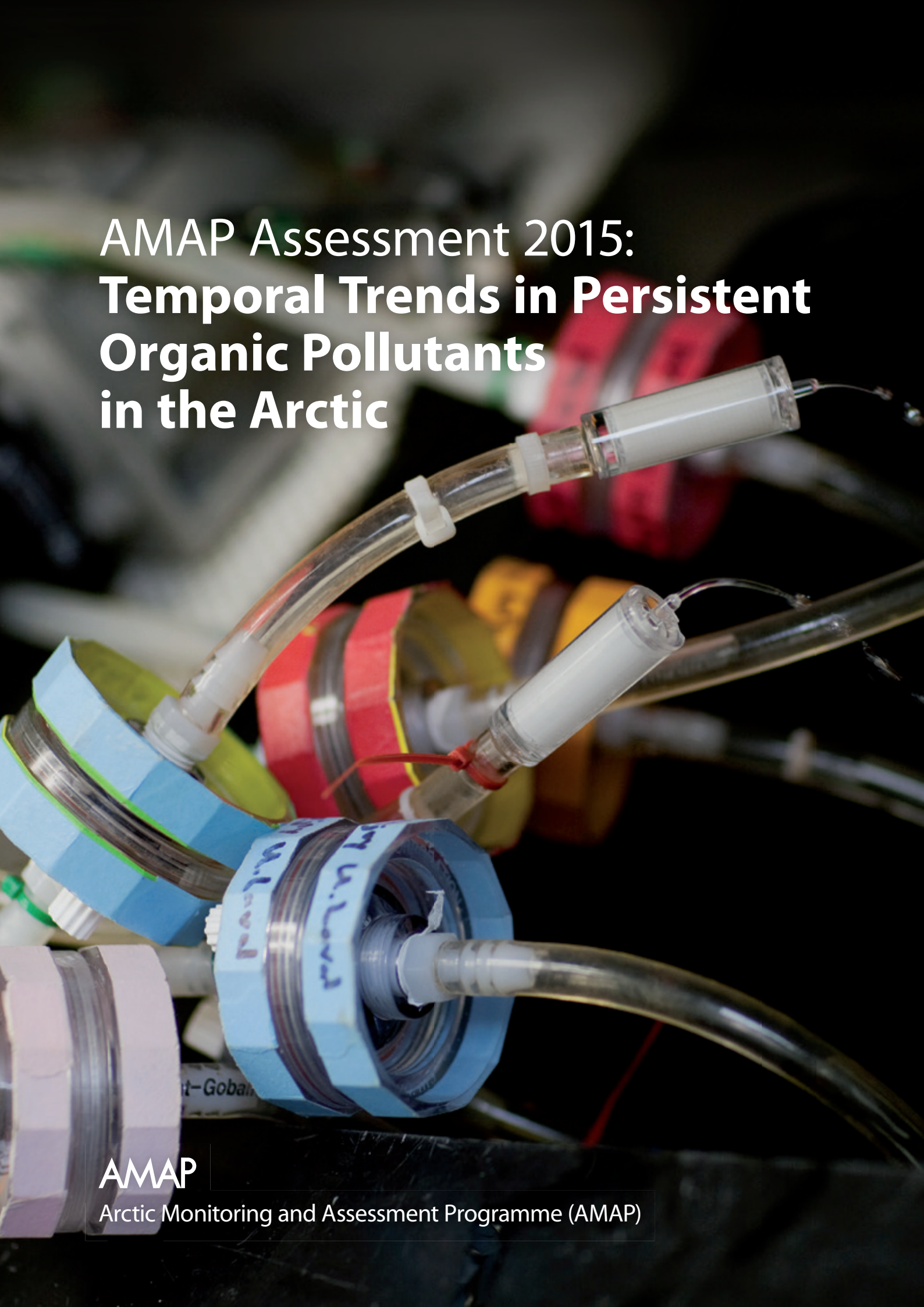

# AMAP Assessment 2015: **Temporal Trends in Persistent Organic Pollutants in the Arctic**

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Oslo, 2016

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# Preface

This assessment report presents the results of the 2015 AMAP Assessment of Temporal Trends in Persistent Organic Pollutants (POPs) in the Arctic. This assessment updates the AMAP POPs assessments delivered in 1998, 2002 and 2009, specifically with respect to information on temporal trends.

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
- recommend actions required to reduce risks to Arctic ecosystems.

This report provides the accessible scientific basis and validation for the statements and recommendations made in the Summary for Policy-makers: Arctic Pollution Issues 2015 report[i] that was delivered to Arctic Council Ministers at their meeting in Iqaluit, Canada in April 2015. It is also the basis for a related AMAP State of the Arctic Environment report Arctic Pollution Issues 2015: Overview[ii]. It includes extensive background data and references to the scientific literature, and details the sources for graphics reproduced in the overview report. Whereas the Summary for Policy-makers report contains recommendations that focus mainly on policy-relevant actions concerned with addressing contaminant impacts on Arctic human populations, 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.

This assessment of temporal trends in Arctic POPs was conducted between 2012 and 2014 by an international group of experts. AMAP POPs expert group members and lead authors are appointed following an open nomination process coordinated by AMAP. A similar process was used to select international experts who independently reviewed this report.

Information contained in this report is fully referenced and based first and foremost on results of research and monitoring undertaken since 2009. It incorporates some new (unpublished) information from monitoring and research conducted according to well established and documented national and international standards and quality assurance/quality control protocols. Care has been taken to ensure that no critical probability statements are based on non-peer-reviewed materials.

Access to reliable and up-to-date information is essential for the development of science-based decision-making regarding ongoing changes in the Arctic and their global implications. Related assessment summary reports[i,ii] have therefore been developed specifically for policy-makers, summarizing the main findings of the assessment. The assessment lead authors have confirmed that both this report and its derivative products accurately and fully reflect their scientific assessment. All AMAP assessment reports are freely available from the AMAP Secretariat and on the AMAP website: www.amap.no, and their use for educational purposes is encouraged.

AMAP would like to express its appreciation to all experts who have contributed their time, efforts and data, in particular the lead authors who coordinated the production of this report. Thanks are also due to the reviewers who contributed to the assessment peer-review process and provided valuable comments that helped to ensure the quality of the report. A list of contributors is included in the acknowledgements at the start of this report and lead authors are identified at the start of each chapter. The acknowledgements list is not comprehensive. Specifically, it does not include the many national institutes, laboratories and organizations, and their staff, which have been involved in various countries in POPs-related monitoring and research. Apologies, and no lesser thanks are given to any individuals unintentionally omitted from the list.

The support from the Arctic countries and non-Arctic countries implementing research and monitoring in the Arctic is vital to the success of AMAP. The AMAP work is essentially based on ongoing activities within these countries, and the countries that provide the necessary support for most of the experts involved in the preparation of the AMAP assessments. In particular, AMAP would like to acknowledge Canada, Kingdom of Denmark and Sweden for taking the lead country role in this assessment and thank Canada and the Nordic Council of Ministers for their financial support to the POPs temporal trends assessment work.

The AMAP Working Group is pleased to present its assessment to the Arctic Council and the international science community.

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Oslo, October 2016

[i] AMAP, 2015. Summary for Policy-makers: Arctic Pollution Issues 2015. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. 12 pp.

[ii] AMAP, 2015. Pollution Issues 2015: Overview report. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

# Contents

# 1. Introduction

The Arctic Monitoring and Assessment Programme (AMAP) has produced a series of assessments of persistent organic pollutants (POPs) in the Arctic. The first of these (AMAP, 1998) documented the presence of POPs in abiotic media (air, water, ice, snow, sediments) and freshwater, terrestrial and marine biota across the circumArctic area. With a few exceptions, this contamination was a result of long-range transport of POPs from source areas in the south. Although levels of contamination were generally low in abiotic media, levels in some biota (high trophic level marine species) and some human populations were very high, raising concerns about possible ecosystem and human health effects. The high levels observed in some biota were associated with the lipophilic properties of many POPs, and their biomagnification in Arctic food webs. The 1998 AMAP assessment established the baseline for follow-up assessments of POPs in the Arctic performed in 2004 (AMAP, 2004) and 2009 (AMAP, 2010); these studies began to follow temporal changes in levels of POPs in the Arctic, and also documented the occurrence of additional groups of POP compounds in the Arctic. At about the same intervals, AMAP also prepared assessments of human health in the Arctic that document levels and effects of POPs in Arctic human populations (AMAP, 1998, 2003, 2009, 2015).

AMAP's current update assessment of POPs in the Arctic has four components:

- An update assessment of temporal trends of POPs in the Arctic (the subject of this document)
- An assessment of chemicals of emerging Arctic concern (to be completed in 2016)
- An update assessment of biological effects of POPs in Arctic biota (to be completed in 2016)
- An assessment of the influence of climate change on Arctic POPs contamination (to be initiated in 2016).

Information reported in the AMAP POPs and human health assessments has been used in processes to establish and further develop international action to reduce emissions and releases of POPs to the environment, including the UNEP Stockholm Convention on POPs (http://chm.pops.int) and the POPs Protocol to the Convention on Long-range Transboundary Air Pollution (www.unece.org/env/lrtap/pops_h1.html). AMAP has an ongoing mandate from the Arctic Council to support the implementation of these international agreements, including their related effectiveness and sufficiency evaluations, and to work to evaluate new compounds for inclusion under the agreements. Information on temporal trends is especially relevant to the effectiveness evaluations, such as those required under Article 16 of the Stockholm Convention. Technical products included in the current AMAP assessment of temporal trends in POPs were therefore provided to the Stockholm Convention groups responsible for the (2015) Article 16 evaluation (AMAP, 2014).

## 1.1 Background

Monitoring undertaken to contribute to the AMAP Trends and Effects Monitoring Programme includes studies that aim to establish long-term trends which can be used (i) to assess the effectiveness of national and international control strategies, (ii) to assess long-range transport of POPs to the Arctic, and (iii) to identify new priority chemicals which may be of concern in the region.

Long-term (multi-annual) changes in levels of POPs in Arctic environmental media and biota are determined by a number of factors, including:

- changes in primary emissions/releases of these chemicals (or their precursors), related to production and uses in industrial applications, agriculture, consumer products, and other uses; and from waste streams associated with such uses
- changes in re-emissions of chemicals that have accumulated in environmental media such as surface soils and surface waters (including possible influences of climate change)
- changes in environmental transport pathways, and processes that affect these, both in abiotic systems (e.g. winds and ocean currents) and in biological systems (ecosystem and food web structure, etc.).

The influence of these factors is also likely to depend on whether contamination is associated with local sources or (long-range transport) from remote sources.

The most rapid route of transport for POPs to the Arctic is via the atmosphere. Ambient air has a short response time to changes in atmospheric emissions and is a relatively well-mixed environmental compartment. It is also a global transport medium and an entry point into food webs via POP deposition and air-surface exchange. Therefore, air is one of the two core media under the Global Monitoring Plan (GMP) for evaluating the effectiveness of the Stockholm Convention on POPs; the other GMP core media are human bio-media (blood and breastmilk). The Stockholm Convention evaluations and reviews focus on results for the GMP core monitoring media but also accommodate supplementary information from studies of other media.

Biota are widely used in environmental monitoring programs around the world, including those specifically designed to monitor temporal trends of POPs. Monitoring of time-trends using biota, in particular freshwater and marine biota, are key components of the Arctic monitoring conducted under the auspices of the AMAP Trends and Effects Monitoring Programme.

The AMAP Trends and Effects Monitoring Programme is a harmonized program for monitoring the trends and effects of contaminants and climate change across the circumArctic region. It includes sub-programs concerned with monitoring atmospheric, marine, terrestrial and freshwater media, and human tissues (in connection with monitoring effects of contaminants on human health). The AMAP monitoring program is based largely on ongoing national monitoring and research activities that comprise AMAP national implementation plans of the eight Arctic countries. AMAP coordinates these activities and works to ensure harmonization, and to promote quality assurance activities, and compiles results

for use in circumArctic assessment activities. The AMAP program is also coordinated with other international programs such as the UNECE European Monitoring and Evaluation Programme (EMEP) and OSPAR's Joint Assessment and Monitoring Programme (JAMP). The AMAP program therefore represents a significant component in the implementation of the GMP in the Arctic.

AMAP's previous POPs temporal trend assessment (AMAP, 2010) included a treatment of time-series in air (Hung et al., 2010) and biota (Rigét et al., 2010) up to 2006/7. Many of these time-series are extended to 2011/12 in the present assessment and these additional years provide a more (statistically) solid foundation for evaluating temporal trends of POPs in Arctic air and biota. See Annex 1 for a discussion of the power of the AMAP biota trends monitoring program for trend detection.

Time-series can be analyzed using different statistical methods. Some approaches to trend detection and analyses incorporate factors that can influence trends differently and to varying degrees. Examples include trend studies that use stable isotope data ($\delta^{15}N$ and $\delta^{13}C$) to indicate possible dietary shifts (Dietz et al., 2013) or parameters such as the Arctic Oscillation Index or sea-ice cover to evaluate the influence of climate change and variation (Bustnes et al., 2010; Gaden et al., 2012; Riget et al., 2013a). This can lead to different interpretations of trend results.

A large number of time-series datasets were considered in this assessment – over 2000 for contaminants in biota and more than 150 for contaminants in air. With so many datasets it is not practical to examine each in the level of detail that would be required to rigorously address all potential confounding factors and supplementary information. The main objective of this assessment has therefore been to apply robust statistical methodologies consistently to all available time-series – to gain a general overview of trends for certain compound groups across the wide range of datasets available, and across the entire geographical region. On this basis, a form of meta-analysis is performed, looking at the consistency (or otherwise) of the apparent trends. In addition, information from more detailed data interpretations produced by those responsible for individual (national) trend studies has also been compiled, and where possible used to qualify the results of the robust trend evaluations.

It should be noted that trend statistics produced in the meta-analysis conducted in this assessment and those produced in other work may differ, depending for example on the statistical methods applied, although these differences are generally small. What is gained from a more careful consideration of individual time-series is, however, better insight into possible reasons for trends, which may be associated with local conditions and/or circumstances pertaining to the specific monitoring studies.

This assessment, therefore, attempts to utilize information from both sources, although the emphasis when describing regional patterns or trends for specific POPs tends to be based mainly on the meta-analysis results. Readers interested in trends at specific locations are encouraged to refer to material published by the data originators; these references can be found in the national commentaries presented in Annexes 2 to 5.

## 1.2 Readers' guide

To facilitate access to information on specific contaminants or groups of contaminants, the following assessment is organized according to contaminants/contaminant groups, each with sub-sections describing trend results for air, biota and then any other media for which information is available. This is followed by a discussion of the results, including where appropriate information on geographical patterns in the observed trends, and possible explanations for the trends (including reference to published results for individual trend studies). See Annexes 2 to 5 for a compilation of contributions provided by experts responsible for specific national trends studies; these commentaries have been harmonized to some degree to include:

- A short commentary on the conclusions highlighted, including for example proposals/ recommendations/ notes on plans for continuing the studies/future work (e.g. new sites or new contaminants being considered for inclusion in the trend monitoring, and why).
- A brief introduction to the datasets available (locations, species/media monitored, time-periods covered), the basis for the monitoring, for example a description (and acknowledgment) of relevant (national/other) monitoring programs under which the trend monitoring is performed, if relevant placing the Arctic work in any wider national context.
- A short description of the methods employed. For example, similar to those used for the meta-analysis (based on PIA).
- A brief summary of trend program results. That is, a selective commentary on the independent trend evaluation work undertaken, highlighting pertinent results, proposing explanations for the trends, or illustrating important factors or processes involved. This could also include reference to other trend studies that may not have been considered in the AMAP meta-data context, such as studies based on sediment or ice cores.

Annex 1 describes the statistical methodologies applied, and includes a discussion of the power of the monitoring programs and the statistical approach applied to the analysis of biota trend datasets to detect trends of interest to policy-makers. Annexes 2 to 5 present more detailed results of trend analyses conducted on individual time-series for some national datasets from Canada, Iceland, United States and Greenland. Annex 6 presents a summary of the contaminant concentrations observed in the various time series considered in this (temporal trend) meta-analyses.

Reference is sometimes made to results on POPs trends in humans. For further information on these studies readers are referred to the latest AMAP Assessment of Human Health in the Arctic (AMAP, 2015).

Chapters 2–4 describe respectively the time series datasets, statistical methodology and temporal trend results obtained for the different contaminants considered. Conclusions and findings of this assessment are summarized in Chapter 5.

# 2. Time-series datasets considered

## 2.1 AMAP Arctic air monitoring

Long time-series of POPs in air are available for trend development from four Arctic air monitoring stations where POPs monitoring has been conducted using high volume air samplers for periods of up to 20 years (Fig. 2.1): Alert (82°30'N, 62°20'W, 200 meters above sea level, masl) on Ellesmere Island as part of Canada's Northern Contaminants Program (NCP); Stórhöfði (63°24'N, 20°17'W, 118 masl) on Iceland; Zeppelin (78°54'N, 11°53'E, 474 masl) at Ny-Ålesund on Svalbard; and Pallas (68°00'N, 24°15'E, 340 masl) in Arctic Finland. The Villum Research Station (formerly Station Nord) on Greenland also has multi-year time-series for POPs but for a shorter period (since 2009) (Bossi et al., 2013). POPs have also been monitored at the Andøya observatory in northern Norway since December 2009; due to the short time-series the data are not included in the present assessment but will be used in future AMAP trend assessments. POPs air concentration data for the Andøya observatory are reported by Aas and Nizzetto (2015) and Nizzetto et al. (2015).

Other POPs air monitoring sites (including Dunai Island, Valkarkai and Tiksi in Arctic Russia, and Tagish/Little Fox Lake in the Yukon, Canada) have operated in past years, but lack the long-term continuity of monitoring necessary for developing long time-series datasets. Work is currently underway to (re-) establish POPs monitoring at the Tiksi and Amderma stations in Russia. Short-term air monitoring results from Little Fox Lake (August 2007–2009) and Valkarkai (2008–2009) are presented here for comparative purposes.

AMAP air monitoring data are compiled in the AMAP atmospheric thematic data center (TDC) database at the Norwegian Institute for Air Research (NILU) and datasets were extracted from the EBAS database (http://ebas.nilu.no) for statistical analysis.

## 2.2 POPs monitoring in biota

For biota, time-series are available from seven countries for a total of 75 location-species-tissue combinations. These include locations in Alaskan marine areas; Arctic Canada; East Greenland (Ittoqqortoormiit area) and West Greenland (Disko Island area and Isortoq), subsequently referred to as eastern and western Greenland; marine areas around Iceland, the Faroe Islands and northern Norway; and lakes in Sweden. No relevant biota time-series datasets are currently available for the Arctic areas of Russia or Finland. Figure 2.1 shows locations where long time-series monitoring of POPs in Arctic biota are conducted.


Figure 2.1 Locations where long time-series monitoring of POPs in Arctic air and biota are conducted. For biota, the species monitored are also indicated.

For the purposes of this assessment, AMAP biota monitoring datasets were selected that included at least six years of data. For 'legacy' POPs (see Box 2.1), priority was also given to time-series that included data from both before and after 2000. The average length of the time-series considered is around 12 years; the longest available time-series has 42 years of data and some time-series include samples collected as early as 1975. Previous AMAP temporal trend assessments (e.g. Rigét et al., 2010) have included terrestrial and marine species. The current assessment includes only marine and freshwater species as no new data were available for the terrestrial components.

Icelandic and Norwegian datasets include data reported to AMAP/OSPAR and archived at the AMAP marine TDC at the International Council for the Exploration of the Sea (ICES) in Denmark. Other AMAP data were collected from lead scientists responsible for relevant temporal trend monitoring studies in Canada, Denmark/ Greenland/ Faroe Islands, Sweden and the United States.

## 2.3 QA/QC considerations

AMAP laboratories responsible for the POPs analyses included in this report participate in a number of laboratory quality assurance / quality control (QA/QC) programs, including the AMAP/NCP Inter-laboratory studies (e.g. Tkatcheva et al., 2013); the AMAP/EMEP/NCP air monitoring inter-laboratory study (Schlabach et al., 2011), the QUASIMEME laboratory performance testing scheme (www.quasimeme.org), and equivalent QA/QC programs run by NOAA/NIST.

**Box 2.1 'Legacy POPs' and 'New POPs'**

The term 'Legacy POPs' has been used to refer to chemicals whose presence in the environment is largely a legacy of past use. This definition covers most of the initial POPs controlled by the Stockholm Convention, even though some, such as polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) have sources that cannot be entirely eliminated.

As of May 2015, 14 POPs had been added to the Stockholm Convention since 2004. They include additional pesticides, and industrial chemicals and by-products such as some polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and perfluorooctane sulfonic acid (PFOS). PBDEs and HBCDD are brominated flame retardants. Within the context of the Convention, these have been referred to as 'New POPs', even though a number of them have been in use in society for decades. Over time, the 'New POPs' should also become 'Legacy POPs'.

# 3. Statistical methods applied

## 3.1 Air

In total, 103 contaminant time-series from the four long-term stations (Alert, Stórhöfði, Zeppelin, Pallas) and 63 contaminant time-series from Little Fox Lake and Valkarkai were statistically analyzed, and trend results compared together with published results from the Villum Research Station.

Air time-series were analyzed using the Digital Filtration (DF) technique (see Hung et al., 2005). DF is a statistical fitting technique that fits seasonal cycles and interannual trends from time-series. This technique has previously been used to derive long-term trends in air concentration for POPs monitored under the AMAP network (Hung et al., 2005, 2010) and was consistently found to be more accurate in estimating long-term trends than two other commonly used methods (Kong et al., 2014).

Briefly described, the method involves determining an approximate long-term trend and an average seasonal cycle by fitting a smoothing Reinsch-type cubic spline and Fourier components to the natural log-transformed concentration data (ln C), respectively, in an iterative manner until the fitted spline function becomes almost unchangeable. Outlier data points that were more than three standard errors away from the fitted curve were successively rejected after each iterative fit. The percentage of data points rejected during this process is dataset specific. Long- and short-term variations of the trend and the seasonal cycle were then extracted using two Butterworth digital filters with two cut-off periods: a short-term cut-off period (set to four months) and a long-term cut-off period (set to 48 months). Variabilities longer than four months and shorter than 48 months were extracted to obtain the overall seasonal cycle, and variabilities longer than 48 months were extracted to obtain the final long-term trend. The cut-off periods, which produced the 'best fit' to the specific dataset, were chosen by trial-and-error based on visual inspection of the fitted seasonal cycle.

A decline in air concentrations over time is often quantified by an apparent first order half-life, $t_{1/2}$, which is estimated by dividing the natural log of two, ln 2, with the negative value of the linear regression slope of the trend line between the natural log of air concentrations, C (pg/m$^3$), and time (year). Note that many POPs do not necessarily decline linearly or consistently in the first order manner throughout the monitoring periods of the four long-term air monitoring stations. The half-lives ($t_{1/2}$) presented here are thus used only to compare the relative rates of decline among the four stations. Readers are advised to use the absolute values of these half-lives with caution (half-lives are summarized in Table A6.1). In the text, $t_{1/2}$ are also given as equivalent annual percentage change (% per year) to facilitate comparisons of rates of change.

## 3.2 Biota

Time-series datasets for POPs in biota samples were analyzed using a robust regression approach (based on Nicholson et al., 1998) testing for both linear and non-linear trend components, using the PIA computer application developed by Anders Bignert and co-workers (Bignert, 2013; see also Annex 1).

Statistical analyses were applied to individual time-series for 65 compounds/compound groups (see Tables 3.1 and 3.2). In all, some 2481 statistical analyses were performed. Of these, a significant number (310) were excluded because the time-series concerned were considered unsuitable for statistical analyses (for example, they included a large proportion of 'less than' values). A selection was also made where alternative runs were performed using different available covariates. Results for some 1809 datasets were eventually summarized and evaluated, comprising time-series that start before 2000 (1074 datasets) as well as time-series that begin in or after 2000, or have years prior to 2000 excluded (735 time-series), see Figure 3.1. The largest number of trend results for a single contaminant was 61 (for CB153 time-series covering the entire period).

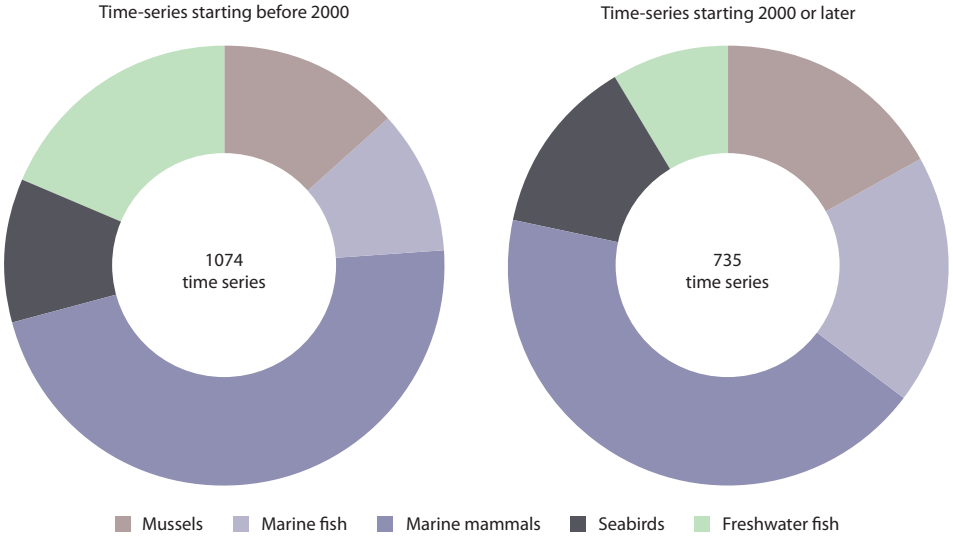



Figure 3.1 Overview of available biota time-series shown by animal group.

Table 3.1 Number of time-series starting before 2000 by compound/compound group and country.

| Compound / Compound Group | Canada | Faroe Islands | Greenland | Iceland | Norway | Sweden | USA | Total |
|---|---|---|---|---|---|---|---|---|
| Dieldrin | 19 | | 3 | | | | | 22 |
| *cis*-Chlordane | 4 | 3 | 3 | 5 | | | 4 | 19 |
| *trans*-Chlordane | 4 | 3 | | 3 | | | 4 | 14 |
| *cis*-Nonachlor | 4 | 3 | 3 | | | | 4 | 14 |
| *trans*-Nonachlor | 8 | 3 | 8 | 11 | | | 5 | 35 |
| Oxychlordane | 4 | 3 | 8 | | 1 | | 4 | 20 |
| ΣCHL | 18 | 3 | 5 | | | | 5 | 31 |
| *o,p'*-DDD | | 2 | | | | | 4 | 6 |
| *o,p'*-DDE | | 2 | | | | | 4 | 6 |
| *o.p'*-DDT | | 2 | | | | | 3 | 5 |
| *p,p'*-DDD | 4 | 3 | | 5 | 3 | | 4 | 19 |
| *p,p'*-DDE | 20 | 3 | 8 | 12 | 11 | 2 | 4 | 60 |
| *p,p'*-DDT | 4 | 3 | | | 1 | | 4 | 12 |
| ΣDDT | 21 | 3 | 8 | 10 | 10 | | 5 | 57 |
| HEPOX | 4 | | 3 | | | | 5 | 12 |
| HCB | 19 | 3 | 8 | 10 | 7 | 2 | 5 | 54 |
| α-HCH | 20 | | 8 | 9 | 2 | 2 | 5 | 46 |
| β-HCH | 16 | 3 | 7 | 2 | | | 5 | 33 |
| γ-HCH | 16 | | 4 | 2 | 3 | 2 | 4 | 31 |
| Mirex | 8 | 3 | | | | | 5 | 16 |
| Pentachlorbenzene | 9 | | | | | | 4 | 13 |
| CB28 | 4 | 3 | 1 | | 10 | | | 18 |
| CB31 | 4 | | 1 | | | | | 5 |
| CB28/31 | | | | | | | 4 | 4 |
| CB52 | 4 | 3 | 7 | | 10 | | 4 | 28 |
| CB99 | | 3 | | | 1 | | | 4 |
| CB101 | 4 | 3 | 8 | | 10 | | 4 | 29 |
| CB105 | 4 | 3 | 2 | | 10 | | 4 | 23 |
| CB118 | 4 | 3 | 2 | | 11 | 2 | 4 | 26 |
| CB128 | | 3 | | | | | | 3 |
| CB138 | 4 | 3 | 8 | | 11 | | 4 | 30 |
| CB153 | 20 | 3 | 8 | 12 | 11 | 2 | 5 | 61 |
| CB156 | 2 | 3 | 1 | | 10 | | 4 | 20 |
| CB158 | | | | | | | 1 | 1 |
| CB170 | | 3 | | | 1 | | | 4 |
| CB180 | 4 | 3 | 8 | | 11 | | 4 | 30 |
| CB183 | | 3 | | | | | | 3 |
| CB187 | | 3 | | | | | | 3 |
| CB209 | | | | | 10 | | 1 | 11 |
| $\Sigma PCB_{10}$ | 20 | 2 | 8 | 10 | 11 | | 5 | 56 |
| Toxaphene Parlar 26 | | 3 | 4 | 9 | | | 1 | 17 |
| Toxaphene Parlar 32 | | | | | | | 1 | 1 |
| Toxaphene Parlar 50 | | 3 | 4 | 9 | | | 1 | 17 |
| Toxaphene Parlar 62 | | 1 | | | | | 1 | 2 |
| BDE47 | 14 | 1 | 6 | | | 1 | 4 | 26 |
| BDE49 | | | | | | | 1 | 1 |
| BDE99 | 9 | 1 | 5 | | | | 4 | 19 |
| BDE100 | | | | | | | 2 | 2 |
| BDE153 | | | | | | 1 | 1 | 2 |
| BDE154 | | | | | | | 1 | 1 |
| BDE155 | | | | | | | 1 | 1 |
| HBCDD | 3 | | 5 | | | | 1 | 9 |
| PFCs | | | | | | | 3 | 3 |
| PFDA | 3 | 1 | 3 | | | | 3 | 10 |
| PFDoA | 3 | | 3 | | | | 3 | 9 |
| PFHxS | | | | | | | 4 | 4 |
| PFNA | 3 | 1 | 3 | | | | 4 | 11 |
| PFOA | | | 3 | | | | | 3 |
| PFOS | 6 | 1 | 6 | | | 2 | 4 | 19 |
| PFOSA | 1 | | 3 | | | | 4 | 8 |
| PFTA | | | | | | | 3 | 3 |
| PFTrA | | | 3 | | | | 4 | 7 |
| PFUnA | 3 | 1 | 3 | | | | 4 | 11 |
| OCS | 8 | | 3 | | | | | 11 |
| **Total** | **329** | **102** | **184** | **109** | **155** | **16** | **186** | **1081** |

Table 3.2 Number of time-series starting in 2000 or later by compound/compound group and country.

| Compound / Compound Group | Canada | Faroe Islands | Greenland | Iceland | Norway | Sweden | USA | Total |
|---|---|---|---|---|---|---|---|---|
| Dieldrin | 10 | | 3 | | | | | 13 |
| *cis*-Chlordane | 2 | 3 | 3 | 10 | | | | 18 |
| *trans*-Chlordane | 2 | 3 | | 2 | | | | 7 |
| *cis*-Nonachlor | 2 | 3 | 3 | | | | | 8 |
| *trans*-Nonachlor | 4 | 4 | 7 | 11 | | | | 26 |
| Oxychlordane | 2 | 3 | 7 | 2 | | | | 14 |
| ΣCHL | 10 | 5 | 4 | | | | 1 | 20 |
| *o,p*'-DDD | | 1 | | | | | | 1 |
| *o,p*'-DDT | | 1 | | | | | | 1 |
| *p,p*'-DDD | 2 | | | 8 | 2 | | | 12 |
| *p,p*'-DDE | 12 | 5 | 7 | 13 | 10 | 2 | 1 | 50 |
| *p,p*'-DDT | 2 | 2 | | | 1 | | | 5 |
| ΣDDT | 11 | 3 | 7 | 12 | 8 | | 1 | 42 |
| HEPOX | 3 | | 2 | | | | 1 | 6 |
| HCB | 10 | 5 | 7 | 11 | 7 | 2 | 1 | 43 |
| α-HCH | 12 | | 7 | 11 | 3 | | 1 | 34 |
| β-HCH | 11 | 3 | 7 | 2 | | | 1 | 24 |
| γ-HCH | 7 | | 4 | 6 | 4 | | 1 | 22 |
| Mirex | 5 | 4 | | | | | 1 | 10 |
| Pentachlorbenzene | 7 | | | | 1 | | | 8 |
| CB28 | 2 | 3 | | | 2 | | | 7 |
| CB31 | 2 | | 1 | | | | | 3 |
| CB52 | 2 | 3 | 7 | | 3 | | | 15 |
| CB99 | | 3 | | | | | | 3 |
| CB101 | 2 | 5 | 7 | | 5 | | | 19 |
| CB105 | 2 | 4 | 2 | | 3 | | | 11 |
| CB118 | 2 | 5 | 2 | | 6 | 1 | | 16 |
| CB128 | | 4 | | | | | | 4 |
| CB138 | 2 | 3 | 7 | | 8 | | | 20 |
| CB153 | 12 | 5 | 7 | 13 | 10 | 2 | 1 | 50 |
| CB156 | 1 | 3 | | | 1 | | | 5 |
| CB163 | | 3 | | | | | | 3 |
| CB170 | | 3 | | | | | | 3 |
| CB180 | 2 | 5 | 7 | | 3 | | | 17 |
| CB183 | | 3 | | | | | | 3 |
| CB187 | | 3 | | | | | | 3 |
| $\Sigma PCB_{10}$ | 12 | 5 | 7 | 11 | 11 | | 1 | 47 |
| Toxaphene Parlar 26 | | 4 | 4 | 12 | | | | 20 |
| Toxaphene Parlar 50 | | 5 | 4 | 13 | | | | 22 |
| Toxaphene Parlar 62 | | 1 | | | | | | 1 |
| BDE47 | 13 | | 6 | | | 1 | 2 | 22 |
| BDE99 | 10 | | 5 | | | | 2 | 17 |
| BDE153 | | | | | | 1 | | 1 |
| HBCDD | | | 1 | | | | | 1 |
| PFDA | 2 | | 3 | | | | | 5 |
| PFDoA | 2 | | 3 | | | | | 5 |
| PFNA | 2 | | 3 | | | | | 5 |
| PFOA | | | 3 | | | | | 3 |
| PFOS | 3 | | 3 | | | 2 | | 8 |
| PFOSA | | | 3 | | | | | 3 |
| PFTrA | | | 3 | | | | | 3 |
| PFUnA | 2 | | 3 | | | | | 5 |
| OCS | 5 | | 3 | | 1 | | 2 | 11 |
| **Total** | **194** | **115** | **162** | **137** | **89** | **11** | **17** | **725** |

Table 3.3 Classification of biota trend results.

| Class | Comment |
|---|---|
| Increasing trend | A statistically significant increasing log-linear trend |
| Increasing trend with non-linear trend component | Both the increasing log-linear and non-linear trend components are statistically significant |
| Decreasing trend | A statistically significant decreasing log-linear trend |
| Decreasing trend with non-linear trend component | Both the decreasing log-linear and non-linear trend components are statistically significant |
| Non-linear trend component | A statistically significant non-linear (fluctuating) trend with no clear increasing or decreasing tendency |
| No trend | The time-series did not exhibit a statistically significant trend |
| Not evaluated | The time-series was unsuitable for trend analysis (for example, it contained too many 'less-than-detection-limit' values) |

The PIA statistical application (Bignert, 2013) provides a robust method for investigating trends in time-series data represented by annual index values (Nicholson et al., 1998). The method employed tests for the presence of (log-)linear trends, and non-linear trend components (for example an increase followed by a subsequent decrease) in the time-series (at a significance level of 5%). Median concentrations were used as the annual index values to minimize the influence of outliers and less-than-detection-limit values. The method also evaluates the number of years required to detect an annual change of 5% with a power of 80% for the particular time-series.

Datasets were handled in a manner similar to previous evaluations (Rigét et al., 2010), taking account of data originators' recommendations for sub-setting animal groups and including covariates. Many time-series were run in different configurations (for example, with and without covariate adjustment) to investigate the influence of factors such as age, sex, and lipid content on the observed trends.

Time-series with a large number of values reported as less-than-detection-limit were examined to consider the pattern of these (for example whether they were concentrated at the end of a time-series exhibiting decreasing trends). Time-series where more than 50% of values were reported as less-than-detection-limit in three or more years and thereby have annual median values of less-than-detection-limit for these years, were generally considered inappropriate for trend analyses, unless these years were concentrated at the start or end of the time-series. 'Less-than' qualified values were replaced by half the reported detection limit.

Biota trend results were classified as shown in Table 3.3.

# 4. Temporal trend analyses results

Persistent organic pollutants are listed under the Stockholm Convention for elimination (Annex A), restriction (Annex B), and reduction of unintentional production (Annex C). Initially, twelve POPs (the so-called 'dirty dozen') were recognized by the Stockholm Convention as causing adverse effects on humans and the ecosystem. Nine further POPs were added to the Stockholm Convention in 2009, one in 2011, one in 2013 and two in 2015. A further four chemicals are currently (February 2016) proposed for listing under the Stockholm Convention. Table 4.1 presents an overview of the chemicals currently listed under the Stockholm Convention and the POPs Protocol to the LRTAP Convention.

The following sections discuss, on a substance-by-substance basis, trend results first in air and then in biota, and then on a more general basis, including, for some substances, a discussion of the geographical patterns in the observed trends. Table 4.1 includes a reference to the sections that pertain to the various listed chemicals. Summary tables presenting the numerical results of the trend analyses in air and biota, respectively, are included in Chapter 5.

Table 4.1 Status of POPs listing under international regulatory Conventions. The Stockholm Convention was agreed in 2001 and entered into force in 2004. The POPs Protocol to the LRTAP Convention was agreed in 1998 and entered into force in 2003; the 2009 amendments to Annexes I-IV, and VI are not yet in force.

| POP | Uses | Date of Stockholm Convention Listing | LRTAP POPs Protocol | Results section |
|---|---|---|---|---|
| Aldrin | Pesticide | Annex A (2004) | Annex I (2003) | 4.1 |
| Chlordane | Pesticide | Annex A (2004) | Annex I (2003) | 4.2 |
| Dichlorodiphenyltrichloroethane (DDT) | Pesticide | Annex B with exemptions for disease vector control (2004) | Annex I and II (2003) (update proposed 2009) | 4.3 |
| Dieldrin | Pesticide | Annex A (2004) | Annex I (2003) | 4.1 |
| Endrin | Pesticide | Annex A (2004) | Annex I (2003) | 4.1 |
| Heptachlor | Pesticide | Annex A (2004) | Annex I (2003) (update proposed 2009) | 4.4 |
| Hexachlorobenzene (HCB) | Pesticide / Industrial / By-product | Annex A and Annex C (2004) | Annex I and III (2003) (update proposed 2009) | 4.5 |
| Mirex | Pesticide | Annex A (2004) | Annex I (2003) | 4.7 |
| Toxaphene | Pesticide | Annex A (2004) | Annex I (2003) | 4.10 |
| Polychlorinated biphenyls (PCBs) | Industrial / By-product | Annex A with specific exemptions and under Annex C (2004) | Annex I and II (2003) (update proposed 2009) | 4.9 |
| Polychlorinated dibenzo-*p*-dioxins (PCDD) | By-product | Annex C (2004) | Annex III and IV (2003) | |
| Polychlorinated dibenzofurans (PCDF) | By-product | Annex C (2004) | Annex III and IV (2003) | |
| Chlordecone | Pesticide | Annex A (2009) | Annex I (2003) | |
| Alpha-hexachlorocyclohexane (α-HCH) | Pesticide / By-product | Annex A (2009) | Annex II (2003) (technical HCH) | 4.6 |
| Beta-hexachlorocyclohexane (β-HCH) | Pesticide / By-product | Annex A (2009) | | 4.6 |
| Lindane (γ-HCH) | Pesticide | Annex A (2009)<br>Specific exemption for use as a human health pharmaceutical for control of head lice and scabies as second line treatment | Annex II (2003) (update proposed 2009) | 4.6 |
| Pentachlorobenzene | Pesticide / Industrial / By-product | Annex A and Annex C (2009) | Amendment proposed (2009) | 4.8 |
| Hexabromobiphenyl | Industrial | Annex A (2009) | Annex I (2003) | |
| Hexabromodiphenyl ether and heptabromodiphenyl ether (commercial octabromodiphenyl ether) | | Annex A (2009) | Amendment proposed (2009) | 4.12 |
| Perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride | | Annex B (2009) | Amendment proposed (2009) | 4.14 |
| Tetrabromodiphenyl ether and pentabromodiphenyl ether (commercial pentabromodiphenyl ether) | | Annex A (2009) | Amendment proposed (2009) | 4.12 |

Table 4.1 continued

| POP | Uses | Date of Stockholm Convention Listing | LRTAP POPs Protocol | Results section |
|---|---|---|---|---|
| Technical endosulfan and its related isomers | | Annex A (2011) | Under review | 4.11 |
| Hexabromocyclododecane (HBCDD) | | Annex A (2013) | Under review | 4.13 |
| Short-chained chlorinated paraffins (SCCPs) | | Proposed | Amendment proposed (2009) | [a] |
| Polychlorinated naphthalenes (PCNs) | | Annex A and C (2015) | Amendment proposed (2009) | [a] |
| Hexachlorobutadiene (HCBD) | | Annex A (2015) | Amendment proposed (2009) | [a] |
| Pentachlorophenol (PCP) | | Annex A (2015) | Under review | [a] |
| Decabromodiphenyl ether | | Proposed | | [a] |
| Dicofol | | Proposed | Under review | [a] |
| Pentadecafluorooctanoic acid, its salts | | Proposed | | [a] |
| Polycyclic aromatic hydrocarbons (PAHs) | | | Annex III (2003) | [a] |

[a]These chemicals and groups of chemicals are further addressed in the (2016) AMAP assessment of Chemicals of Emerging Arctic Concern currently under preparation.

## 4.1 Aldrin, endrin and dieldrin

### 4.1.1 Air trends

Air concentrations of aldrin and endrin measured at Alert (Canada) did not show any consistent trends (Fig. 4.1), with aldrin mostly non-detectable. Alert and Stórhöfði (Iceland) reported time trends of dieldrin (Fig. 4.2) since 1993 and 1995, respectively, and slow decreasing trends were found with $t_{1/2}$ = 15 y (4.6% per year) and $t_{1/2}$ = 25 y (2.8% per year), respectively.

Air concentrations tend to be lowest in the colder months. Slight decreases in the warmest months were observed for dieldrin in all years at Alert which may be related to greater photodegradation during Arctic summer under 24-hour sunlight. The Villum Research Station (Greenland) also reported relatively low concentrations of dieldrin in air (Fig. 4.3) with a statistically significant correlation with temperature (Bossi et al., 2013).

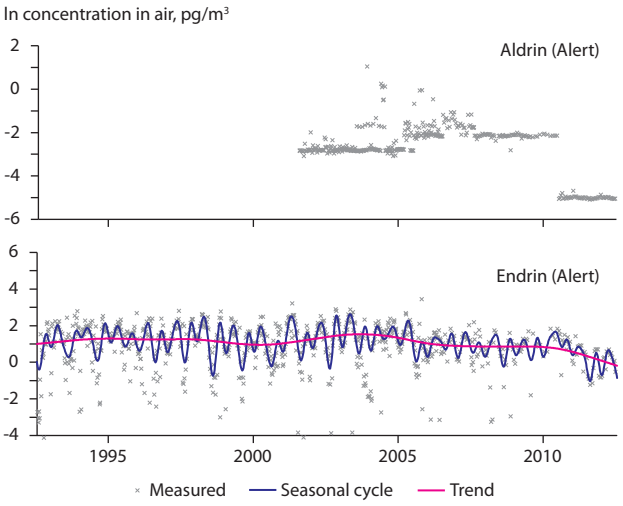



Figure 4.1 Trends in air concentration of aldrin and endrin at Alert (Canada).

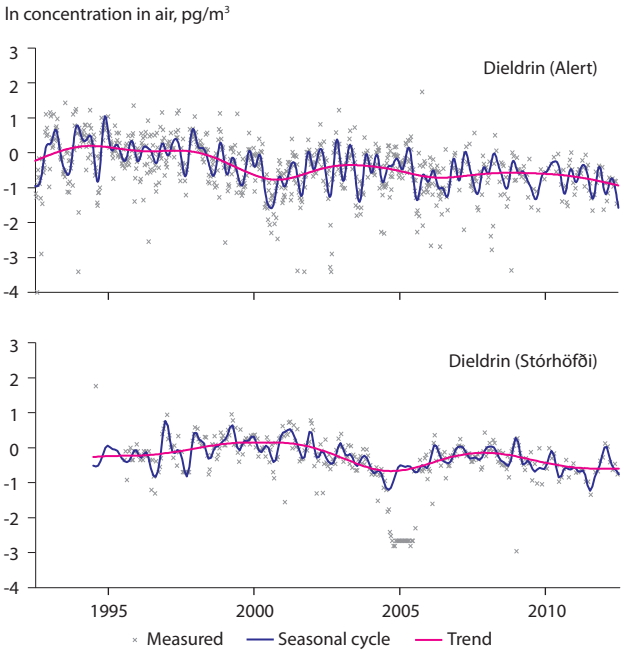



Figure 4.2 Trends in air concentration of dieldrin at Alert (Canada) and Stórhöfði (Iceland).

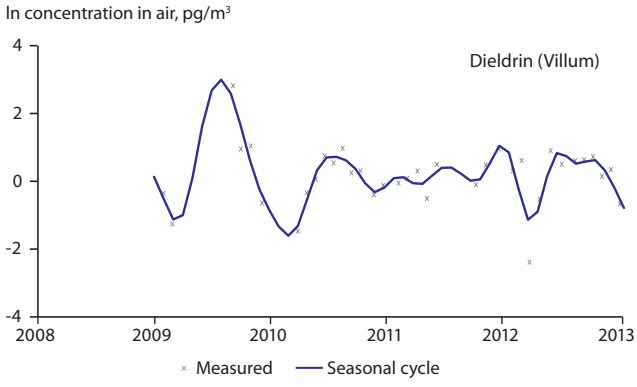



Figure 4.3 Trends in air concentration of dieldrin at the Villum Research Station (Greenland).

### 4.1.2 Biota trends

Twenty-two dieldrin time-series starting before 2000 were assessed; 19 (including freshwater fish, seabird eggs, and marine mammals) were from Arctic Canada, and three (marine mammals) from eastern Greenland. Ten showed significant decreasing trends or decreasing trends with a non-linear trend component; three increasing trends (all <1% per year) were identified, but none were statistically significant. The mean annual decrease for time-series starting before 2000 was 3.0%. For the post-2000 period, only one (ringed seal *Pusa hispida*, Canada) of 13 available time-series showed a significant decreasing trend, and the mean annual decrease was close to zero indicating little if any decrease in levels since 2000.

### 4.1.3 Discussion

Dieldrin in air probably results from re-emission from historical sources and the use of aldrin, which can be converted to dieldrin in the environment. This suggests that dieldrin in air has probably reached some sort of equilibrium with other environmental media as it has been banned under various national and international initiatives worldwide for more than 30 years.

The observed rate of dieldrin decrease in biota (a mean of 3% per year) also reflects very slow declines consistent with the air observations of barely discernible changes over the period since 1993 (a 3% annual change is equivalent to a first order half-life of 23 years). The indications are therefore that Arctic (aldrin and) dieldrin contamination has slowly decreased following bans introduced in the period before 2000. Levels of dieldrin currently observed in Arctic biota are relatively low (means of annual medians since 2010 of about 30–40 ng/g lipid weight (lw) in blubber of Canadian ringed seal populations, about 430 ng/g lw in beluga *Delphinapterus leucas* blubber from Canada, and 150–300 ng/g lw in eastern Greenland polar bear *Ursus maritimus* adipose tissue) and either stable or changing only slowly, again consistent with a pattern of general equilibration between residual historical accumulation of 'drins' in different (Arctic) environmental media and declining entry of dieldrin into Arctic marine food webs.

## 4.2 Chlordanes and nonachlors

### 4.2.1 Air trends

Decreasing trends in *trans*- and *cis*-chlordane were observed at most Arctic air monitoring sites (Fig. 4.4), with $t_{1/2}$ = 11–20 y (3.5–6.3% per year) for *cis*-chlordane and $t_{1/2}$ = 6–11 y (6.3–12% per year) for *trans*-chlordane. *Trans*- and *cis*-nonachlor decrease at $t_{1/2}$ = 17 y (4.1% per year) and $t_{1/2}$ = 10 y (6.9% per year) respectively at Zeppelin (Svalbard), and $t_{1/2}$ = 19 y (3.6% per year) and $t_{1/2}$ = 26 y (2.7% per year) respectively at Alert (Canada) (Fig. 4.4). Similar to the 'drins', slow declines in air with first order half-lives mostly longer than 10 years (<6.9% per year) for all chlordane- and nonachlor-related isomers reflects the fact that chlordane has been banned in western industrialized countries since the 1980s and time trends seem to indicate a tendency towards equilibrium with other media.

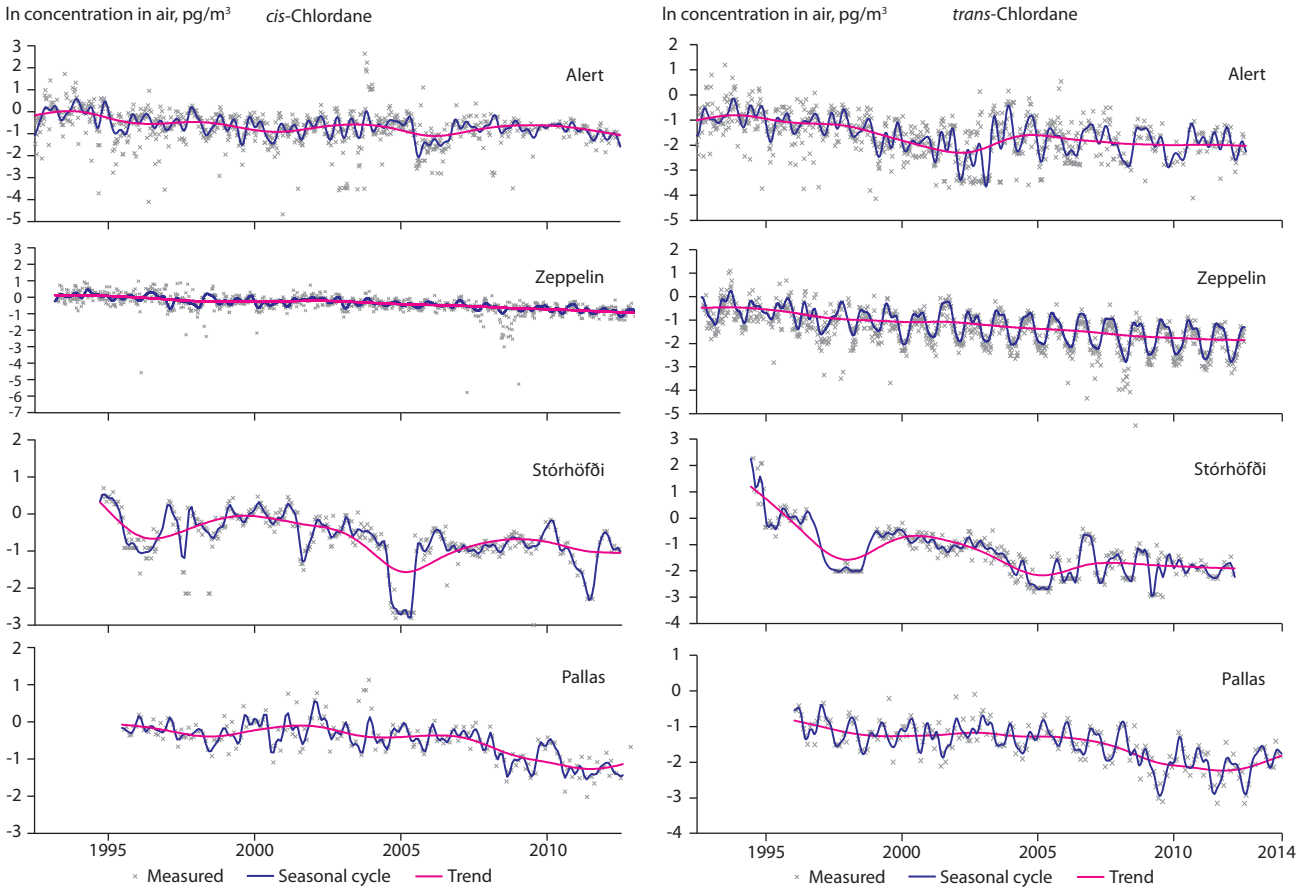
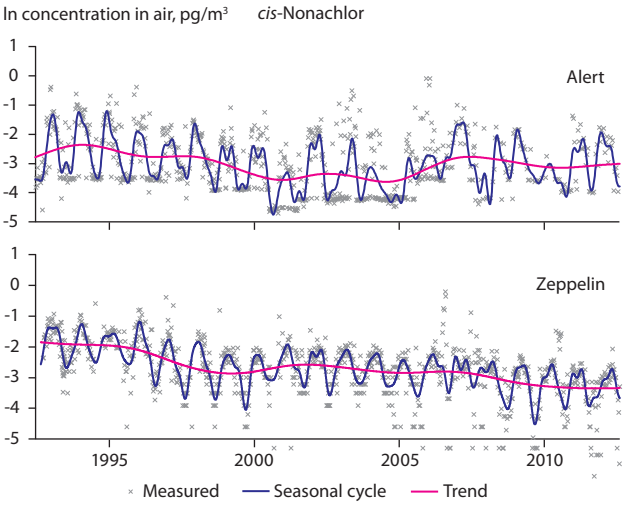



Figure 4.4 Trends in air concentration of chlordanes and nonachlors (see next page) at Arctic air monitoring sites.



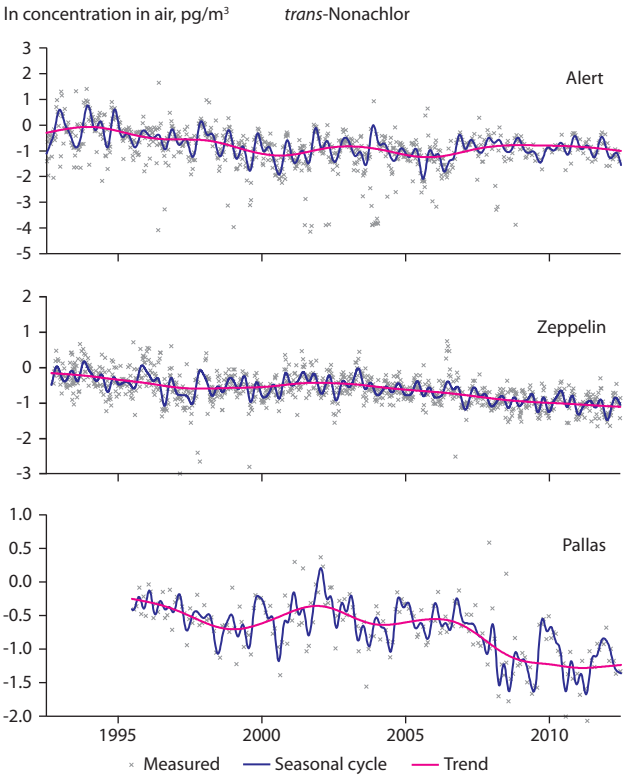




Figure 4.4 Continued.

In terms of seasonality, *trans*-chlordane measured in Arctic air generally showed higher winter concentrations. This seasonality is especially apparent at Zeppelin. Since *trans*-chlordane is less stable than *cis*-chlordane, this seasonality may result from enhanced photodegradation during the Arctic summer when there is 24-hour daylight. No consistent seasonality was observed for *cis*-chlordane in Arctic air, except at Alert where concentrations are slightly higher in spring and autumn. Due to the instability of *trans*-chlordane, it was expected that the *trans*- to *cis*-chlordane ratios would decrease over time. However, no consistent interannual trend was observed in this ratio at the four long time-series air monitoring sites. For the nonachlors, air concentrations were generally higher in the warmer period at most sites, probably due to revolatilization from secondary sources. Measurements of *cis*-chlordane and *cis*-nonachlor at the Villum Research Station (Greenland) are shown in Fig 4.5.

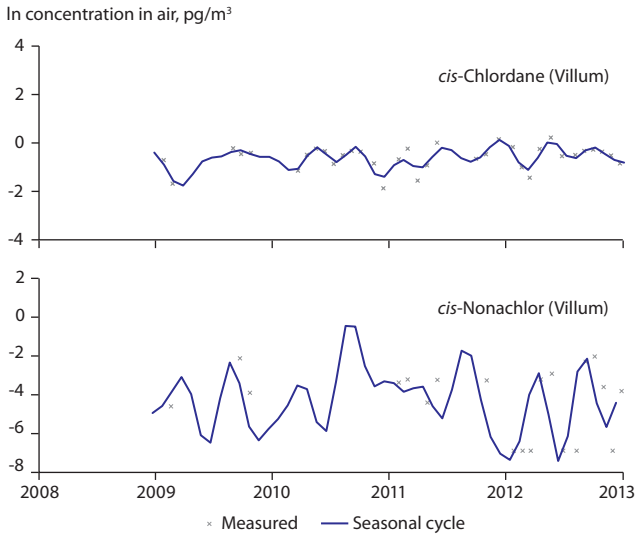



Figure 4.5 Seasonal cycles of *cis*-chlordane and *cis*-nonachlor in air at the Villum Research Station (Greenland).

The chlordanes are chiral chemicals that exist in specific, dual atomic arrangements (enantiomers), such that each enantiomer is the mirror image of the other and cannot be superimposed on each other (i.e. not identical). Most chiral compounds are produced with equal proportions of the two enantiomers (racemic), but microbial processes in soil and water may selectively degrade one of the enantiomers. Bidleman et al. (2015) analyzed air samples collected from Alert during 1994–2000 for enantiospecific *trans*-chlordane and *cis*-chlordane. The enantiomer fractions {EF= (+)/[(+)+(-)] enantiomers} were found to be close to racemic for *cis*-chlordane (0.505±0.004, n=162) and deviated further from racemic for *trans*-chlordane (0.470±0.013, n=165). EF of *trans*-chlordane showed lower values in warm seasons and higher values in cold seasons, in phase with the low and high cycles of the *trans*-/*cis*-chlordane ratio, suggesting greater volatilization of microbially degraded *trans*-chlordane from open ocean in summer-autumn as a result of seasonal ice cover loss in the Arctic versus the greater contribution of termiticides from home air ventilation in winter (Bidleman et al., 2013).

### 4.2.2 Biota trends

Long time-series were assessed for ΣCHL (sum of *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor and oxychlordane) as well as for individual compounds. The largest numbers of time-series starting before 2000 were for ΣCHL (31), *trans*-nonachlor (35) and oxychlordane (20). Close to half of the ΣCHL, *trans*-nonachlor and oxychlordane time-series showed decreasing trends or decreasing trends with a non-linear trend component (Fig. 4.6). This was also the case for *cis*-chlordane and *trans*-chlordane (not shown in the graphic). Although one *trans*-nonachlor time-series did show a significant increasing trend, this particular species-location combination (blue mussel *Mytilus edulis*, from Hvalstod in Hvalfjörður southwestern Iceland) was located near a whale processing site, which is considered a local source of contamination that may be responsible for increasing trends in several POPs at this site (see the Icelandic national report, Annex 3). The trends for chlordane compounds from all sites ranged between annual decreases of 3.6% for *trans*-nonachlor and 9.7% for *trans*-chlordane.

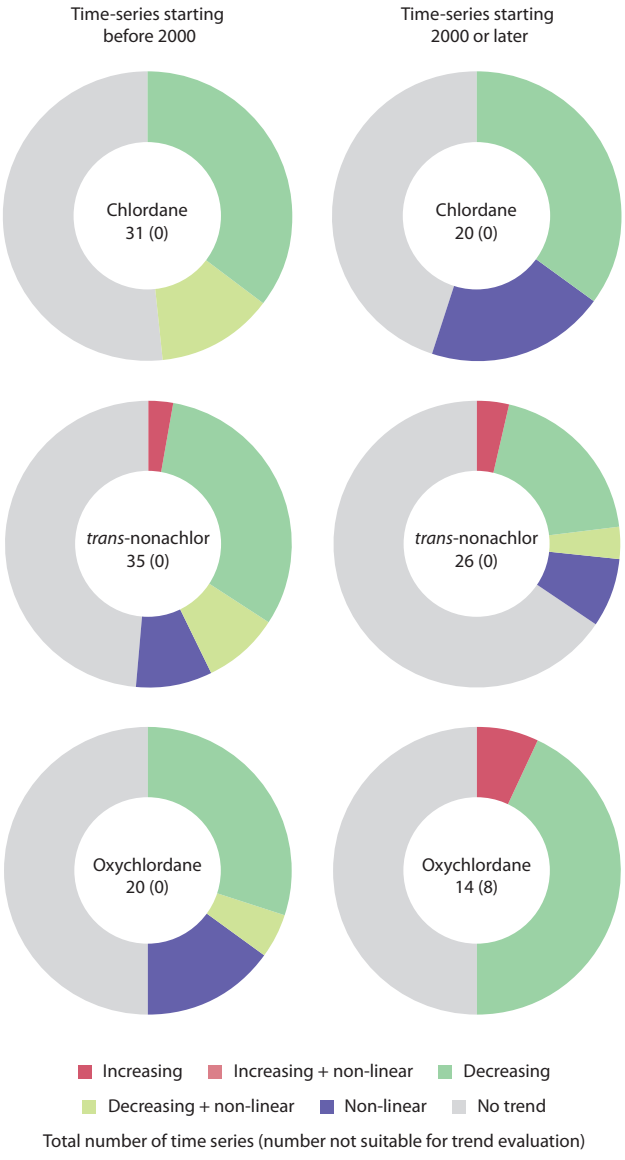



Figure 4.6 Overview of trend results for ΣCHL, *trans*-nonachlor and oxychlordane for time-series starting before 2000 and after 2000.

As for most organochlorines, the proportion of significantly decreasing trends in biota is less and the proportion of no trend time-series is greater when considering time-series starting after 2000. The annual decrease since 2000 ranged from 0.6% for *trans*-nonachlor to 6.8% for *trans*-chlordane.

Oxychlordane is the primary metabolite of chlordane and is very persistent (Bondy et al., 2000); seabirds and marine mammals are able to metabolize *cis*- and *trans*-chlordane (Fisk et al., 2001). Hence, the ratio between oxychlordane and ΣCHL in some species may provide a rough indication of the age of chlordane residues in the environment. Figure 4.7 shows temporal trends in the ratio of oxychlordane to ΣCHL in three (selected) long time-series. Differences between the ratio in the three species (generally increasing in the order beluga < ringed seal < polar bear) reflect the relative metabolic potential among these species. All three time-series show an increase in the ratio over time; however, this increase is only significant for ringed seal (log linear regression of annual medians, $p<0.01$). This indicates that levels of the metabolite oxychlordane increased relative to some of its precursors, for example, *trans*-nonachlor and *trans*-chlordane (Tashiro and Matsumura, 1978).

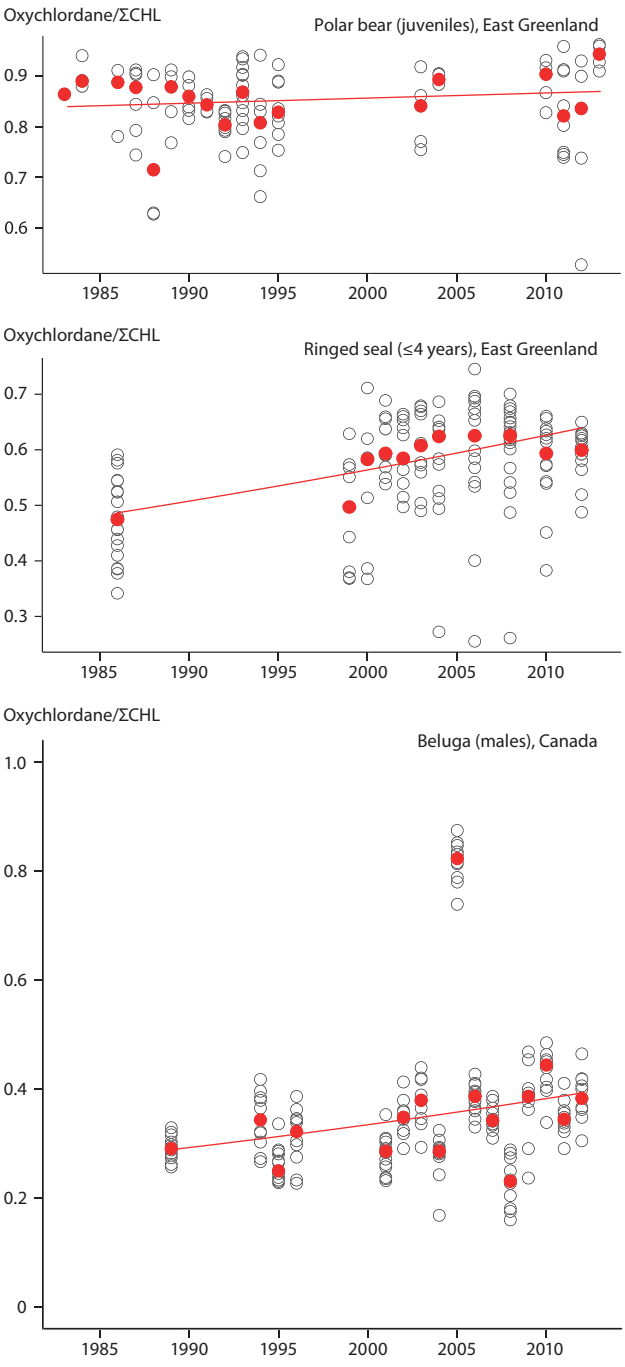



Figure 4.7 Trends in the ratio of oxychlordane to ΣCHL in selected Arctic long time-series.

### 4.2.3 Discussion

Trends in biota are consistent with those in air with approximately similar rates of decrease, indicating that Arctic chlordane contamination has decreased following the bans introduced in western industrialized countries since the 1980s.

The highest recent (post-2010) ΣCHL levels are found in pilot whale (*Globicephala melas*) blubber from the Faroe Islands (1500–2200 ng/g lw) and polar bear adipose tissue from eastern Greenland (1700–2100 ng/g lw). In Canadian and Greenlandic ringed seal populations, recent annual medians were 100–300 ng/g lw in blubber. These can be compared with the highest annual medians in the entire time-series of about 7500 ng/g lw (pilot whale), 4000 ng/g lw (polar bear), and 750 ng/g lw (ringed seal).

Similar to the 'drins', trends in levels of chlordanes in Arctic air and biota appear to reflect the influence of bans and controls on these substances introduced in western industrialized countries, including most countries near the Arctic, as early as the 1970s. The slow declines observed in air, with $t_{1/2}$ = >10 years (<6.9% per year) for all chlordane- and nonachlor-related isomers, are comparable to the annual decreases observed in biota. Levels in Arctic biota are therefore expected to continue to decrease, but only very slowly as chlordanes present in environmental media slowly degrade.

## 4.3 DDTs

### 4.3.1 Air trends

Among the four Arctic sites with long time-series, Zeppelin (Svalbard) is the only station where all DDT isomers show consistent decreasing trends (Figs. 4.8 and 4.9). For *p,p'*- and *o,p'*-DDT, $t_{1/2}$ = 5 y (14% per year) and $t_{1/2}$ = 8.5 y (8.2% per year), respectively, at this site. Declines were also evident at Stórhöfði (Iceland) from the late 1990s to early 2000s for *p,p'*-DDT, DDE and DDD, and at Pallas (Finland) from 1999 to 2011 for *p,p'*-DDT. Air concentrations at other Arctic sites seem to have reached steady state and there are no discernible decreasing trends.

Air concentration maxima for all DDT isomers at Zeppelin and *p,p'*-DDE at Alert (Canada) and Pallas generally occurred in winter. Halsall et al. (1998) reported that DDT-related compounds tend to associate with particles, which show a greater influx to the Arctic in winter/spring during the Arctic Haze season. Higher precipitation rates in summer may also result in lower summer particle concentrations due to enhanced scavenging along the transport pathway to the Arctic. On the other hand, Bossi et al. (2013) reported higher *p,p'*-DDT concentrations in summer at the Villum Research Station (Greenland), showing significant correlation with temperature and negative correlation with ice cover.

Bidleman et al. (2014) analyzed the fractions of different DDT isomers and found the proportion of *p,p'*-DDT to *p,p'*-DDE decreased significantly at Zeppelin, reflecting an enhanced contribution from aged sources. However, the proportion of *p,p'*-DDT to *o,p'*-DDT also decreased with time at Zeppelin, which may indicate a contribution from dicofol (a pesticide that contains DDT impurities that have a greater proportion of *o,p'*-DDT than technical DDT), or soil emissions, or preferential degradation of *p,p'*-DDT relative to *o,p'*-DDT over time. Such declines were not observed at Pallas or Stórhöfði. At Alert, the proportion of *p,p'*-DDT to *p,p'*-DDE increased after 2001 and remained high until 2012 for no apparent reason. Bidleman et al. (2014) suggested this could be due to (i) long-range transport from tropical areas where DDTs are still in use, (ii) transport from China, where high proportions of *p,p'*-DDT to *p,p'*-DDE are observed in air, or (iii) re-emission from soil and oceans due to warming. At Alert, the proportion of *p,p'*-DDT to *o,p'*-DDT [expressed as $F_{DDTO}$ = *p,p'*-DDT / (*p,p'*-DDT + *o,p'*-DDT)] increased from medians of 0.25–0.50 in 1993–2001 to 0.40–0.68 in 2002–2012; which is closer to the estimated vapor composition of WHO technical DDT ($F_{DDTO}$ = 0.58) than that of Chinese dicofol ($F_{DDTO}$ = 0.04).

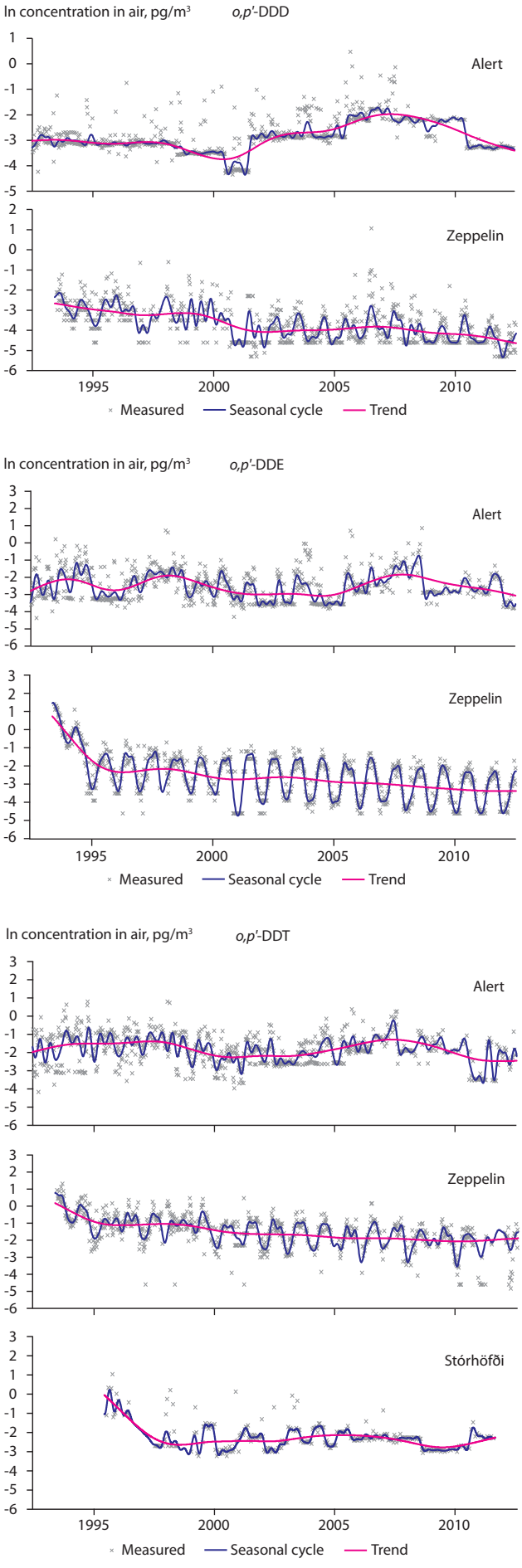



Figure 4.8 Trends in air concentration of *o,p'*-DDTs at Arctic air monitoring sites.

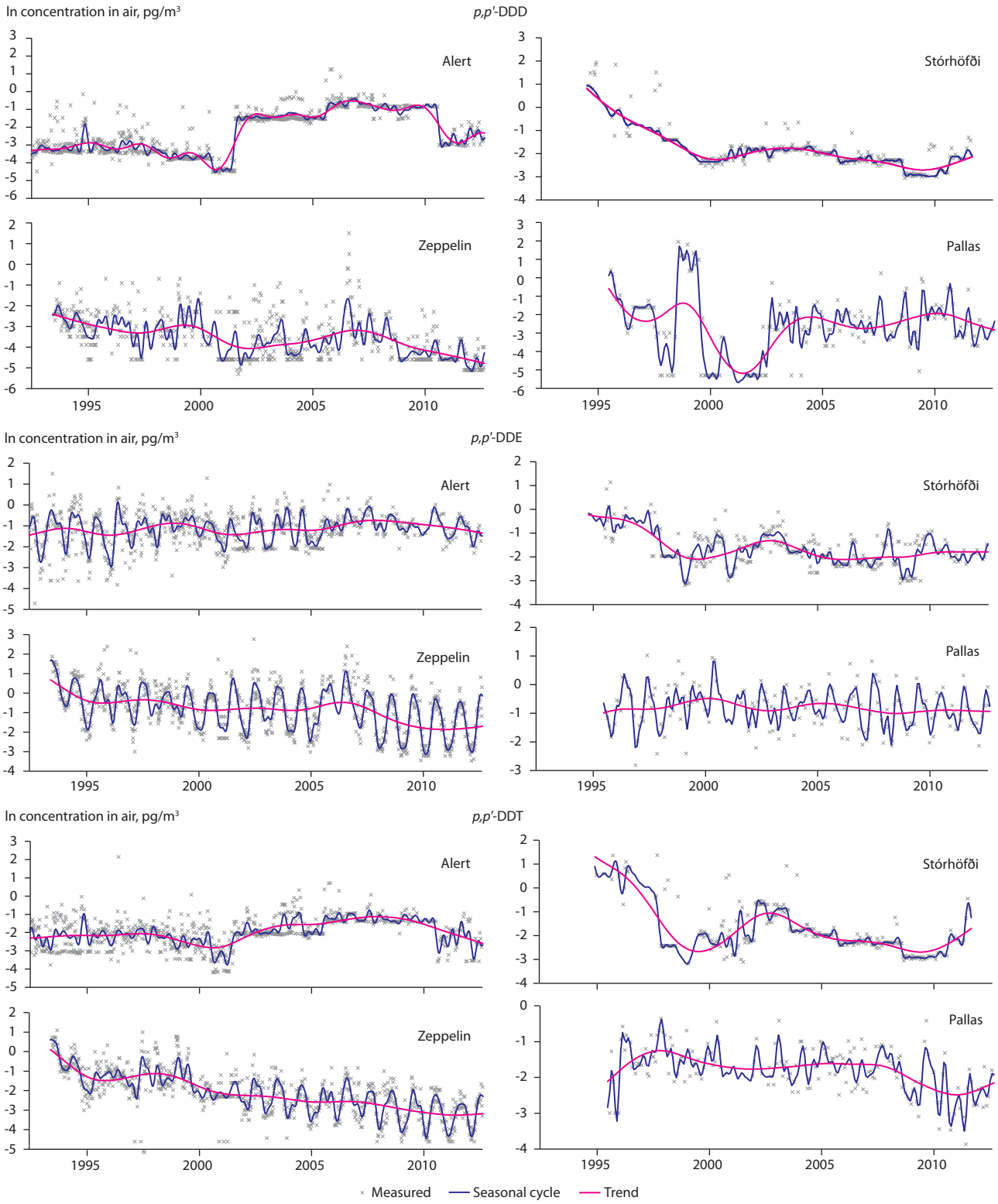



Figure 4.9 Trends in air concentration of *p,p'*-DDTs at Arctic air monitoring sites.

Although this is consistent with measured values from other air sampling campaigns in Canada, the United States and Mexico, the reason for such high and increasing $F_{DDTO}$ values is unknown. Bidleman et al. (2014) noted that many countries manufactured technical DDTs with varying compositions of DDT compounds. Such information on percentages of DDT compounds in technical mixtures and reservoirs (e.g. soil) combined with numerical modelling would help in the understanding of such observations.

### 4.3.2 Biota trends

Time-series of ΣDDT (sum of *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT) and individual *o,p'* and *p,p'* isomers in biota were analyzed, including 165 time-series starting before 2000 and 111 starting 2000 or later, most of these for ΣDDT (71%) and *p,p'*-DDE (82%). Many time-series of *p,p'*-DDD in mussels and some marine fish could not be evaluated owing to several years with medians below the detection limit. 46% of ΣDDT time-series showed a significant decreasing trend, while

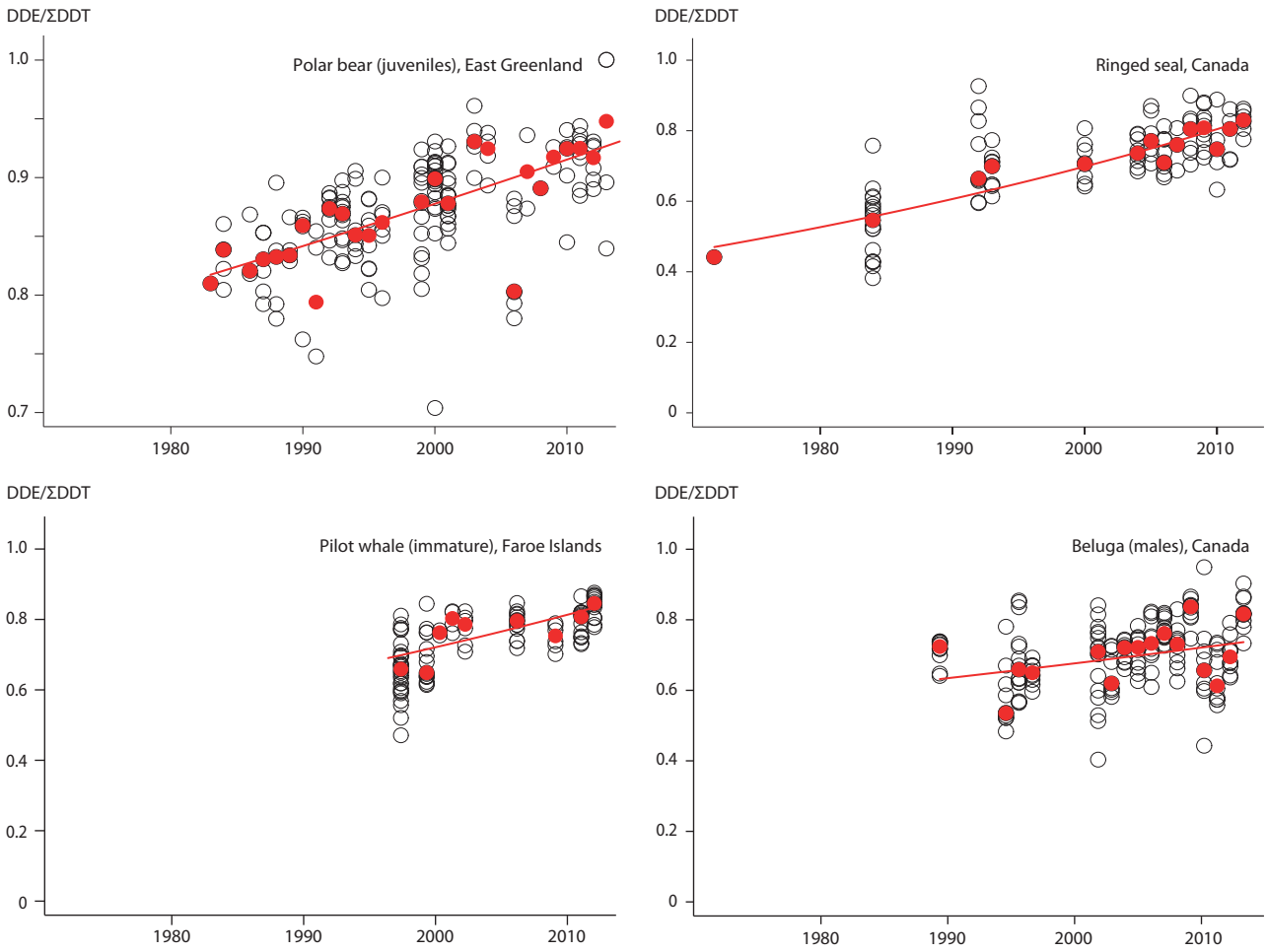



Figure 4.10 Trends in the ratio DDE to Σ*p,p*'-DDTs in selected long time-series.

this was only the case in 32% of *p,p*-DDE time-series. One ΣDDT time-series (blue mussel from Hvalstod in Hvalfjörður southwestern Iceland) showed a significant increasing trend, but this particular site is near a whale processing station that is a possible source of local contamination (Sturludottir et al., 2013).

The mean annual decrease was 4.2% and 4.1% ($t_{1/2}$ = 17 y) for ΣDDT and *p,p*'-DDE, respectively. For species-tissue-location combinations having time-series of both ΣDDT and *p,p*'-DDE, no significant difference in annual change was found (paired t-test, $p = 0.64$). *p,p*'-DDE is the major metabolite of DDT (Kelce et al., 1995). The ratio of *p,p*'-DDE to Σ*p,p*'-DDTs may be a rough indicator of the age of DDT residues in the environment. Figure 4.10 shows the trends of this ratio in selected long time-series. In all four time-series the ratio *p,p*'-DDE to Σ*p,p*'-DDTs increases with time and was significant in polar bear, ringed seal and pilot whale (log-linear regression of annual medians, $p < 0.01$, $p < 0.01$ and $p = 0.03$, respectively), but not in beluga ($p = 0.12$). These results are consistent with an absence of 'fresh' sources of DDT to the Arctic environment. This is also supported by the 12 *p,p*'-DDT time-series with a mean annual decrease of 9.1% ($t_{1/2}$ = 7.6 y), the largest mean rate of decrease of all DDT isomers considered, and a similar rate of decline to that observed in air at Zeppelin.

The proportion of post-2000 ΣDDT time-series showing a significant decreasing trend was much lower than for the time-series starting before 2000 (Fig. 4.11), as was also the case for


Figure 4.11 Overview of trend results for ΣDDT and *p,p*'-DDE for time-series starting before and after year 2000.

PCBs (see Sect. 4.9). In the post-2000 time-series, mean annual decreases were 2.4% and 3.6% ($t_{1/2}$ = 29 and 19 y) for ΣDDT and *p,p'*-DDE , respectively (which is larger than the decreases observed for PCBs).

Seventeen time-series of *o,p'*-DDE, *o,p'*-DDD, and *o,p'*-DDT were available for pilot whales from the Faroe Islands and beluga and northern fur seal (*Callorhinus ursinus*) from the United States. Female and juvenile beluga from East Chukchi / Bering Sea and northern fur seals from St. Pauls Island / Bering Sea showed significant non-linear trends with levels relatively constant, followed by a sharp decrease in most recent samples.

### 4.3.3 Discussion

As a result of national action, levels of DDT and DDE were already strongly declining in biota in western Europe and North America by the mid-1990s (AMAP, 1998). The few available time-series for Arctic biota that extend back to 1970s show that, by 2000, DDE levels had decreased from around 1 to <0.04 ng/g lw in pike (*Esox lucius)* muscle at Storvindeln in northern Sweden (Fig. 4.12), by a factor of about 5 in seabird eggs at Prince Leopold Island (Canadian Arctic Archipelago), and by a factor of about 3-4 in ringed seal blubber (Canadian Arctic Archipelago) and polar bear adipose tissue (Hudson Bay) (AMAP, 2004).

For the time-series starting before 2000, more time-series with a positive annual change were found in the eastern part of the Arctic than in the western part (Fig. 4.13). Most blue mussel time-series from Iceland and Norway had increasing trends although the trend was significant in only one case.

The highest levels of ΣDDT observed in recent years are found in pilot whales from the Faroe Islands with annual medians since 2010 of 5500–8300 ng/g lw, and in beluga from Canada with annual medians of 1000–4000 ng/g lw. Levels in seabird eggs are also still relatively high in recent years when expressed on a lipid weight basis (annual medians of 400–1300 ng/g lw). In polar bears, which have the highest levels of many POPs, recent levels of ΣDDT are lower, at around 200–300 ng/g lw. This probably reflects the ability of polar bears to metabolize the most persistent DDT compounds (Bernhoft et al., 1997).

Although national and global restrictions have resulted in continuing decreases in levels of DDTs in Arctic ecosystems, biota and humans (AMAP, 2014), the major decline occurred before 2000. This was also the case for air where decreases in concentration were no longer apparent for most DDT isomers after 2000 at sites other than Zeppelin. However,

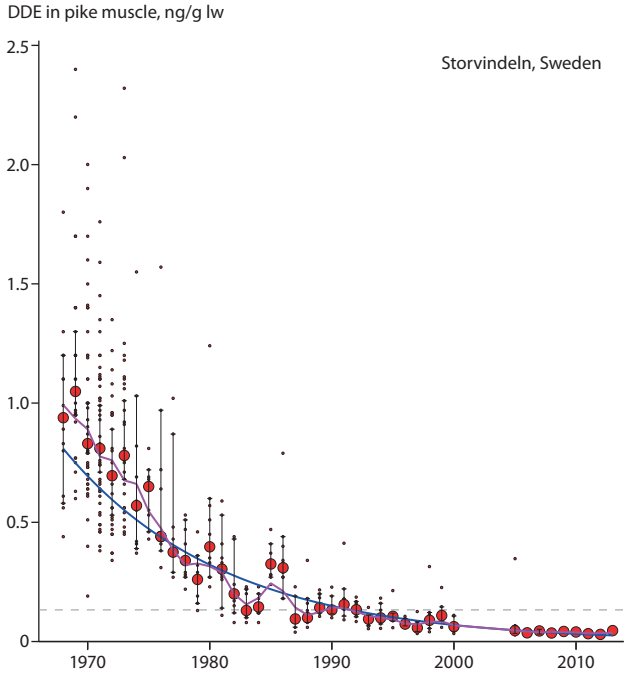



Figure 4.12 Trend in DDE concentrations in pike from Storvindeln (Sweden).

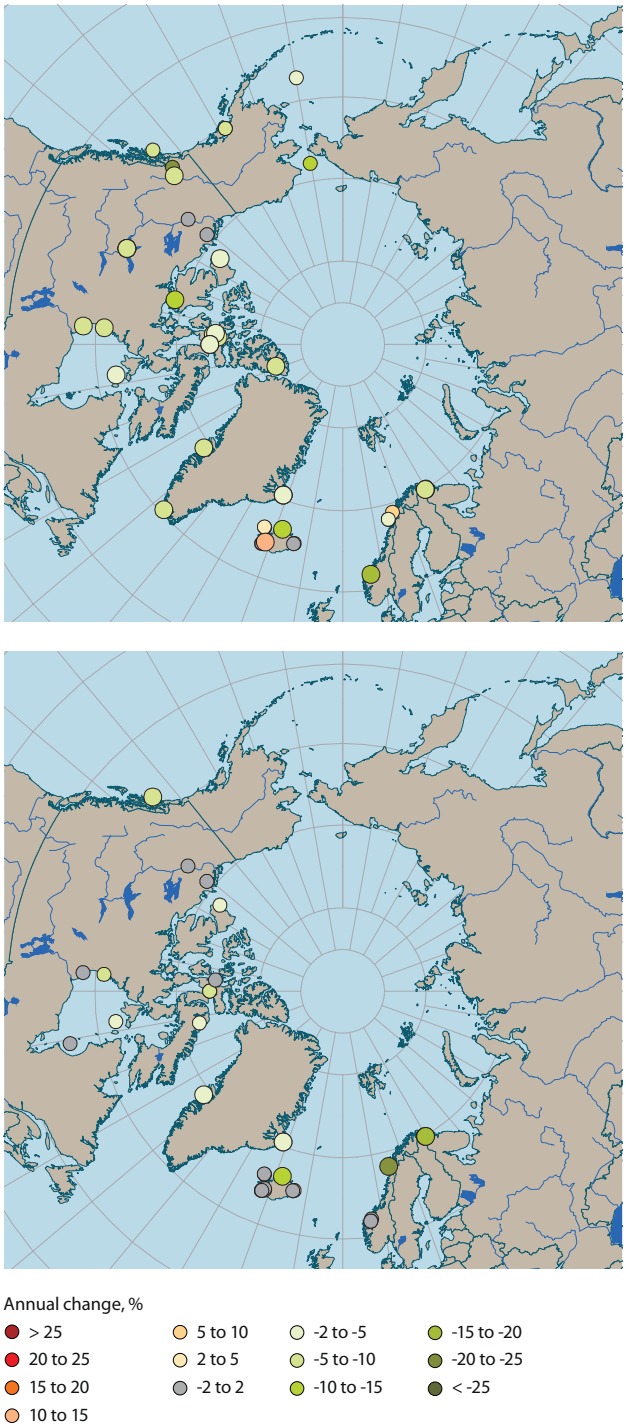



Figure 4.13 Geographical distribution of annual changes in ΣDDT in all time-series (upper) and in time-series starting at or after 2000 (lower) (larger symbols represent trends that are statistically significant or where statistical power was 'adequate').

long-range transport continues to be a source of DDTs to the Arctic, for instance, temperature can only account for about 27% of DDT variability in air at the Villum Research Station meaning that long-range sources cannot be excluded. *p,p'*-DDE concentrations were not correlated with temperature, indicating a predominance of long-range transport rather than re-emission (Bossi et al., 2013).

Concerns still exist regarding the potential for fresh releases of DDTs from contaminated Arctic areas, in addition to long-range transport from continuing-use areas. The Arctic Contaminant Action Program (ACAP) was established to follow-up on the findings of previous AMAP assessments. In 2001, ACAP initiated a project on *Environmentally Sound Management of Stocks of Obsolete Pesticides in the Russian Federation* to compile information and promote environmentally sound management of obsolete pesticide stockpiles in Russian territories in or close to the Arctic. Stocks of about 6800 tonnes of obsolete pesticides (including DDTs, toxaphene and hexachlorocyclohexanes), were identified during the project inventory activities in ten northern regions of the Russian Federation (Altai Krai, Arkhangelsk Region, Komi Republic, Magadan Region, Omsk Region, Tyumen Region, Altai Republic, Republic of Sakha (Yakutia), Tomsk Region, and Krasnoyarsk Krai). Many of the stockpiles were poorly stored. To reduce exposure of humans and the environment, most stocks were repackaged and transported to interim storage facilities to await environmentally sound destruction. Total stocks of obsolete pesticides in the Russian Federation have been estimated at 40,000 tonnes, mostly originating from the Soviet era (ACAP, 2013).

## 4.4 Heptachlor and heptachlor epoxide

### 4.4.1 Air trends

Heptachlor and heptachlor epoxide in air were reported for the Canadian Arctic station of Alert (Fig. 4.14). While heptachlor showed no discernible trend at Alert, heptachlor epoxide showed a very slow decrease ($t_{1/2}$ = 22 y; 3.2% per year). It is known that heptachlor degrades readily to heptachlor epoxide once it is released to the environment. Thus, air concentrations of heptachlor at Alert were generally lower than those of heptachlor epoxide and appear more sporadic. Higher air concentrations of heptachlor epoxide were observed at Alert during the warm period (possibly reflecting enhanced re-emissions) with a slight dip in concentration in mid-summer which may be related to greater photodegradation and enhanced scavenging at this time.

### 4.4.2 Biota trends

Three time-series of heptachlor (seabirds and northern fur seals from the United States) were available, but in all years most values were below detection limits.

Twelve heptachlor epoxide time-series starting before 2000 were evaluated. All time-series concerned concentrations in seabird eggs or marine mammal tissues. Of these, only two showed significant decreasing trends or decreasing trends with a non-linear trend component (annual decreases of 2–3%; $t_{1/2}$ = 23–35 y), the rest showing no statistical trends. None of the six time-series for the post-2000 period showed significant trends other than one with a significant non-linear trend.

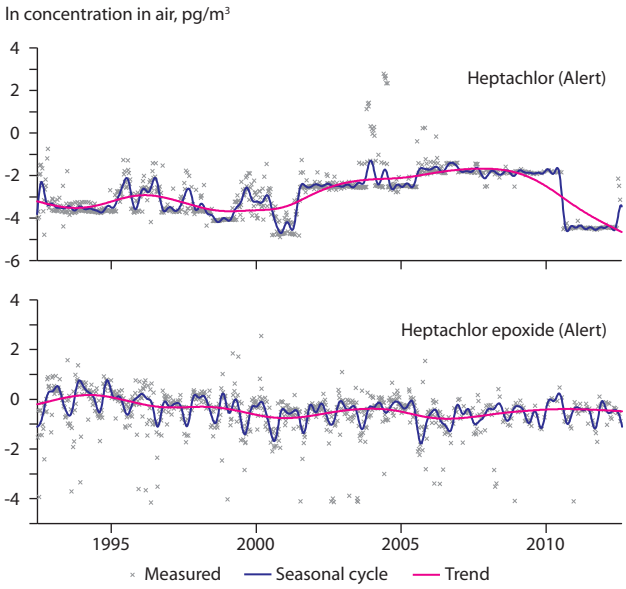



Figure 4.14 Trends in air concentration of heptachlor and heptachlor epoxide at Alert (Canada).

### 4.4.3 Discussion

Heptachlor was mostly non-detectable in Arctic air and biota. The air and biota results point to the long environmental half-life of heptachlor epoxide (the degradation product of heptachlor) and so Arctic contamination by these compounds is likely reflecting the slow and gradually diminishing influence of re-emissions from past contamination and long-range transport.

The highest recent (since 2010) annual medians were in polar bears (180–240 ng/g lw), and were similar to the annual medians in the 1980s and 1990s (up to 260 ng/g lw). In eggs of thick-billed murre (*Uria lomvia*) from Canada, the mean annual medians in the 1975–1979 period, and after 2010 did not significantly differ, reflecting no decrease or only a very slow decrease in heptachlor epoxide.

## 4.5 Hexachlorobenzene

### 4.5.1 Air trends

Hexachlorobenzene (HCB) in air was reported at three Arctic air monitoring stations and showed increasing trends at Zeppelin (Svalbard) and Stórhöfði (Iceland) over the last decade and a very slow decline ($t_{1/2}$ = >20 y; <3.5% per year) at Alert (Canada) (Fig. 4.15). Air concentrations of HCB at Zeppelin were higher during the 1990s, but similar at Alert and Zeppelin after 2000. It should be noted, however, that HCB has a tendency to 'break-through' in air samples collected using polyurethane foam plugs (PUFs); for example, HCB 'broke through' in about 30% of samples at Alert. When 'break-through' occurs, chemicals are lost from the sampling trap due to ineffective retention by the sorbing material with the

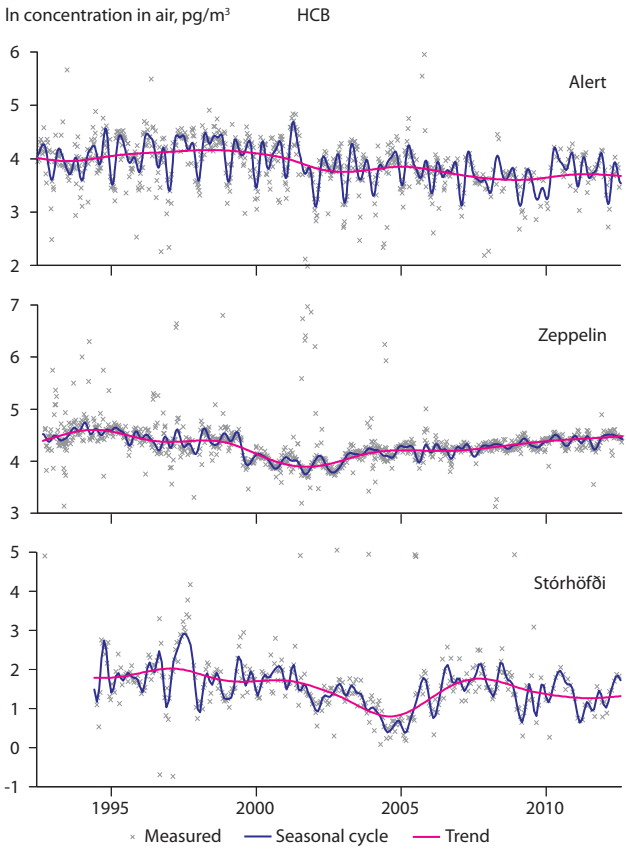



Figure 4.15 Trends in air concentration of HCB at three Arctic air monitoring sites.

result that air concentrations may be underestimated. Among the Arctic sites, the lowest HCB air concentrations were found at Stórhöfði. At the Villum Research Station, where samples should not be affected by break-through because measurements were made using sampling traps that contain both PUF and XAD resin to capture the gas phase, Bossi et al. (2013) found no seasonal variations in HCB air concentration between 2008 and 2010, but a weak correlation with temperature, and no correlation with ice cover. These observations suggest that HCB concentrations were not substantially influenced by re-emission at this site, and may still be controlled by primary sources (e.g. by-products of chlorinated chemicals and incomplete combustion processes). More recent measurements of HCB from this site are shown in Fig. 4.16.

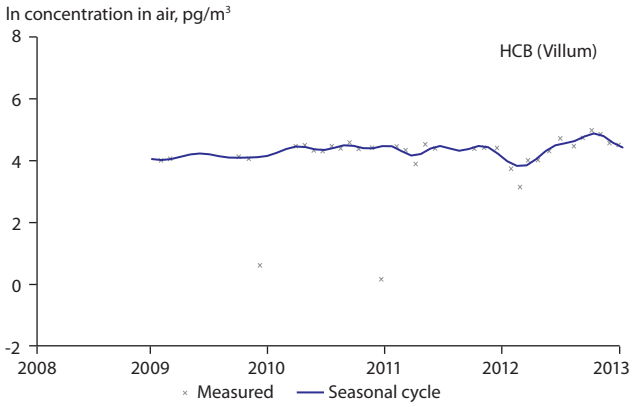



Figure 4.16 Seasonal cycle in HCB air concentration at the Villum Research Station (Greenland).

## 4.5.2 Biota trends

There were 54 and 43 time-series of HCB concentrations in biota starting before and after 2000, respectively. 35% of time-series from the North American and European Arctic starting before 2000 showed significant decreasing trends or a significant decreasing trend with a significant non-linear trend component. One time-series (black guillemot *Cepphus grylle* eggs from eastern Greenland) showed a significant increasing trend, and one time-series (adult male polar bears from eastern Greenland) showed an increasing trend with a significant non-linear trend component. The mean annual decrease for these time-series was 2.6% ($t_{1/2}$ = 27 y), which is lower than for most other organochlorine compounds (Fig. 4.17).

Six time-series starting after 2000 exhibited significant decreasing trends and three (from eastern Greenland) showed significant increasing trends. The remaining time-series showed no trend or had a significant non-linear trend component. The mean annual change for all time-series was 0.0% indicating that no decrease or a limited decrease has occurred since 2000.

## 4.5.3 Discussion

Despite the large proportion of decreasing time-series of HCB in biota observed across the North American and European Arctic over recent decades, the slow mean rate of the declines in biota and increasing trends in both air and biota at some sites since 2000 indicates that, unlike most other 'legacy' organochlorine pesticides, (primary and secondary) emissions and releases of HCB are still occurring. These are associated with by-production during manufacture of chlorinated compounds, the presence of HCB as an impurity in other pesticides, and its (industrial) combustion sources. Owing to its relatively high vapor pressure, the increase in HCB in air has also been associated with the decline in sea-ice cover and volatilization from environmental sinks in a warming Arctic (Hung et al., 2010). It is therefore possible that, despite its inclusion under the Stockholm Convention, HCB contamination, at least at some Arctic locations is increasing. The three biota time-series showing significant increasing trends since 2000 were from eastern Greenland, although one other eastern Greenland time-series (ringed seal) showed a significant decreasing trend.

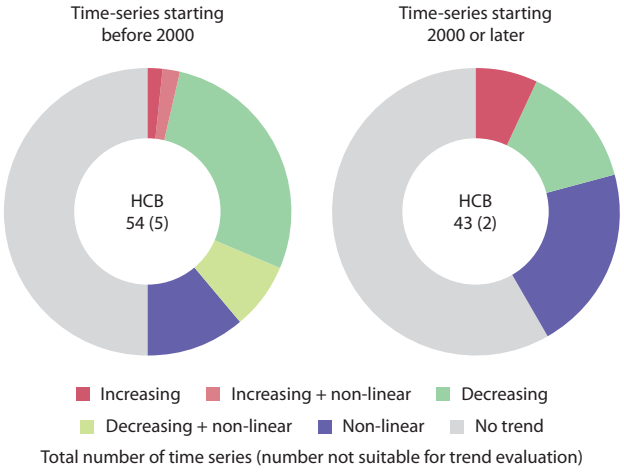



Figure 4.17 Overview of trend results for HCB in biota for time-series starting before and after 2000.

The highest recent annual medians (since 2010) were found in seabird eggs when expressed on a lipid weight basis (230–480 ng/g lw), pilot whales from the Faroe Islands (immature, 380 ng/g lw), and polar bears (90–300 ng/g lw).

## 4.6 Hexachlorocyclohexanes

### 4.6.1 Air trends

For hexachlorocyclohexanes (HCHs) in the Arctic, α-HCH and γ-HCH were found to be declining in air at all long-term monitoring stations: for α-HCH, $t_{1/2}$ = 4.9–5.8 y (12–14% per year) and for γ-HCH, $t_{1/2}$ = ~4 y (17% per year) at all sites, except for Stórhöfði (Iceland) where $t_{1/2}$ = 7.3 y (9.5% per year). While use of technical HCH has declined significantly since the 1980s, lindane (~99% pure γ-HCH) continued to be used in Canada until 2004[1] and the United States until 2009[2]. Atmospheric declines of lindane (γ-HCH) have accelerated in the Arctic with $t_{1/2}$ decreasing from 6.3 y (11% per year) for 1993–2001 to 4.6 y (15% per year) for 2002–2012 (Fig. 4.18) following restrictions on its use in North America (Hung et al., 2010). At the Villum Research Station (Greenland), γ-HCH measured in air (2008–2010) did not show a significant correlation with temperature, implying direct atmospheric transport from sources (Bossi et al., 2013). More recent air concentration data for γ-HCH from this station are given in Fig. 4.19.

β-HCH was also measured in air at Alert (Canada) and Stórhöfði, and showed decreasing concentrations in the late-1990s and early-2000s with $t_{1/2}$ = 3.7 y (1% per year, $r^2$=0.88, 1993–2002) and $t_{1/2}$ = 2.0 y (3% per year, $r^2$=0.72, 1995–2002), respectively (Fig. 4.20). At Alert, occasional episodically high concentrations were measured after 2004. At Stórhöfði, concentrations usually peaked in the warmer months and the temporal trend was more or less unchanged from 2002 to 2012. At Alert, concentrations increased from 2003 to 2007 followed by a decline to 2012 with many samples below detection in recent years. In 2007, when Arctic sea-ice extent reached an (at that time) record low (average extent in September 2007 was 4.3 million km² compared to a September average of 6.52 million km² for 1981–2010; NSIDC, 2015), high concentration episodes of β-HCH were observed throughout the year at Alert. This observation is consistent with the hypothesis that the dominant transport pathway for β-HCH to the Arctic is via the ocean (Li et al., 2002) and its re-emission from oceans due to sea-ice decline sustains levels in Arctic air (Wöhrnschimmel et al., 2012). However, in 2011 and 2012 when sea-ice extent was similarly reduced

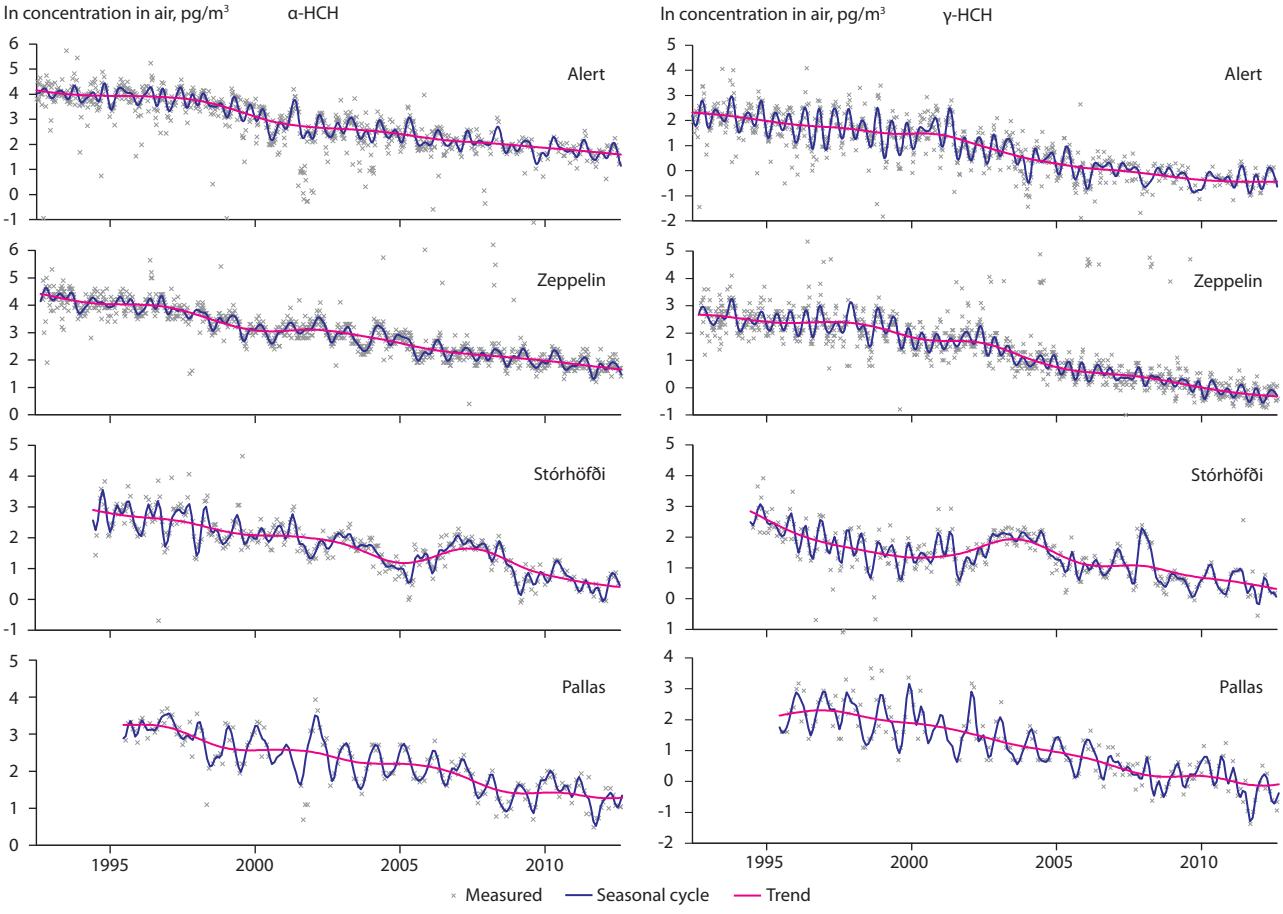



Figure 4.18 Trends in air concentration of α-HCH and γ-HCH at Arctic air monitoring sites.

[1] www.chemicalsubstanceschimiques.gc.ca/fact-fait/lindane-eng.php

[2] www.google.ca/url?sa=t&rct=j&q=&esrc=s&source=web&cd=5&cad=rja&uact=8&ved=0ahUKEwiqvs6Do_HNAhUj6YMKHbNVCr8QFgg2AQ&url=http%3A%2F%2Fwww.pops.int%2Fdocuments%2Fmeetings%2Fpoprc%2Fprepdocs%2FannexFsubmissions%2FLindane%2520USA doc&usg=AFQjCNEWFO1NuWQXJ6Aygo-5OjF667XKjA

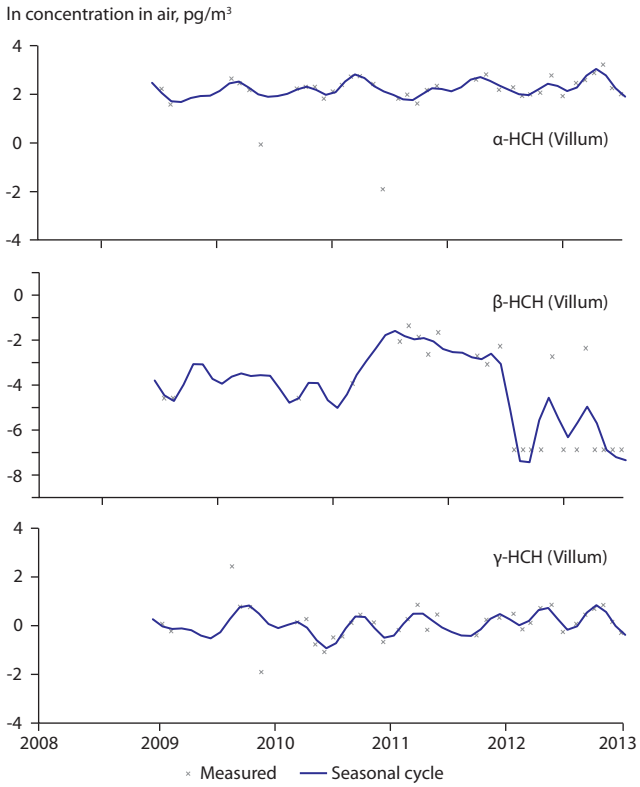



Figure 4.19 Seasonal cycles of α-HCH, β-HCH and γ-HCH in air at the Villum Research Station (Greenland).

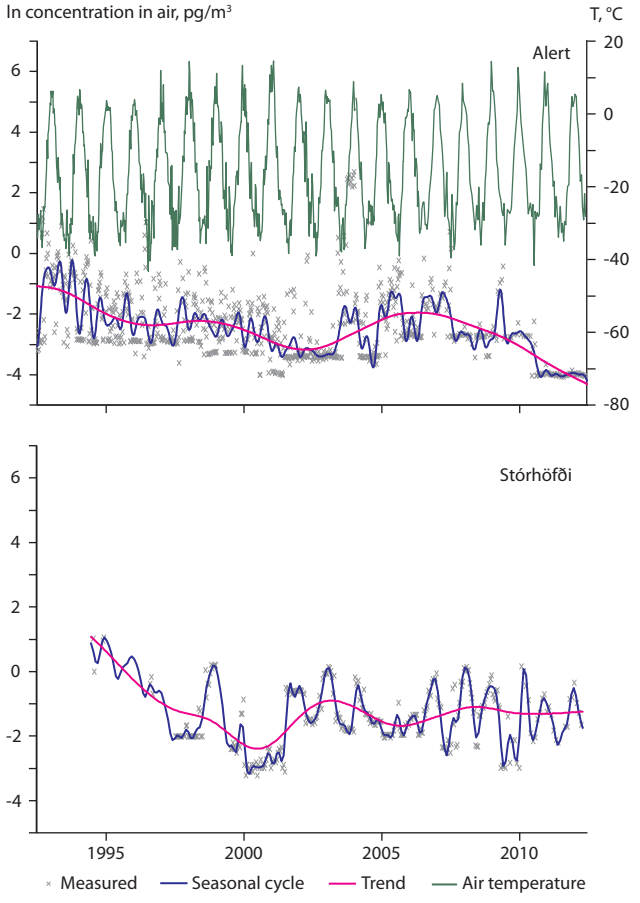



Figure 4.20 Temporal trend of β-HCH in air at Alert (Canada) and Stórhöfði (Iceland). For Alert, when concentrations were below detection limits, the values are replaced with 2/3 instrument detection limit (IDL).

(4.63 and 3.63 million km², respectively; NSIDC, 2015), almost all concentrations were below the detection limit at Alert. At Stórhöfði, summer peak air concentrations from 2002 to 2012 were similar, except for 2005 and 2006 which were lower. The difference in concentration patterns at Stórhöfði and Alert can be explained by the relative proximity of the Stórhöfði site to open ocean year round and that they have different source areas (Gregor et al., 1998). While Alert is still ice-bound throughout the year and high concentration episodes are likely to reflect long-range transport, at Stórhöfði β-HCH peaked in summer due to increased volatilization from adjacent seas.

Bidleman et al. (2013) summarized shipboard studies in the Arctic on the volatilization of α-HCH from seawater. While α-HCH concentrations measured in air in open water areas were non-racemic, those measured in ice-covered regions were almost racemic (see Sect. 4.2.1 for explanations of chiral compounds). In the Canadian Arctic, at Resolute Bay in 1999 (Jantunen et al., 2008) and Banks Island in 2008 (Wong et al., 2011), concentrations of α-HCH in air over the ocean increased during ice breakup in spring/summer, and was accompanied by a switch from almost racemic to non-racemic α-HCH in air. Enantiomer Fractions (EF) of α-HCH in air at Alert from 1994 to 2000 showed seasonal cycles with higher values in the cold season and lower values in the warm season (Bidleman et al., 2015). This observation suggests that Alert air was mostly influenced by background air (EF > 0.5) in winter-spring, while in summer-autumn, partially degraded α-HCH (EF < 0.5) was revolatilized from ice-free areas of the Arctic Ocean and transported to Alert. In 1999, the contribution of α-HCH revolatilized from the sea was estimated at 11% at Alert (a High Arctic land station with a nearby ocean mainly under ice) and 32% at Resolute Bay on Cornwallis Island (74.68°N, 94.90°W). The more southerly location of Resolute Bay with closer proximity to ice-free ocean areas is probably why this site has a greater influence of sea-volatilized α-HCH.

The ratios of γ-HCH to α-HCH (expressed as γ-HCH divided by α+γ-HCH) in air revealed slight increases for 1999–2003 at Zeppelin (Svalbard), 2001–2002 at Alert, and 2002–2004 at Stórhöfði, followed by general decline. A similar increase was not observed at Pallas (Finland) where the ratio declined slightly after 2002. Increases in such ratios observed at the three Arctic sites may reflect use of the lindane stockpile just before its phase-out in many countries and before global control under the Stockholm Convention.

### 4.6.2 Biota trends

In total, 110 α-HCH, β-HCH and γ-HCH time-series starting before 2000 were analyzed and 80 time-series starting after 2000 (Fig. 4.21). 80% of the α-HCH time-series starting before 2000 showed a significant decreasing trend or a significant decreasing trend with a significant non-linear trend component. For the time-series starting after 2000, the corresponding value was 53%. Mean annual decreases for time-series starting before 2000 were 8.9% ($t_{1/2}$ = 7.8 y), which was the highest decrease among the chemicals investigated in this assessment. For time-series starting after 2000 the mean annual decrease was 9.9% ($t_{1/2}$ = 7 y), which indicates that α-HCH has continued to decrease considerably since 2000 (See Table A6.3).

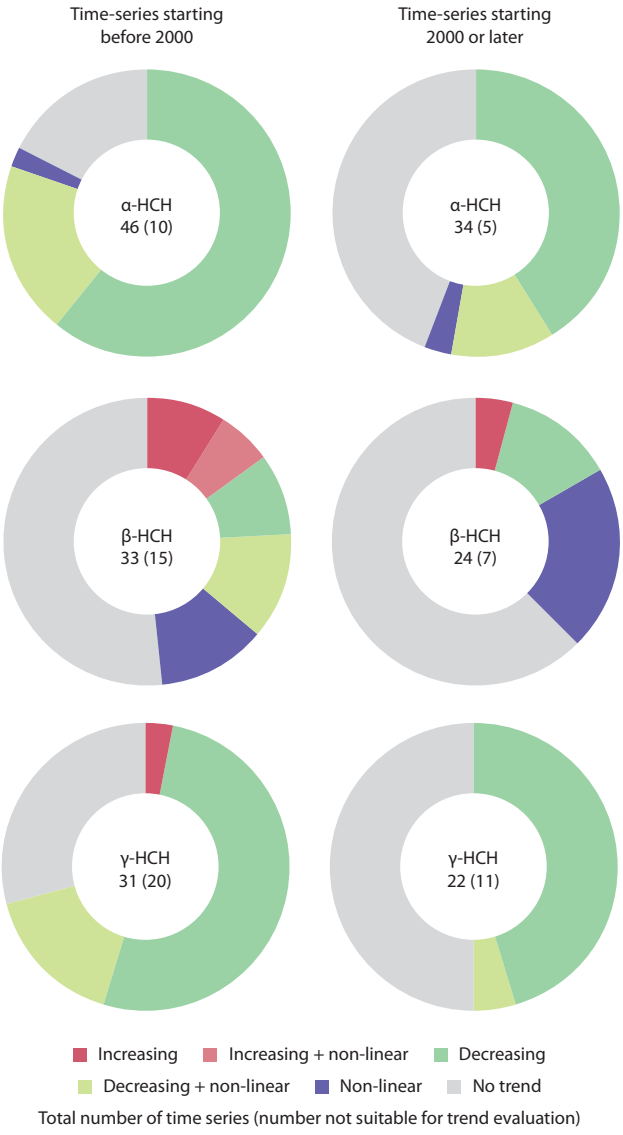



Figure 4.21 Overview of trend results for α-HCH, β-HCH and γ-HCH in biota for time-series starting before and after 2000.

The β-HCH time-series were quite different to those for α-HCH. More than half (64%) the β-HCH time-series starting before 2000 showed no significant trend, or a significant non-linear trend. Both significant decreasing and increasing trends were found. Three time-series showed significant increasing trends and two showed significant increasing trends with a significant non-linear trend component (one seabird time-series and one marine mammals time-series), both from the Canadian Arctic. The mean annual decrease was 1.5% ($t_{1/2}$ = >40 y) for time-series starting before 2000, which is the lowest decrease among organochlorines (see Fig. 5.1). For time-series starting after 2000, 83% showed no trend or a significant non-linear trend component. The difference between α-HCH and β-HCH could be due to different pathways to the Arctic caused by their different physical-chemical properties (Li and Macdonald, 2005). α-HCH and γ-HCH can be more readily metabolized (and so eliminated) than β-HCH, which is recalcitrant in most mammal species (and so biomagnifies) (Moisey et al., 2001). It should be noted that several β-HCH time-series could not be evaluated owing to more than two years with annual medians below detection limits, especially in the case of freshwater fish and blue mussels.

The γ-HCH time-series were similar to those for α-HCH, with 68% of time-series starting before 2000 showing a significant decreasing trend or a significant decreasing trend with a significant non-linear trend component. The corresponding value for the time-series starting after 2000 was 50%. Mean annual decreases were 7.6% ($t_{1/2}$ = 9.1 y) and 6.2% ($t_{1/2}$ = 11 y) for time-series starting before and after 2000, respectively; a smaller decrease than for α-HCH. It should be noted that several γ-HCH time-series could not be evaluated owing to more than two years with annual medians below detection limits.

### 4.6.3 Discussion

Time-series for α-HCH and γ-HCH in air and biota showed decreasing trends. α-HCH is declining faster in air ($t_{1/2}$ = ~5.3 y; 13% per year) than in biota (for time-series starting before 2000, $t_{1/2}$ = 7.8 y; 8.9% per year). The half-life of γ-HCH in biota was almost twice that for air. These observations suggest a lag in decline between air and Arctic biota for those isomers subject to atmospheric transport.

Recent (since 2010) annual medians for α-HCH were relatively low, at less than 10–30 ng/g lw in beluga and polar bears; although recent levels of β-HCH in polar bears and beluga are up to 140–260 ng/g lw. γ-HCH levels were low and in several cases below the detection limit; the highest annual medians occurred in beluga from Canada (~18 ng/g lw).

For time-series starting before 2000, no relationships were found between the magnitude of annual change and longitude, for α-HCH, β-HCH or γ-HCH (Fig. 4.22). However, considering only the northern fur seal and five ringed seal populations, the two seal populations at the highest latitude exhibit lower annual decreases in α-HCH, and there is no obvious tendency with longitude (Fig. 4.23). The two seal populations at the highest latitudes also differ for β-HCH, showing increasing trends, while the other populations show decreasing trends. However, the length of these time-series differed, with two extending back to the early 1970s, two to the mid-1980s, and two to the mid-1990s, so no firm conclusions can be drawn as to why this deviation was observed.

Similar plots were prepared for trend results for freshwater fish and seabirds, but no clear patterns or differences were seen in annual changes for α-HCH and β-HCH by longitude or latitude.

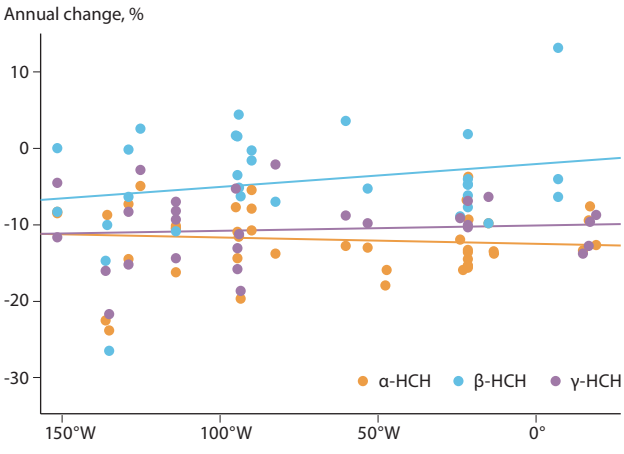



Figure 4.22 Percentage annual change for time-series starting before 2000 including all species for α-HCH, β-HCH and γ-HCH by longitude.

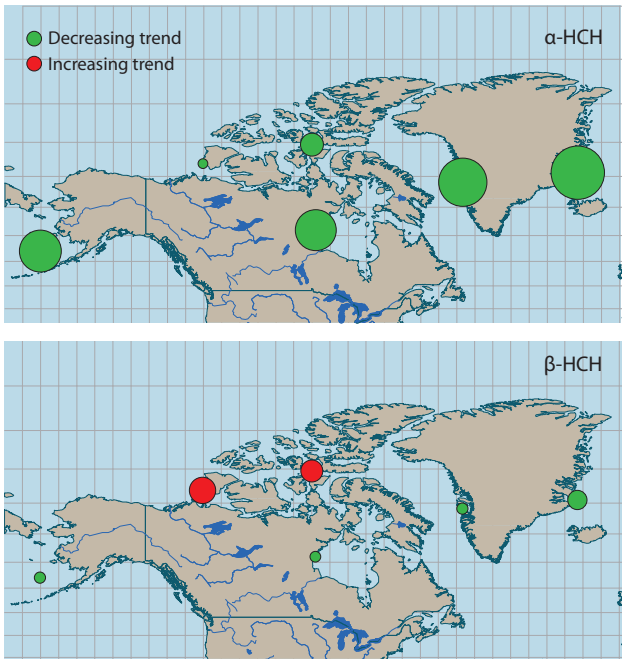



Figure 4.23 Annual decrease and annual increase in α-HCH and β-HCH in seal populations by longitude and latitude. The size of the circle represents the magnitude of the annual change.

## 4.7 Mirex

### 4.7.1 Air trends

Mirex is not routinely measured in air at any of the AMAP stations, so no time trends or concentration data are available. However, mirex concentrations in air have been analyzed at Alert (Canada), with concentrations found to be low or close to the detection limits (Hung et al., 2010).

### 4.7.2 Biota trends

Sixteen time-series for mirex starting before 2000 were available. These were from the Faroe Islands, Canada and the United States and include marine mammals, seabirds and freshwater fish. Four showed significant decreasing trends (one with a significant non-linear trend component), while the others showed either a non-linear trend or no trend. The mean annual decrease for time-series starting before 2000 was 6.7% ($t_{1/2}$ = 10 y). For the ten time-series starting after 2000 only one showed a significant decreasing trend, while the others showed either a non-linear trend or no trend. The mean annual decrease was lower at 1.9% ($t_{1/2}$ = >30 y).

### 4.7.3 Discussion

The highest recent (post-2010) levels of mirex in the time-series considered were found in pilot whale from the Faroe Islands (annual medians of about 120 ng/g lw). In seabird eggs from Canada and the United States, recent levels are low (<0.01–1.5 ng/g lw). With a mean annual decrease in biota of 6.7%, mirex is among the compounds included in this assessment with the highest mean annual decreases (Fig. 5.1). The relatively low annual decreases since 2000 also indicate that the main declines occurred in earlier years.

## 4.8 Pentachlorobenzene

### 4.8.1 Air trends

Pentachlorobenzene is only measured in air at Alert (Canada). However, owing to its high volatility, it is found to break-through in 40–60% of all samples collected with PUFs. Measured results are therefore not reliable.

### 4.8.2 Biota trends

Twelve time-series starting before 2000 were available for pentachlorobenzene, all from Canada or the United States. Many marine fish and blue mussel time-series (from Norway) could not be evaluated because of too many years with medians below the detection limit. Two time-series (one for freshwater fish and one for seabird eggs from Canada) showed significant decreasing trends or decreasing trends with a non-linear trend component. The others had significant non-linear trend components or no trend. The mean annual decreasing trend was 4.7% ($t_{1/2}$ = 15 y). None of the six time-series starting after 2000 showed significant trends other than one (seabird eggs from Canada) which had a significant non-linear trend showing an increase in the initial period followed by a decrease in more recent years.

### 4.8.3 Discussion

The highest recent (post-2010) (annual median) levels of pentachlorobenzene in the time-series considered were found in beluga from Canada (up to 40 ng/g lw). Recent levels are low in Canadian seabird eggs (<0.03 ng/g lw), but higher in seabird eggs from Alaska (0.8 ng/g lw). Few time trend studies of pentachlorobenzene are available. In the Great Lakes region of Canada, outside the Arctic, a dramatic decrease since 1979 was found in eggs from colonies of herring gull (*Larus argentatus*) (Bailey et al., 2009).

## 4.9 Polychlorinated biphenyls

### 4.9.1 Air trends

Polychlorinated biphenyls (PCBs) in Arctic air tend to be declining at all monitoring stations since the 1990s (Fig. 4.24). At Alert (Canada), a laboratory change occurred in 2002 and during the first year of this change the atmospheric PCB data were apparently affected by the sample preparation procedures resulting in low detections of most congeners (Hung et al., 2014). The sample preparation procedures were subsequently modified as described by Su et al. (2011: section 2.3) to correct for this artifact. As a result, PCB air concentration data for 2002 are not included in this analysis. After 2002, the problem did not occur and the long-term trends were reasonably unaffected.

At Alert, Pallas (Finland) and Zeppelin (Svalbard), the decline in PCB air concentrations seems to have slowed in recent years as concentrations became lower resulting in longer half-lives. Negative $t_{1/2}$ for CB52 and CB101 at Stórhöfði (Iceland) indicates steadily increasing trends, especially after 2000 (Fig. 4.24). Since Stórhöfði is a coastal site and near Icelandic ice caps (e.g. the

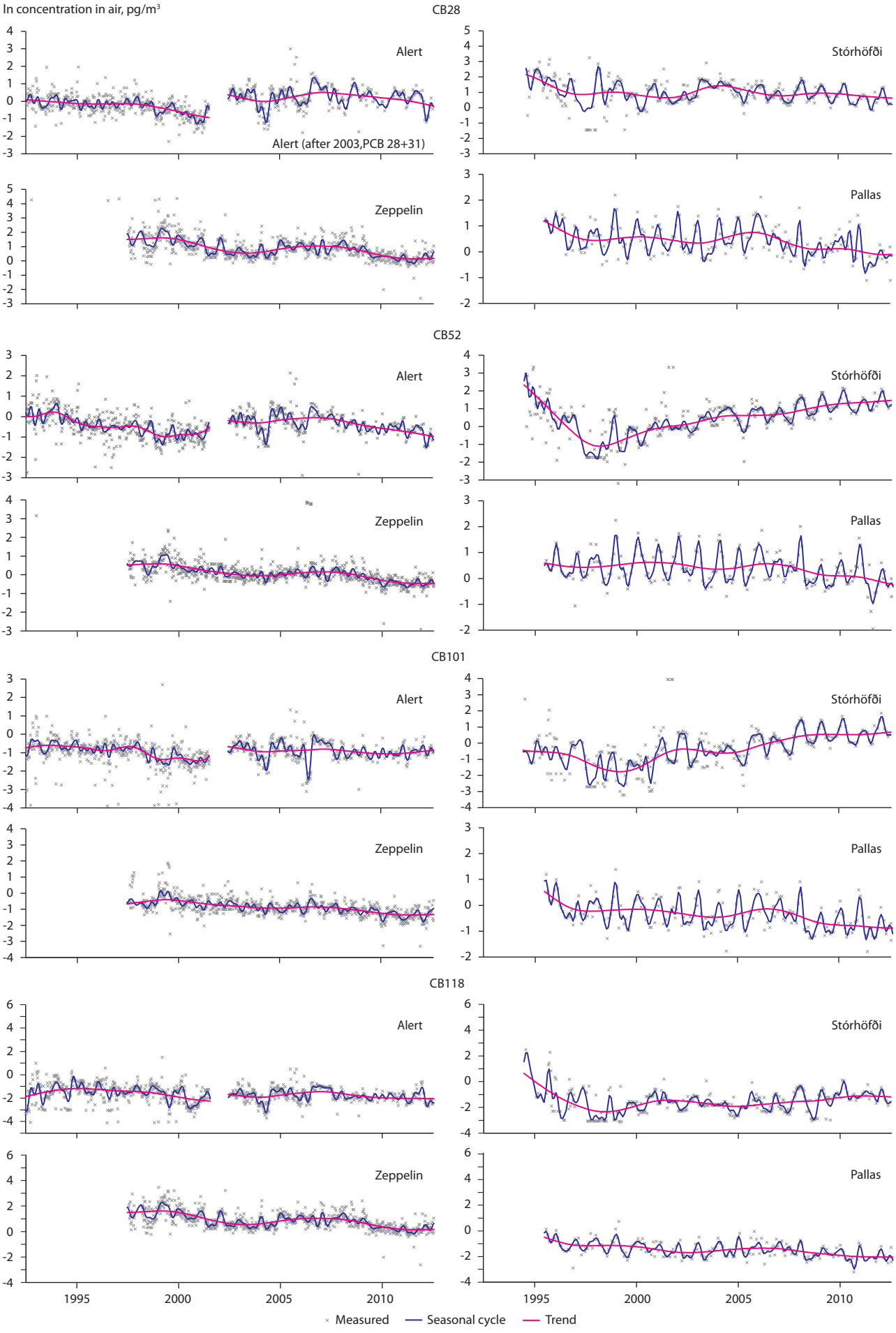
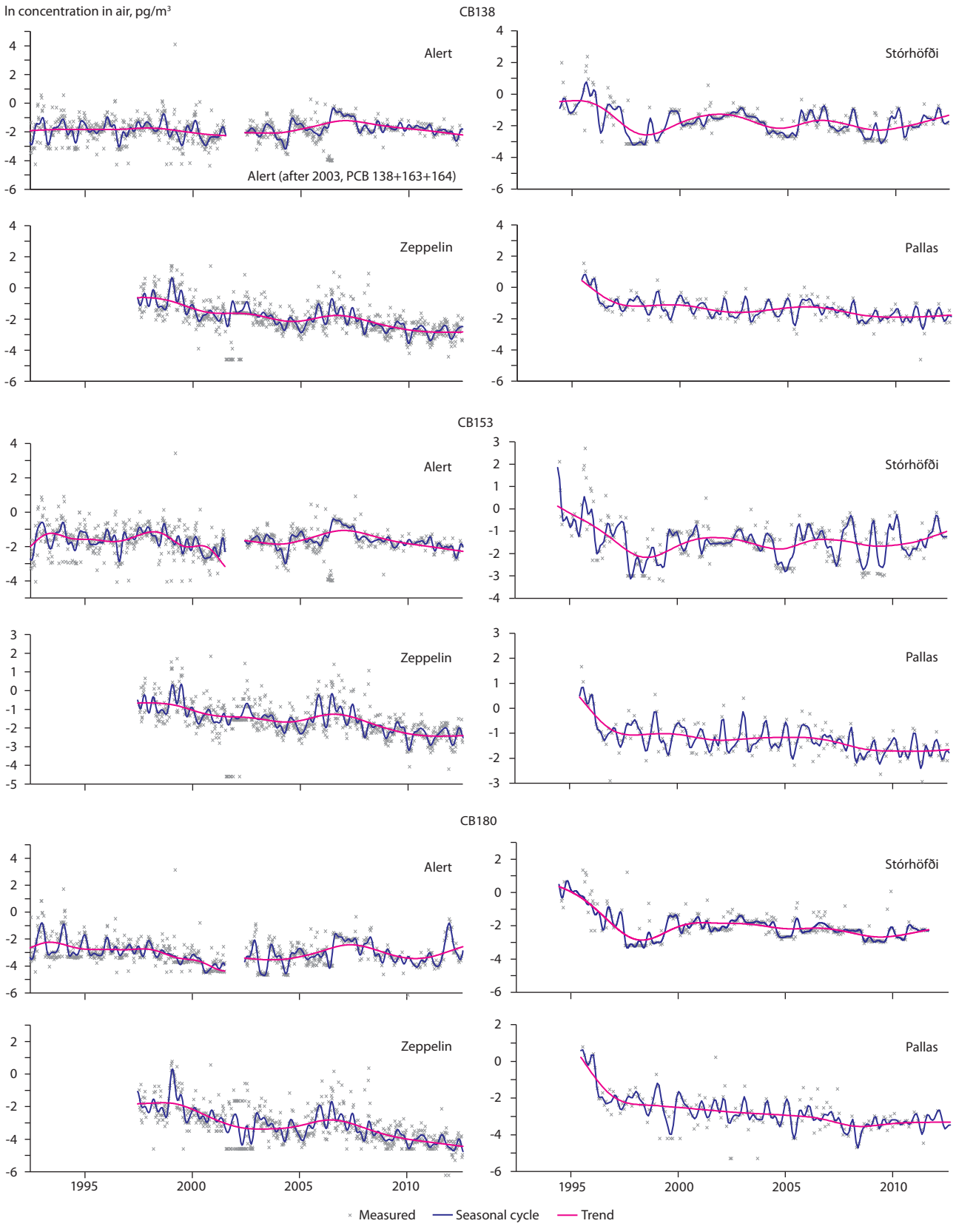



Figure 4.24 Trends in air concentration of PCB congeners CB28, CB52, CB101, CB118, CB138, CB153 and CB180 at Arctic air monitoring sites.




Mýrdalsjökull and Eyjafjallajökull ice caps), sea-ice decline around Iceland and melting of ice caps in a warming Arctic may result in re-emission of previously deposited PCBs from oceans and ice, resulting in increasing concentrations of the lighter PCBs such as CB52 and CB101. Summer maxima were apparent for PCBs measured at Stórhöfði and Pallas, but are not consistently observed at the more northerly stations of Alert and Zeppelin.

### 4.9.2 Biota trends

Time-series of $\Sigma PCB_{10}$ concentrations (CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156, CB180) starting before 2000 and time-series of individual congeners were analyzed for a total of 358 time-series (see Annex 6, Table A6.2 and Fig. 4.25). This discussion focusses on time-series for $\Sigma PCB_{10}$ and CB153. 38% of $\Sigma PCB_{10}$ time-series and 34% of CB153 time-series showed

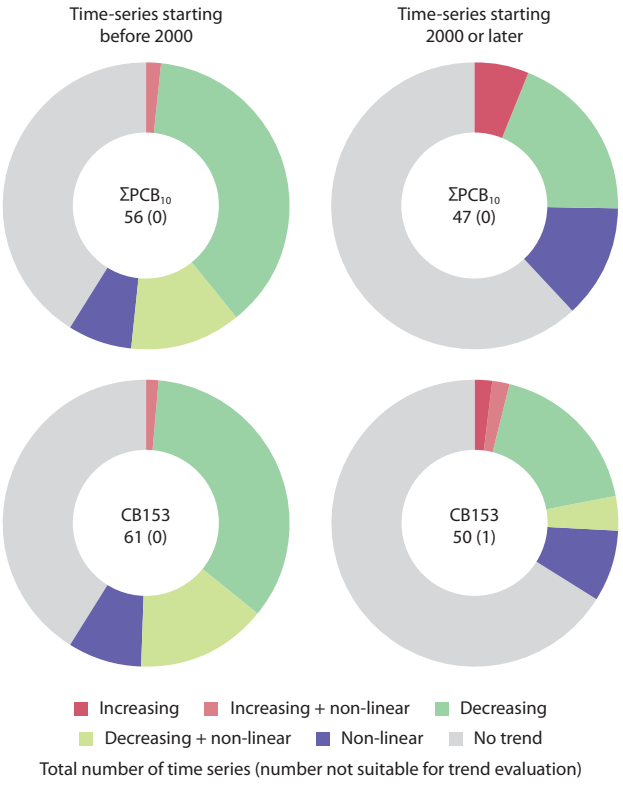



Figure 4.25 Overview of trend results for $\Sigma PCB_{10}$ and CB153 for time-series starting before and after 2000.

a significant decreasing trend, and 13% and 15%, respectively showed a significant decreasing trend with a significant non-linear trend component. One time-series (blue mussel in Mjoifjordur, Iceland) showed a significant increasing trend with a significant non-linear trend component for both $\Sigma PCB_{10}$ and CB153. The mean annual decrease was 3.7% ($t_{1/2}$ = 18) and 3.8% ($t_{1/2}$ = 19 y) for $\Sigma PCB_{10}$ and CB153, respectively.

To compare trends between congeners, 17 species-tissue-location combinations were selected as they all provide time-series of eight PCB congeners (CB28, CB52, CB101, CB105, CB118, CB138, CB153, CB180) starting before 2000, covering all species groups. No significant difference in annual trend was found between congeners (ANOVA, $p$ = 0.73).

For time-series starting after 2000, the proportions showing a significant decrease were 19% ($t_{1/2}$ = 3.6 y) and 18% ($t_{1/2}$ = 3.9 y) and the mean annual decreases were 1.5% ($t_{1/2}$ = >40 y) and 2.5% ($t_{1/2}$ = 28 y) for $\Sigma PCB_{10}$ and CB153, respectively. This indicates that the main decreases in PCB levels occurred in earlier years, and that the decrease has slowed in recent years. Two blue mussel time-series from Iceland (from Hvalstod in Hvalfjordur and Mjoifjordur) and juvenile polar bears from eastern Greenland showed significant increasing trends for $\Sigma PCB_{10}$. The polar bear time-series also showed an increasing trend for CB153, and one of the blue mussel CB153 time-series showed an increasing trend with a significant non-linear trend component.

### 4.9.3 Discussion

Both air and biota time-series showed decreasing trends. They also both showed the decline to have slowed since 2000. Polar bears from Greenland and pilot whales from the Faroe Islands have the highest recent (since 2010) annual medians of 5000–10000 ng/g lw, the highest values in adult males. In beluga from Canada and the United States, recent medians for $\Sigma PCB_{10}$ were 280–1600 ng/g lw, and in ringed seals from Canada and Greenland were 100–500 ng/g lw.

A comparison of PCB trends across the circumpolar Arctic should be treated with caution as time-series from Arctic Russia were not available, with the exception of two years of air measurement from the Valkarkai station. Furthermore, monitoring species differed in the remaining areas. Time-series of PCBs in marine mammals were restricted to Greenland, Canada, the United States and the Faroe Islands with none from Iceland and Norway, while blue mussel and marine fish time-series were restricted to Iceland and Norway. In polar bears from eastern Greenland, an unexpected increase is evident in concentrations of PCBs (and several other POPs) since 2000 (Fig. 4.26); this has been attributed to a shift in polar bear diet as a consequence of sea ice loss (McKinney et al., 2013; Rigét et al., 2016). Time-series with increasing trends of CB153 in biota were found in the region between eastern Greenland and Iceland (Fig. 4.27), most of these were associated with Icelandic blue mussel time-series, only one of which was significant. This concerned blue mussel at Mjoifjordur in Iceland, where the increasing trend has been attributed to a local source (Sturludottir et al., 2013). Sea-ice retreat and de-glaciation leading to increased re-emission from ice and oceans has also been suggested as a possible explanation for increasing air concentrations of CB52 and CB101 at Stórhöfði in Iceland (Hung et al., 2016).

PCB time-series were available for five ringed seal populations and one northern fur seal population. The length of the time-series differed, with two extending back to the early 1970s, two to the mid-1980s, and two to the mid-1990s, which is an important consideration when comparing trend results. Figure 4.28 shows the magnitude of the decreasing and increasing trends for these six seal populations. The smallest annual decrease in CB153 is found in seals in the western Arctic (northern fur seals in Alaska and ringed seal from Sachs Harbour, Canada). There is also a tendency to a smaller annual decrease in the northern (ringed seal) populations. PCB time-series were available for thirteen freshwater fish populations (Arctic char, *Salvelinus alpinus*; burbot, *Lota lota*; lake trout, *Salvelinus namaycush*; and pike). The time-series extend back to the late 1980s or mid-1990s. These time-series did not show any correlation between the annual change and latitude or longitude.

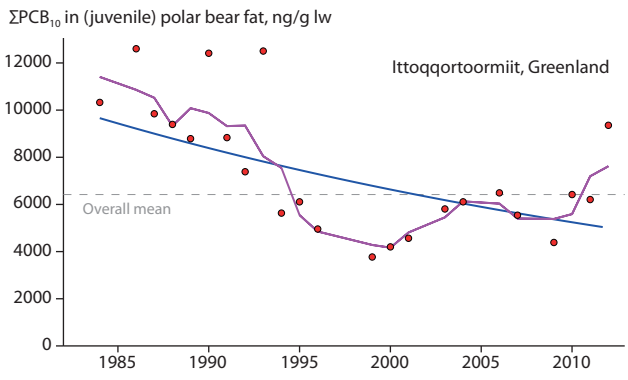



Figure 4.26 Trends in $\Sigma PCB_{10}$ in juvenile polar bears from eastern Greenland.

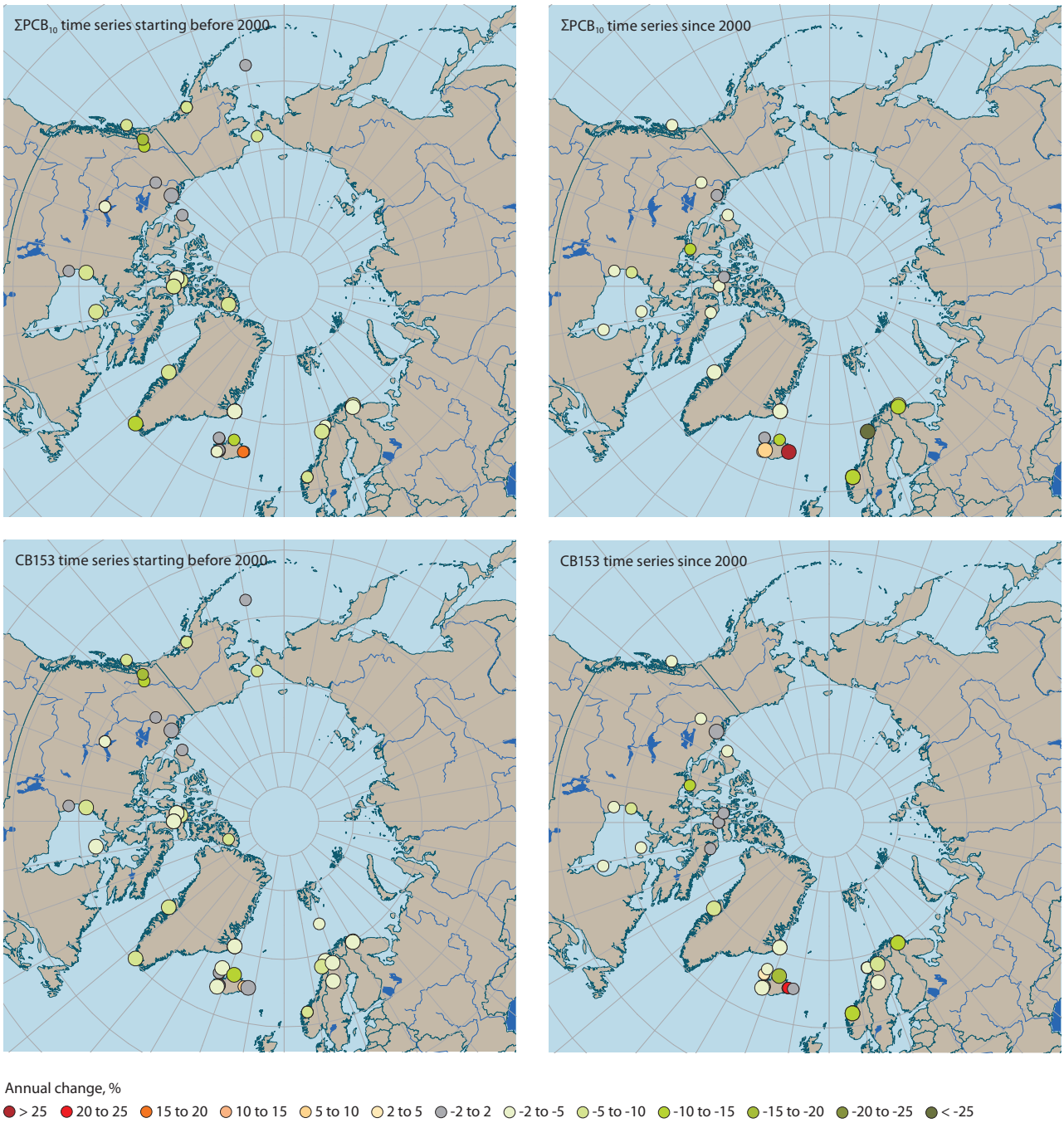



Figure 4.27 Geographical distribution of annual changes of $\Sigma PCB_{10}$ (upper) and CB153 (lower) in all time-series (left) and time-series since 2000 (right). Larger symbols represent trends that are statistically significant or where statistical power was 'adequate'.

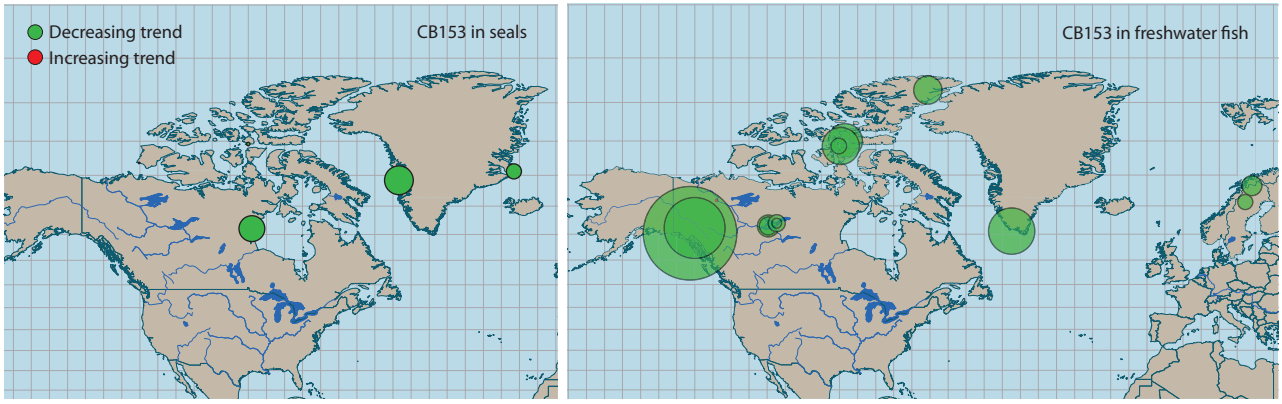



Figure 4.28 Annual decrease and annual increase in PCBs in seals and freshwater fish by latitude and longitude. The size of circle represents the magnitude of the annual change.

## 4.10 Toxaphene

### 4.10.1 Air trends

At Stórhöfði (Iceland), toxaphene Parlars 26, 50 and 62 have been measured in air since 2000. Parlar 62 was below the detection limit in all samples. Figure 4.29 shows the time trend of Parlar (26 + 50) with Parlar 26 being the dominant congener. From 2000 to 2004 air concentrations increased slightly, followed by a continuous decline after the inclusion of toxaphene under the Stockholm Convention, with $t_{1/2}$ = 5.3 y (13% per year) for the period 2000–2012. Toxaphene is not routinely monitored in air at the other AMAP stations.

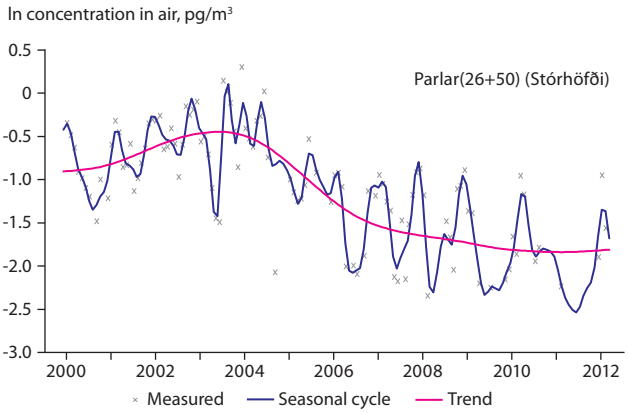



Figure 4.29 Trends in air concentration of toxaphene Parlar (26 + 50) at Stórhöfði (Iceland).

### 4.10.2 Biota trends

Seventeen toxaphene time-series starting before 2000 (Parlars 26 and 50) were available. Time-series were also available for Parlar 32 (one time-series) and Parlar 62 (two time-series). Five of the Parlar-26 and four of Parlar-50 time-series showed significant decreasing trends (two with a significant non-linear trend component), whereas three (two Parlar-26 and one Parlar-50 time-series) showed non-linear trends (Fig. 4.30). The rest showed no trends. The annual decrease of Parlar 26 was 6.0% ($t_{1/2}$ = 12 y), but was considerably lower (0.8%; $t_{1/2}$ = >80 y) for Parlar 50.

Twenty time-series of Parlar 26 starting after 2000 were available of which nine showed significant decreasing trends or decreasing trends with a significant non-linear trend component. In the case of Parlar 50, eight of 22 time-series showed significant decreasing trends. The others showed either non-linear trends or no trend. The mean annual decrease of Parlar 26 was 5.9% ($t_{1/2}$ = 12 y), while Parlar 50 had a mean annual increase of 0.8% ($t_{1/2}$ = >80 y).

### 4.10.3 Discussion

Air concentrations of toxaphene Parlar (26+50) have shown a decreasing trend since 2004 at Stórhöfði. In biota concentrations of Parlar 26 and Parlar 50 in biota also showed generally decreasing trends. The rate of decrease was greater for Parlar 26 than Parlar 50. Parlar 26 showed similar decreases in time-series starting after 2000 as those starting before 2000, while Parlar 50 switched from a slight decrease to a slight increase. Based on levels and congener composition in marine animals from Greenland, Vorkamp et al. (2015) suggested the biotransformation of nona- to octachlorinated toxaphene congeners (here represented by Parlar 50 and Parlar 26, respectively).

The highest recent (2010) annual medians of Parlar 26 and Parlar 50 were found in pilot whale blubber (about 800–1000 and 800–1400 ng/g lw, respectively). In black guillemot eggs, recent medians were about 200 and 300 ng/g lw respectively, and in ringed seal blubber from same location were 2–3 ng/g lw for both Parlar 26 and Parlar 50. For ringed seal, toxaphene levels were generally higher in seals from Canada and Alaska than from Greenland, Svalbard and the western part of Russia (Vorkamp et al., 2015). This likely reflects that the highest use of toxaphene was in the United States (Voldner and Li, 1995). The difference in mean annual change between Parlar 26 (a decrease of about 6% or $t_{1/2}$ = 12 y) and Parlar 50 (a decrease of about 1% or $t_{1/2}$ = 69 y) was mainly due to the Icelandic blue mussel time-series with a mean annual decrease in Parlar 26 of 8.5% ($t_{1/2}$ = 8.2 y) and an increase in Parlar 50 of 3.5% (a doubling of concentration in 20 years).

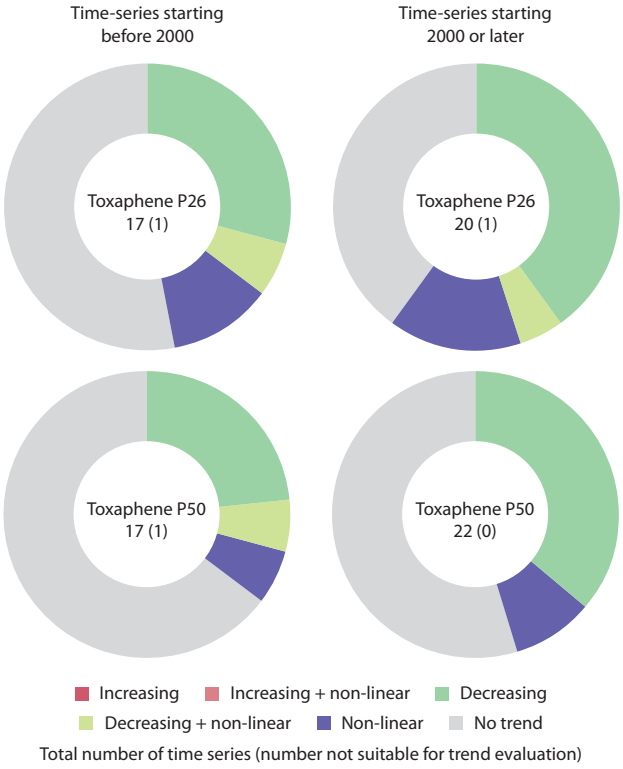



Figure 4.30 Overview of trend results for time-series of toxaphene Parlar 26 and Parlar 50 starting before and after 2000.

## 4.11 Technical endosulfan and its isomers

### 4.11.1 Air trends

Air concentrations of α-endosulfan measured at Alert (Canada; 1992–2012) and Pallas (Finland; 2009–2012) showed non-changing or slightly decreasing trends ($t_{1/2}$ = 19 y at Alert)

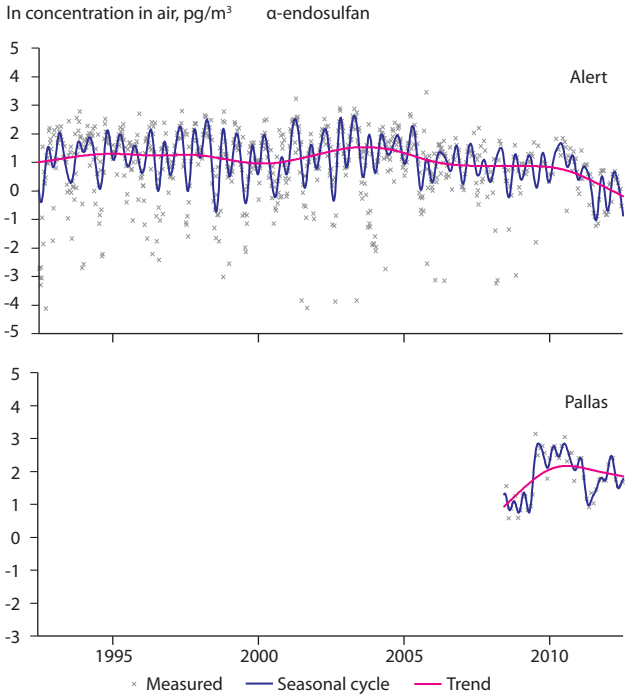



Figure 4.31 Trends in air concentration of α-endosulfan at Alert (Canada) and Pallas (Finland).

(Fig. 4.31), although a slight decrease in concentration was observed in 2012 at Alert. β-endosulfan (not shown) was mostly below the detection limit and no trend or consistent seasonality was observed. At the Villum Research Station (Greenland), it was found that α-endosulfan air concentrations did not correlate with temperature between 2008 and 2010, which indicated transport from direct sources (Bossi et al., 2013).

### 4.11.2 Biota trends

No updated time-series of endosulfan and its isomers in biota were available for this assessment.

### 4.11.3 Discussion

Temporal trends of endosulfan and its isomers were summarized in the last AMAP POPs assessment (see Weber at al., 2010). Endosulfan and its isomers were added to Annex A (compounds should be eliminated) of the Stockholm Convention in 2011.

## 4.12 Polybrominated diphenyl ethers

### 4.12.1 Air trends

Polybrominated diphenyl ethers (PBDEs) were measured in air at Alert (Canada), Zeppelin (Svalbard), Stórhöfði (Iceland) and Pallas (Finland) (Fig. 4.32). BDE47 and BDE99 were the dominant congeners in Arctic air, reflecting influence from the emission of the penta-BDE technical mixture (of which these two congeners are major components). At Stórhöfði, BDE47 was detectable in all samples while BDE99 was only detected in five samples and BDE100 was non-detectable. Concentrations of most PBDE congeners were typically unchanging in air at the Canadian site Alert with apparent summer maxima corresponding to increased volatilization during the warmer months. In contrast, air concentrations at the European Arctic sites of Pallas, Stórhöfði and Zeppelin showed significant decreasing trends with $t_{1/2}$ = 2.6–4.4 y for detectable congeners. Air concentrations at the European sites were also generally much lower than those at Alert and showed no clear seasonality. Air concentrations at Alert were probably influenced by the nearby military base (which contains articles treated with PBDEs) and the generally much higher usage of these compounds in North America compared to the rest of the world.

PBDEs were also measured in air at the Villum Research Station (Greenland) between 2008 and 2013 (Bossi et al., 2016). Concentrations were about ten times lower than those at Alert but in the same range as other remote sites in Greenland (Bossi et al., 2008; Möller et al., 2011). No trend could be identified due to high interannual variability and the short measurement period (Bossi et al., 2016). The authors noted that the air samples collected at the Villum Research Station (located in northern Greenland) contained more BDE28 (which is more volatile than BDE47 and BDE99) and less BDE47 and BDE99 than air samples collected from Nuuk (located in southern Greenland). Other than differences in volatility, the PBDE composition may also be influenced by the fact that BDE99 is more susceptible to photolysis than BDE47 and BDE100 (Bossi et al., 2016).

### 4.12.2 Biota trends

Time-series of several PBDE congeners in biota starting before 2000 were available (BDE47, BDE49, BDE99, BDE100, BDE153, BDE154 and BDE155), however most concerned BDE47 (26 time-series) and BDE99 (19 time-series) and so these are the ones discussed here. For BDE47, nine time-series showed a significant increasing trend or a significant increasing trend with a significant non-linear trend component; for BDE99, this was the case for two time-series. One time-series (adult ringed seals from eastern Greenland) showed decreasing trends for both BDE47 and BDE99. The non-linear trend component was significant in eight BDE47 time-series and the typical pattern was an increase followed by a decrease in recent years (Figs. 4.33 and 5.2).

For time-series starting after 2000, no BDE47 time-series showed increasing trends and an increase was found in only one case for BDE99 (ringed seal from Canada). Decreasing trends were found for four BDE47 time-series and three BDE99 time-series. This indicates that the concentrations of these congeners in biota have peaked and are now mainly decreasing.

### 4.12.3 Discussion

Decreasing trends in air for BDE47 and BDE99 were observed at the European Arctic stations of Pallas, Stórhöfði and Zeppelin. No trends were observed in air at the Canadian Arctic site of Alert, which may be attributed to influence from the nearby military base and the generally higher use of these chemicals in North America.

Mean annual medians (since 2010) of BDE47 were highest in pilot whales from the Faroe Islands (180 ng/g lw), followed

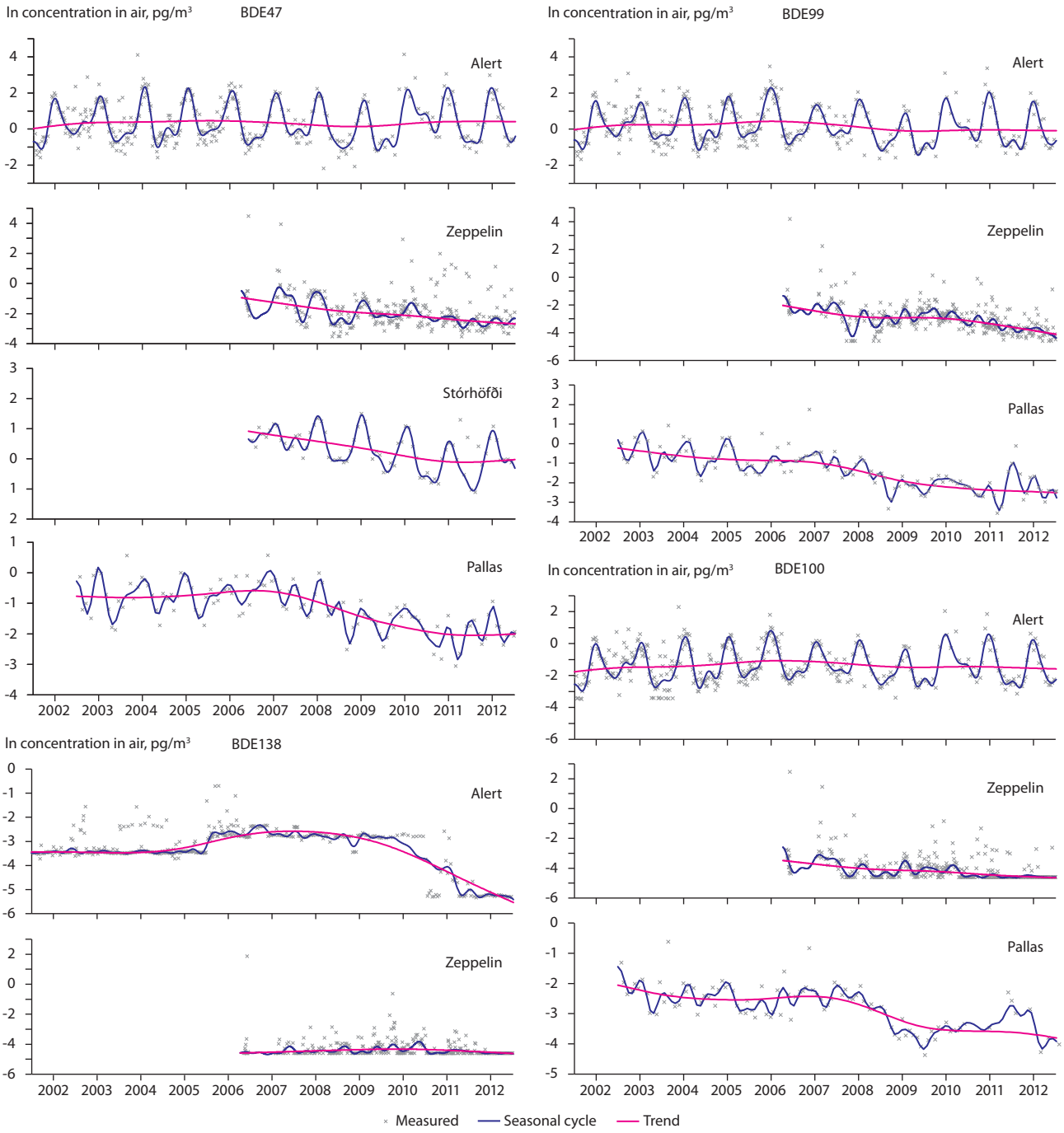



Figure 4.32 Trends in air concentration of PBDEs at Arctic air monitoring sites.

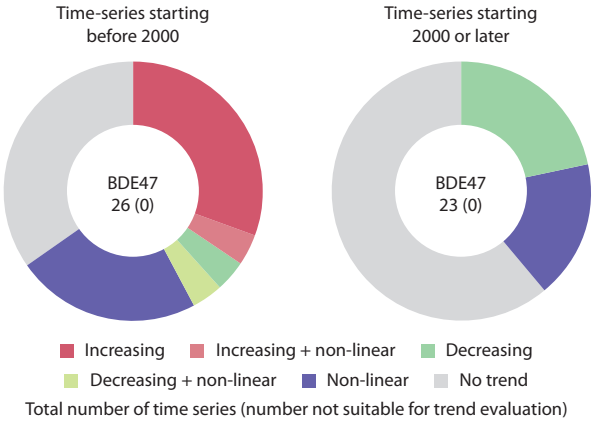



Figure 4.33 Overview of trend results for BDE47 time-series starting before and after 2000.

by polar bears from eastern Greenland (25–35 ng/g lw) and beluga from Alaska (15 ng/g lw). Levels of BDE99 were lower, with recent annual medians of 35 ng/g lw in pilot whale and 1–3 ng/g lw in polar bears.

A parabolic trend, with an increase up to around 2000 followed by a decrease is found in several biota time-series. Similar trend patterns have also been observed in Leach's storm-petrel (*Oceanodroma leucorhoa*) from the Pacific coast of Canada (Miller et al., 2014). That a similar pattern is not found in the air time-series is not inconsistent given that these time-series only extend back to around 2000. In the period since 2000, air time-series show more constant or decreasing trends, which is also observed in a number of biota time-series.

## 4.13 Hexabromocyclododecane

### 4.13.1 Air trends

For air concentrations of hexabromocyclododecane (HBCDD) measured at Zeppelin (Svalbard) (Fig. 4.34), α-HBCDD and γ-HBCDD showed decreasing trends with $t_{1/2}$ = 2.9 y (24% per year) and $t_{1/2}$ = 1.5 y (46% per year), respectively. Total HBCDD air concentrations were reported at Alert (2002–2012), but concentrations were very low and no apparent increase or decrease in trend was observed (Fig. 4.35).

### 4.13.2 Biota trends

Seven time-series of α-HBCDD in marine mammals and seabird eggs were available (Fig. 4.36). Of these, five showed significant increasing trends. Two time-series showed a significant non-linear trend or no trend. The mean annual increase of the seven time-series was 7.6% ($t_{1/2}$ = 9.2 y).

In contrast, time-series starting after 2000 showed decreasing trends. The mean annual decrease of the three post-2000 time-series was 3.6% ($t_{1/2}$ = 19.2 y). One time-series starting after 2000 (thick-billed murre from Canada) showed a significant decreasing trend, while two time-series showed no trend.

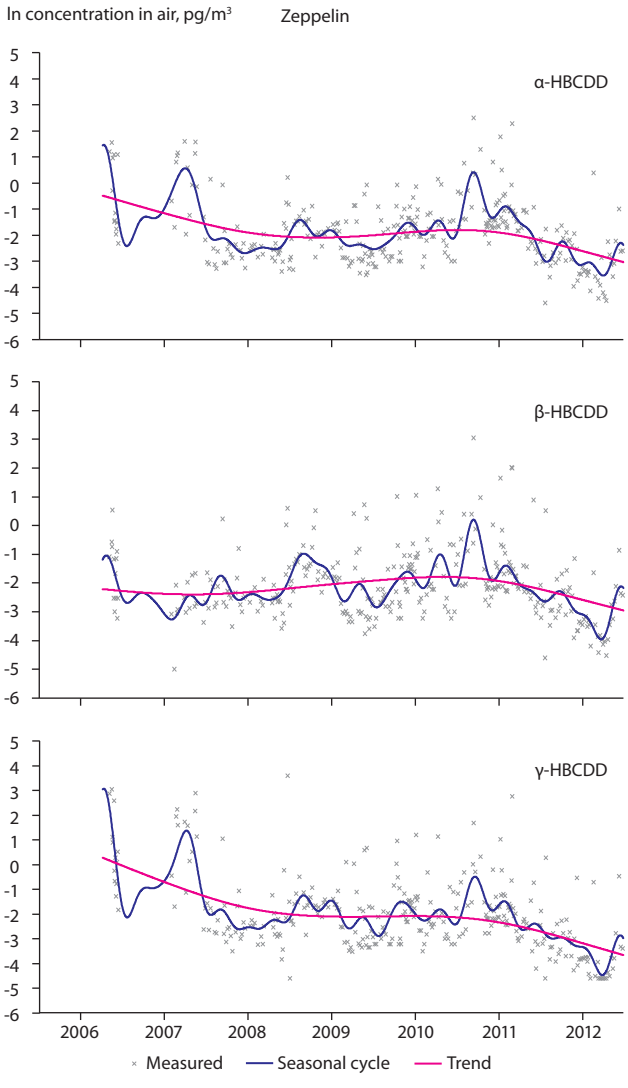



Figure 4.34 Trends in air concentration of HBCDDs at Zeppelin (Svalbard).

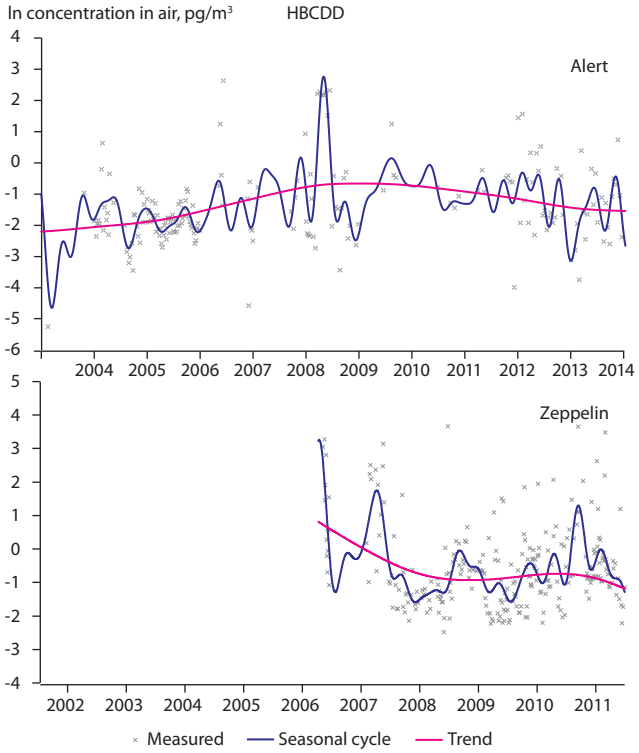



Figure 4.35 Trends in air concentration of total HBCDD at Alert (Canada) and Zeppelin (Svalbard).

### 4.13.3 Discussion

While α-HBCDD in air at Zeppelin showed decreasing trends with a relatively low $t_{1/2}$ of 2.9 (corresponding to an annual decrease of 24%), the majority of biota time-series showed increasing trends. However, the air time-series at Zeppelin only extends back to mid-2006, while the biota time-series in several cases extend back to the mid-1980s. Increasing trends of α-HBCDD have also been found in beluga from Cook Island and the eastern Chukchi Sea (Alaska) covering the period from around 1990 to 2000 and 2005 (Hoguet et al., 2013).

The mean annual medians for the period 2000–2012 were highest in polar bears (25–40 ng/g lw). Levels were lower on seabird eggs (4–7 ng/g lw).

HBCDD was added to Annex A (compounds should be eliminated) to the Stockholm Convention in 2013.

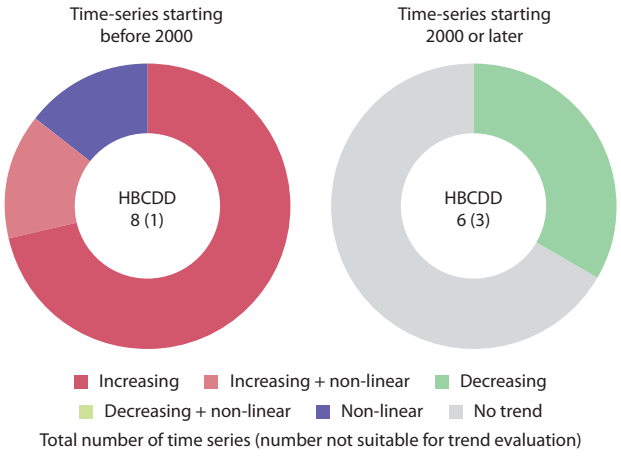



Figure 4.36 Overview of trend results for HBCDD time-series starting before and after 2000.

## 4.14 Perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride

### 4.14.1 Air trends

Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) in airborne particles were measured at Zeppelin from mid-2006 to 2012. The particle phase concentrations for both compounds appear constant throughout the monitoring period and consistent seasonality was not observed (Fig. 4.37). At Alert (Canada), neutral per-and polyfluoroalkyl substances (PFASs) were monitored in air (gas + particle) from August 2006 to 2012 (Fig. 4.38). The 8:2 fluorotelomer alcohol (FTOH) was the dominant compound detected in all air samples at Alert. This is consistent with previous cruise-based measurements performed across the North Atlantic and Canadian Archipelago in 2005 (Shoeib et al., 2006), in the Atlantic Ocean/ Southern Ocean/ Baltic Sea in 2007 and 2008 (Dreyer et al., 2009), as well as globally under the GAPS study at land-based stations (Gawor et al., 2014). Piekarz et al. (2007) calculated the estimated atmospheric residence times for 6:2 FTOH (50 days), 8:2 FTOH (80 days) and 10:2 FTOH (70 days). The relative order of these residence times coincides with their atmospheric concentrations which may explain the relative enhancement of 8:2 FTOH in ambient air. Spring maxima were observed at Alert, particularly for methyl perfluorooctane sulfonamido ethanol (MeFOSE), which may be associated with the increase in particulate input during the Arctic Haze season. Summer maxima were also apparent for FTOH and MeFOSE which may be related to revolatilization due to higher temperatures. PFOS precursors MeFOSE and ethyl perfluorooctane sulfonamide ethanol (EtFOSE) showed non-changing and decreasing trends ($t_{1/2}$ = 1.3 y; 53% per year), respectively (Fig. 4.38); reflecting the voluntary phase-out of the production of PFOS, PFOA, and PFOS-related products by their largest producer 3M in 2000. In contrast, the perfluorinated carboxylic acid (PFCA)

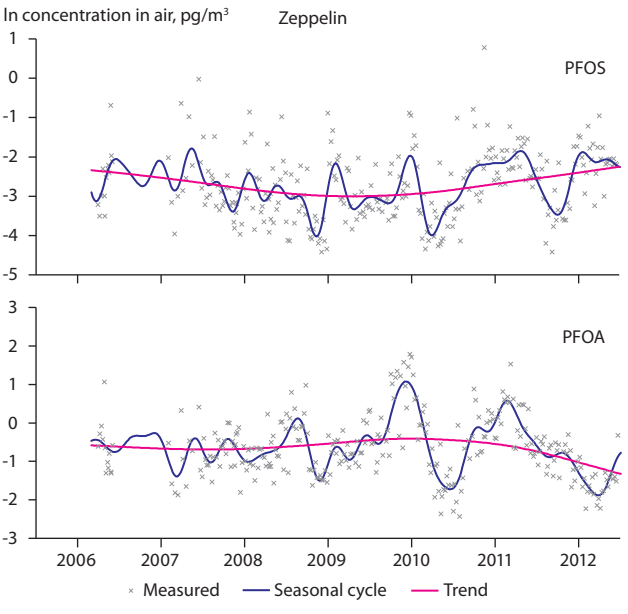



Figure 4.37 Trends in air concentration of PFOS and PFOA (particle phase only) at Zeppelin (Svalbard).

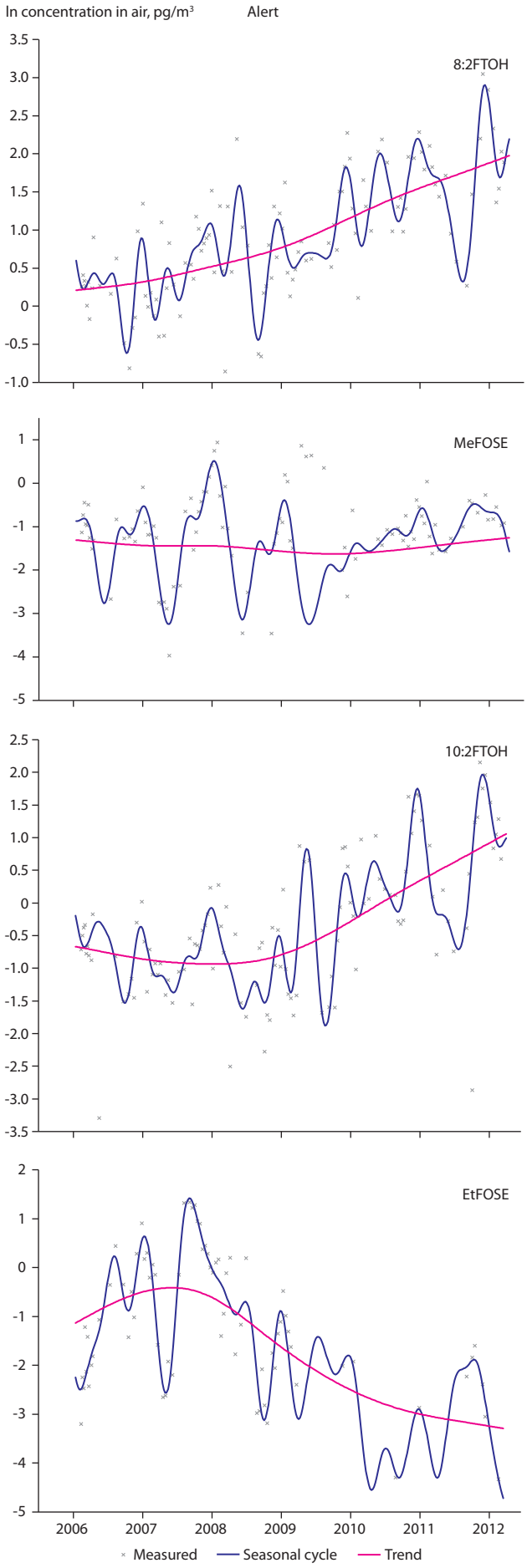



Figure 4.38 Trends in air concentration of 8:2 and 10:2 FTOH and MeFOSE and EtFOSE at Alert (Canada).

precursors 6:2 FTOH, 8:2 FTOH and 10:2 FTOH, which were not regulated at the time of measurement, showed increasing tendencies in air at Alert with doubling times of 2.3–3.3 y (increasing by 21–30% per year).

### 4.14.2 Biota trends

Time-series of the perfluorosulfonates (PFHxS, PFOS), the perfluorocarboxylates (PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTrA, PFTA) and the perfluorosulfonamide (PFOSA) were available; including PFOS time-series from Alaska, Canada, Greenland, the Faroe Islands and Sweden. Most time-series concerned marine mammals, with a few for freshwater fish and seabird eggs. Only PFOS, PFNA, PFDA and PFUnA are discussed here because more than ten time-series for each were available. For PFOS, eight of 16 time-series showed a significant non-linear trend and one (juvenile polar bears from eastern Greenland) showed an increasing trend with a significant non-linear trend component. The others showed no trend. A common trend pattern was an increase until about the mid-2000s followed by a decrease, which was caught by the non-linear trend component (see Fig.4.39). For PFNA (6 of 11), PFDA (4 of 10) and PFUnA (6 of 11), several times-series showed significant increasing trends or significant increasing trends with a significant non-linear trend component (Fig. 4.40).

Eleven PFOS time-series starting after 2000 were available, all having significant non-linear trend components or showing no trend. For PFNA time-series, two of five showed significant increasing trends, the rest showed a significant non-linear component or no trend. All five time-series of PFDA and PFUnA showed a significant non-linear component or no trend.

### 4.14.3 Discussion

No trends were observed in PFOS and PFOA in airborne particles at Zeppelin, while the PFOS precursors MeFOSE and EtFOSE showed no trend and a decreasing trend at Alert, respectively; reflecting the phase-out of PFOS-related production. A decrease in levels in response to this phase-out is also supported by trends in biota. Several of the PFOS time-series in biota extend further back (to the early- or mid-1980s) than those for air, and many show an increase followed by a decrease with a significant peak around the mid-2000s. For northern fur seals from Alaska, however, perfluorinated alkyl acids (PFAAs) continued to increase over the period 1987 to 2007 (Kucklick et al., 2013). Trends in perfluorocarboxylates (PFNA, PFDA, PFUnA) differ from those for PFOS, with a relatively high proportion showing an increasing trend or an increasing trend with a non-linear trend component.

By far the highest levels of PFOS were found in polar bear liver from eastern Greenland. In the mid-2000s, when PFOS in these polar bears peaked, the annual median was almost 3060 ng/g ww. In recent years, the annual median has dropped to about 950 ng/g ww. Levels of PFNA and PFDA were also highest in polar bears, with mean recent (since 2010) annual medians of 130–170 and 45 ng/g ww, respectively. Recent levels of PFUnA in polar bears were 110–160 ng/g ww.

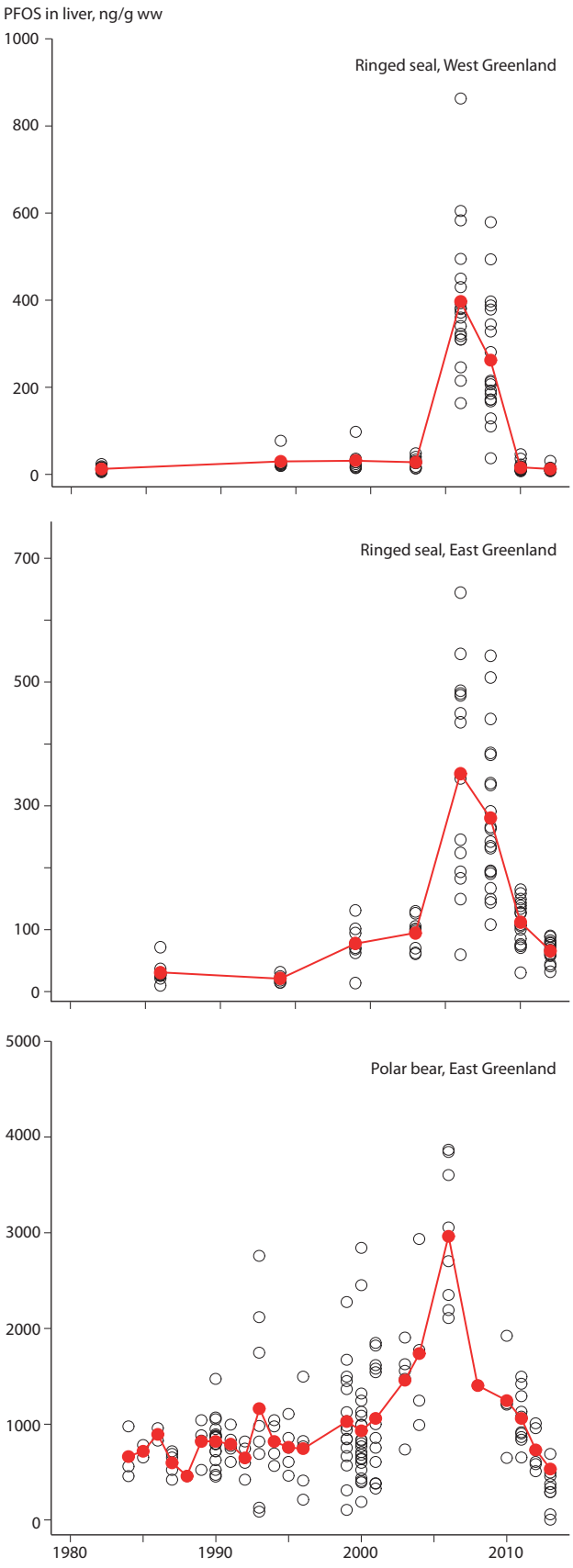



Figure 4.39 Trends of PFOS concentration in ringed seals from western and eastern Greenland and polar bears from eastern Greenland (updated from Rigét et al., 2013b).

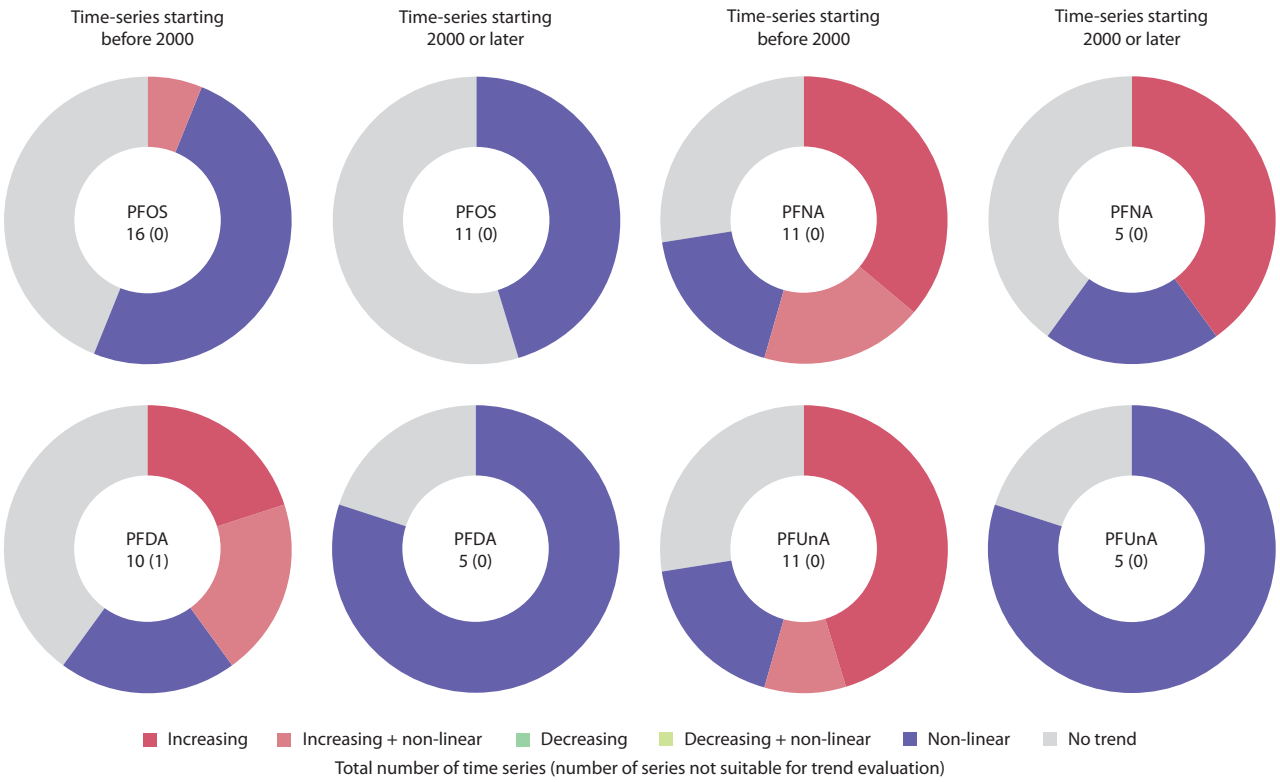



Figure 4.40 Overview of trend results for selected perfluorosulfonate and perfluorocarboxylate time-series starting before and after 2000.

## 4.15 Octachlorostyrene

### 4.15.1 Air trends

Octachlorostyrene is measured in air at Alert and its concentration peaked in the mid-1990s (up to a maximum of 2.96 pg/m$^3$ in 1994) then a decline is observed up to 1997. The concentrations remained more or less the same between 1997 and 2012. Recent levels are quite low (0.01–0.64 pg/m$^3$ in 2012).

### 4.15.2 Biota trends

Eleven octachlorostyrene time-series starting before 2000 were available. Several blue mussel and marine fish time-series from Norway could not be evaluated owing to values below detection limits. Four time-series showed significant decreasing trends. The rest showed no trend or had a significant non-linear trend component. The mean annual decrease was 4.5% ($t_{1/2}$ = 15.4 y). All eleven time-series starting after 2000 showed no trend or had a significant non-linear trend component.

### 4.15.3 Discussion

Levels of octachlorostyrene in air and biota show generally decreasing trends. Biota concentrations are low, with the highest recent (since 2010) annual medians found in polar bears (12–18 ng/g lw), beluga from Canada (6 ng/g lw) and seabird eggs from Canada (<0.1 ng/g lw).

## 4.16 POPs in air at satellite stations

Air measurements of POPs were also conducted at two satellite stations (i.e. air monitoring stations that do not yet have sufficiently long time-series for trend determination) during this reporting period. These sites include the Little Fox Lake station in the Yukon (Canada) and Valkarkai in Russia. Although trends are not available for these sites, the concentration data are useful for comparing with concentrations observed at other sites. Annual mean, median, standard deviation and range in total air concentrations determined at the Little Fox Lake and Valkarkai sites are given in Tables 4.2 and 4.3.

Table 4.2 Arithmetic mean (median ± standard deviation; range, n) of total air concentrations (GFF + PUF) in pg/m$^3$ at Little Fox Lake, Yukon.

| POP | 2007 (mid-August – December) | 2008 | 2009 (January – mid-October) |
|---|---|---|---|
| Aldrin | 0.12 (0.12±0.012; 0.11–0.15, 18) | 0.15 (0.12±0.11; 0.058–0.83, 49) | 0.14 (0.12±0.061; 0.10–0.43, 29) |
| Dieldrin | 0.19 (0.18±0.098; 0.061–0.40, 18) | 0.22 (0.24±0.13; 0.049–0.50, 49) | 0.18 (0.13±0.12; 0.060–0.39, 29) |
| Endrin | 0.11 (0.11±0.011; 0.095–0.14, 18) | 0.17 (0.11±0.21; 0.052–1.4, 49) | 0.17 (0.13±0.10; 0.094–0.50, 29) |
| *cis*-Chlordane | 0.20 (0.20±0.11; 0.054–0.46, 18) | 0.26 (0.13±0.25; 0.058–0.93, 49) | 0.23 (0.21±0.17; 0.053–0.57, 29) |
| *trans*-Chlordane | 0.12 (0.10±0.053; 0.057–0.21, 18) | 0.11 (0.069±0.097; 0.031–0.52, 49) | 0.11 (0.070±0.080; 0.057–0.35, 29) |
| *cis*-Nonachlor | 0.031 (0.030±0.003; 0.026–0.038, 18) | 0.031 (0.030±0.006; 0.014–0.054, 49) | 0.072 (0.031±0.079; 0.026–0.36, 29) |
| *trans*-Nonachlor | 0.20 (0.18±0.069; 0.13–0.37, 18) | 0.19 (0.15±0.16; 0.040–0.68, 49) | 0.11 (0.043±0.11; 0.036–0.42, 29) |
| Oxychlordane | 0.10 (0.090±0.028; 0.078–0.18, 18) | 0.17 (0.095±0.14; 0.060–0.57, 49) | 0.16 (0.12±0.10; 0.077–0.44, 29) |
| *o,p*'-DDD | 0.12 (0.12±0.011; 0.10–0.15, 18) | 0.12 (0.12±0.023; 0.055–0.20, 49) | 0.17 (0.12±0.17; 0.10–1.0, 29) |
| *o,p*'-DDE | 0.12 (0.12±0.027; 0.060–0.16, 18) | 0.088 (0.061±0.049; 0.027–0.27, 49) | 0.088 (0.067±0.047; 0.050–0.21, 29) |
| *o.p*'-DDT | 0.19 (0.17±0.060; 0.13–0.34, 18) | 0.22 (0.16±0.11; 0.14–0.60, 49) | 0.19 (0.15±0.083; 0.13–0.53, 29) |
| *p,p*'-DDD | 0.46 (0.45±0.044; 0.39–0.56, 18) | 0.49 (0.45±0.20; 0.21–1.8, 49) | 0.53 (0.46±0.22; 0.38–1.6, 29) |
| *p,p*'-DDE | 0.61 (0.60±0.16; 0.41–0.96, 18) | 0.45 (0.45±0.14; 0.24–0.78, 49) | 0.38 (0.34±0.21; 0.21–1.2, 29) |
| *p,p*'-DDT | 0.29 (0.26±0.10; 0.23–0.69, 18) | 0.27 (0.26±0.053; 0.12–0.46, 49) | 0.33 (0.27±0.16; 0.22–0.92, 29) |
| Heptachlor | 0.17 (0.16±0.016; 0.14–0.21, 18) | 0.17 (0.17±0.030; 0.078–0.29, 49) | 0.19 (0.17±0.082; 0.14–0.58, 29) |
| Heptachlor epoxide | 0.31 (0.26±0.17; 0.12–0.62, 18) | 0.39 (0.23±0.63; 0.034–3.7, 49) | 0.30 (0.11±0.37; 0.029–1.4, 29) |
| Methoxychlor | 1.6 (1.6±0.16; 1.4–2.0, 18) | 1.9 (1.6±1.7; 0.75–13, 49) | 1.9 (1.6±0.79; 1.4–5.6, 29) |
| Hexachlorobenzene[a] | 26 (28±8.1; 9.3–37, 18) | 28 (25±12; 2.5–51, 49) | 22 (22±9.4; 2.9–41, 29) |
| α-HCH | 5.2 (5.0±4.0; 0.88–14, 18) | 9.1 (9.4±6.4; 0.042–22, 49) | 6.6 (5.7±5.3; 0.34–17, 29) |
| β-HCH | 0.069 (0.068±0.007; 0.059–0.085, 18) | 0.50 (0.070±0.99; 0.032–4.1, 49) | 0.080 (0.069±0.034; 0.058–0.24, 29) |
| γ-HCH | 0.58 (0.59±0.37; 0.099–1.2, 18) | 0.79 (0.33±1.3; 0.028–6.9, 49) | 0.25 (0.043±0.44; 0.026–2.0, 29) |
| Mirex | 0.29 (0.28±0.028; 0.24–0.35, 18) | 0.29 (0.29±0.052; 0.13–0.49, 49) | 0.33 (0.29±0.14; 0.24–1.0, 29) |
| α-Endosulfan | 2.0 (2.1±1.3; 0.23–4.1, 18) | 1.8 (0.60±2.2; 0.021–10, 49) | 1.1 (0.25±1.5; 0.038–7.0, 29) |
| β-Endosulfan | 0.18 (0.17±0.022; 0.15–0.23, 18) | 0.18 (0.18±0.030; 0.12–0.31, 49) | 0.21 (0.18±0.088; 0.15–0.62, 29) |
| BDE47 | 5.9 (1.9±10; 0.72–42, 18) | 1.9 (1.5±1.5; 0.16–6.7, 49) | 2.1 (1.4±1.8; 0.45–7.4, 29) |
| BDE49 | 0.26 (0.11±0.33; 0.11–1.5, 18) | 0.16 (0.11±0.11; 0.052–0.56, 49) | 0.19 (0.15±0.12; 0.094–0.66, 29) |
| BDE66 | 0.14 (0.063±0.16; 0.054–0.71, 18) | 0.079 (0.058±0.043; 0.026–0.22, 49) | 0.12 (0.093±0.079; 0.053–0.41, 29) |
| BDE85 | 0.20 (0.12±0.18; 0.10–0.83, 18) | 0.16 (0.12±0.066; 0.057–0.41, 49) | 0.18 (0.13±0.10; 0.10–0.50, 29) |
| BDE99 | 3.8 (2.0±5.4; 0.69–23, 18) | 2.0 (1.5±1.7; 0.14–7.2, 49) | 2.6 (1.5±2.4; 0.46–8.8, 29) |
| BDE100 | 1.2 (0.45±2.1; 0.15–8.7, 18) | 0.44 (0.32±0.38; 0.057–1.8, 49) | 0.57 (0.37±0.50; 0.099–1.8, 29) |
| BDE153 | 0.21 (0.15±0.16; 0.054–0.64, 18) | 0.15 (0.13±0.094; 0.050–0.42, 49) | 0.19 (0.11±0.20; 0.053–0.96, 29) |
| BDE154 | 0.19 (0.13±0.18; 0.056–0.78, 18) | 0.14 (0.10±0.10; 0.035–0.52, 49) | 0.18 (0.12±0.18; 0.033–0.75, 29) |
| BDE183 | 0.063 (0.061±0.026; 0.042–0.15, 18) | 0.070 (0.049±0.11; 0.028–0.80, 49) | 0.087 (0.049±0.14; 0.041–0.82, 29) |

[a]HCB broke through in about 30% of samples. Concentrations may therefore be underestimated.

Table 4.3 Arithmetic mean (median ± standard deviation; range, n) of total air concentrations (GFF + PUF) in pg/m$^3$ at Valkarkai, Russia.

| POP | 2008 (April – October) | 2009 (mid-January – mid-November) |
|---|---|---|
| Dieldrin | 0.27 (0.28±0.013; 0.24–0.29, 17) | 0.29 (0.27±0.062; 0.19–0.49, 24) |
| Endrin | 0.42 (0.43±0.021; 0.38–0.45, 17) | 0.45 (0.43±0.098; 0.29–0.77, 24) |
| *cis*-Chlordane | 0.25 (0.24±0.027; 0.21–0.32, 17) | 0.26 (0.24±0.055; 0.16–0.44, 24) |
| *trans*-Chlordane | 0.25 (0.26±0.013; 0.23–0.27, 17) | 0.27 (0.26±0.059; 0.18–0.46, 24) |
| *cis*-Nonachlor | 0.12 (0.12±0.006; 0.10–0.12, 17) | 0.13 (0.12±0.031; 0.087–0.21, 14) |
| *trans*-Nonachlor | 0.17 (0.17±0.024; 0.15–0.23, 17) | 0.22 (0.17±0.13; 0.11–0.74, 24) |
| Oxychlordane | 0.35 (0.35±0.035; 0.31–0.47, 17) | 0.40 (0.35±0.11; 0.26–0.78, 24) |
| *o,p*'-DDD | 0.45 (0.46±0.022; 0.40–0.47, 17) | 0.62 (0.45±0.49; 0.34–2.6, 24) |
| *o,p*'-DDE | 0.27 (0.23±0.097; 0.20–0.51, 17) | 0.83 (0.31±0.85; 0.17–3.2, 24) |
| *o.p*'-DDT | 0.58 (0.59±0.029; 0.51–0.61, 17) | 0.78 (0.59±0.32; 0.43–1.7, 24) |
| *p,p*'-DDD | 1.7 (1.8±0.086; 1.5–1.8, 17) | 1.8 (1.7±0.40; 1.2–3.2, 24) |
| *p,p*'-DDE | 1.3 (1.2±0.31; 0.91–1.8, 17) | 1.6 (0.98±1.2; 0.72–5.2, 24) |
| *p,p*'-DDT | 1.1 (1.0±0.17; 0.89–1.5, 17) | 1.3 (1.0±0.43; 0.75–2.6, 24) |
| Heptachlor | 0.63 (0.65±0.032; 0.56–0.67, 17) | 0.68 (0.63±0.15; 0.44–1.2, 24) |
| Heptachlor epoxide | 0.14 (0.13±0.043; 0.12–0.31, 17) | 0.14 (0.13±0.030; 0.090–0.24, 24) |
| Methoxychlor | | 6.9 (6.3±1.6; 4.2–11, 14) |
| α-HCH | 11 (8.2±9.4; 0.15–32, 17) | 4.7 (2.3±6.4; 0.15–22, 24) |
| β-HCH | 0.31 (0.27±0.097; 0.23–0.57, 17) | 0.60 (0.31±0.96; 0.18–4.4, 24) |
| γ-HCH | 0.40 (0.30±0.31; 0.10–1.0, 17) | 1.1 (0.26±2.8; 0.080–13, 24) |
| Mirex | 1.1 (1.1±0.054; 0.96–1.2, 17) | 1.2 (1.1±0.25; 0.75–2.0, 24) |
| CB28 + CB31 | 3.9 (2.6±3.6; 0.89–14, 17) | 1.8 (1.4±1.6; 0.41–8.0, 24) |
| CB52 | 4.8 (4.2±3.0; 1.3–12, 17) | 5.1 (2.7±4.8; 0.87–17, 24) |
| CB101 | 5.6 (4.1±4.3; 0.83–15, 17) | 4.1 (2.3±4.3; 0.97–19, 24) |
| CB105 | 3.8 (3.1±2.8; 0.076–11, 17) | 3.0 (1.9±2.7; 0.59–8.8, 24) |
| CB118 | 7.6 (5.4±4.8; 1.5–15, 17) | 7.4 (3.5±7.9; 1.1–31, 24) |
| CB138[a] | 2.0 (1.6±1.2; 0.52–4.7, 17) | 2.4 (1.2±2.4; 0.077–8.0, 24) |
| CB153 | 2.4 (1.8±1.4; 0.61–4.8, 17) | 2.3 (1.6±2.3; 0.55–7.5, 24) |
| CB156 | 0.16 (0.079±0.26; 0.068–1.1, 17) | 0.40 (0.13±0.46; 0.058–1.5, 24) |
| CB180 | 0.15 (0.079±0.15; 0.072–0.54, 17) | 0.28 (0.13±0.29; 0.058–0.85, 24) |
| α-Endosulfan | 0.17 (0.17±0.008; 0.15–0.18, 17) | 0.18 (0.17±0.039; 0.12–0.31, 24) |

[a]CB138 co-elutes with CB158.

# 5. Summary of trend results

Overview tables presenting trend results for POPs in air and biota are included in Annex 6. The air trend results in Table A6.1 are presented as the effective half-life of the chemicals in air. For biota, the tables show the annual percentage change (average and range) by species for the different contaminants/ contaminant groups. Table A6.2 summarizes the biota trend results for all runs (i.e. for time-series starting before 2000) while Table A6.3 summarizes the trend results for time-series starting in or after 2000. Table A6.4 summarizes the trend results for those time-series with either significant (log-) linear trends or for those time-series that are considered of 'adequate' power (i.e. the ratio of the number of years in the time-series to the number of years required to detect a 5% annual change with a power of 80% is >1), for all runs. Table A6.5 presents results for runs with significant trends or adequate time-series for the time series starting in or after 2000.

Of the more than 1100 statistical runs considered on biota time-series, about 25% were excluded, mainly due to the presence of a high proportion of 'less than' values. Of the non-excluded runs, about 12% of time-series are currently of adequate length to detect a 5% annual change with a statistical power of 80% for the current number of years; this highlights the need to maintain monitoring effort to obtain sufficiently powerful time-series for trend detection.

The results tabulated in Table A6.4 and summarised in Fig. 5.1 show the trends for the different chemical groups ranked from decreasing to increasing based on the mean trend for all biota time series. Also shown are the mean trends if all time series (not just those that are significant or 'adequate') are considered. The figure illustrates that the greatest annual declines (typically 5–10% per year) are observed for 'legacy' organochlorine pesticides, including toxaphenes, HCHs and DDTs. Chlordanes and industrial chemicals such as PCBs and HCB have also declined, but at slightly lower rates, and newer POPs including BDEs, PFOS/PFOA and HBCDD still exhibit increasing trends in many cases (although fewer time-series are currently available for these chemicals).

Figure 5.2 compares the (average) trends observed in air and biota time-series considered in this assessment (for results for all runs starting before 2000).

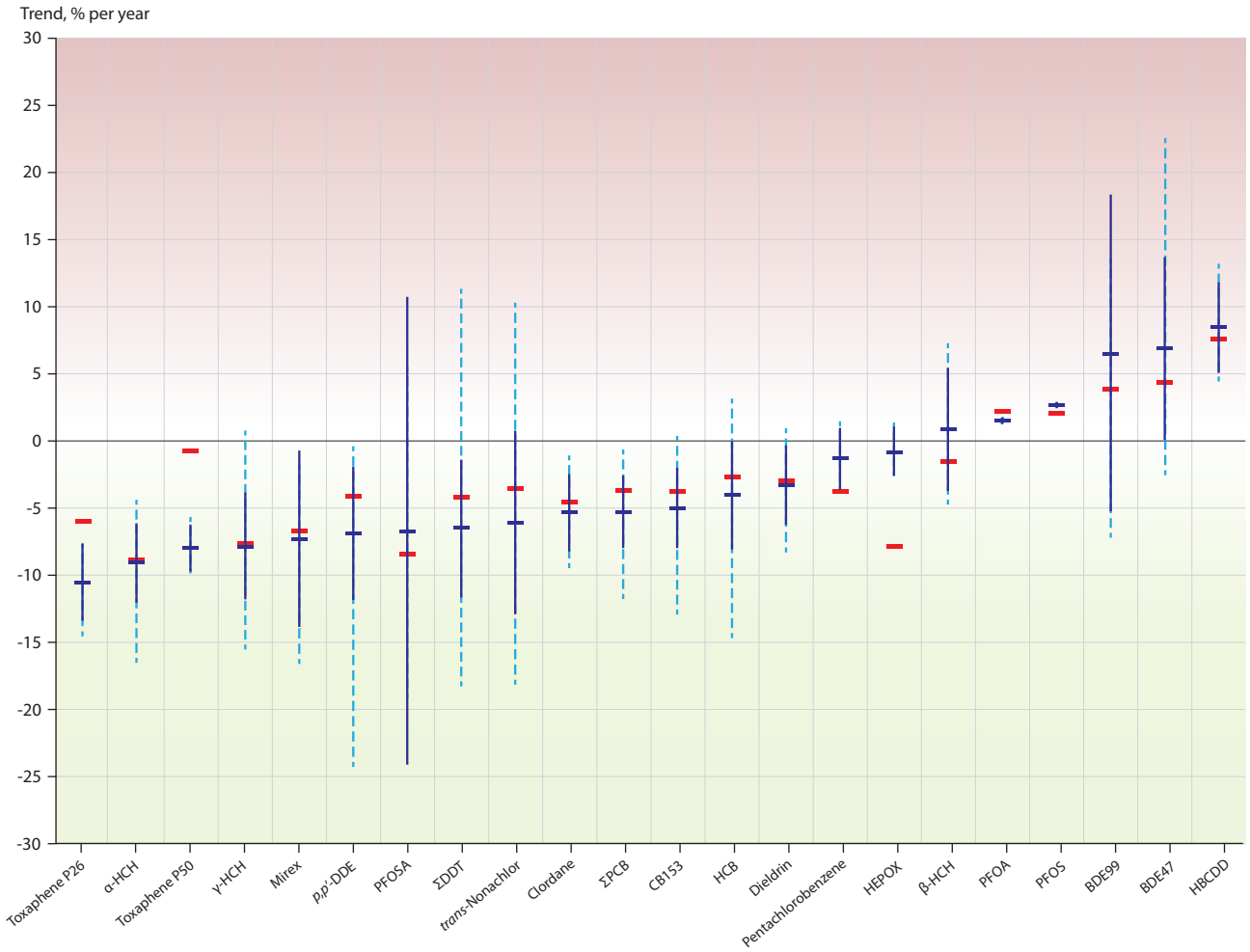



Figure 5.1 Summary of biota trends for different contaminants – results for time-series starting before 2000 where trend results were statistically significant and/or time-series were of 'adequate' power. The graphic shows the ranked mean ±SD (dark blue solid line), range (light blue dashed line); red marks indicate the mean for all runs.

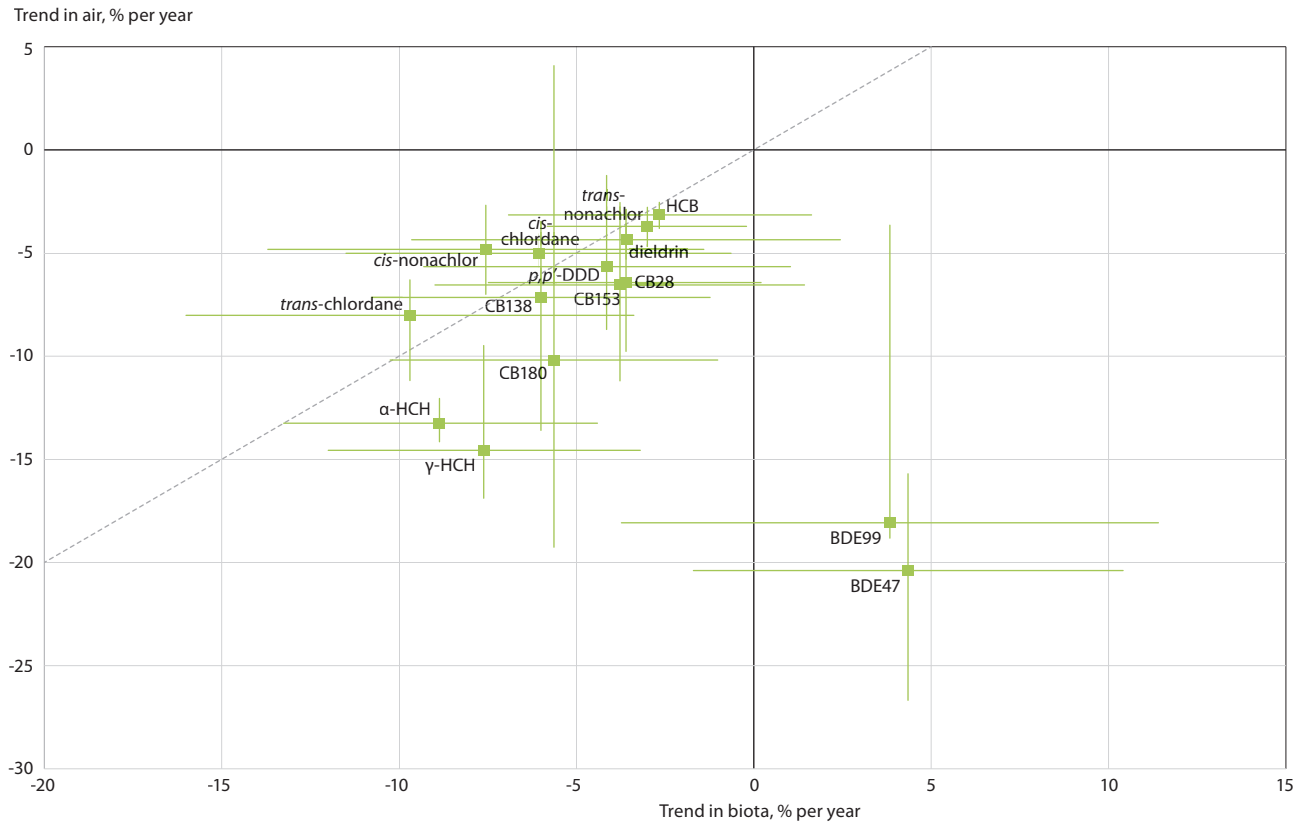



Figure 5.2 Comparison of (average) trends observed in air and biota time-series for selected POPs. The dashed line represents a one-to-one relationship. This graphic shows the mean and range of the results for all air stations with measurements for the chemical concerned and the mean and standard deviation of results for biota time series starting before, in or after 2000 (Annex A6.2).

## Summary Findings

- Downward trends constitute the majority of statistically significant trends of (Stockholm Convention) POPs in Arctic air and biota that have been banned for extended periods (more than 20 to 30 years) in developed countries. For example, DDTs, HCHs, PCBs and chlordanes.
- The downward trend in many of the time-series began decades before the Stockholm Convention entered into force. This probably reflects the impact of control measures introduced at the national level in the 1980s and 1990s, in Arctic countries and in non-Arctic countries in neighboring regions (see Fig. 5.3).
- The rate of decrease in Arctic air for Stockholm Convention POPs that have been subject to regulation for several decades is now slowing, indicating that concentrations are approaching steady state with other environmental media and that secondary sources now dominate. The rate of decrease for Stockholm Convention POPs in Arctic biota is also now slower when comparing time-series that started after 2000 with those that started before 2000.
- 'Newer' POPs that were subject to later regulation in several Arctic countries and have been added to the Stockholm Convention more recently (including the brominated flame retardants PBDEs and HBCDD, and polyfluorinated compounds such as PFOS), show a more mixed pattern of trends, in several cases showing trends increasing until the mid-1990s to 2000 and thereafter decreasing.
- A few time-series for compounds/compound groups that have been subject to regulation for several decades, including *trans*-nonachlor, PCBs ($\Sigma PCB_{10}$ and CB153), ΣDDT and HCB show increasing trends, however most of these occur at sites suspected to be influenced by local sources. This includes time-series of HCB and PCB congeners (CB52 and CB101) in air and HCB (in air and biota) in the eastern Greenland-Iceland-Svalbard region. Increasing trends in air have been attributed to enhanced re-emission from oceans and surfaces and melting ice/snow due to warming. A consistent increase in levels of PCBs and other POPs in polar bears from eastern Greenland in recent years, relative to levels in the mid-2000s seems to be related to a change in diet as a consequence of sea ice loss.
- Some β-HCH time-series from the Canadian Arctic show significant increasing trends, while α-HCH is one of the compounds showing the greatest rate of decrease. The different trends are partly explained by different physical-chemical properties of the two isomers, with α-HCH being transported via air whereas β-HCH partitions to a greater extent into water and so is transported via ocean currents. Also, β-HCH is more persistent and has greater tendency to biomagnify in biota than α-HCH.
- PBDEs in air are declining at the European stations Zeppelin, Stórhöfði and Pallas but not (since the early 2000s) at Alert in Canada. The lack of a decline at Alert may be related to the influence of a nearby military site and generally much higher usage in North America.

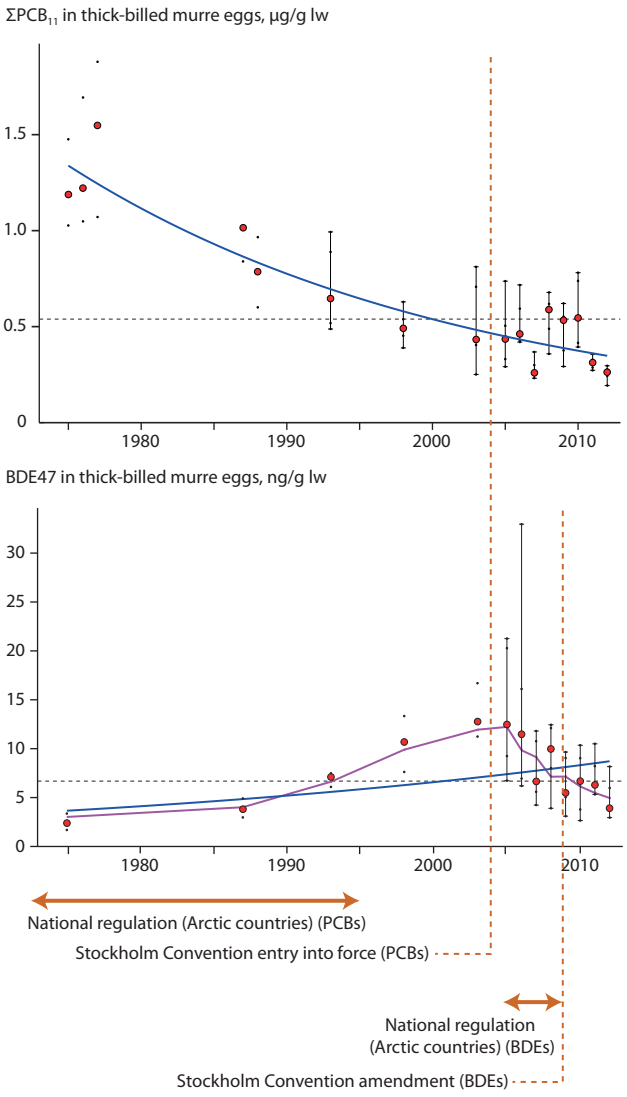



Figure 5.3 Examples of different patterns of change over time for 'legacy POPs' (as illustrated by $\Sigma PCB_{11}$) and 'new POPs' (as illustrated by BDE47) in thick-billed murre eggs collected at Prince Leopold Island (Canada).

Increasing trends in biota time-series starting before 2000 are evident. Controls of hexa- and heptaBDE at both the national and international level were not widely introduced before the late 1990s. The increasing trends in biota are no longer apparent if the time-series for the period after 2000 are considered and, for BDE47 in particular, the proportion of decreasing trends increases (see the example in Fig. 4.33).

- α-HBCDD levels in air have declined at Svalbard since 2006, while increasing trends are evident in biota time-series extending back to the 1980s. HBCDD was included on the Stockholm Convention Annex A list in 2013.
- PFOS precursors showed no trends and decreasing trends in air measured since 2006 at Alert. In biota time-series extending back to the early or mid-1980s, PFOS levels peaked around the mid-2000s. The declines are thought to reflect the voluntary phase-out in 2000 by the US company 3M, of production of PFOS and PFOS-related products. However, particle phase PFOS and PFOA are not showing any decline in Arctic air at Zeppelin, and in biota the three perfluorocarboxylates included in this assessment (PFNA, PFDA, PFUnA) show increasing trends. PFCA-precursors 6:2 FTOH, 8:2 FTOH and 10:2 FTOH show increasing trends in Arctic air.
- Trends in levels in biota (and also in air or human media) are often only interpreted as a response to changes in emission levels. Previous assessments of Arctic trend monitoring data have shown that interpreting trends in this over-simplistic manner is inappropriate. Changes in food-web structure, and in feeding habit and diet can strongly affect levels in biota (including humans), exemplified by the increasing trend of $\Sigma PCB_{10}$ since 2000 in polar bears from eastern Greenland (Fig. 4.26). Trends in levels in air and biota can reflect changes in environmental processes – a number of which can be associated with climate change and variability. A detailed examination of trends in individual datasets is necessary for reliable trend interpretation. Nonetheless, consistency in results from a large number of trend studies, over a large geographical extent, and involving different matrices may provide an indication that global controls on emissions or global processes are responsible for at least some of the observed development. While not necessarily geographically or temporally coincident, there is also a degree of consistency in the trend results obtained from air and biota monitoring (Fig. 5.2) that lends support to the argument that 'other media' should be included in programs such as the Stockholm Convention Global Monitoring Programme that support work to evaluate the effectiveness of that Convention, in addition to the priority media (air and human biomedia).
- Modelling results from studies associated with the EU-ArcRisk project (www.arcrisk.eu) indicate that for most 'legacy' POPs (i.e. POPs which were banned or considerably restricted several decades ago, and where contamination is essentially a legacy of past use) decreases associated with gradual degradation in the environment will continue to exceed any possible increases due to enhanced re-mobilization associated with climate change. Notwithstanding this, however, stockpiles of banned pesticides still exist in some countries which represent a potential source of future, at least local, contamination.
- Results of the ArcRisk studies indicate the need to better characterize primary and secondary sources of POPs and improve models by including indirect effects, such as carbon cycling, catchment hydrology, land use, vegetation cover, etc. (Pacyna et al., 2015). Temperature increases are expected to increase volatilization of chemicals in open-use, including PCBs used as plasticizers in paints and sealants, and PBDEs used as flame retardants.
- Longer time-series are generally more powerful for trend detection, emphasising the need to continue existing time-series and implement new time-series studies in areas not yet covered, and for new chemicals of emerging concern.

# References


Aas, W. and P.B. Nizzetto, 2015. Heavy metals and POP measurements, 2013. NILU EMEP/CCC-Report 3/2015. www.nilu.no/projects/ccc/reports/cccr3-2015.pdf

ACAP, 2013. Environmentally Sound Management of Obsolete Pesticides in the Russian Federation: Final Report for Phases I and II – Inventory and Safe Storage Activities, 2001-2012. Arctic Contaminants Action Program, September 2013.

AMAP, 1998. AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2003. AMAP Assessment 2002: Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2009. AMAP Assessment 2009: Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2010. AMAP Assessment 2009: Persistent Organic Pollutants (POPs) in the Arctic. Science of the Total Environment (SI), 408:2851-3051.

AMAP, 2014. Trends in Stockholm Convention Persistent Organic Pollutants (POPs) in Arctic Air, Human Media and Biota. By: S. Wilson, H. Hung, A. Katsoyiannis, D. Kong, J. van Oostdam, F. Rigét and A. Bignert. AMAP Technical Report No. 7. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2015. AMAP Assessment 2015: Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

Bailey, R.E., D. van Wijk and P.C. Thomas, 2009. Sources and prevalence of pentachlorobenzene in the environment. Chemosphere, 75:555-564.

Bernhoft, A., Ø. Wiig and J.U. Skaare, 1997. Organochlorines in polar bears (*Ursus maritimus*) at Svalbard. Environmental Pollution, 95:159-175.

Bidleman, T.F., L.M. Jantunan, P.B. Kurt-Karakus, F. Wong, H. Hung, J. Ma, G. Stern and B. Rosenberg, 2013. Chiral chemicals as tracers of atmospheric sources and fate processes in a world of changing climate. Mass Spectrometry, 2:S0019. doi:10.5702/massspectrometry.S0019.

Bidleman, T., P. Kurt-Karakus, F. Wong, H. Alegria, L. Jantunen and H. Hung, 2014. Is there still "new" DDT in North America? An investigation using proportions of DDT compounds. In: McConnell, L.L., J. Dachs and C.J. Hapeman (eds.), Occurrence, Fate and Impact of Atmospheric Pollutants, pp. 153-181. ACS Symposium Series. American Chemical Society, Oxford University Press.

Bidleman, T.F., L.M. Jantunen, H. Hung, J. Ma, G.A. Stern, B. Rosenberg and J. Racine, 2015. Annual cycles of organochlorine pesticide enantiomers in Arctic air suggest changing sources and pathways. Atmospheric Chemistry and Physics, 15:1411-1420.

Bignert, A., 2013. The PIA application for statistical analysis of time-series data. www.amap.no/documents/doc/pia-application-version-051113/1026

Bondy, G.S., W.H. Newsome, C.L. Armstrong, C.A.M. Suzuki, J. Doucet, S. Fernie, S.L. Hierlihy, M.M. Feeley and M.G. Barker, 2000. Trans-nonachlor and *cis*-nonachlor toxicity in Sprague-dawley rats: comparisons with technical chlordane. Toxicological Sciences, 58:386-398.

Bossi, R., H. Skov, J. Christensen, K. Vorkamp, S.C. Rastogi, A. Egeløv and D. Petersen, 2008. Atmospheric concentrations of organochlorine pesticides, polybrominated diphenyl ethers (PBDEs) and polychloronaphthalenes (PCNs) in Nuuk, South-West Greenland. Atmospheric Environment, 42:7293-7303.

Bossi, R., C.A. Skjøth and H. Skov, 2013. Three years (2008–2010) of measurements of atmospheric concentrations of organochlorine pesticides (OCPs) at Station Nord, North-East Greenland. Environmental Science: Processes and Impacts, 15:2213-2219.

Bossi, R., K. Vorkamp and H. Skov, 2016. Concentrations of organochlorine pesticides, polybrominated diphenyl ethers and perfluorinated compounds in the atmosphere of North Greenland. Environmental Pollution, 217:4-10.

Bustnes, J.O., G.W. Gabrielsen and J. Verreault, 2010. Climate variability and temporal trends of persistent organic pollutants in the arctic: a study of glaucous gulls. Environmental Science and Technology, 44:3155-3261.

Dietz, R., F.F. Rigét, C. Sonne, E.W. Born, T. Bechshøft, M.A. McKinney, R.J. Drimmie and R.J. Letcher, 2013. Three decades (1983-2010) of contaminant trends in East Greenland polar bears (*Ursus maritimus*). Part 2: Brominated flame retardants. Environment International, 59:494-500.

Dreyer, A., I. Weinberg, C. Temme and R. Ebinghaus, 2009. Polyfluorinated compounds in the atmosphere of the Atlantic and Southern Oceans: Evidence for a global distribution. Environmental Science and Technology, 43:6507-6514.

Fisk, A.T., J. Moisey, K.A. Hobson, N.J. Karnovsky and R.J. Norstrom, 2001. Chlordane components and metabolites in seven species of Arctic seabirds from the Northwater Polynya: relationships with stable isotopes of nitrogen and enantiomeric fractions of chiral components. Environmental Pollution, 113:225-238.

Fryer, R.J. and M.D. Nicholson, 1993. The power of a contaminant monitoring programme to detect linear trends and incidents. ICES Journal of Marine Science, 50:161-168.

Gaden, A., S.H. Ferguson, L. Harwood, H. Melling, J. Alikamik and G.A. Stern, 2012. Western Canadian Arctic ringed seal organic contaminant trends in relation to sea ice break-up. Environmental Science and Technology, 46:4427-4433.

Gawor, A., C. Shunthirasingham, S.J. Hayward, Y.D. Lei, T. Gouin, B.T. Mmereki, W. Masamba, C. Ruepert, L.E. Castillo, M. Shoeib, S.C. Lee, T. Harner and F. Wania, 2014. Neutral polyfluoroalkyl substances in the global atmosphere. Environmental Science: Processes and Impacts, 16:404-413.

Gregor, D.J., H. Loeng and L. Barrie, 1998. The influence of physical and chemical processes on contaminant transport into and within the Arctic. In: Wilson, S.J., J.L. Murray and H.P. Huntington (eds.), AMAP Assessment Report: Arctic Pollution Issues. pp. 25-116. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

Halsall, C.J., R. Bailey, G.A. Stern, L.A. Barrie, P. Fellin, D.C.G. Muir, B. Rosenberg, F. Rovinsky, E. Ya., Kononov and B. Pastukhov, 1998. Multi-year observations of organohalogen pesticides in the Arctic atmosphere. Environmental Pollution, 102:51-62.

Hoguet, J., J.M. Keller, J.L. Reiner, J.R. Kucklick, C.E. Bryan, A.J. Moors, R.S. Pugh and P.R. Becker, 2013. Spatial and temporal trends of persistent organic pollutants and mercury in beluga whales (*Delphinapterus leucas*) from Alaska. Science of the Total Environment, 449:285-294.

Hung, H., P. Blanchard, C.J. Halsall, T.F. Bidleman, G.A. Stern, P. Fellin, D.C.G. Muir, L.A. Barrie, L.M. Jantunen, P.A. Helm, J.Ma and A. Konoplev, 2005. Temporal and spatial variabilities of atmospheric polychlorinated biphenyls (PCBs), organochlorine (OC) pesticides and polycyclic aromatic hydrocarbons (PAHs) in the Canadian Arctic: results from a decade of monitoring. Science of the Total Environment, 342:119-44.

Hung, H., R. Kallenborn, K. Breivik, Y. Su, E. Brorström-Lundén, K. Olafsdottir, J.M. Thorlacius, S. Leppänen, R. Bossi, H. Skov, S. Manø, G.W. Patton, G. Stern, E. Sverko and P. Fellin, 2010. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006. Science of the Total Environment, 408:2854-2873.

Hung, H., P. Kurt-Karakus, L. Ahrens, T. Bidleman, M. Evans, C. Halsall, T. Harner, L. Jantunen, S.C. Lee, D. Muir, M. Shoeib, G. Stern, E. Sverko, Y. Su, P. Vlahos and H. Xiao, 2014. Occurrence and trends in the physical environment. In: Muir, D., P. Kurt-Karakus and J. Stow (eds.), Canadian Arctic Contaminants Assessment Report on Persistent Organic Pollutants – 2013. pp. 147-272. Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada.

Hung, H., A.A. Katsoyiannis, E. Brorström-Lund, K. Olafsdottir, W. Aas, K. Breivik, P. Bohlin-Nizzetto, A. Sigurdsson, H. Hakola, R. Bossi, H. Skov, E. Sverko, E. Barresi, P. Fellin and S. Wilson, 2016. Temporal trends of persistent organic pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP). Environmental Pollution, 217:52-61.

Jantunen, L.M., P.A. Helm, H. Kylin and T.F. Bidleman, 2008. Hexachlorocyclohexanes (HCHs) in the Canadian Archipelago. 2. Air-water gas exchange of α- and γ-HCH. Environmental Science and Technology, 42:465-470.

Kelce, W.R., C.R. Stone, S.C. Laws, E. Gray, J.A. Kemppainen and E.M. Wilson, 1995. Persistent DDT metabolite *p,p'*-DDE is a potent androgen receptor antagonist. Nature, 375:581-585.

Kong, D., M. MacLeod, H. Hung and I.T. Cousins, 2014. Statistical analysis of long-term monitoring data for persistent organic pollutants in the atmosphere at 20 monitoring stations broadly indicates declining concentrations. Environmental Science and Technology, 48:12492-12499.

Kucklick, J., J. Reiner, M. Schantz, J. Keller, J. Hoguet, C. Rimmer, T. Ragland, R. Pugh, A. Moors, C. Rhoderick, J. Ness, D. Peterson and P. Becker, 2013. Persistent organic pollutants and vitamins in northern fur seals (*Callorhinus ursinus*) collected from St. Paul Island, Alaska as part of the Alaska marine mammal tissue archival project. National Institute of Standards and Technology (NISTIR) 7958.

Li, Y. and R. Macdonald, 2005. Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Science of the Total Environment, 342:87-106.

Li, Y.F., R.W. Macdonald, L.M.M. Jantunen, T. Harner, T.F. Bidleman and W.M.J. Strachan, 2002. The transport of β-hexachlorocyclohexane to the western Arctic Ocean: a contrast to α-HCH. Science of the Total Environment, 291:229-246.

McKinney, M.A., S.J. Iverson, A.T. Fisk, C. Sonne, F.F. Rigét, R.J. Letcher, M.T. Arts, E.W. Born, A. Rosing-Asvid and R. Dietz, 2013. Global change effects on the long-term feeding ecology and contaminant exposures of East Greenland polar bears. Global Change Biology, 19:2360-2372.

Miller, A., J.E. Elliott, K.H. Elliott, M.F. Guigueno and L.K. Wilson, 2014. Spatial and temporal trends in brominated flame retardants in seabirds from the Pacific coast of Canada. Environmental Pollution, 195:48-55.

Moisey, J., A.T. Fisk, K.A. Hobson and R.J. Norstrom, 2001. Hexachlorocyclohexane (HCH) isomers and chiral signatures of α-HCH in the Arctic marine food web of the Northwater Polynya. Environmental Science and Technology, 35:1920-1927.

Möller, A., Z. Xie, R. Sturm and R. Ebinghaus, 2011. Polybrominated diphenyl ethers (PBDEs) and alternative brominated flame retardants in air and seawater of the European Arctic. Environmental Pollution, 159:1577-1583.

Nicholson, M.D., R. Fryer and J.R. Larsen, 1998. Temporal trend monitoring: Robust method for analysing contaminant trend monitoring data. ICES Techniques in Marine Environmental Sciences, No. 20.

Nizzetto, P.B., W. Aas and N. Warner, 2015. Monitoring of environmental contaminants in air and precipitation, annual report 2014. NILU report, M-368/2015 www.miljodirektoratet.no/Documents/publikasjoner/M368/M368.pdf

NSIDC, 2015. State of the Cryosphere. Is the cryosphere sending signals about climate change? National Snow and Ice Data Center (NSIDC) Online: http://nsidc.org/cryosphere/sotc/sea_ice.html Updated 8 January, 2015.

Pacyna, J.M., I.T. Cousins, C. Halsall, A. Rautio, J. Pawlak, E.G. Pacyna, K. Sundseth, S. Wilson and J. Munthe., 2015. Impacts on human health in the Arctic owing to climate-induced changes in contaminant cycling – The EU ArcRisk project policy outcome. Environmental Science and Policy, 50: 200-213.

Piekarz, A.M., T. Primbs, J.A. Field, D.F. Barofsky and S. Simonich, 2007. Semivolatile fluorinated organic compounds in Asian and Western U. S. air masses. Environmental Science and Technology, 41:8248-8255.

Rigét, F., A. Bignert, B. Braune, J. Stow and S. Wilson, 2010. Temporal trends of legacy POPs in Arctic biota, an update. Science of the Total Environment, 408, 2874-2884.

Rigét, F., K. Vorkamp, K.A. Hobson, D.C.G. Muir and R. Dietz, 2013a. Temporal trends of selected POPs and potential influence of climate variability in a Greenland ringed seal population. Environmental Science: Processes and Impacts, 15:1706-1716.

Rigét, F., R. Bossi, C. Sonne, K. Vorkamp and R. Dietz, 2013b. Trends of perfluorochemicals in Greenland ringed seals and polar bears: Indications of shifts to decreasing trends. Chemosphere, 93:1607-1614.

Rigét, F., K. Vorkamp, R. Bossi, C. Sonne, R.J. Letcher and R. Dietz, 2016. Twenty years of monitoring of persistent organic pollutants in Greenland biota. A review. Environmental Pollution, 217:114-123.

Schlabach, M., R. Farag-Clement, H. Hung, R. Kallenborn, Y. Su and W. Aas, 2011. AMAP/EMEP/NCP inter-laboratory study for POP analysis 2010. Norwegian Institute for Air Research, Kjeller. EMEP/CCC-Report 7/2011.

Shoeib, M., T. Harner and P. Vlahos, 2006. Perfluorinated chemicals in the arctic atmosphere. Environmental Science and Technology, 40:7577-7583.

Sturludottir, R., H. Gunnlaugsdottir, H.O. Jorundsdottir, E.V. Magnusdottir, K. Olafsdottir and G. Stefansson, 2013. Spatial and temporal trends of contaminants in mussel sampled around the Icelandic coastline. Science of the Total Environment, 454-455:500-509.

Su, Y., H. Hung, G. Stern, E. Sverko, R. Lao, E. Barresi, B. Rosenberg, P. Fellin, H. Li and H. Xiao, 2011. Bias from two analytical laboratories involved in a long-term air monitoring program measuring organic pollutants in the Arctic: a quality assurance/quality control assessment. Journal of Environmental Monitoring, 13:3111-3118.

Tashiro, S. and F. Matsumura, 1978. Metabolism of trans-nonachlor and related chlordane components in rat and man. Archives of Environmental Contamination and Toxicology, 7:113-127.

Tkatcheva, V., B. Ali and E. Reiner, 2013. Northern Contaminants Interlaboratory Quality Assurance Program (NCP III – Phase 7). Northern Contaminants Program, Ottawa. Online: www.aadnc-aandc.gc.ca/eng/1100100035611/1100100035612

Voldner, E.C. and Y.-F. Li, 1995. Global usage of selected persistent organochlorines. Science of the Total Environment, 160/161:201-210.

Vorkamp, K., F.F. Rigét and R. Dietz, 2015. Toxaphene in the aquatic environment of Greenland. Environmental Pollution, 200:140-148.

Weber, J., C.J. Halsall, D. Muir, C. Teixeira, J. Small and K. Solomon, 2010. Endosulfan, a global pesticide: A review of its fate in the environment and occurrence in the Arctic. Science of the Total Environment, 408:2966-2984.

Wöhrnschimmel, H., P. Tay, H. von Waldow, H. Hung, Y.-F. Li, M. MacLeod and K. Hungerbuehler, 2012. Comparative assessment of the global fate of α- and β-hexachlorocyclohexane before and after phase-out. Environmental Science and Technology, 46:2047-2054.

Wong, F., L.M. Jantunen, M. Pućko, T. Papakyriakou, R.M. Staebler, G.A. Stern and T.F. Bidleman, 2011. Air-water exchange of anthropogenic and natural organohalogens on International Polar Year (IPY) expeditions in the Canadian Arctic. Environmental Science and Technology, 45:876-881.

# Annex 1. Observations on the power of the AMAP biota trends monitoring program and use of the PIA statistical application for trend detection

*Author:* Frank Rigét

## How powerful is the time-series?

Statistical power is an important concept when examining temporal trends in monitoring data. The statistical power of a time-series should be sufficiently sensitive to detect a trend of a certain magnitude. When working with time-series with inadequate power, there is a risk of incorrectly suggesting that no change has occurred in a given period. In the case of a log-linear regression the power depends on the magnitude of the trend, the sample size collected each year, the number of years of sampling data and the between-year variability and choice of significance level (Fryer and Nicholson, 1993).

In the following, estimation of power will follow the method described by Fryer and Nicholson (1993). There will be two measures of power: (1) the number of years required to detect a log-linear trend of a 5% annual change with 80% power and a significance level $\alpha<0.05$ with a one-sided test, and (2) the minimum average annual percentage change over a specified period that can be detected with 80% power, $\alpha<0.05$ and a one-sided test. The two measures are essentially the same but provide different information.

Only 12% of the time-series starting before 2000 fulfill the statistical standard requirements (5% annual change, 80% power, 5% significance level) (Fig. A1.1, left). This number would of course increase if the statistical requirements were relaxed (increasing the annual change, decreasing the power, increasing the significance level). Especially in cases with increasing trends and the use of the rather conservative 5% significance level, this means that a test *p*-value should be below 0.05 before the zero hypotheses ($H_0$) of no trend can be rejected. A more precautionary approach with regard to protection of the environment would be to use a significance level of 10% in order to give a warning for, for example, an increasing trend of a contaminant. In the case of the Greenlandic time-series, the number of time-series with 80% power to detect an annual change would increase from 29% to 43% if the significance level was increased from 5% to 10%.

The number of time-series started in or later than 2000 with a power of at least 80% falls to 4% as expected (Fig. A1.1, right). This underlines the fact that time-series must be rather long before sufficient power to detect trends is reached.

The last AMAP POP trend assessment (Rigét et al., 2010) reported that 8% of the time-series had 80% or more power. In this assessment the value increased to 13% for organochlorine time-series excluding the polybrominated diphenyl ethers (PBDEs) and perfluorinated alkylated substances (PFASs) time-series as they were not included in the previous AMAP assessment. Including the BDE and PFA time-series the percentage is 12%. It should be noted that the number of powerful OC time-series has increased from 24 in the previous assessment to 119 in the present assessment. The conclusions drawn in this assessment are therefore more solidly founded. Furthermore, if the number of time-series that showed a significant decreasing or increasing log-linear trend but which did not necessarily meet the statistical requirements are also considered, the percentage and number increase to 44% and 478, respectively.

A consequence of the relatively low power and assuming that the sampling and analytical procedures have been optimized, is that the number of years of the time-series must be high. 53% of the time-series need more than 20 years to obtain 80% power to detect a 5% annual change with a significance level of 5% (Fig. A1.2).

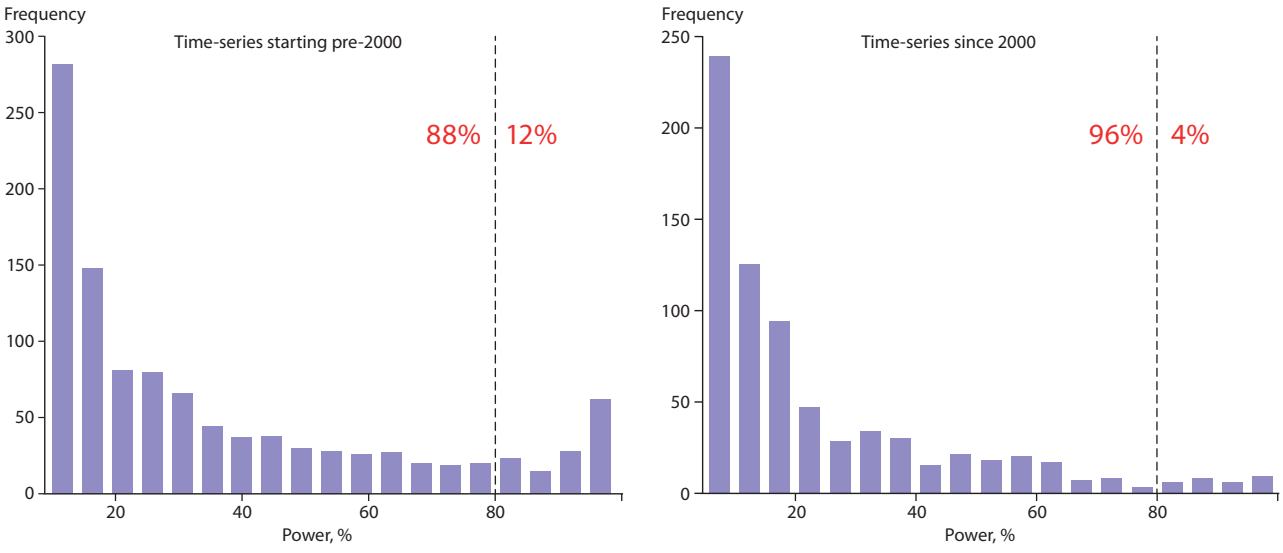



Figure A1.1 Frequency of the power of time-series to detect a 5% annual change with the current number of years with a significance level of 5%. Time-series starting before 2000 (left) and time-series starting after 2000 (right).

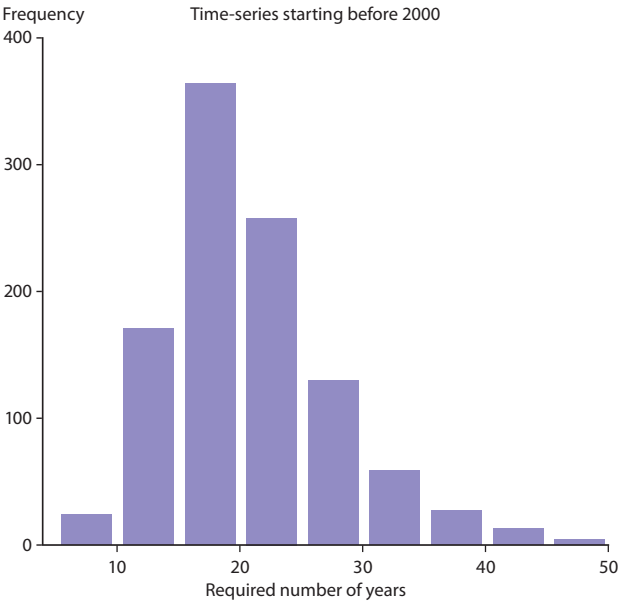



Figure A1.2 Frequency of the number of years required in a time-series to detect a 5% annual change with 80% power and a significance level of 5%.

The power and lowest detectable trend for a 10-year period are compared between media in Table A1.1. The highest power and among the lowest detectable trend are seen for seabird eggs. The reason for this is probably because seabird eggs are easier to sample consistently from year to year, both with regard to time of year and to problems with confounding factors such as age and sex. Low power is seen for fish liver and muscle and may be related to the rather low POP concentrations, where analytical uncertainties play a relatively greater role.

### Comparison between test results of the log-linear regression and Mann-Kendall trend test

The PIA program gives the results of two different trend analyses; the log-linear regression and the Mann-Kendall trend test. The log-linear regression is a parametric test which assumes, besides a log-linear relationship that the errors are normally distributed. The Mann-Kendall trend test is a non-parametric test that measures the association between two variables. If a significance level of 5% is the basis for concluding whether a trend has occurred, the two tests sometimes lead to different conclusions.

In all the time-series analyzed in this assessment that do not have a significant non-linear trend component, the two tests lead to different conclusions in 9% of the time-series. In 6% of the time-series, the Mann-Kendall *p*-values were above 5%, while the log-linear regression *p*-values were below 5% (Fig. A1.3). This can happen if one or more outliers are present at one end of a time-series (or at both ends in opposite directions). In 3% of the time-series, the Mann-Kendall *p*-values were below 5% while the log-linear regression *p*-values were above 5% (Fig. A1.3). This can happen in the presence of outliers especially in the middle of the time-series or when the trend is systematic from year-to-year but the magnitude of the trend is small.

Therefore, when a different result is achieved from each of the two tests, it is recommended to take a closer look at the time-series data, for example the PIA trend plot, before any firm conclusions concerning the trend can be drawn.

Table A1.1 Mean and standard deviation of the statistical power and lowest detectable trend for a 10-year period for each medium. Time-series starting before 2000.

| | Power | Lowest detectable trend |
|---|---|---|
| Freshwater fish | 14 ± 11 | 33 ± 31 |
| liver | 12 ± 5 | 25 ± 12 |
| muscle | 15 ± 12 | 35 ± 35 |
| Marine fish | 16 ± 11 | 22 ± 10 |
| liver | 17 ± 13 | 21 ± 10 |
| muscle | 14 ± 7 | 23 ± 9 |
| Marine mammals | 22 ± 18 | 24 ± 28 |
| adipose tissue | 17 ± 9 | 23 ± 20 |
| blubber | 24 ± 20 | 23 ± 28 |
| liver | 14 ± 12 | 54 ± 56 |
| muscle | 19 ± 7 | 16 ± 6 |
| Mussels | 20 ± 19 | 19 ± 8 |
| soft body | 20 ± 19 | 19 ± 8 |
| Seabirds | 30 ± 23 | 21 ± 29 |
| egg | 32 ± 23 | 21 ± 30 |
| liver | 8 | 34 |
| blood | 17 ± 5 | 18 ± 9 |

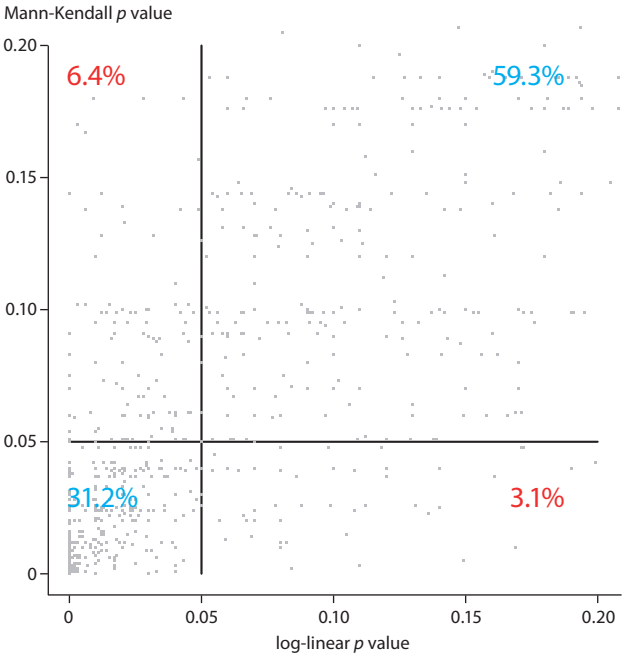



Figure A1.3 Plot of *p*-values from the log-linear regression test versus *p*-values of Mann-Kendall trend test. Time-series with a significant non-linear trend component are excluded.

### Literature cited

Fryer, R.J. and M.D. Nicholson, 1993. The power of a contaminant monitoring programme to detect linear trends and incidents. ICES Journal of Marine Science, 50:161-168.

Rigét, F., A. Bignert, B. Braune, J. Stow and S. Wilson, 2010. Temporal trends of legacy POPs in Arctic biota, an update. Science of the Total Environment, 408, 2874-2884.

# Annex 2. Description and interpretation of results from national trend studies: Canada – Seabirds

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## Introduction

Many persistent halogenated contaminants biomagnify up the food chain and so biota, including seabirds which feed at high trophic positions, are vulnerable to contaminant exposure via their diet (Hobson et al., 2002; Hop et al., 2002; Borgå et al., 2004; Bossi et al., 2005; Kelly et al, 2009). Seabird eggs reflect the contaminant burden in the female at the time of laying (Braune and Norstrom, 1989; Verreault et al., 2006; Gebbink and Letcher, 2012) and are therefore used as a non-invasive means of monitoring contaminants, lipophilic compounds (Bignert et al., 1998; Braune, 2007) as well as perfluoroalkyl acids, such as perfluorooctane sulfonate (PFOS) and perfluorinated carboxylates (PFCAs) (Gebbink et al., 2011; Braune and Letcher, 2013), which associate with proteins rather than partitioning into lipid (Butt et al., 2010).

Eggs of thick-billed murre (*Uria lomvia*), northern fulmar (*Fulmarus glacialis*) and black-legged kittiwake (*Rissa tridactyla*) have been monitored for environmental contaminants at Prince Leopold Island Migratory Bird Sanctuary (74°02'N, 90°05'W) in Lancaster Sound, Nunavut, Canada, since 1975 (1975, 1976, 1977, 1987, 1988, 1993, 1998, 2003, annually since 2005), and thick-billed murre has also been monitored at Coats Island (62°98'N, 82°00'W) in northern Hudson Bay since 1993 (1993, 1998, 2003, annually since 2005). All samples have been archived in the National Wildlife Specimen Bank at the National Wildlife Research Centre in Ottawa, Canada. Archived samples have allowed for retrospective analyses to be carried out for new, emerging compounds. The Northern Contaminants Program (NCP) of Indigenous and Northern Affairs Canada has supported the monitoring of contaminants in seabirds as well as a variety of other biota in the Canadian Arctic since its inception in 1991. These data have been included in assessments of contaminants in the Canadian Arctic under the NCP, as well as in assessments of contaminants in the circumpolar Arctic under the Arctic Monitoring and Assessment Programme (AMAP).

## Methods

Interpretation of contaminant concentrations in biota may be confounded if populations vary their diet over trophic levels through time (see Hebert et al., 1997, 2000). Determining trophic position, as a reflection of dietary change, is possible by measuring naturally occurring stable isotopes of nitrogen ($^{15}N/^{14}N$, expressed as $\delta^{15}N$) (Hobson and Welch, 1992; Hobson et al., 1994; Hebert et al., 1999). This is also true of seabird eggs since stable isotope ratios in egg material are expected to reflect the diet of the female prior to or during egg-laying (Hobson, 1995; Hebert et al., 1999). For details of methodology used by data originator/custodian, see Braune and Letcher (2013) for perfluorinated sulfonate and carboxylate compounds, and Braune (2007) for legacy organochlorines. Only those compounds for which more than 90% of samples had detectable concentrations for a given species were statistically analyzed. Non-detect values (<0.1 ng/g ww) were set to one half of the detection limit for the statistical analyses but were set to zero for calculating annual means and the sums of the major organochlorine groups (i.e. $\Sigma PCB_{10}$, $\Sigma PCB_{11}$, ΣDDT, ΣChordanes).

## Summary of program results

With the exception of β-HCH, most of the legacy persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs) and *p,p'*-DDE, have decreased in eggs of thick-billed murre, northern fulmar and black-legged kittiwake monitored at Prince Leopold Island since 1975 (Braune, 2007; NCP, 2013), and concentrations of most declining POPs now appear to be levelling off, albeit with some interannual variability. Likewise, concentrations of legacy POPs have decreased in murre eggs at Coats Island in northern Hudson Bay (Braune et al., 2015a). Concentrations of the legacy organochlorines in eggs of the ivory gull (*Pagophila eburnea*) collected from Seymour Island also either decreased or showed little change between 1976 and 2004 (Braune et al., 2007).

Concentrations of β-HCH, however, have increased in eggs of all three seabird species at Prince Leopold Island due to a combination of the compound's recalcitrant nature in biota and its physical-chemical characteristics which have resulted in its delayed transport to Arctic waters via ocean currents from source regions in southeastern Asia (Braune, 2007). It has been suggested that the lower concentrations of β-HCH reported in murre eggs at Coats Island compared to Prince Leopold Island are due to water flow dynamics in the eastern Canadian Arctic (Braune et al., 2014). No significant temporal trend was found for β-HCH at Coats Island.

Although no significant temporal trends were found for the polybrominated diphenyl ethers (PBDEs) in eggs of thick-billed murre and northern fulmar from Prince Leopold Island over the period 1975 to 2012, concentrations of ΣPBDE (sum of BDE17, BDE28, BDE49, BDE47, BDE66, BDE100, BDE99, BDE85, BDE153, BDE138, BDE183 and BDE190) steadily increased from 1975 to 2003 in both species after which levels started to decrease, driven largely by concentrations of BDE47 (NCP, 2013; Braune et al., 2015b). An increase in ΣPBDE concentrations between 1976 and 2004 is also reflected in ivory gull eggs collected from Seymour Island in the Canadian Arctic (Braune et al., 2007). BDE47 is a major component of the commercial Penta-BDE product used in polyurethane foam for which North America accounted for most of the global demand (Hale et al., 2003). BDE47 was the predominant congener in the eggs of thick-billed murre and northern fulmar, as well as ivory gull eggs (Braune et al., 2007), followed by BDE99 and BDE100 in all years analyzed. The increase in ΣPBDE concentrations in the seabird eggs between the mid-1970s and 2003/2004 agrees well with the pattern of significant increase seen during the

period spanning the early 1970s to the early 2000s in North America (Hale et al., 2003; Law et al., 2003). The declining concentrations observed in the fulmar and murre eggs after 2003 may reflect the phasing out of Penta-mix BDE usage in North America after 2005 (de Wit et al., 2010).

Concentrations of PFCAs in liver of thick-billed murre and northern fulmar from Prince Leopold Island increased significantly from 1975 to 2003/2004 (Butt et al., 2007a), and concentrations of total PFCAs [ΣPFCA= sum of PFHxA ($C_6$), PFHpA ($C_7$), PFOA ($C_8$), PFNA ($C_9$), PFDA ($C_{10}$), PFUnA ($C_{11}$), PFDoA ($C_{12}$), PFTrA ($C_{13}$), PFTeA ($C_{14}$) and PFPA ($C_{15}$)] increased from 1975 to 2011 in eggs of both northern fulmar and thick-billed murre from Prince Leopold Island (Braune and Letcher, 2013). A more recent analysis of the data has shown that concentrations of ΣPFCA increased significantly from 1975 to 2008 in northern fulmar eggs and from 1975 to 2010 in thick-billed murre eggs followed by significant declines for both species to 2012 (Braune and Letcher, 2014). Significant increases in concentrations of all longer-chained PFCAs ($C_9$–$C_{15}$) contributed to the increase in ΣPFCA in the murres and the fulmars. PFUnA ($C_{11}$) and PFTrA ($C_{13}$) were the predominant PFCAs measured in eggs of both species (Braune and Letcher, 2013, 2014). The increase in ΣPFCA concentrations in both murre and fulmar eggs up to 2008–2010 is consistent with observed increases in other Arctic species monitored into the early 2000s, including herring gull (*Larus argentatus*) eggs from northern Norway (Verreault et al., 2007), polar bears (*Ursus maritimus*) from Alaska and Baffin Island (Smithwick et al., 2006), ringed seals (*Pusa hispida*) from the Canadian Arctic (Butt et al., 2007b) and beluga (*Delphinapterus leucas*) from Alaska (Reiner et al., 2011). PFCAs are not regulated under the Stockholm Convention on Persistent Organic Pollutants. However, the United Nations Environment Programme (UNEP) Strategic Approach for International Chemicals Management (SAICM) includes an initiative to reduce emissions, particularly of the longer-chain PFCAs (i.e. carbon chain lengths $C_8$ or higher) (Martin et al., 2013). The recently observed declines in concentration of ΣPFCA in fulmar and murre eggs may be a reflection of this recent UNEP initiative.

Perfluorooctane sulfonate levels did not change significantly in eggs of either thick-billed murre or northern fulmar from Prince Leopold Island over the period 1975 to 2011 (Braune and Letcher, 2013) or 2012 (Braune and Letcher, 2014). However, PFOS levels have shown significant declines since 2009 in fulmars and since 2008 in murres (Braune and Letcher, 2014). The recent declines in PFOS concentration may represent variability in the temporal trend data set, or they may indicate a delayed response to the manufacturing phase-out of PFOS by the 3M Company between 2000 and 2002 (Butt et al., 2010). Other studies from the Arctic and elsewhere have also shown recent declines in PFOS levels in response to the phase-out (Butt et al., 2010; Houde et al., 2011). Variability in the PFOS data may also be due to annual differences in the contribution of the degradation of PFOS precursors to PFOS. PFOS precursors such as *N*-Et-FOSA can be dealkylated to FOSA, which can in turn be degraded to PFOS, as has been reported *in vitro* in liver microsomal studies with polar bears and Arctic ringed seals (Letcher et al., 2014). There still remain many exemptions for PFOS under the Stockholm Convention resulting in PFOS and related compounds remaining in global commerce, as well as continued production of PFOS in China and Brazil (Martin et al., 2013). Therefore, it is not inconceivable that the recent declines of PFOS levels in the Arctic seabird data sets are just an artifact of data variability. Continued monitoring will determine whether the recent declines in PFOS concentrations in the Arctic seabird eggs constitute a real downward trend.

## Conclusions

It has long been recognized that seabirds are good monitors of contaminants in the marine environment. Their rapid response to changes in emission patterns of contaminants further endorses their utility as effective biomonitors. For example, the patterns of organochlorine decline documented to date in these migratory seabirds probably reflect overall lower contamination of the food chain resulting from restrictions placed on the use of many of these compounds in the 1970s and 1980s. There are many factors which affect contaminant cycling in the Arctic including changes in the structure and dynamics of food webs as reflected through $\delta^{15}N$ values. It is critical to understand the mechanisms driving temporal trends in contaminants in order to interpret them correctly. Further research is needed to investigate the complex links between climate change and biogeochemical and food-web processes in Arctic marine ecosystems.

## Literature cited

Bignert, A., M. Olsson, W. Persson, S. Jensen, S. Zakrisson, K. Litzén, U. Eriksson, L. Häggberg and T. Alsberg, 1998. Temporal trends of organochlorines in Northern Europe, 1967-1995. Relation to global fractionation, leakage from sediments and international measures. Environmental Pollution 99:177-198.

Borgå, K., A.T. Fisk, P.F. Hoekstra and D.C.G. Muir, 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in Arctic marine food webs. Environmental Toxicology and Chemistry, 23:2367-2385.

Bossi, R., F.F. Rigét, R. Dietz, C. Sonne, P. Fauser, M. Dam and K. Vorkamp, 2005. Preliminary screening of perfluorooctane sulfonate (PFOS) and other fluorochemicals in fish, birds and marine mammals from Greenland and the Faroe Islands. Environmental Pollution, 136: 323-329.

Braune, B.M., 2007. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975 to 2003. Environmental Pollution, 148:599-613.

Braune, B.M. and R.J. Letcher, 2013. Perfluorinated sulfonate and carboxylate compounds in eggs of seabirds breeding in the Canadian Arctic: temporal trends (1975-2011) and interspecies comparison. Environmental Science and Technology, 47:616-624.

Braune, B.M. and R.J. Letcher, 2014. Temporal trends of perfluorinated sulfonate and carboxylate compounds in seabird eggs from the Canadian Arctic. Organohalogen Compounds, 76:138-141.

Braune, B.M. and R.J. Norstrom, 1989. Dynamics of organochlorine compounds in herring gulls: III. Tissue distribution and bioaccumulation in Lake Ontario gulls. Environmental Toxicology and Chemistry, 8:957-968.

Braune, B.M., M.L. Mallory, H.G. Gilchrist, R.J. Letcher and K.G. Drouillard, 2007. Levels and trends of organochlorines and brominated flame retardants in ivory gull eggs from the Canadian Arctic, 1976 to 2004. Science of the Total Environment, 378:403-417.

Braune, B.M., A.J. Gaston, R.J. Letcher, H.G. Gilchrist, M.L. Mallory and J.F. Provencher, 2014. A geographical comparison of chlorinated, brominated and fluorinated compounds in seabirds breeding in the eastern Canadian Arctic. Environmental Research, 134:46-56.

Braune, B.M., A.J. Gaston, K.A. Hobson, H.G. Gilchrist and M.L. Mallory, 2015a. Changes in trophic position affect rates of contaminant decline at two seabird colonies in the Canadian Arctic. Ecotoxicology and Environmental Safety, 115:7-13.

Braune, B.M., R.J. Letcher, A.J. Gaston and M.L. Mallory, 2015b. Trends of polybrominated diphenyl ethers and hexabromocyclododecane in eggs of Canadian Arctic seabirds reflect changing use patterns. Environmental Research, 142:651-661.

Butt, C.M., S.A. Mabury, D.C.G. Muir and B.M. Braune, 2007a. Prevalence of long-chained perfluorinated carboxylates in seabirds from the Canadian Arctic between 1975 and 2004. Environmental Science and Technology, 41:3521-3528.

Butt, C.M., D.C.G. Muir, I. Stirling, M. Kwan and S.A. Mabury, 2007b. Rapid response of arctic ringed seals to changes in perfluoroalkyl production. Environmental Science and Technology, 41:42-49.

Butt, C.M., U. Berger, R. Bossi and G.T. Tomy, 2010. Levels and trends of poly- and perfluorinated compounds in the arctic environment. Science of the Total Environment, 408:2936-2965.

de Wit, C.A., D. Herzke and K. Vorkamp, 2010. Brominated flame retardants in the Arctic environment – trends and new candidates. Science of the Total Environment, 408:2885-2918.

Gebbink, W.A. and R.J. Letcher, 2012. Comparative tissue and body compartment accumulation and maternal transfer to eggs of perfluoroalkyl sulfonates and carboxylates in Great Lakes herring gulls. Environmental Pollution, 162:40-47.

Gebbink, W.A., R.J. Letcher, C.E. Hebert and D.V.C. Weseloh, 2011. Twenty years of temporal change in perfluoroalkyl sulfonate and carboxylate contaminants in herring gull eggs from the Laurentian Great Lakes. Journal of Environmental Monitoring, 13:3365-3372.

Hale, R.C., M. Alaee, J.B. Manchester-Neesvig, H.M. Stapleton and M.G. Ikonomou, 2003. Polybrominated diphenyl ether flame retardants in the North American environment. Environment International, 29:771-779.

Hebert, C.E. and K.A. Keenleyside, 1995. To normalize or not to normalize? Fat is the question. Environmental Toxicology and Chemistry, 14:801-807.

Hebert, C.E., J.L. Shutt and R.J. Norstrom, 1997. Dietary changes cause temporal fluctuations in polychlorinated biphenyl levels in herring gull eggs from Lake Ontario. Environmental Science and Technology, 31:1012-1017.

Hebert, C.E., J.L. Shutt, K.A. Hobson and D.V.C. Weseloh, 1999. Spatial and temporal differences in the diet of Great Lakes herring gulls (*Larus argentatus*): evidence from stable isotope analysis. Canadian Journal of Fisheries and Aquatic Science, 56:323-338.

Hebert, C.E., K.A. Hobson and J.L. Shutt, 2000. Changes in food web structure affect rate of PCB decline in herring gull (*Larus argentatus*) eggs. Environmental Science and Technology, 34:1609-1614.

Hobson, K.A., 1995. Reconstructing avian diets using stable-carbon and nitrogen isotope analysis of egg components: patterns of isotopic fractionation and turnover. Condor, 97:752-762.

Hobson, K.A. and H.E. Welch, 1992. Determination of trophic relationships within a high Arctic marine food web using $\delta^{13}C$ and $\delta^{15}N$ analysis. Marine-Ecology Progress Series, 84:9-18.

Hobson, K.A., J.F. Piatt and J. Pitocchelli, 1994. Using stable isotopes to determine seabird trophic relationships. Journal of Animal Ecology, 63:786-798.

Hobson, K.A., A. Fisk, N. Karnovsky, M. Holst, J.M. Gagnon and M. Fortier, 2002. A stable isotope ($\delta^{13}C$, $\delta^{15}N$) model for the North Water food web: implications for evaluating trophodynamics and the flow of energy and contaminants. Deep-Sea Research II, 49:5131-5150.

Hop, H., K. Borgå, G.W. Gabrielson, L. Kleivane and J.U. Skaare, 2002. Food web magnification of persistent organic pollutants in poikilotherms and homeotherms from the Barents Sea. Environmental Science and Technology, 36:2589-2597.

Houde, M., A.O. De Silva, D.C.G. Muir and R.J. Letcher, 2011. An updated review of monitoring and accumulation of perfluorinated compounds in aquatic biota. Environmental Science and Technology, 45:7962-7973.

Kelly, B.C., M.G. Ikonomou, J.D. Blair, B. Surridge, D. Hoover, R. Grace and F.A.P.C. Gobas, 2009. Perfluoroalkyl contaminants in an Arctic marine food web: trophic magnification and wildlife exposure. Environmental Science and Technology, 43:4037-4043.

Law, R.J., M. Alaee, C.R. Allchin, J.P. Boon, M. Lebeuf, P. Lepom and G.A. Stern, 2003. Levels and trends of polybrominated diphenyl ethers and other brominated flame retardants in wildlife. Environment International, 29:757-770.

Letcher, R.J., S.G. Chu, M.A. McKinney, G.T. Tomy, R. Dietz and C. Sonne, 2014. Comparative hepatic *in vitro* depletion and metabolite formation of major perfluorooctane sulfonate precursors in arctic polar bear, beluga whale, and ringed seal. Chemosphere, 112:225-231.

Martin, J.W., S.A. Mabury, K.R. Solomon and D.C.G. Muir, 2013. Progress toward understanding the bioaccumulation of perfluorinated alkyl acids. Environmental Toxicology and Chemistry, 32:2421-2423.

NCP, 2013. Canadian Arctic Contaminants Assessment Report on Persistent Organic Pollutants – 2013. Muir, D., P. Kurt-Karakus and J. Stow (eds.), Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, pp. 377-381.

Peakall, D.B. and P. Gilman, 1979. Limitations of expressing organochlorine levels in eggs on a lipid-weight basis. Bulletin of Environmental Contamination and Toxicology, 23:287-290.

Reiner, J.L., S.G. O'Connell, A.J. Moors, J.R. Kucklick, P.R. Becker and J.M. Keller, 2011. Spatial and temporal trends of perfluorinated compounds in beluga whales (*Delphinapterus leucas*) from Alaska. Environmental Science and Technology, 45:8129-8136.

Smithwick, M., R.J. Norstrom, S.A. Mabury, K. Solomon, T.J. Evans, I. Stirling, M.K. Taylor and D.C.G. Muir, 2006. Temporal trends of perfluoroalkyl contaminants in polar bears (*Ursus maritimus*) from two locations in the North American Arctic, 1972-2002. Environmental Science and Technology, 40:1139-1143.

Verreault, J., R.A. Villa, G.W. Gabrielsen, J.U. Skaare and R.J. Letcher, 2006. Maternal transfer of organohalogen contaminants and metabolites to eggs of Arctic-breeding glaucous gulls. Environmental Pollution, 144:1053-1060.

Verreault, J., U. Berger and G.W. Gabrielsen, 2007. Trends of perfluorinated alkyl substances in herring gull eggs from two coastal colonies in northern Norway: 1983-2003. Environmental Science and Technology, 41:6671-6677.

# Annex 3. Description and interpretation of results from national trend studies: Iceland – Marine fish and shellfish

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## Introduction

Annual monitoring of trace metals in marine biota around Iceland began in 1989 and monitoring of organochlorine compounds a couple of years later, in 1991. The monitoring program includes annual monitoring of heavy metals and organochlorine compounds in blue mussel (*Mytilus edulis*) collected along the coast around Iceland, and cod (*Gadus morhua*) fished in the Icelandic exclusive economic zone (EEZ) using sampling guidelines by OSPAR and the International Council for the Exploration of the Sea (ICES). Mussels are collected from 11 sites along the Icelandic coastline where each sample comprises 50 individuals. Cod is fished at three sites in the Icelandic EEZ; two northwest of Iceland and one northeast of Iceland and each sample comprises 25 individuals. For mussels, one pooled sample is prepared that represents each site each year. For cod, the 25 livers were divided into four to six sub-samples according to the weight of the livers; livers of similar weight were pooled, except in 1992 and 1995 when all livers were pooled into one group. The project, overseen by the Environment Agency of Iceland, is to fulfil agreements under OSPAR and AMAP. The project has been funded by the Icelandic Ministry for the Environment and Natural Resources as well as the Icelandic Ministry of Industries and Innovation. The data obtained are a part of Iceland's contribution to the ICES databank (www.ices.dk). Each year, the Environment Agency of Iceland agrees a contract with Matís Ltd (or its predecessor) for coordination of monitoring the marine biosphere around Iceland. Under this this contract, Matís is responsible for methods relating to sampling, preparation and analysis of samples. Samples are collected by the Icelandic Marine Research Institute and are analyzed at Matís and the Department of Pharmacology and Toxicology at the University of Iceland. An annual report has been prepared by Matís and made available via the Matís (www.matis.is) and Environment Agency (www.ust.is) homepage (Jörundsdóttir et al., 2013).

## Methods

### *Mussels*

As reported by Sturludóttir et al. (2013), to test for an overall pattern of change in contaminant concentration and to test whether the pattern of change and concentration differs between locations, analyses with random coefficient models were carried out using a method by Verbeke and Molenberghs (2000). They recommended starting with a saturated model, including both fixed and random effects. A polynomial model of order four was considered to be a saturated model. The percentage of fat in the mussels was included in the model with a quadratic term to reduce variability. Concentrations were log-transformed prior to analysis to meet the normal assumption. The full model was thus:

$$Y_{it}=\beta_0+\beta_1 t+\beta_2 t^2+\beta_3 t^3+\beta_4 t^4+\beta_5 f_{it}+\beta_6 f_{it}^2+b_{0i} +b_{1i}t+b_{2i}t^2+b_{3i}t^3+b_{4i}t^4+b_{5i}f_{it}+b_{6i}f_{it}^2+\epsilon_{it}$$

$$b\sim N(0,G)$$
$$\epsilon\sim N(0,\sigma^2 I)$$

where $Y_{it}$ is the log-concentration of a contaminant at location *i* and year *t*. The 'β's are fixed effect coefficients, the 'b's are coefficients describing random effects and $f_{it}$ is the percentage of fat in the sample at location *i* and year *t*. The G matrix was fitted as a diagonal matrix, that is, the random effects were assumed independent. Random effects were tested first using the restricted likelihood ratio test, which is known to be conservative (Pinheiro and Bates, 2000). Subsequently the fixed effects were tested using likelihood ratio tests. Terms not significant at α=0.05 were removed from the model. If fixed effects could be dropped the random effects were tested again. The modelling procedure was done using the nlme package (Pinheiro et al., 2012) in the statistical software R (R Development Core Team R, 2013).

The random effects indicate whether and how much variation exists between locations in concentration and pattern of change with time. If none of the random effects are significant then there is not a significant difference in concentration between locations or in the pattern of change. If for example only the intercept, $b_{0i}$ is significant then there is no significant difference in the pattern of change between locations but there is a significant difference in average concentration. The expected values of the 'b's are zero so the fixed effects can be interpreted as the average concentration and pattern of change of all locations. The best linear unbiased predictions (BLUPs) of random effects were used to determine the pattern of change at each location. The average concentration for each individual sampling location was calculated from the predicted values for the years sampled, adjusted for average fat percentage (0.41%). All values are given on a dry weight basis.

### *Cod*

As reported by Sturludóttir et al. (2014), to test whether contaminant concentrations in cod had changed over the sample periods a method of mixed models was applied (Pinheiro and Bates, 2000). The change in concentration over time was modelled as a polynomial and a polynomial model of order four was considered a saturated model. The average age, length, liver fat content and liver weight of the subsamples were used as covariates to adjust for biological variation. Interactions between location and the fixed effects were added to the model to test whether the change in concentration was different between locations and to test whether the relationship between concentrations and covariates differed between the two locations. It is well established that biological attributes of marine animals caught near each other tend to be more similar than attributes of animals caught at very different locations. In the context of general random effects models

this is called the intra-class correlation or, for marine fish surveys, the intra-haul correlation (Pennington and Volstad, 1991, 1994) and is typically taken into account by using the discrete sampling location as a random effect. Therefore, a random year–location interaction was also included in the model to account for correlation between observations from the same year at each location.

The saturated model of the change in concentration of the contaminants (polychlorinated biphenyls, DDE, hexachlorobenzene, hexachlorocyclohexanes, chlordanes, toxaphenes, arsenic, cadmium, copper, selenium and zinc in cod liver) is as follows:

$$y_{ijt}=\beta_{0j}+\beta_{1j}t+\beta_{2j}t^2+\beta_{3j}t^3+\beta_{4j}t^4+\beta_{5j}a_{ijt}$$
$$+\beta_{6j}le_{ijt}+\beta_{7j}f_{ijt}+\beta_8 lw_{ijt}+\gamma_{jt}+\epsilon_{ijt}$$
$$\gamma\sim N(0,G)$$
$$\epsilon\sim N(0,\sigma^2 I)$$

where $y_{ijt}$ is the log-concentration of a contaminant in sub-sample $i$ at location $j$ in year $t$, $a_{ijt}$ is the average age, $le_{ijt}$ is the average length, $f_{ijt}$ is the average fat content of the livers and $lw_{ijt}$ is the log of the average liver weight in the sub-samples. The $\gamma_{jt}$ is the random effect for the intra-haul correlation and the G matrix was fitted as a diagonal matrix, that is, the random effects were assumed independent. The model for mercury in cod muscle was different owing to the different data structure, it did not have a random term and only had age and length as covariates. The models were fitted with restricted maximum likelihood (REML) using the nlme package (Pinheiro et al., 2013) in the statistical software R (R Development Core Team R, 2013). Fixed effects were tested using the conditional t-test which is an approximate test (Pinheiro and Bates, 2000), terms not significant at α=0.05 were removed from the model.

### Summary of program results

A detailed statistical evaluation of the Icelandic monitoring program for the period 1991–2010 can be found in two publications by Sturludóttir and co-workers (Sturludóttir et al., 2013, 2014). For mussels, in general, there are no major changes over time for the inorganic trace elements. On the other hand, concentrations of persistent organic pollutants (*p,p*'-DDE, HCB, α-HCH, PCB-153 and *trans*-nonachlor) in mussels have decreased at most sampling locations in Iceland in recent years. However, an increasing trend was found at a few locations that could be explained by anthropogenic activity. In this regard three noteworthy events were observed. First, for one mussel sample site, Hvalstöð located north of Reykjavík in Hvalfjörður, the concentration of 4,4´-DDE and *trans*-nonachlor showed a sudden large increase in 2009 and 2010. In 2011, the concentration dropped again to a level similar to 2008. This was linked to a whale processing site near the mussel sampling site. Whales were hunted and processed in both 2009 and 2010, after a period of no whale hunting, whereas no whales were hunted in 2011. Even though whales are drained of blood at sea to cool the hunted animal as soon as possible, waste products still drain out of the animal at the processing site into the sea during the cutting of the whale. Minke whale (*Balaenoptera acutorostrata*) and fin whale (*B. physalus*) are the whale species hunted and processed at the site. Second, the arsenic concentration in mussels from Úlfsá, near Ísafjörður in the Westfjords, is very high compared to other sites investigated, generally three to four times higher than in mussels from other sites. The arsenic concentration showed an increasing trend in mussels from Úlfsá until 2001 and then a decreasing trend. The HCB concentration in mussels from Úlfsá was also higher than in mussels from other sampling sites, up to double the concentration. The HCB concentration also showed an increasing trend over the entire period investigated (2% annual change). Thus, HCB and arsenic show a different temporal trend at the Úlfsá site. For other analytes investigated, the results at Úlfsá are no different from other sampling sites. There was a waste incineration facility located near Úlfsá operating between 1994 and 2011 that was exempt from the current EU regulations covering waste incineration facilities. After dioxins were found in milk from a dairy farm nearby, the waste incineration facility was closed. It is unclear whether this site was the source of arsenic in mussels from Úlfsá, although it is likely that the higher HCB concentration was linked to the waste incineration facility. Further monitoring should reveal whether the HCB concentration decreases after the site closure 2011. The third event is a sharp decrease in cadmium in a fjord in eastern Iceland (Botn, Mjóifjörður) with an 11% annual decrease. There are two other sample sites in the same fjord, also showing an annual decrease of 8%, while the cadmium concentration in mussels from Botn (at the inner end of the fjord) was three to four times higher than in mussels from other sample sites at the start of the period investigated. There is no known industrial source of cadmium in Iceland, especially in Mjóifjörður which is a narrow fjord with about 50 inhabitants. The cadmium found in Iceland is generally considered to be from natural sources.

For the cod investigated, all POPs showed a decreasing trend. For the inorganic trace elements, arsenic showed an increasing trend during the study period with a 2% annual increase. Zinc showed a decreasing trend with a 1% annual decrease. Other elements showed some fluctuations in the trend but the average revealed no change.

### Conclusions

It is important to monitor environmental pollutants, even at sites considered non-impact sites with limited effects due to human activities, as changes are still expected to occur but just more slowly than at sites near known sources of pollutants. The example of Hvalstöð shows there are events that can lead to unforeseen impacts on environmental quality. In other cases, such as at Úlfsá, there are unknown sources of pollutants. Unfortunately, funding for the annual monitoring of the marine biosphere has decreased considerably over the past decade and no funds were allocated for this monitoring program during the period 2012–2015. However, funding is now available to analyze all the cod samples collected during the period when funding for analysis was not available and so, despite this gap in funding, the time-series for cod will be intact. Mussels were not collected during the non-funded years (2014 and 2015) so there will be a short gap in the mussel time series. Because the Icelandic monitoring program results indicate that the annual change in pollutant concentration is low, the sampling period needed for statistical evaluation is very long, which means interruptions in the time-series are challenging.

**Literature cited**

Jörundsdóttir, H., N. Desnica, Þ. Ragnarsdóttir and H. Gunnlaugsdóttir, 2013. Monitoring of the marine biosphere around Iceland 2011 and 2012. Matis report 22-13.

Pennington, M. and J.H. Volstad, 1991. Optimum size of sampling unit for estimating the density of marine populations. Biometrics, 47:717-723.

Pennington, M. and J.H. Volstad, 1994. Assessing the effect of intra-haul correlation and variable density on estimates of population characteristics from marine surveys. Biometrics, 50:725-732.

Pinheiro, J.C. and D.M. Bates, 2000. Mixed-effects models in S and S-PLUS. Springer Verlag.

Pinheiro, J.C., D.M. Bates, S. DebRoy, D. Sarkar and the R Development Core Team, 2012. nlme: Linear and Nonlinear Mixed Effects Models. R package version 3.1-103.

Pinheiro, J.C., D.M. Bates, S. DebRoy, D. Sarkar, and the R Development Core Team, 2013. nlme: Linear and Nonlinear Mixed Effects Models. R Package Version3:1-109.

R Development Core Team R, 2013. A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria.

Sturludóttir, E., H. Gunnlaugsdottir, H. Jörundsdottir, E. Magnusdottir, K. Ólafsdottir and G. Stefansson, 2013. Spatial and temporal trends of contaminants in mussel sampled around the Icelandic coastline. Science of the Total Environment, 454-455:500-509.

Sturludóttir, E., H. Gunnlaugsdóttir, H.Ó. Jörundsdóttir, E.V. Magnúsdóttir, K. Ólafsdóttir and G. Stefánsson, 2014. Temporal trends of contaminants in cod from Icelandic waters. Science of the Total Environment, 476-477:181-188.

Verbeke, G. and G. Molenberghs, 2000. Linear Mixed Models for Longitudinal Data. Springer Verlag.

# Annex 4. Description and interpretation of results from national trend studies: US-Alaska – Seabirds

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## Introduction

The Seabird Tissue Archival and Monitoring Project (STAMP) has banked over 1900 seabird eggs collected from Alaska since 1999. Common murre (*Uria aalge*) and thick-billed murre (*U. lomvia*) make ideal biomonitors as abundant, circumpolar, essentially non-migrating, piscivores that only lay one egg (Vander Pol and Becker, 2007). Glaucous gull (*Larus hyperboreus*) and glaucous-winged gull (*L. glaucescens*) egg clutch banking began in 2004 at the request of subsistence harvesters and to correlate with other circumpolar studies. STAMP began as a joint project of the US Fish and Wildlife Service Alaska Maritime National Wildlife Refuge (USFWS-AMNR), the US Geological Survey Biological Resources Division (USGS-BRD), and the National Institute of Standards and Technology (NIST) to monitor long-term trends in environmental quality by (1) collecting seabird tissues (mainly eggs) at Alaskan seabird colonies without inadvertently contaminating them, (2) processing and banking the samples under conditions that ensure chemical stability during long-term (decadal) storage, and (3) analyzing subsamples of the stored material for anthropogenic contaminants (see York et al., 2001). Egg collections and processing are conducted by USFWS-AMNR and collaborating researchers (including Bureau of Indian Affairs Alaska Regional Subsistence Branch, BIA/ARSB; and local subsistence harvesters), and the banking of egg contents and contaminant analyses are conducted by NIST in its facilities at the Hollings Marine Laboratory, Charleston, South Carolina. In addition, carbon and nitrogen isotope analysis for food-web determination is now being conducted by Environment and Climate Change Canada and all eggshells are being archived at the Museum of the North, University of Alaska in Fairbanks. Additional support has been provided by the North Pacific Research Board.

## Methods

Eggs are collected, measured and contents harvested using standard protocols (York et al., 2001; Rust et al., 2010). All egg contents are banked at the national Marine Environmental Specimen Bank (ESB) at NIST where until 2005 (for gull eggs) or 2008 (for murre eggs), the contents were cyohomogenized and aliquoted into Teflon jars (see York et al. 2001; Vander Pol et al., 2003, 2009a), after which the contents were fresh homogenized using various hand blenders and finally a bag-mixer (Interscience, St Nom la Bretêche, France) to increase the 'real-time analysis' potential into both Teflon jars and polypropylene cryovials to allow for analysis of perfluorinated chemicals (PFCs; see Vander Pol et al., 2009a,b, 2012; Rust et al., 2010). All aliquots are stored in liquid nitrogen vapor freezers and are available to other researchers under a formal access policy. Sample metadata are available[1].

Since 1999, 475 eggs have been analyzed for persistent organic pollutants (POPs) and mercury (Hg) by scientists at NIST to help advance the third goal of STAMP. Mercury analysis originally comprised microwave digestion followed by isotope dilution cold vapor inductively coupled plasma mass spectrometry (ID-CV-ICPMS; see Christopher et al., 2002; Day et al., 2006, 2012a). Since 2010, Hg analysis has moved to direct measurements (Milestone, Shelton, Connecticut; Bryan et al., in prep.) again to allow for more 'real-time analysis' potential. The creation and use of a control material (Vander Pol et al., 2007) ensures consistency between instrumental methods and batches (prior to the creation of this matrix-matched control material, SRM 1946 Lake Superior Fish Tissue was used). Likewise, for analysis of organic contaminants – polychlorinated biphenyls (PCBs), organochlorine pesticides, and since 2001 polybrominated diphenyl ethers (PBDEs) – methods have changed from pressurized fluid extraction (PFE) followed by size exclusion chromatography (SEC) and semi-preparative aminopropylsilane liquid chromatography (LC) fractionation with gas chromatography (GC) coupled to dual electron capture detectors (ECD; see Vander Pol et al., 2003, 2004), to PFE followed by SEC and solid phase extraction (SPE) with pressure temperature vaporization (PTV) GC coupled to mass spectrometer detector (MSD; see Vander Pol et al., 2009a,b, 2011, 2012) to decrease preparation time and increase analysis sensitivity.

Methylmercury (Davis et al., 2004), butyltin (Vander Pol et al., 2009a) and Hg isotopes (Point et al., 2011; Day et al., 2012a) have also been analyzed in selected samples. Stable carbon and nitrogen isotopes have been analyzed at Environment and Climate Change Canada to aid in the interpretation of the contaminant results (Vander Pol et al., 2011, 2012; Day et al. 2012a,b). Future work will include analysis of other heavy metals and trace elements and genetic analysis to distinguish eggs from unidentified murre species as these sympatric species nest in some of the same areas, and the eggs appear similar.

Data reported here have been analyzed using principal components analysis (PCA) for the compounds with no values below detection limit (PCB congeners CB28+CB31, CB66, CB99, CB105, CB118, CB138, CB146, CB153+CB132, CB163, CB170, CB180+CB193, CB183, CB187); 4,4'-DDE; oxychlordane; mirex; hexachlorobenzene, HCB; Hg). The compounds were summed for each egg and the proportion of that compound to the total was used for the PCA. The resulting eign value for each principal component (PC) compound was multiplied by the proportion of that compound to the sum in the egg. These values were summed to obtain the PC for each egg. Multivariate Analysis of Variances (MANOVA) was conducted to compare these compounds by combined region/species to limit type 1 errors. As the MANOVA was significant, individual analysis of variances (ANOVAs) and post-hoc Tukey HSD tests were used to further determine differences among

[1] Sample metadata are available at: http://batchgeo.com/map/2e4a316a78e6f5a19b0c56b81db323fb

colonies. All statistics were analyzed with JMP (SAS Institute, Cary, North Carolina) software and plotted using SigmaPlot (SPSS Inc., Chicago, Illinois) software. In addition to the PIA temporal analysis, means and standard errors of the colony data per year were plotted against the Pacific Decadal Oscillation (PDO; http://jisao.washington.edu/pdo/PDO.latest) index mean and standard error between February and May (when the eggs should be forming) to compare changes observed with oceanographic changes.

## Summary of program results

Based on the PCA, Norton Sound murre eggs were generally separated from the Bering and Chukchi seas eggs due to higher proportions of Hg compared to mid-chlorinated PCBs (PC1 explained 41.1% of the total variation; Fig. A4.1). The gull eggs are also slightly separated along PC1, but are also dominated by higher proportions of heavier chlorinated PCBs, similar to Gulf of Alaska murre eggs, which are generally separated from Being and Chukchi seas murre eggs along PC2 (24.3% of the total variation). PC2 appears to follow the global distillation/fractionation theory of heavier chemicals dominating lower latitudes and lighter chemicals moving toward the polar regions.

The MANOVA comparing region/species combinations was highly significant (Wilk's $\lambda = 0.0123, F_{234,4269} = 9.40, p < 0.0001$). All of the post-hoc ANOVAs were also highly significant ($p < 0.0001$). The 3-Cl PCBs were higher in the Chukchi Sea region and Norton Sound glaucous gull eggs and lower in the Norton Sound common murre and Bering Sea and Gulf of Alaska common and thick-billed murre eggs. The 7-Cl PCBs were highest in the Chukchi Sea glaucous gull and Gulf of Alaska thick-billed murre eggs and lowest in the Aleutian Islands thick-billed murre and Norton Sound and Bering Sea murre eggs (see Table A4.1). The Gulf of Alaska thick-billed murre eggs had the highest levels of 4,4'-DDE while the Bering Sea and Norton Sound murre eggs had the lowest. Eggs from these regions/species also had the lowest oxychlordane levels while the greatest were in the Chukchi Sea glaucous gull eggs. HCB is a lighter compound, and the lowest levels were observed in the Gulf of Alaska thick-billed and glaucous-winged gull eggs, while the highest were in the Chukchi Sea murre eggs.

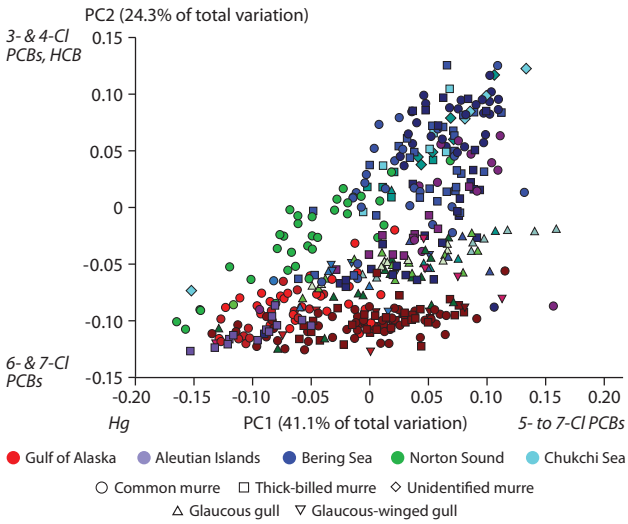


Figure A4.1 Principal Components Analysis (PCA) for Alaskan seabird eggs collected between 1999 and 2010.

Table A4.1 Post-hoc Tukey HSD results following highly significant ($p<0.0001$) ANOVAs. Region/species that do not have overlapping letters were statistically ($p<0.05$) different.

| Region/Species | 3-Cl PCBs (28+31) | 4-Cl PCBs (66) | 5-Cl PCBs (99, 105, 118) | 6-Cl PCBs (138, 146, 153+132, 163) | 7-Cl PCBs (170, 180+193, 183, 187) | 4,4'-DDE | Oxychlordane | HCB | Mirex | Mercury | ΣPCBs | Σpesticides | Σcontaminants | BDE47 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Chukchi Sea | | | | | | | | | | | | | | |
| thick-billed murre | A | A-E | C-E | C-F | CD | EF | D-F | AB | C-G | C-F | C-F | C-E | B-D | BC |
| unidentified murre sp. | A | A-C | C-E | C-F | CD | D-F | C-E | A | B-E | C-F | D-F | B-E | B-D | BC |
| glaucous gull | A | A | A | A | AB | EF | A | B-D | A | A-D | A | AB | A | A |
| Norton Sound | | | | | | | | | | | | | | |
| glaucous gull | A | A | B | B | A-C | BC | B | C-G | A | A | B | A-C | A | BC |
| common murre | B | E | D-E | E-F | D | F | F | C-E | C-G | BC | EF | E | CD | C |
| Bering Sea | | | | | | | | | | | | | | |
| glaucous-winged gull | AB | A-D | B-D | B-E | A-D | B-F | BC | C-G | A-C | AB | B-F | A-E | A-C | BC |
| thick-billed murre | B | C-E | DE | E-F | D | F | F | C-G | F | EF | EF | DE | D | C |
| common murre | B | D-E | E | F | D | F | F | C | E-G | F | F | DE | D | C |
| Aleutian Islands | | | | | | | | | | | | | | |
| glaucous-winged gull | B | A-E | B-D | A-D | A-D | A-E | B-F | E-G | AB | A-F | A-E | A-E | A-D | BC |
| thick-billed murre | B | A-D | C-E | D-F | D | B-E | D-F | C-G | D-G | A | D-F | B-D | AB | BC |
| common murre | AB | B-E | C-E | C-F | CD | C-F | D-F | BC | FG | D-F | C-F | C-E | CD | BC |
| Gulf of Alaska | | | | | | | | | | | | | | |
| glaucous-winged gull | AB | AB | B-E | B-F | A-D | A-D | C-F | FG | C-G | A-E | B-F | A-E | A-C | A |
| thick-billed murre | B | A-C | C | BC | AB | A | D-F | G | CD | AB | BC | A | A | B |
| common murre | B | B-D | C | CD | BC | B | D-F | D-G | C-G | A | CD | BC | A | BC |

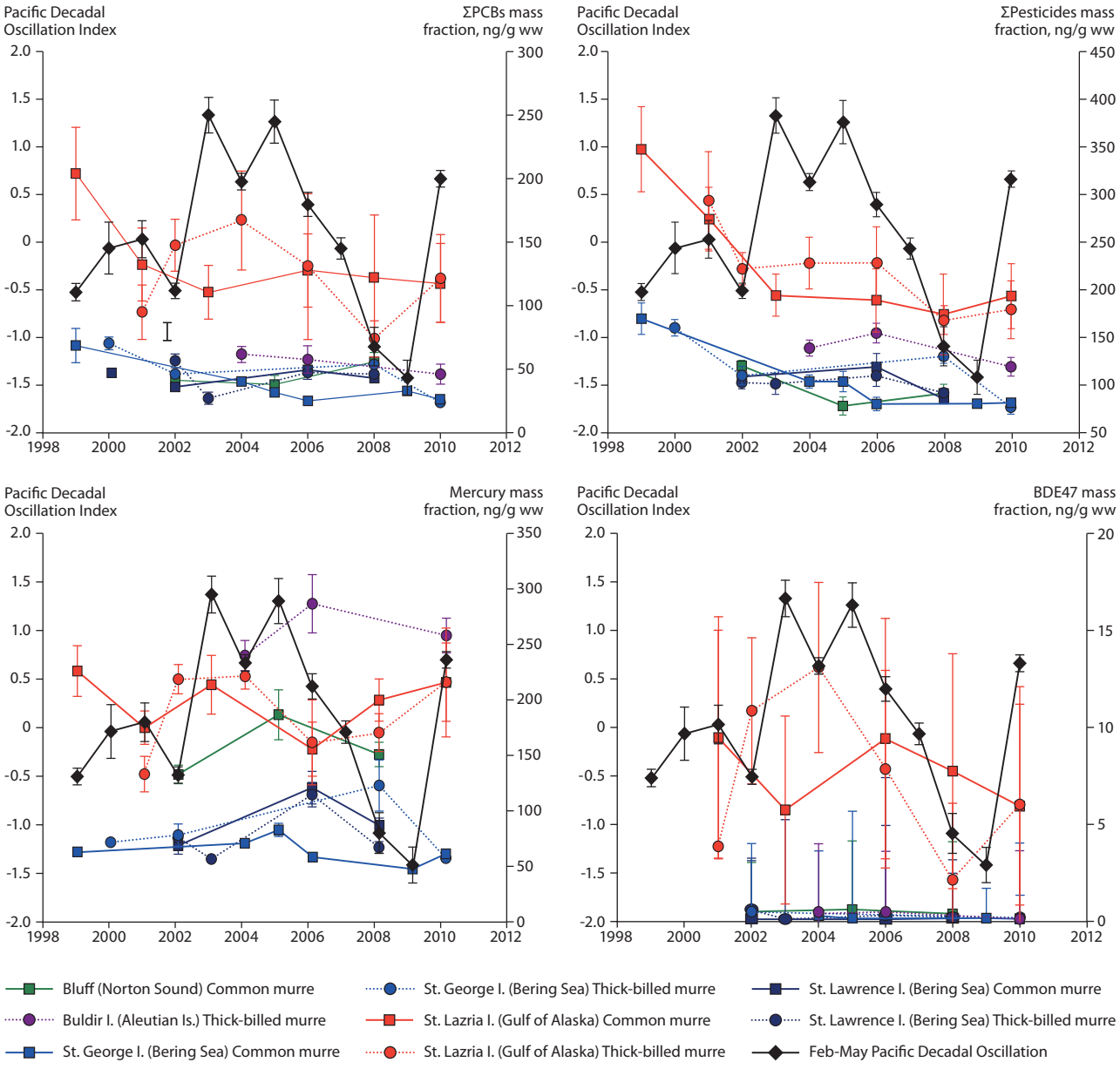


Figure A4.2 Mean concentrations (and standard errors) for a range of contaminants in Alaskan murre (*Uria* spp.) eggs compared to the February–May Pacific Decadal Oscillation.

The highest levels of mirex were in the Chukchi Sea and Norton Sound glaucous gull eggs while the lowest were in the Bering Sea and Aleutian Islands murre eggs. Mercury was highest in the Norton Sound glaucous gull, Aleutian Islands thick-billed murre, and Gulf of Alaska common murre eggs and lowest in the Bering Sea murre eggs. While, BDE47 was not included in the MANOVA due to missing values for the 1999–2001 eggs, a separate ANOVA was conducted for this important compound. The highest levels were in Chukchi Sea glaucous gull and Gulf of Alaska glaucous-winged gull eggs, and lowest in the Bering Sea and Norton Sound murre eggs. Overall, the highest contaminant levels were found in the Chukchi Sea and Norton Sound glaucous gull and Gulf of Alaska murre eggs and the lowest in the Bering Sea murre eggs.

As species differences, especially at St. Lazaria Island in the Gulf of Alaska, seemed to vary over the past decade (see Fig. A4.2), potential relationships with oceanographic conditions (probably affecting prey availability) were examined. Several contaminants in thick-billed murre eggs at St. Lazaria Island generally correlated with the spring (February–May, when eggs should be forming) PDO indices, while common murre eggs at this location exhibited an opposing trend. None of the other long-term monitoring colonies exhibited this correlation with the PDO. However, this could be due to a lack of time sampling points. Possible explanations for the correlation are being examined and will be published once all the carbon and nitrogen stable isotope data are available.

## Conclusions

The Seabird Tissue Archival and Monitoring Project –STAMP – has been successfully banking and monitoring seabird eggs in the Alaskan region since 1999. Aliquots of the eggs are available to other researchers. Contaminant patterns and concentrations differ by region and species and some of the temporal trends appear to be correlated with the PDO. STAMP will continue to use murre eggs to monitor changing contaminant trends in this region.

**Literature cited**

Christopher, S.J., S.S. Vander Pol, R.S. Pugh, R.D. Day and P.R. Becker, 2002. Determination of mercury in the eggs of common murres (*Uria aalge*) for the seabird tissue archival and monitoring project. Journal of Analytical Atomic Spectrometry, 17:780-785.

Davis, W.C., S.S. Vander Pol, M.M. Schantz, S.E. Long and S.J. Christopher, 2004. An accurate and sensitive method for the determination of methylmercury in biological specimens using GC-ICPMS with solid phase microextraction. Journal of Analytical Atomic Spectrometry, 19:1546-1551.

Day, R.D., S.S. Vander Pol, S.J. Christopher, W.C. Davis, R.S. Pugh, K.S. Simac, D.G. Roseneau and P.R. Becker, 2006. Murre eggs (*Uria aalge* and *Uria lomvia*) as indicators of mercury contamination in the Alaskan marine environment. Environmental Science and Technology, 40:659-665.

Day, R.D., D.G. Roseneau, S. Berail, K.A. Hobson, O.F.X. Donard, S.S. Vander Pol, R.S. Pugh, A.J. Moors, S.E. Long and P.R. Becker, 2012a. Mercury stable isotopes in seabird eggs reflect a gradient from terrestrial geogenic to oceanic mercury reservoirs. Environmental Science and Technology, 46:5327-5335.

Day, R.D., D.G. Roseneau, S.S. Vander Pol, K.A. Hobson, O.F.X. Donard, R.S. Pugh, A.J. Moors and P.R. Becker, 2012b. Regional, temporal, and species patterns of mercury in Alaskan seabird eggs: Mercury sources and cycling or food web effects? Environmental Pollution, 166:226-232.

Point, D., J.E. Sonke, R.D. Day, D.G. Roseneau, K.A. Hobson, S.S. Vander Pol, O.F.X. Donard and P.R. Becker, 2011. Methylmercury photodegradation influenced by sea-ice cover in Arctic marine ecosystems. Nature Geoscience, 4:188-194.

Rust, L.B., R.S. Pugh, A.J. Moors, S.S. Vander Pol, D.G. Roseneau and P.R. Becker, 2010. Seabird Tissue Archival and Monitoring Project: Project Overview and Updated Protocols for Collecting and Banking Seabird Eggs. National Institute of Standards and Technology, NISTIR 7678.

Vander Pol, S.S. and P.R. Becker, 2007. Monitoring contaminants in seabirds: The importance of specimen banking. Marine Ornithology, 35:113-118.

Vander Pol, S.S., S.J. Christopher, D.G. Roseneau, P.R. Becker, R.D. Day, J.R. Kucklick, R.S. Pugh, K.S. Simac and G.W. York, 2003. Seabird Tissue Archival and Monitoring Project: Egg Collections and Analytical Results for 1999–2002. National Institute of Standards and Technology, NISTIR 7029.

Vander Pol, S.S., P.R. Becker, J.R. Kucklick, R.S. Pugh, D.G. Roseneau, K.S. Simac and G.W. York, 2004. Persistent organic pollutants in Alaskan murre (*Uria* spp.) eggs: geographical, species, and temporal comparisons. Environmental Science and Technology, 38:1305-1312.

Vander Pol, S.S., M.B. Ellisor, R.S. Pugh, P.R. Becker, D.L. Poster, M.M. Schantz, S.D. Leigh, B.J. Wakeford, D.G. Roseneau and K.S. Simac, 2007. Development of a murre (*Uria* spp.) egg control material. Analytical and Bioanalytical Chemistry, 387:2357-2363.

Vander Pol, S.S., P.R. Becker, R.D. Day, M.B. Ellisor, A. Guichard, A.J. Moors, D. Point, R.S. Pugh and D.G. Roseneau, 2009a. Seabird Tissue Archival and Monitoring Project: Egg Collections and Analytical Results for 2002–2005. National Institute of Standards and Technology, NISTIR 7562.

Vander Pol, S.S., P.R. Becker, M.B. Ellisor, A.J. Moors, R.S. Pugh and D.G. Roseneau, 2009b. Monitoring organic contaminants in eggs of glaucous and glaucous-winged gulls (*Larus hyperboreus* and *Larus glaucescens*) from Alaska. Environmental Pollution, 157:755-762.

Vander Pol, S.S., K.A. Hobson, P.R. Becker, R.D. Day, M.B. Ellisor, R.S. Pugh and D.G. Roseneau, 2011. Geographic differences in organic contaminants and stable isotopes ($\delta^{13}C$, $\delta^{15}N$) in thick-billed murre (*Uria lomvia*) eggs from Alaska. Journal of Environmental Monitoring, 13:699-705.

Vander Pol, S.S., P.R. Becker, S. Berail, R.D. Day, O.F.X. Donard, K.A. Hobson, A.J. Moors, R.S. Pugh, L.B. Rust and D.G. Roseneau, 2012. Seabird Tissue Archival and Monitoring Project: Egg Collections and Analytical Results for 2006-2009. National Institute of Standards and Technology, NISTIR 7872.

York, G.W., B.J. Porter, R.S. Pugh, D.G. Roseneau, K. Simac, P.R. Becker, L. K. Thorsteinson and S.A. Wise, 2001. Seabird Tissue Archival and Monitoring Project: Protocol for Collecting and Banking Seabird Eggs. National Institute of Standards and Technology, NISTIR 6735.

# Annex 5. Description and interpretation of results from national trend studies: Greenland – Marine mammals, seabirds, freshwater fish

*Authors:* Frank Rigét

## Introduction

Monitoring of contaminants in biota is conducted as a part of the Danish/Greenlandic AMAP implementation program funded by the Danish Cooperation for Environment in the Arctic of the Danish Environmental Protection Agency. The program was initiated in 1994 and the contaminants and species selected mainly follow AMAP recommendations. The program constitutes three parts; continuous monitoring of selected contaminants, screening studies of emerging compounds, and retrospective studies based on tissue bank samples. The program design has changed over time, particularly by switching from sampling every five years to sampling every two years.

## Methods

The following examination of temporal trends in Greenlandic biota is based on the time trend analyses done during this assessment. The statistical methods applied are described in Sect. 3. For many of the time-series, parts were analyzed in more depth by taking into account covariates such as stable isotopes and climatic indices (see cited literature). In this assessment, marine mammals were divided into two age groups in order to minimize the effect of age accumulation and also to avoid modeling or making assumptions about age accumulation relationships. In the case of polar bears (*Ursus maritimus*), sex was accounted for by dividing adults into males and females.

## Time-series available

Time-series of legacy persistent organic pollutant (POP) concentrations are available for landlocked Arctic char (*Salvelinus alpinus*) from southwestern Greenland, ringed seals (*Pusa hispida*) from western and eastern Greenland, and black guillemot (*Cepphus grylle*) and polar bears, both from eastern Greenland. Furthermore, a retrospective study of POPs in eggs of peregrine falcon (*Falco peregrinus*) from southern Greenland covering the period 1986 to 2003 has been reported (Vorkamp et al., 2009, 2014), which reports trends in accordance with those described here. The time-series extend back to the mid-1990s, however polar bears and ringed seal from eastern Greenland also include data from the mid-1980s. The number of years included in the time-series thus varies between 7 and 25. Of 183 full time-series, 28% and 34% had a statistical power of more than 80% and 70%, respectively, to detect a potential annual change of 5%. In general, the time-series for young ringed seal and polar bear were more powerful than those for adults probably owing to the larger number of samples each year and to greater individual variation in adults than juveniles. The results from these times-series (or parts of time-series) have been published in the scientific literature and associations with climate change, feeding habits and biological variables were examined: Arctic char (Rigét et al., 2010), ringed seal (Rigét et al., 2008, 2013a, 2016; Vorkamp et al., 2008, 2011), and polar bear (Dietz et al., 2013a). For the present POPs assessment, the time-series have been updated with data for the most recent year(s).

## Summary of program results

### *Dieldrin*

The only time-series for dieldrin in Greenland biota are for polar bears. These show significantly decreasing trends for juveniles and adult females and no trend for adult males.

### *CHLs*

The time-series of ΣCHL (*cis*-chlordane, *trans*-chlordane, *cos*-nonachlor, *trans*-nonachlor, oxychlordane), *trans*-nonachlor and oxychlordane in Arctic char, and juvenile ringed seal from western and eastern Greenland show significantly decreasing trends. The annual decrease ranges from 4% to 7%. This is also the case for the time-series since 2000. For polar bear time-series (juvenile, adult female and adult male) significantly decreasing trends are only found for *trans*-nonachlor in juvenile and adult females. The trends for oxychlordane since 2000 in polar bears are increasing but are only significant for adult females.

### *PCBs and DDTs*

Most of the polychlorinated biphenyl (PCB) and DDT time-series from the mid-1980s show significantly decreasing trends, with fewer decreasing trends found in time series sets extending back to 2000. The annual decrease for the sum of ten PCB congeners (CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156, CB180; $\Sigma PCB_{10}$) and the sum of DDTs (*p,p*-DDE, *p,p*-DDD, *p,p*-DDT; ΣDDTs) was very similar and ranged from less than 1% to 11% with the lowest decrease in adult male polar bears and the highest decrease in landlocked Arctic char. For time-series starting in 2000 the decrease was lower and increasing trends were even found for $\Sigma PCB_{10}$ in juvenile, adult male and adult female polar bears and eggs of black guillemot. This positive trend observed in recent years in polar bears may be influenced by changing feeding habits, i.e. switching from Arctic seal species to more sub-Arctic seal species during warmer periods (Dietz et al., 2013a; McKinney et al., 2013). Comparing the $\Sigma PCB_{10}$ and DDTs time-series for ringed seal from western and eastern Greenland, showed a tendency for higher decreases in ringed seal from western Greenland.

### *HCB*

The results of the hexachlorobenzene (HCB) trend analyses in Greenland are more ambiguous as some time-series show significant increases (adult male polar bears and black guillemot), while others (Arctic char and juvenile seals from western and eastern Greenland) show significant decreases. In terms of time-series since 2000, both significant increases and decreases are found. Therefore, no firm conclusions can be drawn for temporal trends of HCB in Greenland biota.

*HCHs*

All time-series of α-hexachlorocyclohexane (α-HCH) show significantly decreasing trends with annual decreases of 10–13%. The time-series since 2000 show even higher annual decreases of 11–20%. This contrasts with β-HCH where the annual decrease was 1–5% and only significant for the longest time-series. The annual decrease of γ-HCH was between those for α-HCH and β-HCH ranging from 4% to 7%. This difference between HCH isomers is probably caused by different pollutant transfer pathways to Greenland as a consequence of different physical and chemical properties (Rigét et al., 2008).

*Toxaphene*

Time-series for concentrations of toxaphene congeners Parlar 26 and Parlar 50 in Arctic char and ringed seal from western and eastern Greenland and black guillemot in general show decreasing trends, both for the full time-series and for those since 2000. The annual decrease ranges from no change to 10%.

*PBDEs*

Time-series of polybrominated diphenyl ethers (PBDEs) in Greenland biota are available for ringed seals from western and eastern Greenland and polar bears from eastern Greenland. A retrospective study of POPs in peregrine falcon eggs from southern Greenland for the period 1986 to 2003 and generally showing increasing concentrations is also available (Vorkamp et al., 2005).

The results from the analyses of PBDE trends are ambiguous. In western Greenland, ringed seal had a significantly increasing trend for BDE47, while a significantly decreasing trend was found for ringed seal in eastern Greenland. For juvenile and adult female polar bear, both BDE47 and BDE99 peaked in the years around 2000, which is reflected by the significant non-linear trend component (Dietz et al., 2013b). However, BDE47 concentrations appear to have increased in recent years, which may be due to changing food availability caused by declining sea ice cover (Dietz et al., 2013a; McKinney et al., 2013).

*HBCDD*

Time-series of hexabromocyclododecane (HBCDD) concentrations are available for glaucous gull (1994–2012), ringed seal (1986–2012) and polar bear (1983–2012) from eastern Greenland (Vorkamp et al., 2012; Dietz et al., 2013b). A significantly increasing trend is found for ringed seal and polar bear with an annual increase of 4.5% to 14.5%. For glaucous gull a significant non-linear trend component was found caused by a relatively high annual median in one year in the middle of the time-series, which may be an outlier year. The Mann-Kendall trend test showed a significant decreasing trend.

*PFASs*

Levels of perfluorinated alkylated substances (PFASs) in ringed seals from western and eastern Greenland and young polar bears from eastern Greenland have been monitored for temporal trends. The time-series extend back to the early 1980s and include seven to eight years of data for ringed seals and 23 years of data for polar bears. Rigét et al. (2013b) analyzed these time-series and found that several PFASs in ringed seals and polar bears peaked around 2006. Adding one more data point to these time-series for 2012 further highlights this. Temporal trend analyses for perfluorooctane sulfonic acid (PFOS) shows a significant non-linear trend component reflecting the previously increasing and now decreasing trend. Data for the other PFASs in ringed seals do not allow a statistical analysis of the temporal trend as too many years have annual median concentrations below detection limits. For polar bear the time-series of PFASs in juveniles are considered more reliable than the adult (male and female) time-series, because the time-series are much longer and comprise a larger sample size each year. For juvenile polar bears the non-linear trend component was also significant for perfluorooctanesulfonamide (PFOSA) and perfluorodecanoate (PFDA) indicating that these peaked similarly to PFOS. Perfluorooctanoic acid (PFOA), perfluorononanoate (PFNA), perfluroundecanoic acid (PFUnA), perfluorododecanoic acid (PFDoA) and perfluorotridecanoic acid (PFTrA) all show significant increases during the period from 1984 to 2012.

*Octachlorostyrene*

The only time-series for octachlorostyrene (OCS) in Greenland biota are for polar bears. In juvenile and adult males the non-linear trend components were significant while no trend was found for adult females. The trend curves show a wave-like pattern with periods of small increases followed by periods of small decreases.

## Conclusions

The main part of the legacy POPs time-series in Greenlandic biota show a significantly decreasing trend. The temporal trends in Greenlandic biota for brominated flame retardants are ambiguous as both increasing and decreasing trends are found. Furthermore, some time-series indicated that brominated flame retardants peaked in the years around 2000. Several perfluorinated alkylated substances (PFOS, PFHxS, PFOSA) peaked in ringed seals and polar bears from Greenland around 2006, while for long-chain perfluorinated carboxylic acids the trends are less clear. It is important to continue monitoring these compounds, including the legacy POPs although it may be argued that the international regulatory actions have worked successfully and that concentrations are likely to decrease even more in the future. However, the time-series now cover such a long period that the impacts of mechanisms such as climate change and food web structure on temporal trends can be investigated. Continuing the monitoring will increase understanding of these interactions, which benefits the future management of contaminants in Arctic ecosystems.

## Literature cited

Dietz, R., F.F. Rigét, C. Sonne, E.W. Born, T. Bechshøft, M.A. McKinney and R.J. Letcher, 2013a. Three decades (1983-2010) of contaminant trends in East Greenland polar bears (*Ursus maritimus*). Part 1: Legacy organochlorine contaminants. Environment International, 59:485-493.

Dietz, R., F.F. Rigét, C. Sonne, E.W. Born, T. Bechshøft, M.A. McKinney, R.J. Drimmie and R.J. Letcher, 2013b. Three decades (1983-2010) of contaminant trends in East Greenland polar

bears (*Ursus maritimus*). Part 2: Brominated flame retardants. Environment International, 59:494-500.

McKinney, M.A., S.J. Iverson, A.T. Fisk, C. Sonne, F.F. Rigét, R.J. Letcher, M.T. Arts, E.W. Born, A. Rosing-Asvid and R. Dietz, 2013. Global change effects on the long-term feeding ecology and contaminant exposures of East Greenland polar bears. Global Change Biology, 19:2360-2372.

Rigét, F., K. Vorkamp, R. Dietz and D.C.G. Muir, 2008. Levels and temporal trends of HCH isomers in ringed seals from West and East Greenland. Journal of Environmental Monitoring, 10:935-940.

Rigét, F., K. Vorkamp and D. Muir, 2010. Temporal trends of contaminants in Arctic char (*Salvelinus alpinus*) from a small lake, southwest Greenland during a warming climate. Journal of Environmental Monitoring, 12:2252-2258.

Rigét, F., K. Vorkamp, K.A. Hobson, D.C.G. Muir and R. Dietz, 2013a. Temporal trends of selected POPs and potential influence of climate variability in a Greenland ringed seal population. Environmental Science: Processes and Impact, 15:1706-1716.

Rigét, F., R. Bossi, C. Sonne, K. Vorkamp and R. Dietz, 2013b. Trends of perfluorochemicals in Greenland ringed seals and polar bears: Indications of shifts to decreasing trends. Chemosphere, 93:1607-1614.

Rigét, F., K. Vorkamp, R. Bossi, C. Sonne, R.J. Letcher and R. Dietz, 2016. Twenty years of monitoring of persistent organic pollutants in Greenland biota. A review. Environmental Pollution, 217:114-123.

Vorkamp, K., M. Thomsen, K. Falk, H. Leslie, S. Møller and P.B. Sørensen, 2005. Temporal development of brominated flame retardants in peregrine falcon (*Falco peregrinus*) eggs from South Greenland (1986-2003). Environmental Science and Technology, 39:8199-8206.

Vorkamp, K., F.F. Rigét, M. Glasius, D.C.G. Muir and R. Dietz, 2008. Levels and trends of persistent organic pollutants in ringed seals (*Phoca hispida*) from central West Greenland, with particular focus on polybrominated diphenyl ethers (PBDEs). Environment International, 34:499-508.

Vorkamp, K, M. Thomsen, S. Møller, K. Falk and P.B. Sørensen, 2009. Persistent organochlorine compounds in peregrine falcon (*Falco peregrinus*) eggs from South Greenland: levels and temporal changes between 1986 and 2003. Environment International, 35:336-341.

Vorkamp, K., F.F. Rigét, R. Bossi and R. Dietz, 2011. Temporal trends of hexabromocyclododecane, polybrominated diphenyl ethers and polychlorinated biphenyls in ringed seals from East Greenland. Environmental Science and Technology, 45:1243-1249.

Vorkamp, K., K. Bester and F.F. Rigét, 2012. Species-specific time trends and enantiomer fractions of hexabromocyclododecane (HBCD) in biota from East Greenland. Environmental Science and Technology, 46:10549-10555.

Vorkamp, K., S. Møller, K. Falk, F.F. Rigét, M. Thomsen and P.B. Sørensen, 2014. Levels and trends of toxaphene and chlordane-related pesticides in peregrine falcon eggs from South Greenland. Science of the Total Environment, 468-469:614-621.

# Annex 6. Overview of trend results for POPs in air and biota

Table A6.1 First order half-lives ($t_{1/2}$, y) for the four long-term air monitoring stations under AMAP. Negative values indicate increasing trends.

| POP | Alert | Alert | Pallas | Stórhöfði | Zeppelin |
|---|---|---|---|---|---|
| | 1993–2012 | | 1996–2012 | 1995–2012 | 1993–2012 |
| Dieldrin | | | | | |
| $t_{1/2}$ | 15.0 | | - | 24.6 | - |
| $r^2$ | 0.71 | | - | 0.34 | - |
| Endrin | | | | | |
| $t_{1/2}$ | 15 | | - | - | - |
| $r^2$ | 0.71 | | - | - | - |
| *cis*-Chlordane | | | | | |
| $t_{1/2}$ | 20 | | 11 | 13 | 14 |
| $r^2$ | 0.51 | | 0.75 | 0.40 | 0.95 |
| *trans*-Chlordane | | | | | |
| $t_{1/2}$ | 11 | | 9.7 | 6.2 | 9.3 |
| $r^2$ | 0.63 | | 0.81 | 0.59 | 0.98 |
| *cis*-Nonachlor | | | | | |
| $t_{1/2}$ | 26 | | - | - | 9.9 |
| $r^2$ | 0.19 | | - | - | 0.85 |
| *trans*-Nonachlor | | | | | |
| $t_{1/2}$ | 19 | | 13 | - | 17 |
| $r^2$ | 0.44 | | 0.71 | - | 0.84 |
| *o,p*'-DDT | | | | | |
| $t_{1/2}$ | ND | | - | ND | 8.5 |
| $r^2$ | - | | - | - | 0.83 |
| *o,p*'-DDE | | | | | |
| $t_{1/2}$ | ND | | - | - | 6.0 |
| $r^2$ | - | | - | - | 0.69 |
| *o,p*'-DDD | | | | | |
| $t_{1/2}$ | ND | | - | - | 8.3 |
| $r^2$ | - | | - | - | 0.85 |
| *p,p*'-DDT | | | | | |
| $t_{1/2}$ | ND | | 15 | 4.8 | 5.0 |
| $r^2$ | - | | 0.57 | 0.45 | 0.92 |
| *p,p*'-DDE | | | | | |
| $t_{1/2}$ | ND | | 56 | 9.8 | 8.0 |
| $r^2$ | - | | 0.22 | 0.45 | 0.74 |
| *p,p*'-DDD | | | | | |
| $t_{1/2}$ | ND | | ND | 5.1 | 8.8 |
| $r^2$ | - | | - | 0.67 | 0.63 |
| Heptachlor epoxide | | | | | |
| $t_{1/2}$ | 26 | | - | - | - |
| $r^2$ | 0.38 | | - | - | - |
| HCB | | | | | |
| $t_{1/2}$ | 26 | | - | 19 | ND |
| $r^2$ | 0.71 | | - | 0.34 | - |

| POP | Alert | Alert | Pallas | Stórhöfði | Zeppelin |
|---|---|---|---|---|---|
| | 1993–2012 | | 1996–2012 | 1995–2012 | 1993–2012 |
| α-HCH | | | | | |
| $t_{1/2}$ | 4.9 | | 5.8 | 5.4 | 4.9 |
| $r^2$ | 0.97 | | 0.97 | 0.94 | 0.99 |
| γ-HCH | | | | | |
| $t_{1/2}$ | 4.4 | | 4.3 | 7.3 | 4.1 |
| $r^2$ | 0.98 | | 0.97 | 0.78 | 0.97 |
| CB28 | | | | | |
| $t_{1/2}$ | 7.1[c] | 39[d,e] | 15 | 24 | 8.2[f] |
| $r^2$ | 0.76[c] | 0.07[d] | 0.64 | 0.23 | 0.68[f] |
| CB52 | | | | | |
| $t_{1/2}$ | 4.6[c] | 9.8[d] | 17 | -8.7 | 4.4[f] |
| $r^2$ | 0.81[c] | 0.60[d] | 0.62 | 0.27 | 0.71[f] |
| CB101 | | | | | |
| $t_{1/2}$ | 6.9[c] | 35[d] | 14 | -6.2 | 11[f] |
| $r^2$ | 0.77[c] | 0.40[d] | 0.76 | 0.65 | 0.90[f] |
| CB118 | | | | | |
| $t_{1/2}$ | 8.3[c] | 23[d] | 11 | - | 6.1[f] |
| $r^2$ | 0.28[c] | 0.21[d] | 0.80 | - | 0.91[f] |
| CB138 | | | | | |
| $t_{1/2}$ | 20[c] | -[d,g] | 10.0 | 15 | 5.1[f] |
| $r^2$ | 0.17[c] | - | 0.66 | 0.20 | 0.60[f] |
| CB153 | | | | | |
| $t_{1/2}$ | 8.3[c] | 16[d] | 11.0 | 27 | 6.2[f] |
| $r^2$ | 0.32[c] | 0.14[d] | 0.72 | 0.09 | 0.84[f] |
| CB180 | | | | | |
| $t_{1/2}$ | 3.6[c] | -17[d] | 5.9 | 8.4 | 4.4[f] |
| $r^2$ | 0.84[c] | 0.10[d] | 0.71 | 0.35 | 0.81[f] |
| α-endosulfan | | | | | |
| $t_{1/2}$ | 19 | 5[a] | ND[b] | - | - |
| $r^2$ | 0.39 | 0.86[a] | - | - | - |
| BDE47 | | | | | |
| $t_{1/2}$ | ND[h] | | 4.4[i] | 3.7[j] | 2.6[k] |
| $r^2$ | - | | 0.79[i] | 0.94[j] | 0.98[k] |
| BDE99 | | | | | |
| $t_{1/2}$ | 19[h] | | 2.9[i] | - | 2.6[k] |
| $r^2$ | 0.40[h] | | 0.96[i] | - | 0.92[k] |
| BDE100 | | | | | |
| $t_{1/2}$ | ND[h] | | 4.0[i] | - | 4.1[k] |
| $r^2$ | - | | 0.89[i] | - | 0.97[k] |
| BDE138 | | | | | |
| $t_{1/2}$ | 7.0[h] | | - | - | ND[k] |
| $r^2$ | 0.19[h] | | - | - | - |

ND: Trend cannot be determined. [a]2004–2012; [b]1996–2012 and 2004–2012; [c]1993–2001; [d]2003–2012; [e]after 2003, CB28 coelutes with CB31; [f]1998–2012; [g]after 2003, CB138 coelutes with CB163 and CB164; [h]2002–2012; [i]2003-2012; [j]2007–2012; [k]mid-2006–2012.

Table A6.2 Summary of trend results (mean and range of annual change, %) for time-series starting before 2000.

| | Dieldrin | *trans*-Nonachlor | ΣCHL | *p,p'*-DDE | ΣDDT | Heptachlor-expoxide | HCB | α-HCH | β-HCH | γ-HCH |
|---|---|---|---|---|---|---|---|---|---|---|
| Blue mussel | | 2.6 (-4.9 to 10.3) | | -0.9 (-8.3 to 8.4) | 1.1 (-10.9 to 11.3) | | 0.03 (-6.2 to 4.0) | -8.3 (-14.8 to -0.8) | | -9.7 |
| Arctic char | -6.0 (-8.3 to -3.8) | -8.3 | -6.0 (-9.5 to -0.8) | -6.9 (-9.5 to -1.6) | -8.2 (-10.3 to -2.1) | | -3.0 (-6.4 to 1.0) | -11.3 (-16.5 to -7.9) | 1.7 (-3.3 to 6.4) | -9.9 (-15.5 to -5.7) |
| Burbot | -3.4 (-8.0 to 0.8) | -5.4 | -5.4 (-6.7 to -4.1) | -1.4 (-3.7 to 1.0) | -3.4 (-5.7 to 0.5) | | -2.4 (-5.0 to 0.5) | -10.7 (-13.1 to -7.5) | -3.4 | -9.9 (-12.1 to -6.3) |
| Atlantic cod | | -5.0 (-7.0 to -3.0) | | -5.8 (-9.2 to -0.4) | -5.3 (-8.8 to -1.4) | | -3.5 (-5.1 to -2.3) | -8.7 (-10.3 to -6.8) | -6.4 (-6.8 to -5.9) | -7.7 (-10.7 to -3.4) |
| European plaice | | | | -14.6 (-15.6 to -13.5) | -11.1 (-15.5 to -6.6) | | -13.8 (-14.7 to -12.9) | | | |
| Lake trout | -3.9 (-6.4 to -1.2) | -17.3 (-18.1 to -16.5) | -3.9 (-8.2 to 0.5) | -10.0 (-24.3 to -2.5) | -10.8 (-20.9 to -7.1) | | -4.7 (-12.1 to 2.9) | -13.6 (-20.6 to -7.1) | -14.2 (-23.2 to -7.8) | -10.2 (-18.5 to -4.0) |
| Pike | | | | -7.4 | | | -2.9 | -4.6 | | -6.6 |
| Black guillemot | | 0.7 | 0.1 | 2.1 | 1.5 | | 3.0 | -10.2 | 4.7 | -3.9 |
| Black-legged kittiwake | -2.0 | -0.5 | -2.0 | -5.8 | -5.8 | -1.1 | -4.1 | -2.5 | 2.6 | |
| Common murre | | -5.8 | -5.9 | | -7.1 | -1.5 | -2.5 | -5.7 | -7.0 | |
| Glaucous gull | | | | 1.5 | | | -2.8 | | | |
| Northern fulmar | -0.9 | -1.0 | -1.6 | -4.4 | -4.5 | -0.7 | -2.6 | -7.7 | | |
| Thick-billed murre | -2.7 (-4.5 to -0.8) | -4.5 (-8.5 to -0.4) | -3.4 (-5.2 to -1.6) | -3.6 (-4.3 to -2.8) | -3.5 (-4.3 to -2.7) | -1.7 (-3.1 to -0.2) | -2.7 (-2.9 to -2.4) | -7.8 (-10.7 to -4.9) | -1.4 (-4.0 to 1.3) | 0.8 |
| Beluga | 0.9 | -7.3 (-18.5 to -1.4) | -7.6 (-19.9 to -1.0) | -3.9 (-8.9 to 0.1) | -4.9 (-10.3 to -0.7) | -27.6 (-39.3 to -16.0) | -5.4 (-13.9 to -0.2) | -3.5 (-5.5 to 1.2) | -1.2 (-5.3 to 2.9) | -6.8 (-11.5 to -1.6) |
| Northern fur seal | | -3.2 | -3.8 | -4.2 | -4.8 | -1.2 | -4.2 | -8.6 | -2.3 | -7.3 |
| Pilot whale | | -6.0 (-6.6 to -5.3) | -4.9 (-8.1 to -0.3) | -4.8 (-15.7 to 4.6) | -6.9 (-18.3 to 2.1) | | -0.6 (-3.5 to 5.1) | | 3.8 (-3.4 to 15.8) | |
| Polar bear | -1.3 (-2.5 to 0.6) | -2.4 (-3.0 to -1.6) | -1.2 | -3.3 (-5.8 to -2.2) | -3.7 (-6.1 to -2.6) | -1.1 (-2.6 to 1.5) | 1.4 (-1.4 to 5.4) | -11.4 (-12.5 to -8.5) | 0.6 (-1.8 to 7.2) | |
| Ringed seal | -1.6 (-4.3 to -0.1) | -5.7 (-5.8 to -5.7) | -3.6 (-7.5 to 0.8) | -3.2 (-5.8 to -0.8) | -4.2 (-6.3 to -2.3) | | -1.6 (-3.7 to 1.2) | -7.8 (-11.4 to -2.0) | -0.4 (-4.7 to 5.4) | -5.3 (-8.2 to 0.1) |
| All species | -3.0 (-8.3 to 0.9) | -3.6 (-18.5 to 10.3) | -4.6 (-19.9 to 0.8) | -4.1 (-24.3 to 8.4) | -4.2 (-20.9 to 11.3) | -7.8 (-39.3 to 1.5) | -2.6 (-14.7 to 5.4) | -8.9 (-20.6 to 1.2) | -1.5 (-23.2 to 15.8) | -7.6 (-18.5 to 0.8) |

Cell shading: ≤-10% | -10 to -3% | >-3 to <3% | 3 to 10% | ≥10% annual change

| Mirex | Pentachloro-benzene | CB153 | $\Sigma PCB_{10}$ | Toxaphene Parlar-26 | Toxaphene Parlar-50 | BDE47 | BDE99 | HBCDD | PFOA | PFOS | PFOSA |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | -1.3 (-12.8 to 19.6) | -0.1 (-10.0 to 20.7) | -8.5 (-14.5 to -1.5) | 3.5 (-2.7 to 7.4) | | | | | | |
| | | -6.2 (-9.5 to -3.2) | -7.0 (-10.7 to -3.0) | -7.7 | -9.9 | 8.7 (2.1 to 22.5) | 7.5 (3.6 to 13.5) | | | 0.8 | |
| -1.2 | -3.8 | -2.0 (-4.5 to 0.6) | -3.4 (-5.3 to -2.0) | | | 7.4 | | | | -3.4 | |
| | | -5.4 (-9.3 to 0.3) | -7.4 (-10.6 to -4.2) | | | | | | | | |
| | | -9.9 (-11.3 to -8.4) | -9.1 (-11.7 to -6.5) | | | | | | | | |
| -13.9 (-16.6 to -11.2) | -6.5 (-9.8 to -3.2) | -9.7 (-19.2 to -3.5) | -9.6 (-17.9 to -2.6) | | | -5.7 (-6.3 to -5.0) | | | | | |
| | | -3.1 | | | | | | | | | |
| | | 1.9 | 1.5 | -2.1 | -3.9 | | | | | | |
| -3.5 | -2.1 | -6.5 | -6.8 | | | | | | | | |
| -7.3 | | -6.1 | -5.9 | | | | | | | | |
| | | -4.0 | | | | | | -4.1 | | | |
| -1.7 | -1.6 | -3.8 | -4.2 | | | 2.4 | 0.3 | | | | |
| -7.0 (-10.4 to -3.6) | -1.8 (-2.4 to -1.2) | -3.9 (-4.6 to -3.1) | -4.4 (-5.1 to -3.6) | | | -1.3 (-5.0 to 2.4) | -1.3 (-1.3 to -1.2) | | | -0.5 | |
| -9.5 (-28.0 to -1.8) | -7.1 (-27.9 to 1.6) | -4.5 (-7.7 to -0.8) | -4.4 (-7.1 to -0.7) | | | 8.4 (5.3 to 11.5) | 11.9 (1.6 to 25.1) | 10.1 | | 3.1 (0.9 to 8.1) | -1.3 (-6.6 to 5.6) |
| -15.1 | -3.4 | -0.6 | | -7.9 | -7.9 | 12.1 | 13.2 | 13.3 | | 1.9 | -7.0 |
| 0.4 (-3.7 to 5.1) | | -2.8 (-5.7 to 1.8) | -2.5 (-5.3 to 0.4) | -2.4 (-5.3 to 1.8) | -3.0 (-5.3 to -0.1) | 6.6 | 7.3 | | | 2.0 | |
| | 6.7 | -0.9 (-2.3 to 1.3) | -1.2 (-2.8 to 1.0) | | | 3.7 (0.8 to 9.3) | 3.6 (0.5 to 5.8) | 9.7 (6.6 to 14.5) | 2.2 (-1.2 to 6.4) | -0.2 (-3.3 to 2.7) | -18.4 (-28.8 to -7.4) |
| | | -3.1 (-5.9 to -0.3) | -3.5 (-5.7 to -1.2) | -0.3 (-1.4 to 0.9) | -7.1 (-8.4 to -5.7) | 2.6 (-2.5 to 5.3) | -2.9 (-7.2 to 1.2) | 4.5 | | 6.7 (4.2 to 8.5) | |
| -6.7 (-28.0 to 5.1) | -3.8 (-27.9 to 6.7) | -3.8 (-19.2 to 19.6) | -3.7 (-17.9 to 20.7) | -6.0 (-14.5 to 1.8) | -0.8 (-9.9 to 7.4) | 4.4 (-6.3 to 22.5) | 3.8 (-7.2 to 25.1) | 7.6 (-4.1 to 14.5) | 2.2 (-1.2 to 6.4) | 2.0 (-3.4 to 8.5) | -8.5 (-28.8 to 5.6) |

Table A6.3 Summary of trend results (mean and range of annual change, %) for time-series starting in or after 2000.

| | Dieldrin | *trans*-Nonachlor | ΣCHL | *p,p'*-DDE | ΣDDT | Heptachlor-expoxide | HCB | α-HCH | β-HCH | γ-HCH |
|---|---|---|---|---|---|---|---|---|---|---|
| Blue mussel | | 6.3 (-0.1 to 19.0) | | -0.3 (-15.1 to 15.5) | 0.6 (-14.0 to 16.5) | | 3.8 (-1.3 to 7.7) | -11.4 (-21.9 to -2.0) | | -3.7 (-16.2 to 8.8) |
| Arctic char | 3.2 (1.7 to 4.6) | -20.1 | -10.8 (-24.3 to 2.8) | -2.1 (-20.3 to 12.5) | -2.4 | | -3.7 (-21.0 to 2.8) | 8.1 (2.0 to 14.2) | 5.3 (0.6 to 10.0) | 2.9 (-3.5 to 9.3) |
| Burbot | -4.6 | -6.4 | | -2.0 | -1.8 | | -1.9 | -12.2 | | -9.1 |
| Atlantic cod | | 0.5 (-2.9 to 6.2) | 3.1 | -4.0 (-11.1 to 1.9) | -0.5 (-9.7 to 5.0) | | -1.9 (-6.0 to 0.8) | -11.1 (-12.1 to -10.5) | -1.8 (-2.0 to -1.6) | -9.5 (-10.8 to -8.7) |
| European plaice | | | | -18.8 (-27.2 to -13.2) | -11.3 (-22.9 to -1.8) | | -14.5 (-17.3 to -10.7) | -17.5 | | -17.8 |
| Lake trout | | | | | | | | | | |
| Pike | | | | -8.0 | | | -1.7 | | | |
| Black guillemot | | 0.7 | -0.3 | 2.4 | 2.0 | | 3.3 | -10.1 | 6.1 | -4.1 |
| Common murre | | | | | | | | | | |
| Northern fulmar | 4.3 | 3.9 | -1.8 | -1.6 | -1.2 | -3.3 | 5.1 | -14.4 | 17.1 | |
| Thick-billed murre | 2.7 (1.1 to 4.3) | -7.2 | -5.6 (-7.3 to -4.4) | -4.8 (-5.1 to -4.3) | -4.9 (-5.2 to -4.4) | -1.5 (-2.9 to 0.2) | 1.5 (-0.7 to 3.3) | -6.5 (-13.4 to -2.7) | 4.3 (-6.2 to 13.6) | -12.8 |
| Beluga | 1.6 | -0.3 | -1.4 | 2.5 | 1.3 | | -0.4 | -2.9 | -1.0 | -1.4 |
| Pilot whale | | -11.8 (-14.6 to -8.9) | -9.2 (-16.8 to -1.7) | -10.0 (-27.3 to 2.7) | -2.5 (-6.3 to 1.3) | | -2.2 (-7.2 to 7.2) | | 11.9 (-4.2 to 40.3) | |
| Polar bear | 0.9 (-0.3 to 2.2) | -1.9 (-2.2 to -1.3) | 2.6 (-0.4 to 5.6) | -2.3 (-7.1 to 2.5) | -2.9 (-7.2 to 2.0) | 0.9 (-1.6 to 3.0) | 9.0 (6.1 to 12.3) | -13.4 (-20.7 to -1.6) | 3.0 (-0.1 to 5.9) | |
| Ringed seal | -4.6 (-7.5 to -2.8) | -4.7 (-5.8 to -3.1) | -4.2 (-7.2 to -1.0) | -3.5 (-7.0 to -0.6) | -3.9 (-7.0 to -1.0) | | 0.6 (-4.6 to 5.5) | -10.5 (-11.9 to -9.0) | -1.7 (-5.5 to 6.2) | -7.7 (-9.3 to -5.5) |
| All species | 0.2 (-7.5 to 4.6) | -0.6 (-20.1 to 19.0) | -4.3 (-24.3 to 5.6) | -3.6 (-27.3 to 15.5) | -2.4 (-22.9 to 16.5) | -0.7 (-3.3 to 3.0) | 0.02 (-21.0 to 12.3) | -9.9 (-21.9 to 14.2) | 3.4 (-6.2 to 40.3) | -6.2 (-17.8 to 9.3) |

Cell shading: ≤-10% -10 to -3% >-3 to <3% 3 to 10% ≥10% annual change

| Mirex | Pentachloro-benzene | CB153 | $\Sigma PCB_{10}$ | Toxaphene Parlar-26 | Toxaphene Parlar-50 | BDE47 | BDE99 | HBCDD | PFOA | PFOS | PFOSA |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | 1.8 (-15.3 to 28.0) | 6.1 (-10.9 to 33.0) | -7.9 (-11.6 to -1.5) | 8.3 (1.6 to 16.0) | | | | | | |
| | | -9.6 (-19.8 to -0.4) | -12.0 (-22.1 to -2.9) | | -29.4 | -12.5 (-14.8 to -8.9) | -17.2 (-25.5 to -8.8) | | | -2.1 (-5.2 to 1.1) | |
| -6.2 | -1.0 | -3.9 | -3.7 | | | 0.2 | | | | 5.9 | |
| 7.4 | | -1.6 (-8.7 to 4.2) | -2.2 (-10.8 to 2.6) | -2.2 (-5.2 to 2.8) | 0 (-5.7 to 10.1) | | | | | | |
| | -5.4 | -16.3 (-30.8 to -5.5) | -12.6 (-27.5 to -4.5) | | | | | | | | |
| | | | | | | -2.8 (-7.0 to 1.5) | -2.4 (-3.7 to -1.0) | | | -36.9 (-44.9 to -28.9) | |
| | | -3.8 | | | | 1.1 | | | | -5.2 | |
| | | 2.5 | 2.3 | -2.7 | -4.2 | | | | | | |
| | | | | | | -8.2 | -4.1 | | | | |
| 0.1 | 7.4 | | -1.1 | | | -13.6 | 14.8 | -7.5 | | 4.7 | |
| -8.0 (-14.3 to -2.6) | 5.2 (3.5 to 6.9) | -3.1 (-4.3 to -1.8) | -3.8 (-4.7 to -2.9) | | | -11.4 (-13.1 to -9.3) | 40.0 (3.0 to 104.5) | -6.9 | | 4.9 | |
| -3.9 | 2.3 | 0.4 | -0.5 | | | | | | | | |
| 2.7 (-4.6 to 13.5) | | -7.1 (-14.1 to 2.3) | -7.3 (-13.4 to 1.0) | -5.5 (-8.5 to -0.2) | -7.3 (-9.8 to -2.9) | | | | | | |
| | 11.1 (0.2 to 22.0) | 1.0 (-4.6 to 5.4) | 0.6 (-4.3 to 3.7) | | | -3.2 (-9.4 to 4.6) | -10.7 (-12.7 to -8.0) | | 3.1 (0.2 to 8.2) | -2.1 (-4.7 to -0.2) | -25.3 (-30.7 to -20.4) |
| | | -3.8 (-8.5 to 1.4) | -4.5 (-9.6 to 0.3) | -4.6 (-5.1 to -4.2) | -6.4 (-6.8 to -5.5) | 1.7 (-3.7 to 12.4) | 4.5 (-8.7 to 44.1) | 3.6 | | | |
| -1.9 (-14.3 to 13.5) | 4.5 (-5.4 to 22.0) | -2.5 (-30.8 to 28.0) | -1.5 (-27.5 to 33.0) | -5.9 (-11.6 to 2.8) | 0.8 (-29.4 to 16.0) | -4.5 (-14.8 to 12.4) | 4.8 (-25.5 to 104.5) | -3.6 (-7.5 to 3.6) | 3.1 (0.2 to 8.2) | -6.7 (-44.9 to 5.9) | -25.3 (-30.7 to -20.4) |

Table A6.4 Summary of trend results (mean and range of annual change, %) for 'adequate' and/or significant runs for time-series starting before 2000.

| | Dieldrin | *trans*-Nonachlor | ΣCHL | *p,p'*-DDE | ΣDDT | Heptachlor-expoxide | HCB | α-HCH | β-HCH | γ-HCH |
|---|---|---|---|---|---|---|---|---|---|---|
| Blue mussel | | 10.3 | | -7.3 (-8.3 to -6.2) | 0.2 (-10.9 to 11.3) | | | -9.1 (-10.7 to -6.3) | | |
| Arctic char | -6.0 (-8.3 to -3.8) | -8.3 | -8.5 (-9.5 to -6.9) | -8.3 (-9.5 to -7.4) | -9.4 (-10.3 to -8.4) | | -3.8 (-6.4 to -1.0) | -12.0 (-16.5 to -9.6) | | -9.9 (-15.5 to -5.7) |
| Burbot | -5.5 (-8.0 to -3.0) | -5.4 | | -3.7 | -5.4 (-5.7 to -5.0) | | | -9.5 (-11.4 to -7.5) | | -9.2 (-12.1 to -6.3) |
| Atlantic cod | | -5.0 (-7.0 to -3.0) | | -3.4 (-5.3 to -0.4) | -8.8 | | -3.2 (-3.8 to -2.3) | -8.7 (-10.3 to -6.8) | | -9.1 (-10.7 to -6.1) |
| European plaice | | | | -15.6 | -15.5 | | -13.8 (-14.7 to -12.9) | | | |
| Lake trout | | -17.3 (-18.1 to -16.5) | -8.2 | -17.1 (-24.3 to -9.9) | -7.5 (-7.9 to -7.1) | | -9.8 | -7.3 (-7.4 to -7.1) | | -12.9 |
| Pike | | | | -7.4 | | | -2.9 | -4.6 | | |
| Black guillemot | | | | | | | 3.0 | -10.2 | | |
| Black-legged Kittiwake | -2.0 | | | -5.8 | -5.8 | | -4.1 | | | |
| Common murre | | | -5.9 | | | | | -5.7 | | |
| Northern fulmar | -0.9 | | -1.6 | -4.4 | -4.5 | | -2.6 | | | |
| Thick-billed murre | -0.8 | -8.5 | -3.4 (-5.2 to -1.6) | -3.6 (-4.3 to -2.8) | -3.5 (-4.3 to -2.7) | -0.2 | -2.9 | -4.9 | | 0.8 |
| Beluga | 0.9 | | -1.0 | | | | -0.8 | -4.9 (-5.5 to -4.3) | 2.7 | -5.3 |
| Northern fur seal | | | | | | | | -8.6 | | -7.3 |
| Pilot whale | | | | | -18.3 | | | | -3.4 | |
| Polar bear | -1.3 (-2.5 to 0.6) | -2.9 (-3.0 to -2.7) | | -4.3 (-5.8 to -2.8) | -4.7 (-6.1 to -3.2) | -1.1 (-2.6 to 1.5) | 0.1 | -11.4 (-12.5 to -8.5) | 2.7 (-1.8 to 7.2) | |
| Ringed seal | -4.3 | -5.7 (-5.8 to -5.7) | -5.0 (-5.6 to -4.4) | -4.9 (-5.8 to -3.8) | -4.2 (-6.3 to -2.3) | | -2.9 (-3.7 to -2.0) | -9.0 (-11.4 to -4.7) | 0.5 (-4.7 to 5.4) | -6.3 (-8.2 to -2.3) |
| All species | -3.3 (-8.3 to 0.9) | -6.1 (-18.1 to 10.3) | -5.3 (-9.5 to -1.0) | -6.9 (-24.3 to -0.4) | -6.4 (-18.3 to 11.3) | -0.9 (-2.6 to 1.5) | -4.0 (-14.7 to 3.0) | -9.1 (-16.5 to -4.3) | 0.8 (-4.7 to 7.2) | -7.9 (-15.5 to 0.8) |

Cell shading: ≤-10% -10 to -3% >-3 to <3% 3 to 10% ≥10% annual change

| Mirex | Pentachloro-benzene | CB153 | $\Sigma PCB_{10}$ | Toxaphene Parlar-26 | Toxaphene Parlar-50 | BDE47 | BDE99 | HBCDD | PFOA | PFOS | PFOSA |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | -6.5 (-12.8 to -2.9) | -6.5 (-7.2 to -4.7) | -12.4 (-14.5 to -10.5) | | | | | | | |
| | | -6.3 (-9.5 to -3.2) | -7.0 (-10.7 to -3.0) | -7.7 | -9.9 | 14.9 (7.3 to 22.5) | 13.5 | | | | |
| | -3.8 | | | | | | | | | | |
| | | -3.0 (-4.8 to 0.3) | -4.8 (-5.3 to -4.2) | | | | | | | | |
| | | -11.3 | -11.7 | | | | | | | | |
| -16.6 | | | | | | | | | | | |
| | | -3.1 | | | | | | | | | |
| | | | | | | | | | | | |
| | | -6.5 | -6.8 | | | | | | | | |
| -7.3 | | | | | | | | | | | |
| -1.7 | -1.6 | -3.8 | -4.2 | | | | | | | | |
| | -1.2 | -3.9 (-4.6 to -3.1) | -4.4 (-5.1 to -3.6) | | | | | | | | |
| | 1.6 | -0.8 | -0.7 | | | 8.4 (5.3 to 11.5) | | 10.1 | | | 5.6 |
| | | | | -7.9 | -7.9 | 12.1 | 13.2 | 13.3 | | | |
| -3.7 | | | | | | | | | | | |
| | | -2.1 | -2.0 (-2.8 to -0.8) | | | 6.0 (2.6 to 9.3) | | 7.3 (6.6 to 8.0) | 1.5 | 2.7 | -19.1 |
| | | -4.4 (-5.9 to -2.7) | -4.6 (-5.7 to -3.6) | | -7.1 (-8.4 to -5.7) | 1.4 (-2.5 to 4.1) | -7.2 | 4.5 | | | |
| -7.3 (-16.6 to -1.7) | -1.3 (-3.8 to 1.6) | -5.0 (-12.8 to 0.3) | -5.3 (-11.7 to -0.7) | -10.6 (-14.5 to -7.7) | -8.0 (-9.9 to -5.7) | 6.9 (-2.5 to 22.5) | 6.5 (-7.2 to 13.5) | 8.5 (4.5 to 13.3) | 1.5 | 2.7 | -6.8 (-19.1 to 5.6) |

Table A6.5 Summary of trend results (mean and range of annual change, %) for 'adequate' and/or significant runs for time-series starting in or after 2000.

| | Dieldrin | *trans*-Nonachlor | ΣCHL | *P,P'*-DDE | ΣDDT | Heptachlor-expoxide | HCB | α-HCH | β-HCH | γ-HCH |
|---|---|---|---|---|---|---|---|---|---|---|
| Blue mussel | | 6.3 (-0.1 to 19.0) | | -15.1 | 0.6 (-14.0 to 16.5) | | | -18.2 (-21.9 to -14.5) | | -9.3 |
| Arctic char | | -20.1 | -24.3 | -12.0 (-20.3 to -3.7) | -2.4 | | -21.0 | | | |
| Burbot | | -6.4 | | | -1.8 | | | -12.2 | | -9.1 |
| Atlantic cod | | 0.5 (-2.9 to 6.2) | | | -0.5 (-9.7 to 5.0) | | -1.4 (-2.7 to 0.5) | -12.1 | -1.6 | -9.5 (-10.8 to -8.7) |
| European plaice | | | | -18.8 (-27.2 to -13.2) | | | -14.5 (-17.3 to -10.7) | -17.5 | | -17.8 |
| Pike | | | | -8.0 | | | | | | |
| Black guillemot | | 0.7 | | | 2.0 | | 3.3 | | | |
| Northern fulmar | | | | | -11.3 (-22.9 to -1.8) | | | | | |
| Thick-billed murre | | -1.9 (-2.2 to -1.3) | -7.3 | -5.0 | -2.9 (-7.2 to 2.0) | | | | | |
| Beluga | | -0.3 | | | 1.3 | | | | | -1.4 |
| Pilot whale | | 3.9 | -13.0 (-16.8 to -9.1) | -27.3 | -1.2 | -4.6 | -6.6 | | -4.2 | |
| Polar bear | | | | | | | 9.2 (6.1 to 12.3) | -16.3 (-20.7 to -6.3) | 5.9 | |
| Ringed seal | -7.5 | -11.8 (-14.6 to -8.9) | -5.0 (-5.2 to -4.8) | -3.9 (-4.1 to -3.7) | -2.5 (-6.3 to 1.3) | | -2.6 (-4.6 to -0.6) | -10.8 (-11.9 to -9.7) | -5.4 (-5.5 to -5.3) | -7.8 (-9.1 to -6.4) |
| All species | -7.5 | -4.7 (-5.8 to -3.1) | -10.3 (-24.3 to -4.8) | -13.1 (-27.3 to -3.7) | -3.9 (-7.0 to -1.0) | -4.6 | -4.5 (-21.0 to 12.3) | -14.1 (-21.9 to -6.3) | -2.1 (-5.5 to 5.9) | -8.9 (-17.8 to -1.4) |

Cell shading: ≤-10% | -10 to -3% | >-3 to <3% | 3 to 10% | ≥10% annual change

| Mirex | Pentachloro-benzene | CB153 | $\Sigma PCB_{10}$ | Toxaphene Parlar-26 | Toxaphene Parlar-50 | BDE47 | BDE99 | HBCDD | PFOA | PFOS | PFOSA |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | -9.4 (-11.0 to -7.2) | -14.0 | -8.8 (-15.3 to -2.3) | -7.9 (-11.6 to -1.5) | 8.3 (1.6 to 16.0) | | 12.2 (-6.2 to 33.0) | | | 19.0 | -4.7 (-21.9 to 33.0) |
| | | | -12.9 (-19.8 to -5.9) | | -29.4 | | -22.1 | | -29.4 | -20.1 | -18.5 (-29.4 to -3.7) |
| | | | | | | | | | | -6.4 | -9.2 (-12.2 to -6.4) |
| | -4.7 (-5.2 to -4.2) | 5.0 | | -2.2 (-5.2 to 2.8) | 0 (-5.7 to 10.1) | | | | -5.1 (-5.7 to -4.4) | | -4.7 (-12.1 to 5.0) |
| | | -19.2 (-22.9 to -15.5) | -21.7 (-30.8 to -12.6) | | | | -19.9 (-27.5 to -12.3) | | | | -18.4 (-30.8 to -10.7) |
| | | | -3.8 | | | | | | | | -5.9 (-8.0 to -3.8) |
| | | | | -2.7 | -4.2 | | | | | | 3.3 |
| -13.6 | | | | | | | | | | | -13.6 |
| -11.7 | | -5.0 | | | | | | -6.9 | | | -7.2 (-11.7 to -5.0) |
| | | | 0.4 | | | | | | | | -0.5 (-1.4 to 0.4) |
| | -8.5 | | -14.1 | | | | -11.4 (-13.4 to -9.4) | | -9.5 (-9.8 to -9.2) | -14.6 | -11.4 (-27.3 to -4.2) |
| | | -7.2 | 4.2 | | | -11.4 | 3.7 | | | | -4.7 (-20.7 to 12.3) |
| -3.5 (-3.7 to -3.2) | -4.8 (-5.1 to -4.5) | -4.2 (-4.5 to -3.9) | -5.1 (-5.4 to -4.7) | -5.5 (-8.5 to -0.2) | -7.3 (-9.8 to -2.9) | 9.2 (-8.2 to 44.1) | -4.4 (-4.7 to -4.1) | | -6.4 (-6.8 to -5.5) | -5.5 (-5.8 to -5.1) | -4.8 (-11.9 to 44.1) |
| -8.1 (-13.6 to -3.2) | -7.0 (-11.0 to -4.2) | -8.5 (-22.9 to 5.0) | -9.2 (-30.8 to 4.2) | -4.6 (-5.1 to -4.2) | -6.4 (-6.8 to -5.5) | 4.1 (-11.4 to 44.1) | -4.8 (-27.5 to 33.0) | -6.9 | -9.7 (-29.4 to -4.4) | -5.5 (-20.1 to 19.0) | -8.1 (-30.8 to 44.1) |

# Acronyms and Abbreviations

| | |
|---|---|
| ΣCHL | Sum of *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor and oxychlordane |
| ΣDDT | Sum of *p,p*'-DDE, *p,p*'-DDD, and *p,p*'-DDT |
| $\Sigma PCB_{10}$ | Sum of CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156, CB180 |
| $\Sigma PCB_{11}$ | Sum of CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156, CB180, CB209 |
| AMAP | Arctic Monitoring and Assessment Programme |
| DDD | Dichlorodiphenyldichloroethane |
| DDE | Dichlorodiphenyldichloroethylene |
| DDT | Dichlorodiphenyltrichloroethane |
| DF | Digital filtration |
| EF | Enantiomer fraction |
| EtFOSE | Ethyl perfluorooctane sulfonamide ethanol |
| FTOH | Fluorotelomer alcohol |
| GMP | Global Monitoring Plan |
| HBCD | Hexabromocyclododecane |
| HCB | Hexachlorobenzene |
| HCH | Hexachlorocyclohexane |
| HEPOX | Heptachlor epoxide |
| ln C | Natural log-transformed concentration data |
| lw | Lipid weight |
| masl | Meters above sea level |
| MeFOSE | Methyl perfluorooctane sulfonamido ethanol |
| NCP | Northern Contaminants Program (Canada) |
| NILU | Norwegian Institute for Air Research |
| OCS | Octachlorostyrene |
| PBDEs | Polybrominated diphenyl ethers |
| PCBs | Polychlorinated biphenyls |
| PFAAs | Perfluorinated alkyl acids |
| PFASs | Per-and polyfluoroalkyl substances |
| PFCs | Perfluorinated compounds |
| PFCAs | Perfluorinated carboxylic acids |
| PFDA | Perfluorodecanoate |
| PFDoA | Perfluorododecanoic acid |
| PFHxS | Perfluorohexane sulfonate |
| PFNA | Perfluorononanoate |
| PFOA | Perfluorooctanoic acid |
| PFOS | Perfluorooctane sulfonic acid |
| PFOSA | Perfluorooctanesulfonamide |
| PFTA | Perfluorotetradecanoic acid |
| PFTrA | Perfluorotridecanoic acid |
| PFUnA | Perfluroundecanoic acid |
| POP | Persistent organic pollutant |
| PUF | Polyurethane foam plug |
| $t_{1/2}$ | First order half-life |
| TDC | Thematic Data Centre |
| ww | Wet weight |

# Biota

| | |
|---|---|
| Invertebrates | |
| Blue mussel | *Mytilus edulis* |
| Fish | |
| Arctic char | *Salvelinus alpinus* |
| Burbot | *Lota lota* |
| Cod | *Gadus morhua* |
| European plaice | *Pleuronectes platessa* |
| Lake trout | *Salvelinus namaycush* |
| Pike | *Esox lucius* |
| Birds | |
| Black guillemot | *Cepphus grylle* |
| Black-legged kittiwake | *Rissa tridactyla* |
| Common murre | *Uria aalge* |
| Glaucous gull | *Larus hyperboreus* |
| Glaucous-winged gull | *Larus glaucescens* |
| Herring gull | *Larus argentatus* |
| Ivory gull | *Pagophila eburnea* |
| Leach's storm-petrel | *Oceanodroma leucorhoa* |
| Northern fulmar | *Fulmarus glacialis* |
| Peregrine falcon | *Falco peregrinus* |
| Thick-billed murre | *Uria lomvia* |
| Marine mammals | |
| Beluga | *Delphinapterus leucas* |
| Fin whale | *Balaenoptera physalus* |
| Minke whale | *Balaenoptera acutorostrata* |
| Northern fur seal | *Callorhinus ursinus* |
| Pilot whale | *Globicephala melas* |
| Polar bear | *Ursus maritimus* |
| Ringed seal | *Pusa hispida* |

**Arctic Monitoring and Assessment Programme**

The Arctic Monitoring and Assessment Programme (AMAP) was established in June 1991 by the eight Arctic countries (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden and the United States) to implement parts of the Arctic Environmental Protection Strategy (AEPS). AMAP is now one of six working groups of the Arctic Council, members of which include the eight Arctic countries, the six Arctic Council Permanent Participants (indigenous peoples' organizations), together with observing countries and organizations.

AMAP's objective is to provide 'reliable and sufficient information on the status of, and threats to, the Arctic environment, and to provide scientific advice on actions to be taken in order to support Arctic governments in their efforts to take remedial and preventive actions to reduce adverse effects of contaminants and climate change'.

AMAP produces, at regular intervals, assessment reports that address a range of Arctic pollution and climate change issues, including effects on health of Arctic human populations. These are presented to Arctic Council Ministers in 'State of the Arctic Environment' reports that form a basis for necessary steps to be taken to protect the Arctic and its inhabitants.

This report has been subject to a formal and comprehensive peer review process. The results and any views expressed in this series are the responsibility of those scientists and experts engaged in the preparation of the reports.

The AMAP Secretariat is located in Oslo, Norway. For further information regarding AMAP or ordering of reports, please contact the AMAP Secretariat (Gaustadalléen 21, N-0349 Oslo, Norway) or visit the AMAP website at www.amap.no.

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