Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota

Frank Rigét1,2*, Anders Bignert3, Birgit Braune4, Maria Dam5, Rune Dietz1, Marlene Evans6, Norman Green7, Helga Gunnlaugsdóttir8, Katrin S. Hoydal9, John Kucklick9, Robert Letcher4, Derek Muir10, Stacy Schuur9, Christian Sonne1, Gary Stern11, Gregg Tomy12, Katrin Vorkamp13, Simon Wilson14

1 University of Aarhus, Department of Biosciences, Arctic Research Centre, PO Box 358, DK-4000, Roskilde, Denmark
2 Greenland Institute of Natural Resources, Kivioq 2, PO Box 570, Nuuk, 3900, Greenland.
3 Dep. of Environmental Research and Monitoring, Swedish Museum of Natural History, PO Box 50 007, S-104 05 Stockholm, Sweden
4 Environment and Climate Change Canada, National Wildlife Research Centre, Carleton University, Raven Road, Ottawa, Canada K1A 0H3
5 Environment Agency, Traðagøta 38, P.O. box 2048, FO-165 Argir, Faroe Islands
6 Aquatic Ecosystem Protection Research Division, Environment Canada, 11 Innovation Blvd, Saskatoon, Canada, SK S7N 3H5
7 Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, NO-0349 Oslo, Norway
8 Matís, Icelandic Food and Biotech R&D, Vinlandsleið 12, 113 Reykjavík, Iceland
9 National Institute of Standards and Technology, Hollings Marine Laboratory, 331 Fort Johnson Road, Charleston, SC 29412, USA
10 Environment and Climate Change Canada, Aquatic Contaminants Research Division, 867 Lakeshore Road, Burlington, Ontario, Canada L7R 4A6
11 Centre for Earth Observation Sciences (CEOS), Clayton H. Riddell Faculty of Environment, Earth and Resources, University of Manitoba, 586 Wallace Bld, 125 Dysart Rd. Winnipeg, Manitoba, Canada, R3T 2N2
12 Centre for Oil and Gas Research and Development, University of Manitoba, Department of Chemistry, Winnipeg, Manitoba, Canada R3T 2N2
13 University of Aarhus, Department of Environmental Chemistry and Microbiology, Arctic Research Centre, PO Box 358, DK-4000, Roskilde, Denmark
14 Arctic Monitoring and Assessment Programme (AMAP) Secretariat, PO Box 8100 Dep. N-0032 Oslo, Norway

*Corresponding author: Frank Rigét, frri@natur.gl

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Abstract

More than 1000 time-series of persistent organic pollutants (POPs) in Arctic biota from marine and freshwater ecosystems some extending back to the beginning of 1980s were analyzed using a robust statistical method. The Arctic area encompassed extended from Alaska, USA in the west to northern Scandinavian in the east, with data gaps for Arctic Russia and Arctic Finland. The aim was to investigate whether temporal trends for different animal groups and matrices were consistent across a larger geographical area. In general, legacy POPs showed decreasing concentrations over the last two to three decades, which were most pronounced for α-HCH and least pronounced for HCB and β-HCH. Few time-series of legacy POPs showed increasing trends and only at sites suspected to be influenced by local source. The brominated flame retardant congener BDE-47 showed a typical trend of increasing concentration up to approximately the mid-2000s followed by a decreasing concentration. A similar trend was found for perfluorooctane sulfonic acid (PFOS). These trends are likely related to the relatively
recent introduction of national and international controls of hexa- and hepta-BDE congeners and the voluntary phase-out of PFOS production in the USA in 2000. Hexabromocyclododecane (HBCDD) was the only compound in this study showing a consistent increasing trend. Only 12 % of the long-term time-series were able to detect a 5 % annual change with a statistical power of 80 % at $\alpha < 0.05$. The remaining 88 % of time-series need additional years of data collection before fulfilling these statistical requirements. In the case of the organochlorine long-term time-series, 45 % of these would require more than 20 years monitoring before this requirement would be fulfilled.

1. Introduction

Persistent organic pollutants (POPs) are chemicals that have a long lifetime in the environment and due to their physical-chemical properties, they are transported over long distances. POPs enter food-webs and accumulate, and in some cases biomagnify in wildlife and humans. Several global and regional conventions were developed with the goal of eliminating or reducing emissions of POPs. The Stockholm Convention on POPs initially addressed twelve priority POPs and since then these have been extended to 28 POPs as of 2017 (http://chm.pops.int). Despite the fact that several of these POPs were banned in the 1970’s and 1980’s and others have been restricted, they are still found in the environment at levels that may cause adverse effects to the health of top predators in Arctic food chains (Letcher et al., 2010). Humans living in the Arctic and consuming significant amounts of high trophic traditional food are exposed to legacy POPs, which may lead to adverse health effects (AMAP, 2009).

Temporal trend studies are an important means of assessing the fate of contaminants in ecosystems. They can provide a first warning that potentially harmful compounds may be increasing in the environment, e.g. in biota regarded as indicator organisms. Temporal trend studies may also indicate whether regulatory actions aimed at reducing harmful chemicals in the environment are proving successful, or whether environmental levels are approaching or exceeding threshold values for biological and possibly toxic effects. Several Arctic countries perform POP monitoring in biota, focusing on freshwater and marine ecosystems, resulting in time series of varying statistical power. Statistical power is an important consideration in relation to temporal trend monitoring. The power of a temporal trend represents the statistical probability of detecting a change of a given magnitude when this change actually occurs. It is desirable that monitoring data series have sufficient statistical power to minimise false negatives, i.e. the risk of incorrectly concluding that no change has occurred.

Rigét et al., (2010) assessed temporal trends of POPs in Arctic biota based on the time-series available up until 2008 and in the context of the Arctic Monitoring and Assessment Programme (AMAP). A large number of new time-series data has become available since then and additional years have been added to the time-series included in Rigét et al., (2010). Therefore, it was expected that this update would lead to a more robust assessment than the previous one. The objective of this study was to analyze trends for POPs in all time-series available from the Arctic, including an evaluation of their statistical power.

2. Datasets and statistical analyses
Time-series of POPs were available from seven Arctic countries for a total of 64 location-species-tissue combinations. Figure 1 shows the locations, together with the species or species group monitored. No series in biota are currently available for the Arctic areas of Russia and Finland. Time-series were available for marine mammals, seabirds, marine and freshwater fish and blue mussels.

Asmund et al. (2004) focused early on the quality assurance/quality control (QA/QC) of the Danish and Canadian laboratories, which were involved in the Greenlandic AMAP trend program. AMAP has worked towards harmonized programs with respect to methodologies and QA/QC in order to ensure the quality and credibility of AMAP assessments. For example, AMAP has established guidelines that cover all aspects of data generation from sample collection, handling and processing, to analysis, and data management, and that must be followed by the laboratories contributing to AMAP assessments. The laboratories responsible for the POP analyses included in this study participate in a number of QA/QC programs including the AMAP/NCP inter-laboratory studies (e.g. Tkatcheva et al. 2013), the QUASIMEME laboratory performance testing scheme (www.quasimeme.org), and equivalent QA/QC programs run by NOAA/NIST. All laboratories have established common internal QA/QC procedures and measures such as the use of internal and certified reference materials, analyses of blank and duplicates etc. Each individual time-series has been analysed by the same laboratory with the same method over several years.

Figure 1. Map of locations with long-term time-series. Symbols indicate animal group. Included are Air monitoring stations. From AMAP (2016).

Time-series were separated into long-term time-series starting before the year 2000 and short-term time-series starting in or after the year 2000. Several of the short-time time-series were derived by deleting data from years before 2000 of the long-term time-series. In total, 1074 long-term time-series and 735 short-term time-series were available covering the following POPs: Polychlorinated biphenyls...
congeners, dichlorodiphenyltrichloroethane (DDT) and its transformation products, hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and pentachlorobenzene (PeCB), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and pentachlorobenzene (PeCB), chlordane-related pesticides (CHL) and heptachlor, dieldrin, mirex, octachlorostyrene (OCS), toxaphene, polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and perfluoroalkyl substances (PFASs). Octachlorostyrene (OCS) was included although not included in the Stockholm Convention because time-series were available and only few time-series study has been published. The distribution of available time-series by country, species group and time-series type are shown in Table 1.

Table 1. Number of long-term time-series (starting before year 2000) and short-term time-series (starting in or after year 2000) by country and species group.

<table>
<thead>
<tr>
<th>Country</th>
<th>Long-term time-series</th>
<th>Short-term time-series</th>
</tr>
</thead>
<tbody>
<tr>
<td>United States (Alaska)</td>
<td>186</td>
<td>17</td>
</tr>
<tr>
<td>Canada</td>
<td>323</td>
<td>200</td>
</tr>
<tr>
<td>Greenland</td>
<td>184</td>
<td>163</td>
</tr>
<tr>
<td>Faroe Islands</td>
<td>102</td>
<td>115</td>
</tr>
<tr>
<td>Iceland</td>
<td>109</td>
<td>137</td>
</tr>
<tr>
<td>Norway</td>
<td>155</td>
<td>89</td>
</tr>
<tr>
<td>Sweden</td>
<td>15</td>
<td>14</td>
</tr>
<tr>
<td>Total</td>
<td>1074</td>
<td>735</td>
</tr>
</tbody>
</table>

Datasets were treated as in previous assessments of temporal trends of contaminants in Arctic biota (Rigét et al., 2010; AMAP, 2014; 2016). Only time-series with six or more years of data were included. Generally, time-series were considered inappropriate for trend analyses if three or more years had annual medians that were less than the reported detection limit. However, time-series were included if these years were concentrated in the end of the time-series, indicating a decreasing trend or in the beginning, indicating an increasing trend. In these cases, half of the detection limit was used to represent the actual annual medians.

A statistically robust method was applied to the time-series data. Covariates such as age, sex and lipid content were treated as recommended by data originators such as selecting a subset of similar characteristics e.g. sex and/or age. All time-series were treated with the same statistical method and therefore highly comparable results have been generated.

The statistical analyses were performed using the software package PIA (Plot and Image Analyses) (Bignert, 2013). Briefly, median concentrations were used as the annual index values to minimise the influence of outliers and values below detection limits. The method tests for the presence of a log-linear trend and/or non-linear trend by separating the total variance over time into a log-linear component and a non-linear component (Nicholson et al., 1998). The log-linear trend was tested by log-linear regression. A 3-point running smoother was applied to describe the non-linear trend component and
tested by mean of an analysis of variance (ANOVA). In general, $\alpha < 0.05$ was applied. The results of the trend analyses were classified into six classes as follows (AMAP, 2016):

- **Increasing**, a statistically significant increasing log-linear trend.
- **Increasing with non-linear trend component**, both an increasing log-linear and the non-linear trend components are statistically significant.
- **Decreasing**, a statistically significant decreasing log-linear trend.
- **Decreasing with non-linear trend component**, both a decreasing log-linear and the non-linear trend components are statistically significant.
- **Non-linear component**, a statistically significant non-linear (fluctuating) trend with no significant increasing or decreasing trend.
- **No trend**. The time-series did not exhibit a statistically significant trend.

Statistical power is defined as the probability of rejecting the null hypotheses in a case where a temporal trend exists, and thereby accepting the alternative hypotheses that a temporal trend exists (Cohen, 1977). For time-series, this describes the probability to detect a temporal trend, when the trend is in fact real.

The determination of statistical power of the time-series followed the method described by Fryer and Nicholson (1993). Two measures of power were estimated: 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. 2) the minimum average annual percent change over a 10-year 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 as the latter is not locked at a specific 5% annual change.

### 3. Results

#### 3.1 Polychlorinated biphenyls (PCBs)

PCBs are an industrial and by-product chemical that were produced in global total quantities of about 1.3 million tons (Breivik et al., 2007). They were listed in both Annex A (eliminate production and use) and Annex C (unintentional production) of the Stockholm Convention on POPs in 2004, but most industrial countries banned PCBs already back in the 1970s/1980s (AMAP, 1998). Nevertheless, there still are important sources of PCB emissions in the cities of Western Europe and North America due to legacy PCB use in closed systems such as electrical transformers (Diamond et al., 2010; Gasic et al., 2010).

A total of 347 long-term time-series were analyzed for concentrations of $\Sigma_{10}$PCB (CB-28, -31, -52, -101, -105, -118, -138, -153, -156 and -180) and individual congeners, mainly CB-153. Thirty-eight (38) % of $\Sigma_{10}$PCB and 34 % of CB-153 time-series showed a significant decreasing trend, and 13 % and 15 %, respectively, showed a significant decreasing trend together with a significant non-linear trend component (Figure 2). The mean annual decrease for all time-series was 3.7 % for $\Sigma_{10}$PCB and 3.8 % for CB-153. These time-series were spread over the entire Arctic area, for which data were available. Only one time-series (blue mussels, one location in Iceland) showed a significant increasing trend together
with a significant non-linear component for both $\Sigma_{10}$PCB and CB-153. However, this particular site is likely affected by a local pollution source and several other POPs (e.g. DDTs) likewise showed increasing trends (Sturludottir et al., 2013).

In seventeen long-term time-series covering all species groups (Table 1) and having analyzed eight of the same PCB congeners (CB-28, -52, -101, -105, -118, -138, -153 and -180), no significant difference was found between annual trends of individual congeners (ANOVA, $p = 0.73$).

For short-time time-series of $\Sigma_{10}$PCB and CB-153 fewer time series showed significant decreasing trends (20 % and 18 %, respectively) (Figure 2). The mean annual decrease was also lower (1.5 % and 2.5 %, respectively). Three time-series showed significant increasing trends of $\Sigma_{10}$PCB, one of these also for CB-153. These results indicate that the main decrease of PCBs occurred in earlier years and that the decrease has been less pronounced in recent years after 2000.

PCB time-series were available from five ringed seal ($Pusa hispida$) populations and one northern fur seal ($Callorhinus ursinus$) population covering the area from Alaska to East Greenland. The largest annual decrease of CB-153 was found in the eastern populations (East Greenland) and there was also a tendency to smaller annual decrease in the northern populations (Figure 3). However, no firm conclusion should be drawn as the length and the period covered by the time-series differed.

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**Figure 2.** Overview of the trend results for POPs in long-term time-series (left) and short-term time-series (right). The colors represent the percentage of time-series with a given trend for each contaminant. Red = increasing trend, Pink = increasing with a non-linear trend. Green = decreasing trend, Light green = decreasing with a non-linear trend, Gray = non-linear trend, Black = no trend.
Figure 3. The relative size of the annual decrease of CB-153 in seal populations across the Arctic from Alaska to East Greenland. The double ring represents the juvenile (outer circle) and the adult seals (inner circle) from the same location.

The mean annual decrease of $\Sigma_{10}$PCB in muscle tissue of freshwater fish was higher (-8.2 %) but not significantly (t-test, $p = 0.15$) than in muscle tissue of marine fish (-5.3 %).

3.2 Dichlorodiphenyltrichloroethane (DDT) and its transformation products

DDT (insecticide) was listed in Annex B (restrict production and use) of the Stockholm Convention on POPs in 2004, but like for PCBs most industrial countries banned DDT already back in the 1970s/1980s (AMAP, 1998). In recent years, the global annual production was approximately 3300 metric tons and used to control insects that spread diseases such as malaria primarily in tropical countries (UNEP/POPS/COP.8/INF/6, 2017).
Time-series of ΣDDT (pp'-DDE, pp'-DDD, pp'-DDT) and individual o,p' and p,p' isomers were analyzed in a total of 165 time-series mostly of ΣDDT and p,p'-DDE (71%). Several time-series of p,p'-DDD in blue mussels (Mytilus edulis) and some marine fish could not be evaluated due to several years with median values below the detection limit. Forty-six (46) six % of the long-term time-series of ΣDDT showed a significant decreasing trend, while only 32 % of the p,p'-DDE time-series showed a significant decrease. One ΣDDT time-series showed a significant increasing trend; this was the same time-series from Iceland that also showed an increasing trend for PCBs. The significant decreasing trends and those showing no significant trends were spread over the entire area from Alaska to northern Norway.

The mean annual decrease was 4.2 % for both ΣDDT and p,p'-DDE. For 55 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 between p,p'-DDE and Σ DDT may be a rough indicator of the age of DDT residues in the environment. Figure 4 shows the trends of this ratio in selected long-term time-series. In all four time-series the ratio p,p'-DDE/Σ DDT increased with time, and this increase was significant in polar bear (Ursus maritimus), ringed seal, and pilot whale (Globicephala melas) (log-linear regression of annual medians, p < 0.01 and p = 0.03, respectively), but was not significant in belugas (Delphinapterus leucas; p = 0.12). Therefore, no indication of a presence of ‘fresh’ sources of DDT to the Arctic environment were found. This is also supported by the twelve time-series of p,p'-DDT that have a mean annual decrease of 10.5 %, the largest mean decrease of all of the DDT isomers considered.

A total of seventeen long-term time-series of o,p'-DDE, o,p'-DDD, o,p'-DDT were available from pilot whales from the Faroe Islands and beluga and northern fur seal from the Alaska. None of these showed significant trends, except one (for female and juvenile beluga from East Chukchi/Bering Sea) where levels were relatively constant up to 1998 followed by a sharp decline giving a significant non-linear trend.

The proportion of short-term time-series showing significant decreasing trends was considerably lower than that of the long-term time-series (Figure 2). The mean annual decreases of the short-term time-series were 2.4 % and 3.6 % for ΣDDT and p,p'-DDE, respectively.

The mean annual decrease of $\Sigma$DDT in muscle tissue of freshwater fish was higher (-9.1 %) but not significantly (t-test, $p = 0.18$) than in muscle tissue of marine fish (-5.2 %).

3.3 Hexachlorocyclohexanes (HCHs)

Technical hexachlorocyclohexane (consisting of ca. 55-70 % $\alpha$-HCH, 5-15 % $\beta$-HCH, 10-20 % $\gamma$-HCH (lindane) and small amounts of $\delta$-HCH) was widely used as an insecticide prior to its phase out in a number of countries (including the USA, Canada and Nordic countries) in the late 1970s. (Li et al., 2005). There are only two lindane producing countries left (India and Romania) (Vijgen, 2006). $\alpha$-, $\beta$-HCH and lindane were listed in Annex A of the Stockholm Convention in 2009, however, lindane had a specified exemption for cases where human health might be affected (http://chm.pops.int).

In total 110 long-term time-series of $\alpha$-, $\beta$- and $\gamma$-HCH and 80 short-term time-series were available. However, several HCH time-series could not be evaluated because of more than three years with annual medians below detection limits. Thirty seven (80 %) of the long-term $\alpha$-HCH time-series showed a significant decreasing trend or a significant decreasing trend together with a significant non-linear trend component (Figure 2). For the short-term time-series, the corresponding amount was 18 (53 %) (Figure 2).
The mean annual decrease of the long-term time-series was 8.9 %, which was among the highest decrease of all the compounds (Figure 5). For the short-term time-series, the mean annual decrease was 9.9 %, showing that α-HCH has decreased after the year 2000 with a similar or higher rate than before 2000.

**Figure 5.** Mean of annual change (± SD) of POPs in the 1074 long-term Arctic time-series of compounds by increasing order.

The results for the time-series of β-HCH were quite different from those of α-HCH. More than half (64 %) of the β-HCH long-term time-series showed no significant trend or a significant non-linear trend, and both significant decreasing and increasing trends were found. In Canada, two seal populations showed significant increasing trends and time series for beluga and seabird eggs showed significant increasing trends together with a significant non-linear trend component. The mean annual decrease of 1.5 % for the long-term time-series is the lowest decrease among organochlorines, except the toxaphene congener Parlar_50 (Figure 5). Among the short-term time-series, 83 % showed no trend or a significant non-linear trend component.

The results of the γ-HCH time-series were closer to those of α-HCH than β-HCH with 70 % of the long-term time-series showing significant decreasing trend or significant decreasing trend together with a significant non-linear trend component. The corresponding percentage for the short-term time-series was 50 %. The mean annual decreases were 7.6 % and 6.2 % for long-term and short-term time-series, respectively, a somewhat smaller decrease than for α-HCH (Figure 5).

No general relationships between size of annual change in the long-term time-series and longitude were found for α, β and γ-HCH (linear regression, p = 0.30, p = 0.12 and p = 0.51, respectively). However, when considering only the seal populations, there appeared to be a tendency of lowest annual decrease of α-HCH in the two populations at highest latitude and in case of β-HCH an annual increase was found for these two populations (Figure 6).
Figure 6. Annual decrease (green) and annual increase (red) in seal populations related to longitude and latitude. The size of circle represents the size of annual change. The double ring represents the juvenile (inner circle) and the adult seals (outer circle) from the same location.

3.4 Hexachlorobenzene (HCB) and pentachlorobenzene (PeCB)

HCB has had several uses in industry and agriculture (Becker et al., 2012). PeCB is not known to be manufactured for any commercial uses (Bailey et al., 2009). In the past, PeCB was one component of chlorobenzenes mixture (Bailey et al., 2009). HCB and PeCB have been listed in Annex A and Annex C of the Stockholm Convention (http://chm.pops.int). While HCB was among the initial POPs regulated in 2004, PeCB was added to the Stockholm Convention on POPs in 2009 (UNEP/POPS/POPRC.3/20/Add.2, 2007). (http://chm.pops.int).

Fifty-five (55) long-term time-series of HCB were available of which 36 % showed significant decreasing trends, some of these also with a significant non-linear trend component. Two time-series showed a significant increasing trend, one of them also with a significant non-linear component. The mean annual decrease for HCB time-series was 2.6 %, somewhat lower than for most other organochlorine compounds (Figure 5).
Six (6) out of 43 short-term time-series exhibited a significant decreasing trend and three (3); all from East Greenland show significant increasing trends. The remaining time-series showed no trend or with a significant non-linear trend component. The mean annual change for short-term time-series was close to 0 % indicating that no or limited change has occurred since 2000.

Despite the large proportion of declining trends of HCB observed across the North American and European Arctic over recent decades, the relative low mean rate of the declines compared to other organochlorines and the increasing trend at some sites after the year 2000 may indicate that HCB emissions (primary and secondary) and releases are still occurring.

Thirteen (13) long-term time-series of PeCB existed from Canada and the United States of which two (2) showed significant decreasing trend, one of them also with a significant non-linear trend component. The other time-series show either no trend (9) or a significant non-linear trend (2). The mean annual decrease change was 3.8 %.

3.5 Chlordane-related pesticides (CHL) and heptachlor

Complex mixtures of chlordane-related compounds were used as a broad-spectrum insecticide in seed dressing and have been banned or restricted for decades. Heptachlor is an insecticide used to control soil insects, termites and crop pests, and is also a constituent of technical chlordane. Heptachlor degrades in the environment to heptachlor epoxide, which is more persistent (Gannon and Bigger, 1958). Chlordane and heptachlor were listed in Annex A of the Stockholm Convention on POPs in 2004 (http://chm.pops.int) Cis- and trans-chlordane and cis- and trans-nonachlor, together with heptachlor are primary constituents of the more than 120 compounds in the technical chlordane mixture (Dearth and Hites, 1991).

There were 31 long-term time-series of ΣCHL (sum of cis-chlordane, trans-chlordane, cis-nonachlor, trans-nonachlor and oxychlordane), 35 of trans-nonachlor and 20 of oxychlordane. Decreasing trends or decreases together with a non-linear trend component were found in 48%, 41% and 35% of the long-term time series for ΣCHL, trans-nonachlor and oxychlordane, respectively. Only one trans-nonachlor time-series showed an increasing trend, which was the same site also showing increasing trends of PCBs and DDTs influenced by local pollution sources. The mean annual decreases for ΣCHL, trans-nonachlor and oxychlordane were 4.6 %, 3.6 % and 3.0 %, respectively.

As for most organochlorines, the proportion of significantly decreasing trends in biota was smaller and the proportion of time-series showing no trend greater when considering the short-term time-series. The annual decrease since 2000 ranged between 0.6 % for trans-nonachlor and 4.3 % for ΣCHL.

Marine mammals and seabirds metabolize cis- and trans-chlordane (Fisk et al., 2001). Oxychlordane is the primary metabolite of cis- and trans-chlordane and is persistent in the environment (Tashiro and Matsumura, 1978; Bondy et al., 2000). The ratio between oxychlordane and ΣCHL may be a rough indicator of the age of chlordane residues in the environment. Figure 7 shows the temporal trends of the ratio between oxychlordane and ΣCHL in three long-term time-series. The general increasing levels of the ratio in the order beluga < ringed seal < polar bear express the relative metabolic potential among
these species. All three time-series show an increase in the ratio of oxychlordane/ΣCHL with time, however, only significantly in case of ringed seals (log linear regression, p < 0.01). This indicates that the metabolite oxychlordane increases relative to some of its precursors.

Figure 7. Trends of the ratio of oxychlordane/ΣCHL in juvenile polar bear (black) and ringed seal (red) both from East Greenland and beluga (blue) from northwest Canada.

Three time-series of heptachlor (seabirds and northern fur seals from Alaska) were available; however, in all years the annual medians were below detection limits. Twelve long-term time-series of heptachlor epoxide were evaluated; all from seabird eggs or marine mammal tissue. Of these, only two time-series showed significant decreasing trends or decreases together with a non-linear trend component; the other time-series showed no statistical trends. Heptachlor was one of the organochlorines with the highest annual decrease (Figure 5). None of the seven short-term time-series showed significant decreasing trends.

3.6 Dieldrin

Dieldrin is a pesticide used to control insect-borne diseases. It is also the degradation product of the pesticide aldrin (http://chm.pops.int). Both dieldrin and aldrin have been restricted or voluntarily withdrawn from use by most developed countries. Both pesticides were among the original compounds listed in Annex A of the Stockholm Convention on POPs in 2004 (http://chm.pops.int).

Twenty-two (22) long-term time-series were available for dieldrin. Ten (10) of these (45 %) showed significant decreasing trends or decreases together with a non-linear trend component. The mean annual decrease was 3.0 %. Among the 13 short-term time-series only one showed a significant decreasing trend, and the mean annual decrease was close to zero indicating little if any change in levels since 2000. This indicates that dieldrin (and aldrin) contamination in the Arctic has slowly decreased following bans introduced in the period before the year 2000.

3.7 Mirex
Mirex is an insecticide primarily used in the 1960s and 1970s in the United States, but it was also used as a fire retardant (ATSDR, 1995). Mirex was among the original compounds listed in Annex A of the Stockholm Convention on POPs in 2004 (http://chm.pops.int).

Sixteen (16) long-term time-series for mirex were available originating from the Faroe Islands, Canada and USA (Alaska) and included marine mammals, seabirds and freshwater fish. Four (4) of these time-series showed significant decreasing trends, whereas the others showed either a non-linear trend or no trend. The mean annual decrease was 6.7 %. For short-term time-series, a total of ten (10) time-series existed, but only one showed a significant decreasing trend and the mean annual decrease was 1.9 %.

3.8 Octachlorostyrene (OCS)

Octachlorostyrene has never been used as a commercial product, but it may be produced during incineration and combustion process involving chlorinated compounds (Chu et al., 2003). Octachlorostyrene is not among the listed compounds in the Stockholm Convention on POPs (http://chm.pops.int).

Eleven (11) long-term time-series, all from Canada or Greenland, were available. Several marine fish and blue mussel time-series from northern Norway could not be evaluated because nearly all annual medians were below detection limits. Four (4) of the 11 time-series included in the evaluation showed significant decreasing trends, while the others showed non-linear or no trends. Considering the eleven (11) time-series from year 2000 and onwards, none showed a significant decreasing trend and the mean annual change was raised from a decrease of 4.5 % to an increase of 0.2 %.

3.9 Toxaphene

Toxaphene is an insecticide consisting of a complex mixture of polychlorobornanes and camphenes (De Geus et al., 1999). Toxaphene was among the initial 12 compounds listed by the Stockholm Convention on POPs in 2004.

Seventeen (17) long-term time-series of toxaphene congeners Parlar 26 and 50 were available including blue mussels, marine mammals, seabirds and freshwater fish. Of those six (6; 35 %) and five (5; 29 %) showed significant decreasing trends or decreasing together with a non-linear trend component for Parlar 26 and 50, respectively. No time-series showed significant increasing trends. The mean annual decrease was 6.0 % for Parlar 26 but was considerably lower for Parlar 50 (0.8 %).

Nine (9) out of 20 (45 %) and eight (8) out of 22 (36 %) short-time time-series of Parlar 26 and 50, respectively, showed significant decreasing trends one together with a significant non-linear trend component. The mean annual decrease was 5.9 % of Parlar 26 while Parlar 50 had a mean annual increase of 0.8 %.

3.10 Polybrominated diphenyl ethers (PBDEs)

PBDEs were used as flame retardants. Tetra-, penta-, hexa-, and hepta-BDE were added to Annex A of the Stockholm Convention in 2009, while deca-BDE was added in 2017 (UNEP/POPS/POPRC.10/10/2,
2014), with specified exemptions. There are still emissions of PBDEs to the environment because of the stock of in-use products (Abbasi et al., 2015). PBDEs can be subject to debromination and the products are lower brominated PBDE congeners (Söderström et al., 2004).

Most long-term time-series were available for the congeners BDE-47 (26) and BDE-99 (19) and only few for other congeners. BDE-47 was the compound with highest number (8) of time-series showing a significant increasing trend and in one case together with a non-linear trend component (35 %) (Figure 2). Only two time-series showed significant decreasing trends, one also having a significant non-linear trend component. Considering the short-term time-series of BDE-47, none showed an increasing trend and five (5) out of 23 (22 %) showed significant decreasing trends. The non-linear trend component was significant in 31 % of the long-term time-series of BDE-47 and a typical trend was an increase up until year 2005 followed by a decrease in recent years, which is quite different from the general trend of organochlorines (Figure 8). Only two (2) out of 19 (11 %) and one (1) out of 17 (5.9 %) long-term and short-term BDE-99 time-series, respectively, showed significant increasing trends. The mean annual change of long-term time-series of BDE-47 and BDE-99 were among the highest increase observed (Figure 5).

**Figure 8.** Pattern of change over time of BDE-47 in thick-billed murre from Prince Leopold Island in Canada (updated time-series published by Braune et al., (2015a). Red points = annual medians. Red-line = running three points average.

3.11 Hexabromocyclododecane (HBCDD)

HBCDD is used as an additive flame retardant and was listed in Annex A of the Stockholm Convention with specific exemptions in 2013 (UNEP/POPs/POPRC.6/13, 2010). Time-series were only available of the α-HBCDD diastereomer. All time-series deal with α-HBCDD in marine mammals or seabirds (one)
from Canada and Greenland; in total, seven (7) long-term time-series were available. Six (6; 86 %) of
those showed significant increasing trends, one of them also with a significant non-linear trend
component. HBCDD was the compound found with the highest annual mean increase (7.6 %).

3.12 Perfluoroalkyl substances (PFASs)

PFAS substances have had widely use in numerous industrial and commercial applications to make e.g.
products more stain-resistant and waterproof (Buck et al., 2011). Perfluorooctane sulfonic acid (PFOS),
its salts and perfluorooctane sulfonyl fluoride (PFOS-F) were added to Annex B (restriction) of the
Stockholm Convention on POPs in 2009 (http://chm.pops.int). Other PFASs such as perfluorooctanoic
acid (PFOA) and perfluorohexane sulfonic acid (PFHxS) are currently proposed for listing
(http://chm.pops.int). Time-series of several PFAS substances were available, however only substances
with more than ten (10) time-series are included here (PFOS, perfluorononanoic acid (PFNA),
perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUnA). This restriction was made
because the other PFAS compounds had several years with concentrations below limit of detection and
were limited to cover only a few species.

Sixteen (16) long-term time-series of PFOS were available of which nine (9; 56 %) had a significant non-
linear trend component. The others showed no trend except one also having a significant increasing
trend (Figure 2). A common trend was a concentration increase until about the mid-2000s followed by
decreasing concentrations (Figure 9). This trend is somewhat similar to the trend observed for BDE-47
and different from that of most organochlorine compounds. For the other compounds, about half of the
time-series showed significant increasing trends or significant increasing trends together with a
significant non-linear trend component (six (6) out of 11 (55%) PFNA, four (4) out of 10 (40 %) PFDA, six
(6) out of 11 (55 %) PFUnA). None of these compounds showed a significant decreasing trend indicating
that these compounds had not decreased as observed in several PFOS time-series.
Figure 9. Patterns of change over time of PFOS in ringed seal from West Greenland (red) and ringed seal (blue) and polar bear (black) from East Greenland. Points represent annual medians. Updated time-series published by Rigét et al., (2013a).

For the short-term time-series of the selected PFAS compounds, none showed significant increasing or decreasing trends except two increasing trends of PFNA.

3.13 Statistical power

In order to compare the statistical power of different time-series it is necessary to apply or define statistical standard requirements. We use the requirements to be able to detect a 5 % annual change with a statistical power of 80 % at $\alpha < 0.05$ (AMAP, 2016). Only 12 % of the long-term time-series fulfilled these criteria and for the short-time time-series this number decreased to 4 % (not shown). Thirteen percent of the organochlorine (OC) long-term time-series had 80 % or more power to detect a 5 % annual change with $\alpha < 0.05$.

This also means that there was still a large part (88 %) of the long-term time-series not fulfilling these statistical requirements. The number of years in time-series with data is proportional to the power of the time-series. Figure 10 shows the frequencies of required number of years with data in cases of the long-term time-series of OC to being able to fulfill the set of statistical requirements. Forty-five percent (45 %) of the OC time-series need more than 20 years of data to fulfill this requirement.

Figure 10. Histogram of the long-term OC time-series according to the number of years required to detect a 5 % annual change with 80 % power with $\alpha < 0.05$.

In order to compare the power of the long-term time-series among animal groups and tissues the power to detect a 5 % annual change with $\alpha < 0.05$ for a 10-year period was estimated together with the lowest detectable trend, which could be detected in a 10-year period with a power of 80 % (Table 2). The highest power and lowest detectable trend were seen for seabird eggs and marine mammal blubber and...
the lowest power was found for fish liver and muscle. This may be related to the lower POP concentrations in fish where analytical uncertainties play a relatively larger role.

**Table 2.** Mean and CV of the statistical power of long-term time-series and lowest detectable trend (LDT) for a 10-year period for each media.

<table>
<thead>
<tr>
<th></th>
<th>Mean (CV %) power</th>
<th>Mean (CV %) LDT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mussels, soft tissue</td>
<td>19.7 (94)</td>
<td>18.7 (43)</td>
</tr>
<tr>
<td>Freshwater fish</td>
<td>13.9 (69)</td>
<td>31.7 (93)</td>
</tr>
<tr>
<td>Liver</td>
<td>12.7 (43)</td>
<td>24.1 (41)</td>
</tr>
<tr>
<td>Muscle</td>
<td>14.2 (74)</td>
<td>34.0 (97)</td>
</tr>
<tr>
<td>Marine fish</td>
<td>15.7 (69)</td>
<td>21.7 (45)</td>
</tr>
<tr>
<td>Liver</td>
<td>16.8 (76)</td>
<td>21.2 (45)</td>
</tr>
<tr>
<td>Muscle</td>
<td>14.0 (54)</td>
<td>22.6 (38)</td>
</tr>
<tr>
<td>Seabirds</td>
<td>30.5 (74)</td>
<td>20.9 (140)</td>
</tr>
<tr>
<td>Blood</td>
<td>16.8 (30)</td>
<td>17.9 (48)</td>
</tr>
<tr>
<td>Egg</td>
<td>32.0 (73)</td>
<td>21.1 (69)</td>
</tr>
<tr>
<td>Liver</td>
<td>7.9 (-)</td>
<td>34.3 (-)</td>
</tr>
<tr>
<td>Marine mammals</td>
<td>22.2 (83)</td>
<td>24.1 (118)</td>
</tr>
<tr>
<td>Adipose tissue</td>
<td>16.7 (53)</td>
<td>22.8 (89)</td>
</tr>
<tr>
<td>Blubber</td>
<td>23.9 (84)</td>
<td>23.3 (121)</td>
</tr>
<tr>
<td>Liver</td>
<td>13.8 (84)</td>
<td>54.2 (104)</td>
</tr>
<tr>
<td>Muscle</td>
<td>18.5 (38)</td>
<td>16.0 (36)</td>
</tr>
</tbody>
</table>

**Discussion**

The approach of analyzing a large number of time-series of POPs applying the same robust statistical methodologies consistently to all available time-series provides an overview of trends in POP developments across a large geographical Arctic area. On this basis a meta-analysis has been performed, addressing the direction and rates of the trends as well as the consistency among species and locations. The time trend analyses of 1074 long-term time series (beginning before the year 2000) show that PCBs and organochlorine pesticides (OCs) have decreased in Arctic biota during the last 20 to 30 years. The same development of decreasing trends has been observed for most OCs in Arctic air (Hung et al., 2016). However, for HCB and β-HCH, the decrease in Arctic biota has been rather slow or in some locations, the concentrations even increased. Emissions and releases of HCB may continue to a larger degree than those of most other OCs, associated with by-production of HCB in chemical processes (Barber et al., 2005). This is supported by the trend of HCB in Arctic air, for which Hung et al., (2016) reported increasing concentrations or a very slow decline at three Arctic monitoring stations over the last decade.
The trend of β-HCH is quite different from those of α-HCH and γ-HCH, although α- and β-HCH followed the same emission pattern, peaking in the early 1980s (Li and Macdonald, 2005). The large differences observed in the biota time trends are likely related to differences in chemical properties, more specifically the Henry’s Law Constant, which means that α- and β-HCH partition differently between air and water, and the pathways to the Arctic are different (Li and Macdonald, 2005). The main pathway to the Arctic of β-HCH is by the ocean current while α-HCH is transported by air, causing a delay in the transport of β-HCH compared to α-HCH (Li et al., 2002). Furthermore, β-HCH is recalcitrant in most mammal species, while α-HCH and γ-HCH can be more readily metabolized (and so eliminated) (Moisey et al., 2001).

The relative few time-series of PCBs and OCs in Arctic biota extending back as far as the 1970s show that the downward trend of many of these compounds began decades before the Stockholm Convention on POPs entered into force. This is illustrated by the time-series of POPs in pike (Esox lucius) from a Swedish Arctic lake (Nyberg et al., 2014) and eggs of northern fulmar (Fulmaris glacialis) and thick-billed murre (Uria lomvia) from the Canadian Arctic (Braune et al., 2019). It also becomes evident when comparing the decrease rates in time-series starting after the year 2000 with those starting before the year 2000. The temporal trends of PCBs and OCs follow the schematic representation by Loganathan et al. (2016), where POP levels under national control and regulations in the 1980s and 1990s first decline relatively rapidly, followed by a long period with only slow declining concentrations. The global emissions of PCBs peaked in the mid-1970s, mainly related to the usage of PCB containing products (Breivik et al., 2002). This decrease in emissions is likely reflected in the first rapid decrease of PCBs in Arctic biota. The second phase of slow decrease might be related to secondary emissions e.g. volatilization from soil and vegetation (Ashraf, 2017) or from the ocean (as a results of sea-ice retreat and rising temperatures) (Ma et al., 2011) and potential releases by melting sea ice and glaciers (Bogdal et al., 2009). Furthermore, ongoing primary emissions exist from sources which are difficult to control (Diamond et al., 2010; Gasic et al., 2010), e.g. PCBs in building materials (Brown et al., 2016) and releases from e-waste (discarded electrical and electronic equipment) (Breivik et al., 2016). PCBs can also be formed unintentionally, e.g. in combustion processes (Ballschmiter et al., 1987), as reflected by their Annex C regulation in the Stockholm Convention. Despite these ongoing primary and secondary emissions the projected global atmospheric emissions in 2020 of 22 individual PCB congeners is predicted to be only ~2-5 % of the levels in 1970, when the emission peaked (Breivik et al., 2007). The time trend analysis indicates that current metabolisation and shedding (hair, feathers etc.) of most of the PCBs and OCs compounds in Arctic biota still exceeds these ongoing emissions, but that the rate of decrease today are minor.

The temporal trends of BDE-47 and PFOS were systematically different from those of many organochlorines as a period with increasing concentrations was followed by a period of a decreasing trend in many time series. For BDE-47, this is clearly seen in eggs of northern fulmar and thick-billed murre from the Canadian Arctic (Braune et al., 2015a): From 1975 to 2003, ΣPBDE concentrations increased exponentially followed by a rapid decrease to levels not significantly different from those from the early years. A similar trend was also found in ringed seals from East Baffin although the peak occurred later, i.e. in 2008 (Houde et al., 2017). Also in seabird eggs from the Pacific coast of Canada, the ΣPBDE concentrations peaked in the years around 2000 (Miller et al., 2014). The peak of the ΣPBDE
concentrations occurred at approximately the same time as the phase-out of the commercial Penta-BDE and Octa-BDE mixtures in the EU and the U.S. (EU, 2003; Tullo, 2003) and prior to inclusion of PBDEs in the Stockholm Convention in 2009. Abbasi et al. (2015) estimated the stocks of Penta- and Octa-BDE in in-use products in the U.S. and Canada to have peaked in 2004. Therefore, it may be interpreted that at least some Arctic animal populations responded rapidly to reduced production and/or use of PBDEs despite the wide occurrence of PBDEs in consumer products with potential lifetimes of several years and the persistency of PBDEs. In the review by de Wit et al., (2010) of PBDEs in the Arctic environment, a few time-series showed signs of leveling off or decline. Today this appears to be even more pronounced. Although the lag between national regulations and inclusion in the Stockholm Convention is much shorter than for PCBs and OCs, the time trends of BDE-47 also indicate a rapid reaction to national regulations. However, the decrease of PBDEs in biota was not yet observed in the Antarctic, indicating the relevance of proximity to sources (Markham et al., 2018).

This observation of a rapid response to regulations is also valid for PFOS: The decline of PFOS concentrations were connected to the phase-out of POSF-based chemicals by a large US company in 2000 (3M company, 2000). The decline in Arctic biota has been characterized as a rapid response to this phase-out, as documented for Canadian ringed seals (Butt et al., 2007), Northern sea otters (Enhydra lutris) (Hart et al., 2008) and Greenland ringed seals and polar bears (Rigét et al., 2013a). The trend of PFOS in Arctic biota may be expected to further decrease in the coming years. Based on modelling studies Zhang et al. (2017) estimated that in 2015 30% of the cumulative PFOS discharges from North America and Europe had entered the Arctic Ocean. The large-scale overturning of the North Atlantic Ocean transports PFOS to deep waters of the oceans, which is the terminal PFOS sink. The surface concentration was modeled to have decreased since year 2000, while the deep waters (> 1000 m) concentrations was modeled to have increased since 2000.

Although fewer time-series were available, the long-chain perfluoroalkyl carboxylic acids (PFCA) PFNA, PFDA and PFUnA did not show a similar decrease to that of PFOS, but rather a continuing increase. Increasing trends of long-chain PFCAs in Arctic biota were reported in several cases (e.g. Rotander et al., 2012; Braune and Letcher, 2013; Vorkamp et al., in press). Wang et al. (2014) estimated the global emissions of C6-C14 (PFCA) homologues to have increased in the period 1951-2002, followed by a decrease and then another increase in the period 2002-2012. Since 2002, the production sites of long-chain PFCAs and fluoropolymers has shifted from US, Japan and Western Europe to the continental Asia (Wang et al., 2014), which likely will change the spatial and temporal trends in Arctic biota in the future. Furthermore, PFOA, which was the main PFCAs in the emission inventories by Wang et al. (2014) has been reviewed for inclusion in the Stockholm Convention. The Review Committee concluded that inclusion in Annex A or B was recommended (UNEP/POPs/POPRC.13/7/Add.2, 2017).

The trend of α-HBCDD was different from those of PBDEs showing an increasing trend in five out of seven time-series. Increasing trends of α-HBCDD have also been reported for two subpopulations of beluga from Alaska (Hoguet et al., 2013) and in southwest Greenland peregrine falcon although not significantly (Vorkamp et al., 2018). Houde et al., (2017) studying several ringed seals populations from the Canadian Arctic found also increasing concentrations at several sites over the past decade. HBCDD
was listed for elimination in the Stockholm Convention in 2013, however, with specific exemptions that relate to its main use in expanded and extruded polystyrene (UNEP/POPs/POPRC.6/13, 2010). Furthermore, its presence in recycled polystyrene has been shown (Abdallah et al., 2018). These circumstances may still lead to primary emissions of HBCDD. However, time trends in Europe show decreasing concentrations of HBCDD (Esslinger et al., 2011), whereas HBCDD in fish increased, possibly following the phase-out of PBDEs (Chen et al., 2011). It may therefore be expected that the increasing trend observed in Arctic biota will turn to more stable levels or even a decreasing trend in the coming years.

As discussed above, some trends in levels of POPs in Arctic biota seem to be a direct response to changes in emission levels. However, multiple factors can affect the accumulation of POPs in Arctic biota. For example, climate changes affecting sea ice coverage, animal migration patterns and distribution areas, and changes in feeding habits can substantially affect the POP concentrations in biota. There is growing evidence of the importance of the interactions between climate change and POPs and that climate change affects contaminant trends as reviewed by Ma et al. (2016). The number of studies showing that climate change or variability influence the temporal trend of POPs in Arctic biota is increasing (e.g. Bustnes et al., 2010; Rigét et al., 2013b; Braune et al., 2015b; Cabrerizo et al., 2018). Including climate variables in temporal trend analyses of individual time-series are therefore advisable in ongoing and future time trend studies. Ecological tracers such as stable isotope and/or fatty acids are also useful for reliable trend interpretation. Isotopic data are useful to control for variation in trophic level over time and are widely incorporated into contaminant-monitoring (Hebert and Popp, 2018). Fatty acids is another dietary tracer that can provide insights to changes of food web structure over time (McKinney et al., 2013; 2017).

But also in the absence of climate variables and/or ecological tracers in temporal trends established so far, a large number of time-series covering a large geographical area and including different matrices show consistency and agreement in the POP trends. These results provide indications of the effectiveness of national and global controls. This indication is particularly strong in the time series of PBDEs and PFOS for which voluntary measures and regulations (as well as closely following concentration maxima) occurred during the monitoring phase. In both cases, relatively rapid changes in the trends took place. For the organochlorines, most regulations occurred prior to the establishment of the time-series. The subsequent concentration decreases are less clearly visible in biota as most time-series were established later, as discussed above.

The statistical power analyses showed that only 12 % of the long-term time-series would be able to detect a 5 % annual change with a statistical power of 80 % at $\alpha < 0.05$, and also that many years (more than 10) with data are needed before any firm statistical conclusion can be drawn. This emphasizes that large efforts in terms of sampling and chemical analyses have to be invested in monitoring temporal trends, especially in remote areas such as the Arctic. However, it should be noted that often the annual changes are larger than 5 % - maybe closer to 10 %, and that the statistical power is higher to detect a
10% annual change than a 5% annual change. Other monitoring programmes, such as the AMAP mercury trend assessments, previously drew similar conclusions (Bignert et al., 2004; Rigét et al., 2011).

The chosen statistical requirements of the power analyses have a large impact on the outcome of a time trend. A more precautionary approach with regard to protection of the environment would be to use a significance level of 10% in order to be able to give an early warning in a timely manner; i.e. in case of increasing trends of a contaminant. For the Greenlandic organochlorine time-series (in total 141), the number of time-series with 80% power to detect a 5% annual change would increase from 29% to 43% if the significance level was increased from 5% to 10%.

Sampling design, sample handling and analytical methods should be standardised as much as possible in temporal trend monitoring in order to avoid undesirable year-to-year variation. However, relatively long time-series are still needed because of the occurrence of a random year-to-year variance component beside a possible systematic trend component (Fryer and Nicholson, 1993). Random year-to-year variation in contaminant concentrations can arise from seasonal variations between years in e.g. prey item occurrence or climate variability.

In this study, bird eggs showed the highest statistical power of all animal tissues to detect trends. Using eggs in monitoring contaminants has several advantages over internal tissues (Furness and Greenwood, 2013 and references therein), which may add to the observed higher power. Eggs have a highly consistent composition, they can be sampled from the same location each year and there are no problems with confounding factors such as sex. The lowest power was found for fish liver and muscle and may be related to the rather low POP concentrations where analytical uncertainties play a relatively larger role.

Several time-series included in this study have a rather low power to detect temporal trends even if the number of years increase in the coming years. The objectives of individual trend monitoring should be stated based on sound statistical considerations and described in terms of quantitative measures, such as the number of years required to statistically detect a given rate of change, rather than the more general qualitative statements that are often employed. Furthermore, the trend study should be carefully designed and standardized as much as possible in order to be able to meet its objectives. This is especially important when working in the Arctic where sampling often is carried out in remote areas and is both costly and time consuming compared to working in temperate areas.

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