}\text{N}$  stable isotope ratio (standardized against air).
- $\Sigma\text{CBz}$  . . . . . Sum of tetra-, penta- and hexachlorobenzene (concentrations).
- $\Sigma\text{CHL}$  . . . . . Sum of *cis*- and *trans*-chlordanes and *cis*- and *trans*-nonachlor (concentrations).
- $\Sigma\text{DDT}$  . . . . . Sum of DDT, DDD, and DDE (concentrations).
- $\Sigma\text{HCH}$  . . . . . Sum of (concentrations of)  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH isomers.
- $\Sigma\text{nPCB}$  . . . . . Sum of non-*ortho* PCBs (concentrations).
- $\Sigma\text{PCB}$  . . . . . Sum of a number of individual polychlorinated (PCB) congeners. A method of expressing the PCB content of a sample by measuring and summing the quantities of specific PCB congeners. The number and identification of the congeners determined varies from laboratory to laboratory and for different types of samples. It is generally given in explanatory notes. (See also Total PCB).
- $\Sigma\text{PBDE}$  . . . . . Sum of a number of individual polybrominated diphenyl ether (PBDE) congeners.
- $\Sigma\text{PCDD}$  . . . . . Sum of a number of polychlorinated dibenzo-p-dioxin (PCDD) congeners (concentrations).
- $\Sigma\text{PCDF}$  . . . . . Sum of a number of polychlorinated dibenzofuran (PCDF) congeners (concentrations).
- $\Sigma\text{TEQ}$  . . . . . Sum of TCDD equivalents (TEQs) from all dioxin-like compounds.
- a.s.l. . . . . Above sea level.
- ACAP . . . . . Arctic Council Action Plan to eliminate pollution of the Arctic.
- ADEC . . . . . Alaskan Department of Environmental Conservation.
- AE . . . . . Aldrin epoxidase.
- Aerosol . . . . . Dispersion of solid or liquid particles of microscopic size in gaseous media. The particles are so small that their fall speeds are small compared with the vertical component of the air motion. Haze and cloud are the commonest atmospheric aerosols, fall speeds being fractions of 1 cm per second.
- Ah receptor . . . . . Aryl hydrocarbon receptor, also known as the dioxin receptor.
- AHH . . . . . Aryl hydrocarbon hydroxylase.
- AMAP . . . . . Arctic Monitoring and Assessment Programme (Arctic Council Working Group).
- APND . . . . . Aminopyrine N-demethylase.
- Arctic Circle . . . . . Latitude of 66°32' N.
- Arctic haze . . . . . A condition in which visibility is reduced in horizontal and slant directions, but remains unimpeded vertically; encountered by aircraft over Arctic regions.
- Aroclor 1254 and 1260 . . . . . Technical PCB mixtures.
- Assessment . . . . . A (regional) assessment is both a *process* (i.e., the procedure by which the information is collected and evaluated) undertaken periodically to assess the state of knowledge, and a *product* (i.e., a document presenting and synthesizing the findings of the assessment and an action plan for future work).
- B[a]P . . . . . Benzo[a]pyrene (PAH).
- B[e]P . . . . . Benzo[e]pyrene (PAH).
- BDE . . . . . Brominated diphenyl ether. Typically used in association with an IUPAC number to identify a particular BDE congener (e.g. BDE47).
- Beaufort Gyre . . . . . Major current system in the Arctic Ocean: clockwise surface circulation in the Beaufort Sea.
- BGS . . . . . Brain growth spurt.
- Bioaccumulation . . . . . The net accumulation of POPs from all exposure routes, usually expressed as the concentration of a POP in an organism on a lipid weight basis divided by the concentration found in water (truly dissolved) or air (gas phase).
- Biomagnification . . . . . The increased accumulation of POPs with each trophic level in a food web, expressed as the concentrations in the organisms divided by the concentrations in its food, both on a lipid weight or organic carbon (sediments, soils) basis.
- BMF . . . . . Biomagnification factor.
- Carcinogenic . . . . . Cancer-causing.
- CB . . . . . Chlorobiphenyl. Typically used in association with an IUPAC number to identify a particular PCB congener (e.g. CB153).
- CBz . . . . . Chlorinated benzene.
- CFC . . . . . Chlorofluorocarbons.
- CHL . . . . . Chlordane.
- COE . . . . . (US) Corps of Engineers.
- Congener . . . . . An individual PCB, PBDE, PCDD, PCDF or toxaphene.
- Contamination . . . . . See pollution and contamination.
- Coplanar PCBs . . . . . PCB molecules that can take on a planar configuration and that are dioxin-like. These include non-*ortho* and mono-*ortho* PCBs.
- CP . . . . . Chlorinated paraffins.
- CYP . . . . . Cytochrome P450-dependent enzymes.
- d . . . . . Day.

- DBT . . . . . Dibutyltin.
- DBP . . . . . Dimethyl bipyroles.
- DDD . . . . . 1,1-dichloro-2,2-bis (4-chlorophenyl) ethane.
- DDE . . . . . 1,1-dichloro-2,2-bis (4-chlorophenyl) ethylene.
- DDT . . . . . Dichlorodiphenyltrichloroethane (an organochlorine pesticide). 1,1,1-trichloro-2,2-bis (4-chlorophenyl) ethane.
- DeBDE . . . . . Decabromodiphenyl ether.
- DHH . . . . . Dihydroheptachlor.
- DiBDE . . . . . Dibromodiphenyl ether.
- DiCN. . . . . Dichloronaphthalene.
- DMBA . . . . . 7,12-dimethylbenz[a]anthracene.
- DNA . . . . . Deoxyribonucleic acid.
- Dry deposition . . . . . Delivery of airborne contaminants to the surface by particle fallout, diffusion, and air movements (including irregular air movements in the turbulent boundary layer close to the ground and diffusion through the laminar surface layer).
- dw . . . . . Dry weight – basis of determination of concentration.
- EC<sub>50</sub> . . . . . Effective concentration causing a response in 50% of the treated organisms.
- EF . . . . . Enantiomeric fraction. This is defined as:  $A_+/A_+ + A_-$  and a racemic mixture has an EF of 0.5. See also ER.
- ER . . . . . Enantiomer ratio. This is defined as:  $A_+/A_-$  and a racemic mixture has an ER of 1. See also EF.
- ERL . . . . . Effects Range Low.
- ERM . . . . . Effects Range Median (or Effects Range Medium).
- EROD . . . . . Ethoxyresorufin-O-deethylase.
- Estrogenic . . . . . Estrogen-like.
- EU . . . . . European Union.
- Fetotoxic . . . . . Toxic to the fetus.
- Food web. . . . . A series of interconnected food chains.
- Fram Strait . . . . . Strait between Greenland and Svalbard; major exit route for water and ice exiting the Arctic Ocean.
- Freshet . . . . . The sudden overflowing of a river caused by heavy rain or melting snow, the latter more likely in an Arctic context.
- fSU . . . . . Former Soviet Union.
- FT3. . . . . Free triiodothyronine.
- FT4. . . . . Free thyroxine.
- FUDS. . . . . Formerly used defense sites.
- GJIC . . . . . Gap junctional intercellular communication.
- HLC . . . . . Henry's Law constant.
- h, hr . . . . . Hour.
- HBCD . . . . . Hexabromocyclododecane.
- HCB, HCBz . . . . . Hexachlorobenzene.
- HCBd . . . . . Hexachlorobutadiene.
- HCFC . . . . . Hydrochlorofluorocarbons.
- HCH . . . . . Hexachlorocyclohexane (organochlorine insecticides; including the  $\gamma$ -HCH isomer, lindane).
- HDBP. . . . . Halogenated dimethyl bipyroles.
- HELCOM . . . . . Helsinki Commission.
- HpBDE . . . . . Heptabromodiphenyl ether.
- HpCDD . . . . . Heptachlorodibenzo-*p*-dioxin.
- HpCDF. . . . . Heptachlorodibenzofuran.
- HpCN . . . . . Heptachloronaphthalene.
- HxBDE. . . . . Hexabromodiphenyl ether.
- HxCB. . . . . Hexachlorobiphenyl.
- HxCDD . . . . . Hexachlorodibenzo-*p*-dioxin.
- HxCDF. . . . . Hexachlorodibenzofuran.
- HxCN . . . . . Hexachloronaphthalene.
- IC<sub>50</sub> . . . . . The concentration at which 50% inhibition occurs.
- IJC . . . . . International Joint Commission.
- In ovo* . . . . . Experiments carried out in eggs.
- In vitro* . . . . . Experiments carried out 'in glass', i.e. outside of living organisms.
- In vivo* . . . . . Experiments carried out in living organisms.
- IPCS . . . . . (WHO) International Programme on Chemical Safety.
- IRP . . . . . Installation Restoration Programme.
- Isomer . . . . . Member of a homologue group. Compounds having the same 'skeleton' and same number of chlorine atoms, e.g., 1,2,7,8-TeCDF and 2, 3, 7, 8-TeCDF are isomers.
- ITEQ . . . . . International TCCD equivalents.
- IUPAC . . . . . International Union of Pure and Applied Chemistry.
- K<sub>ow</sub> . . . . . Partitioning coefficient between octanol and water.
- kt. . . . . Kilotonnes.
- LD<sub>50</sub> . . . . . Lethal dose that kills 50% of the organisms.
- Lipophilic. . . . . Affinity for lipid; lipophilic substances exhibit a preference to accumulate in fat and fatty tissues.
- LOAEC. . . . . Lowest-(observed)-adverse-effect-concentration.
- LOAEL. . . . . Lowest-(observed)-adverse-effect-level.
- LOEC . . . . . Lowest-(observed)-effect-concentration.
- LOEL. . . . . Lowest-(observed)-effect-level.
- LORAN . . . . . Long Range Aid to Navigation.
- LRTAP . . . . . (UN-ECE Convention on) Long-range Transboundary Air Pollution.
- lw. . . . . Lipid weight – basis of determination of concentration.
- m<sup>3</sup> . . . . . Cubic meter.
- MBT . . . . . Monobutyltin.
- MCCP . . . . . Medium-chain chlorinated paraffins.
- MeSO<sub>2</sub> . . . . . Methylsulfone.
- MFO . . . . . Mixed function oxidase.
- mo . . . . . Month.
- MoCN . . . . . Monochloronaphthalene.
- mol. . . . . Mole.
- Mono-*ortho* PCBs . . . . . PCB molecules with one chlorine in the *ortho* position; i.e. with 2,3,3',4,4' chlorine substitution

- (e.g. CBs 118 and 105). (See also coplanar PCBs.)
- mRNA . . . . . Messenger ribonucleic acid.
- MROD . . . . . Methoxy-*O*-resorufin deethylase.
- MS . . . . . Mass spectrometer.
- n . . . . . Number (of individuals in a sample, observations contributing to a mean, etc.).
- NAP . . . . . Naphthalene.
- NOAEC . . . . . No-(observed)-adverse-effects-concentration.
- NOAEL . . . . . No-(observed)-adverse-effects-level.
- NOEC . . . . . No-(observed)-effects-concentration.
- NOEL . . . . . No-(observed)-effects-level.
- NOW . . . . . Northwater Polynya study.
- nPCBs,  
non-*ortho* PCBs . . . . . PCB molecules with no chlorines in the *ortho* position; i.e., non-*ortho* substituted PCBs; PCB congeners with 3,3',4,4' chlorine substitution (e.g. CBs 77, 126 and 169). (See also coplanar PCBs.)
- NWT . . . . . Northwest Territories, Canada.
- OC . . . . . Organochlorine.
- OcBDE . . . . . Octabromodiphenyl ether.
- OCDD . . . . . Octachlorodibenzo-*p*-dioxin.
- OCDF . . . . . Octachlorodibenzofuran.
- OcN . . . . . Octachloronaphthalene.
- OCS . . . . . Octachlorostyrene.
- OH . . . . . Hydroxyl radical.
- Ortho* PCBs . . . . . PCB molecules with chlorine substituents in the 2 and 6 positions.
- OSPAR . . . . . Oslo and Paris Convention for the Protection of the Marine Environment of the North East Atlantic, 1992.
- OSPARCOM . . . . . Oslo and Paris Commissions.
- p . . . . . Probability.
- Pa . . . . . Pascals (unit of pressure).
- Parlar . . . . . An individual toxaphene congener.
- PAHs . . . . . Polyaromatic hydrocarbons (polycyclic aromatic hydrocarbons; polynuclear aromatic hydrocarbons). In a strict sense, a group of chemicals comprised of all aromatic hydrocarbon molecules containing three or more benzene rings, but often also used to include hydrocarbon molecules with two benzene rings.
- PBB . . . . . Polybrominated biphenyl.
- PBDEs . . . . . Polybrominated diphenyl ethers.
- PCA . . . . . Polychlorinated n-alkanes
- PCBs . . . . . Polychlorinated biphenyls. (See also CB).
- PCDDs . . . . . Polychlorinated dibenzo-*p*-dioxins.
- PCDE . . . . . Polychlorinated diphenyl ethers.
- PCDFs . . . . . Polychlorinated dibenzofurans.
- PCEs . . . . . Perchloroethylenes.
- PCNs . . . . . Polychlorinated naphthalenes.
- PCP . . . . . Pentachlorophenol.
- PeBDE . . . . . Pentabrominated diphenyl ethers.
- PeCA . . . . . Pentachloroanisole.
- PeCB . . . . . Pentachlorobiphenyl.
- PeCBz . . . . . Pentachlorobenzene.
- PeCDD . . . . . Pentachlorodibenzo-*p*-dioxin.
- PeCDF . . . . . Pentachlorodibenzofuran.
- PeCN . . . . . Pentachloronaphthalene.
- PFA . . . . . Perfluorinated acids.
- PFOA . . . . . Perfluorooctanoic acid.
- PFOS . . . . . Perfluorooctane sulfonate.
- PHA . . . . . Phytohemagglutinin.
- Planar PCBs . . . . . Collective name for non-*ortho* PCBs and mono-*ortho* PCBs.
- Plankton . . . . . Animals and plants, many of them microscopic, that float or swim very feebly in fresh- or salt water bodies. They are moved passively by winds, waves or currents.
- Poikilotherm . . . . . Cold-blooded animal.
- Pollution and  
contamination . . . . . Contamination implies the presence of a substance where it would normally not be found.
- Marine pollution is defined by GESAMP as: 'a harmful effect on marine life, human health, resources, amenities or other beneficial use of the sea caused, directly or indirectly, by substances and wastes (including energy) derived from human activity'. The GESAMP definition of (marine) pollution introduces the concepts of 'harmful effects' and 'human activity' in distinguishing the terms contamination and pollution.
- In AMAP's assessment, the GESAMP approach to defining 'pollution' as opposed to 'contamination' has been adopted for the most part. However, the use of the two terms in this report is not always entirely consistent in reflecting this. This is particularly so in, e.g., cases where the word pollution is in common usage; such as 'air pollution' or the use of the term 'persistent organic pollutants' rather than 'persistent organic contaminants'.
- Polynya . . . . . Open water regions, ranging in area up to thousands of square kilometers, that persist within closed sea-ice cover.
- POP . . . . . Persistent organic pollutant.
- PROD . . . . . Pentoxyresorufin-*O*-dealkylase.
- PUF . . . . . Polyurethane foam plug.
- PVC . . . . . Polyvinyl chloride plastic.
- QA . . . . . Quality assurance.
- QC . . . . . Quality control.
- $r^2$  . . . . . Coefficient of correlation.
- RAIPON . . . . . Russian Association of Indigenous Peoples of the North.
- RBP . . . . . Retinol-binding protein.
- RPSI . . . . . Relative penis size index.
- ROSHYDROMET . . . . . Federal Service of Russia on Hydro-meteorology and Environment.

- SCCP . . . . . Short-chain chlorinated paraffins.  
SD . . . . . Standard deviation.  
SE . . . . . Standard error.  
SHEBA . . . . . Surface Heat Budget of the Arctic Project.  
SOM . . . . . Soil organic matter.  
SPMD . . . . . Semipermeable membrane device.  
Subarctic  
(or forest tundra) Transition zone between the boreal forest and the treeless tundra.  
T3 . . . . . Triiodothyronine.  
T4 . . . . . Thyroxine.  
TAG . . . . . Triacylglycerols.  
TBT . . . . . Tributyltin.  
TBTO . . . . . Tributyltin oxide.  
TBBPA . . . . . Tetrabromobisphenol A.  
TCDD . . . . . 2,3,7,8-tetrachlorodibenzo-*p*-dioxin.  
TCPM-OH . . . . . tris(4-chlorophenyl)methanol.  
TCPMe . . . . . tris(4-chlorophenyl)methane.  
TeBDE . . . . . Tetrabrominated diphenyl ethers.  
TeCB . . . . . Tetrachlorobiphenyl.  
TeCBz . . . . . Tetrachlorobenzene.  
TeCDD . . . . . Tetrachlorodibenzo-*p*-dioxin.  
TeCDF . . . . . Tetrachlorodibenzofuran.  
TeCN . . . . . Tetrachloronaphthalene.  
TEF . . . . . Toxic equivalency factor.  
TEQ . . . . . TCDD equivalents.  
TNF- $\alpha$  . . . . . Tumor necrosis factor alpha.  
TNW . . . . . Tundra Northwest expedition.  
Total PCB . . . . . An (older) method for expressing the PCB content of a sample in which the sample is quantitated against a technical PCB product (such as Aroclor 1254) as the standard. The analysis is carried out using packed column chromatography. Also termed PCB on a formulation basis.  
Toxaphene . . . . . Polychlorobornanes and camphenes.  
TrCB . . . . . Trichlorobiphenyl.  
TrCN . . . . . Trichloronaphthalene.  
Transpolar Drift . . . . . Major current system in the Arctic Ocean: surface current traversing the Arctic Ocean from the Chukchi Sea to the North Greenland Sea. (See also Beaufort Gyre.)  
TriBDE/TrBDE . . . . . Tribromodiphenyl ether.  
TPhT . . . . . Triphenyltin.  
TSH . . . . . Thyroid-stimulating hormone.  
TTR . . . . . Transthyretin.  
TTR-RBP . . . . . Transthyretin-retinol-binding protein.  
UDPGT . . . . . Uridine diphosphate glucuronosyl transferase.  
UGMS . . . . . Russian regional administrations for hydrometeorology and monitoring of the environment.  
UK . . . . . United Kingdom.  
U.N. . . . . United Nations.  
UN ECE . . . . . United Nations Economic Commission for Europe.  
UNEP . . . . . United Nations Environment Programme.  
US, U.S., USA . . . . . United States (of America).  
USEPA . . . . . United States Environmental Protection Agency.  
UV . . . . . Ultraviolet radiation (100-400 nm).  
VDSI . . . . . Vas deferens sequence index. The VDSI scale ranges from 0 (normal) to 6 (last stages of development of vas deferens and penis in females leading to sterility).  
Wet deposition . . . . . Delivery of airborne contaminants to the surface as a result of their incorporation into rain or snow which subsequently falls to the ground.  
WHO . . . . . (UN) World Health Organization.  
wk . . . . . Week.  
ww . . . . . Wet weight – basis of determination of concentration.  
Xenobiotic . . . . . Chemical compound that is foreign to an organism; normally a synthetic chemical compound.  
yr . . . . . Year.
- Prefixes*  
Di . . . . . Di-  
Tr . . . . . Tri-  
Te . . . . . Tetra-  
Pe . . . . . Penta-  
Hx . . . . . Hexa-  
Hp . . . . . Hepta-  
Oc . . . . . Octa-