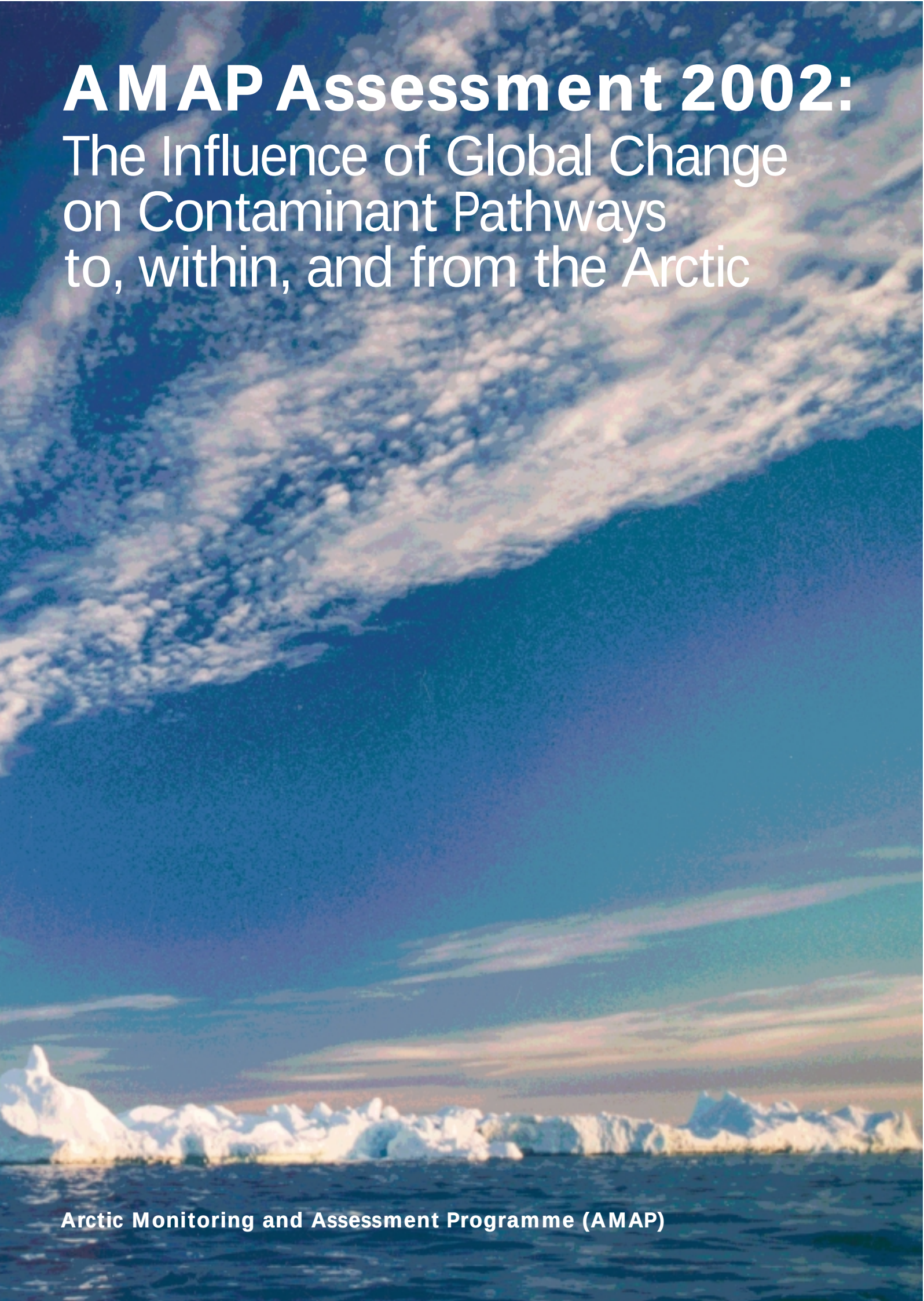
AMAP Assessment 2002:

The Influence of Global Change on Contaminant Pathways to, within, and from the Arctic



Arctic Monitoring and Assessment Programme (AMAP)

AMAP Assessment 2002:
The Influence of Global Change on Contaminant Pathways to, within, and from the Arctic

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Preface

This assessment report details the results of the 2002 AMAP assessment of the Influence of Global Change on Contaminant Pathways to, within, and from the Arctic. It builds upon the previous AMAP assessment of pathways of contaminants that was presented in 'AMAP Assessment Report: Arctic Pollution Issues' that was published in 1998. In covering issues relating to the influence of climate change on Arctic systems, this report also constitutes part of the AMAP input to the Arctic Climate Impact Assessment (ACIA) that is currently under preparation and due to be published in 2004.*

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.*

*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 (Persistent Organic Pollutants, Heavy Metals, Radioactivity, and Human Health), is reproduced in this report on pages vii to xi.

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 reponsibility of the scientific experts involved in the preparation of the assessment. The lead country for the AMAP Contaminant Pathways Assessment under AMAP Phase II was Canada. 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 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 author of this report, Robie Macdonald, and to his co-authors, and 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 iv of this report. Apologies, and no lesser thanks, are given to any individuals unintentionally omitted from the list.

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 for undertaking a lead role in supporting the Contaminant Pathways assessment.

The AMAP Working Group that was established to oversee this work, and the author of the AMAP contaminant pathways assessment are pleased to present their assessment.

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Oslo, April 2003

* 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.

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 negotation 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 toxicity 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

Predicting how climate change will alter contaminant transport in the global environment poses an exceptional challenge. It requires detailed knowledge of the physical and chemical properties of contaminants, subjects in which much progress has been made during the past decade. It also requires a profound understanding of environmental pathways and how they might respond to change forced by, for example, alteration of the atmosphere's greenhouse gas composition. This depth of understanding is currently lacking. It is clear that aerosols and greenhouse gases such as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) and chlorofluorocarbons (CFCs) are being released to the atmosphere by human activities (IPCC, 1990, 1995, 2002), that the world's hydrological cycle is being massively altered by damming for power and irrigation (Dynesius and Nilsson, 1994), and that humans are assaulting the global marine food web (Pauly *et al.*, 1998). Sufficient evidence has been assembled from paleorecords and much shorter instrumental observations to convince most – but not all – climate scientists that these types of disturbance will contribute to global change if they have not already done so (e.g., IPCC, 1990, 2002; Parsons, 1996; Showstack, 2001).

As a component of the cryosphere with a large seasonal amplitude in ice and snow cover, the Arctic is pivotal both as a region sensitive to change (sentinel) and as an exporter of change to other parts of the world (amplifier) (Aagaard and Carmack, 1989; Vörösmarty *et al.*, 2001; Walsh, 1995; Walsh and Crane, 1992). The 0°C isotherm is an especially important threshold of change because shifts between liquid and solid water have immense consequences for physical and biological systems and for humans.

Two major difficulties exist in understanding change in the Arctic and projecting its future. Firstly, recent trends are difficult to detect and comprehend due to short, sparse instrumental records. This is especially true of the Arctic where any climate trend must be discriminated from an enormous seasonal amplitude in weather, and natural variation at time scales from annual to five years to centuries and longer (Fischer *et al.*, 1998; McGhee, 1996; Polyakov and Johnson, 2000; Proshutinsky and Johnson, 1997; Stirling *et al.*, 1999; Tremblay *et al.*, 1997; Vanegas and Mysak, 2000; Wang and Ikeda, 2001). Secondly, the understanding of environmental processes in the Arctic is not sufficient to establish, with confidence, the link between primary changes (sea-level air pressure, air temperature, ice cover) and those of higher complexity but of much greater significance (e.g., thermohaline circulation, ecological structure and function, the hydrological cycle). These difficulties currently form an insurmountable hurdle to making reliable projections of how, exactly, exposure of Arctic biota to contaminants will be affected by global change. Nevertheless, sufficient evidence has accumulated during the past decade to demonstrate that contaminant pathways can and will continue to change. Experience strongly suggests that the environment is likely to deliver surprises (Macdonald *et al.*, 2000b), one of these being the abruptness of change – something for which the Arctic has recently shown an unexpected predisposition (Alley *et al.*, 2002; Dickson, 1999; Macdonald, 1996; Mysak, 2001; Rothrock *et al.*, 1999). Other surprises lurk in the sometimes subtle and non-intuitive connections between global and regional pathways that put the Arctic at risk from contaminants in the first place (Figure 1·1; AMAP, 1998; Macdonald *et al.*, 2000a,b).

Figure 1·1. The major physical pathways (wind, rivers and ocean currents) that transport contaminants to the Arctic.

To project how global change may alter contaminant exposure in the Arctic, this report builds on the foundation developed in previous assessments (AMAP, 1998; Jensen *et al.*, 1997), particularly those components addressing pathways (Barrie *et al.*, 1998; Macdonald *et al.*, 2000a). This makes it possible to consider the implications of model predictions that the world will experience a mean global air temperature rise of 3 to 5°C during the coming century (IPCC, 2002). Increased temperature

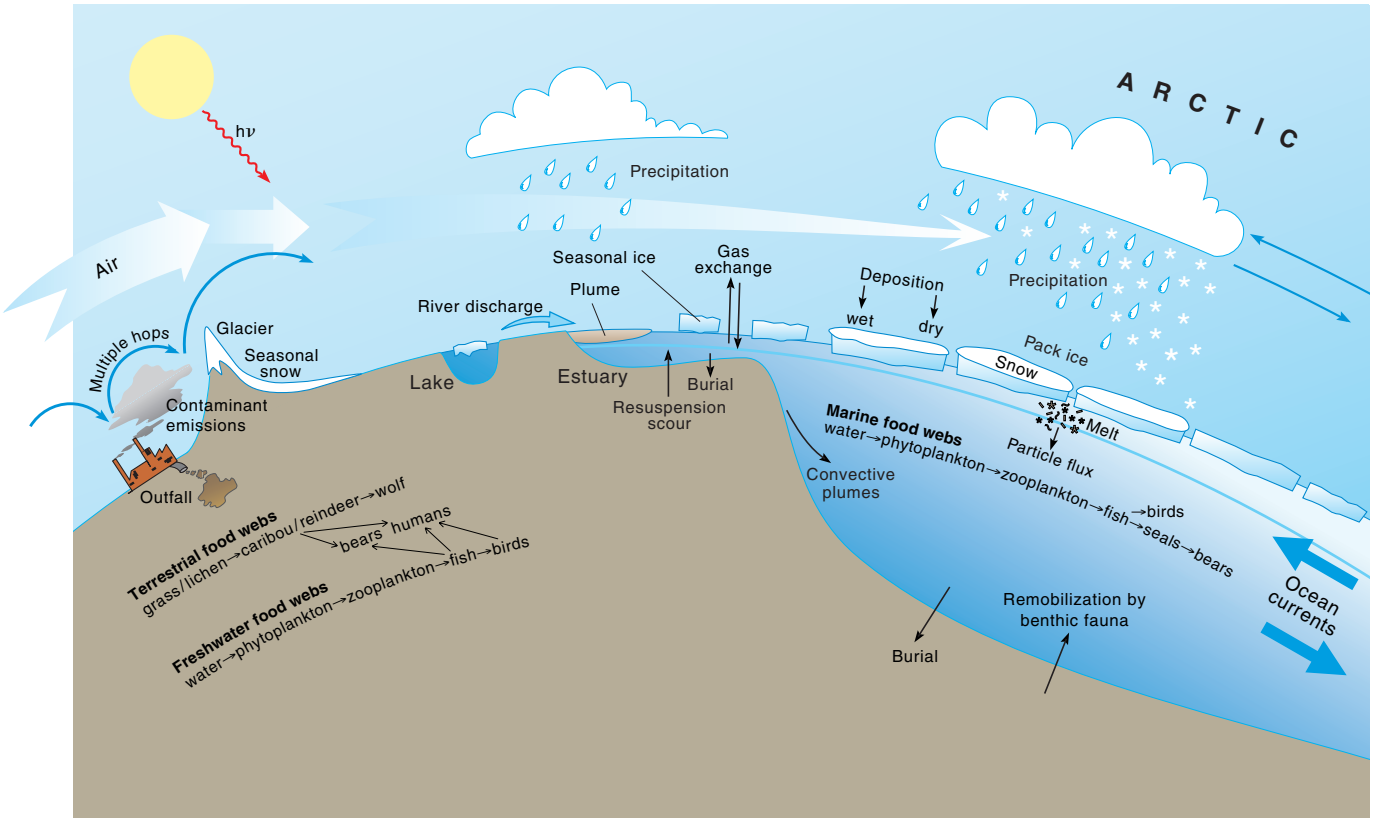

Figure 1·2. A simplified schematic diagram showing how physical pathways deliver contaminants emitted from northern industrial regions to the Arctic where they may be concentrated in biota or removed through degradation and burial (after Stewart, 2000; Muir *et al.*, 1999).

will have direct effects on contaminants (e.g., enhanced volatility, more rapid degradation, altered partitioning between phases) and on the environment (e.g., loss of permafrost, change in the seasonal cycle of snow or ice). However, a general air-temperature projection such as this is an insufficient basis for addressing the more important changes that will inevitably occur in the connections between the Arctic and the regions to the south and in the detail of pathways within the Arctic. Within the context of Arctic ecosystems and the humans who depend on them, it will not be temperature rise that is most important but, rather, the consequent change in the timing of seasons. During a contaminant's voyage to the Arctic, it may spend varying proportions of time in air, soil, water, ice, and food webs or it may become degraded (Figure 1·2). Each step along the path and every point of transfer can be altered by global change, which for a contaminant may mean dilution, concentration, transformation, bifurcation, shortcut, or delay.

1.1. Objectives of this report

This synthesis focuses on the question of how global change might alter contaminant pathways to and within the Arctic. As such, this is neither a review of change nor is it a review of contaminant pathways or of newly emerging contaminants. These topics have been, or are being, thoroughly reviewed elsewhere (AMAP, 1998; IPCC, 1995, 2002; Macdonald *et al.*, 2000a; Ruddiman, 2000). Rather, this report seeks to highlight observations and projections of global change that appear most likely to play a significant role in the life history of contaminants headed for an Arctic destination.

1.2. Organization of this report

Change in *physical pathways* is the first subject discussed, with the emphasis on observations during the past decade. Recent dramatic changes, many of which can be related directly to variation in atmospheric pressure fields (popularly referred to as the Arctic Oscillation (AO) (Wallace and Thompson, 2002)), include changes in winds, sea-ice drift and cover, ocean currents, precipitation, and other environmental pathway components (Morison *et al.*, 2000; Serreze *et al.*, 2000). The contrast between high (positive) and low (negative) AO indices allows authoritative discussions on some of the ways in which the Arctic actually has changed. Secondly, consideration is given to the likely consequences of these physical changes for *biological pathways*, noting that the organic carbon cycle and the food web are crucial pathway components for many contaminants (e.g., mercury, cadmium, and most organochlorine compounds). Brief consideration is then given to how *human responses* to global change are likely to alter contaminant pathways in the Arctic. The detailed review of physical and biological pathway changes that have occurred, or are likely to occur, then enables a discussion of the consequences for each category of contaminant of concern to AMAP (i.e., metals, persistent organic pollutants (POPs), radionuclides, and hydrocarbons). Lastly, the difficulty of interpreting time-series data is discussed, within the context that contaminant trends observed in sediment, water, air, and biota collected from the Arctic may include aspects related to changing pathways as well as aspects related to changes in contaminant emissions.

Chapter 2

Long-term Change in the Arctic

2.1. The distant past, and recorded history

During the last 400 000 years, the Earth has experienced four ice ages which have left records in glacial ice accumulating in Antarctica (Petit *et al.*, 1999) and in Greenland (Dansgaard *et al.*, 1993; Sowers and Bender, 1995). The overall surface air-temperature change between glacial and interglacial periods is thought to have been about 12°C, but perhaps more significant than temperature were the accompanying changes in continental ice masses, sea-ice climate and global ecosystems. In particular, sea-ice cover has proven to be a master variable in the equation of change. During the last glacial maximum, sea ice was locked within the Arctic and seasonal or perennial sea ice extended well south into the North Atlantic Ocean (Darby *et al.*, 1997; de Vernal *et al.*, 1993). The change from glacial to de-glacial to interglacial can be seen widely in Arctic sediments, both in terms of sedimentation rate and in the amounts and sources of organic material derived from marine primary production or land vegetation (see for example, Darby *et al.*, 2001; Nørgaard-Petersen *et al.*, 1998; Phillips and Grantz, 1997; Stein *et al.*, 1994, 2001).

Sea level dropped by about 120 m during the last glacial maximum (Fairbanks, 1989). This exposed much of the Arctic Ocean's enormous continental shelves, forcing rivers to cut channels across them to enter the interior sea directly, and severing the connection between the Arctic and Pacific Oceans. With sea-level rise, about 15 000 years ago the Bering land bridge was flooded (Hopkins, 1979) and then gradually submerged (Dyke *et al.*, 1996b) allowing the Pacific Ocean access to the Arctic Ocean. This sequence of events together with inundation of the continental shelves must have had enormous consequences for the oceanography and regional biogeography of the western Arctic and the Canadian Arctic Archipelago (Dunton, 1992; Dyke *et al.*, 1996a,b; Hequette *et al.*, 1995).

Although the climate has been described as 'exceptionally stable' during the past 10 000 years (Dansgaard *et al.*, 1993), it has actually continued to undergo substantial fluctuations. Indeed, it seems that very small shifts in temperature, perhaps of only a degree or two, account for the so-called Medieval Warm Period (1100-1400 AD) and subsequent Little Ice Age (1450-1850 AD) (for the relevance of these terms see Bradley and Jones, 1993; Crowley and Lowery, 2000). Both of these minor and sporadic deviations in the temperature record had dramatic consequences for humans – especially those living on the margins of northern oceans (Alley *et al.*, 2002; McGhee, 1996; Ogilvie and Junsson, 2000). During the past two centuries, small changes in ice and watermass distribution have continued to have an impact on humans and ecosystems, sometimes leading to migration or abandonment of locations, but certainly requiring adaptation (Miller *et al.*, 2001; Vibe, 1967).

For most of the past 10 000 years (the Holocene), climate change was not accompanied by the added complexity of anthropogenic pollutants. However, over the past two millennia and especially during the past two centuries, Arctic glacial ice has recorded the transient rise in the levels of virtually every contaminant emitted by human activities (Boutron *et al.*, 1995, 1998; Gregor *et al.*, 1995; Hong *et al.*, 1994; Masclet and Hoyau, 1994; Rosman *et al.*, 1997). These include the greenhouse gasses (GHGs) that force atmospheric temperature change (Petit *et al.*, 1999), and it is the dramatic rise in the levels of GHGs during the past several decades that make future projections based on past climates subject to such uncertainty.

2.2. The present and future

The twentieth century has been the warmest in the Arctic for the past 400 years (Overpeck *et al.*, 1997). The Intergovernmental Panel on Climate Change (IPCC) suggests that over the past century the global mean surface temperature has increased by about 0.3 to 0.6°C, mostly attributable to human activities, and will probably further increase by 1.4 to 5.8°C between 1990 and 2100 (Houghton *et al.*, 1995; IPCC, 1995, 2002; Showstack, 2001). According to models, warming will be more pronounced in polar regions (Figure 2·1); perhaps

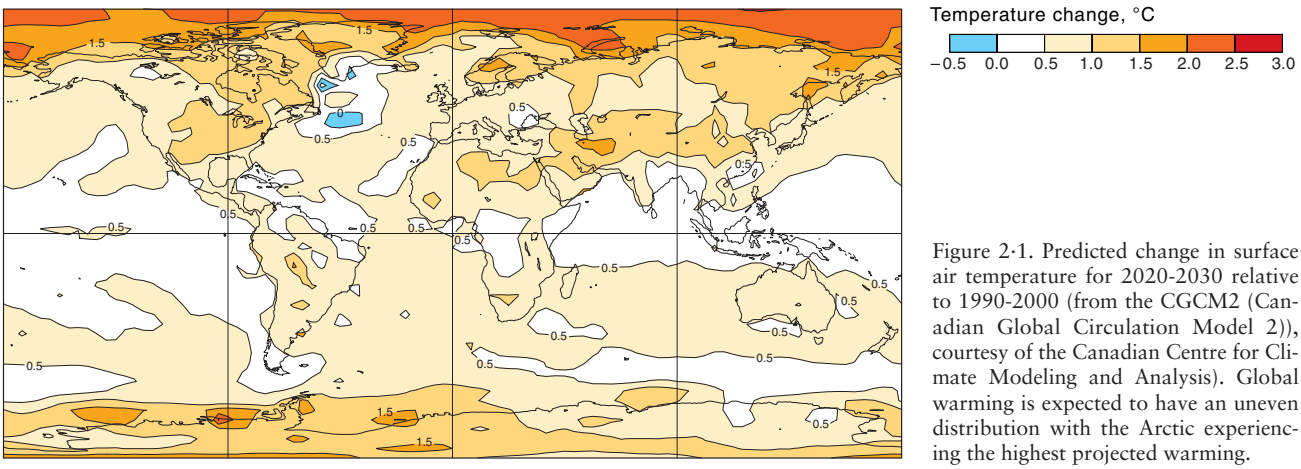

Figure 2·1. Predicted change in surface air temperature for 2020-2030 relative to 1990-2000 (from the CGCM2 (Canadian Global Circulation Model 2)), courtesy of the Canadian Centre for Climate Modeling and Analysis). Global warming is expected to have an uneven distribution with the Arctic experiencing the highest projected warming.

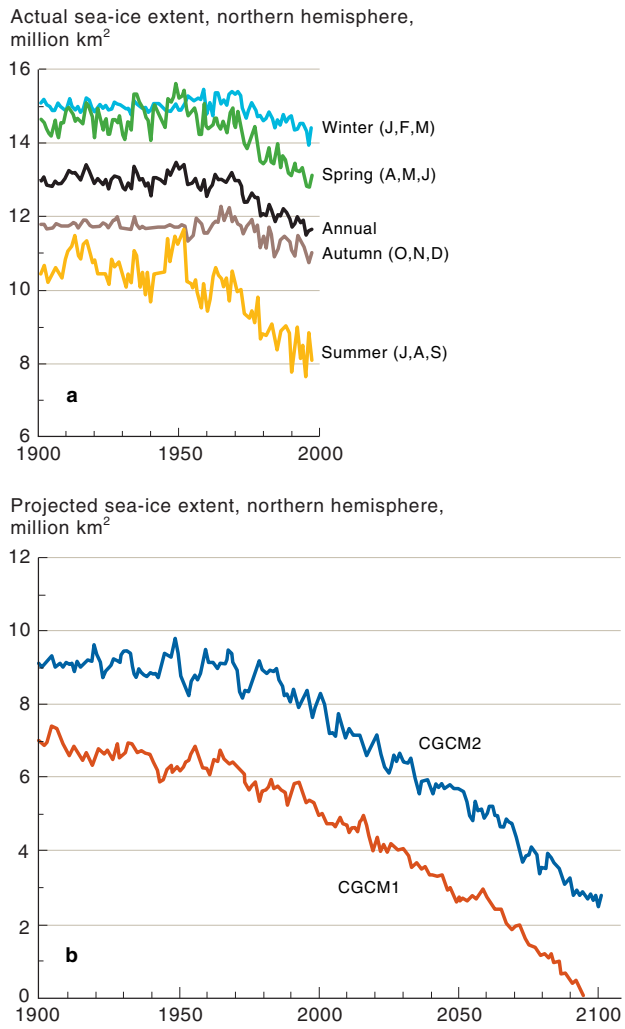

Figure 2·2. Sea-ice extent in the Northern Hemisphere. This figure illustrates a) time series of 'actual' annual and seasonal sea-ice extent between 1990 and 2000 derived from long-term observations and satellite images (adapted from Walsh and Chapman, 2000) and b) simulations of annual mean sea-ice extent from the models CGCM1 and CGCM2 (Canadian Global Circulation Models 1 and 2 of the Canadian Centre for Climate Modeling and Analysis), where the latter differs from the former in mixing parameterization. (After Flato and Boer, 2001.)

5°C or more near the pole and 2 to 3°C around the margins of the Arctic Ocean, with a decreasing temperature contrast between poles and the equator (Manabe *et al.*, 1992; Mitchell *et al.*, 1995; Zwiers, 2002). For gases emitted to the atmosphere, climate warming will increase inter-hemispheric exchange times, mixing times, and mean transit times by perhaps 10% (Holzer and Boer, 2001). Furthermore, the greatest warming will occur in the autumn–winter period due to delay in the onset of sea-ice cover (Manabe *et al.*, 1992; Serreze *et al.*, 2000). Continental interiors will become dryer and sea level will continue to rise, perhaps by a further 50 cm in addition to the estimated rise of 10 to 25 cm during the past century (Proshutinsky *et al.*, 2001; Serreze *et al.*, 2000).

Models predict that after about 80 years of atmospheric CO_2 increase at 1% per year, precipitation will increase within the Arctic and subpolar regions to 0.5 to 1 m/yr, which would more than double the current moisture flux convergence north of 70°N estimated at about 20 cm/yr (Manabe *et al.*, 1992; Walsh, 2000), making the Arctic a considerably 'wetter' place. Over the past four decades sea-ice extent in the Arctic Ocean has decreased in summer by as much as 25%. By the end of the twenty-first century, GHG forcing might produce an Arctic Ocean seasonally clear of ice (Figure 2·2, Flato and Boer, 2001).

Simulations based on GHG forcing predict that mean annual river discharge will increase by about 20% for the Yenisey, Lena and Mackenzie Rivers, but decrease by 12% for the Ob (Arora and Boer, 2001; Miller and Russell, 1992). Furthermore, the projection that high-latitude rivers will undergo marked changes in amplitude and seasonality of flow due to decreased snowfall and earlier spring melt (Arora and Boer, 2001) may already have some support in observations (Lammers *et al.*, 2001).

The coupling of the runoff cycle with northern lake hydrology is probably one of the points most sensitive to climate change (see for example, Vörösmarty *et al.*, 2001) but the understanding of the processes involved is not yet sufficient to make confident projections. If Arctic lakes become more 'temperate' in character, productivity is likely to be enhanced due to less ice cover and more mixing, and there will be greater opportunity for runoff to mix into the lake during freshet, further supporting a more vigorous aquatic food web.

With these primary changes, permafrost melting can be expected to accelerate, disrupting vegetation and enhancing nutrient, organic carbon and sediment loading of rivers and lakes (Vörösmarty *et al.*, 2001). The loss of sea ice in the marginal seas, together with sea-level rise will promote further erosion of poorly bonded, low-gradient coasts, particularly during the period of autumn storms.

Chapter 3

Recent Change in the Arctic and the Arctic Oscillation

3.1. The Arctic Oscillation

During the 1990s, a quiet revolution took place in the perception of the Arctic (Carmack *et al.*, 1997; Dickson, 1999; Johannessen *et al.*, 1995; Johnson and Polyakov, 2001; Kerr, 1999; Levi, 2000; Macdonald, 1996; Macdonald *et al.*, 1999a; Maslanik *et al.*, 1996; Maslowski *et al.*, 2000; McPhee *et al.*, 1998; Morison *et al.*, 1998, 2000; Parkinson *et al.*, 1999; Polyakov and Johnson, 2000; Quadfasel *et al.*, 1991; Smith, 1998; Steele and Boyd, 1998; Vanegas and Mysak, 2000; Vörösmarty *et al.*, 2001; Walsh, 1991; Welch, 1998; Weller and Lange, 1999). Despite early evidence of cyclical change in northern biological populations and ice conditions (e.g., Bockstoce, 1986; Gudkovich, 1961; Vibe, 1967), the general view among many western physical scientists throughout the 1960s to 1980s was that the Arctic was a relatively stable place (Macdonald, 1996). This view has been replaced by one of an Arctic where major shifts can occur in a very short time, forced primarily by natural variation in the atmospheric pressure field associated with the Northern-hemisphere Annual Mode.

The Northern-hemisphere Annual Mode, popularly referred to as the Arctic Oscillation (AO) (Wallace and Thompson, 2002), is a robust pattern in the surface manifestation of the strength of the polar vortex (for a very readable description, see Hodges, 2000). The AO correlates strongly (85-95%) with the more commonly used indicator of large-scale wind forcing, the North Atlantic Oscillation (NAO) (the NAO is the normalized gradient in sea-level air pressure between Iceland and the Azores – see for example, Deser, 2000; Dickson *et al.*, 2000; Hurrell, 1995; Serreze *et al.*, 2000). In this report the AO and NAO are used more or less interchangeably because they carry much the same information. It is recognized, however, that in both cases the term 'oscillation' is rather misleading because neither index exhibits quasi-periodic behaviour (Wallace and Thompson, 2002). The AO captures more of the hemispheric variability than does the NAO which is important because many of the recent changes associated with the AO have occurred in the Laptev, East Siberian, Chukchi and Beaufort Seas – a long way from the NAO's center of action (Thompson and Wallace, 1998). Furthermore, the Bering Sea and the Mackenzie Basin are both influenced to some degree by atmospheric processes in the North Pacific (e.g., the Pacific Decadal Oscillation; see also Bjornsson *et al.*, 1995; Niebauer and Day, 1989; Stabeno and Overland, 2001), whereas the Baffin Bay ice climate appears to have an association with the Southern Oscillation (Newell, 1996), and the Canadian Arctic Archipelago and Hudson Bay probably respond to various atmospheric forcings as yet not fully understood.

Although the AO is an important component of change in Arctic climate, it accounts for only 20% of the variance in the atmospheric pressure field and many other factors can determine the atmospheric forcing. Because the AO is usually 'smoothed', it does not adequately represent events and short-term variations which are known to be important for the delivery of contaminants to the Arctic and possibly also locally important in forcing ice and surface water motion (see for example, Sherrell *et al.*, 2000; Welch *et al.*, 1991). It is important to note that one of the projections of climate change is that cyclonic activity will increase; extreme events may therefore become a prominent component of atmospheric transport in the coming century. The earliest significant rain event on record (May 26, 1994), which was observed widely throughout the Canadian Arctic Archipelago may have represented an example of this (see also Graham and Diaz, 2001; Lambert, 1995).

Around 1988 to 1989, the AO entered a positive phase of unprecedented strength (Figures 3·1 a and c, next page). The sea-level pressure (SLP) distribution pattern of the AO for winter and summer (Figures 3·1 b and d) shows that this positive shift in AO is characterized by lower than average SLP distributed somewhat symmetrically over the pole (the blue-isoline region on Figures 3·1 b and d) and higher SLP over the North Atlantic and North Pacific in winter and over Siberia and Europe in summer. As might be expected from examination of the AO SLP pattern (Figures 3·1 b and d), when the AO index is strongly positive conditions become more 'cyclonic' – i.e., atmospheric circulation becomes more strongly counterclockwise (Proshutinsky and Johnson, 1997; Serreze *et al.*, 2000).

In discussing change it is important to distinguish between variability, which can occur at a variety of time scales (Fischer *et al.*, 1998; Polyakov and Johnson, 2000) and trends caused, for example, by GHG warming. It has been suggested that locking the AO into a positive mode might actually be one way that a trend forced by GHGs can manifest itself in the Arctic (Shindell *et al.*, 1999). Others, however, consider that the extraordinary conditions of the 1990s were produced naturally by a reinforcing of short (5-7 yr) and long (50-80 yr) time-scale components of SLP variation (Polyakov and Johnson, 2000; Wang and Ikeda, 2001), and that GHG forcing will affect the mean property fields rather than alter the AO itself (Fyfe, 2003; Fyfe *et al.*, 1999). Longer records of the NAO index (Figure 3·16 a, page 18) indeed suggest that there have been other periods of high AO index during the past 150 years (e.g., 1900-1914), but none as strong as that experienced during the early 1990s. Recent data suggest that the AO index has decreased and that the Arctic system has to some degree begun to return to 'normal conditions' (Björk *et al.*, 2002; Boyd *et al.*, 2002; Johnson *et al.*, 1999).

The contrast in conditions between the Arctic in the 1960s, 1970s and 1980s (with a generally low/negative AO index) and the Arctic in the early 1990s (with an exceptionally high/positive AO index) provides an extraordinary opportunity to investigate how the Arctic might respond to climate change. Similarity between climate-

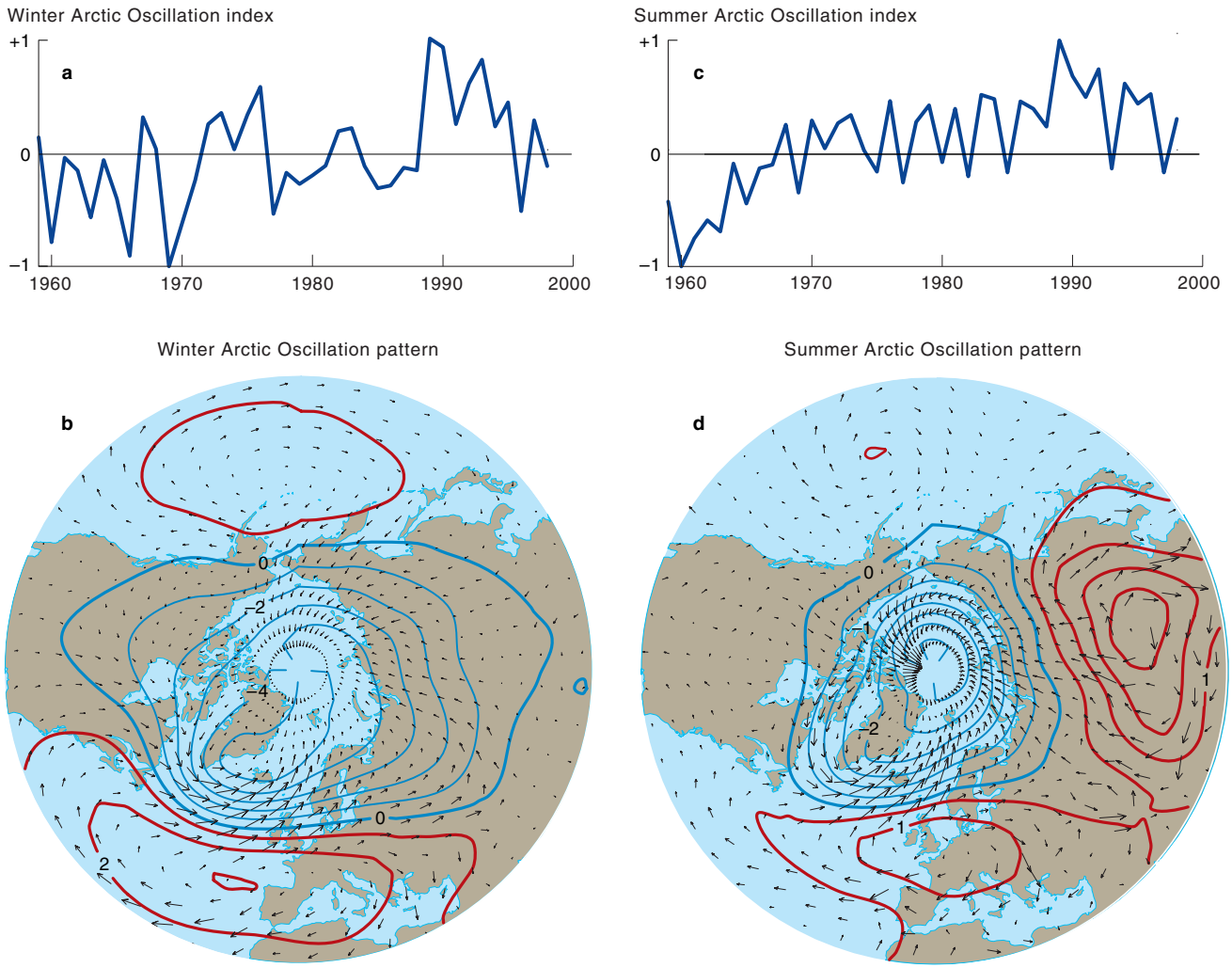

Figure 3·1. The Arctic Oscillation. The figure illustrates a) variability in the AO index between 1958 and 1998 in winter; b) the associated winter sea-level pressure pattern; c) variability in the AO index between 1958 and 1998 in summer; and d) the associated summer sea-level pressure pattern. To recreate atmospheric pressure patterns during the period in question, the winter (b) and summer (d) AO patterns would be multiplied by their respective indices (a, c) and added to the mean pressure field. Thus the high winter AO index in the early 1990s implies anomalously low pressure over the pole in the pattern shown in (b). Small arrows show the geostrophic wind field associated with the AO pattern with longer arrows implying stronger winds.

change projections and AO-induced change suggests that examining the differences between AO^- and AO^+ states should provide insight into the likely effects of climate change forced by GHG emissions. Variation in SLP, as reflected by the AO index, demonstrates that the Arctic exhibits at least two modes of behaviour (Morison *et al.*, 2000; Proshutinsky and Johnson, 1997) and that these modes cascade from SLP into wind fields, ice drift patterns, watermass distributions, ice cover and probably many other environmental parameters.

The Arctic is to a large degree constrained by overarching structures and processes in how it can respond to change. As illustrated in previous assessments, the Arctic Ocean is, and will remain, a 'mediterranean' sea, much influenced by land–ocean interaction and with restricted exchange with other oceans (Figure 1·1). Topography, bathymetry and global distribution of salinity in the ocean, require that water from the Pacific Ocean will predominantly flow *in* to the Arctic and the shallow sill at Bering Strait (50 m) guarantees that only surface water will be involved in this exchange. Pacific water will remain above Atlantic Layer water which is denser. Deep-basin water communicates predominantly with the Atlantic Ocean through the deep connection at Fram Strait. Ocean circulation within the Arctic is tightly tied to bathymetry through topographic steering of currents (Rudels *et al.*, 1994). Considering these kinds of constraints, rapid change can occur in ocean-current pathways or in the source or properties of the water carried by currents when, for example, fronts shift from one bathymetric feature to another (McLaughlin *et al.*, 1996; Morison *et al.*, 2000), when a given current strengthens or weakens (Dickson *et al.*, 2000), when source-water composition alters (Smith *et al.*, 1998; Swift *et al.*, 1997), or when relative strength of outflow varies between the Canadian Arctic Archipelago and Fram Strait (Macdonald, 1996), but not by reversal of flow in boundary currents or reversal of mean flow in the Bering Strait or out through the Archipelago.

Change associated with the Northern-hemisphere Annual Mode requires that consideration be given to large-scale variability in the Arctic; the fact that physical pathways can change rapidly needs to be recognized in greater detail, and the potential effects of GHG emissions against this naturally variable background should be assessed.

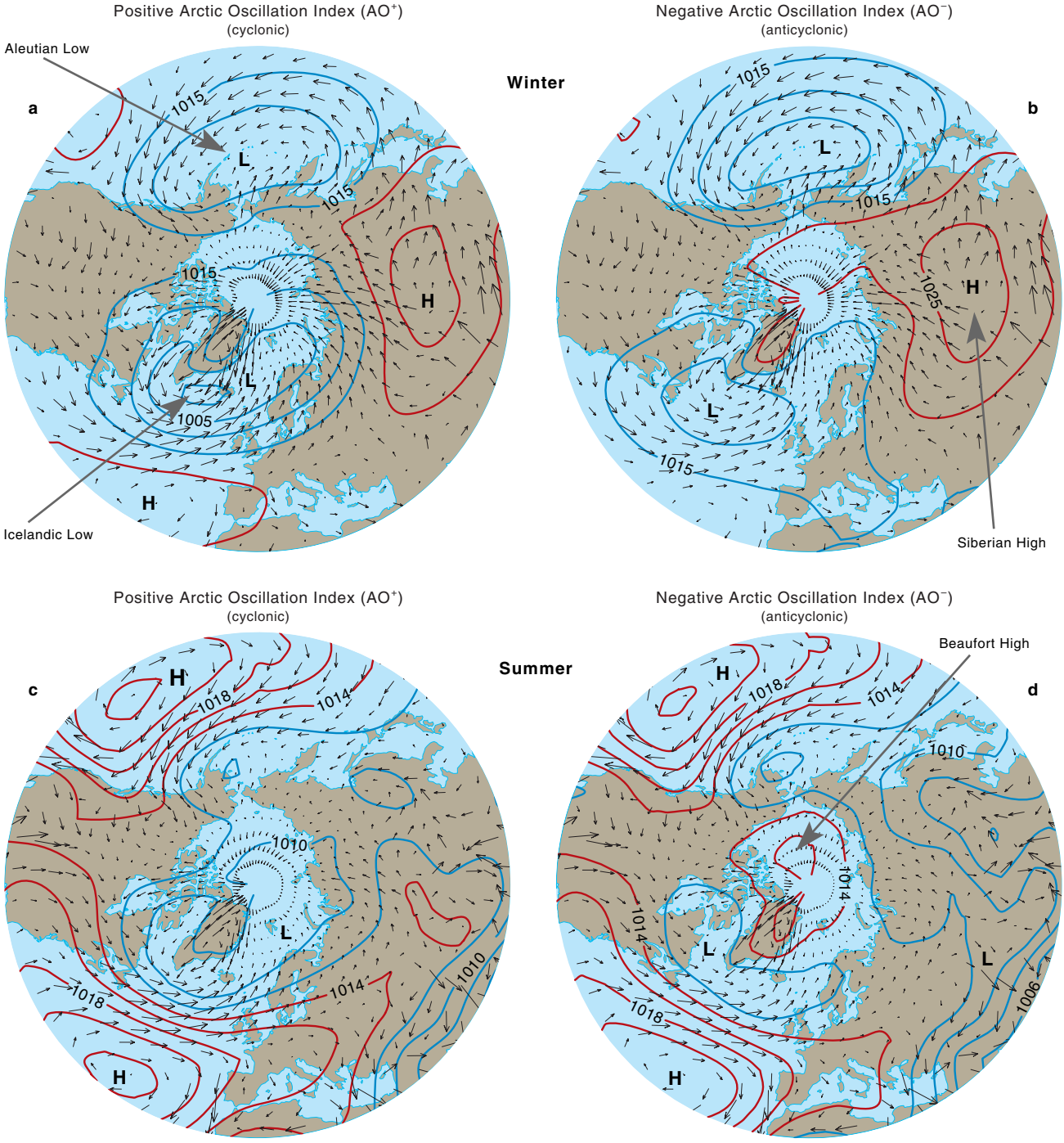

Figure 3·2. Atmospheric pressure fields and wind stream lines in the Northern Hemisphere. The figure illustrates a) strong AO+ conditions in winter; b) strong AO− conditions in winter; c) strong AO+ conditions in summer; and d) strong AO− conditions in summer.

3.2. Winds

Winds transport contaminants directly to the Arctic by delivering volatile and semi-volatile chemicals, and chemicals attached to fine particulates, from the south in timescales as short as a few days (Bailey *et al.*, 2000; Barrie *et al.*, 1998; Halsall *et al.*, 1998; Hung *et al.*, 2001; Stern *et al.*, 1997). Over the longer term, spanning months to years, winds deliver volatile and semi-volatile contaminants through a series of hops, as airborne chemicals become deposited onto surfaces (water, soil or vegetation) and then re-volatilized during, for example, summer warming. Winds also provide the primary forcing for ice drift and surface ocean currents (Mysak, 2001; Proshutinsky and Johnson, 1997) thereby indirectly affecting transport by these two media as well.

To understand how swings in the AO can affect atmospheric circulation, AO+ and AO− wind field/SLP maps have been constructed for winter (Figures 3·2 a and b) and summer (Figures 3·2 c and d) by adding (AO+) or subtracting (AO−) the patterns in Figures 3·1 b and d to/from the mean pattern for the period of record in the time series (1958-1998). The changes discussed in the following paragraphs can be considered generally as representing the difference between conditions during the 1960s to 1970s (low/negative AO index) and during the early 1990s (high/positive AO index) (see for example, Proshutinsky and Johnson, 1997).

In winter (Figures 3·2 a and b), the lower tropospheric circulation is dominated by high pressure over the continents and low pressure over the northern Pacific Ocean (Aleutian Low) and Atlantic Ocean (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 to the High Arctic. 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 second conduit by which airborne contaminants from eastern North America and Europe can rapidly reach the Arctic. 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%), over Eastern Europe/Siberia (15%), and over the Bering Sea (25%) account for about 80% of the annual south-to-north air transport (Iversen, 1996).

With a higher AO index (Figure 3·2 a), the Icelandic Low intensifies and extends farther into the Arctic across the Barents Sea and into the Kara and Laptev Seas (Johnson *et al.*, 1999). This has the effect of increasing the wind transport east across the North Atlantic, across southern Europe and up into the Norwegian Sea. During high NAO winters, westerlies onto Europe may be as much as 8 m/s (~700 km/day) stronger (Hurrell, 1995). At the same time, strong northerly winds are to be found over the Labrador Sea (Mysak, 2001).

The extension of the Icelandic Low into the Arctic also implies an effect of the AO on storm tracks. During the strong AO^+ conditions of the early 1990s, there was a remarkable increase in the incidence of deep storms, to around 15 per winter, and these storms penetrated farther into the Arctic (Dickson *et al.*, 2000; Maslanik *et al.*, 1996; Semiletov *et al.*, 2000). Increased cyclone activity increases poleward transport of heat and other properties carried by the air masses involved. Anomalous southerly airflow over the Nordic Seas enhances the connection between industrial regions of North America and Europe and the Arctic. At the same time, increased cyclones enhance transfer of contaminants from the atmosphere to the surface as a consequence of increased precipitation. Deep within the Arctic, the high SLP ridge that extends across Canada Basin during AO^- conditions (the Beaufort High), disappears and withdraws toward Russia (Johnson *et al.*, 1999; Morison *et al.*, 2000). It is worth noting that the Pacific mean atmospheric pressure field and wind patterns appear to change little between strong positive and strong negative phases of the AO in winter. Penetration of air from the Pacific into the Arctic is hindered by the mountain barrier along the west coast of North America where intensive precipitation also provides a mechanism to transfer contaminants and aerosols to the surface (Figures 3·2 a and b).

Summer pressure fields and air-flow patterns are markedly different from those of winter (compare Figures 3·2 a, b, c, and d). 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 (Figures 3·2 c and d). 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%)). The streamlines show that winds provide a means to transport contaminants from industrialized North America and Europe to the North Atlantic but penetration into the Arctic weakens. In the North Pacific, there remain atmospheric pathways to move air masses into the Gulf of Alaska from the east coast of Asia (Figures 3·2 c and d). During AO^+ conditions in particular, the Beaufort High weakens or disappears (Figures 3·2 a and c), altering mean wind fields.

3.3. Surface air temperature

A strong trend of warming has been observed in the Arctic for the period from 1961 to 1990 (Figure 3·3 a). This warming, which has been especially evident over northwestern North America and Siberia, has been accompanied by cooling in northeastern Canada, Baffin Bay, and West Greenland. An almost identical pattern of warming to that shown in Figure 3·3 a is produced by taking the difference between mean surface air temperatures during periods of AO^+ and AO^- conditions (Wallace and Thompson, 2002). Due to an extensive temperature record collected from drifting buoys, manned drifting stations, and land stations, direct relationships can be drawn between air surface temperature over sea and land in the Arctic and the changes in pressure field discussed in section 3.2. Over the period 1979 to 1997, a trend of +1°C per decade was found for winter surface air temperature (SAT) in the eastern Arctic Ocean, offset by a trend of –1°C per decade in the western Arctic Ocean (Rigor *et al.*, 2000). However, in spring almost the entire Arctic Ocean shows significant warming – as much as 2°C per decade in the eastern Arctic where a trend toward lengthened melt season was also observed. The trend in increasing SAT over the ocean is matched by temperature increases over Arctic land masses of 2°C per decade during winter and spring and annual increases of perhaps 0.8°C per decade (Figure 3·3 a). Records of annual temperature anomalies since 1900 (Figure 3·3 b) clearly show the warming trend since the 1970s, but note also that a similar episode of warming occurred in the 1930s to 1940s. Taken together, the trends in SAT over the central Arctic Ocean suggest that warming has occurred predominantly during January to July (Figure 3·3 c). Over half of the change in SAT in Alaska, Eurasia and the eastern Arctic Ocean has been ascribed to the AO, but less than half in the western Arctic (see Dickson *et al.*, 2000; Rigor *et al.*, 2000; Serreze *et al.*, 2000). The temperature changes associated with the AO are considered large enough to have an immediate effect on polar circulation (Morison *et al.*, 2000).

3.4. Precipitation and runoff

Precipitation is a key pathway for contaminant transport (Figure 1·2); rain, snow and fog scavenge aerosols and gasses from the atmosphere to deposit them at the surface (Chernyak *et al.*, 1996; Li *et al.*, 2002; Macdonald *et al.*, 2000a; Mackay and Wania, 1995; Malcolm and Keeler, 2002; Wania and Mackay, 1999). Scavenging by precipitation may be relatively weak in the desert-like

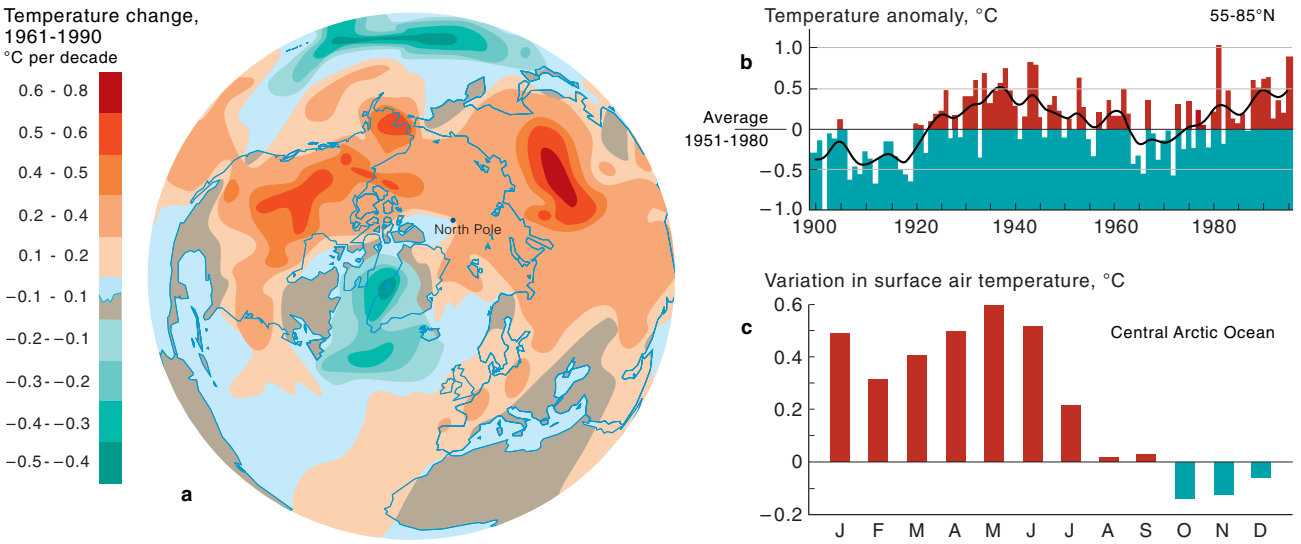

Figure 3·3. Temperature trends for the Arctic. This figure illustrates a) surface temperature trends over the Northern Hemisphere between 1961 and 1990 (courtesy of the Climate Monitoring and Data Interpretation Division of the Atmospheric Environment Service of Canada; Stewart *et al.*, 1998); b) annual temperature anomalies (55-85°N) for the period 1900 to 1995 set against the average for 1951 to 1980, showing that the high temperatures of the late 1980s and 1990s are matched by equally high temperatures during the 1930s and 1940s (adapted from Serreze *et al.*, 2000); and c) the average monthly variation in surface air temperature of the central Arctic Ocean between 1979 and 1995 showing that recent warming is mainly a winter–spring phenomenon (adapted from Serreze *et al.*, 2000).

conditions that prevail over much of the High Arctic. For example, mean precipitation for the Arctic Ocean is estimated to be about 25.2 cm/yr and evaporation about 13.6 cm/yr, yielding a net moisture flux to ground of 11.9 cm/yr (Barry and Serreze, 2000). The precipitation over land in the Arctic drainage basins is somewhat greater as implied by runoff yield (precipitation minus evaporation (P−E)) estimates of 21.2 cm/yr from the network of gauged discharge by rivers (Lammers *et al.*, 2001).

Given the changes in winds (Figure 3·2) and temperature that occur with shifts in the AO, it is to be expected that precipitation and evaporation within the Arctic will also be affected, both in amount and seasonality (Serreze *et al.*, 2000). Due to sparse monitoring networks and short time-series, it is difficult to assess with confidence the spatial or temporal variation of precipitation within the Arctic. Nevertheless, records suggest that precipitation has increased over northern Canada by about 20% during the past 40 years (Serreze *et al.*, 2000). The increase in southerly winds in the Norwegian Sea in winter and the penetration of cyclones from the Atlantic into the Barents, Kara and Laptev Seas, when the AO (or NAO) index is high, is reflected in increased moisture flux and precipitation during autumn and winter, especially in the area between 10°W and 50°E (Figure 3·4 a; Dickson *et al.*, 2000; Ser-

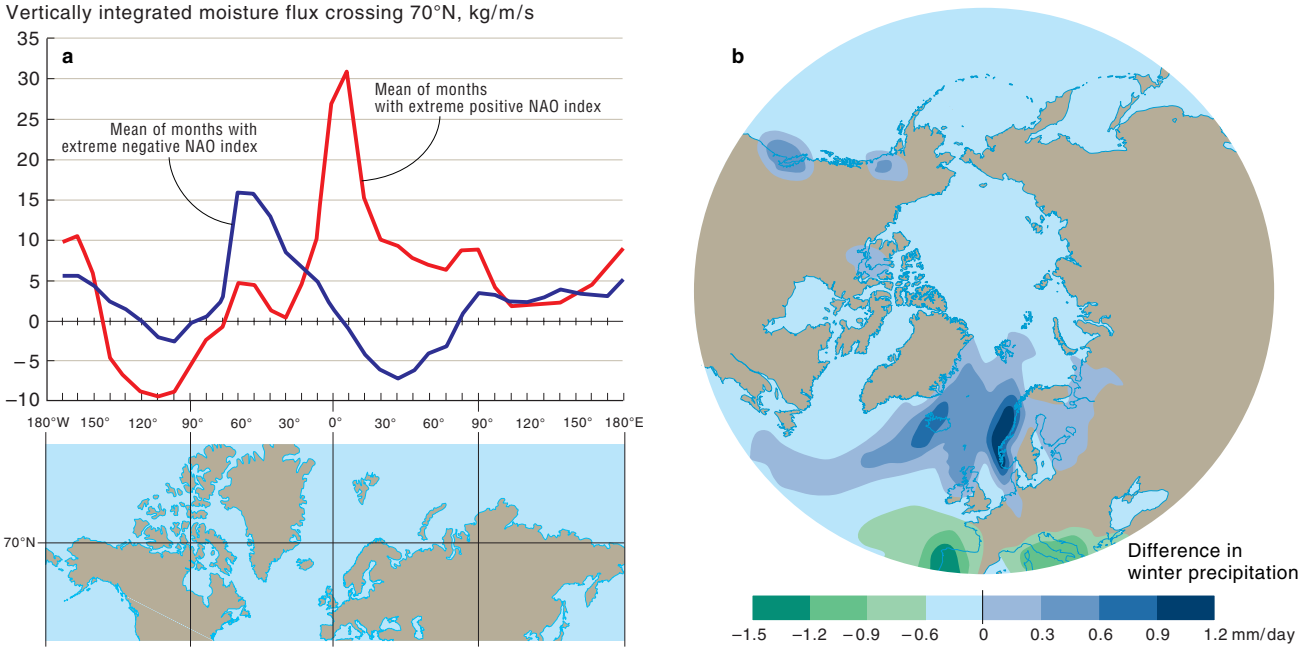

Figure 3·4. The effect of the North Atlantic Oscillation/Arctic Oscillation on precipitation in the Arctic; the NAO and AO are highly correlated and this figure is based on the NAO for which a longer time series exists. The figure illustrates a) the mean vertically integrated meridional flux crossing 70°N in winter for extreme NAO⁻ conditions (blue) and extreme NAO⁺ conditions (red) and b) the change in winter precipitation between extreme NAO⁻ and extreme NAO⁺ conditions (modified from Dickson *et al.*, 2000).

reze *et al.*, 2000; Walsh, 2000). The composite difference in precipitation (Figure 3·4 b), which may actually underestimate the change between index extremes (Dickson *et al.*, 2000), shows an increase of up to 15 cm/yr precipitation during winter in the Norwegian–Greenland Sea atmosphere–ocean conduit to the Arctic when the NAO is strongly positive. The response over the central Arctic to changes in the AO/NAO index is clearly much less, but it is likely that conditions there become wetter during index highs (Serreze *et al.*, 2000). Overall, it is estimated that P–E north of 70°N is 36% higher during periods of high index compared to low index (Serreze *et al.*, 1995). Over central and northern Canada, flux of moisture out of the Arctic increases when the AO/NAO is high, but toward the western Beaufort Sea moisture flux into the Arctic again increases (Figure 3·4 a).

Whether precipitation falls as snow or as rain, and how long snow covers surfaces are important components of climate that control the interaction of contaminants with the hydrological cycle (Macdonald *et al.*, 2002c; Wania, 1997). Snow cover in the Arctic varies from a maximum of about 46×10^6 km^2 to as little as 4×10^6 km^2 (Serreze *et al.*, 2000). As might be predicted from recent warming trends over Arctic land masses (Figure 3·3 a), there is evidence that the average area covered by snow has been decreasing by about 2% (450 000 km^2) per decade between 1979 and 1999 (Armstrong and Brodzik, 2001). A correlation between the AO and snow cover in Eurasia for the period from 1972 to 1997 suggests that a change from minimum to maximum AO index is accompanied by a loss of about 4×10^6 km^2 of snow cover, which could account for much of the trend described above (Vörösmarty *et al.*, 2001). The snow-cover anomalies plotted by Armstrong and Brodzik (2001) show a downward step around 1989 when the AO index sharply increased. The late 1980s up to at least 1998 has been identified as a period of low snow cover for both Eurasia and North America with the largest changes occurring in spring–summer (Serreze *et al.*, 2000); for Canada, there has been a decrease in snow depth, especially in spring, since 1946 (Brown and Goodison, 1996).

Precipitation minus evaporation integrated over a drainage basin should be equivalent to river discharge for the basin (if changes in groundwater or glacial storage are ignored). Arctic rivers exhibit large interannual variation (Semiletov *et al.*, 2000; Shiklomanov *et al.*, 2000; Stewart, 2000) making it difficult to link river flow to precipitation or temperature trends or to climatic variables such as the AO. For example, Shiklomanov *et al.* (2000) suggested little change in mean annual discharge for Arctic rivers between the 1920s and 1990s, whereas Semiletov *et al.* (2000) found recent increases for several Eurasian rivers, and Lammers *et al.* (2001) found evidence of increased winter discharge from rivers in Siberia and Alaska in the 1980s relative to the 1960s and 1970s. Within Can-

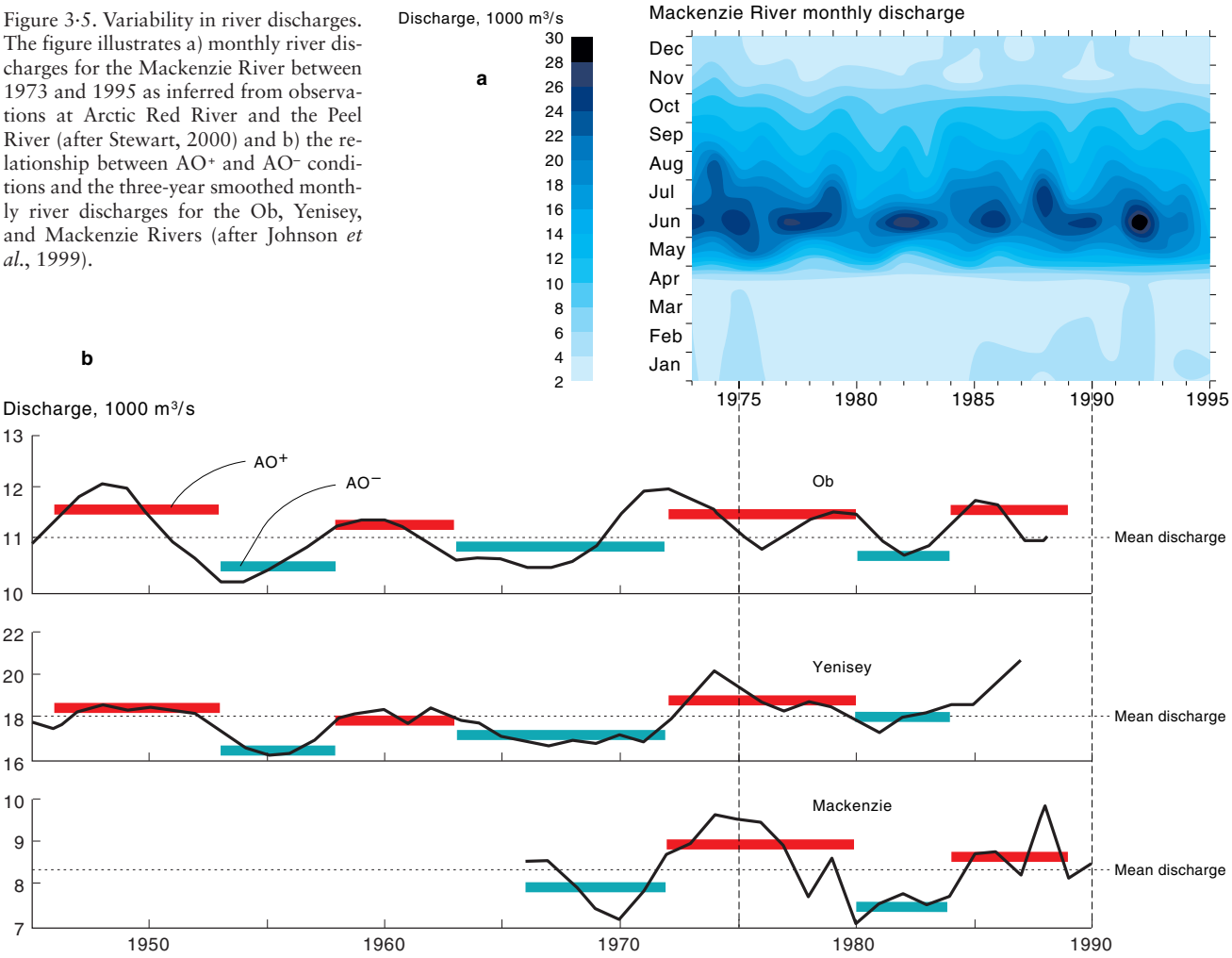

Figure 3·5. Variability in river discharges. The figure illustrates a) monthly river discharges for the Mackenzie River between 1973 and 1995 as inferred from observations at Arctic Red River and the Peel River (after Stewart, 2000) and b) the relationship between AO$^+$ and AO$^-$ conditions and the three-year smoothed monthly river discharges for the Ob, Yenisey, and Mackenzie Rivers (after Johnson *et al.*, 1999).

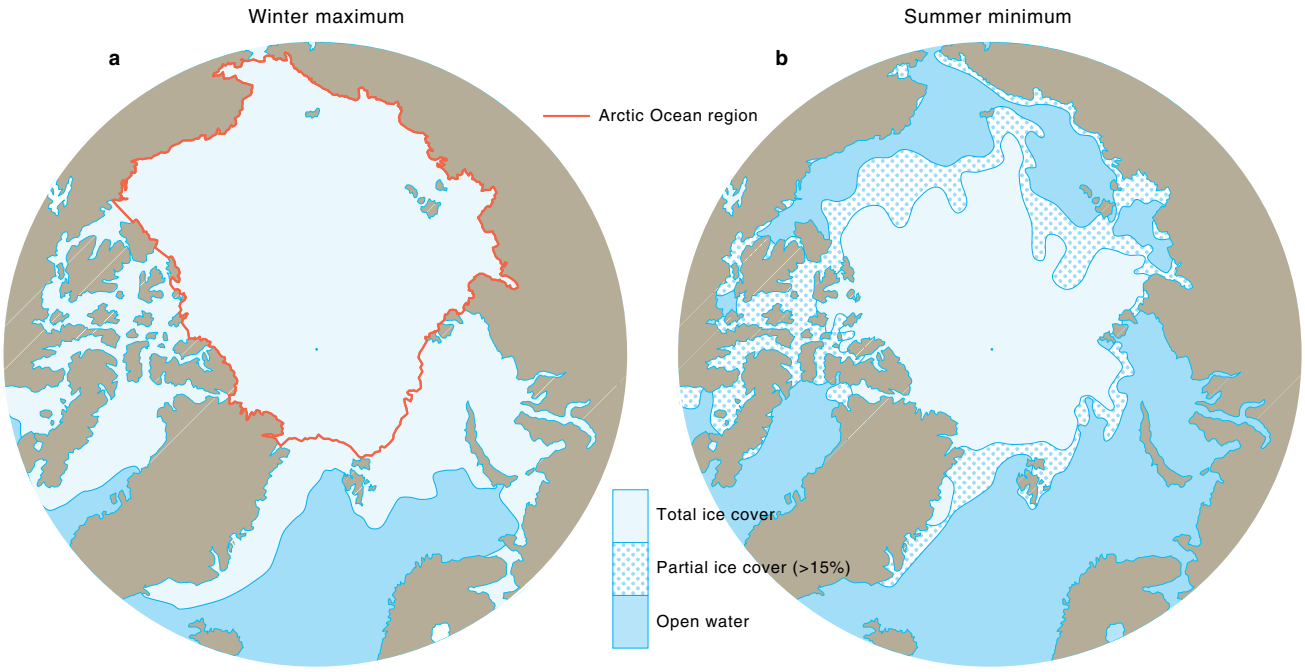

Figure 3·6. Arctic sea-ice cover derived from satellite imagery at the time of a) the winter maximum and b) the summer minimum (Johannessen and Miles, 2000). The red line delimits the Arctic Ocean area as defined for the ice trends shown in Figure 3·7 (Parkinson *et al.*, 1999).

ada, the Mackenzie Basin has undergone an exceptional warming between 1961 and 1990 (Figure 3·3 a); nevertheless, increased basin temperatures are not obviously evident in this river's hydrology (Figure 3·5 a) or in other Arctic rivers (Shiklomanov *et al.*, 2000). Instead, there is evidence of 3- to 4-year periodicity in peak flow and alterations in the seasonal shape of the hydrograph with higher flows delayed well into August, suggesting changes in both total annual discharge and its seasonality and possibly also changes in the relative importance of the river's sub-drainage basins. Such patterns appear to be only partially related to the AO, as evidenced by significant correlations between runoff and precipitation for the Mackenzie Basin and variation in North Pacific storm tracks (Bjornsson *et al.*, 1995). These correlations suggest that trans-Pacific transport of airborne contaminants may be the dominant component of contaminant loading for northwestern Canada, which is supported by air monitoring time-series data collected at Tagish in the Yukon (Bailey *et al.*, 2000). Hence, change related to atmospheric contaminant pathways for this region is more likely to come from the North Pacific, and it is possible that such change might be manifested as an alteration in the domains of influence of Pacific air masses versus Eurasian air masses.

Discharges for the Ob, Yenisey, and Mackenzie rivers appear to show a positive relationship with the North Pole pressure anomaly, with a lag in discharge of about 0.5 to 0.7 years (Figure 3·5 b), but such a relationship runs counter to the enhanced precipitation observed during AO+ (cyclonic) conditions (Figure 3·4). Even if all variation in Arctic river discharge at the 4- to 5-year time scale is assigned to shifts in AO/NAO index, the maximum effect on annual flow would be about 5 to 15% which is within the range of interannual variability (for example see Johnson *et al.*, 1999; Semiletov *et al.*, 2000).

3.5. The Arctic Ocean

3.5.1. Sea ice

3.5.1.1. Sea-ice cover

Sea ice controls the exchange of heat and other properties between the atmosphere and the ocean and, together with snow cover, determines the penetration of light into the sea. Ice also provides a surface for particle and snow deposition, a biological habitat above, beneath and within the ice and, when it melts in summer, creates stratification of the upper ocean.

During the 1990s, the science community recognized (with some alarm) that Arctic sea ice had been undergoing retreat over the previous three decades. Observed changes include: a reduction in area covered by sea ice (Johannessen *et al.*, 1999; Levi, 2000; Maslanik *et al.*, 1996; Parkinson *et al.*, 1999; Vinnikov *et al.*, 1999), an increase in the length of the ice-melt season (Rigor *et al.*, 2002; Smith, 1998), a loss of multi-year ice (Johannessen and Miles, 2000), a general decrease in the thickness of ice over the central Arctic Ocean (Rothrock *et al.*, 1999), and an increase of ice melt in the Beaufort Sea (Macdonald *et al.*, 1999a; McPhee *et al.*, 1998).

Analyses of satellite data from 1978 to 1987 indicate a decrease in Arctic sea-ice area of about 2.4% per decade (Gloersen and Campbell, 1991). Subsequent analyses have revised that figure upward to 4% per decade for the period from 1987 to 1994 with an estimated average loss during the entire period (1978 to 1997) of 3% per decade, which corresponds to the disappearance of 0.3×10^6 km^2 per decade of sea ice (Cavalieri *et al.*, 1997; Parkinson *et al.*, 1999). The Siberian shelves contribute significantly to the estimated ice losses. Multiyear ice is apparently being lost at an even greater rate, estimated at 7% per decade, partly replaced by first-year ice (Johannessen and Miles, 2000).

The large seasonal amplitude in area covered by ice (Figure 3·6) makes it difficult to assess trends. Further-

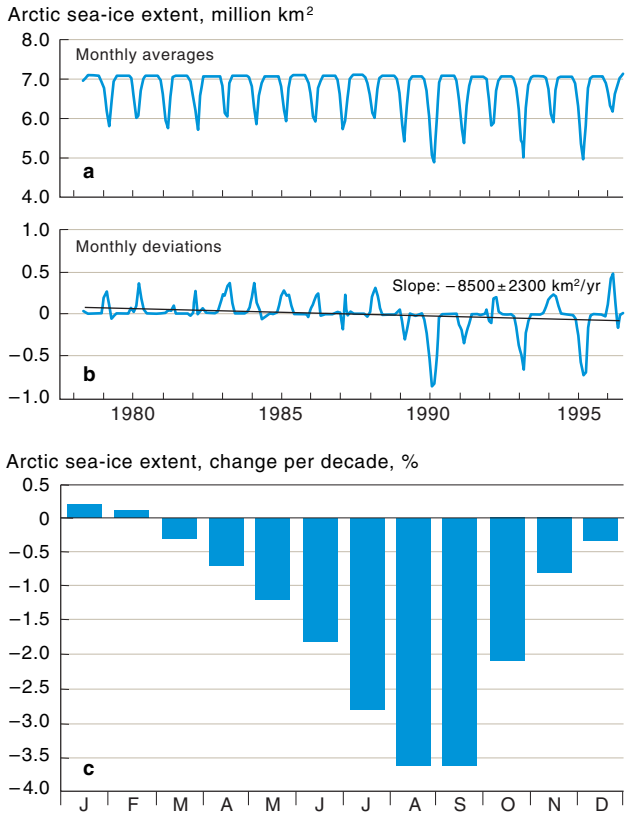

Figure 3·7. Trends in sea-ice cover in the Arctic Ocean. The figure illustrates a) monthly average sea-ice cover between 1979 and 1996 for the Arctic Ocean as delimited by Parkinson *et al.* (1999), see Figure 3·6 a; b) monthly deviations in sea-ice cover for this area showing the transition in 1990 to seasonally clear shelves; and c) the change in monthly sea-ice extent in percent per decade (1979-1995) showing ice loss to be predominantly a spring–summer phenomenon (after Serreze *et al.*, 2000).

more, various authors have partitioned the Arctic differently to assess changes in ice cover or have compared different years and/or different seasons (Dickson *et al.*, 2000; Johannessen and Miles, 2000; Maslanik *et al.*, 1996, 1999; Parkinson *et al.*, 1999). Despite these difficulties, the satellite data available since the late 1970s clearly indicate a reduction of 2% per decade of total ice area in winter (Johannessen *et al.*, 1999), and a significant shift in the marginal seas toward first-year ice which is easier to melt than multi-year ice because it is thinner and saltier. The total area of Arctic sea ice, including the marginal seas, varies from about 13×10^6 km² in winter to 5×10^6 km² in summer, and has shrunk by about 0.6×10^6 km² between 1978 and 1997 (Johannessen and Miles, 2000). The Arctic Ocean component, as defined by Parkinson *et al.* (1999) (Figure 3·7), which is about 7×10^6 km² in area, began to exhibit a much stronger seasonal modulation in ice cover in about 1989 (Figures 3·7 a and b) with the East Siberian and Beaufort Seas experiencing anomalous areas of open water in late summer at various times during the 1990s (Maslanik *et al.*, 1999; Parkinson *et al.*, 1999; Rigor *et al.*, 2002; Serreze *et al.*, 1995). That the loss of sea-ice cover is predominantly a spring–summer phenomenon is clearly shown by seasonal monthly trends for which June to October show the greatest change (Figure 3·7 c; Serreze *et al.*, 2000).

What part does the AO play in the variation of Arctic sea-ice distribution? The trends in ice cover with time (Figures 3·7 a and b) suggest that the wholesale clearing of ice from shelves is a phenomenon of the 1990s, timed with (Russian shelves) or slightly delayed from (Beaufort shelves) the shift to strong AO⁺ conditions in 1989. In the Beaufort Sea, Macdonald *et al.* (1999a) used stable isotope data ($\delta^{18}O$) collected from 1987 to 1997 to show that amounts of ice melt contained in the water column increased substantially at the same time as the AO index increased in 1989. During such conditions, the cyclonic circulation leads to greater ice divergence, more new ice in leads, enhanced heat flux, and reduced ridging, all of which imply thinning (Flato and Boer, 2001; Macdonald *et al.*, 1999a; Rigor *et al.*, 2002). Maslanik *et al.* (1996) draw the connection between increased penetration of cyclones, which is observed during AO⁺ conditions, and increased poleward transport of heat, and the absence of ice in late summer over the Siberian shelves. Based on results of a coupled sea/ice/ocean model, Zhang *et al.* (2000) suggest that there is a strong correlation between sea-ice thinning and the AO (~80%) due to dynamical effects, and that the Eurasian and Canada Basins respond differently to the AO forcing. The removal of the supply of ice from the Beaufort to the East Siberian Sea when the index becomes strongly positive (discussed in section 3.5.1.2.) results in depletion of thick ice in the eastern Arctic Ocean but may enhance thick ice-buildup in the western Arctic. This is important in light of the findings from repeat submarine surveys that ice thickness has decreased over the central Arctic by about 1.3 m between 1958 and 1976 and the 1990s (Rothrock *et al.*, 1999; Wadhams, 1997, 2000). According to several models (Holloway and Sou, 2002; Polyakov and Johnson, 2000; Zhang *et al.*, 2000), the submarine observations may have been conducted primarily in that part of the ocean that underwent thinning in response to a shift to AO⁺ conditions. The conclusion concerning reduction of ice thickness, while valid for the domain of submarine measurements, is not necessarily true for the whole Arctic Ocean and an alternative hypothesis that ice-thickness *distribution* changed but ice *volume* may not have changed in response to the AO needs to be carefully evaluated. The loss of ice cover between NAO⁻

Figure 3·8. The contrast in ice cover between pronounced AO⁻ conditions (1958) and pronounced AO⁺ conditions (1990s) (Dickson *et al.*, 2000; Maslanik *et al.*, 1999; Serreze *et al.*, 1995).

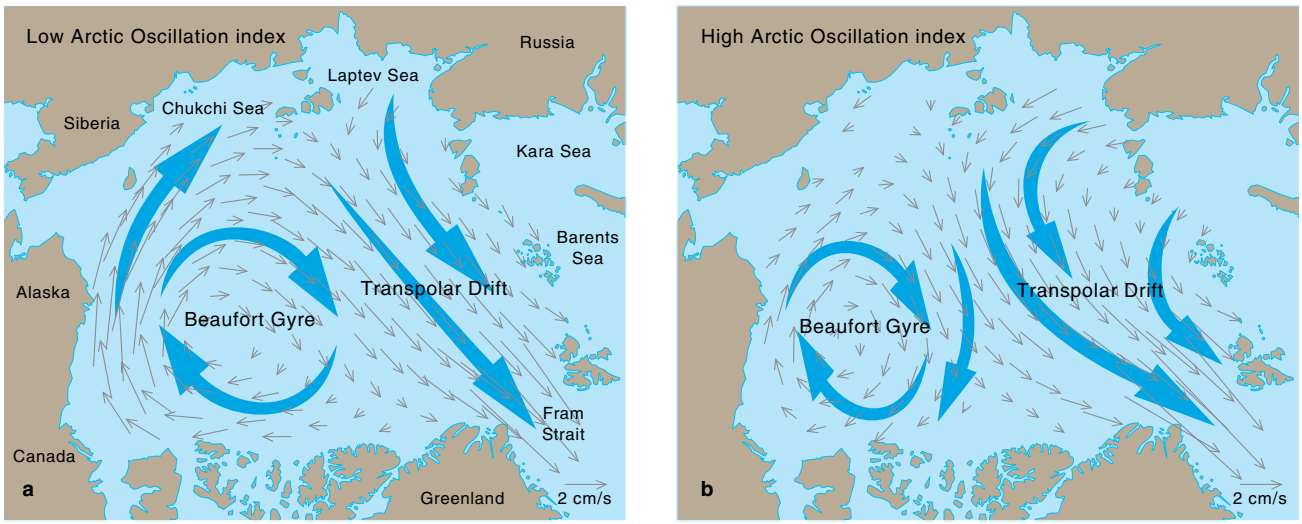

Figure 3·9. Ice drift patterns for a) years with pronounced AO⁻ (anticyclonic) conditions and b) pronounced AO⁺ (cyclonic) conditions (after Maslowski *et al.*, 2000; Polyakov and Johnson, 2000; Rigor *et al.*, 2002). The small arrows show the detailed ice drift trajectories based on an analysis of sea level pressure (Rigor *et al.*, 2002). The large arrows show the general ice drift patterns long recognized as the Beaufort Gyre to the left and the Transpolar Drift to the right.

and NAO⁺ conditions is estimated at 590 000 km^2 in the Barents and Greenland Seas (Dickson *et al.*, 2000), and if the remarkably open ice in the East Siberian Sea in 1990 and the Beaufort Sea in 1998 (Figure 3·8) is a product of the strong AO⁺ conditions of the early 1990s then perhaps half as much again ice loss occurred over the Russian and North American shelves due to AO forcing.

In light of the changes observed in ice cover during the 1990s, it is worth noting that over a century ago the Pacific whaling fleet experienced similar dramatic changes in ice conditions in the western Arctic. Extraordinarily open water from 1861 to 1867 may have contributed to a complacency that resulted in the loss of 32 ships, crushed in the ice along the Alaskan coast in 1871 (Bockstoce, 1986). In this respect it is interesting to remember the caution given by Polyakov and Johnson (2000), that both short (decadal) and long (60-80 yr) time-scale variations are associated with the AO.

From data collected between 1979 and 1997, Rigor *et al.* (2000) determined that sea-ice melt begins in the marginal seas by the first week of June and advances rapidly to the pole within two weeks. Freezing begins at the pole on 16 August, returning to the marginal seas by late September for a total melt season length of about 58 days at the pole and 100 days toward the margin. Based on satellite data (SSMR and SSM/I) predominantly from the Beaufort Sea, Smith (1998) estimated that the length of the melt season has been increasing by about 5.3 days per decade during the period 1979 to 1996. In contrast, Rigor *et al.* (2000) found a shortening of the melt season in the western Arctic of 0.4 days per decade and an increase of about 2.6 days per decade in the eastern Arctic. These trends in length of melt season parallel the general observations of a 1°C per decade *decrease* in temperature for the Beaufort Sea compared to a 1°C per decade *increase* in the eastern Arctic for the same time period (Rigor *et al.*, 2000).

Change in ice cover and its seasonality are especially important for contaminants like hexachlorocyclohexanes (HCHs), toxaphene, and polychlorinated biphenyls (PCBs) where air–sea exchange is a significant component of regional budgets (Macdonald *et al.*, 2000a,b). Furthermore, change in sea-ice cover, which alters light penetration and mixing, may also alter primary production and carbon flux (Gobeil *et al.*, 2001b) which then alters the vertical flux of particle reactive and bio-active contaminants from the ocean surface to depth.

3.5.1.2. Sea-ice drift

General ice motion in the Arctic Ocean follows the Transpolar Drift (TPD) on the Eurasian side of the ocean and the Beaufort Gyre in Canada Basin (Figure 1·1; Barrie *et al.*, 1998). Although it has long been recognized that large-scale ice-drift patterns in the Arctic undergo change (Gudkovich, 1961), it was not until the International Arctic Buoy Programme (IABP) that sufficient data became available to map the ice drift in detail and thereby directly evaluate the role of the AO in changing ice drift trajectories. The IABP data from 1979 to 1998 suggest two characteristic modes of Arctic ice motion, one during low index (AO⁻) and the other during high index (AO⁺) periods (Figures 3·9 a and b; Proshutinsky and Johnson, 1997; Rigor *et al.*, 2002). The ice-motion scheme shown by drifting buoys is reasonably well corroborated by models that are being used to investigate the influence of the atmospheric variability inherent in the AO (Maslowski *et al.*, 2000; Polyakov and Johnson, 2000). There are two overarching differences between the two ice circulation modes:

1) during AO⁻ conditions (Figure 3·9 a), ice in the TPD tends to move directly from the Laptev Sea across the Eurasian Basin and out into the Greenland Sea, whereas during strong AO⁺ conditions (Figure 3·9 b) ice in the TPD takes a strong cyclonic diversion across the Lomonosov Ridge and into Canada Basin (Mysak, 2001); and

2) during AO⁺ conditions (Figure 3·9 b), the Beaufort Gyre shrinks back into the Beaufort Sea and becomes more disconnected from the rest of the Arctic Ocean, exporting less ice to the East Siberian Sea and importing little ice from the region to the north of the Canadian Arctic Archipelago – a region known to contain the Arctic's thickest multi-year ice (Bourke and Garrett, 1987).

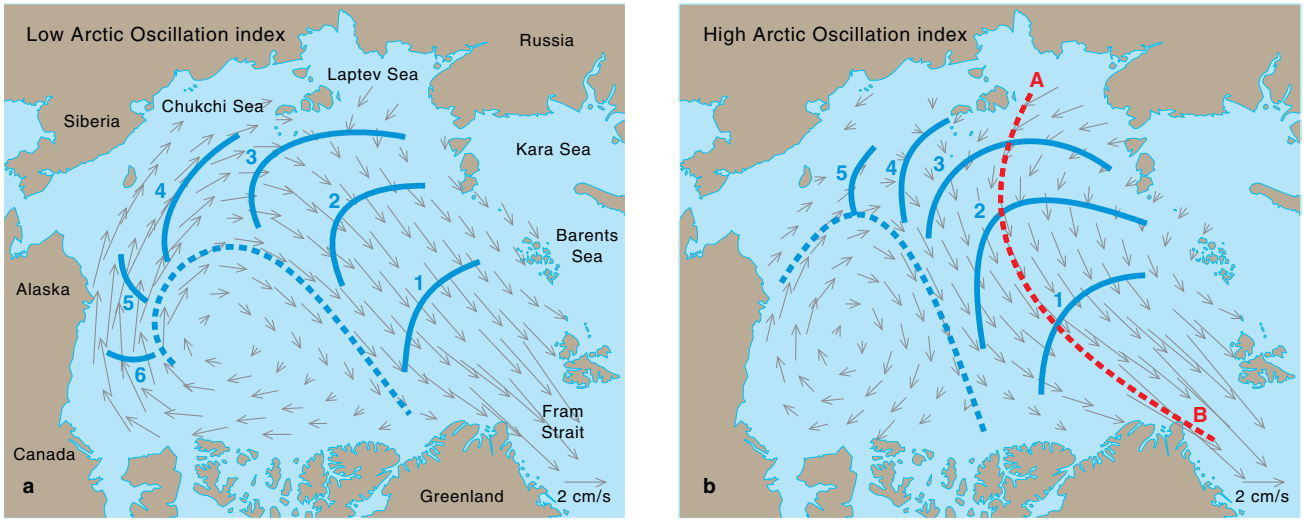

Figure 3·10. Time taken in years for sea ice at that location to reach Fram Strait during a) pronounced AO⁻ conditions and b) pronounced AO⁺ conditions (after Rigor *et al.*, 2002). Line A-B represents the transect used to describe change in sea ice during drift shown in Figure 3·12.

There are also changes in the time required for ice to transit the ocean (Figures 3·10 a and b) and in the destinations of ice exported from shelves. During winter, under AO⁺ conditions there is an increase in ice advection away from the East Siberian and Laptev Sea coasts, leading to the production of more new, thin ice in the coastal flaw leads (Figure 3·11; Polyakov and Johnson, 2000; Rigor *et al.*, 2002), a decrease in the advection of ice from the western Arctic into the eastern Arctic, possibly an increased advection of ice from the Arctic Ocean to the Barents Sea through the Svalbard–Franz Josef Land passage (Polyakov and Johnson, 2000), and an increase in the 900 000 km^2 of ice advected out of the Arctic through Fram Strait (Morison *et al.*, 2000; Rigor *et al.*, 2002). Interestingly, increased ice export through Fram Strait can be produced by shifts to both negative and positive AO states (Dickson *et al.*, 2000).

Comparing the two modes of ice drift (Figures 3·9 a and b), it is apparent that during AO⁻ conditions the East Siberian Sea imports much of its ice from the Beaufort Sea and that there is an efficient route to carry ice clockwise around the Arctic margin of the East Siberian Sea and out toward Fram Strait. Under the strong AO⁺ conditions of the early 1990s, the Beaufort Sea ice be-

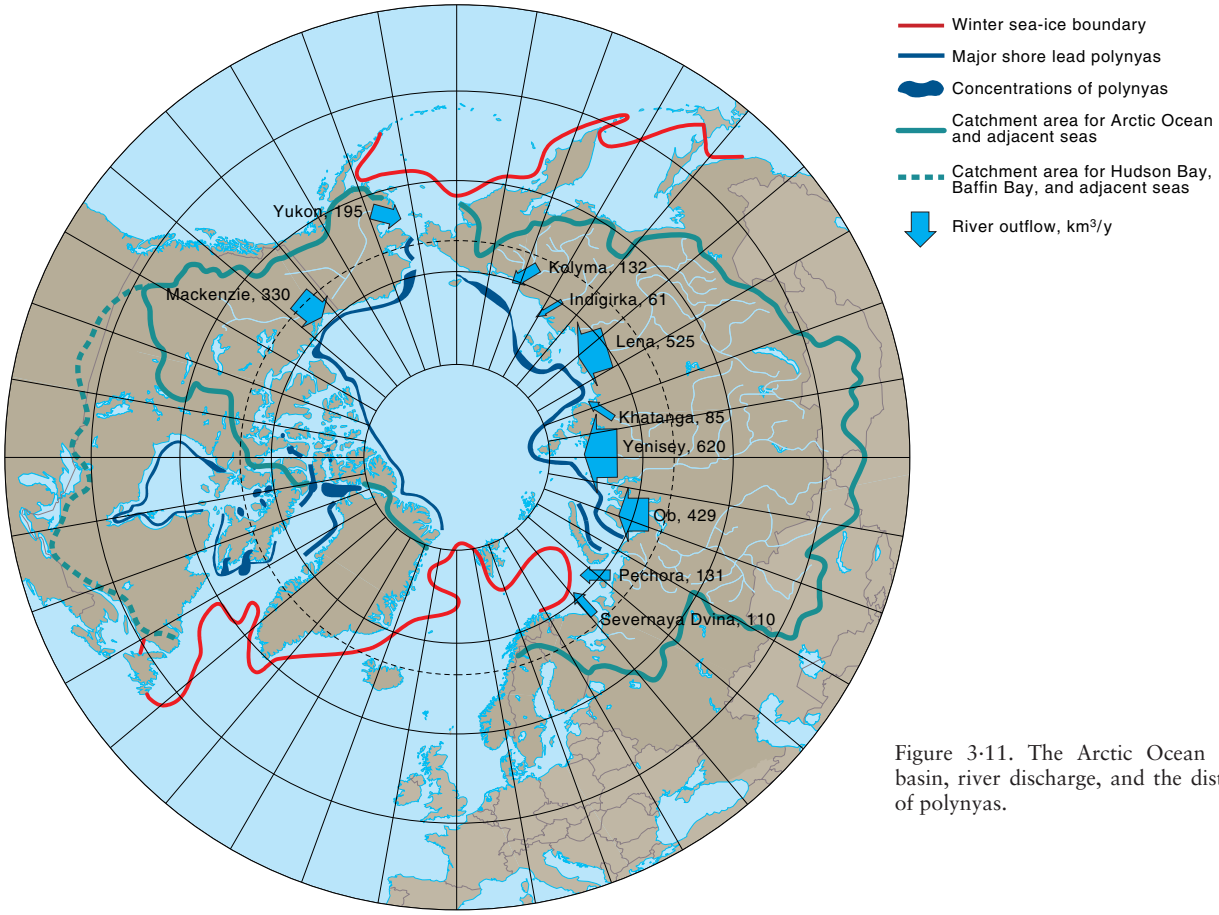

Figure 3·11. The Arctic Ocean drainage basin, river discharge, and the distribution of polynyas.

came more isolated, whereas ice from the Kara, Laptev and East Siberian Seas was displaced into the central Arctic and toward the Canadian Arctic Archipelago. It is not clear from the IABP data how much ice from the Russian shelves might transport into the Canadian Arctic Archipelago or the Beaufort Gyre under AO⁺ conditions, but models (Maslowski *et al.*, 2000; Polyakov and Johnson, 2000), paleo-studies of Eurasian wood (Dyke *et al.*, 1997; Tremblay *et al.*, 1997), and sediment records (Darby *et al.*, 2001) all suggest that such transport is likely and may at times be important.

Sea ice provides a rapid means to accumulate and transport contaminants long distances without dilution (Pfirman *et al.*, 1995; Wadhams, 2000). The response in ice-drift trajectories to change in AO index (Rigor *et al.*, 2002) therefore carries immense implication for the connectivity between contaminant source and sink regions for ice pathways within the Arctic Ocean.

3.5.1.3. Sea-ice transport of material

Sea ice is an important mechanism for the transport of coastal and continental shelf sediments to the interior Arctic Ocean and out into the Greenland Sea (Barrie *et al.*, 1998; Dethleff *et al.*, 2000b; Nürnberg *et al.*, 1994). Sediments become incorporated into ice formed over shelves. Although all the shelves of the Arctic are implicated in this process, the Laptev Sea has proven so far to be the most efficient exporter of sediment-laden ice (Eicken *et al.*, 1997, 2000; Reimnitz *et al.*, 1992, 1993, 1994). This transport process involves several steps including: 1) the delivery of sediment to the shelf by rivers or from coastal erosion where much of it may become trapped; 2) the incorporation of sediment into the ice, either through ice grounding or through suspension freezing in mid-shelf flaw polynyas; 3) the export of ice from the shelf to the interior ocean; 4) the transport of ice across Arctic basins, potentially with some loss of sediment during transport; and 5) the release of sediment at the location where the ice melts (Figure 3·12). During transport, the ice 'weathers', ablating at the surface during summer and incorporating more ice on the bottom during winter, with the consequence that some of the sediment entrained over the shelf migrates to the surface of the ice. Additionally, atmospheric particulates deposit and accumulate on the ice along its transport route. Consequently, an increase or decrease in the time taken for ice to cross the Arctic Ocean (Figure 3·10) respectively increases or decreases the time for accumulation of atmospheric aerosols and sediments at the ice surface. Each step in the ice pathway can be altered by climate change. For example, fine river sediments (known to carry contaminants) become trapped in estuaries by the so-called 'marginal filter' (Lisitzin, 1995). Sea level rise, change in the ice climate, or change in the river's hydrology can all alter the location of this filter. The process of suspension freezing might be enhanced by larger amounts of open water over shelves in the autumn whereas more sediment might be lost from the ice during transport due to predominance of thin, first-year ice and augmented melting. Finally, the location at which ice melts and drops its particulate and dissolved loads can change. There are no direct data on how these components of the ice-transport pathway respond either individually or collectively to the AO; however, long-term sediment records (Darby *et al.*, 2001), disequilibria in sediments (Gobeil *et al.*, 2001b), and the distribution of sediments within the Arctic Ocean (Stein, 2000) suggest that climate forcing akin to the AO probably occurs.

3.5.2. Ocean currents and water properties

3.5.2.1. Surface water

For ocean currents that deliver contaminants to Arctic ecosystems, surface water is most important because it interacts more directly with biota and ecosystems. Surface water pathways will to some extent reflect ice-drift trajectories (Morison *et al.*, 2000), responding in like manner to the state of the AO (Figure 3·9). In strong AO⁺ conditions, water in the TPD makes a diversion into the Makarov Basin and the Beaufort Gyre contracts and retreats into Canada Basin. However, the AO results in other critical changes in surface water not represented by ice drift. With the enhanced inflow and spreading of water in the Atlantic Layer, a retreat of the cold halocline in the Eurasian Basin under AO⁺ conditions was also

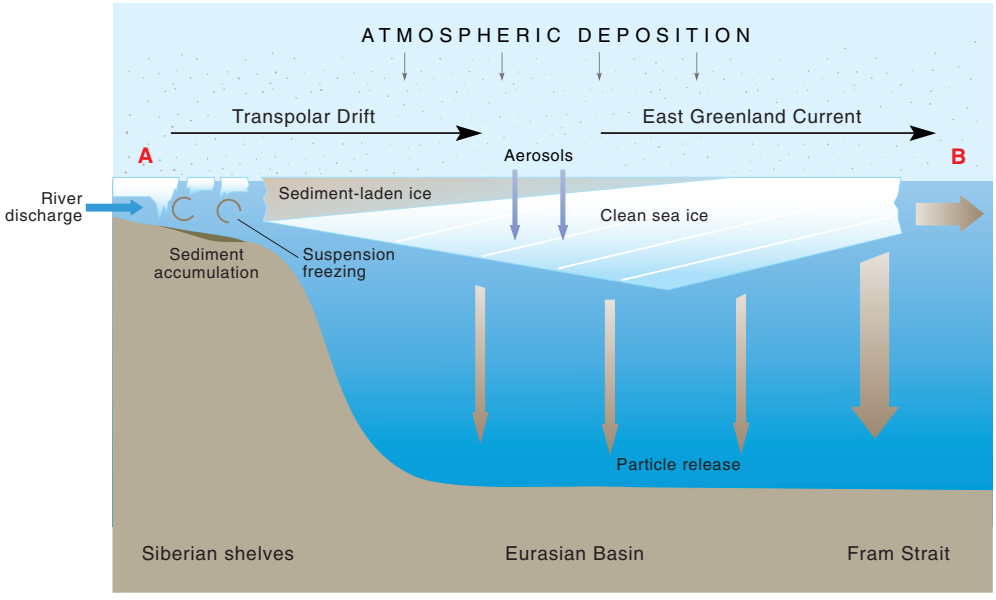

Figure 3·12. A schematic diagram showing the accumulation and transport of sediments and contaminants by sea ice over the transect marked A-B on Figure 3·10 b (modified from Lange and Pfirman, 1998).

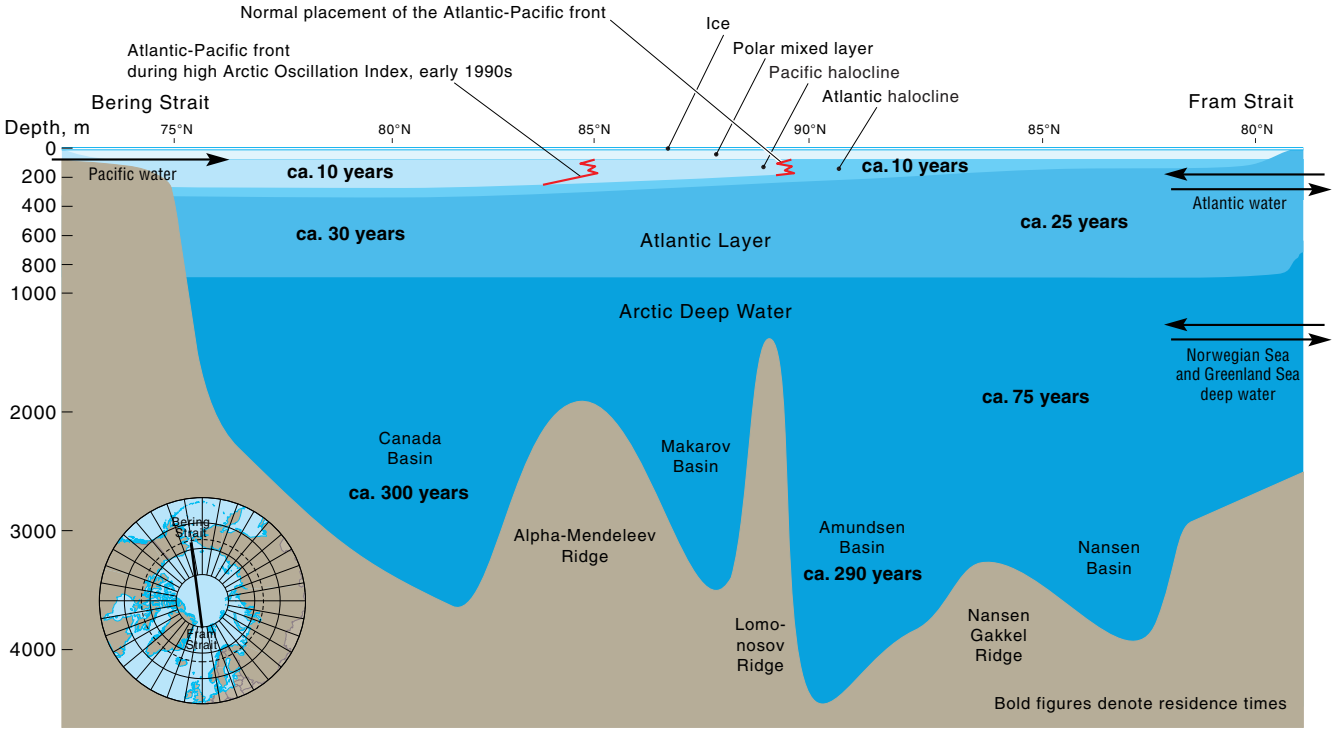

Figure 3·13. The stratification of the Arctic Ocean, showing the polar mixed layer, the Pacific and Atlantic domains of influence and the haloclines. The red lines show the normal placement and the displacement of the Atlantic-Pacific front during the high Arctic Oscillation index of the early 1990s.

noted (Steele and Boyd, 1998). The halocline (Figure 3·13) provides stratification between the Atlantic Layer and surface water thereby preventing or reducing the transfer of properties such as heat or contaminants between deep and surface layers. The increase in salinity of surface water in the Eurasian Basin noted by Steele and Boyd (1998), however, was not due to enhanced inflow from the Atlantic, which actually freshened slightly with the high AO/NAO index of the late 1980s, but rather to the diversion of river inflow at the margins of the Arctic Ocean.

Models (Figures 3·14 a and b; Dickson, 1999; Johnson and Polyakov, 2001; Maslowski *et al.*, 1998) and geochemical measurements (Ekwurzel *et al.*, 2001; Guay *et al.*, 2001; Macdonald *et al.*, 1999a, 2002a; Schlosser *et al.*, 2002) clearly show that with the high AO⁺ index of the late 1980s, river water entering the Laptev and Kara shelves was forced to the east rather than directly off the shelf and into the TPD. Under strong AO⁺ conditions, perhaps 1000 km³/yr or more of runoff from the Lena, Ob, and Yenisey rivers stopped entering the Eurasian Basin and entered, instead, the East Siberian shelf and thence the Canadian Basin, possibly to exit the Arctic Ocean via the Canadian Arctic Archipelago (Figures 3·14 c and d) (Morison *et al.*, 2000). A consequence of this diversion was a reduction of stratification in the Eurasian Basin (Steele and Boyd, 1998) and an increase in stratification in the Canadian Basin (Macdonald *et al.*, 1999a, 2002a). The drop in the AO index toward the end of the 1990s (Figure 3·1) appears to have initiated a return to the former pathways for river water in the Eurasian Basin (Björk *et al.*, 2002; Boyd *et al.*, 2002).

At the same time, Atlantic surface water invaded the Makarov Basin, displacing water of Pacific origin from the top 200 m of the water column (McLaughlin *et al.*, 1996); this represents a rapid change of water source and properties for about 20% of the Arctic Ocean's area (Figures 3·13 and 3·15). Although there does not appear to be a strong AO signal in the Pacific inflow through Bering Strait (~0.8 Sv, there has been a general decline of about 15% since the early 1940s (Coachman and Aagaard, 1988; Roach *et al.*, 1995) and the flow may also have freshened due to runoff and precipitation in the Bering Sea (Weingartner, pers. comm., 2001).

3.5.2.2. The Atlantic Layer

Repeat hydrographic surveys of Arctic basins, commencing in 1987 (Aagaard *et al.*, 1996; Anderson *et al.* 1989; Carmack *et al.*, 1995; McLaughlin *et al.*, 1996; Morison *et al.*, 1998; Quadfasel *et al.*, 1991; Swift *et al.*, 1997), have revealed an Arctic Ocean in transition. The timing of that transition in the late 1980s implicates the AO (or NAO) as a major source of forcing that has altered connections between the Atlantic and the Arctic Oceans and so changed the distribution of Atlantic water within the Arctic, both in the surface layer, as discussed in section 3.5.2.1, and in the deeper Atlantic Layer water (Dickson, 1999; Macdonald, 1996). Ironically, some of the clearest evidence of these changes has come from contaminant time series, in particular the tracing of artificial radionuclides released from European reprocessing plants into the waters of the eastern North Atlantic (Carmack *et al.*, 1997; Smith *et al.*, 1998).

A major change, starting in about 1989, was a possible intensification of flow from the Atlantic into the Arctic through Fram Strait and the Barents Sea in response to the shift to strong AO⁺ or NAO⁺ conditions (Figure 3·15 – for detailed reviews see Dickson *et al.*, 2000; Morison *et al.*, 2000; Serreze *et al.*, 2000). The winds associated with AO⁺ conditions (Figure 3·2) increased the rate of northward transport of surface water in the Norwegian Sea and produced warmer air temperatures,

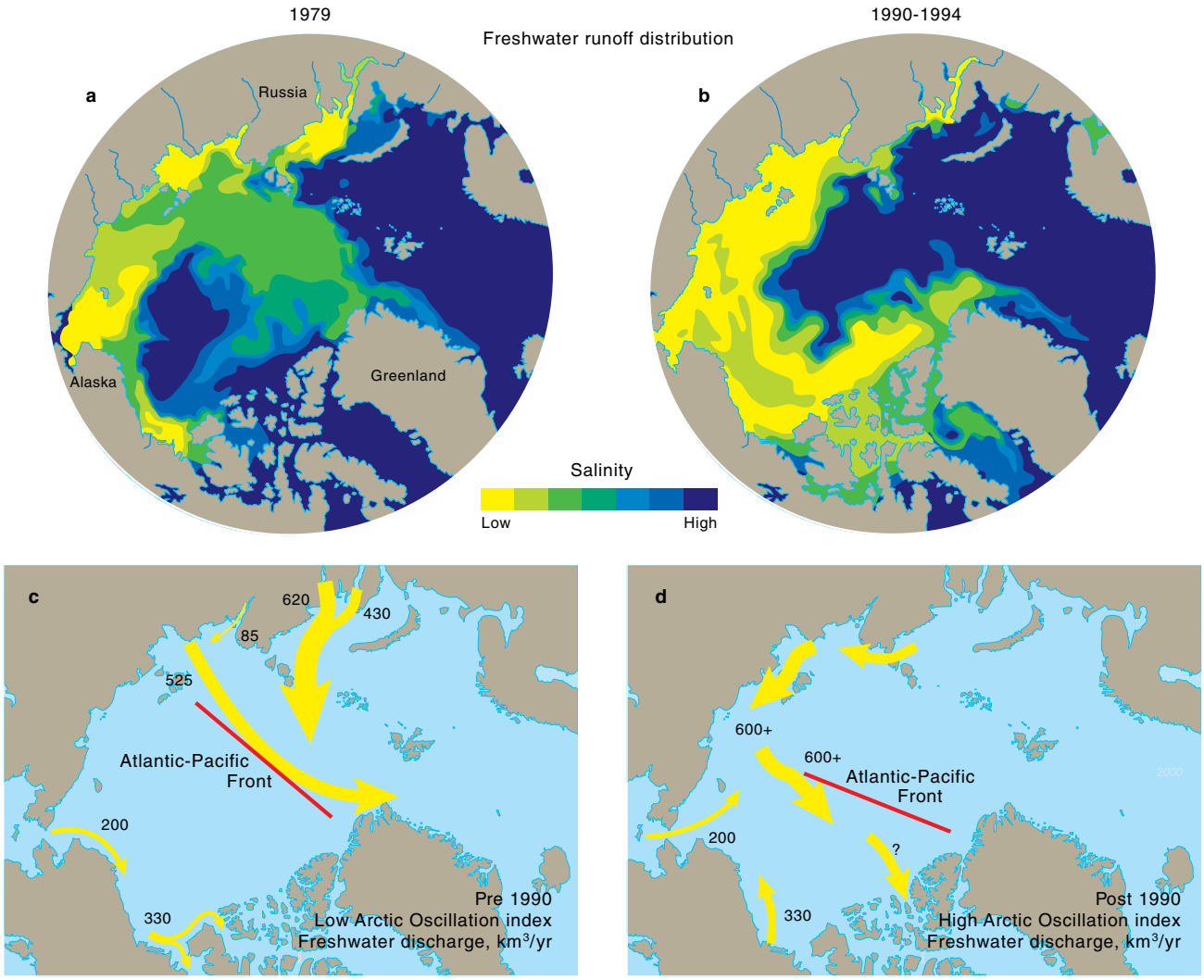

Figure 3·14. Transport of freshwater runoff across the Arctic Ocean. This figure illustrates a) freshwater pathways during pronounced AO⁻ conditions (1979); b) freshwater pathways during pronounced AO⁺ conditions (1990-94) (both a and b are based on model results by W. Maslowski reproduced in Dickson, 1999); c) the amounts and changes in pathways for freshwater inflows during AO⁻ conditions; and d) the amounts and changes in pathways for freshwater inflows during AO⁺ conditions.

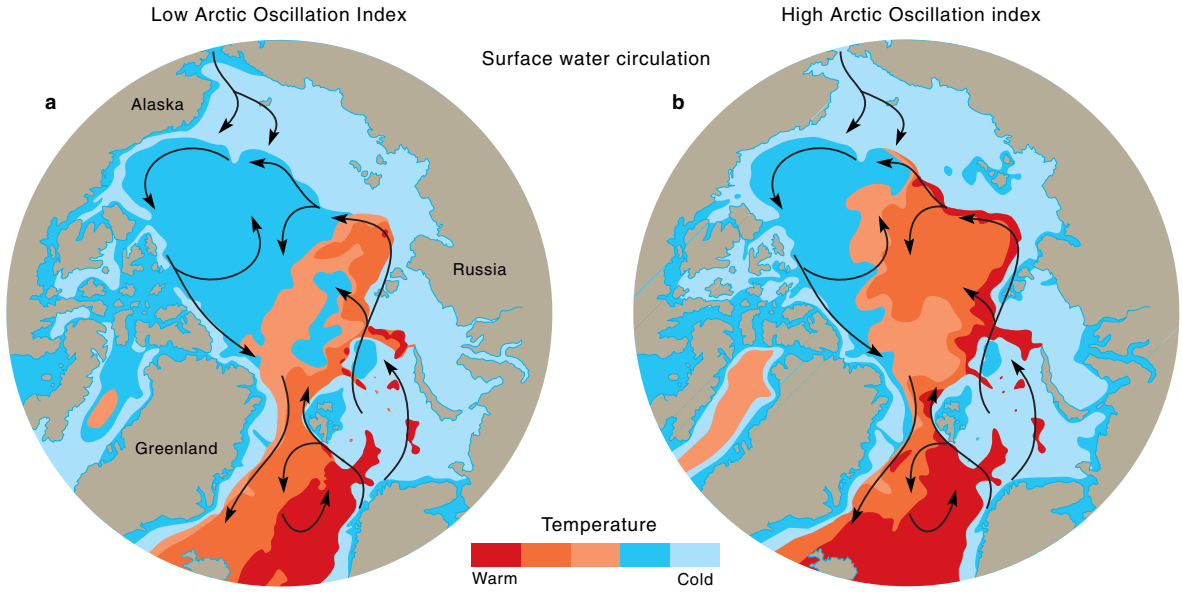

Figure 3·15. The change in Atlantic water inflow to the Arctic and distribution within the Arctic produced by the exceptionally strong shift to AO⁺/NAO⁺ conditions around 1989. The figure illustrates a) the distribution of Atlantic Layer water prior to the late 1980s and b) the distribution of Atlantic Layer water during the early to mid 1990s. The distribution of the Atlantic Layer (see Figure 3·13) is based on Hodges (2000), Maslowski *et al.* (2000), McLaughlin *et al.* (1996), and Morison *et al.* (1998, 2000). The Atlantic Layer boundary currents, which are relatively fast, transport properties along basin margins at about 1-5 cm/s (300-1600 km/yr; Woodgate *et al.*, 2001).

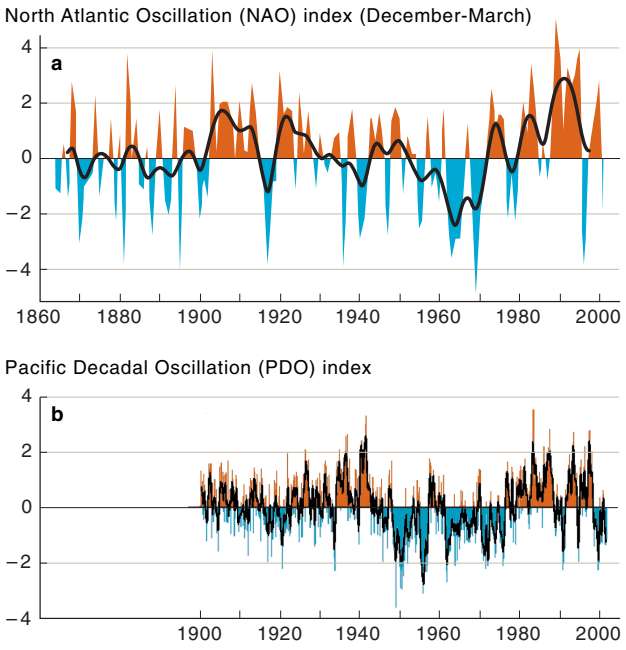

Figure 3·16. a) The North Atlantic Oscillation index from 1860 to 2000 (source: Hurrell, 2002) and b) the Pacific Decadal Oscillation index 1900 to 2000.

which, together with the shorter transit times, contributed to warming by about 2.3°C of the Atlantic water entering the Arctic Ocean (Swift *et al.*, 1997). The Atlantic water also exhibited slightly decreased salinity (by 0.03-0.05), probably reflecting increased precipitation in the Nordic Seas during NAO⁺ conditions (Figure 3·4 b).

Within the Arctic Ocean, the changes in the distribution and composition of the Atlantic Layer water were spectacular when set against the traditional perception of a quiet, steady-state ocean (see Figures 3·13 and 3·15 for the position of the Atlantic Layer within the water column). The front between Atlantic water and Pacific water, traditionally located over the Lomonosov Ridge was forced over to the Alpha-Mendeleev Ridge (Figure 3·13 and see McLaughlin *et al.*, 1996; Morison *et al.*, 2000). At the same time, the inflowing water could be detected in the Atlantic Layer by an approximately 1.5°C temperature rise above the climatological norm (Carmack *et al.*, 1995). The changes in volume and composition of Atlantic water entering the Arctic Ocean through Fram Strait continue to cascade through the Arctic basins, first as changes in properties along the boundaries (McLaughlin *et al.*, 2002; Newton and Sotirin, 1997), then as changes propagated into the basin interiors along surfaces of constant density (Carmack *et al.*, 1997) (Figure 3·13). Woodgate *et al.* (2001) estimated that in 1995 to1996, the boundary flow over the southern margin of the Eurasian Basin was transporting 5±1 Sv at about 1 to 5 cm/s (300-1600 km/yr). When water in the boundary current reached the Lomonosov Ridge, the flow split with around half entering the Canadian Basin along its margin and half returning toward Fram Strait along the Lomonosov Ridge. The high NAO index of the late 1980s (Figure 3·16 a) also strengthened and warmed the inflowing Barents Sea branch of Atlantic water, perhaps by as much as 25% relative to 1970 (Dickson *et al.*, 2000), which probably led to a parallel warming and increase in salinity of the Barents Sea (Zhang *et al.*, 2000).

3.6. Adjacent polar seas and regions

3.6.1. The Nordic and Barents Seas

The Nordic Seas (Greenland, Iceland and Norwegian Seas) are dominated by a northward flow of warm Atlantic water on the eastern side and a southward flow of cold Arctic water on the western side (Figure 3·17). The northward flows are large, estimated at 7 to 8 Sv in the Norwegian Atlantic Current (NwAC). About 2 Sv of the NwAC enters the Barents Sea and the remainder continues north where part enters the Arctic Ocean through Fram Strait and part re-circulates toward the west (Barrie *et al.*, 1998; Dickson *et al.*, 2000).

The currents that transport warm Atlantic water to the Barents Sea are important for regional climate, keeping the entire Norwegian Sea and large areas of the Barents Sea ice free and open for biological production. These same currents provide a significant pathway for contaminants from the western coast of Europe and perhaps from as far as the eastern North American seaboard. The volume flux for these currents and the distribution of Atlantic water in the Norwegian Sea is strongly influenced by wind forcing (e.g., Blindheim *et al.*, 2000; Hansen *et al.*, 2001; Mork and Blindheim, 2000; Orvik *et al.*, 2001), with a large component of variation accounted for by the NAO index (Figure 3·17).

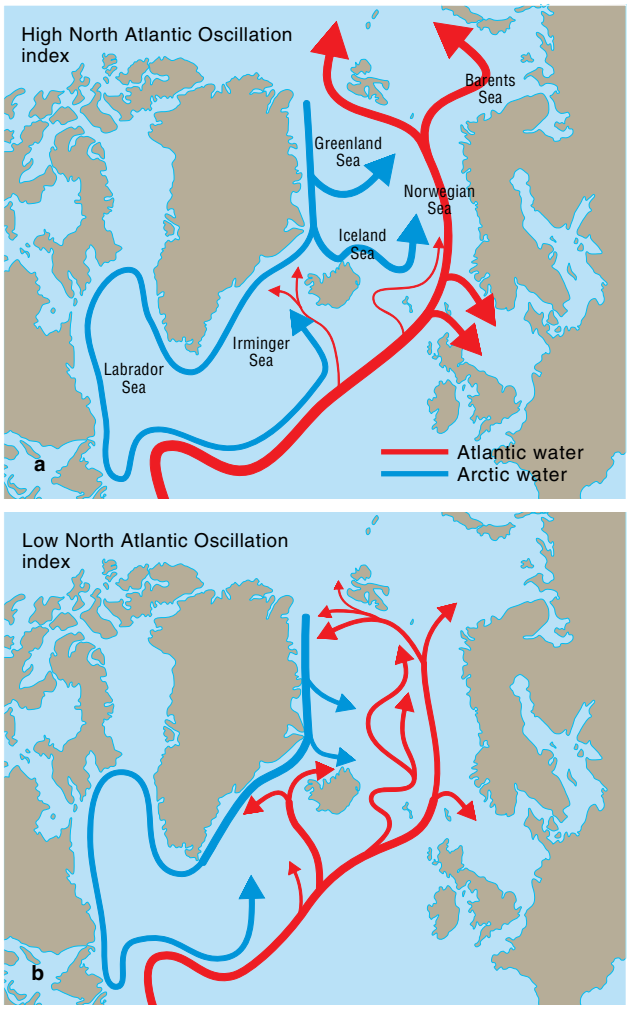

Figure 3·17. Main features of the circulation of Atlantic waters (red) and Arctic waters (blue) in the northern North Atlantic and Nordic Seas under a) pronounced NAO⁺ conditions and b) pronounced NAO⁻ conditions (source: Blindheim *et al.*, 2000).

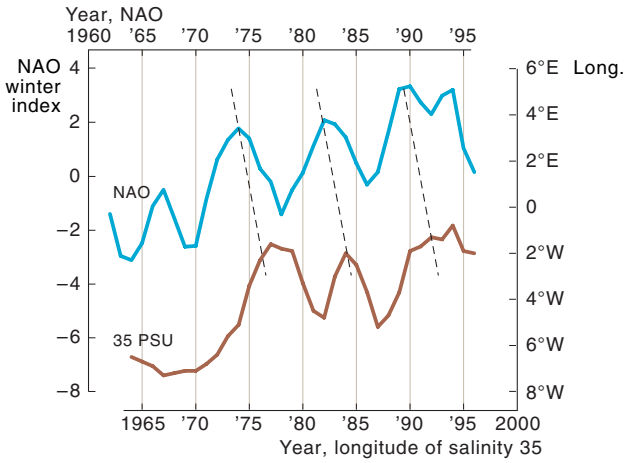

Figure 3·18. A comparison of the western extent of Atlantic Water (brown curve) and the NAO index (blue curve). The brown curve reflects three-year moving averages of the longitude of maximum western extent of water with salinity of 35 in the section along 65°45'N (source: Blindheim *et al.*, 2000). The blue curve reflects a three-year moving average of NAO winter values (Dec-March) (updated from Hurrell, 1995). The two lines are strongly correlated with a time-lag of ca. 2-3 years between respective peaks.

A high winter NAO index (December-March) is associated with more south-westerly winds and storms which increase the volume flux of the inner (eastern) branch of the NwAC (compare Figure 3·17a with Figure 3·17b; Orvik *et al.*, 2001). Under these conditions, more Atlantic water reaches the Barents Sea and the Arctic Ocean, and less Atlantic water is transported into the central Norwegian Sea (Blindheim *et al.*, 2000). Coincidentally, more Arctic water from the East Greenland Current, which is fresher and cooler, enters the central Norwegian Sea. During periods of low NAO index (Figure 3·17b), weaker south-westerlies result in a weaker inner branch of the NwAC and a greater extension of Atlantic water to the west in the Norwegian Sea. The east–west extent of Atlantic water passing through the Norwegian Sea predictably follows the winter NAO index with a lag of 2 to 3 years (Figure 3·18).

Large-scale atmospheric features, as represented by the NAO, appear to affect the strength of the Atlantic inflow to the Barents Sea (Figure 3·17; Dippner and Ottersen, 2001; Ingvaldsen *et al.*, 2002) but this inflow seems also to be closely related to regional atmospheric circulation (Ådlandsvik and Loeng, 1991). Low atmospheric pressure over the inflow area favours increased inflow of Atlantic water resulting in higher than average temperatures in the Barents Sea.

From the mid-1960s the NAO index has increased progressively with relatively high values in the early- to mid-1990s (Figure 3·16a). Increased atmospheric carbon dioxide (CO_2) is projected to increase storm frequencies and weaken thermohaline circulation (IPCC, 2002) probably producing upper ocean circulation more like that observed during NAO+ conditions (Figure 3·17a). The increased wind-induced transport may be partly compensated for by a reduced thermohaline circulation with the net effect of a relatively large transport of Atlantic water to the Barents Sea and the Arctic Ocean, and increased influence of Arctic water masses on the western and central Norwegian Seas.

Vertical mixing of water masses is important for biological production and for redistribution of contaminants. In the Greenland Sea, deep or intermediate water masses are created by winter cooling of the upper layer, a process which has weakened or even ceased since the beginning of the 1970s (e.g., Bönisch *et al.*, 1997). In the Barents Sea, vertical mixing takes place down to 300 m in cold years, which means the whole water column is well mixed from the surface to the bottom. In addition to vertical mixing due to cooling in the open sea, vertical mixing also takes place in frontal zones of the Nordic and Barents Seas. Cold and warm water masses meet and mix to create a very productive area where there is likely to be a high organic sedimentation rate. The fronts also act as a barrier for distribution of both plankton and fish.

3.6.2. The Bering and Chukchi Seas

The eastern Bering and Chukchi Seas are contiguous shelves that extend nearly 2000 km northward from the Alaskan Peninsula to the continental slope of the Arctic Ocean's Canada Basin (Figure 3·19). Both shelves are broad and shallow with typical depths on the Bering and Chukchi shelves being <100m and <60m, respectively. The Bering Strait provides a continuous pathway by which approximately 25 000 km^3 of water from the North Pacific Ocean enter the Arctic Ocean annually (Roach *et al.*, 1995). The nutrient-rich, but moderately

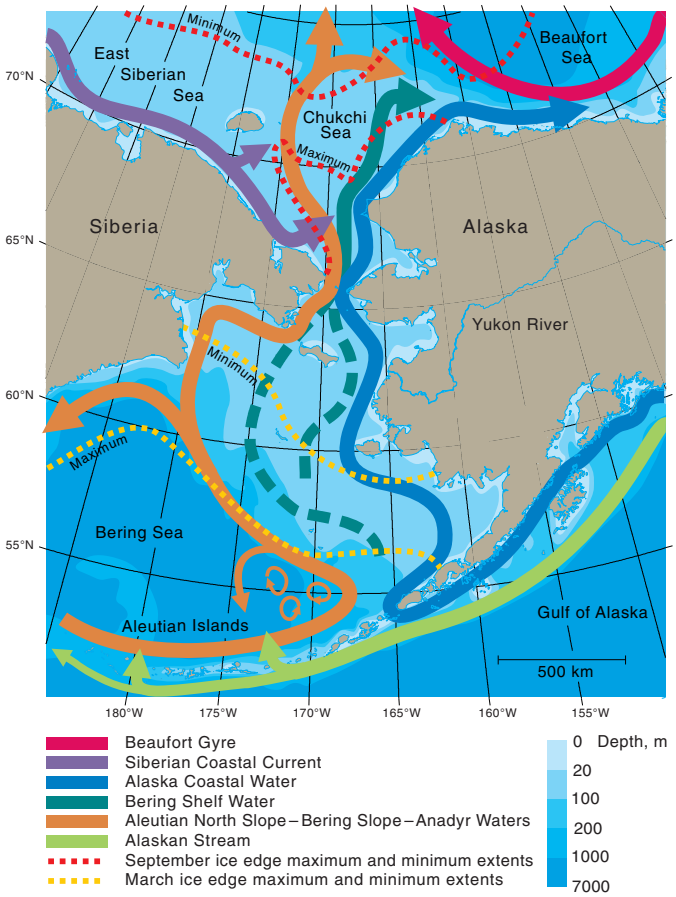

Figure 3·19. A schematic diagram of circulation in the Bering–Chukchi region of the Arctic. Flow over the Bering Sea shelf consists of waters from the Alaskan Stream, which feeds the Bering Slope Current, and fresher water from the Gulf of Alaska shelf which contributes to northward transport over the eastern Bering Shelf. In the Chukchi region, the Siberian Coastal Current transports fresh, cold water from the East Siberian Sea. Maximum (March) and minimum (September) ice coverage and their variations are shown by dashed lines (adapted from NOCD, 1986).

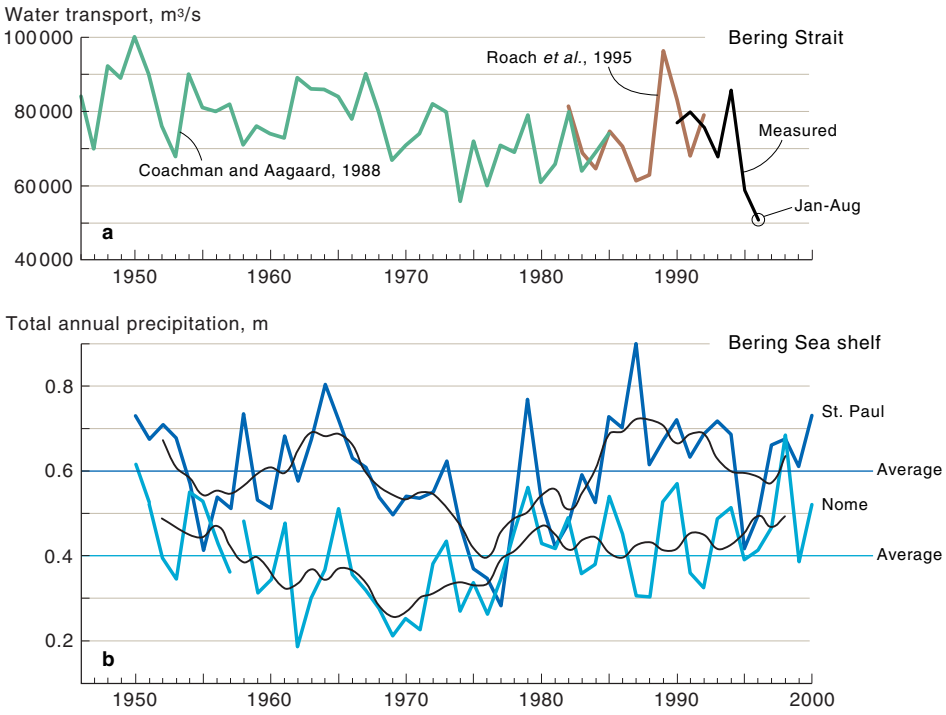

Figure 3·20. Time series of a) mean annual transport of water through Bering Strait from 1946 to 1996 (adapted from Roach *et al.*, 1995) and b) precipitation over the Bering Sea shelf based on annual precipitation measurements.

fresh Pacific inflow plays an important role in stratifying the upper 200 m of the Canada Basin (Carmack, 1986; Coachman and Aagaard, 1974) and can be detected as far away as Fram Strait (Jones and Anderson, 1986; Newton and Sotirin, 1997). The freshness of Pacific waters is supported by river runoff onto the Bering Sea shelf, relatively fresh inflow from the Gulf of Alaska (Royer, 1982), and greater precipitation than evaporation over the North Pacific Ocean (Warren, 1983).

Any alteration in the global thermohaline circulation (occurring at time scales of hundreds of years) will probably lead to change in the amount and composition of water passing through Bering Strait (see, for example, Wijffels *et al.*, 1992). Interannual and decadal scale variations in water transport, which may account for 40% of the variation in the long-term mean, are predominantly forced by the regional winds (Figure 3·20 a) whose strength and direction depends on the intensity and position of the Aleutian Low (Figure 3·2). Variation on the decadal scale (water transport was 15% lower during 1973 to 1996 than during 1946 to 1972) has also been inferred from wind records (Figure 3·20 a).

Upwelled Pacific waters support one of the world's most productive ecosystems in the northern Bering–southern Chukchi Seas (Springer *et al.*, 1996; Walsh *et al.*, 1989), including both oceanic fauna and shelf fauna transported within the surface water flowing northward over the Bering shelf (Walsh *et al.*, 1989). These shelves also serve as an important migratory pathway and critical habitat for a rich and diverse group of marine mammals, birds, and fish that move between polar, temperate, and tropical waters (Ainley and DeMaster, 1990; Tynan and DeMaster, 1997).

Pacific waters are substantially modified over the shelves through exchanges with the land, atmosphere, seabed, ice cover, and through biogeochemical transformations within the water column. These processes, which vary tremendously on seasonal and interannual time scales, affect the fate and distribution of contaminants deposited in the northeast Pacific and the Bering and Chukchi Seas following atmospheric transport across the North Pacific from Asia (Li *et al.*, 2002; Macdonald and Bewers, 1996; Macdonald *et al.*, 2002b; Wilkening *et al.*, 2000).

Salinity variations of ~1 in the inflowing Pacific water (Roach *et al.*, 1995) correspond to a range in injection depth into the Arctic Ocean halocline of 80 m or more, which is significant in determining whether, or how, contaminants from the Pacific Ocean might enter Arctic Ocean food webs. Salinity variation over the Bering shelf is controlled partly by sea-ice production and melting and partly by net precipitation, this latter process being, itself, an important pathway for some contaminants (e.g., Li *et al.*, 2002). Precipitation exhibits interannual variations that are as high as 50% of the long-term average, and also decadal or longer cycles with, for example, the 1960s to 1970s being relatively dry (Figure 3·20 b).

The ice edge in the Bering–Chukchi Seas migrates annually as much as 1700 km between the southern Bering shelf in winter (lowest dashed line on Figure 3·19) to the northern Chukchi shelf in summer (highest dashed line on Figure 3·19) with interannual variability of as much as 400 km (Niebauer, 1998; Walsh and Johnson, 1979). Variability in ice cover is governed by the strength and position of the Aleutian Low and East Siberian High (Figure 3·2) which influence storm tracks across the North Pacific (Niebauer, 1998; Overland and Pease, 1982). In the Chukchi Sea, the summer and autumn ice edge position can vary by as much as 200 km from its seasonally adjusted mean location (NOCD, 1986), with this variability associated with wind anomalies (Maslanik *et al.*, 1999). A significant decrease in ice cover over the Bering Sea appears to have coincided with a shift in the late 1970s from the 'cold' phase of the Pacific Decadal Oscillation (PDO) (Figure 3·16 b) to its 'warm' phase (Figure 3·21, and see also Macklin, 2001; Niebauer, 1998).

In the Bering Strait, late summer and autumn water temperatures can exceed 7°C, with substantial year-to-year variability. This variability, which affects ice melt on the Chukchi shelf and possibly the developmental rates

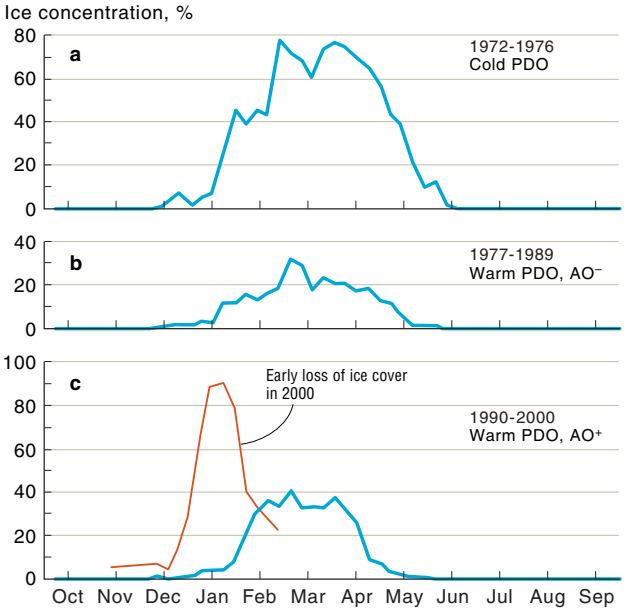

Figure 3·21. Ice concentration over the southeastern Bering Sea between 57°N and 58°N for a) a cold PDO phase (1972-76), b) a warm PDO and an AO⁻ phase (1977–89), and c) an intermediate regime with a warm PDO and an AO⁺ phase (1990-2000). Red line shows the early loss of ice cover in 2000 (adapted from Macklin, 2001).

of fish and other marine organisms, is related to change in large-scale advection and/or the summer radiation balance over the Bering Sea. Most of the ice in the Bering Sea grows and melts *in situ* such that brine rejection and melting strongly influence this shelf's water properties and stratification. During a spring and summer following heavy ice cover, the shelf water column is more strongly stratified and bottom temperatures are considerably lower (Azumaya and Ohtani, 1995), exerting a substantial influence on species composition and the distribution of marine fish, birds, and mammals (Wyllie-Echeverria and Ohtani, 1998; Wyllie-Echeverria and Wooster, 1998).

Ice cover also affects the carbon cycle. In spring, when nutrient concentrations are high, meltwater from sea ice stratifies the euphotic zone and initiates an ice edge bloom. Because zooplankton concentrations are low due to cold water temperatures, ungrazed phytoplankton settle to the seabed. In the absence of ice, water temperatures are warmer, phytoplankton are consumed by zooplankton, and less carbon is delivered to the benthos (Niebauer and Alexander, 1985).

Throughout spring and summer, waters of the central Bering shelf are strongly stratified owing both to the previous winter's ice history and to summer surface warming by solar radiation. Wind mixing is required to erode the stratification and replenish the euphotic zone with nutrients from depth. Springer (pers. comm., 2001.) shows that there is large interannual variability in wind speed, which provides the energy for mixing, and wind speeds over the Bering Sea seem to have diminished since the late 1970s by ~40%, coincident with the shift in the PDO from its cold phase to its warm phase.

The northward advection of warm water from the Bering Strait causes ice melt to occur earlier over the Chukchi shelf and freeze-up to be delayed, thereby prolonging the ice-free season relative to most other Arctic shelves (Martin and Drucker, 1997; Paquette and Bourke, 1981). Variability in flow through the Bering Strait (Figure 3·20 a), therefore, affects ice climate in the Chukchi Sea. Furthermore, variations in summer water properties and/or transport could lead to changes in the structure of shelf and slope food webs, insofar as the warm water provides suitable seasonal habitat for Pacific organisms advected into the Arctic (Melnikov *et al.*, 2002).

The volume and salinity of dense water formed over the Chukchi shelf each winter is affected by the extent of the seasonal retreat and advance of sea ice, which is variable and may change with warming. A thin ice cover in the autumn promotes early cooling of shelf waters, new ice growth, and the formation of multiple high-salinity water mass modes (Figure 3·22 a), whereas thick ice cover delays cooling, inhibits new ice growth, and leads to low-salinity water mass modes (Figure 3·22 b). These water masses then exchange differently with the interior ocean.

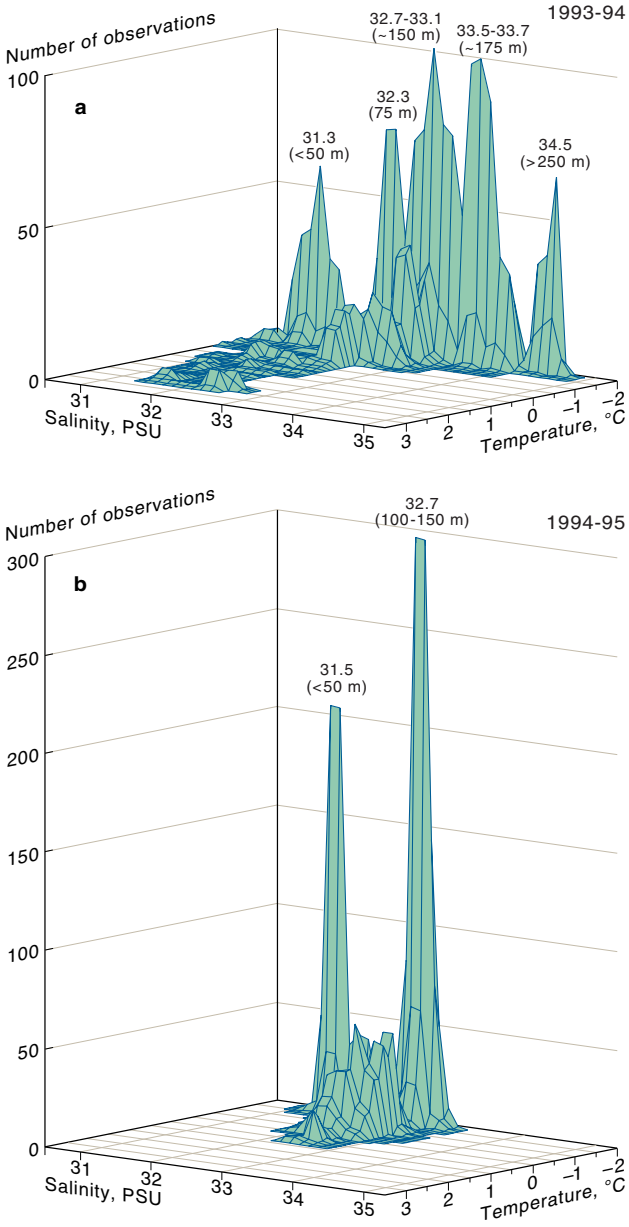

Figure 3·22. Water properties (T, S) on the northeastern Chukchi shelf in a) 1993-1994, with light autumn ice cover, large polynyas in winter, and multiple high-salinity water mass modes; and b) 1994-1999, with heavy autumn ice cover, small polynyas, and low-salinity water mass modes. The depths in parentheses refer to the depth to which each mode would sink along the continental slope if no mixing occurred.

The Siberian Coastal Current (SCC; Figure 3·19), flowing eastward along the Siberian coast for ~2000 km, also contributes low-salinity water to the Chukchi shelf (Coachman *et al.*, 1975; Weingartner *et al.*, 1999). From its probable origin in the western East Siberian Sea, the SCC draws water from as far as the Laptev Sea including ice melt and river discharges from the Lena, Indigirka, Kolyma and other Russian rivers along the way (Figure 1·1, Proshutinsky *et al.*, 1995). Zooplankton aggregate along the front between the SCC and the warmer, saltier water flowing through Bering Strait, providing important feeding opportunities for bowhead whales (*Balaena mysticetus*) in the western Chukchi Sea (Moore *et al.*, 1995) and potentially a critical pathway for contaminants to enter the food web at that point. Although transport in the SCC is relatively small (~3000 km^3/yr), its low-salinity waters enriched by runoff from Russian rivers could substantially dilute the inflow through Bering Strait and influence the distribution of Pacific waters in the Arctic Ocean.

The SCC exemplifies several ways in which changes in Arctic coastal currents can affect the transport and dispersal of contaminants (dissolved or sediment-bound). Firstly, coastal currents have large seasonal variability because 75 to 90% of the river discharge occurs within a three-month summer period. Change in the timing or amount of freshwater inflow will be expressed in the seasonal structure of the shelf circulation (Omstedt *et al.*, 1994). Secondly, coastal currents provide a vector to transport contaminants along a vast shoreline that can be dramatically altered by windfield change accompanying the AO (see Figure 3·14, section 3.5.2.1 and Guay *et al.*, 2001; Johnson and Polyakov, 2001; Weingartner *et al.*, 1999). Thirdly, river ice breakup typically occurs before the landfast ice melts with the result that little mixing initially occurs between the river water and the ambient shelf water and the inner shelf becomes strongly stratified (Ingram, 1981; Macdonald *et al.*, 1995). After the landfast ice melts, plume behaviour depends critically on the time-integrated effects of the wind velocity. Under 'upwelling' favourable winds, freshwater plumes can easily spread to interior basins carrying contaminants far beyond the shelf break (Macdonald *et al.*, 1999a). Under 'downwelling' favourable winds coastal currents are more likely to be formed (Melling, 1993; Weingartner *et al.*, 1999). Lastly, the dispersal and storage of freshwater over Arctic shelves in late summer set limits on the density of water that can be formed by sea-ice production the following winter (Macdonald, 2000; Melling, 1996; Weingartner *et al.*, 1999).

3.6.3. The Canadian Arctic Archipelago

The Canadian Arctic Archipelago provides one of the important outlets for Arctic Ocean surface water (Figure 1·1). Therefore, both changes in Arctic Ocean surface water contaminant burdens and changes in the source of water flowing out through the Archipelago have the potential to alter contaminant concentrations within the Archipelago's channels. There are few data with which to evaluate how seawater within the Archipelago channels responds to the AO. However, changes in distribution of surface water properties (Figures 3·13 and 3·14) and ice drift trajectories (Figure 3·9) in the Arctic Ocean itself, together with non-uniform spatial distribution of properties including river water and contaminants (e.g., see Carmack *et al.*, 1997; Guay and Falkner, 1997) should indicate the potential for upstream basin changes to be recorded as variable contaminant loadings in water flowing through the Archipelago. Furthermore, bowhead whale remains and driftwood on Archipelago shores suggest that ice-drift trajectories and ice cover have both varied greatly over time (Dyke *et al.*, 1996b, 1997; Dyke and Savelle, 2000, 2001) implying that the Canadian Arctic Archipelago is sensitive to rapid and dramatic change.

3.6.4. Hudson Bay

Hudson Bay is a large, shallow, semi-enclosed sea strongly influenced by seasonal runoff. The annual discharge (710 km^3/yr) is equivalent to a freshwater yield of about 65 cm (Prinsenberg, 1991). Presently, this sea exhibits a complete cryogenic cycle with summer (August-October) being ice free and winter fully ice covered. Climate models suggest that a doubling of CO_2 may lead to the virtual disappearance of ice from Hudson Bay thereby raising winter air temperatures and leading to the thawing of permafrost in adjacent land areas (Gough and Wolfe, 2001). These same models predict that the complete loss of ice will be preceded by years exhibiting earlier break-up and later freeze-up. According to Stirling and co-workers (1999), some of these projected changes may already be occurring (Figure 3·23), putting considerable stress on the western Hudson Bay polar bear (*Ursus maritimus*) population.

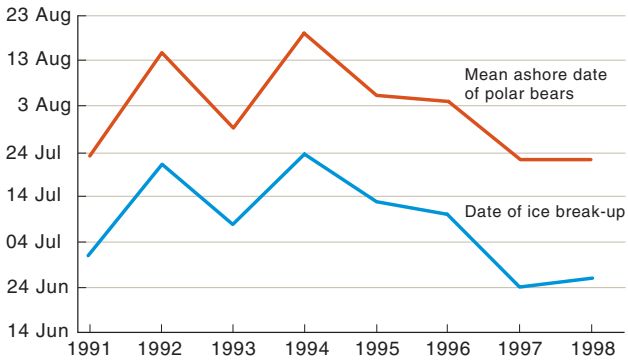

Figure 3·23. Temporal trends in the date of ice break up and the mean date at which polar bears return to shore in Hudson Bay. The break-up dates refer to that region in Hudson Bay where satellite-tagged female polar bears spent at least 90% of their time (modified from Stirling *et al.*, 1999).

The hydrological cycle of Hudson Bay has been strongly altered through immense damming projects in the drainage basin, leading to an increase in winter runoff to Hudson Bay of over 50% (Prinsenberg, 1991). Not only do such changes have an impact on stratification and hence nutrient cycling in this sea, but newly-flooded reservoirs are well known for their secondary effect of releasing mercury to downstream aquatic environments (Bodaly and Johnston, 1992).

Due to its southern location, Hudson Bay is clearly in the vanguard of Arctic change and is, therefore, a vital region to collect time series. According to Figure 3·3 a, Hudson Bay lies on a divide between warming and cooling. Regional temperature maps and other evidence

(Gilchrist and Robertson, 2000; Skinner *et al.*, 1998) confirm that between 1950 and 1990, the western side has warmed at about the same rate as the eastern side has cooled. In agreement with this observation, bears on the eastern side of Hudson Bay do not show the same pattern of weight loss as the bears on the western side (Stirling *et al.*, 1999), further emphasizing the importance of this region as a laboratory to study detailed consequences of change by contrast.

3.6.5. Baffin Bay, Davis Strait and the Labrador Sea

Baffin Bay, Davis Strait and the Labrador Sea occupy a unique position in that they may receive contaminants both from ice and water that exit the Arctic Ocean in the East Greenland Current and also from water and ice passing through the Canadian Arctic Archipelago. Change, therefore, can be produced by variation of contaminant composition within either of these sources or by altering their relative strength and the strength of direct exchange with the atmosphere. Furthermore, decadal-scale modulation probably differs for the various sources, with the AO perhaps influencing Archipelago through-flow or Fram Strait outflow, whereas the ice cover in Baffin Bay is more closely associated with the Southern Oscillation (Newell, 1996). In agreement with spatial temperature patterns (Figure 3·3 a), whereas ice season has been getting shorter within the Arctic Ocean and its marginal seas, Davis Strait and the Labrador Sea have recently exhibited an increase in the length of ice season (Parkinson, 1992).

Long-period cycles in the ice climate of this region (50 years or more) appear to have had dire consequences for both terrestrial and marine biological populations – including humans (Vibe, 1967). Like Hudson Bay, this appears to be an important region to study in the context of contaminants (Fisk *et al.*, 2001a), biogeographical variation (Johns *et al.*, 2001) and the impact of change on humans (Woollett *et al.*, 2000). Furthermore, surface water from Baffin Bay is exported to the south via Davis Strait to feed the Labrador Current (along with outflow from Hudson Bay and the Canadian Arctic Archipelago – Figure 1·1). Baffin Bay, therefore, provides a region of transition which can export change in ocean properties (freshwater, contaminants, biota) to the Northwest Atlantic.

3.7. Lake and river ice

Arctic lakes and rivers are likely to provide sensitive sentinels of climate change in their freeze, melt and hydrological cycles (Vörösmarty *et al.*, 2001). Whereas there appear to be no studies showing a relationship between freshwater ice cover and the AO, significant trends in these properties over the past 150 to nearly 300 years have been demonstrated (Magnuson *et al.*, 2000; Semiletov *et al.*, 2000). During the period between 1846 and 1995, there has been a mean delay of 5.8 days per century for freeze-up and a corresponding 6.5 days per century advance in break-up. This change in the freeze/melt cycle implies increasing temperatures of about 1.2°C per century.

Most Arctic lakes receive their contaminant burdens from the atmosphere, with the catchment area acting as a receptor through snow fall in winter, and a conveyor through runoff in spring. From a very limited set of studies, it appears that Arctic lakes presently retain only a small fraction of contaminant inputs because the main runoff pulse, which precedes lake turnover and peak primary production, simply traverses the lake surface under the ice (Macdonald *et al.*, 2000a). With lakes exhibiting more temperate characteristics, the coupling of runoff with lake mixing and primary production will change, probably allowing lakes to capture more of the inflowing contaminant burden. In particular, the potential for snow surfaces to enhance contaminant fugacity in lake settings is extremely large (Macdonald *et al.*, 2002c). However, quantitative measurements of contaminant–snow interactions are required because the significance of snow in contaminant cycling cannot be projected simply from hydrological measurements.

3.8. Permafrost

Permafrost underlies about 25% of the land in the Northern Hemisphere, including large areas of Canada, Russia, China and Alaska (Figure 3·24, Zhang *et al.*, 1999). Permafrost can also be found in sediments of the continental shelves (not shown). Especially vulnerable to change are regions of discontinuous permafrost which include large parts of northern Canada, Alaska and Russia. The IPCC (2002) suggests that permafrost area could be reduced by 12 to 22% by 2100 with perhaps as much as half of the present-day Canadian permafrost disappearing.

In regions of permafrost, the active layer of soil, typically limited to the top 1 m, supports almost all of the biological processes. The loss of permafrost, therefore, alters these biological processes, including affecting the kind of vegetation that can grow, and changes the way soil interacts with the hydrological cycle (Osterkamp *et al.*, 2000; Vörösmarty *et al.*, 2001) both of which have consequences for contaminant transport. In particular, thawing frozen ground releases sediment, nutrients, and

Figure 3·24. The distribution of permafrost in northern landmasses (source: IPA, 2001).

organic carbon which then enter ground water, rivers and lakes to impact upon biological cycles (see, for example, the studies done in the Mackenzie Basin; Cohen, 1997a). The observed thawing trends in Alaska and Russia, but not in northeastern Canada, appear to match the observed trends in SAT (see Figure 3·3 a and Rigor *et al.*, 2000). Accelerated permafrost degradation during the 1990s can probably be ascribed at least partly to the AO (Morison *et al.*, 2000) with, for example, the advection of warm air into the Russian Arctic during strong AO^+ conditions contributing to thawing in that region.

In addition to the changes in biogeochemical pathways that will accompany permafrost degradation, there will also be the widespread problem of re-mobilization of contaminants (see, for example, conference proceedings dedicated to the issue of contaminants in frozen ground; Anon., 2001a,b). Historical disposal of waste substances has occurred in the form of sewage lagoons, dump sites at DEW line sites (Distant Early Warning Line; a chain of defense radar stations, many now abandoned, along 66°N in Canada, also extending into Alaska and Greenland), solid waste dumps in small Arctic communities, mine tailings, and oil drilling sumps. A large component of the containment strategy for these sites is the presence of permafrost. With permafrost degradation, landfills can become washed directly into rivers or the ocean, or runoff can leach contaminants into local groundwater. In locations such as river deltas and coastal plains, low relief may provide a shortcut between such waste sites and drinking water.

3.9. Glacial ice

Most Arctic glaciers have experienced net loss in ice mass over the past few decades (Dowdeswell *et al.*, 1997). The Greenland ice mass appears presently (1994-1999) to be decreasing, predominantly at lower elevations, at a rate of about 51 km^3/yr (Krabill *et al.*, 2000). Data also point clearly to loss of ice mass for small glaciers in the Arctic during the interval between 1961 and 1993 (Arendt *et al.*, 2002; Dyurgerov and Meier, 1997; Serreze *et al.*, 2000). Since about 1960, glacial melt-back in the Canadian Arctic alone (Figure 3·25) is estimated at over 800 km^3 – around half of the melt-back estimated for the whole Arctic (Dyurgerov and Meier, 1997). In conformity with the strong AO^+ conditions of the early 1990s, the loss of glacial ice mass in the Canadian Arctic Archipelago was exceptionally strong in the early 1990s, amounting to 390 km^3 (Figure 3·25).

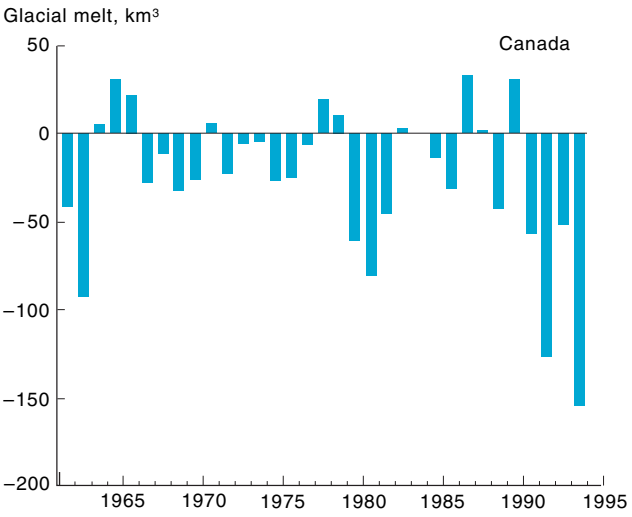

Figure 3·25. The loss of glacial ice mass, km^3/yr, in the Canadian Arctic between 1961 and 1993 based on data compiled by Serreze *et al.* (2000) and Dyurgerov and Meier (1997).

Glaciers may act as long-term reservoirs, sequestering and preserving airborne contaminants during peak emission years (1950-1970) later to release them during periods of melt-back (Blais *et al.*, 1998).

Chapter 4

Biological Responses to Climate Change

An exhaustive consideration of the biological consequences of the kinds of physical change that are predicted for the Arctic is not feasible at this time, nor is it warranted for identifying how biological changes might effect major change in contaminant pathways. Here, ecosystem changes are highlighted that appear to have a strong potential to alter the exposure of Arctic biota to contaminants or to alter their resilience to that exposure. There is general agreement that the kinds of changes discussed below have, or will, take place in the Arctic but much less agreement concerning their probable scope and timing. The primary intent, therefore, is to provide examples of processes that ought to be included explicitly in models and to help focus future attention on biological connections of significance to contaminants.

Whereas aquatic food webs in the Arctic exhibit endemic contamination from biomagnifying chemicals, terrestrial food webs are among the world's cleanest (AMAP 2003b; de March *et al.*, 1998). Therefore, apex feeders that adapt to change by switching between land-based and aquatic food webs have a particularly large potential to change their exposure to contaminants such as organochlorine compounds (OCs) and mercury. Humans probably provide the best example of such flexibility but other animals (e.g., Arctic foxes (*Alopex lagopus*) and grizzly bears (*Ursus arctos*)) can also adjust diet to opportunity.

4.1. Terrestrial systems

In this report terrestrial systems are defined as including forests, grasslands, tundra, agricultural crops, and soils. Surface-air exchange between airborne contaminants and terrestrial systems is important in the overall fate and long-range transport of chemicals, especially for the semi-volatile chemicals which are split between the gaseous and condensed states. As a result of their high organic content, terrestrial phases (e.g., soils, forests, grasslands) act as reservoirs for many persistent organic pollutants (POPs) (Simonich and Hites, 1994), particularly polychlorinated biphenyls (PCBs), DDT, hexachlorohexane (HCH) and chlorobenzenes (AMAP, 2003b). Air-surface exchange of POPs into terrestrial phases is a dynamic process that controls air burdens of chemicals. Thus any change in the extent of vegetation cover associated with global warming will have implications for contaminant fate and transport. Wania and McLachlan (2001) have shown that forests have a unique ability to mitigate atmospheric concentrations of OCs by 'pumping' chemicals from the atmosphere into foliage and thence to a long-term reservoir in forest soil. This process is likely to be most important for OC compounds with log K_{OA} of ~9 to 10 and log K_{AW} ~ –2 to –3 (where K_{OA} and K_{AW} are octanol–air and air–water partition coefficients – see Wania, 2001). Because these key properties are strongly temperature-dependent (see section 6.3.4. for greater detail) even a small change in climate may alter the dynamics of this process and thus the cycling of contaminants. Terrestrial vegetation also has an indirect impact on contaminants by altering snow accumulation and soil temperature (Sturm *et al.*, 2001).

Arctic terrestrial animals have provided some of the clearest examples of large cycles in their populations (Krebs *et al.*, 2001; Predavec *et al.*, 2001) and it is against this natural background variability that the effects of global change will have to be evaluated. Warmer winter temperatures promote the growth of woody shrubs and encourage the northward migration of the tree line (MacDonald *et al.*, 1993; Serreze *et al.*, 2000; Vörösmarty *et al.*, 2001). Although the advance of the tree line (estimated at 100 km per °C warming (IPCC, 2002)) might be expected to occur slowly over time scales measured in centuries, the particular sensitivity of tundra to water-table fluctuations and permafrost melt could produce widespread alteration in ground cover more rapidly with, for example, the replacement of tundra by vascular plants (Gorham, 1991; Rouse *et al.*, 1997; Weller and Lange, 1999). Gradual climate change can affect species distribution, abundance, morphology, behavior, population diet and community structure (Easterling *et al.*, 2000; Predavec *et al.*, 2001). Although there appears to be no compelling evidence of recent large change in the Arctic tundra ecosystem, models suggest that tundra may decrease to one third of its present size (Everett and Fitzharris, 1998).

Warmer summer temperatures are likely to promote forest fires which will be accompanied by direct emissions of polyaromatic hydrocarbons (PAHs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and other POPs produced by combustion (see for example, Gribble, 1994; Yunker *et al.*, 2002). Forest fires will also damage terrestrial soils leading to erosion and an increased release of organic carbon, which in turn affect aquatic systems.

4.2. Aquatic systems

4.2.1. Lakes, rivers and estuaries

The changes in snow and ice cover and in the hydrological cycle will alter the light and nutrient climate of freshwater systems. These changes together with loss of permafrost, which will enhance the supply of nutrients and particulates to lakes, will increase aquatic productivity and particle flux (Douglas *et al.*, 1994; McDonald *et al.*, 1996; Schindler, 1997). Although the spring bloom will probably advance with early loss of ice cover, hydrological processes in a lake's drainage basin will probably also advance. Increased summer temperatures will disadvantage fish such as trout (*Salmo* spp.) and grayling (*Thymallus arcticus*) whereas winter temperature increase may enhance microbial decomposition. Shifts in the seasonal light/temperature cycle may also advantage or disadvantage species lower in the food web including phytoplankton, zooplankton and insects. Change in water level will have obvious effects on important fish

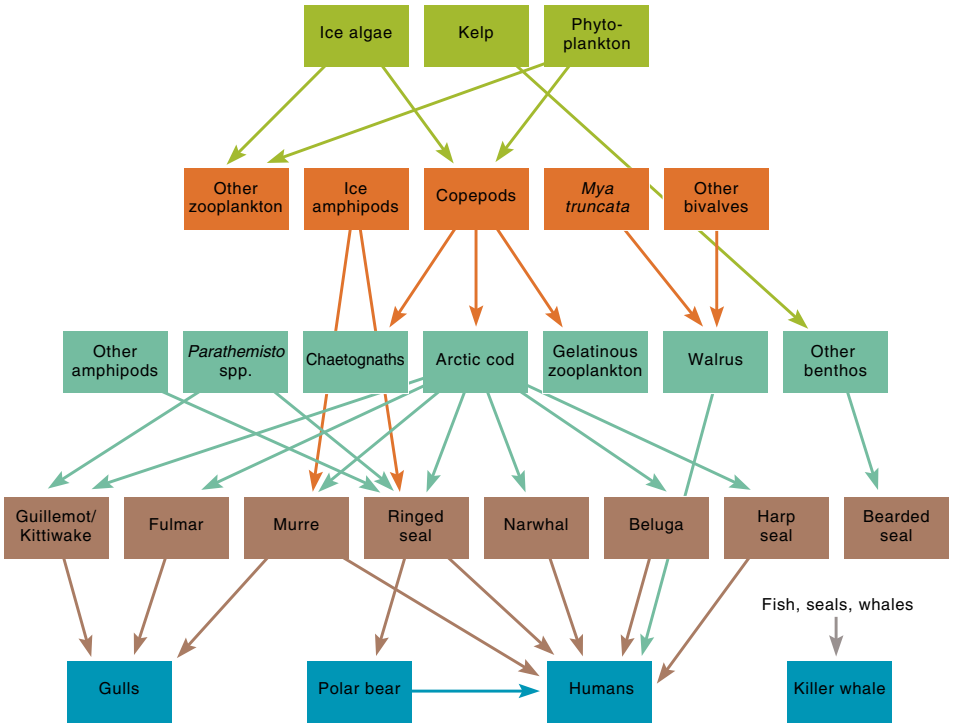

Figure 4·1. A simplified schematic diagram showing the marine food web (based on Welch *et al.*, 1992).

stocks, especially species dependent on small refugia for over-wintering (Hammar, 1989) or species dependant on freshwater coastal corridors for their life cycle; the Arctic cisco (*Coregonus autumnalis*) provides a relevant example of the latter (see Gallaway *et al.*, 1983). Warming and loss of nearshore or estuarine ice in the Beaufort Sea may eliminate indigenous fish which are then replaced by anadromous fish from the Pacific Ocean (see Babaluk *et al.*, 2000). Although warming is likely to result in widely-distributed shifts in zoogeographic distributions that have the potential to affect every step in the freshwater food chain, prediction will probably founder on 'counter-intuitive' surprises (Schindler, 1997).

4.2.2. The ocean

The effect of ice on Arctic marine ecosystems has long been understood by those who harvest the sea (Bockstoce, 1986; McGhee, 1996; Scoresby, 1969; Vibe, 1967). Change in ice climate, therefore, has a large potential to modify marine ecosystems, either through a *bottom-up* reorganization of the food web by altering the nutrient or light cycle, or a *top-down* reorganization by altering critical habitat for higher trophic levels (see, for example, Parsons, 1992). Any reorganization that changes the number of trophic levels in the food web or alters the flow of carbon between, for example, pelagic and benthic food webs would have particular significance for contaminants that biomagnify, such as mercury and the OCs; the complexity of the interaction between ice and aquatic ecosystems provides much scope for alterations in contaminant pathways (Figure 4·1). Arctic and sub-arctic marine ecosystems are also altered by ocean climate changes such as regime shifts involving the displacement of water masses and associated populations or temperature change (Figure 4·2; Dippner and Ottersen, 2001; Hare and Mantua, 2000; Helland-Hansen and Nansen, 1909; Hunt *et al.*, 1999; Loeng, 2001; Saar, 2000; Sakshaug *et al.*, 1991, 1994).

There are too many examples of how ice climate variation can affect ecosystem structure to list them all (see for example Sakshaug and Slagstad, 1992; Tynan and DeMaster, 1997) and it is not likely that all changes that have occurred in Arctic systems have been observed. The thickness and distribution of ice can influence the amount of organic carbon produced, the types of algae that produce it, and connections between the algal production and communities in the water column or sediments (Niebauer and Alexander, 1985). Ice controls wind mixing and light penetration especially when covered with snow, and it may also support upwelling at the ice edge but suppress upwelling beneath the ice. Through its annual cycle, ice formation decreases stratification in winter but increases stratification when the ice melts in spring. These physical factors impact upon the nutrient supply to surface water, the light climate, and the water stability which together control primary production. Furthermore, mats of algae that grow on the bottom of the ice support an epontic food web that ultimately feeds Arctic cod (*Boreogadus saida*), ringed seals (*Phoca hispida*) and polar bears (*Ursus maritimus*) or, alternatively, by being shed from melting ice in spring, support a benthic food web that feeds molluscs, walrus (*Odobenus rosmarus*), bearded seals (*Erignathus barbatus*) and king eiders (*Somateria spectabilis*). Similarly, primary production within the water column may be partially grazed to support a pelagic food web, or descend ungrazed and, together with fecal pellets and zooplankton carcasses, feed the benthos (Grebmeier and Dunton, 2000). The bifurcation between pelagic and benthic food webs is strongly influenced by the distribution of ice and its impact on nutrient and light climates. These processes, which have great potential to alter the timing and amounts of vertical particle flux in the ocean, are likely to have a greater impact on the sequestering of POPs into the Arctic Ocean than air-sea exchange or the so-called 'cold-condensation' effect. Dachs *et al.* (2002) show that in mid-latitudes, sinking particulate matter, which is

the dominant export pathway for POPs from the ocean surface layer, drives deposition at the ocean surface.

Shifts in benthic species distribution due to temperature, carbon flux or other climate-related change have the potential to alter completely the coupling between sediments and bottom water. In one well-documented example from a temperate region, the invasion of Echiura (*Listriolobus pelodes*) into coastal benthic communities off California, for as yet unknown reasons, resulted in aerated and biomixed sediments that reduced the evidence of wastewater impacts regionally (Stull *et al.*, 1986).

4.2.2.1 Bottom-up trophic change

The projected loss of ice for the Arctic Ocean, particularly over the shelves, intuitively should increase primary production in the marginal seas through enhanced mixing, light penetration and upwelling. In other words, Arctic shelves would begin to look more 'temperate'. Greater new production implies greater particle flux and greater secondary production, but the complexity of marine ecosystems should forewarn of possible surprises. Massive blooms of jellyfish were observed in the Bering Sea during in the 1990s (Brodeur *et al.*, 1999; Hunt *et al.*, 1999) and their emergence was ascribed to sea-surface temperature increase and loss of ice cover – the same two key changes poised over the Arctic Ocean.

Parsons (1979) has drawn attention to the fundamental ecological differences between western seaboards in the Northern Hemisphere, where coastal water exhibits divergence and upwelling, and eastern seaboards which are convergent. The former have been of greater commercial interest but are also characterized by jellyfish (Parsons, 1979). The Arctic Oscillation (AO) does not cause reversal of large-scale wind circulation but does produce more divergent Arctic Ocean margins under AO⁻/anticyclonic conditions and less divergent margins under AO⁺/cyclonic conditions. The inherently noisy events of coastal upwelling and downwelling could then act together with the AO in a form of 'stochastic resonance' (Rahmstorf and Alley, 2002) to enhance upwelling during AO⁻ conditions. This enhanced upwelling might then have the capacity to produce large-scale modal shifts in shelf ecosystems and their commercial potential. Changes in ocean climate, such as those associated with the AO/NAO, have long been known to affect fisheries in sub-polar seas either directly through water property changes (T, S) or indirectly through changes in community structure (Hare and Mantua, 2000; Klyashtorin, 1998; Marteinsdottir and Thorarinsson, 1998).

A dramatic example of large-scale, bottom-up biological change was witnessed during the SHEBA drift across the Beaufort and Chukchi Seas in 1997 to 1998 (Melnikov *et al.*, 2002). Compared to Soviet observations from drifting stations that passed over the same region 20 years earlier, there was a marked decrease in large diatoms in the water column and microfauna within the ice. The freshening and strong stratification of the surface water, due to river discharge diverted into the basin under the strong AO⁺ conditions of the early 1990s, reduced the supply of nutrients from below, and promoted species more typical of freshwater ecosystems. Consequentially, there was a high proportion of recycled production and less new production. The loss of relatively large diatoms could reduce the size of herbivores, potentially inserting an extra 'small-carnivore' step at the bottom of the food web which would increase the number of trophic levels. Because biomagnification of OCs is often exponential (Fisk *et al.*, 2001a), slightly higher concentrations at low trophic levels (e.g., copepods) can have a large impact on apex feeders. Stratification, which is altered at the basin scale under AO/NAO shifts, affects plankton composition and vertical flux dramatically as evident from studies in the Barents Sea (Wassmann, 2001). For example, Wassmann *et al.* (1990) showed that algal blooms by *Phaeocystis* sp. along the Greenland coast and in the Barents Sea tend not to get grazed resulting in a large transfer of organic carbon to the benthos. Climate change in the form of either loss of ice cover or increase in stratification has the potential to alter the quantity of available food and to redistribute its flow between epontic, pelagic and benthic habitats.

The Bering Sea provides another outstanding example of recent change from the bottom-up permeating an entire ecosystem. In view of the Bering Sea's vulnerability to airborne contaminants from Asia (Bailey *et al.*, 2000; Li *et al.*, 2002), it is particularly regrettable that the observations of ecosystem change since the 1970s were not matched by contaminant pathway studies. Evidence from stable isotope records in bowhead whale (*Balaena mysticetus*) baleen suggests that the carrying capacity of the Bering Sea ecosystem began to decline in the mid 1970s (Figure 4·2a, Schell, 2000). This change may relate to a larger picture of change throughout the North Pacific (Hare and Mantua, 2000) and, in particu-

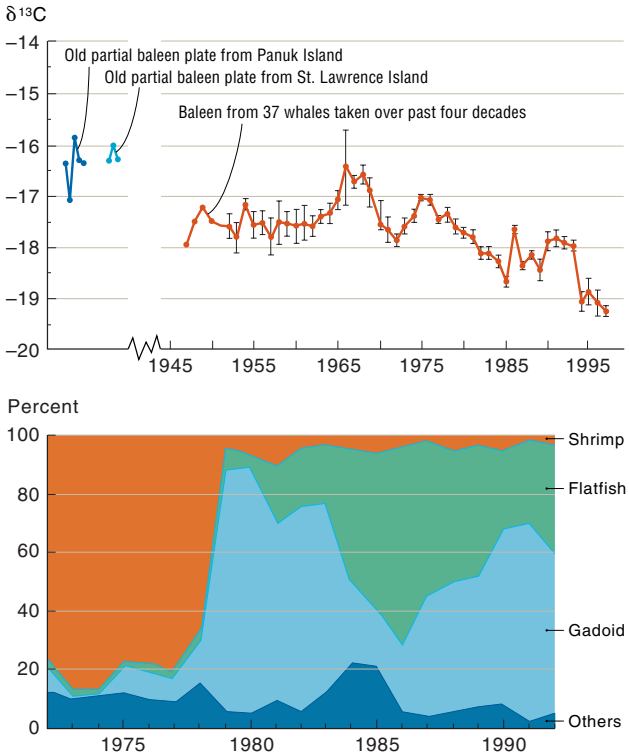

Figure 4·2. Examples of significant change within Arctic and subarctic ecosystems. This figure illustrates a) a change in the food web structure probably commencing during the 1970s as reflected by a decrease in the $\delta^{13}C$ of baleen from bowhead whales (Schell, 2000) and b) significant fish population changes in the Gulf of Alaska since 1970 (modified from Anderson and Piatt, 1999).

lar, to the switch in the Pacific Decadal Oscillation (PDO) from cold to warm phase in the mid 1970s. The change in regime rapidly permeated the entire ecosystem of the Bering Sea altering fish community structure (shrimp and crab populations declined while pollock, cod and flatfish populations increased significantly Figure 4·2 b), and seabird and mammal populations (Springer, 1998). More recently, blooms of small phytoplankton (*Emiliania huxleyi*) were observed in 1997 and 1998 (Saar, 2000). Because these phytoplankton are smaller than the diatoms that typically bloom in the Bering Sea, they were grazed by smaller copepods instead of larger euphausids which in turn probably led to die-offs of the short-tailed shearwaters (*Puffinus tenuirostris)* that feed on the latter (Stockwell *et al.*, 1999). Similarly, the alteration of primary production both in quantity and distribution probably decreased food availability for fish, whale, seal and walrus populations forcing die-offs, migration or redistribution throughout the food web (Botsford *et al.*, 1997; Grebmeier and Cooper, 1995; Grebmeier and Dunton, 2000; Hare and Mantua, 2000; Rugh *et al.*, 1999; Stabeno and Overland, 2001). Large as these ecosystem changes appear to have been, they may pale in comparison to the natural fluctuations that have occurred during the past two millennia (Finney *et al.*, 2002). Furthermore, these long-term proxy data provide a strong warning that relationships between biological populations and physical forcing established from short observational records may not hold up over a longer period when other non-linear factors may have a chance to operate (initial conditions, or other cyclical forcing, for example). Clearly, the dramatic changes in the Bering Sea system could spill over into the Chukchi Sea, and the decline of Bering inflow by ~15% since the 1940s (Figure 3·20 a) suggests a matched decline in new and advected production in the Chukchi Sea simply due to reduced nutrient and organic carbon supply.

In the Barents and Nordic Seas it has long been recognized that fish populations respond to climate variability (Helland-Hansen and Nansen, 1909). Indeed, the distribution of capelin (*Mallotus villosus*), the single most important food species for Arcto-Norwegian cod, is known to vary from year to year dependent on the inflow of Atlantic water (Sakshaug *et al.*, 1994). Fluctuations in large- and regional-scale atmospheric pressure conditions affect winds and upper ocean currents (Figures 3·2 and 3·17), modify water temperature, alter drift patterns of fish larvae, and change availability of prey items. Mixing during summer alters the nutrient cycle and the coupling between primary production and benthos (Peinert *et al.*, 2001; Wassmann, 2001). Details are important. For example, while long and unrestricted larval drift is crucial for the Arcto-Norwegian and Icelandic component of cod stocks at West Greenland, larval retention on favourable banks is the key for recruitment to stocks residing in small and open systems (Ottersen, 1996). The 600 to 1200 km drift of Arcto-Norwegian cod larvae from spawning grounds to nursery grounds where they settle on the bottom provides much opportunity for interannual variation; pelagic juveniles in the Barents Sea exhibit a typical westerly distribution in some years while, in other years they distribute to the east (Ådlandsvik and Sundby, 1994). Ottersen and Sundby (1995) showed that southerly wind anomalies during the period of pelagic drift from the main spawning grounds in the Lofoten area in northern Norway to the nursery grounds in the Barents Sea leads to above average year-class strength. This was attributed in part to temperature and in part to added supply of zooplankton-rich water from the Norwegian Sea into the feeding areas of the Barents Sea.

For older fish, other factors may contribute to interannual variation. During periods of high abundance, fish density may cause the geographic range of Arcto-Norwegian cod to expand or shift. Temperature has been reported to cause displacement of Arcto-Norwegian cod – toward the east and north during warm periods and toward the south-western part of the Barents Sea in cold periods (Ottersen *et al.*, 1998). These shifts may not be caused by temperature itself but, rather, by temperature-induced changes in the distribution of prey organisms (Ottersen, 1996).

Loeng (2001) has discussed the types of change that may well occur in the Nordic seas should ocean temperature rise, as projected, by 1 to 2°C (Figure 4·3). In the Barents Sea the feeding area of capelin will be displaced to the northeast and the spawning ground may move eastward along the northern coast of Russia. Cod will distribute more toward the northeast partly because acceptable ocean temperature will be found there and partly because their main food item, capelin, will move in that direction. These displacements will put such stocks closer to contaminant sources in the eastern Barents Sea. In the Norwegian Sea, the Norwegian spring spawning herring (*Clupea harengus*) may return to the migration route they used prior to the mid 1960s, when ocean temperature around Iceland was over 1°C higher than today. Presently, adult herring over-winter in a Norwegian fjord before commencing their spawning migration along the Norwegian coast followed by a feeding migration into the Norwegian Sea. With temperature increase, the herring may over-winter after the feeding migration just east of Iceland as they did before 1965,

Figure 4·3. Possible changes in the distribution of selected fish species in the Nordic and Barents Seas resulting from an increase in sea temperature of 1 to 2°C (modified from Loeng, 2001).

and not return to the Norwegian fjord, completely altering their exposure to contaminants. New species may invade. Presently, mackerel (*Scomber scombrus*) is scarce along the coast of northern Norway, but with ocean warming might migrate as far as the Barents Sea. Blue whiting (*Micromesistius poutassou*), bluefin tuna (*Thunnus thynnus*) and sharks (Elasmobranchia) may also become more frequent visitors to this area.

4.2.2.2. **Top-down trophic change**

Ice-covered seas have a unique capacity for top-down trophic change. To understand and predict how the partial or complete loss of ice will impact upon the trophic structure requires a detailed understanding of how top predators take advantage of ice (Carmack and Macdonald, 2002; Lowry, 2000; Vibe, 1967). In an incisive review, Tynan and deMaster (1997) discuss how whales, walrus, seals, bears and cod, are likely to be affected by change in ice climate and show that their response to change depends on how 'plastic' their dependence on ice might be.

Change in the *landfast ice* may give the advantage to either seals or to bears (Carmack and Macdonald, 2002) with the result that Arctic cod would be subject to more, or less, predation, respectively. Walrus use *drifting ice* to haulout in winter because it provides better access to benthos, but they also use terrestrial haulouts in ice-free periods, perhaps with detrimental energy costs (Lowry, 2000; Tynan and DeMaster, 1997). In contrast, eiders (*Somateria* spp.) and other benthic-feeding birds prefer *open water* with a relatively shallow bottom (<50 m) (Dickson and Gilchrist, 2002; Grebmeier *et al.*, 1988; Suydam *et al.*, 2000). Loss of ice (landfast or drifting) in critical regions or at critical times of the year, or movement of the ice edge to deeper water where benthos can no longer be accessed, therefore, can mean a substantial rearrangement of the top of the food web advantaging some animals, disadvantaging others and possibly causing wholesale migration (Dyke *et al.*, 1996b, 1999; Dyke and Savelle, 2001; Fay, 1982; Lowry, 2000; Moore and Clarke, 1986; Tynan and DeMaster, 1997; Woollett *et al.*, 2000). With benthos not readily available, walrus might turn to predation on seals thereby raising their trophic position considerably (Muir *et al.*, 1999), or with absence of ice, killer whale (*Orcinus orca*) predation on bowhead whales might decimate their population leaving their prey (zooplankton) as food for something else. Early breakup in the Bering and Beaufort Seas during 1995 to 1998 probably led to the observed abandonment of seal pups in 1998 and the decline or starvation of walrus.

The Hudson Bay polar bear population provides perhaps one of the clearest warnings of the consequence of change. Polar bears rely on ringed seals for food, and ringed seals prefer landfast or stable first-year ice for pupping (Finley *et al.*, 1983; Stirling, 2002; Wiig *et al.*, 1999). The loss of landfast ice in spring, the loss of food supply for seals, or the inability of bears to access seals during the few critical weeks in spring when pupping occurs, means life or death and can produce large population shifts (Harwood *et al.*, 2000; Smith and Harwood, 2001; Stirling *et al.*, 1999). In Hudson Bay, bears probably accumulate most of their annual energy requirements during the few months of late spring prior to breakup when they can access older pre-weaning ringed-seal pups or naïve post-weaning pups – exactly the period of time that has seen recent dramatic change (Figure 3·23). Furthermore, permafrost is a critical habitat for bears because they dig maternity dens in frozen peat, and this habitat is threatened by warming or increased incidence of fire initiated by more frequent lightning strikes. In Hudson Bay, at the southern limit of their population, polar bears presently appear to be in a very precarious position (Stirling and Derocher, 1993; Stirling *et al.*, 1999).

Arctic cod is the most important forage fish in the Arctic Ocean food web (Figure 4·1; Bradstreet *et al.*, 1986; Tynan and DeMaster, 1997; Welch, 1995). The loss of ice, in either the marginal seas or, as projected by models, for the entire ocean (Figure 2·2b, Flato and Boer, 2001), would have a massive impact on the distribution and life history of Arctic cod and, therefore, on seals, beluga (*Delphinapterus leucas*) and birds who depend heavily on them. One thing is clear: the ice edge is an especially critical habitat for cod and marine mammals and it is this region that is most vulnerable to change.

Finally, climate change can alter the routes and destinations of migratory species. For example, under the AO^+ conditions of the early 1990s, Pacific salmon (*Oncorhynchus* spp.) began to enter Arctic rivers (Babaluk *et al.*, 2000). Similarly, bowhead whales and belugas range widely in search of food and their range varies enormously in time and space with changes in ice climate (Dyke *et al.*, 1996b; Dyke and Savelle, 2001; McGhee, 1996). Nor are long migrations limited to whales. Harp seals (*Phoca groenlandica*) of the Northwest Atlantic undergo 8000 km round trips to feed on Arctic cod in Baffin Bay (Finley *et al.*, 1990) and bird species migrate inordinately long distances often depending on critical areas along their migration pathways where they may ingest contaminants (see, for example, Braune *et al.*, 1999; Savinova *et al.*, 1995; Springer, 1998). The extent to which migratory species are able to adapt to potentially rapid changes in key staging areas may be of critical importance to their future (Carmack and Macdonald, 2002).

Chapter 5

The Effect of Climate Change on Human Activities

There are at least six very different ways in which climate change may lead to an alteration of contaminant pathways through a modification of human activities. First, people on the margins of the Arctic Ocean will make dietary choices, as they have always done, based on the availability of traditional country foods including terrestrial and marine animals (Krupnik, 2000; McGhee, 1996; Vibe, 1967). Second, a marginal sea that clears of ice for large portions of the year will encourage shipping, tourism, oil exploration and other industrial activities each of which brings with it associated contaminants. Furthermore, enhanced shipping increases the risk of introducing exotic species or diseases which then affect indigenous species. Third, the encroachment of commercial fisheries into the Arctic could alter the food web structure in oceans (Bockstoce, 1986; Parsons, 1992; Pauly *et al.*, 1998, 2001) and lakes (AMAP 2003b; de Graff and Mychasiw, 1994). Fourth, climate change may promote the spread of insect pests globally forcing some countries to re-introduce or increase the use of pesticides. Fifth, climate change toward conditions suitable for domestic crops may encourage further expansion of agriculture or silviculture within the Arctic drainage basin along with increased industry. Lastly, the various changes listed above will probably contribute to demographic shifts and population increases in northern regions, which will in turn lead to increased local releases of contaminants (for example from burning, power and fuel consumption, use of industrial or agricultural products).

It is well known that the dietary composition of the human 'food basket' (e.g., marine versus terrestrial foods, fat versus protein, older fish or seals versus younger fish or seals) controls the amounts and kinds of contaminants ingested (AMAP, 1998; Kinloch *et al.*, 1992; Van Oostdam *et al.*, 1999). Dietary changes can be forced by fluctuations in the populations of target species as discussed in chapter 4 (e.g., beluga (*Delphinapterus leucas*), bowhead whales (*Balaena mysticetus*), walrus (*Odobenus rosmarus*), seals, bears, birds, fish, caribou/reindeer (*Rangifer tarandus*), muskox (*Ovibos moschatus*)) or by changes in access to the species (early melt, permafrost degradation, open water, loss of multi-year ice, late freeze-up (see, for example, Fast and Berkes, 1998; Riedlinger, 2001)). Because the manner in which contaminants enter and concentrate in these two food webs is so different, the balance between terrestrial and aquatic food items in the food basket will be a pivotal point of change in exposure to biomagnifying contaminants.

With marginal seas clear of ice for long periods of the year, it is inevitable that the Arctic will become a favoured shipping route between Europe, Asia and North America either via the Northern Sea Route (Russia) or the Canadian Arctic Archipelago. Shipping brings with it specific, well-known contaminants such as hydrocarbons and marine antifoulants (e.g., tributyltin) and, potentially, non-indigenous species in ballast water. The major concern, however, is likely to come from accelerated oil exploration and development on the Arctic continental shelves of North America and Eurasia (Bakke *et al.*, 1998).

Outside the Arctic, global warming and alteration of hydrological cycles will probably cause insects and other pests to flourish in some locations. Many of the Arctic's problematic pesticides (toxaphene, DDT, hexachlorohexane (HCH)) continue to be used in central America, Africa and Asia, particularly by developing countries, and it is these countries that may be forced to rely more heavily on pesticides in coming decades (Harner, 1997).

Within Arctic drainage basins, warming may expand the area suitable for agriculture. Much of the southern portion of the Mackenzie Basin in North America is presently cultivated; under a global warming scenario this region is projected to contribute an additional 10 million hectares of land suitable for small grain crops (Cohen, 1997b), an area that might be further expanded with the development of new 'climate' resistant crops. In Russia, most of the major Arctic river basins contain agricultural land, particularly within the river valleys and as far north as 65°N (including the Severnaya Dvina, Ob, Yenisey and Lena River basins). Agriculture brings with it pesticides and other chemicals and, should pests thrive in a warmer Arctic climate, farmers may resort to increased reliance on pesticides to protect crops.

Chapter 6

The Effects of Climate Change on Contaminant Pathways

This chapter considers how the changes described in the previous chapters will affect specific groups of contaminants – heavy metals, radionuclides, organochlorine compounds (OCs), and hydrocarbons. For each group, the significance of the recent shift to high Arctic Oscillation (AO) index will be discussed and then consideration will be given to the more general, long-term changes thought likely. To avoid repetition, direct, unsupported statements are made for which the arguments and citations have been provided in previous chapters. Connections in the contaminant pathways (Figure 1·2) will be emphasized, such as: 1) mobilization from global primary or secondary contaminant sources and/or a change in delivery pathways to Arctic ecosystems; 2) entry into the base of the food web from water, snow pack, ice, soil, and runoff; 3) shifts in the relative importance of the source of primary productivity in aquatic systems (ice versus aquatic or coastal versus deep ocean); 4) change in food web structure affecting the degree of biomagnification (bottom-up effects); 5) change in the feeding ecology of key higher order consumers (top-down effects); and 6) change in the age structure of higher trophic order populations where contaminant concentrations in tissue are age-dependent.

6.1. Heavy metals

6.1.1. Lead, cadmium, zinc

Lead (Pb), cadmium (Cd) and zinc (Zn) are commonly released to the atmosphere through high-temperature processes or, in even greater quantities, to water through runoff, municipal discharges and dumping (Pacyna *et al.*, 1995). In addition, anthropogenic Pb has had a unique, predominant source in leaded gasoline combustion. Although leaded gasoline has been largely phased-out in North America and Europe, it is still used in other regions, including much of Russia. Due to the strong atmospheric connection in winter between Eurasia and the High Arctic (Figures 3·2 a and b), long recognized in events such as Arctic haze (Hileman, 1983) and brown snow (Welch *et al.*, 1991), much attention has been focused on the atmosphere as a means of transporting contaminant metals to the Arctic (Akeredolu *et al.*, 1994; AMAP, 1998; Boutron *et al.*, 1995; Pacyna, 1995; Rosman *et al.*, 1993; Sirois and Barrie, 1999; Sturges and Barrie, 1989). Based on back trajectories, models, and stable lead isotope composition, the sources of atmospheric metals have been established primarily as Eurasia, including the industrialized areas of Western and Eastern Europe and the Urals (in particular Norilsk), and secondly North America – each of which supplies air masses to the Arctic at particular times during the year (Figure 3·2). The shift between AO^- and AO^+ conditions alters mean wind fields thereby effecting change in the balance and timing of air movement from these various source regions, but the connections remain intact. Air transport from eastern North America and Western Europe strengthens under strong AO^+ conditions, especially in winter, due to the intensification and extension northward of the Icelandic Low. Air mass trajectory changes, while probably shifting the pathway and rate of transport between temperate sources and Arctic sinks, will probably not change the net delivery of airborne contaminants substantially. This hypothesis could be tested by running transport models (e.g., see Akeredolu *et al.*, 1994) under AO^+ and AO^- conditions.

The greatest leverage for change with aerosol metals resides in the wet and dry removal processes within the Arctic for which present knowledge is relatively incomplete. Because the Arctic is a poor trap for atmospheric metals associated with aerosols and particulates, sequestering <10% of the emissions that pass through it (Akeredolu *et al.*, 1994; Pacyna, 1995), there is considerable scope to enhance the deposition of airborne contaminants to surfaces by altering location and intensity of precipitation (Figures 3·4 a and 3·4 b) and/or by changing the balance between snow and rain (see, for example, Sherrell *et al.*, 2000). Under AO^+ conditions, the atmospheric corridor from eastern North America and Western Europe will become a more efficient trap for particulates, raining them out in the Nordic Seas and in the southern portion of the Eurasian Basin. Particle scavenging will generally increase wherever higher precipitation prevails, such as over northern Europe and the Eurasian Basin in general. Contaminants deposited on the eastern side of the Nordic Seas will then enter the Barents Sea and the Eurasian Basin via ocean currents. Contaminants deposited to the west will be delivered back into the North Atlantic via the East Greenland Current. Given this scenario, it seems likely that under AO^+ conditions, metal-contaminated aerosols entering the Arctic near the prime meridian will be subject to enhanced scavenging *en route*. Larger areas of open water (Figure 3·8) mean that scavenging will tend to place a greater proportion of these airborne contaminants directly into the surface ocean rather than on the sea ice. The decline of aerosol metal concentrations at Alert after about 1991, ascribed by Sirois and Barrie (1999) to the collapse of industry following the break-up of the former Soviet Union, could also be explained partly by changes in wind and precipitation patterns at the end of the 1980s (Figures 3·2 and 3·4). Enhanced loadings to sea-ice surfaces under AO^+ conditions are most likely to occur over the southern Eurasian Basin and this ice would then be exported back into the Greenland Sea.

The focus on the atmosphere as a pathway for the transport of metals from anthropogenic sources to the Arctic has to some degree diverted attention from the ocean. Sediment cores collected along the margins of the Eurasian and Canadian Basins (Figure 6·1) suggest that a major route for contaminant Pb to the Arctic Ocean

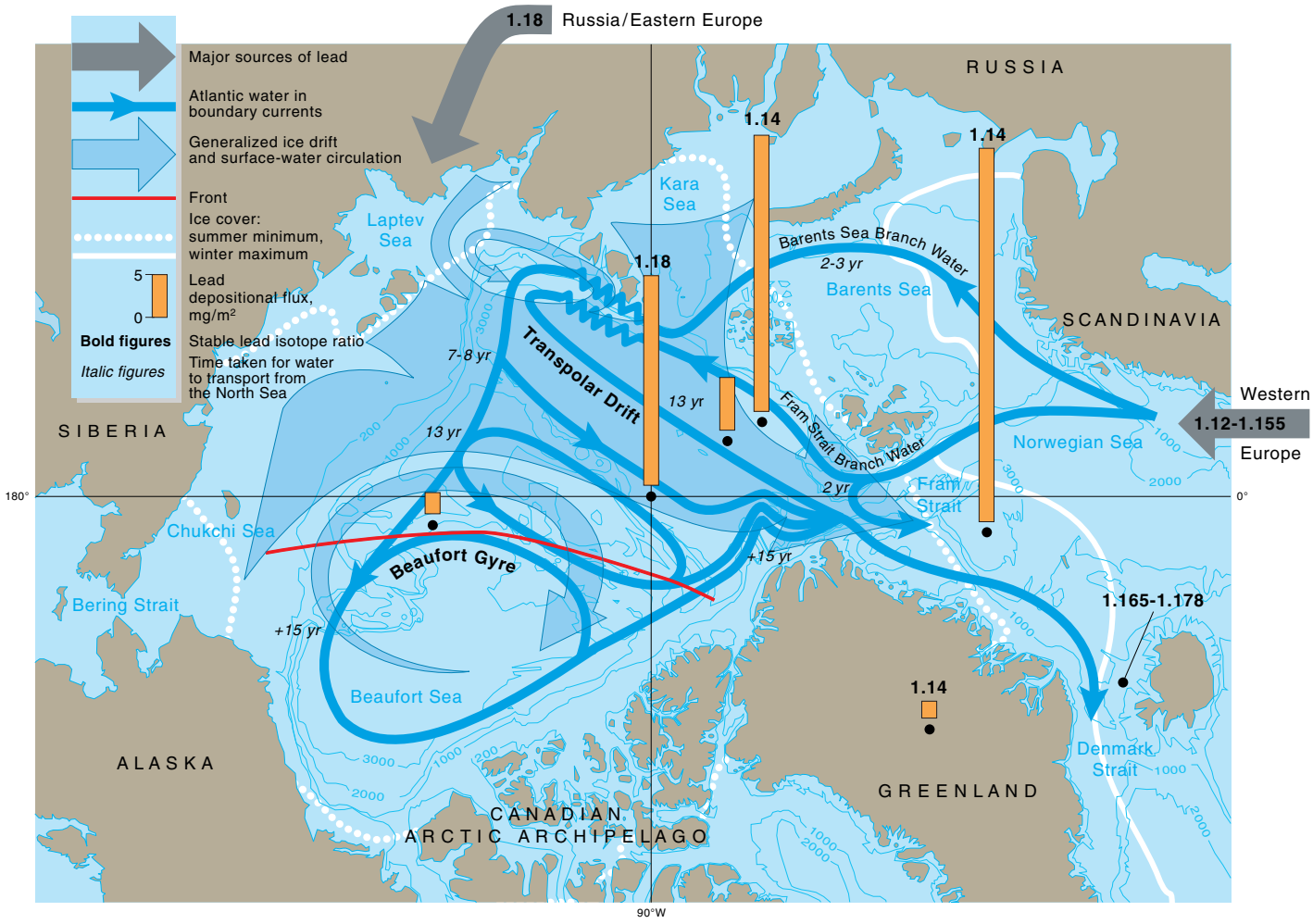

Figure 6·1. The major sources of contaminant lead transported to the Arctic are indicated by the stable lead isotope composition of sediment cores as being either Western Europe (indicated by a $^{206}Pb:^{207}Pb$ ratio of 1.14) or Eurasia (indicated by a $^{206}Pb:^{207}Pb$ ratio of 1.18) (adapted from Gobeil *et al.*, 2001a).

has been the same ocean current that transports radionuclides northward from the European reprocessing plants (Gobeil *et al.*, 2001a). The residence time of Pb in surface water, which is relatively short (<5 years), is still long enough to permit transfer of contaminant Pb from the North Atlantic and Nordic Seas into the Arctic (Gobeil *et al.*, 2001a). Under AO⁺ conditions an even more efficient transfer of metals may be expected from Western Europe to the Arctic either via rainout in the air transport corridor to the northwest of Europe (Figures 3·2 and 3·4) or via coastal discharges to the North or Baltic Seas. Lead comprises four stable isotopes: ^{204}Pb (1.48%), ^{206}Pb (23.6%), ^{207}Pb (22.6%) and ^{208}Pb (52.3%), with the composition varying between geological reservoirs (Sangster *et al.*, 2000). This variation has provided an incisive means of determining the sources of contaminant Pb in global environmental media, including Arctic aerosols and ice (Rosman *et al.*, 1993; Sturges and Barrie, 1989; Sturges *et al.*, 1993). Accordingly, Gobeil *et al.* (2001a) were able to relate the contaminant Pb accumulating in sediments along the Barents Sea margin to a western European source ($^{206}Pb:^{207}Pb \sim 1.14$) with ocean currents acting as the major transporting mechanism (Figures 3·17 and 6·1). Anthropogenic Pb in sediments near the North Pole, however, had an Eastern European or Russian composition ($^{206}Pb:^{207}Pb \sim 1.18$) (Figure 6·1). Based on the distribution of anthropogenic Pb in the Arctic sediments, these authors proposed a transport scheme wherein Pb from these source areas enters the Arctic Ocean via the Laptev Sea, either entrained in ice or, perhaps more likely, in water carried in the Transpolar Drift (TPD) (Gobeil *et al.*, 2001a). The observation that anthropogenic Pb was conspicuous in the Eurasian Basin margins but not in the Canadian Basin, led these authors to conclude that ocean and ice transport pathways during the peak Pb emission years (~ the last 60 years) must have been predominantly those associated with AO⁻ conditions. Under AO⁺ conditions, pathways for ice, ocean currents and runoff change dramatically (Figures 3·2, 3·5, 3·14) such that contaminant metals entering the Russian shelves (atmospherically or by runoff) would be diverted to the east into the Canada Basin and toward the Canadian Arctic Archipelago (Gobeil *et al.*, 2001a; Mysak, 2001). The pathways indicated by Pb contamination are probably relevant for other particulate-bound contaminants – for example some of the more highly chlorinated polychlorinated biphenyls (PCBs).

Atmospheric aerosols of Cd and Zn will to some degree behave like Pb, except that the predominant source of anthropogenic Pb is leaded gasoline as opposed to metallurgical industries and stationary combustion for Cd and Zn. Time series of aerosol composition at Alert (Sirois and Barrie, 1999) and records from ice cores

(Boutron *et al.*, 1991, 1995) and glacial snow (Sherrell *et al.*, 2000) reveal contamination due to industrial activity in Asia, Europe and North America. Like Pb, Cd and Zn are poorly captured within the Arctic (<15%; Akeredolu *et al.*, 1994; Pacyna, 1995) and changes in precipitation patterns probably have the greatest potential to change metal delivery to surfaces.

Of these three heavy metals, Cd provides the greatest risk to wildlife and human health, as a result of bioaccumulation and biomagnification (Figure 6·2), especially into liver and kidney of marine and terrestrial mammals (Braune *et al.*, 1999; Muir *et al.*, 1999). Observed high concentrations of Cd in Arctic biota, however, appear to be natural and not obviously related to human activities except, possibly, at locations close to sources (<100 km). In consequence, significant changes in Cd exposure are likely to occur through changes in the natural cycle of Cd and not by changes in anthropogenic Cd pathways. An exception to this rule may occur locally when Cd contamination is accompanied by, or followed by, system changes that alter Cd biogeochemistry. Croteau *et al.* (2002) provide a clear example of where reductions in Cd loadings to a contaminated lake were accompanied by increases in pH with the consequence that organisms actually exhibited increasing Cd uptake.

In the Arctic Ocean, natural cycles completely dominate Cd fluxes and budgets (Macdonald *et al.*, 2000a). Cadmium follows soft body parts in the marine biogeochemical cycle and exhibits a strong correlation with phosphate (Boyle, 1988; de Baar *et al.*, 1994). The interaction between the biogeochemical cycle and circulation of the world ocean results in sub-surface water of the North Pacific containing naturally higher Cd concentrations than that of the North Atlantic (by a factor of about 5 – see Bruland and Franks, 1983) which in turn makes the Pacific inflow through Bering Strait by far the dominant source of Cd to surface waters of the Arctic Ocean (Macdonald *et al.*, 2000a). Reduced Bering inflow since the 1960s (Figure 3·20 a) probably entails a similar (15%) reduction of the supply of Cd to the Arctic Ocean from that source. The encroachment of Atlantic water into the surface of the Makarov Basin, seen under recent AO^{+} conditions, will further reduce the domain of Cd-rich water within the Arctic (Figures 3·13 and 3·15). The accompanying increased stratification and recycled production in the smaller Pacific domain of the Canada Basin will, however, tend to maintain Cd from runoff or atmospheric deposition at the surface. It is noteworthy that the Canadian Arctic Archipelago is the downstream recipient of water from the Pacific Ocean and, therefore, the recipient of Cd and nutrients from that source.

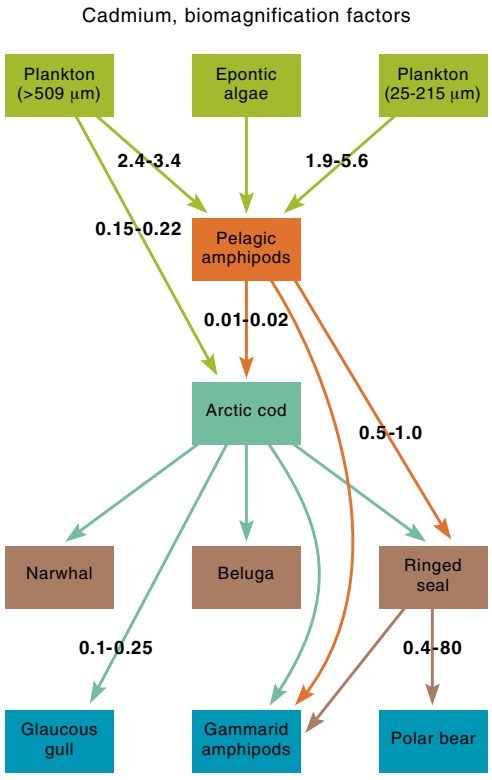

Figure 6·2. Pathways and biomagnification factors for cadmium in the Arctic marine food web. Biomagnification factors are based on dry weight concentrations in whole organisms for biomagnification to invertebrates and fish, and on wet weight concentrations for fish to muscle in top predators (for polar bear, liver was used instead of muscle) (from Muir *et al.*, 1999). In the case of the biomagnification factors from ringed seal to polar bear, the lower value is for seal blubber to polar bear liver, and the higher value is for ringed seal liver to polar bear liver.

Recent work on metal-impacted lakes near a copper-smelting center in Quebec, Canada, shows that metal loadings (Cd, Cu, Zn) can alter ecosystem structure, causing the demise of medium to large benthic invertebrates and producing fish populations shifted to smaller sizes (Sherwood *et al.*, 2000, 2001). This suggests the strong possibility that contamination by these heavy metals could interact with the accumulation and biomagnification of other contaminants like mercury (Hg) and OCs, potentially reducing concentrations of the latter in apex feeders.

6.1.2. Mercury

Due to its volatility and tendency to undergo biogeochemical transformation, Hg must be considered separately from other heavy metals (see, for example, Fitzgerald *et al.*, 1998; Mason and Fitzgerald, 1996; Mason *et al.*, 1994). Particular attention must be given to aquatic environments because it is there that Hg poses its greatest threat through biomagnification (Atwell *et al.*, 1998; Evans and Lockhart, 1999; Muir *et al.*, 1999). To a certain extent, processes leading to enhanced Hg concentration in the environment can be considered as either 'solvent switching' or 'solvent depletion' (Macdonald, *et al.*, 2002c). In the former, Hg moves between phases such as air, water and particles based simply on partition coefficients, whereas in the latter Hg may achieve high fugacity through the loss of surfaces or through chemical reactions mediated by photons or microbes (see, for example, processes described by Lindberg *et al.*, 2002; Malcolm and Keeler, 2002). The natural Hg cycle has been enhanced by human activities such that two to three times as much Hg is presently cycling through the atmosphere and upper ocean than before the rise of industry (Lamborg *et al.*, 2002; Mason and Fitzgerald, 1996; Mason *et al.*, 1994; Pacyna and Keeler, 1995). Because atmospheric Hg is almost entirely gaseous (Hg^0), it is tempting to assume that the polar regions might be global sinks simply due to low tempera-

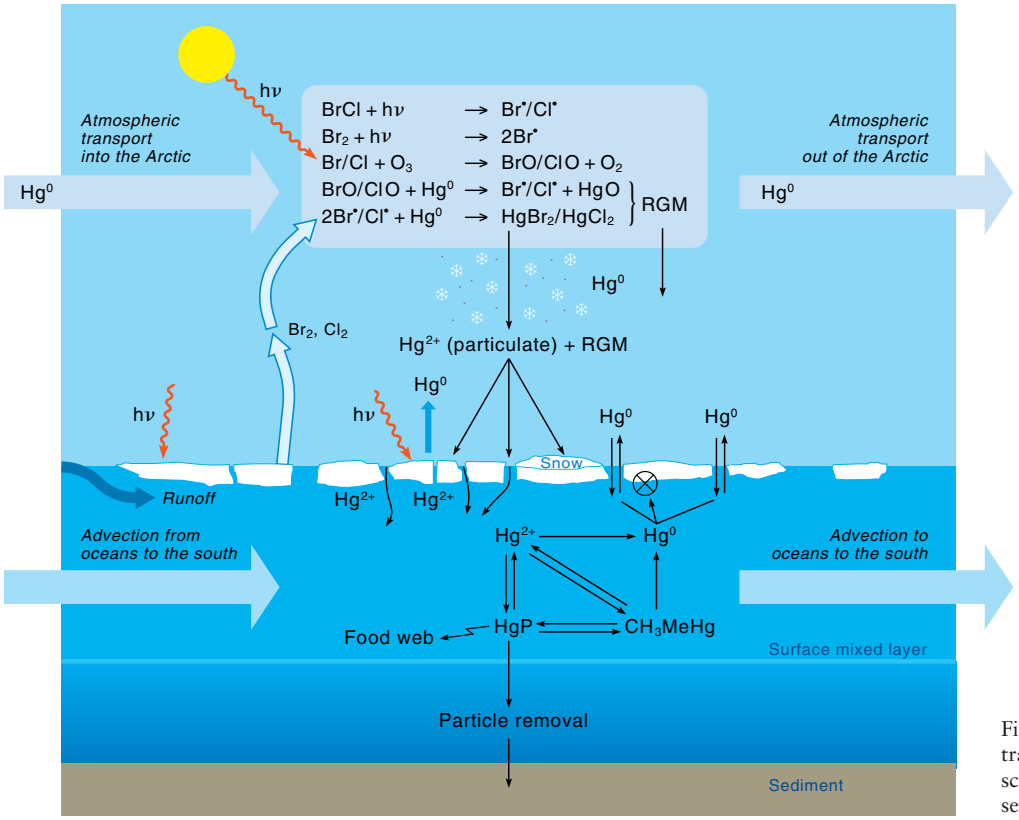

Figure 6·3. A schematic diagram illustrating how atmospheric mercury is scavenged after polar sunrise and subsequently enters surface waters.

ture and, accordingly, to make projections of the effects of climate change based on this fact alone. In fact, thermal forcing for Hg^0 to Arctic aquatic systems, either by rain or by air–water exchange, is weak due to a relatively high Henry's Law constant (Macdonald *et al.*, 2000a; Mason *et al.*, 1994). Nevertheless, the Arctic may be especially vulnerable to Hg because of an extraordinary set of circumstances at polar sunrise that results in the deposition of reactive (and bioavailable) Hg to the surface (Figure 6·3; Lindberg *et al.*, 2002; Lu *et al.*, 2001; Schroeder *et al.*, 1998). A relatively long residence time for Hg^0 of one to two years in the atmosphere (Lamborg *et al.*, 2002) ensures that winds can transport Hg to the Arctic from sources throughout the Northern Hemisphere. With the increase of UV radiation at polar sunrise, Hg^0 is converted to reactive gaseous mercury (RGM) through reaction sequences in which bromine, chlorine and ozone, and compounds like BrO and ClO appear to have prominent roles (Figure 6·3 and see Lindberg *et al.*, 2002; Lu *et al.*, 2001). The RGM thus formed is very effectively removed from the atmosphere by particles/snow, with this process estimated to account for the deposition of ~50 t (>90% of the annual total) on the Arctic Ocean and Hudson Bay during spring (Lu *et al.*, 2001) and perhaps as much as 150 to 300 t/yr (Lindberg *et al.*, 2002). Using an air transport model, Christensen *et al.* (AMAP, 2003a) estimated Hg deposition north of the Arctic Circle at 180 t when mercury depletion event (MDE) chemistry was included in the model, as opposed to ca. 80 t in the absence of MDEs. Based on snow samples, and the BrO distribution as determined from satellite measurements, reactive or particulate Hg is probably deposited mainly in areas subject to marine influence (marine aerosol being the source of the bromine/chlorine compounds involved in MDEs). The marginal seas of the Arctic appear to be especially vulnerable, because it here that sea salt (and therefore bromine) can most easily interact with snow through aerosols or frost flower formation associated with flaw leads or first-year ice. With spring warming, about two-thirds of the Hg deposited in snow is estimated to re-volatilize (Brooks, pers. comm., 2001) with the remaining third entering aquatic environments through meltwater.

Before projecting the impact of global change on the Hg cycle in the Arctic, it is necessary to understand how the invasion of Hg to global aquatic reservoirs via wet and dry deposition of reactive forms is balanced by gaseous evasion of reduced forms of Hg such as Hg^0 or methylmercury (MeHg) (Mason *et al.*, 1994). Dissolved Hg or Hg associated with suspended particulates in meltwater and runoff can drain into surface water below the ice cover whereas the evasion of Hg^0 is partly or completely blocked by ice cover. Indeed, this set of circumstances may provide the foundation for the elevated Hg levels which have been observed in Arctic biota (Evans and Lockhart, 1999; Macdonald *et al.*, 2000a; Muir *et al.*, 1999; Wagemann *et al.*, 1995, 1996). Unfortunately, appropriate geochemical studies investigating the cycling of Hg in Arctic aquatic systems have not yet been conducted and this is only speculation at the present time.

The vulnerability of the Arctic to global Hg emissions, therefore, probably lies in the mismatch between invasion and evasion processes. Climate change can affect both the invasion and evasion routes for Hg. Springtime depletion of atmospheric Hg depends on the availability of sea salt, calm weather, a temperature inversion, the presence of sunlight and sub-zero temperatures (Lindberg *et al.*, 2002; Lu *et al.*, 2001). Initially with climate change, it is likely that increased amounts of first-year ice around the polar margins will contribute to generally saltier ice and snow in spring which will enhance the production of BrO/ClO. Depending on what con-

trols the rate of supply of Hg to the Arctic, increased BrO/ClO will either enhance scavenging or maintain it at present levels, possibly extending the area of springtime Hg depletion beyond that implied by recent satellite measurements of the distribution of BrO/ClO (see Lu *et al.*, 2001). Considering that global emissions of Hg have generally decreased over the last 20 years, Lindberg *et al.* (2002) proposed that the recent increases observed in Hg levels in Arctic biota in some areas are, in fact, evidence that MDEs may be a recent phenomenon associated with change in sea-ice climate and atmospheric chemistry over the past decade or two. However, the entire Hg cycle must be considered before linking MDEs with Hg concentrations in apex aquatic feeders. Larger areas of open water in spring, either for ocean or lakes, will enhance exchange allowing gaseous forms of Hg to escape back into the atmosphere (Figure 6·3). With further warming, parts of the Arctic will become more temperate in character implying that atmospheric Hg depletion would decrease and evasion from the water increase leading eventually to lower Hg concentrations in water.

Aquatic food webs have strong leverage for change in Hg exposure in the Arctic because MeHg biomagnifies, exhibiting a concentration increase of about 1000- to 3000-fold from particulate organic matter to apex predators (Figure 6·4; Atwell *et al.*, 1998; Kidd *et al.*, 1995a; Muir *et al.*, 1999). Mercury concentration also increases with age/size in predatory fish, such that large, old fish often exceed thresholds considered safe for unrestricted human consumption (Lockhart and Evans, 2000), containing anywhere from two to five times the Hg concentration of smaller fish of the same species. Therefore, adding an extra step in the food web could reasonably be expected to enhance Hg concentrations in higher trophic levels by a factor of about five. Likewise, altering the population distribution of fish in a lake could produce a change rivaling or exceeding any change caused by alteration of physical pathways.

With warming (Figure 3·3) will come the loss of permafrost (Figure 3·24) causing altered hydrology, potentially more wetland, and enhanced fluxes of soil and organic carbon to rivers, lakes and estuaries. Warming of drainage basins in the Arctic, therefore, would appear to provide a widespread mechanism to increase Hg fluxes to northern aquatic environments and to the atmosphere. A recent study of the pathway of Hg from snow-covered land to streams in Vermont (Stanley *et al.*, 2002) showed that Hg export from soils correlated with particulate organic carbon, and that Hg concentrations in runoff increased with flow – unlike most solutes (see also Bishop *et al.*, 1995). These two factors together suggest that episodic, large releases of organically-bound Hg (of both anthropogenic and natural origin) may become a dominant feature accompanying permafrost degradation. Clearly, Arctic lakes would be most vulnerable to this process, but enhanced input of terrestrial carbon is projected to occur to Arctic seas as well (Kabat *et al.*, 2001), suggesting that Hg loadings there may, similarly, be increased. In the ocean, Hudson Bay would seem especially vulnerable, partly due to its large drainage basin, already affected by reservoir flooding (Bodaly and Johnston, 1992), and partly due to the likelihood of permafrost melting within that drainage basin (Gough and Wolfe, 2001). It seems noteworthy that enhanced

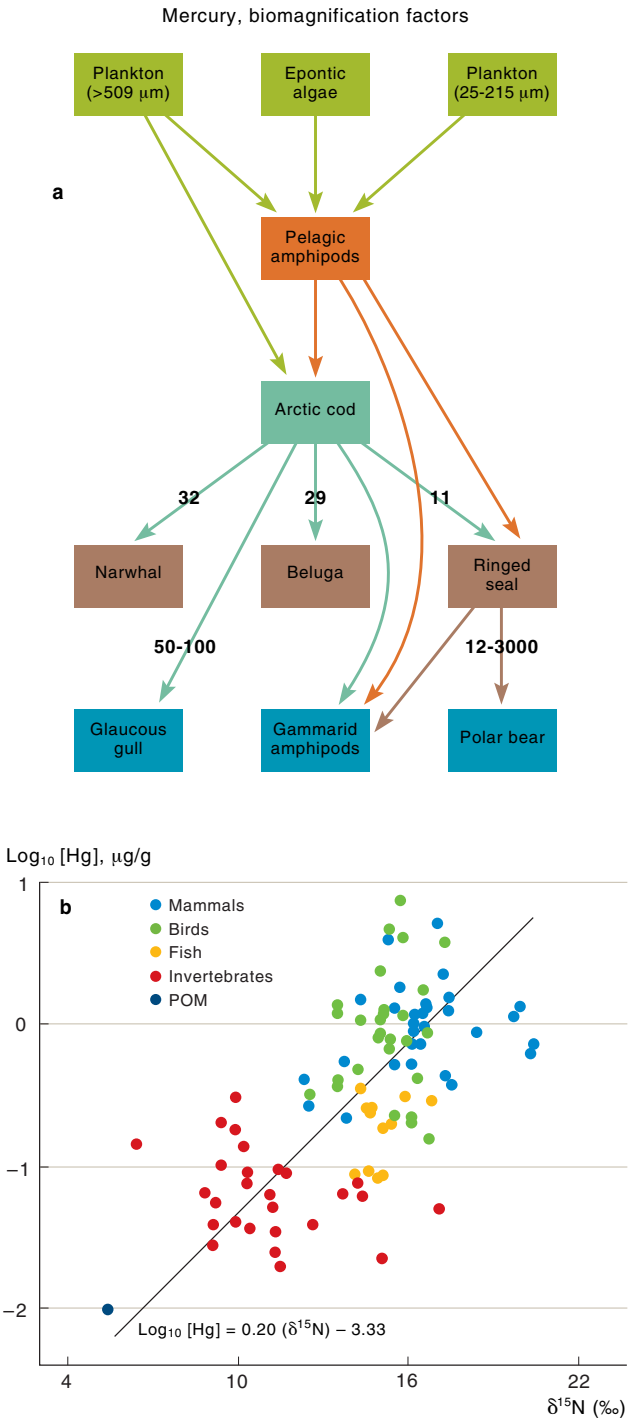

Figure 6·4. Biomagnification of mercury in Arctic food webs. This figure illustrates a) biomagnification factors for a simplified food web (based on dry weight concentrations in whole organisms (invertebrates and fish) and on a wet weight concentration for fish to muscle in top predators. For polar bear, liver was used instead of muscle) (Muir *et al.*, 1999) and b) biomagnification as a function of trophic level based on $\delta^{15}N$ measurements (adapted from Atwell *et al.*, 1998). In the case of the biomagnification factors from ringed seal to polar bear, the lower value is for seal blubber to polar bear liver, and the higher value is for ringed seal liver to polar bear liver.

concentrations of Hg in snow are observed generally in that region (Lu *et al.*, 2001) and that a recent increase in Hg flux to Hudson Bay sediments has, likewise, been observed (Lockhart *et al.*, 1995).

Historical records from dated sediment cores have been used to infer Hg fluxes increasing by factors of 3 to 7 in some Arctic lakes during the past two centuries

(Bindler *et al.*, 2001; Landers *et al.*, 1995; Lockhart *et al.*, 1998). What is not so clear is whether such increases are due to increased atmospheric deposition or to alteration of processes that transfer Hg from wetlands to atmosphere or from the drainage basin into lakes or transfer Hg from the water to the sediments. In regard to the latter, Gajewski *et al.* (1997) have shown major increases in diatom fluxes to varved sediments from a lake on Devon Island which they attribute to climate change (i.e., longer, ice-free summers). Not only could such a mechanism explain enhanced Hg fluxes to Arctic lake sediments but it could also have the non-intuitive result of *reducing* the exposure of higher trophic levels to Hg through bloom dilution at the algal stage (Pickhardt *et al.*, 2002).

Mercury depletion events leading to the production of bioavailable Hg have been confirmed at Barrow, Alaska (Lindberg *et al.*, 2002). Given that this region receives airborne contaminants from Asia (Li *et al.*, 2002; Wilkening *et al.*, 2000), that China is increasing its reliance on coal for energy, and that coal is a leading source of anthropogenic Hg (Nriagu and Pacyna, 1988), it seems likely that the Bering/Chukchi Sea region may be especially vulnerable to further increased Hg loadings. In general, the pathways of anthropogenic Hg might be expected to change as a result of shifts in major sources, for example, as European and North American emissions decrease and Asian emissions increase.

6.1.3. Arsenic

The sediment geochemistry of arsenic (As) can, like Hg, be altered by changing the carbon cycle. Mining and smelting have been major sources of As to the environment (Nriagu, 1989), where it may accumulate in aquatic sediments (Martin and Pedersen, 2002). In the Arctic, it has also been suggested that underwater nuclear weapons testing may have provided a significant anthropogenic As source to the Pechora Sea (Loring *et al.*, 1995). Large natural surface sediment enrichment of As is often produced by diagenesis; solid-phase As(V) at depth in sediments is remobilized through reduction to As(III), which then diffuses upward to re-precipitate through reactions with nitrate, oxides of manganese or oxygen (Figure 6·5). This natural redistribution of As in sediments makes it exceptionally precarious to infer contamination from sediment surface As enrichments (Loring *et al.*, 1995, 1998; Siegel *et al.*, 2001). However, strong sediment-surface enrichments serve as a warning that alteration of the organic geochemistry of aquatic sediments by enhanced organic carbon fluxes may have unanticipated consequences for As mobility (cf. Figures 6·5 a and b).

A geochemical study of a temperate, seasonally ice-covered lake contaminated by As, Cu, nickel and Zn is especially instructive (Martin and Pedersen, 2002). When action was taken to reduce metal loadings, the lake's response was enhanced phytoplankton production which then invigorated carbon fluxes to sediments. Through metabolism in sediments, the enhanced carbon fluxes then reduced oxygen, thus mediating the conversion of solid-phase As(V) to dissolved-phase As(III) which then diffused back into the bottom water. The unanticipated result of decreasing metal loadings to the lake was to produce higher As concentrations. If one of the responses to change in Arctic aquatic environments is enhanced aquatic productivity or enhanced organic carbon loadings, release of solid-phase As, especially from sediments with high natural or anthropogenic contaminant burdens (e.g., see Loring *et al.*, 1995, 1998; Siegel *et al.*, 2001) is a likely response.

6.2. Radionuclides

Previous assessments have outlined the atmospheric and oceanic pathways that transport artificial radionuclides to the Arctic (AMAP, 1998; Macdonald, *et al.*, 2000). Atmospheric artificial radionuclides derive mainly from atmospheric weapons testing predating the mid 1960s, and the Chernobyl accident in 1986. Accordingly, predominant food web contamination from the atmosphere has occurred to terrestrial systems through fallout (AMAP, 1998) and there appears little opportunity for

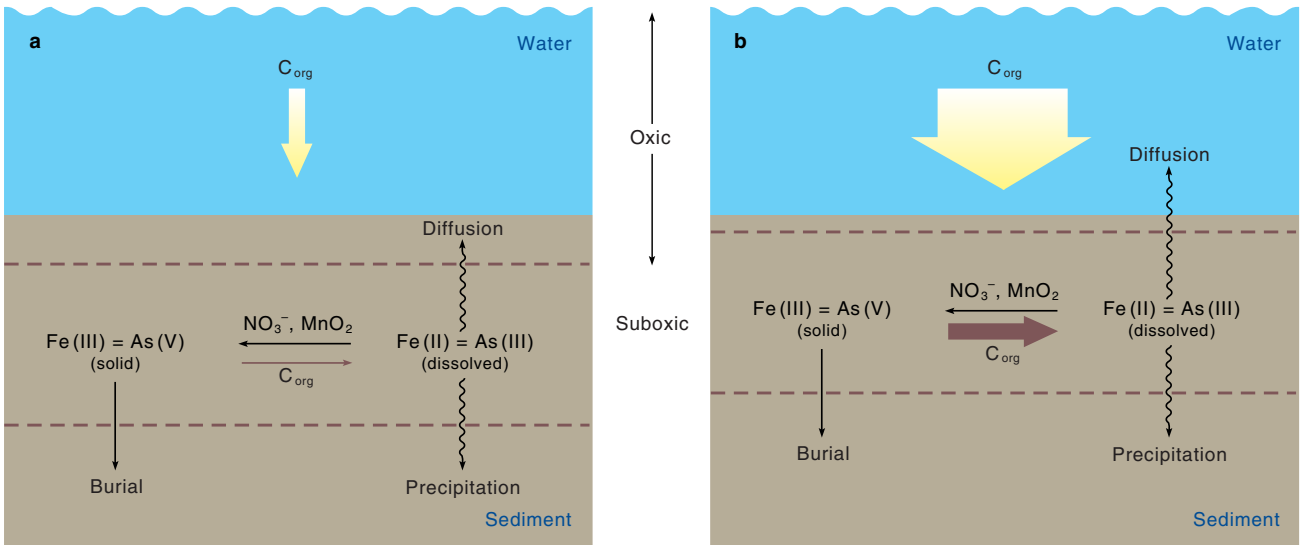

Figure 6·5. A schematic diagram showing how arsenic cycles in sediments (modified from Sullivan and Aller, 1996). The solid-phase form of arsenic (As(V)) is released to pore water through reduction to As(III) which may then diffuse back out of sediments. Enhanced fluxes of organic carbon to sediments, shown in the right-hand panel, are reflected by enhanced diffusion of dissolved arsenic back into bottom waters.

present or future climate changes to have much impact on historical atmospheric sources which are decaying with half-lives of about 30 years (^{137}Cs and ^{90}Sr, predominantly). The distribution of fallout from Chernobyl was very much controlled by wind and rainfall patterns (see Figures 8.4 and 8.5 in AMAP, 1998), and these pathways are clearly subject to climate change. An important lesson to be learned from the Chernobyl accident is that climate patterns can predispose the Arctic in how it will receive contaminants from such accidents and, for example, wetter conditions in the Nordic Seas and Northern Europe during AO$^+$ conditions (Figure 3·4 b) would favour deposition of ^{137}Cs fallout in that region.

In the ocean, the predominant artificial radionuclides (^{137}Cs, ^{90}Sr, $^{239+240}$Pu) do not biomagnify sufficiently (except perhaps for plutonium (Pu) in brown macroalgae; see Berrow *et al.*, 1998; Fisher *et al.*, 1999) to contribute significantly to the radiation dose for humans (Layton *et al.*, 1997; Macdonald and Bewers, 1996). This suggests that, in order to find pathway changes that might be cause for concern, it is the ice and surface water motion of the Arctic Ocean that must be considered, both of which exhibit variability resulting from the AO/NAO (Figures 3·9 and 3·14).

Clearly, the enhanced northward transport of water in the Nordic Seas under the AO$^+$ conditions of the 1990s (Figure 3·15) strengthened the delivery to the Arctic of radionuclides discharged by the European nuclear reprocessing plants where they continued to spread into the surface waters of the Makarov Basin (Figures 3·13 and 3·15; Smith *et al.*, 1998).

The transport routes for radionuclides in the Nordic Seas are generally known (Figure 3·17). A shift in the climate regime toward increased NAO index and stronger wind fields will probably lead to radionuclides undergoing a faster transport closer to the Norwegian coast, with a larger proportion entering the Barents Sea. It is expected that other contaminants entering the North Sea and southern Norwegian Sea from sources in Europe will encounter similar change in their oceanic transport route as that proposed for radionuclides.

The enhanced coupling between release points for European reprocessing plant nuclear wastes and the Arctic Ocean will be more than offset by reduction in releases of the major radionuclides that have occurred since the 1970s (Macdonald *et al.*, 2000a), and input of tracers such as ^{137}Cs to the Arctic Ocean should continue to decline. Recent increased discharges of technetium, however, provide a reminder that not all radionuclide discharges from European reprocessing plants are declining (AMAP, 2003c). Extensive data collection under the Arctic Nuclear Waste Assessment Program (ANWAP; Layton *et al.*, 1997) and from icebreakers (Smith *et al.*, 1998) has provided sufficient information on the distribution of artificial radionuclides in Arctic surface waters to show that they pose little risk to human or ecosystem health. The conclusion of the ANWAP assessment was that the largest radiation doses to individuals living on the Alaskan coast and consuming subsistence seafoods were, in order of importance, ^{210}Po (a natural radionuclide), followed by ^{137}Cs and ^{90}Sr from atmospheric fallout. It seems that the nuclear reprocessing radionuclides have made elegant tracers of water motion and, although they will reflect recent changes in that motion, they will continue to provide almost no risk to Arctic marine biota (Macdonald and Bewers, 1996).

The ice and surface water pathway changes forced by the AO (Figures 3·10 and 3·14) strongly suggest that radionuclides discharged to the Russian shelves may, under AO$^+$ conditions, enter the Canadian Basin and subsequently find their way into the Canadian Arctic Archipelago. The estimated total release of radionuclides to the Kara Sea via river water (Ob, Yenisey) is about 1.1×10^{15} Bq (^{90}Sr and ^{137}Cs) (Paluszkiewicz *et al.*, 2001), or about an order of magnitude less than the fallout and reprocessing plant sources to the Arctic Ocean (Aarkrog, 1994). It seems likely, therefore, that diversion of Russian river runoff from the Eurasian Basin to the Canadian Basin under AO$^+$ conditions will be matched by a diversion of associated radionuclides (see Cooper *et al.*, 1999) which, nevertheless, will provide little threat to ecosystems there.

Ice drift, the remaining transport pathway, provides a distinct, but difficult to quantify risk. Sediments from the Russian shelves, known to have been contaminated by weapons testing and accidental and deliberate radioactive waste discharges, have been found to be heavily contaminated at several locations (Josefsson, 1998; Matishov *et al.*, 1999; Smith *et al.*, 2000). Suspension freezing provides an efficient mechanism to entrain fine sediments into newly-formed ice in the Russian seas (Eicken *et al.*, 2000) and ice has been shown to carry radioactive sediments (Dethleff *et al.*, 2000a; Landa *et al.*, 1998; Meese *et al.*, 1997). High radioactivity has been found in ice-entrained sediments in the Canada Basin (>70 Bq/kg; Cooper *et al.*, 1998) and in the Canadian Arctic Archipelago (Darby, pers comm., 2001) but the origin of the sediment in the ice, based on mineralogy, has not been assigned to Russian shelves. Given the very few samples together with their uncertain provenance, it is impossible to quantify risks to biota in the Canada Basin and the Canadian Arctic Archipelago from contaminated ice, except to say that the AO$^+$ conditions of the early 1990s appear to produce ice transport pathways conducive to carrying sediment and surface water from the Russian shelves into the Archipelago.

Perhaps the most significant increase in radioactivity exposure to northern residents will come from the natural ^{226}Ra decay series that supports ^{222}Rn, ^{210}Pb and ^{210}Po. ^{210}Pb in aquatic systems derives partly from *in-situ* production supported by ^{226}Ra and partly from ^{222}Rn which has diffused out of soils and, with a short (3.8 day) half-life, decays to ^{210}Pb which is scavenged by particles. This latter component, called excess ^{210}Pb, often exceeds the 'supported' ^{210}Pb in aquatic sediments. Presently, excess ^{210}Pb tends to be very low in the Arctic because ^{222}Rn remains trapped in the soil by permafrost and snow/ice cover. With warming, ^{222}Rn evasion will increase as will excess ^{210}Pb activity matched by the activity of ^{210}Po, its granddaughter. Since ^{210}Po and ^{222}Rn together account for about 75% of the radiation dose to native northern residents (Layton *et al.*, 1997; Macdonald and Bewers, 1996) any substantive increase in ^{222}Rn evasion due to warming/permafrost melting would have a widespread and substantial (doubling or tripling) effect on the radiation dose.

6.3. Organochlorine compounds

Of all the contaminants, the organochlorines (OCs) provide the greatest challenge to predict consequences of change because they have been so widely released, comprise so many compounds and exhibit such a wide range of physical chemical properties. Furthermore, the important chemical properties – volatility, phase partitioning, and degradation kinetics – are all sensitive to temperature and hydrological change. Efforts to determine where in the environment these compounds end up have improved the understanding of global pathways enormously, but new surprises give an indication that intuition often fails due to an incomplete grasp of environmental processes (Macdonald *et al.*, 2000b; Oreskes *et al.*, 1994; Schindler, 1997). Recent OC budgets underscore the importance of the atmosphere–ocean coupling in the transfer of OCs to the Arctic from their temperate and tropical release points (Li *et al.*, 2002; Macdonald *et al.*, 2000a,b). These same budgets show that the relative importance of atmosphere versus ocean in transporting contaminants will vary widely among the OCs and over time. Therefore, change forced by the AO or by general global change will have a similarly varied impact depending on the particular OC and the time period in question. All the OCs of concern (DDT, toxaphene, chlordane, PCBs, hexachlorocyclohexanes (HCHs)) have transient emissions: that is, they were first released in the 1930s to 1940s, emissions peaked sometime in the 1970s to 1990s, after which they have generally declined or ceased. Continued declines, or possibly increases, will depend on bans, further controls, or new use following invigorated assaults by pests linked to climate change. As a general rule, particularly with transient releases of chemicals that partition strongly into water, the atmosphere is initially the dominant transporting medium, but as aquatic reservoirs (lakes, rivers, upper ocean) become loaded, these then contain the important if not dominant budget and flux terms (Li *et al.*, 2002; Macdonald *et al.*, 2000a,b) and it is dominant budget terms that posses the greatest leverage for change.

Before discussing how climate change is likely to impact upon OCs, it is important to understand the manner in which OCs become concentrated in the environment. Building on earlier perspectives developed by Wania (1999), Macdonald *et al.* (2002c) suggest there to be two fundamental concentrating processes which they termed *solvent switching* and *solvent depletion*. The distinction between these two processes is particularly important in the context of change. Solvent switching is a natural process wherein a particular contaminant distributes itself between various phases (solid, liquid, gas) according to well-described thermodynamic rules. This process can lead to elevated concentrations – for example, the partitioning of HCH into cold water will produce concentrations in the water that far exceed those in the air, and indeed this process alone can cause non-intuitive divergence of chemicals over large scales (Li *et al.*, 2002). But this process cannot cause concentrations to exceed thermodynamic equilibrium (that is, solvent switching does not increase fugacity). Another very important solvent-switching process is the transfer of a POP from water to lipid at the bottom of the food web; again, this process enhances POP concentrations in phytoplankton or small zooplankton strictly according to thermodynamic forcing.

Solvent depletion, however, is a very different process in that it can lead to concentrations in selected media that exceed thermodynamic equilibrium (i.e., fugacity is increased), which requires a source of energy. Perhaps the clearest example of solvent depletion occurs in the food web where lipid transfers, going from one trophic level to a higher one, are inefficient. Metabolism effectively burns much of the lipid, leaving the contaminant to accumulate in a decreasing volume of stored fat. This process can lead to OC concentrations in aquatic apex feeders that are well above thermodynamic equilibrium with the water, and the situation can be exacerbated by starvation cycles during which individuals reduce their fat content further. Arctic and other cold environments offer several solvent depletion processes by which contaminants can be ramped up above thermodynamic equilibrium (Macdonald *et al.*, 2002c). In particular, a POP can be scavenged by adsorption onto snow and thence transported to the ground. The initial large snow surface (0.1 m^2/g) can be reduced by a factor of over 100 and, indeed, during melting, the snow surface can disappear entirely effectively removing the solvent (the snow's surface) from under the contaminant. The fugacity can be increased enormously during snow sintering/melt with the consequence that the OC is forced to diffuse back into the atmosphere, enter the meltwater or adsorb onto other particles. Similarly, fog water droplets can provide a temporarily large surface area which is thermodynamically attractive – upon coalescence, most of the surface is lost and contaminant fugacity increases in the remaining large water droplets. Finally, cryo-concentration may occur in shallow lakes or seas that form a thick ice cover. The withdrawal of water into the ice, leaving behind most of the POPs, can easily increase concentrations in the water beneath the ice to values that exceed thermodynamic equilibrium with the atmosphere by factors of 2 to 5. Due to the ice cover, diffusion back into the atmosphere is not an option.

Under change scenarios, the solvent-switching processes can be modeled by taking into account changes in partition coefficients and vapour pressures with temperature. However, changes in the solvent-depletion processes are much harder to project especially since the elevated fugacities imply transports that will be sensitive to time. For example, the effect of snow on contaminants entering an Arctic lake will depend on when the snow accumulates, how long it sinters, how porous and how deep the snow is, how and when snow melt occurs, and how quickly snow melt enters the lake. None of these transfer processes have been investigated in sufficient detail to provide guidance for modellers.

6.3.1. The influence of the Arctic Oscillation

There appear to be several consequential ways that physical pathways could change in response to the AO. In the Nordic Seas, especially under AO^+ conditions, the atmospheric coupling of eastern North America and Western Europe with the Arctic becomes more intense, in particular during winter/spring (Figures 3·2 a and b) suggesting that spraying pesticides in these regions will, if anything, intensify 'events' such as those seen at Alert

for α- and γ-HCH (see, for example, Figure 22 in Macdonald *et al.*, 2000a). Furthermore, re-emission of old OC residues in soils and aquatic reservoirs of Europe or eastern North America will enter these same air pathways to be transported north. In this regard it is worth noting that the highest cumulative use of PCBs was in Western Europe and eastern North America (see Figure in AMAP 2003b) – both of which are source regions for air masses entering the Arctic between Greenland and Europe (Figures 3·2 a and b). The higher precipitation in the Nordic Seas and southern Eurasian Basin (Figure 3·4) will provide more effective scavenging of particle-associated OCs (high molecular weight PCBs, for example) and for OCs with low Henry's Law constants (HLCs) that partition strongly into water (HCHs, toxaphene).

In the Bering Sea, gas exchange with the ocean or washout by rain can provide a mechanism to remove β-HCH selectively from the air as it moves northward, simply due to its exceptionally low HLC (Li *et al.*, 2002). This process does not prevent the entry of this contaminant to the Arctic Ocean; rather, it switches the mode of delivery from winds to ocean currents and, as a consequence, slows the rate of transport from m/s to cm/s. Under AO$^+$ conditions, therefore, a more rapid atmospheric transport of OCs into the Arctic from Western Europe is likely, with the delivery shifting toward the ocean pathway for OCs that partition strongly onto particles or have low HLCs. On the Pacific side, OCs will continue to enter the Arctic via the atmosphere and ocean currents (see, for example, Bailey *et al.*, 2000), but the 15% reduction in Bering Sea inflow over the past several decades would effect a proportional reduction in this pathway. Variation in precipitation over the North Pacific and Bering Sea (Figure 3·20 b) will alter the balance between atmosphere and ocean as transport pathways to the Arctic.

Larger areas of open water under AO$^+$ conditions or from general climate change (Figures 3·6, 3·7 and 3·8) will accelerate equilibrium between air and sea by an amount equivalent to the expanded open areas. Furthermore, increased numbers of polynyas in winter will enhance the production of fog over sea ice, acting to scavenge and deposit contaminants to surface (Chernyak *et al.*, 1996) at locations known to be important for biota (Stirling, 1997). Due to the drastic reduction in atmospheric concentrations as a result of emission controls, α-HCH has become oversaturated in ice-covered areas of the Arctic Ocean (Jantunen and Bidleman, 1995; Macdonald *et al.*, 2000b). The opening of the pack and seasonal clearance of shelves will, in this case, result in evasion and drawdown of HCH from the upper ocean. In contrast, PCBs and toxaphene are still loading into the Arctic Ocean via the atmosphere (Macdonald *et al.*, 2000b) and, therefore, the same loss of ice cover will lead to increased loading of these substances in surface seawater. A PCB budget for the Arctic Ocean (Macdonald *et al.*, 2000b) estimated a net gas exchange into the ocean of about 20 t/yr. The reduced ice cover evident in Figure 3·8 might lead to as much as a doubling of the area of open water which would similarly double net exchange. Not only does change in ice cover alter the air-sea exchange of OC gases, but the consequent loading or unloading of the interior Arctic Ocean with such chemicals will later be felt in the composition of water exiting the Arctic through the Canadian Arctic Archipelago and in the East Greenland Current and points farther downstream.

Table 6·1. Russian river loadings for selected organochlorine compounds.

	Load, t/yr	Percent of Arctic Ocean input budget	Reference
α-HCH	25	13	Alexeeva *et al.*, 2001; Macdonald *et al.*, 2000a
γ-HCH	44	51	Alexeeva *et al.*, 2001; Macdonald *et al.*, 2000a
ΣPCBs	15	23	Macdonald *et al.*, 2000b
ΣDDT	18		Alexeeva *et al.*, 2001

The diversion of Russian river inflow toward the east under AO$^+$ conditions (Figure 3·14) will have a significant effect on OC pathways within the Arctic Ocean. Inherent with this diversion is a shift of all the OC loadings from these Russian rivers out of the Eurasian Basin and into the Canada Basin (Table 6·1, and see Macdonald *et al.*, 2000a; Sericano *et al.*, 2001). As the Canada Basin has a longer residence time (10 years, compared to 2 years in the TPD), there would be an added consequence of increased contaminant inventories for the Arctic Ocean in general and the Canada Basin in particular. Instead of tracking across the Eurasian Basin to exit into the East Greenland Current, OCs discharged by the Russian rivers might now exit via the Canadian Arctic Archipelago (see Figure 3·14). The changes here are consequential to budgets (Table 6·1) and to distribution within the water column, keeping in mind that the same river water that delivers contaminants also stratifies the ocean and potentially reduces new production and vertical particle flux, which together will act to maintain river-borne contaminants near the surface where they can partition into algae. Although there are few data from which to evaluate the relative importance of OC pathways in the ocean, the findings of Andersen *et al.* (2001) provide a strong warning that there are sources of PCBs in the region around the Kara Sea and Franz Josef Land, possibly as a consequence of riverine inputs.

Water in the Canadian Arctic Archipelago channels, supplied from surface water in the Arctic Ocean (~0-200 m), has the potential to undergo change in its OC content due to alterations in the distribution of water masses in the Canada or Eurasian Basins. As shown in several studies (Carmack *et al.*, 1997; Li *et al.*, 2002; Macdonald *et al.*, 2000a,b), HCHs are not distributed uniformly within the Arctic Ocean and it is likely that other OCs are, likewise, not uniformly distributed. For example, α-HCH is highest near the surface, decreasing to very low concentrations in water deeper than several hundred meters, and the Canada Basin under the permanent pack ice exhibits much higher HCH concentrations than are observed in the Chukchi Sea or the Eurasian Basin surface waters. The redistribution of Pacific and Atlantic water masses in surface water of the Arctic Ocean (Figures 3·13 and 3·15) may therefore have been accompanied by change in the composition of water flowing into the Canadian Arctic Archipelago. Such change could occur in two ways, either by horizontal

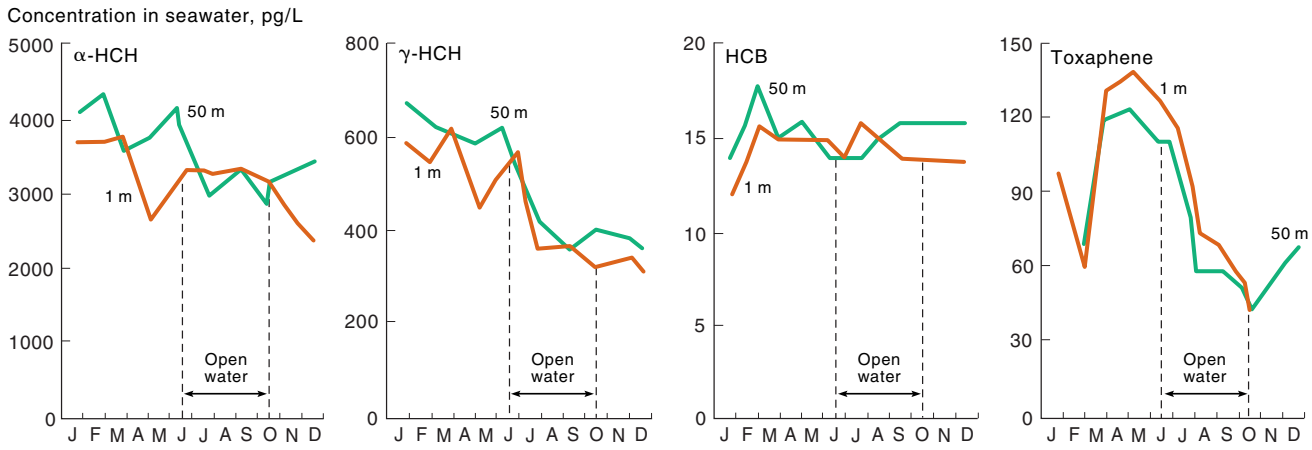

Figure 6·6. Time series of organochlorine concentrations measured at Resolute (Canadian Arctic Archipelago) during 1993 at the surface and 50 m water depth for α- and γ-HCH, hexachlorobenzene and toxaphene (source: Hargrave *et al.*, 1997).

displacement of water mass domains or by vertical displacement of water properties. Although there are few data from which to evaluate how the water composition in the Archipelago channels might respond to the AO, an extraordinary set of data collected by Hargrave *et al.* (1997) illustrates clearly that change in upstream water composition can have important consequences for contaminant concentrations in water flowing through the Archipelago. Seasonal measurements of OCs made at Resolute in 1993 (Figure 6·6) reveal a coherence between the surface and 50 m water depth in contaminant trends. This coherence, together with patterns differing for the various OCs, argues strongly that the observed time trends at Resolute are produced by variation in the composition of upstream water drawn into the Archipelago from the Canada Basin. The concentration variations in this single season's data exceed a factor of two.

6.3.2. The effect of glacial melt back

Glacier ice-mass loss and snow melt back due to warming (cyclical or trend) can release archived contaminants accumulated during years of higher fluxes (Blais *et al.*, 1998). Based on the total amount of glacial melt back (Figure 3·25) and the range in concentration of OC contaminants measured in ice and snow during the 1960s to 1990s for the Agassiz Ice Cap (Table 6·2) the maximum total inputs are expected to range from about 3 kg for PCBs to as much as 400 kg for ΣDDT. For 1993, the year exhibiting the most substantial melt back on record, the maximum inputs of PCBs and DDT were estimated at 0.5 kg and 74 kg, respectively. The HCH and PCB inputs are minuscule compared to Arctic Ocean budgets (Macdonald *et al.*, 2000b) and are also relatively small compared to the flux of these contaminants through the Canadian Arctic Archipelago (Table 6·2). For DDT, however, glacial melt appears potentially to provide an important, climate-modulated source. For Arctic glaciers, most of the melt occurs in zones where old ice of pre-industrial age is emerging. This ice would contain little or no contaminant burden and, indeed, would act to dilute any contaminants released to aquatic environments. For smaller ice caps, more recent layers of snow and ice might be involved. Thus, glacial melt back appears to be of significance only for DDT, and even then is likely to be of only local and short-lived significance.

6.3.3. The effect of warming on organochlorine cycling in lakes

Arctic lakes presently tend to retain only a small fraction of the contaminants they receive, a fact which should indicate the potential for change. Studies and models (Helm *et al.*, 2002; Macdonald *et al.*, 2000a) show that the snowmelt and runoff cycle connects with the lake's hydrological cycle such that most of the contaminants deposited in the drainage basin throughout winter are transported across the lake surface in a low density layer under the ice to exit in out-flowing water. The lack of a strong particle flux due to oligotrophic conditions further decouples deep lake water from contaminants at the surface. Reduced ice cover and loss of permafrost, leading to greater mixing and stronger primary production, will enhance the ability of Arctic lakes to retain OCs. How this added retention will be expressed in the food web is less certain. Enhanced primary production and settling of ungrazed phytoplankton in early spring might draw down contaminant burdens in lake surface water and thereby reduce entry of contaminants into the food web or act so as to dilute the OC concentration in algae as has been shown for Hg (Pickhardt *et al.*, 2002). How-

Table 6·2. Potential input of selected organochlorine compounds from glacial melt.

	Concentration, pg/L	Total glacial input, kg	Glacial input for 1993, kg/yr	Flux through the Canadian Arctic Archipelago[a], kg/yr
α-HCH[b]	256	205	39	195 000
γ-HCH[b]	115	92	18	27 900
ΣDDT[b]	480	384	74	161
Chlordane[b]	35	28	5	96
HCB[b]	65	52	10	810
PCBs[c]	3.5	2.8	0.5	2700

[a] Flux through the Canadian Arctic Archipelago was estimated assuming a mean flow of 54 000 km³/yr (Macdonald *et al.*, 2000a) and concentration data collected in the Archipelago during 1993 (HCH, HCB, Chlordane; Hargrave *et al.*, 1997) or Canada Basin in 1997–98 (PCB, DDT; Macdonald *et al.*, 2001);
[b] Concentration data from Franz *et al.* (1997); samples collected for 1987, 1990 and 1992 in snow layers after first year loss;
[c] Concentration data from Gregor *et al.* (1995); average concentration over a 30-year period from 1964/65 to 1992/93 (n = 34).

Table 6·3. Physical parameters sensitive to temperature change.

	Unit	Description/application
Henry's Law Constant (HLC)	Pa m^3/mol	Partitioning between air and water
Air–water partition coefficient (K_{AW})	unitless	$K_{AW} = HLC / RT$
Octanol-air partition coefficient (K_{OA})	unitless	Used as a proxy to model partitioning between air and organic phases such as vegetation, soil, sediment organic carbon, and particles in air and water
Vapour pressure (p^o)	Pa	Describes the tendency of a chemical to volatilize
Particle-gas partition coefficient (K_{PG})	m^3/µg	The ratio of chemical concentration on atmospheric particles ng/µg) to concentration in the gas phase

ever, if a given lake has a very low sedimentation rate and most of the organic carbon depositing to sediments becomes metabolized, it is likely that OCs associated with the particle flux will be released to the bottom water and re-mixed within the lake as has been observed in Lake Superior (Jeremiason *et al.*, 1994). This process of drawdown and remineralization could slowly ramp up water column concentrations below the thermocline, especially if ice cover is sufficient to hinder exchange during lake turnover. It appears that climate change has the potential to cause substantive physical and biological changes in northern lakes that would alter OC pathways, but current knowledge is insufficient to predict what these changes might be.

6.3.4. The effect of warming on chemical partitioning and degradation

Physical–chemical properties sensitive to temperature include vapour pressure (p^o), Henry's Law Constant (HLC; alternatively expressed as the air-water partition coefficient, K_{AW}), octanol-air partition coefficient (K_{OA}) and the octanol-water partition coefficient (K_{OW}) (Table 6·3). The particle-gas partition coefficient (K_{PG}), which depends on both particle composition and chemical composition, also varies with temperature.

The extent to which chemicals are associated with aerosols is fundamental to their atmospheric transport to the Arctic. Association with particles may, on the one hand, slow or reduce transport to the Arctic through temporary or permanent deposition to surfaces; while on the other hand, the association may protect a chemical from oxidation during transit to the Arctic. The potential for temperature to alter partitioning between gas and aerosol phases appears greatest for chemicals that exhibit log K_{OA} values in the range of 11 to 14 (Figure 6·7). For example, DDT varies from being over 70% particulate-bound in winter (–30°C) to occurring almost entirely in

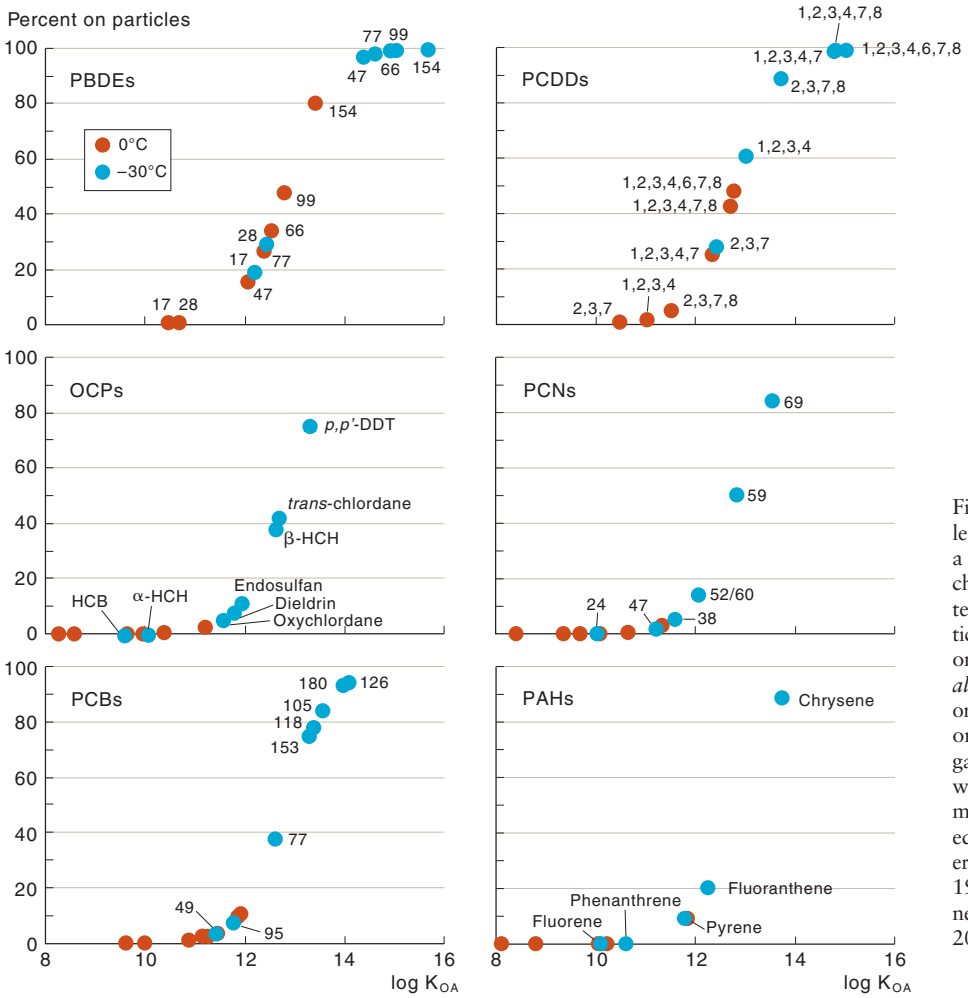

Figure 6·7. The partitioning of selected persistent organic pollutants as a function of K_{OA}. The figure shows changes in partitioning between winter (–30°C) and summer (0°C). Particulate fractions were calculated based on equations developed by Finizio *et al.* (1997) from field measurements of organochlorines. Calculations are based on measurements of particulate organic carbon in air at Alert during winter and summer 1998-1999 (Sharma *et al.*, 2002). Temperature-adjusted K_{OA} values were taken from the literature (Harner and Bidleman, 1996, 1998; Harner and Shoeib, 2002; Harner *et al.*, 2000; Shoeib and Harner, 2002).

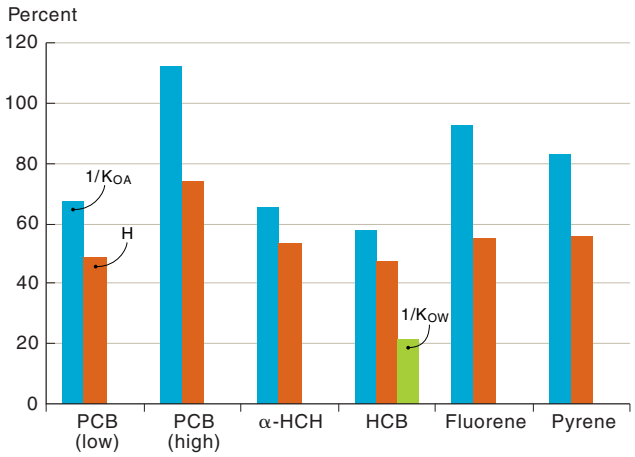

Figure 6·8. Predicted percent changes in H, $1/K_{OA}$ and $1/K_{OW}$ associated with a 5°C increase in temperature for selected chemicals. 'High' and 'low' represent the upper and lower values reported for PCBs. Values are based on ΔH_H (enthalpy of phase change associated with transfer from water to air, kJ/mol), ΔH_{OA} (enthalpy of phase change from octanol to air), and ΔH_{OW} (enthalpy of phase change from octanol to water) (Bidleman *et al.*, 2003).

the gas phase in summer (0°C). Similar changes in partitioning are evident for many of the other OCs illustrated in Figure 6·7. Chemicals with K_{OA} values at –30°C of about 11 to 12, therefore, are most sensitive to change in atmospheric transport as a result of temperature rise.

At 0°C most of the chemicals shown in Figure 6·8 occur in the gas phase, implying that they are easily advected by air but will be prone to photolytic degradation. Timing is important: the transport processes that produce Arctic haze in spring may alter substantially if warming comes earlier in the season, with consequent change to the particle–gas partitioning. Several of the higher molecular weight polybrominated diphenyl ethers (PBDEs) and polychlorinated dibenzo-*p*-dioxins (PCDDs) are appreciably associated with particles even at 0°C. Depending on their susceptibility to photolysis, particle-association may actually protect these compounds during transport which might explain why the increase in PBDEs in the Arctic closely follows the production and usage to the south (Ikonomou *et al.*, 2002) – which is not the case for the PCBs and other OCs which are mostly in the gas-phase during summer. Despite cold winter temperatures, many PCB, organochlorine pesticide (OCP) and polychlorinated naphthalene (PCN) compounds remain in the gas phase. Winter warming projected by climate models may therefore facilitate their transport during the period of year when reduced sunlight makes them less vulnerable to degradation.

Chemical partitioning between environmental media (air, water, soil, biota) can be described using three partition coefficients – K_{OA}, K_{AW}, and K_{OW} (Gouin *et al.*, 2000). K_{OW}, which is a ratio of two solubilities that both increase with temperature, tends to vary weakly with temperature, as shown by the modest 20% increase for hexachlorobenzene (HCB) in response to a 5°C increase (Figure 6·8; Bahadur *et al.*, 1997). A 5°C temperature rise produces a more substantive (60-100%) increase in $1/K_{OA}$, which will be manifest as an increase in volatility and greater potential for atmospheric transport. Under warming conditions, more chemical will partition out of surface soils and aerosols to enter the gas phase. Thus, if global warming occurs, cycling of chemicals through the atmosphere will increase. On the other hand, models suggest that temperature contrast between the equator and the pole will decrease (Figure 2·1) (Zwiers, 2002). Accordingly, kinetic processes will increase with temperature rise but the overall thermodynamic forcing toward polar regions will decrease with reduced global thermal contrast. K_{AW} also increases significantly (40-70%) under warming conditions, which would favor evasion from surface waters. This would be particularly important for OCs such as the HCHs which are at, or in the case of α-HCH over, saturation in Arctic waters. The predicted increase in K_{AW} associated with a 5°C increase in water temperature corresponds to tonnes of HCHs that could be forced back into the atmosphere each year. However, temperature increases of 5°C are unlikely to occur in partially ice-covered regions where temperatures will be buffered by melting ice. Nevertheless, recent changes in the AO index (Figure 3·1) and model projections (Figures 2·1 and 2·2) indicate that a large number of lakes and major areas of Arctic shelves could be subject to such changes.

Loss of chemicals occurs during transport in the atmosphere through reactions with hydroxyl (OH) radicals, nitrate radicals (NO_3) or ozone (O_3), through photolytic oxidation and through sorptive partitioning to other phases (e.g., aerosols, precipitation, vegetation) with subsequent deposition. Although photolytic reactions do not have a strong dependence on temperature they will be affected by cloud cover which is predicted to increase with global warming (IPCC, 1995). Increased cloud cover will also result in lower OH radical concentrations and less chemical removed by this and other photolytic pathways.

The dominant removal processes in soil and water include hydrolysis, photolysis, redox reactions, microbial degradation and removal through soil-surface–air partitioning. Of these, only photolysis is not strongly dependent on temperature. The influence of temperature on the rate constant, k, is usually described using the Arrhenius expression,

$$k = Ae^{-E_a/RT} \qquad \text{Eqn 1}$$

where A is a constant, R is the gas constant, T is temperature, and E_a is the activation energy. Based on hydrolysis activation energies of 78 to 85 kJ/mol (Ngabe *et al.*, 1993) for α-HCH and γ-HCH respectively, a 5°C increase would increase removal rates by ~85 to 95%. The increase would be even greater if it is considered that the dissociation constant for water (K_W) (e.g., at pH = 8) increases with temperature, resulting in more OH^- ions. Activation energies associated with redox reactions are not reported for OCs but are usually assumed to be about 50 kJ/mol (Tratnyek and Macalady, 2000) which imply about a 50% increase in reaction rate with a 5°C rise in temperature.

Microbial degradation also follows the Arrhenius equation, but few studies report E_a. As a general rule, the biological activity in the mesophilic range (5-35°C) doubles for every 10 to 15°C temperature rise which implies an E_a of 30 to 45 kJ/mol (Viessman and Hammer, 1985). Arctic microbial populations exhibit a large diversity and abundance (Ravenschlag *et al.*, 2001; Sahm and Berninger, 1998) and are typically cold-adapted, being able to maintain efficient rates of organic degradation and mineralization down to the freezing point of

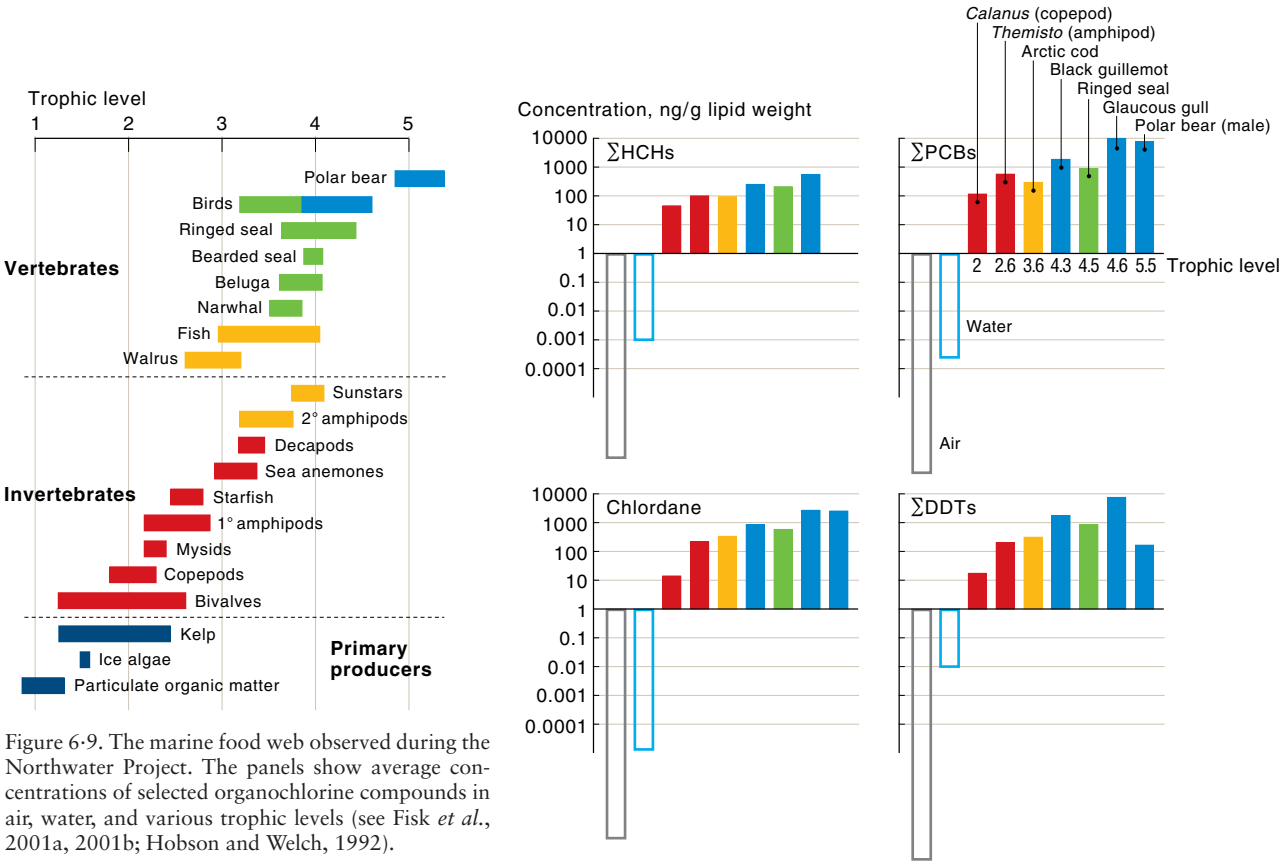

Figure 6·9. The marine food web observed during the Northwater Project. The panels show average concentrations of selected organochlorine compounds in air, water, and various trophic levels (see Fisk *et al.*, 2001a, 2001b; Hobson and Welch, 1992).

seawater (–2°C) (Arnosti, 1998; Sagemann *et al.*, 1998). This suggests that warming may be accompanied by consequences of adaptation or population change, but not necessarily that microbial degradation rates will increase. For example, reduced degradation of methyl dichlorprop was observed in experiments where field plots were warmed to a temperature of 5°C above normal for several years (Peterjohn, 1994).

Harner *et al.* (1999) found *in situ* microbial removal rates for α- and γ-HCH in cold Arctic Ocean waters to be surprisingly fast ($t_{1/2}$ for (+)α-HCH, 5.9 yr; (–)α-HCH, 22.8 yr; γ-HCH, 18.8 yr). Assuming an E_a of 50 kJ/mol, a 2 to 5°C temperature rise in the upper Arctic Ocean would imply a reduction of these half-lives by 20 to 50%. Microbial degradation was estimated to account for over 30% of the removal of HCH suggesting that a small temperature rise could push this proportion to over 50%.

Aside from temperature, alterations in other environmental characteristics (e.g., soil moisture, soil and water pH, nutrient levels, vegetation cover and type) will be tied to global warming and will affect the composition and density of microbial populations. For instance Lewis *et al.* (1999) found differences in microbial preference for microbes inhabiting forested versus pasture soils. They also showed that the enantioselectivity (preference of the microbial population for a + or – enantiomer of a chiral compound, i.e., a compound exhibiting mirror-image forms) changed with organic nutrient enrichment. Other studies have observed high rates of microbial degradation of HCHs in Arctic lakes (Law *et al.*, 2001) and watersheds (Helm *et al.*, 2000). Law *et al.* (2001) found that enantioselective degradation of α-HCH was greater in small, High Arctic lakes and streams compared to temperate lakes and wetlands. They concluded that low nutrient levels in the Arctic systems resulted in an adapted microbial population that was more capable of degrading organic contaminants. Thus biodegradation of chemical residues in soil and water will be altered as microbial populations adapt to changing climate. The complexity and uncertainty associated with these changes however, does not, at the present time, allow prediction of whether global warming will enhance or diminish chemical removal by this pathway.

Putting many of these concepts into a numerical model, McKone *et al.* (1996) investigated the effect of a 5°C temperature rise on the health risk from HCB. They found surprisingly little consequence from the projected 5°C temperature increase. Indeed, their results showed that warming would actually reduce exposure as it would enhance degradation and tend to force HCB out of water and into air. The critical step, controlled by the sensitivity of air–water partitioning to temperature, acts at the bottom of aquatic food webs which, due to biomagnification, are still of great significance for dietary exposure and related health risks.

6.3.5. The effect of altering food web structure

Biomagnification can concentrate fat-soluble compounds, such as the OCs, by factors as high as 10^5 to 10^9 from water to apex predators (Fisk *et al.*, 2001a; Kidd *et al.*, 1995b; Muir and Norstrom, 1994; Muir *et al.*, 1999). The distribution of contaminants in air, water, and the first step in the food web (phytoplankton, particulate organic carbon), can be predicted simply by applying appropriate partition coefficients (e.g., K_{AW} and K_{OW}) (Figure 6·8). Chemical partitioning, which is based solely on thermodynamics, provides a crucial platform upon which biomagnification can then operate (Figure 6·9). A chemical at equilibrium will have identical fugacity in

the media in question (e.g., air, water, oil) and this makes it relatively simple to predict how temperature will alter its distribution.

Biomagnification, however, cannot be explained solely by thermodynamics, and it requires energetic processes to produce the elevated concentrations in top predators. These processes can be considered in part as a reduction of fat – the solvent containing OCs – through metabolism. The processes are complex and can lead to variability simply due to bio-energetics; the offloading of contaminants to offspring by nursing mammals is a classic example (Addison and Smith, 1998). It is not the intent here to conduct a thorough review of bioaccumulation and biomagnification except to state that the complexity of the process offers the opportunity for climate change to act in subtle ways. The data shown in the four panels of Figure 6·9 show an almost linear relationship between the log (contaminant burden) and trophic level. The slope of this relationship indicates the multiplication factor involved for each step in trophic level so that, for example, one step would multiply DDT concentration by perhaps as much as 6 (neglecting polar bears) whereas the other contaminants experience factors of 2 (ΣHCH), 3 (ΣPCB) and 4 (chlordane). The removal or addition of trophic levels in the food web mediated by climate change, therefore, will not have the same effect for all contaminants; for example, Figure 6·9 suggests that DDT will be the most sensitive to this kind of change.

Another way in which food web structure can effect change is by bifurcation. For instance, altering the coupling between pelagic and ice production and the benthos can change the relative proportions of organic carbon (and contaminant) that enter pelagic or benthic food webs. However, these changes in pathway do not alter the relationship between contaminant concentration and trophic level. In this context it can be seen that walrus (*Odobenus rosmarus*) feeding on a benthos enriched by strong coupling with primary production will be exposed to lower (by factors of 10 or more) OC concentrations than if they switch to predation on seals.

6.3.6. The epontic food web and changes in ice climate

The entry of contaminants to a stratifying surface layer from ice melt in spring offers a mechanism vulnerable to climate change. However, recent studies do not appear to show higher concentrations in epontic fauna than in zooplankton (Borgå *et al.*, 2002). Organochlorine concentrations in epontic amphipods (*Apherusa glacialis*, *Gammarus wilkitzkii*, *Onisimus* spp.) and zooplankton (*Calanus hyperboreus*, *Thysanoessa inermis*, *Parathemisto libellula*, and Chaetognatha) from the marginal ice zone near Svalbard could mostly be explained by diet, with habitat (sea-ice underside versus the pelagic zone) accounting for a smaller part of the variance. Epontic amphipods had higher concentrations of HCB, γ- and α-HCH, while DDTs, PCBs and chlordanes did not differ between epontic and pelagic habitats. This pattern of uptake can be explained by the vertical distribution of OCs in the water column: higher concentrations of HCHs and HCB are found near the sea surface (Harner *et al.*, 1999; Jantunen and Bidleman, 1998; Tanabe and Tatsukawa, 1983) whereas particle reactive compounds like DDTs and highly-chlorinated PCBs adsorb onto sinking particles to produce a more homogenous vertical distribution (Tanabe and Tatsukawa, 1983). Furthermore, most particles transported by sea ice are not available to epontic biota because they are released in the marginal ice zone when the ice melts (Ramseier *et al.*, 1999) and descend rapidly, carrying the adsorbed contaminants with them. For OCs that exhibit strong gradients in the upper ocean, the loss of ice and hence of epontic fauna can alter the dietary exposure of higher trophic levels like seabirds and seals.

6.3.7. Food deprivation or shifts in diet

Many of the Arctic top predators undergo periods of fasting forced by lack of food, seasonality of food, or inability to access food. Perhaps the best documented example is the stress to the Hudson Bay polar bear (*Ursus maritimus*) population deprived of their ability to hunt seals during spring due to change in the spring ice climate (see Figure 3·23 and Stirling, 2002; Stirling and Lunn, 1997; Stirling *et al.*, 1999). The burning of stored fat through metabolism results in release of archived fat-soluble contaminants and, potentially, an increase of contaminant burden in the remaining fat reservoir. Longer periods of starvation due to change in ice or change in prey populations could lead to higher doses of OCs sequestered in fat – usually at a time when the animal can least afford it. Although the concern with nourishment-deprived polar bears has received much attention, similar circumstances probably apply to other species such as common eider (*Somateria mollissima*; Olafsdottir *et al.*, 1998) and Arctic char (*Salvelinus alpinus*; AMAP, 2003b).

Species that have dietary flexibility may respond to ecosystem change by switching to alternate prey, again with consequences on their OC intake. For example, the large variation in OC concentrations in the livers of glaucous gulls (*Larus hyperboreus*) from the western Barents Sea correlates with nitrogen isotopic ($\delta^{15}N$) composition (AMAP, 2003b). Nitrogen isotopes provide a well-established, reliable chemical indicator of the trophic level of an animal's prey (Hobson and Welch, 1992) therefore implying that much of the variation in OC concentration in glaucous gull livers can be explained by the dietary choices the birds make – or are forced to make. Recent decreases in PCB concentrations in Svalbard minke whales (*Balaenoptera acutorostrata*) might superficially be ascribed to the banning of PCB manufacture during the 1970s. However, it seems more likely that this decline reflects a dietary switch from capelin (*Mallotus villosus*), whose stocks collapsed in 1992 to 1993, to krill which are lower down the food chain (AMAP, 2003b). Polar bears also display a range in prey that can explain regional variation in OC burdens. For example, Chukchi and Bering Sea bears feed more heavily on Pacific walrus which are less contaminated than ringed seals (*Phoca hispida*) because they are at a lower trophic level, whereas bears from Svalbard feed on more heavily contaminated harp seals (*Phoca groenlandica*). These latter results indicate that climate variables expressed through prey availability and biological condition can have a considerable influence on the exposure of apex feeders to OCs.

6.3.8. Altered migration pathways and invading species

Migratory species, including whales, fish, and birds, can obtain contaminant loadings in one location and release them in another, and migrating animals can be subject to varied exposure as they feed along their migration path. For one Alaskan lake, the loadings of OC contaminants returned by anadromous fish exceeded those entering the drainage basin from the atmosphere (Ewald *et al.*, 1998). The recent expansion in the range of Pacific salmon (*Oncorhynchus* spp.) into rivers farther north in the Arctic (Babaluk *et al.*, 2000) could, likewise, have an impact on contaminant budgets for the rivers or lakes that they enter. Certainly, these fish provide a new 'vector' for delivering contaminants to species that predate upon them or depend upon a food web supplied by their carcasses. Given the higher concentrations of β-HCH in the North Pacific Ocean than in the Arctic (Li *et al.*, 2001) it seems likely that anadromous fish may provide an important, climate-sensitive vector for β-HCH into Arctic freshwater environments.

If the spatial distribution of contaminants is controlled by processes subject to climate change, then exposure during population migrations can also alter through climate change. An intriguing example of how such a process may operate has recently been described for the Bering/Beaufort bowhead whale (*Balaena mysticetus*) migration. The whales reflect in their body burdens the change in α- and β-HCH composition between the Bering and Beaufort Seas (Hoekstra *et al.*, 2002). The ocean composition for the HCHs is probably controlled by large-scale physical processes (e.g., rainfall and air–sea partitioning – see Li *et al.*, 2002) as are migratory routes (Dyke *et al.*, 1996b; Moore *et al.*, 1995), each of which is sensitive to climate change. Therefore, alteration in HCH loading of the ocean caused by change in winds, ice cover or rainfall pattern, or alteration in the feeding locations of the whales caused by change in ice distribution, will similarly alter the exposure to HCHs.

Finally, invasions of new species fostered by climate change, overfishing or by introduced exotic species also have the potential to re-structure food webs. An elegant example of how dramatic such change can be, both in trophic organization and contaminant pathways, was provided by the invasion of the zebra mussel (*Dreissena polymorpha*) into the Great Lakes (Morrison *et al.*, 1998, 2000; Whittle *et al.*, 2000). Following their invasion in 1988, zebra mussels have led to a decline in phytoplankton and rotifer densities, the water has become clearer (by the removal of particulates), and the deposition of nutrient-rich pseudofeces at the bottom has altered benthic habitat density and structure. In turn, the zebra mussels have become an important prey item altering the trophic structure within the lake as reflected by the stable isotopic composition of fish. In another example, devastating change in the Black Sea ecosystem has resulted from the population explosions of phytoplankton and jellyfish (Daskalov, 2002). In this case, top-down trophic change may have been initiated by overfishing of apex feeders. Clearly, the migration northward even of a humble filter feeder or jellyfish, potential products of changing water properties or organic carbon supply, could have unexpected impact on contaminant cycling in coastal seas.

6.3.9. Organochlorine compounds, disease, and epidemics

During the past decade or so, there has emerged much evidence that mass mortalities in marine mammals may occur as a result of a combination of factors including disease vectors, population stress and contaminants, each of which may be affected by climate change (see Lavigne and Schmitz, 1990; Ross, 2002). The complexity of this interaction provides fertile ground for surprises. Some disease outbreaks have been observed following migrations associated with large-scale ecological change, and some have derived from the introduction of viruses from domestic animals. But it is the addition of immunotoxic chemicals, such as many of the POPs, that may provide the trigger for disease to emerge (Ross *et al.*, 2000; Vos and Luster, 1989). The widespread distribution of the canine distemper virus, or a closely related morbillivirus in seals from Greenland led Dietz *et al.* (1989) to speculate on the possibility that large-scale migration of harp seals from the Barents Sea to northern Europe in 1986 to 1987 might have provided a disease vector. The co-factors of a naïve marine mammal (seal) population in coastal Europe manifesting suppressed immune systems through high contaminant PCB burdens would then have provided the foundation for the epidemic (Heide-Jørgensen *et al.*, 1992).

Within the Arctic, top predators would be at greatest risk due to their high exposure to contaminants, and marine mammals probably face the added stress of changes in ice climate. Accordingly, indications of immunosuppression have been found in polar bears, northern fur seals (*Callorhinus ursinus*) and glaucous gulls (AMAP, 2003b). In particular, the polar bears of the Kara Sea, Franz Josef Land, East Greenland and Svalbard would seem especially vulnerable. Firstly, these bears exhibit inordinately high contaminant burdens (Andersen *et al.*, 2001; Norstrom *et al.*, 1998) and these high burdens may well derive partly from the enhanced connection between this region and Europe/North America under the strong AO^{+}/NAO^{+} conditions of the 1990s (Figures 3·2 and 3·17). Secondly, as previously discussed, change in ice climate and in marine ecosystems may have provided the added stress of malnourishment. Lastly, it seems that these bears already have sufficient contaminant burdens to exhibit health effects (Bernhoft *et al.*, 2000; Skaare *et al.*, 2001).

6.4. Hydrocarbons

Hydrocarbons in the Arctic derive from combustion and petrogenic sources (Yunker *et al.*, 1995). The pathways of these two sources of hydrocarbon differ substantially as will their sensitivity to climate change. Hydrocarbons of anthropogenic origin pose two kinds of problem; polyaromatic hydrocarbons (PAHs) and their oxidation products are toxic (Zedeck, 1980), and spilled oil has direct, well-known effects on biota, especially those that inhabit interfaces between water, air, ice and sediments. (Patin, 1999; Wolfe *et al.*, 1994).

6.4.1. Combustion PAHs

Combustion PAHs are well-known products of natural fires and human-related combustion processes (e.g., au-

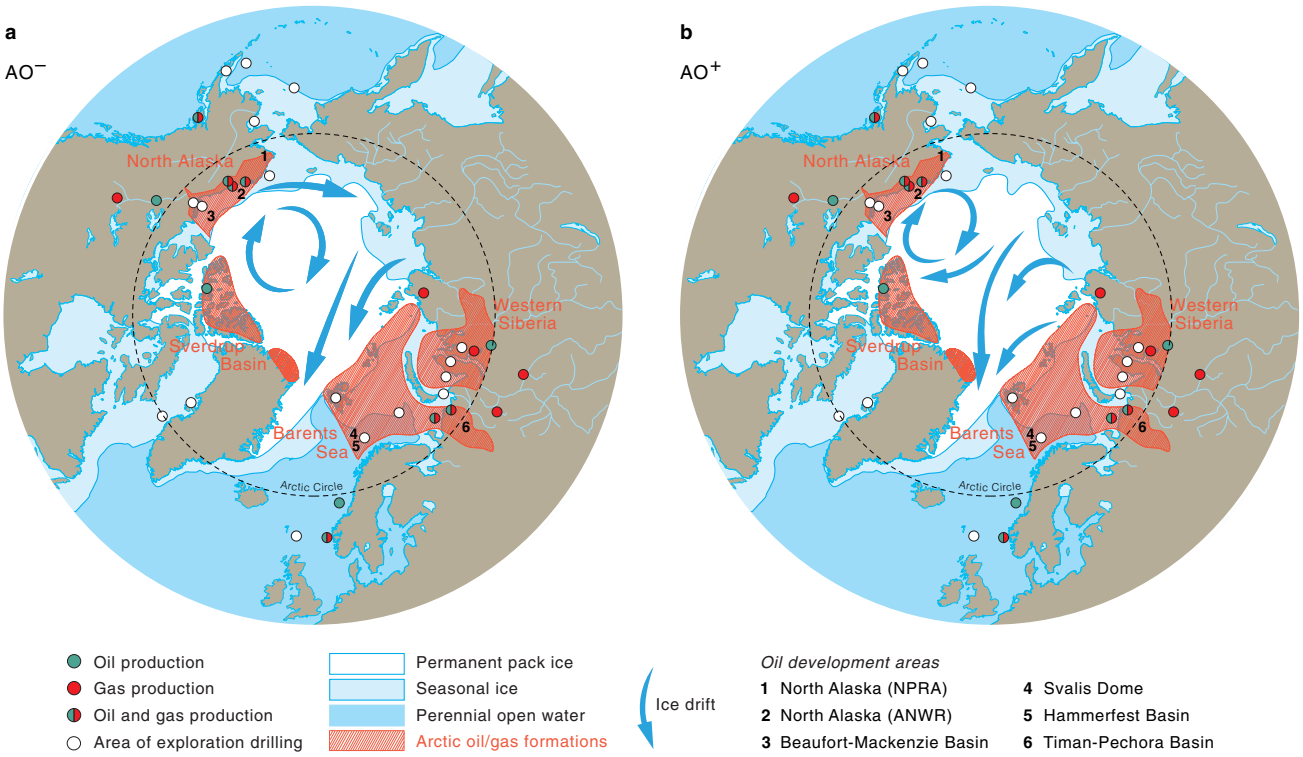

Figure 6·10. Oil/gas-bearing regions and locations of oil/gas production together with the ice-drift field under a) AO⁻ conditions and b) AO⁺ conditions (ice motion based on Rigor *et al.* (2002) and oil data from Bakke *et al.* (1998)).

tomobiles, liquid and solid fuel burning, waste incineration, metallurgy, etc.). In the atmosphere, PAHs partition between the vapor phase and particulates (see for example Figures 14 and 15 in Macdonald *et al.*, 2000a) and are detected at remote Arctic locations following transport over long distances (Halsall *et al.*, 1997; Macdonald *et al.*, 2000a; Patton *et al.*, 1991). There is a strong seasonality in PAH concentrations in air at Alert (Ellesmere Island, Canada) with colder months (October-April) displaying concentrations about ten times higher than in warmer months (May-September). This suggests that the winter haze phenomenon that transports heavy metals across the pole from Eurasia also transports industrial combustion products. Therefore, much of what has been said about aerosol metals and climate change in section 6.1.1 relates directly to PAHs; altered wind patterns (Figure 3·2) and enhanced precipitation (Figure 3·4) have the potential to change pathways and to deposit PAH aerosols over parts of the Arctic Ocean especially toward the southern Eurasian Basin. Furthermore, temperature increases may shift the equilibrium from particulate to vapor phase for PAHs like pyrene, fluoranthene, phenanthrene and anthracene, which at Arctic temperatures are partially distributed between air and solid phase (Figure 6·7).

In addition to the strong seasonal signal of industrial PAHs observed in the Arctic, outliers (samples with abnormally high PAH concentrations) are also observed during summer months, particularly at Tagish (southwest Yukon, Canada), and these have been assigned to forest fires (Macdonald *et al.*, 2000a). Forest fires are projected to increase through climate change as a result of warmer continental temperatures (Figure 2·1) and less precipitation in continental interiors. A general increase in atmospheric PAHs in the Arctic deriving from such biomass burning is thus likely, and this increase will probably have an impact on small rivers, e.g., in northern Canada, which already receive almost all their PAHs from combustion sources (Yunker *et al.*, 2002). Loss of permafrost and enhanced erosion of peat may also contribute enhanced amounts of relict PAHs to lakes and rivers (Yunker *et al.*, 1993).

6.4.2. Petrogenic hydrocarbons and oil

A leading concern for the Arctic is the risk of oil spills from both onshore and offshore exploration and production (Bakke *et al.*, 1998; Patin, 1999). Climate change that produces an ocean margin substantially clear of ice will undoubtedly encourage further offshore exploration, perhaps in more remote locations. Producing oil from the Arctic reserves, especially those on the remote shelves, has the associated problem of transporting produced oil to markets in the south, either by ship or pipeline.

It is clear that changes in the ice drift associated with changes in the AO index will have a dramatic influence on where spilled oil will go if it enters the ice pack (Figure 6·10). For example, in the Canadian and Alaskan sector of the Arctic Ocean during AO⁻ conditions, the oil will follow ice into the East Siberian Sea to traverse the Russian shelves and then exit to the Greenland Sea. During AO⁺ conditions, oil from the same location would tend to remain within the Beaufort Gyre potentially to return within a few years to where it was spilled.

Oil spilled over the Russian shelves, or entering their coastal seas from spills into rivers, would tend to track directly across the Arctic under AO⁻ conditions (Figure 6·10 a). However, oil spilled under AO⁺ conditions could move more to the east, with a slight chance that it might

reach the Canada Basin and then the Canadian Arctic Archipelago through which it would then have to pass. It is uncertain how viable this latter route might be for oil spilled in the Kara or Laptev Seas, but evidence from tree dendrology suggests that there have been periods during the Holocene when communication from Siberia to the Canadian Arctic Archipelago has been mediated by ice drift (Dyke and Savelle, 2000; Dyke *et al.*, 1997). Indeed, the transport of trees by Arctic sea ice and the change in that transport with time provides some of the best evidence of where oil spilled in Arctic coastal regions is likely to travel (see for example, Eggertsson, 1994a,b).

Pipelines transporting oil across land in the Arctic are vulnerable to enhanced permafrost degradation. Regions such as the Komi Republic, which experienced large oil spills onto the tundra in 1994, exemplify the difficulty. Warming and melting of frozen ground will put corroded pipelines, many working beyond their design life, at greater risk. Oil already spilled onto land within the Arctic drainage basin may become more mobile, entering water courses as the hydrological cycle becomes more vigorous.

Another connection between spilled oil and climate change derives from the projected increase in incident UV radiation (Weatherhead and Morseth, 1998) which could lead to an increase in photo-enhanced toxicity of spilled oil (see, for example Barron and Ka'aihue, 2001; Pelletier *et al.*, 1997). Toxicological assessments of oil made in the presence of UV light reveal a toxicity of up to 1000 times greater than that measured under the traditional fluorescent light. Furthermore, photo-enhanced toxicity of oil can occur at the intensities and wavelengths measured for UV in aquatic water columns suggesting that increased incident UV radiation projected for polar regions may, in addition to many other effects on ecosystems (Weatherhead and Morseth, 1998), enhance damage done by spilled oil.

Chapter 7

Time Series

The value of time series, both for climate-related variables (Hare and Mantua, 2000; McGowan, 1990) and for contaminants (AMAP, 1998), is undisputed. However, recognition of the potential of climate variables to produce variance in contaminant time series has all but been neglected (Macdonald *et al.*, 2002b). This report has discussed numerous examples of how global change can alter delivery of contaminants to and within the Arctic; alteration in wind fields and precipitation forced by the Arctic Oscillation (AO) being but one simple example. That the leading 'global distillate', water, provides one of the clearest detectors of global temperature change in its isotopic composition (Fischer *et al.*, 1998) should generate considerable anxiety about time series of volatile and semi-volatile compounds established from, e.g., atmospheric concentrations, ice cores, or sediment cores, even if the pathway between emission and point of measurement is reasonably direct. Examples closer to contaminant time series can be found. For instance, the mercury (Hg) record in ancient Antarctic ice has been suggested as a proxy for ocean productivity (Vandal *et al.*, 1993) and cadmium (Cd) has been applied in paleo ocean-productivity histories (Saager and deBaar, 1993; Shen *et al.*, 1987).

The environment can be monitored at many points (Figure 7·1), each of which will tell a separate story. For example, PCB concentrations might be measured as follows: in air at Alert every two weeks; in an ocean profile collected annually; in biota or surface sediments collected every five years; or in a dated sediment or ice core. What these PCB time-series data will indicate depends on how many environmental processes have an opportunity to operate on the original signal (the emission), and how the recorder (the medium being monitored) itself actually functions to modify the signal. Much research has been conducted to understand and account for the latter (e.g., studies of organochlorine compound (OC) cycles in female mammals; increase in Hg with age of fish, diagenesis in sediment cores; food-web studies, etc.) but the difficulties associated with climate variability creeping into the record have been mostly ignored. Climate variability by itself may cause aliasing. Aliasing, the result of sampling being conducted at a rate that does not resolve the natural frequencies (or period) of the system, can lead to situations where either the phenomenon is missed completely, or interpreted to have a lower frequency (or longer period). In the worst case, a cycle may be interpreted as a trend. For example, the

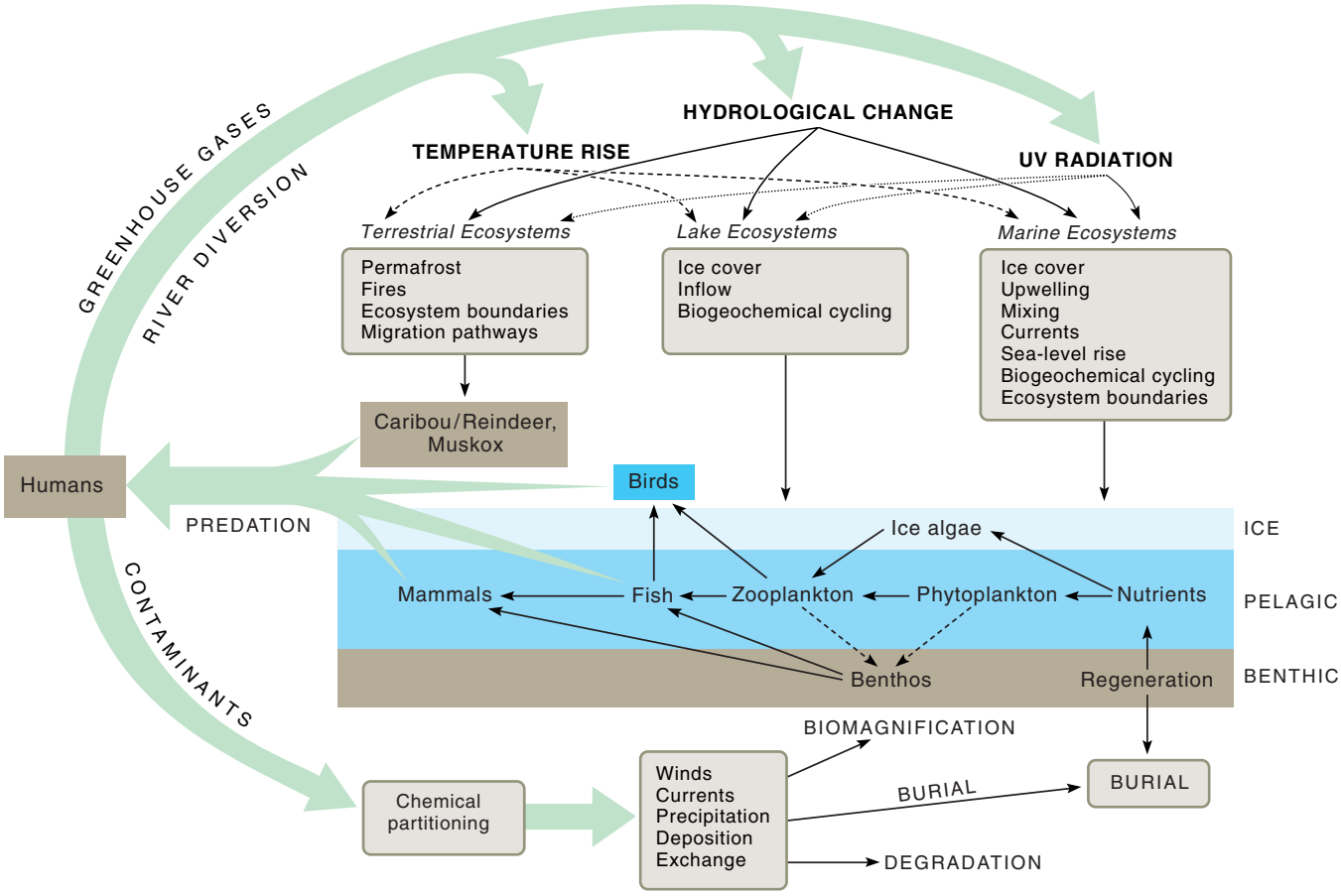

Figure 7·1. A schematic diagram of the pathways contaminants must traverse between emission and points at which environmental time series data are collected for the Arctic.

AO's time scale of five to seven years (Proshutinsky and Johnson, 1997) makes it very difficult to assess the role of such sub-decadal variation in data collected at five-year intervals. Furthermore, a trend from a time series collected from, for example, 1980 to 2000 will potentially carry a large bias, either positive or negative, produced by a switch in the middle of the record from generally AO^- to strong AO^+ conditions. Climate change often may provide an alternate hypothesis for time-series contaminant data. A few examples are described briefly in sections 7.1 to 7.3.

7.1. Time series derived from sediment-core records and surface sediments

Well-dated cores from lakes and oceans provide an established means of estimating historical fluxes of contaminants (Lockhart *et al.*, 1995, 1998; Muir *et al.*, 1996). Provided the uneven spatial distribution of lake sedimentation rate (focusing) is taken into account, fluxes to sediments can be used to infer the flux to the surface of the water and thereby allow comparison with fluxes at other sites or with emissions. Increases or decreases in contaminant concentrations with depth in a sediment core are then often used to infer historical changes in emissions. However, between emission and burial at the bottom of the lake or ocean lie atmospheric transport, deposition to surface water or the drainage basin, attachment to particles, settling to accumulate at the bottom and, possibly, remobilization (Figure 7·1). Considering atmospherically transported contaminants, climate change may operate within this system to 1) change wind fields (important if there are atmospheric gradients); 2) change the efficiency of the air to ground transport (e.g., by altering precipitation or temperature); 3) change the efficiency of capture to sediments (by processes outlined in section 6.3.3.); 4) change the pathways of water currents; 5) change the supply of particles; and 6) change the carbon flux and metabolism in sediments thereby changing preservation.

Possibly the best example of how climate-related variables could interact with contaminant records in sediments has been revealed in lake sediments on Bjørnøya (Bear Island). The sediments of the lake Ellasjøen contain anomalously high OC concentrations that are attributed to large inputs of bird guano (AMAP, 2003b). What would a dated sediment core from such a lake show? It is clear that the record would contain a component deriving from the emission histories of various OC chemicals, but much of that signal would be mediated by birds. The population dynamics of birds, then, will alter the contaminant delivery over time depending on nesting locations and populations, and on the source and trophic level of their food which derives from the Barents and Nordic Seas (AMAP, 2003b), all of which are subject to climate variation. This example, which is particularly compelling in the way local and perhaps regional PCB patterns have been affected by biological sources (guano – see AMAP, 2003b; Enge *et al.*, 1998), is probably not an isolated example. There is also the importance of fish as agents for transporting OCs and Hg (Ewald *et al.*, 1998; Zhang *et al.*, 2001) which, together with high and variable anadromous fish escapements (Finney *et al.*, 2000, 2002), probably imprint an as yet uninvestigated influence on the contaminant records in coastal lakes of Alaska.

Finally, natural variability in organic carbon flux to sediments can enhance sediment foraging (including small infauna and large animals such as diving birds, walrus, seals and belugas). A richer supply of metabolizable carbon feeds a more vigorous benthos (bivalves, worms, amphipods, etc.) and these, in turn, provide food to benthic feeders like birds, grey whales (*Eschrichtius robustus*) and walrus (*Odobenus rosmarus*). Variation in organic carbon flux, together with the potential for episodic colonization of sediments by new species under changing ocean climate (see for example Stull *et al.*, 1986), provides a strong caution on using surface sediment contaminant distributions to infer spatial or temporal trends in contaminants even where normalizing factors such as aluminum or organic carbon have been applied.

7.2. Time series in atmospheric concentrations

Data collections from air monitoring stations in the Arctic are of relatively short duration, extending back a couple of decades for metals (Sirois and Barrie, 1999), and less for OCs (see, for example, AMAP 2003a,b; Bailey *et al.*, 2000; Halsall *et al.*, 1998; Hung *et al.*, 2001; Macdonald *et al.*, 2000a; Stern *et al.*, 1997). There are fewer processes between the emission and the recorder for these measurements (Figure 7·1) but as discussed in previous sections, changes inherent in the AO (winds, ice cover, precipitation) are sufficient to imprint themselves on the emission before it reaches this recorder.

7.3. Time series in biological tissue residues

Marine and terrestrial biota have been collected to monitor bioaccumulating substances, especially the OCs and Hg (Addison and Smith, 1998; AMAP, 1998; Braune *et al.*, 1999; Wagemann *et al.*, 1995). Some programmes are based on an annual sampling strategy, however many utilize samples collected at very sparse intervals (typically 3-6 periods over a period of 20-30 years). α-HCH concentrations in seals collected at Holman Island clearly do not follow atmospheric emissions (see Figure 7·2) nor do they follow Arctic air concentrations which mimic emissions quite closely (see Figure 9 in Macdonald *et al.*,

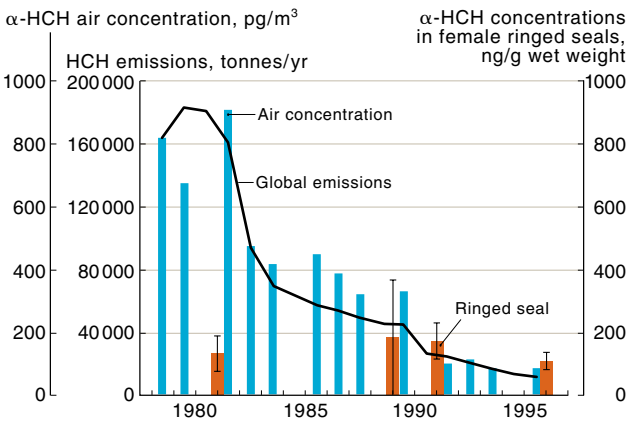

Figure 7·2. A comparison of time series concerning global emissions of α-HCH, mean concentrations of α-HCH in Arctic air and α-HCH concentrations in Holman Island seals. The air concentration data were measured at different stations by several research groups (for data sources see Addison and Smith, 1998; Li *et al.*, 2002).

2000a). Instead, they approximately follow Canada Basin surface water concentrations, which makes sense given that seals obtain their HCH burden from a marine diet. The burden of α-HCH is large in the surface waters of the Canada Basin (estimated at 1750 tonnes in the early 1990s, Macdonald *et al.*, 2000a), and not quickly changed. However, the burden can be altered by diversion of Russian rivers or by removal of ice cover, both of which occurred in the 1990s. Furthermore, change in the seal burdens can be caused by a regional change in the food web – something that has probably occurred as a consequence of changing ice cover and stratification (Melnikov *et al.*, 2002). The food web factor might be 'controlled for' by monitoring trophic level through isotopic measurements ($\delta^{15}N$) or by monitoring other components of the food web (e.g., Arctic cod (*Boreogadus saida*)) over the same time interval. The advantage of monitoring species at high trophic levels – that they are very sensitive to OCs and important to human diets – is somewhat offset by their sensitivity to variance from many factors other than emission strength (Figure 7·1). Given the five- to seven-year time scale inherent in change forced by the AO, time series data for biological tissue will be vulnerable to an unaccounted aliasing.

Chapter 8

Conclusions and Recommendations

The previous AMAP assessment (AMAP, 1998) described in detail the physical and chemical contaminant pathways to and within the Arctic (Barrie *et al.*, 1998). The present assessment is neither an extension nor a revision of that report; rather, it is a review of the environmental changes in the Arctic that are likely to have significant consequences for contaminant pathways. The remarkable changes recognized to have occurred in the Arctic during the 1990s were not addressed in the previous AMAP assessment. Nevertheless, that overview of how contaminants arrive at the Arctic and subsequently move through its physical systems is not incorrect, just incomplete. The synthesis provided here is also incomplete, for two reasons. Firstly, coherent studies of the alteration of pathways as they affect contaminants have not been conducted. This, therefore, means that the consequences of observed or projected change for contaminant pathways has to be inferred on the basis of the state of knowledge as outlined in the previous assessment. Secondly, change will continue to occur globally, especially within the Arctic, and the inevitable surprises will highlight that the current understanding of complex environmental systems is still very incomplete.

In the previous AMAP assessment, physical and chemical transport processes were considered separately from biological processes. In the equation of change, physical and biological systems interact with one another and both may have dramatic influences on the fate of a contaminant headed for the Arctic. Accordingly, in addition to the physical/chemical pathway changes that have occurred, or may occur in the Arctic, the present assessment also examines biogeochemical pathway changes. These have been considered in this report only in terms of organic carbon cycles or food webs.

In the previous AMAP assessment, the atmosphere was deemed the most important pathway relative to ocean and terrestrial/freshwater pathways for single-hop compounds. The most important message of the present assessment is that a contaminant's journey from its point of emission to its accumulation in an Arctic ecosystem is a conspiracy that cannot work without the compliance of many steps all of which can be altered by global change (Figure 7·1). The risks posed by contaminants to Arctic ecosystems cannot be fully understood by isolating components, and no one step can be considered more or less important than another. Chains break at the weakest link and so it is with contaminants; rapid transport to the Arctic in air may or may not be followed by entry into Arctic food webs (Macdonald, 2001).

The response time of natural systems is a key factor in establishing the role of a particular process in the equation of change (Ruddiman, 2000). Almost all contaminants exhibit a transient rise in their emission followed by a dramatic decrease after bans or controls are instituted. Depending on environmental response times and reservoir sizes, the unloading stage for a given contaminant, following bans on its emission, is not a simple reverse of its loading stage. The atmosphere may predominate initially as a transporting medium, but with controls, other slower-moving media (water, soil) may take over, especially if they comprise the largest reservoir of contaminant.

The previous AMAP assessment highlighted the importance of models for understanding contaminant transport, but stated that many processes are insufficiently understood to be included realistically in those models. That conclusion deserves strong re-emphasis and extension. Many of the transport steps affected by change (Figure 7·1) are not captured by models. Even if complete physical models were to be constructed, it would still not be possible to produce realistic scenarios for contaminants by virtue of the importance of organic systems. Eventually, fully-coupled atmosphere–ice–ocean–ecosystem models will be required to predict how natural systems will respond when contaminants are released or when contaminants are banned.

The previous AMAP assessment also highlighted the lack of spatial and temporal time series. These are, if anything, even more urgent now. These time series must include observations both of contaminants and the systems they enter. For this reason, it is crucial that scientists working on contaminant issues collaborate with climate change scientists to design appropriate observation networks. Such networks provide validation data sets for models and contribute to a systematic understanding of processes that are vulnerable to change. Many of the important climate variations occur at time scales from a few years to centuries; the dramatic shift associated with the Arctic Oscillation (AO) that occurred during the late 1980s is just one example. As a consequence, the short time series presently available for most contaminants are subject to aliasing from natural cycles and time-series data will need to be collected for some time before it will be possible to discriminate between sources of variance associated with climate change or variability. It is certain that assembling time-series data by collecting samples every five years or so would be a poor strategy in the Arctic in view of the natural time period of the AO (5-7 years).

All factors that can influence the contaminant transport system can change, including temperature, winds, precipitation, ocean currents, ice cover (on lakes, rivers and oceans), streamflows, and ecosystems. Perhaps physical/chemical changes (such as temperature and physical partitioning) are the easiest to model and understand, but they are probably the least important in the equation of change. For the Arctic, it is clear that changes associated with ice and precipitation (the 0°C isotherm), and with ecosystems (trophic structure, pathway bifurcation) have the greatest potential to alter contaminant pathways and exposure.

The previous AMAP assessment identified ice as a dominant, multi-compartment medium of the Arctic, but stated that its role in modulating contaminant fate was poorly understood. The changes that have recently occurred in ice climate, and that are projected to occur during the coming century make it even more imperative to fill this gap.

Natural climate variation, such as the AO, provides an opportunity to study change in the Arctic and to improve the ability to project the consequences of change. The marginal seas appear to be the important locations for observations and study because it is here that change is likely to occur first, and it is here that change is most likely to affect ecosystems that are valued by humans. It is clear that Hudson Bay, the Bering/Chukchi Sea and the Barents Sea have all have undergone substantial variation, especially during the 1980s and 1990s. The changes that have occurred throughout physical and biological systems are alarming but, as yet, only partially understood. The East Siberian Sea stands out as a region sensitive to change, having experienced large variation in ice cover and in runoff pathways along its coastal corridor during the 1990s. However, there is virtually no information on contaminant cycling in that region, no time series data and very little information on the response of this shelf's ecosystem to climate change.

Alteration of the ice climate in the coastal margin is likely to increase the risk of local contamination. Increased shipping due to more economic transport corridors and increased oil exploration and transport are two probable consequences of projected change. Furthermore, local contaminated sites (sewage lagoons, dumps, tailings ponds, soils contaminated by spilled oil) and pipelines whose strategy of containment strongly depends on permafrost stability are likely to be at risk. These sites and any future such sites should be improved/developed in the light of projected change.

8.1. Contaminants of concern

8.1.1. Heavy metals

Of the metals, mercury (Hg) continues to generate the greatest concern. Despite declining anthropogenic emissions, at least in the period between the 1980s and the 1990s, the Arctic ecosystem appears to be increasingly exposed to Hg. It is unclear why this is so because the complete Hg pathway has not been adequately studied. The connection between atmospheric transport and deposition to Arctic surfaces (Hg depletion events) shows the Arctic to possess a unique, climate-sensitive process that may explain much of its susceptibility to Hg contamination. However, the pathway for Hg between its deposition to surfaces, especially following polar sunrise, and its concentration in apex aquatic feeders is very poorly known. It is recommended that study continue on the Hg cycle in the Arctic with emphasis on the processes implicated in Hg depletion events and in the biogeochemical cycling of Hg in ice-covered environments. Such studies should consider the role of ice cover and primary production in the Arctic biogeochemistry of Hg.

Increase in the distribution of wetlands, e.g., with the melting of permafrost, may also provide a mechanism exhibiting the 'reservoir effect' by which Hg presently trapped in organic carbon phases of frozen ground is released. It is recommended that the effect of permafrost destruction and accompanying change in surface hydrology be studied within the Arctic for its potential to affect regional Hg cycles.

Increased erosion or permafrost melting in drainage basins may produce enhanced fluxes of Hg to Arctic lakes and coastal zones. It is recommended that studies of Hg cycling be conducted at lakes or along coasts that are undergoing change if such can be identified.

8.1.2. Radionuclides

Artificial radionuclides in the Arctic are not generally considered a radiological hazard, to humans at least, but, rather, excellent tracers of ocean pathways. For this reason, it is recommended that time series for artificial radionuclides in the Arctic Ocean continue to be maintained, because the pathways and changes in pathways that these tracers indicate, are relevant for other contaminants. One pathway for artificial radionuclides continues to prove difficult to assess – sea ice. Evidence for the concentration of radionuclides in ice far from known sources suggests that this vector may cause exposure to humans and animals that use sea-ice surfaces. Furthermore, this pathway appears vulnerable to climate change. It is therefore recommended that further measurements of radioactivity in ice should be made and that these should be accompanied by measurements (geochemical properties) and models (back trajectories) to shed light on the origin of the ice.

8.1.3. Organochlorine compounds

Organochlorine compounds (OC) continue to pervade global ecosystems despite success in curtailing many of the emissions. The risk they provide to Arctic ecosystems is predicated on a conspiracy between physical, chemical and biological pathways that produces relatively high concentrations in apex feeders far from known sources. Given the length and complexity of the OC pathways into top predators of aquatic systems in the Arctic, exposure to these chemicals is particularly sensitive to global change. Furthermore, the subtle effects of OCs (immune function, reproduction and endocrine disruption) can be exacerbated by, and work together with, nutritional stress brought about by ecosystem and ice climate change. The significance of contaminants as added stressors to predators already suffering from habitat and ecosystem change, while widely suspected, is not confidently understood. Key species and study areas in this context appear to be marine mammal populations (bears and seals) from the Kara Sea, Franz Josef Land, Svalbard and Hudson Bay.

8.1.4. Hydrocarbons and PAHs

Change accompanying the AO offers interesting divergence in ice and coastal current pathways which have relevance to spilled oil. However, the greatest concern from hydrocarbons will probably come from the growing attractiveness of Arctic marginal seas for oil production and transport with reduced ice cover. Even though ice may clear from substantial portions of the marginal seas annually, the perspective must be maintained that these are seasonally ice-covered regions. It is recommended that regulation appropriate for ice-covered waters be secured for offshore production and in designing ships or pipelines for oil transport. The rapid change in ice conditions experienced during the 1990s may be the initial stages of a trend toward a more temperate Arctic Ocean but it may also be an extreme manifestation of a natural cycle which will be followed by a return to heavier ice conditions.

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Abbreviations

$\delta^{18}O$	$^{18}O/^{16}O$ stable isotope ratio (standardized against Standard Mean Ocean Water)
$\delta^{15}N$	$^{15}N/^{14}N$ stable isotope ratio (standardized against air)
ΣHCH	Sum of (concentrations of) α-, β- and γ-HCH isomers
ΣPCB	Sum of a number of individual PCB congeners
AMAP	Arctic Monitoring and Assessment Programme
ANWAP	Arctic Nuclear Waste Assessment Program
AO	Arctic Oscillation
AO^-	Conditions of low AO index values
AO^+	Conditions of high AO index values
As	Arsenic
Cd	Cadmium
CFCs	Chlorofluorocarbons
CHB	Toxaphene
CH_4	Methane
CO_2	Carbon dioxide
Cs	Cesium
Cu	Copper
DDT	Dichlorodiphenyltrichloroethane (an organochlorine pesticide)
GHGs	Greenhouse gasses
HCB	Hexachlorobenzene
HCH	Hexachlorohexane
Hg	Mercury
Hg^0	Gaseous mercury
HLC	Henry's Law constant
IABP	International Arctic Buoy Programme
IPCC	Intergovernmental Panel on Climate Change
K_{AW}	Air-water partition coefficient
K_{OA}	Octanol-air partition coefficient
K_{OW}	Octanol-water partition coefficient
K_{PG}	Particle-gas partition coefficient
K_W	Dissociation constant for water
MeHg	Methylmercury
MDE	Mercury Depletion Event
N_2O	Nitrous oxide
NAO	North Atlantic Oscillation
NwAC	Norwegian Atlantic Current
OC	Organochlorine compound
OCPs	Organochlorine pesticides
OH	Hydroxyl radical
PAHs	Polyaromatic hydrocarbons
Pb	Lead
PBDEs	Polybrominated diphenyl ethers
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo-*p*-dioxins
PCDFs	Polychlorinated dibenzofurans
PCNs	Polychlorinated naphthalenes
PDO	Pacific Decadal Oscillation
P-E	Precipitation minus evaporation
Po	Polonium
POM	Particulate organic matter
POP	Persistent organic pollutant
Pu	Plutonium
Ra	Radium
RGM	Reactive gaseous mercury
Rn	Radon
S	Salinity
SAT	Surface air temperature
SCC	Siberian Coastal Current
SLP	Sea-level pressure
Sr	Strontium
SSM/I	Special Sensor Microwave/Imager
SSMR	Scanning Multichannel Microwave Radiometer
Sv	Sverdrup, 1 Sv = 10^6 m^3/s (approximately 32 000 km^3/yr)
T	Temperature
Tc	Technetium
TPD	Transpolar Drift
UV	Ultraviolet radiation
Zn	Zinc