Science of the Total Environment 408 (2010) 2874–2884

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Temporal trends of legacy POPs in Arctic biota, an update☆

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ARTICLE INFO

*Article history:*
Received 13 May 2009
Received in revised form 10 July 2009
Accepted 20 July 2009
Available online 15 August 2009

*Keywords:*
Temporal trends
Arctic
POPs
Biota

ABSTRACT

A statistically robust method was applied to 316 time-series of 'legacy' persistent organic pollutants (POPs) in Arctic biota from marine, freshwater and terrestrial ecosystems with the purpose of generating a 'meta-analysis' of temporal trend data collected over the past two to three decades for locations from Alaska in the west to northern Scandinavian in the east. Information from recently published temporal trend studies was tabulated and comparisons were also drawn with trends in arctic air. Most of the analysed time-series of legacy POP compounds showed decreasing trends, with only a few time-series showing significantly increasing trends. Compounds such as α-HCH, γ-HCH and ΣDDT had a relatively high proportion of time-series showing significantly decreasing trends; ΣCHL had the lowest proportion. β-HCH was an exception, where long-range transport through the ocean, and not the atmosphere, may explain several increasing trends that were detected in the Canadian Arctic. Moving east from the Canadian Arctic there was a trend towards a greater proportion of significantly decreasing trends. Several time-series for DDE and ΣDDT showed significantly non-exponential trends, most often with a period of relative stability followed by a decrease. The median 'minimum detectable annual change within a 10-year period' for all of the time-series considered was 12% which did not meet the desirable level of statistical power capable of detecting a 5% annual change with a significance level of 5% within a 10-year period. The trends observed in the biota were consistent with decreasing trends of legacy POPs reported for Arctic air which appear to follow historic decreases in emissions. However, recent decreases in air are also starting to show signs of levelling off which may be an indication that atmospheric concentrations and, consequently those in the biota, are being less driven by primary sources and more by environmental processes and degradation.

## 1. Introduction

Persistent organic pollutants (POPs) are chemicals that have a long-lifetime in the environment, and therefore have the potential to be transported over long distances. Many POPs can enter food-webs, accumulating in wildlife and people. The Arctic is believed to act as a sink for such chemicals (Macdonald et al., 2000). Several global and regional conventions have been developed with the goal of eliminating or reducing emissions of persistent organic pollutants. The Stockholm Convention on Persistent Organic Pollutants addresses twelve priority POPs, while the POPs Protocol to the UN-ECE Convention on Long-range Transboundary Air Pollution (UN-ECE LRTAP) covers an additional four. POPs that have been banned or regulated are sometimes referred to as 'legacy' POPs, because present day contamination is largely a 'legacy' of past releases; this review deals with some of these legacy POPs. Despite the fact that use of these chemicals has either been phased-out or restricted, they are still found in the environment at levels that may cause negative effects to the health of individual animals and in some cases severe impacts on animal populations. Humans living in the Arctic and eating certain traditional foods can receive high dietary exposure to some legacy POPs and may also suffer adverse health effects from these compounds (Van Oostdam et al., 2005).

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 selected biota (indicator organisms) in the ecosystem. Temporal trend studies can also indicate whether regulatory actions aimed at reducing inputs of harmful chemicals to the environment are proving successful, or whether environmental levels are approaching threshold values. As indicated above, the objectives of temporal trend studies can differ and it should be stressed that an individual temporal trend study should be carefully designed to meet its intended objective based on sound statistical considerations. Also, the objectives should be

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☆ This paper is a contribution to the AMAP POPs assessment.
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doi:10.1016/j.scitotenv.2009.07.036

described in terms of quantitative measures, such as the number of years required to statistically detect a given rate of change etc., rather than the more general qualitative statements that are often employed.

The first circumpolar assessment of legacy POPs carried out by the Arctic Monitoring and Assessment Programme (AMAP) in 1997/98 contained largely baseline information, with very little information on temporal trend studies. At that time, only a few time-series for POPs in biota existed and, in most cases, these included only data from a few years. One recommendation from that assessment was to continue monitoring in order to obtain longer time-series (AMAP, 1998). By the of the second AMAP assessment of POPs, in 2002, the situation had improved with more and longer time-series datasets becoming available. That assessment concluded that, the levels of some legacy POPs were decreasing in most Arctic species included in temporal trend monitoring studies, but that the rates varied between locations and species studied (de Wit et al., 2004). Again, the recommendation from the 2002 AMAP assessment was to continue trend monitoring of POPs in key indicator biota, not least so that the results of these Arctic studies would be available for use in 'effectiveness' evaluations of the ENEP Stockholm Convention and UN ECE LRTAP POPs Protocol. Monitoring of Arctic biota has been complemented by atmospheric POPs monitoring at several air stations in the Arctic since 1992, providing a more direct measure of long-range atmospheric transport from source regions outside of the Arctic.

In this paper, we update the knowledge base regarding temporal trends of legacy POPs in Arctic biota. A statistically robust method has been applied to time-series data collected across a large part of the Arctic, covering terrestrial, freshwater and marine ecosystems. The data were provided to AMAP by several Arctic scientists, who are responsible for running national trend monitoring studies. These experts also provided guidance regarding potential confounding factors which should be taken into consideration. The advantage of this approach is that all time-series are treated with the same statistical method and therefore highly comparable results are generated. We also include a literature review of relevant temporal trend studies published within the past 4 to 5 years. In addition to analysis of trends in biota, a separate review of trends in arctic air has also been carried out (Hung et al. 2010-this issue), and is referred to periodically in this review to draw some comparison between atmospheric trends and those found in biota. Long-range transport through the atmosphere and deposition in the Arctic is the main pathway by which POPs are introduced to Arctic ecosystems (Macdonald et al., 2000). There are, however many processes that control uptake, accumulation and trophic transfer of POPs, all of which may change over time and can influence the biotic trends discussed here. Comparisons between trends in air and biota, therefore, are not meant to imply a direct functional link between the two media; rather the atmospheric trends are intended as a rough indicator of change in the source of POPs feeding Arctic ecosystems.

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 a monitoring data series have sufficient statistical power that the change of incorrectly concluding that no change has occurred when in fact it has is minimised. In the work reported here, the statistical power of the legacy POPs data series to detect temporal changes is also evaluated.

## 2. Statistical method for temporal trend analysis

Time trend analyses were performed on 316 time-series of legacy POPs (Table 1). Only time-series with at least 6 years of data and having data for years both before and after 2000 were included in this analysis. The compounds included were α-, β- and γ-HCH (lindane); total chlorobenzenes ($\Sigma$CBz = sum of tetra-, penta- and hexachlorobenzene) and hexachlorobenzene (HCB); total chlordanes ($\Sigma$CHL = sum of *trans*-nonachlor, *cis*-nonachlor, *trans*-chlordane, *cis*-chlordane and oxychlordane) as well as *trans*-nonachlor and heptachlor epoxide; total DDTs ($\Sigma$DDT = sum of *p,p'*-DDD, *p,p'*-DDE, *p,p'*-DDT) and *p,p'*-DDE; sum of 10 PCB (polychlorinated biphenyl) congeners ($\Sigma_{10}$PCB = sum of congeners 28, 31, 52, 101, 105, 118, 138, 153, 156, 180) and PCB-153; dieldrin; mirex and toxaphene (technical or sum of single congeners).

The distribution of data sets (see Fig. 1) by country was as follows:

| | |
|---|---|
| United States (Alaska) | 4 |
| Canada | 113 |
| Greenland | 64 |
| Faroe Islands | 35 |
| Iceland | 78 |
| Norway | 3 |
| Sweden | 19 |

The distribution by biota was:

| | |
|---|---|
| blue mussel | 55 |
| freshwater fish | 49 |
| terrestrial mammals | 7 |
| marine fish | 42 |
| seabirds | 46 |
| marine mammals (including polar bear). | 117 |

The data concerned different tissues and organs, depending on the species.

The analyses of temporal trends followed the procedure used in temporal trend assessments of ICES (International Council for the Exploration of the Sea). The method employs a robust regression-based analysis to detect temporal trends (Nicholson et al., 1998). Annual median concentrations were used as the yearly contaminant index value. The median was preferred over the geometric mean because it is less sensitive to outliers and is not influenced by how different laboratories report less than detection limit values. The total variation in the contaminant index values over time is divided into a linear and a non-linear component. Log-linear regression analysis was applied to describe the linear component and a simple 3-point running mean smoother was applied to describe the non-linear component. The linear and non-linear components were tested by means of an ANOVA. A detailed worked example of this temporal trend analysis is provided by Nicholson et al. (1998). The use of smoothers in temporal trend analyses is described by Fryer and Nicholson (1999). The statistical package employed by AMAP for its temporal trend analyses is available from: http://amap.no/documents/index.cfm?dirsub=%2FPIA%20Trend%20Analysis%20Package&sort=default.

The power of the temporal trend test applied in this paper is discussed and described in detail by Fryer and Nicholson (1993). They advocate the incorporation of random between-year variation in addition to the within-year variation in the error structure of the trend analysis. They also showed that the results of, e.g., regressions of contaminant level on year of collection could be misleading if a random between-year variation was present. The random (i.e. thus far unexplained) between year-variation may derive from fluctuations in the timing of physiological or behavioural changes that may be associated with contaminant levels in biota even if samples are collected at the same time in each year.

The time trend data used in this analysis were supplied to the AMAP POPs assessment group by national key experts. Data originators decided whether their data should be adjusted for covariables such as age/size and sex, and whether the analyses should be done on lipid weight, wet weight or dry weight concentrations.

Additionally, results of time trends studies of legacy POPs in Arctic biota published in the scientific literature during the past 4 to 5 years have been compiled. It should be noted that a number of the data series included both in the temporal trend analyses are also reported

**Table 1**
Summary of results from the statistical time trend analyses.

| Compound | *N* of time-series | Sign linear increasing trend | Sign non-linear increasing trend | Sign linear decreasing trend | Sign non-linear decreasing trend | No trend | Non-monotonic trend | Many below QA level prevent analysis | Mean annual (%) change and (SE) | Mean lowest detectable trend (%) in 10 years | Mean power (%) in 10 years to detect 5% with sign 5% |
|---|---|---|---|---|---|---|---|---|---|---|---|
| $\Sigma_{10}$PCB | 16 | | | 5 | 1 | 7 | 3 | | −1.9 (0.8) | 9.4 | 36 |
| PCB-153 | 40 | 3 | | 12 | 4 | 21 | | | −1.2 (0.9) | 11.9 | 28 |
| ΣDDT | 19 | | | 6 | 4 | 7 | 1 | 1 | −4.4 (0.7) | 12.4 | 31 |
| *p,p'*-DDE | 40 | 1 | | 6 | 5 | 21 | 6 | 1 | −1.9 (0.8) | 14.6 | 24 |
| α-HCH | 32 | | | 15 | 4 | 13 | | | −7.4 (0.9) | 13.9 | 26 |
| β-HCH | 24 | 2 | 1 | 4 | 2 | 10 | 2 | 3 | −2.9 (1.2) | 20.5 | 27 |
| γ-HCH | 17 | | | 8 | 4 | 3 | | 2 | −7.3 (1.5) | 17.3 | 31 |
| ΣCBz | 3 | | | 1 | | 2 | | | −4.0 (3.5) | 13.5 | 38 |
| HCB | 40 | | | 13 | 1 | 25 | 1 | | −2.5 (0.6) | 12.5 | 32 |
| ΣCHL | 17 | | | 2 | | 15 | | | −1.8 (0.7) | 10.5 | 33 |
| *trans*-nonachlor | 29 | 1 | | 5 | | 19 | 4 | | −1.0 (0.8) | 17.5 | 26 |
| Heptachlor Epoxide | 9 | | | | | 4 | 2 | 3 | −0.7 (0.6) | 12.1 | 33 |
| Dieldrin | 11 | | | 3 | 1 | 6 | 1 | | −2.1 (0.8) | 9.2 | 47 |
| Mirex | 12 | | | 4 | 1 | 6 | 1 | | −0.7 (1.1) | 13.8 | 26 |
| Toxaphene | 10 | | | 2 | | 7 | 1 | | −0.8 (2.5) | 13.9 | 33 |

SE = standard error.

in the published literature, though in some cases covering different time periods and using different statistical analysis methods.

## 3. Temporal trends of legacy POPs

### 3.1. PCB-153 and $\Sigma_{10}$PCB (sum of congeners 28,31,52,101,105,118, 138,153,156,180)

In total, 40 time-series of PCB-153 and 16 time-series of $\Sigma_{10}$PCB were available (Table 1). The mean annual change for all time-series analysed was an annual decrease of 1.2% (PCB-153) and 1.9% ($\Sigma_{10}$PCB). 40% of the PCB-153 and 38% of the $\Sigma_{10}$PCB time-series showed statistically significant decreasing trends. Most of the significant trends were best described by a log-linear decrease (i.e., an exponential decrease on a non-log concentration scale). Significant decreasing trends and non-significant trends were found across the Arctic area covered by the studies (Fig. 2). Significant decreasing trends were seen in freshwater fish populations from northern Canada and northern Sweden and in one time-series of marine fish from Iceland. Several time-series of PCB-153 in blue mussels from around Iceland showed significant decreasing trends, however one time-series from a fjord system showed a significant increase. Decreasing trends were also found in seabirds from the Canadian Arctic Archipelago, in marine mammals from Greenland and terrestrial mammals (reindeer) from Sweden. Significant increasing trends of PCB-153 concentrations were found in two time-series beside the above-mentioned blue mussel dataset from Iceland; i.e. one in a freshwater fish population in Canada and one in a marine mammal population from the Faroe Islands.

The majority of time trends in the literature report significantly decreasing trends for PCBs (Table 2). Beside the above-mentioned time-series, significant decreasing trends are reported in three lake trout populations in Canada while one of three burbot populations from the same Canadian lakes showed an increasing trend (Ryan et al., 2005). Decreasing trends were also found in seabird populations from Canada, Iceland and northern Norway, and in marine mammal populations from Canada and Svalbard, Norway (Table 2). No significant changes were found for non-*ortho*-, mono-*ortho*- and di-*ortho*-substituted-PCBs in ringed seals from Holman, Canada, during the period 1981 to 2000 (Addison et al., 2005). However, ΣPCB levels decreased markedly in this ringed seal population earlier, from 1972 to 1981 (Braune et al., 2005). In ringed seals from Lancaster Sound, Canada, ΣPCB and ΣDDT levels have steadily declined similar to many of the time-series analysed here, and paralleled well the global trends in emissions, which for PCBs peaked around 1970, compared with the 1960s for DDTs (Lohmann et al., 2007). In northern Swedish reindeer, significantly decreasing trends were reported for PCB-118, PCB-138, PCB-153 and PCB-180 (Bignert et al., 2008). Air monitoring at the four long-term Arctic stations of Alert, Pallas, Zeppelin and Storhofdi, showed decreasing trends for all 10 PCB congeners, with half-lives generally ranging between three and ten years (Hung et al. 2010-this issue). It should be noted, however, that the trends were not consistent over the entire time period and there were some cases where the calculated half-life of the congener was significantly higher than 10 years.

### 3.2. DDE and ΣDDT (sum of p,p'-DDE, p,p'-DDD, p,p'-DDT)

Forty and 19 time-series were available for *p,p'*-DDE and ΣDDT, respectively (Table 1). In the case of reindeer from Sweden, data were not suitable for analyses of time trends due to the number of values below level of quantification. The mean change for all time-series was an annual decrease of 1.9 and 4.4% for DDE and ΣDDT, respectively. Statistically significant decreasing trends were found in 28% (DDE) and 53% (ΣDDT) of the time-series with approximately half showing a log-linear decrease and the other half a non-linear trend. Several of the time-series showing a non-linear trend exhibited a more or less stable level during the first part of the time period followed by a decrease towards the end of the period. Significant decreasing trends were found in freshwater fish populations from northern Sweden while only one of the time-series for freshwater fish in northern Canada showed a significantly decreasing trend for ΣDDT. One marine fish population from Iceland and one from Greenland showed a significant decreasing trend for DDE and ΣDDT, respectively. Decreasing trends were also found in seabird populations from Arctic Canada, and among marine mammal populations from Arctic Canada, Greenland and the Faroe Islands. Of the eleven Icelandic blue mussel time-series, only one showed a significant decreasing trend while another showed a significant increasing trend; the latter being the time-series that also showed increasing PCB-153. These increasing trends have been noted by the Icelandic monitoring agency, but as yet no explanation for the increases has been found.

The majority of time trends in the literature report significant decreasing trends (Table 2). Beside the above-mentioned data, significantly decreasing trends in ΣDDT concentrations were found in three lake trout populations from Arctic Canada (Table 2). Burbot

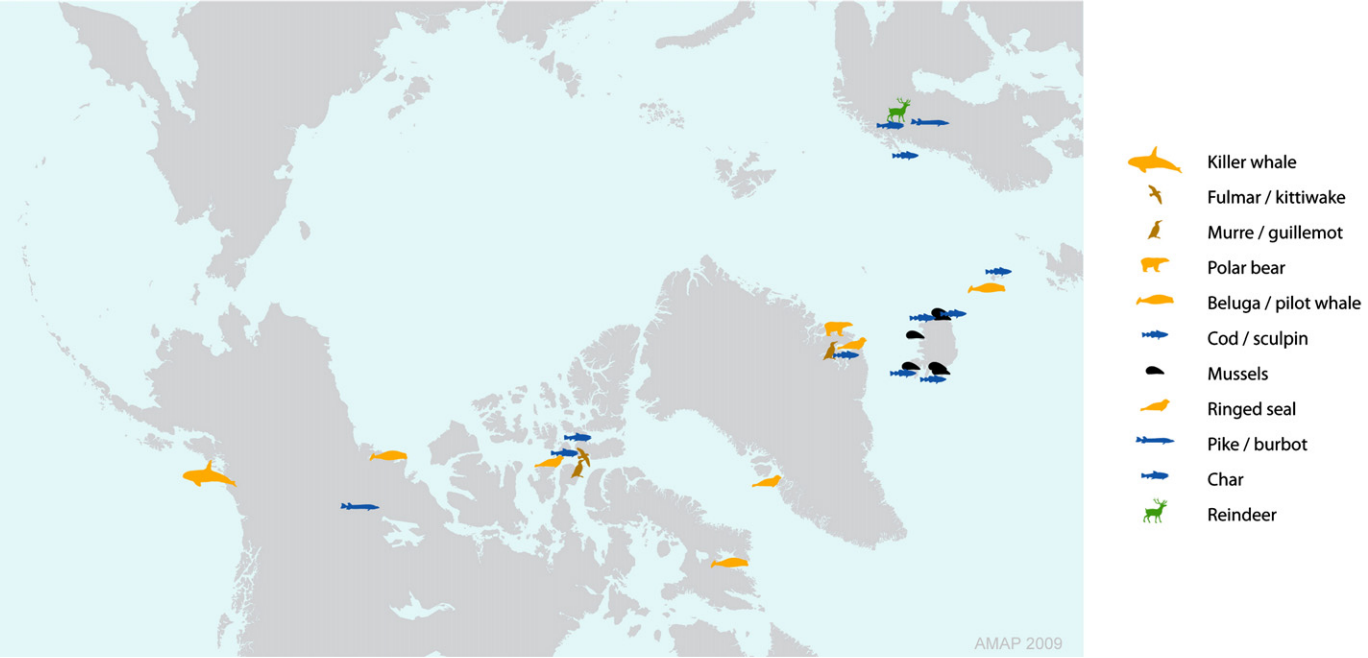

**Fig. 1.** Map of locations with POP time-series data.

sampled from eight Russian Arctic rivers near their outflows to the Arctic Ocean also showed $\Sigma$DDT levels decreasing during the period 1988 to 1994 (Zhulidov et al., 2002). DDD concentrations, but not DDE, showed a decreasing trend in Arctic cod from northern Norway over the period 1987–1998 (Sinkkonen and Paasivirta, 2000). Decreasing trends of DDE in seabirds are reported from Alaska, Canada, Iceland and northern Norway except of glaucous gull from Svalbard (Table 2). In marine mammals, decreasing $\Sigma$DDT levels from Canada are reported in beluga from St. Lawrence, ringed seals from Lancaster Sound and Holman Island, and polar bears from Hudson Bay (Table 2). Hung et al. (2010-this issue) report that concentration of DDT related compounds in arctic air were generally quite close to the detection limit and did not display a discernable trend at three of the four long-term monitoring stations. Zeppelin was the exception, where DDT related compounds decreased between 1993 and 2006 with half-lives ranging from four to eleven years.

### 3.3. $\alpha$-HCH

In total, 32 time-series of $\alpha$-HCH were analysed (Table 1). The mean change for all time-series was an annual decrease of 7.4%. Fifty-nine percent of the time-series showed a significantly decreasing trend and, in most cases, the trend could be described by an exponential decrease. Significant decreasing trends were seen in freshwater fish populations from northern Canada and northern Sweden. One of eleven blue mussel time-series from Iceland showed significantly decreasing trends. Also marine fish time-series from Iceland and Greenland showed a significant decrease. In seabirds and marine mammals, significantly decreasing trends were found in populations from Arctic Canada and Greenland. Also, reindeer from northern Sweden, the only available time-series from the terrestrial ecosystem, showed a significant decrease.

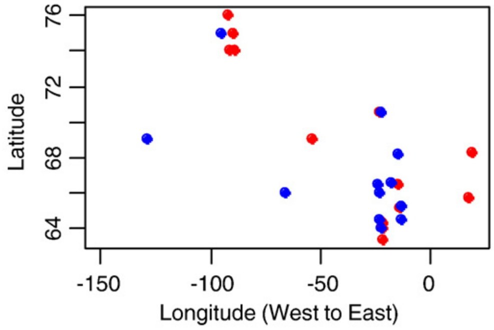

**Fig. 2.** Geographical distribution of significantly decreasing trends of PCB-153 (red) and non significant trends (blue).

The majority of time trends in the literature report significantly decreasing trends. Beside the above mentioned time-series, significantly decreasing trends are reported from one marine fish population from northern Norway, one seabird population from Iceland and additional marine mammal populations from Arctic Canada (Table 2). The sum of $\alpha$- and $\gamma$-HCH concentrations in burbot liver from eight Arctic Russian rivers showed decreasing trends during the period 1988 to 1994 (Zhulidov et al., 2002). However, in ivory gull eggs from the Canadian Arctic, no trend was found for the sum of $\alpha$- and $\gamma$-HCH concentrations. Consistent with the trends in biota, atmospheric concentrations of $\alpha$-HCH have also decreased at all four of the long-term air monitoring stations in the Arctic. Results from Alert, however, suggest that the decrease may have slowed since 2001 (Hung et al. 2010-this issue).

### 3.4. $\beta$-HCH

In total, 24 time-series were available, 25% of which showed a significantly decreasing trend (Table 1). In three cases, time trend analyses could not be performed as several of the annual median values were below the level of quantification, especially for freshwater fish populations. The mean change for the 21 time-series analyzed showed an annual decrease of 2.9%. Decreasing trends were seen in marine fish from Iceland, marine mammals from Greenland, and reindeer from Sweden, while increasing trends were seen among seabird populations and one marine mammal population from Arctic Canada. The percent annual decrease of $\beta$-HCH increased significantly

**Table 2**
Legacy POPs temporal trend literature review.

| Area | Species | Tissue | Period (*n*) | Trend | Significantly trend according to author | |
|---|---|---|---|---|---|---|
| *PCBs (number of congeners included varies)* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in two lakes | Muscle | 1993–2003(4,6) | → | Not sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1992–2002(4) | ↑ | Sign | Ryan et al. (2005) |
| Holman, Canada | Ringed seal | Blubber | 1972–2001(4) | ↓ | Sign | Braune et al. (2005) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | → | Not sign | Braune et al. (2005) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | ↓ | ? | Braune et al. (2007) |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| Lancaster Sound, Canada | Ringed seal | Blubber | 1975–2006(8) | ↓ | ? | Lohmann et al. (2007) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Not sign | Vorkamp et al. (2008) |
| East Greenland | Ringed seal | Blubber | 1986–2004(8) | ↓ | Sign | Rigét et al. (2006) |
| Svalbard, Norway, PCB-153 | Glaucous gulls | Blood | 1995–2004 | ↓ | | Verreault et al. (2006) |
| Svalbard, Norway, PCB-153 | Polar bear | Blood | 1990–2002 | ↓ | | Verreault and Gabrielsen (2006) |
| Northern Norway | Herring gull, Puffin, Kittiwake, Common guillemot | Egg | 1983–2003(3) | ↓ | Sign | Helgason et al. (2008) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | → | Not sign | Sinkkonen and Paasivirta (2000) |
| Iceland, PCB-153 | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| Sweden, PCB-118,138,153,180 | Reindeer | Muscle | 1987–2006(16) | ↓ | Sign | Bignert et al. (2008) |
| | | | | | | |
| *ΣDDT* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in two lakes | Muscle | 1993–2003(4,6) | → | Not sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1992–2002(4) | ↑ | Sign | Ryan et al. (2005) |
| Holman, Canada | Ringed seal | Blubber | 1972–2001(5) | ↓ | Sign | Braune et al. (2005) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | ↓ | ? | Braune et al. (2007) |
| St. Lawrence, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| Lancaster Sound, Canada | Ringed seal | Blubber | 1975–2006(8) | ↓ | ? | Lohmann et al. (2007) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | ↓ | Sign | Braune et al. (2005) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Sign | Vorkamp et al. (2008) |
| | | | | | | |
| *p,p'-DDE* | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | ↓ | Sign | Pol et al. (2004) |
| St George, Alaska | Common murre | Egg | 1973–1999(2) | ↓ | Sign | Pol et al. (2004) |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| Svalbard, Norway | Glaucous gulls | Blood | 1995–2004 | ↑↓ | | Verreault et al. (2006) |
| Northern Norway | Herring gull, Puffin, Kittiwake, Common guillemot | Egg | 1983–2003(3) | ↓ | Sign | Helgason et al. (2008) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | → | Not sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *p,p'-DDD* | | | | | | |
| Lancaster Sound, Canada | Fulmar | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | ↓ | Sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *p,p'-DDT* | | | | | | |
| Lancaster Sound, Canada | Fulmar | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| | | | | | | |
| *ΣHCH* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in two lakes | Muscle | 1992–2003(4,6) | → | Not sign | Ryan et al. (2005) |
| Holman, Canada | Ringed seal | Blubber | 1981–2001(4) | → | Not sign | Braune et al. (2005) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | → | ? | Braune et al. (2007) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | ↓ | Sign | Braune et al. (2005) |
| Northern Norway | Herring gull, Puffin, Kittiwake, Common guillemot | Egg | 1983–2003(3) | ↓ | Sign | Helgason et al. (2008) |
| | | | | | | |
| *α-HCH* | | | | | | |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| Cumberland Sound, Canada | Beluga | Blubber | 1982–2002(5) | ↓ | Sign | Braune et al. (2005) |
| Lancaster Sound, Canada | Ringed seal | Blubber | 1975–2006(8) | ↓ | ? | Lohmann et al. (2007) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | ↓ | ? | Braune et al. (2005) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Sign | Rigét et al. (2008) |
| East Greenland | juvenile ringed seal | Blubber | 1986–2006(9) | ↓ | Sign | Rigét et al. (2008) |
| East Greenland | adult ringed seal | Blubber | 1994–2006(8) | ↓ | Sign | Rigét et al. (2008) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | ↓ | Sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *β-HCH* | | | | | | |
| Lancaster Sound, Canada | Kittiwake | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↑ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Ringed seal | Blubber | 1975–2006(8) | ↑↓ | | Lohmann et al. (2007) |
| Cumberland Sound, Canada | Beluga | Blubber | 1982–2002(5) | ↓ | Sign | Braune et al. (2005) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | → | Not sign | Braune et al. (2005) |

**Table 2** (*continued*)

| Area | Species | Tissue | Period (*n*) | Trend | Significantly trend according to author | |
|---|---|---|---|---|---|---|
| *β-HCH* | | | | | | |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Sign | Rigét et al. (2008) |
| East Greenland | juvenile ringed seal | Blubber | 1986–2006(8) | → | Not sign | Rigét et al. (2008) |
| East Greenland | adult ringed seal | Blubber | 1994–2006(7) | ↓ | Not sign | Rigét et al. (2008) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| | | | | | | |
| *γ-HCH* | | | | | | |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1993–2002(4) | ↓ | Sign | Ryan et al. (2005) |
| St. Lawrence, Canada | Beluga male | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| St. Lawrence, Canada | Beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| Cumberland Sound, Canada | Beluga | Blubber | 1982–2002(5) | → | Not sign | Braune et al. (2005) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Sign | Rigét et al. (2008) |
| East Greenland | juvenile ringed seal | Blubber | 1986–2006(9) | ↓ | Sign | Rigét et al. (2008) |
| East Greenland | adult ringed seal | Blubber | 1994–2006(8) | ↓ | Not sign | Rigét et al. (2008) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | ↓ | Sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *ΣCBz* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in two lakes | Muscle | 1993–2003(4,6) | → | Not sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1992–2002(4,) | ↑ | Sign | Ryan et al. (2005) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | ↑↓ | | Braune et al. (2005) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | ↓ | ? | Braune et al. (2007) |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| | | | | | | |
| *HCB* | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | → | Not sign | Pol et al. (2004) |
| St George, Alaska | Common murre | Egg | 1973–1999(2) | → | Not sign | Pol et al. (2004) |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| St. Lawrence, Canada | Beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Sign | Vorkamp et al. (2008) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | ↓ | Sign | Ólafsdóttir et al. (2005) |
| Svalbard, Norway | Glaucous gulls | Blood | 1995–2004 | → | | Verreault et al. (2006) |
| Northern Norway | Herring gull, Puffin, Kittiwake, Common guillemot | Egg | 1983–2003(3) | ↓ | Sign | Helgason et al. (2008) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | → | Not sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *ΣCHL* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1993–2003(6) | → | Not sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1992–2002(4) | ↑ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1993–2002(4) | ↓ | Sign | Ryan et al. (2005) |
| Holman, Canada | Ringed seal | Blubber | 1981–2001(3) | ↑ | | Braune et al. (2005) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | → | ? | Braune et al. (2007) |
| Lancaster Sound, Canada | Kittiwake | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| Hudson Bay, Canada | Polar bear | Adipose | 1968–2002(12) | ↑↓ | | Braune et al. (2005) |
| West Greenland | Ringed seal | Blubber | 1999–2006(6) | ↓ | Not sign | Vorkamp et al. (2008) |
| Vestertana Fjord, Norway | Arctic cod | Liver | 1987–1998(9) | → | Not sign | Sinkkonen and Paasivirta (2000) |
| | | | | | | |
| *cis-Chlordane* | | | | | | |
| Lancaster Sound, Canada | Kittiwake, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| St. Lawrence, Canada | Beluga female | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| | | | | | | |
| *trans*-Chlordane | | | | | | |
| St. Lawrence, Canada | Beluga male | Blubber | 1986/87–2002(14) | ↓ | Sign | Lebeuf et al. (2007) |
| St. Lawrence, Canada | Beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| | | | | | | |
| *trans-Nonachlor* | | | | | | |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| West Greenland | Ringed seal | Blubber | 1994–2006(7) | ↓ | Not sign | Vorkamp et al. (2008) |
| Iceland | Black guillemot | Muscle | 1976–1996(13) | → | Not sign | Ólafsdóttir et al. (2005) |
| | | | | | | |
| *cis*-Nonachlor | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | ↓ | Sign | Pol et al. (2004) |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| | | | | | | |
| *Oxychlordane* | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | → | Not sign | Pol et al. (2004) |
| St George, Alaska | Common murre | Egg | 1973–1999(2) | → | Not sign | Pol et al. (2004) |
| Lancaster Sound, Canada | Kittiwake | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| *Lancaster Sound, Canada* | Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| Iceland | Black guillemot | Muscle | 1976–1992(12) | → | Not sign | Ólafsdóttir et al. (2005) |

(*continued on next page*)

**Table 2** (*continued*)

| Area | Species | Tissue | Period (*n*) | Trend | Significantly trend according to author | |
|---|---|---|---|---|---|---|
| *Oxychlordane* | | | | | | |
| Svalbard, Norway | Glaucous gulls | Blood | 1995–2004 | ↑↓ | | Verreault et al. (2006) |
| Northern Norway | Herring gull, Puffin, Kittiwake, Common guillemot | Egg | 1983–2003(3) | ↓ | Sign | Helgason et al. (2008) |
| | | | | | | |
| *Heptachlor epoxide* | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | → | Not sign | Pol et al. (2004) |
| St George, Alaska | Common murre | Egg | 1973–1999(2) | ↓ | Sign | Pol et al. (2004) |
| Lancaster Sound, Canada | Kittiwake | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar | Egg | 1975–2003(7) | ↑ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| | | | | | | |
| *Dieldrin* | | | | | | |
| Bogoslof, Alaska | Common murre | Egg | 1973–2000(2) | → | Not sign | Pol et al. (2004) |
| St George, Alaska | Common murre | Egg | 1973–1999(2) | → | Not sign | Pol et al. (2004) |
| Seymour Island, Canada | Ivory gull | Egg | 1976–2004(3) | ↓ | ? | Braune et al. (2007) |
| Lancaster Sound, Canada | Kittiwake, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| Cumberland Sound, Canada | Beluga | Blubber | 1982–2002(5) | ↓ | ? | Braune et al. (2005) |
| | | | | | | |
| *ΣMirex (sum of mirex and photomirex)* | | | | | | |
| Lancaster Sound, Canada | Kittiwake | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| Lancaster Sound, Canada | Fulmar, Thick-billed murre | Egg | 1975–2003(7) | → | Not sign | Braune (2007) |
| | | | | | | |
| *Mirex* | | | | | | |
| Lancaster Sound, Canada | Kittiwake, Fulmar, Thick-billed murre | Egg | 1975–2003(7) | ↓ | Sign | Braune (2007) |
| St. Lawrence, Canada | Beluga male, beluga female | Blubber | 1986/87–2002(14) | → | Not sign | Lebeuf et al. (2007) |
| | | | | | | |
| *Toxaphene (sum of chlorinated bornanes, number of congeners varies)* | | | | | | |
| Yukon lakes, Canada | Lake trout in three lakes | Muscle | 1992–2003(4,4,6) | ↓ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1993–2003(6) | ↑↓ | | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1992–2002(4) | ↑ | Sign | Ryan et al. (2005) |
| Yukon lakes, Canada | Burbot in one lake | Muscle | 1993–2002(4) | → | Not sign | Ryan et al. (2005) |
| West Greenland | Ringed seal | Blubber | 2000–2006(6) | ↓ | Not sign | Vorkamp et al. (2008) |

(linear regression, $p = 0.03$) from west to east while no trend with longitude was found for α-HCH (linear regression, $p = 0.56$) (Fig. 3).

In addition to the above-mentioned time-series, the literature reports significant decreasing trends in beluga from Cumberland Sound, Canada, and in black guillemots from Iceland (Table 2). Ringed seal from Lancaster Sound, Canada, showed an increasing trend before 1995 followed by a decrease (Lohmann et al., 2007).

The difference in the observed pattern of the time trends between α- and β-HCH may be explained by the different pathways of these two isomers to the Arctic. Air concentrations of α-HCH responded rapidly to reductions in global HCH emissions whereas atmospheric data for β-HCH do not reflect global emission patterns (Li and Macdonald, 2005). Due to the different physical and chemical properties of the two isomers, β-HCH partitions more strongly into water than α-HCH (Li and Macdonald, 2005) resulting in a delayed arrival of β-HCH into the Arctic via ocean currents, particularly through the Bering Strait. The fact that the major pathway for β-HCH to the Arctic is via ocean currents would also explain why concentrations in freshwater fish and terrestrial (i.e. reindeer) populations were below quantification levels. Concentrations of β-HCH are highest in the Bering Sea, decreasing northward into the Chukchi Sea, and decrease northward (Li and Macdonald, 2005). From the Bering Strait, β-HCH is transported eastward into the Beaufort Sea and then to the Canadian Archipelago. Furthermore, unlike α- and γ-HCH which are readily metabolized (Willet et al., 1998), β-HCH appears to be recalcitrant in most biota leading to food web biomagnification (Hoekstra et al., 2003) and a slower response to changes in levels in the abiotic environment. The overall west to east pattern for β-HCH trends in biota shown in Fig. 2 has been discussed in more detail in the recent literature (Rigét et al., 2008; Braune, 2007; Li and Macdonald, 2005). It should be noted that Hung et al. (2010-this issue) do not report trends for β-HCH.

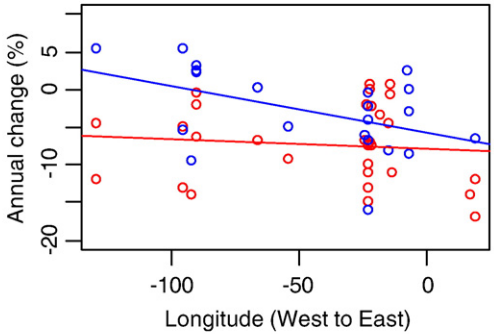

**Fig. 3.** Annual change (%) of α-HCH (red) and β-HCH (blue) versus longitude.

### 3.5. γ-HCH

The mean change among 17 time-series for γ-HCH was an annual decrease of 7.3% (Table 1). In two cases, time trend analyses could not be performed as several of the annual median values were below the level of quantification. The majority (71%) of time-series showed a decreasing trend and the mean rate of decrease was among the highest, along with that for α-HCH, found for the legacy POP compounds (Table 1). Although γ-HCH is still used in some areas of the world, compared with the α-HCH isomer it has a shorter lifetime in the atmosphere and may even be photochemically transformed to α-HCH (Ding et al., 2007). Also, γ-HCH is rapidly metabolized (Willet et al., 1998) limiting its detection in the upper trophic levels of the food web. Decreasing trends were found in Arctic char populations from Canada and Sweden, and in marine fish populations from Iceland and Greenland. Also, in seabird and marine mammal populations from Arctic Canada and Greenland, decreasing trends were observed.

In addition to the above-mentioned time-series, the literature reports significant decreasing trends in Arctic cod from northern Norway, black guillemot from Iceland, and male beluga from St. Lawrence Estuary, Canada (Table 2). Hung et al. (2010-this issue) also report decreases in γ-HCH from each of the long-term air monitoring stations and they note an acceleration in this trend at Alert and Zeppelin after 2002, which is around the same time that Canada implemented a series of restriction on lindane use, eventually banning it entirely, between 2001 and 2004. These trends, particularly at Zeppelin, were also likely influenced by use of lindane in Europe which prior to a complete cessation of use in 1998 had been a major user (Li and Bidleman, 2003).

### 3.6. ΣCBz (sum of tetra-, penta- and hexachlorobenzene) and HCB

The mean change for 3 ΣCBz (all from Canada) and 40 HCB time-series was an annual decrease of 4% and 2.5% for ΣCBz and HCB, respectively (Table 1). One ΣCBz time-series and 35% of the HCB time-series showed statistically significant decreasing trends and for all except one, the trend could be described as a log-linear (i.e. exponential) decrease. The remaining time-series showed no significant trend. Significant decreasing trends were seen among freshwater fish from northern Canada and northern Sweden and among marine fish populations from Iceland and Norway. Seabird populations from Arctic Canada also showed a significantly decreasing trend. Among the available time-series for HCB in marine mammal populations, only ringed seals from Greenland showed significantly decreasing trends.

Beside the above-mentioned time-series, significant decreasing trends are reported in seabirds from northern Canada and northern Norway. No trend was seen in seabirds from Alaska and in glaucous gull from Svalbard, Norway. Decreasing trend is reported in male beluga from St. Lawrence Estuary, Canada (Table 2). In the recent review of HCB in the global environment, Barber et al. (2005) found that, although dependent on media and locations studied, a consistent downward trend has been seen in the environment over the past 20 years from peaks occurring in the 1960s. However, based on observed opposite trends in abiotic media, such as lake sediments, ice core and precipitation in the Arctic, Barber et al. (ibid) also suggested that the poles may still act as sinks for the global movement of HCB. Hung et al. (2010-this issue) report that HCB concentrations decreased in arctic air through the 1990s but since 2000 have shown a slight increase at Alert and Zepplin. The increasing trend cannot be directly attributed to increased emissions as HCB was largely banned as a pesticide during the 1970s, however, it is a known contaminant in some current use pesticides that have seen increasing growth since 2000. Alternatively, Hung et al. hypothesize that increasingly ice-free arctic sea-water may be acting as a source to the atmosphere, thereby influencing the atmospheric trend. The general trend in biota, however, appears to follow the longer term trend in air, which is decreasing.

### 3.7. ΣCHL (sum of trans-nonachlor, cis-nonachlor, trans-chlordane, cis-chlordane and oxychlordane), trans-nonachlor and heptachlor epoxide

The mean change for 17 ΣCHL time-series analysed was an annual decrease of 1.8%, and for 29 *trans*-nonachlor time-series an annual decrease of 1.0% (Table 1). The mean change for 6 heptachlor epoxide time-series, all from Canada, was an annual decrease of 0.7%. Twelve and 26% of the time-series showed significant decreasing trends for ΣCHL and *trans*-nonachlor, respectively. None of the heptachlor epoxide time-series showed a significant non-linear decreasing trend. All significantly decreasing ΣCHL or *trans*-nonachlor time-series could be described as log-linear trends. One time-series of blue mussels from Iceland showed a significant increasing trend of *trans*-nonachlor; the same series that also showed increasing trends of PCB-153 and *p,p'*-DDE. The remaining time-series showed no significant trend. Significant decreasing trends were seen in freshwater fish populations from northern Canada and marine fish populations from Iceland. One population of marine mammals from Greenland showed a significant decreasing trend. No significant trends were observed in marine mammals, neither from the Faroe Islands, nor in seabirds and marine mammals from Arctic Canada despite nine time-series of both ΣCHL and *trans*-nonachlor being available.

Burbot from three lakes in the Yukon, Canada, showed opposite trends for ΣCHL concentrations; however the authors suspected that individual ecosystem dynamic and biotic factors such as condition factor and lipid content mass rather than atmospheric input were the primary factors affecting the contaminant concentrations (Ryan et al., 2005). In eggs of black-legged kittiwakes from Lancaster Sound, Canada, *cis*-chlordane, oxychlordane and the sum of five chlordanes were reported to decrease, whereas no trends were found for the sum of five chlordanes in eggs of fulmars and thick-billed murres from the same area (Table 2). In black guillemots from Iceland no trends were found for *trans*-nonachlor and oxychlordane. In common murres from Alaska, *cis*-nonachlor was reported as decreasing whereas no trend was observed for oxychlordane. In four different seabird species from northern Norway, oxychlordane was reported as decreasing. However no trend was found in glaucous gull from Svalbard. Significant decreasing trends of *trans*-chlordane were reported in male beluga from St. Lawrence Estuary, Canada, while no trends were observed for *trans*-nonachlor, *cis*-nonachlor and the sum of five chlordanes. In seabirds from Canada, both significantly increasing and decreasing trends of heptachlor epoxide were reported (Table 2). A decreasing trend of heptachlor epoxide has also been reported in one seabird population from Alaska. The trends in biota reported here and in the literature are consistent with the reported trends in arctic air (Hung et al. 2010-this issue). While the general trend in air was towards decreasing levels of chlordane compounds, the trend appears to be levelling off in recent years and at some stations concentration are at or below the detection limit.

### 3.8. Dieldrin

The mean change for 11 time-series from both Canada and Greenland was an annual decrease of 2.1% (Table 1). Approximately half of the time-series showed statistically significant linear or non-linear decreasing trends. These series include freshwater fish and seabirds from Canada, and one marine mammal population from Greenland.

Beside the above-mentioned time-series, there are reports of decreasing trends in ivory gulls and one population of beluga from Canada and no trend in two seabird populations from Alaska (Table 2). The decreases in biota are consistent with decreasing trends measured in arctic air during the late 1990s at two stations in the Canadian Arctic (Hung et al. 2010-this issue).

### 3.9. ΣMirex (sum of photomirex and mirex) and mirex

The mean change for 12 mirex time-series from Canada and the Faroe Islands was an annual decrease of 0.7% (Table 1). Both significantly increasing and decreasing trends are observed in biota from Canada. The decreasing trend was found in one freshwater fish population, seabird populations and one marine mammal population. The increasing trends were found in one freshwater fish population and one marine mammal population.

In eggs of black-legged kittiwakes, fulmars and thick-billed murres from Lancaster Sound, Canada, mirex concentrations were reported to decrease significantly, while ΣMirex concentrations only decreased significantly in the black-legged kittiwake (Braune, 2007). Mirex was occasionally measured at concentrations above the detection limit in air at Alert, however, trend analysis was not possible (Hung et al. 2010-this issue).

### 3.10. Toxaphene (technical, sum of congeners 26, 40, 41, 44, 50, 52 and sum of 50 and 62)

The mean change for 10 time-series from Canada, Greenland, Iceland and the Faroe Islands was an annual decrease of 0.8% (Table 1). Two of the ten time-series (one freshwater fish population from Canada and one seabird population from Greenland) showed significantly decreasing log-linear trends.

Besides the above-mentioned time-series, three lake trout populations from Canada show decreasing trends while one burbot population in one of the same lakes shows a significantly increasing trend (Table 2). For one Greenland ringed seal population, a non-significant decreasing trend is reported (Table 2).

## 4. Statistical power of temporal trend datasets

Statistical power is an important issue to deal with when analysing temporal trends of contaminant concentrations. Statistical power is connected to hypotheses testing, and can be defined as the probability of rejecting the zero hypotheses ($H_0$) and accepting the alternative hypotheses ($H_A$) when the alternative hypotheses is in fact true (Cohen 1977). Applied to temporal trend analyses, it relates to rejection of the hypotheses that no trend is existing ($H_0$) and acceptance that a real trend exists ($H_A$). The statistical power of a given time-series depends on the length (in the present context, the number of years) of the time-series, the number of samples in each year, the magnitude of change, the magnitude of the random (unexplained) between-year variation, and the significance level (one- or two sided) that is applied.

The number of years of data included in the time-series analysed ranged from 6 to 24. To compare the power of the time-series, we calculated the statistical power of the time-series (given 10 years of data) to detect a 5% annual change in contaminant concentrations with a significance level of 5%. On this basis, 89% of the time-series had a power of less than 60% and the majority had power from 10 to 25% (Fig. 4). Only 15 (5%) of the available time-series had a power of 80% or more. It may therefore be concluded that, in most cases, more than 10 years of data are required before sufficient power is obtained to detect a 5% annual change with a significance level of 5%. Similar results had been obtained for other contaminant monitoring programmes e.g. from the Baltic (Bignert et al., 1997), the International Council for the Exploration of the Sea (ICES) Contaminant Monitoring Programme data series (Fryer and Nicholson 1993), and the AMAP Phase II mercury temporal trend assessment (Bignert et al., 2004). The power would increase if its definition were to employ, e.g. a higher annual change and/or a weaker level of significance testing. However, the parameters used in this power analyses are those typically used in other similar assessments (Bignert et al., 2004; ICES, 2002).

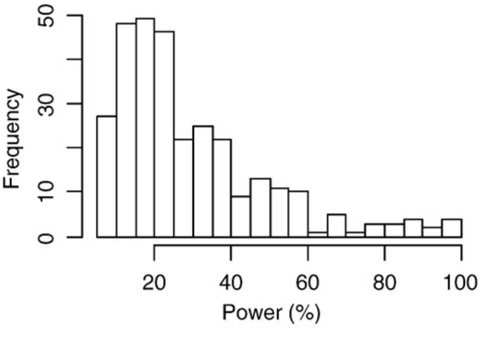

**Fig. 4.** Frequency of the power (%) to detect a 5% annual change in a 10 years period with a significance level of 5%.

A question often asked is “How long should a time-series be before sufficient statistical power is obtained?” For each time-series, therefore, we also calculated the minimum numbers of years necessary to obtain a statistical power of 80% and to detect a 5% annual change with a significance level of 5%. The ‘adequacy’ of each time-series was then determined, defined as the number of years of data in a time-series divided by the minimum number of years necessary to meet the above criteria. An adequacy value equal to or greater than 1 indicates that the time-series is ‘powerful’ enough with respect to the statistical requirements. On the other hand, an adequacy value below 1 indicates that additional years of data are necessary for the time-series to fulfil the requirements of statistical power.

In Fig. 5, the time-series falling at or below the solid line are those with an adequacy value of 1 or above. Only few (8%) fulfil the defined statistical requirements. These included the longest time-series, together with a single time-series with less than 10 years of data. The time-series with adequate power included series for different POP compounds and also from biota belonging to terrestrial, freshwater and marine ecosystems. By this analysis it can be concluded that, if the analysed time-series are extended for the next five years, it is likely that 32% of them will obtain adequate statistical power as indicated by the dotted line in Fig. 5.

In the above power considerations, an annual change of 5% has been applied. For each time-series, the lowest detectable change was calculated for a 10-year period with a significance level of 5% and a power of 80%. The median minimum detectable change was 12% annually, with 60% in the interval from 5 to 15% (Fig. 6). An annual change of 12% corresponds to doubling or halving of contaminant concentrations in about 6 years.

## 5. Conclusions

In general, most time-series of concentrations of legacy POP compounds in biota show significantly decreasing trends over the past

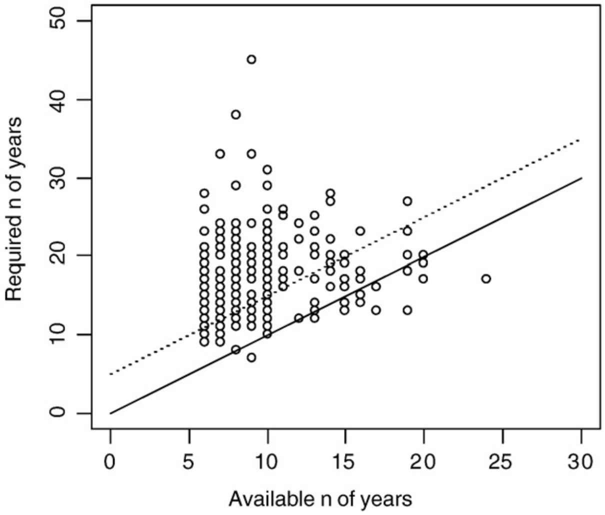

**Fig. 5.** Plot of the number of years in the time-series versus the required number of years to detect a 5% annual change with a power of 80% and a significance level of 5%. The solid line divides the points where the number of years are above and below the required number of years. The broken line predicts the same division given an additional five years of data.

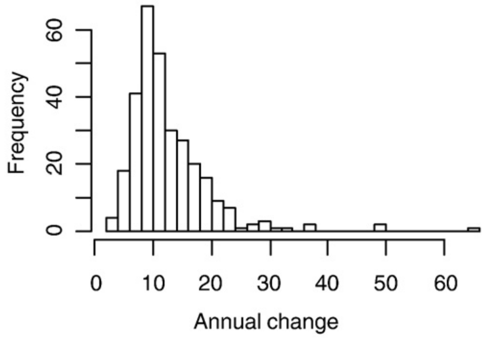

**Fig. 6.** Frequency of lowest detectable trend in a 10 year period with a power of 80% and significance level of 5%.

two to three decades, with only very few examples of significantly increasing trends. Decreasing trends of PCBs, DDTs, α-HCH, γ-HCH and HCB are found across the Arctic area covered by the available time-series, and in all groups of animals studied. The highest mean rate of decrease in this analysis is found for α-HCH, γ-HCH, and DDTs while the lowest rate was found for CHLs and toxaphene. β-HCH concentrations showed a geographical trend, with a low or non-existent rate of decrease in the western Arctic, including some increasing trends, and greater decreasing trends to the east. The rate of decrease varies among species and geographical areas. The relatively low or lack of decline of POPs observed in many Arctic time-series may be explained by the fact that the most intense reductions following regulation of emissions of legacy POPs likely occurred prior to the time periods that are covered by monitoring. The continuing elimination of POPs from the environment may be the reason for the majority of significant time-series showing an exponential rather than a non-exponential decreasing trend. The trends observed in biota are consistent with trends reported for Arctic air, where concentrations of legacy POPs are also decreasing (Hung et al. 2010-this issue). Decreasing trends in Arctic air appear to follow historic decreases in emissions (Li and Macdonald, 2005), however, recent decreases in air are also starting to show signs of levelling off which may be an indication that atmospheric concentrations are being less driven by primary sources and more by environmental processing and degradation. In general, the statistical power of the POPs time-series to detect an annual change of the magnitude typically observed is rather low, and longer data time-series are therefore needed. In the mean time, further attempts could be made to reduce the unexplained between-year variation e.g. by standardizing sample collections as much as possible and/or by adjusting contaminant concentrations for confounding factors when appropriate.

## Acknowledgements

On behalf of the Arctic Monitoring and Assessment Programme, the authors would like to thank the following persons who contributed time-series data for this meta-analysis or provided additional comments on the results, etc.: Maria Dam (Environmental Agency, Faroe Islands), Helga Gunnlaugsdóttir (Matis, Iceland), Derek Muir (Environment Canada, Canada), Kristin Ólafsdóttir (Department of Pharmacology and Toxicology, University of Iceland, Iceland), Gary Stern (Department of Fisheries and Oceans, Canada) and Rune Dietz (National Environmental Research Institute, Denmark). ICES in preparing data archived in the AMAP marine thematic data centre at ICES is also gratefully acknowledged. We would also like to thank the national programs in the circumpolar countries for their funding and support of this work, as well as the community councils and hunters and trappers organizations in many circumpolar regions for their cooperation and active participation in the collection of biological samples.

## References

Addison RF, Ikonomou MG, Fernandez MP, Smith TG. PCDD/F and PCB concentrations in Arctic ringed seals (*Phoca phoca*) have not changed between 1981 and 2000. Sci Tot Environ 2005;351–352:301–11.

AMAP 1998. AMAP assessment report: Arctic pollution issues. Arctic monitoring and assessment programme (AMAP). Oslo, Norway, xii+859 pp.

Barber JL, Sweetman AJ, van Wilk D, Jones KC. Hexachlorobenzene in the global environment: emissions, levels, distribution, trends and processes. Sci Tot Environ 2005;349:1-44.

Bignert A, Cleemann M, Dannenberger D, Gaul H, Roots O. Baltic Sea environment proceedings. Helsinki Commission 1997;64B:130–8.

Bignert A, Riget F, Braune B, Outridge P, Wilson S. Recent temporal trend monitoring of mercury in Arctic biota — how powerful are the existing data sets? J Environ Monit 2004;6:351–5.

Bignert A, Danielsson S, Odsjö T, Remberger M. Organic contaminants in moose (*Alces alces*) and reindeer (*Rangifer tarandus*) in Sweden from the past twenty years. Swedish monitoring programme in terrestrial biota. Rapport till Naturvårdsverket 2008:43 09-25.

Braune B. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975–2003. Environ Pollut 2007;148:599–613.

Braune BM, Outridge PM, Fisk AT, Muir DC, Helm PA, Hobbs K, Hoekstra PF, Kuzyk ZA, Kwan M, Letcher RJ, Lockhart WL, Norstrom RJ, Stern GA, Stirling I. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: an overview of spatial and temporal trends. Sci. Total Environ 2005;351–352:4-56.

Braune BM, Mallory ML, Gilchrist HG, Letcher RJ, Drouillard KG. Levels and trends of organochlorines and brominated flame retardants in Ivory Gull eggs from the Canadian Arctic, 1976 to 2004. Sci. Total Environ 2007;378:403–17.

Cohen J. Statistical power analysis for the behavioural sciences. New York: Academic Press; 1977.

De Wit C, Fisk AT, Hobbs KE, Muir DCG. AMAP assessment 2002: persistent organic pollutants in the Arctic. Oslo, Norway: Arctic Monitoring and Assessment Programme (AMAP; 2004. xvi + 310 pp. Available at the AMAP website: www.amap.no.

Ding X, Wang X-M, Xie Z-Q, Xiang C-H, Mai B-X, Sun L-G, Zheng M, Sheng G-Y, Fu J-M. Atmospheric hexachlorocyclohexanes in the North Pacific Ocean and the adjacent Arctic region: spatial patterns, chiral signatures, and sea–air exchanges. Environ Sci Technol 2007;41:5204–9.

Fryer RJ, Nicholson MD. The power of a contaminant monitoring programme to detect linear trends and incidents. ICES J Mar Sci 1993;50:161–8.

Fryer RJ, Nicholson MD. Using smoothers for comprehensive assessments of contaminants time-series in marine biota. ICES J Mar Sci 1999;56:779–90.

Helgason LB, Barrett R, Lie E, Polder A, Skaare JU, Gabrielsen GW. Levels and temporal trends (1983–2003) of persistent organic pollutants (POPs) and mercury (Hg) in seabird eggs from Northern Norway. Environ Pollut 2008;155:190–8.

Hoekstra PF, O'Hara TM, Fisk AT, Borgå K, Solomon KR, Muir DCG. Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas. Environ Pollut 2003;124:509–22.

Hung H, Kallenborn R, Breivik K, Su Y, Brorström-Lundén E, Olafsdottir K, Thorlacius JM, Leppänen S, Bossi R, Skov H, Manø S, Patton GW, Stern G, Sverko E, Fellin P. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic monitoring and assessment programme (AMAP); 1993–2006. Sci Total Environ 2010; 408:2854–2873 (this issue).

ICES CM 2002. ACME:01. Report of the ICES/AMAP Study Group for the Assessment of AMAP POPs and Heavy Metals data.

Lebeuf M, Noël M, Trottier S, Measures L. Temporal trends (1987–2002) of persistent, bioaccumulative and toxic chemicals in beluga whales (*Delphinapterus leucas*) from St. Lawrence Estuary, Canada. Sci Total Environ 2007;383:216–31.

Li YF, Bidleman TF. Usage and emissions of organochlorine pesticides. In: Bidleman TF, Macdonald RW, Stow JS, editors. Canadian Arctic contaminants assessment report II: sources, occurrence, trends and pathways in the physical environment. Ottawa: Indian and Northern Affairs Canada; 2003. p. 49–70.

Li YF, Macdonald RW. Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Sci Total Environ 2005;342:87-106.

Lohmann R, Breivik K, Dachs J, Muir D. Global fate of POPs: current and future research directions. Environ Pollut 2007;150:150–65.

Macdonald RW, Barrie LA, Bidleman TF, Diamond ML, Gregor DJ, Semkin RG, Strachan WMJ, Li YF, Wania F, Alaee M, Alexeeva LB, Backus SM, Bailey R, Bewers JM, Gobeil C, Halsall CJ, Harner T, Hoff JT, Jantunen LMM, Lockhart WL, Mackay D, Muir DCG, Pudykiewicz J, Reimer KJ, Smith JN, Stern GA, Schroeder WH, Wagemann R, Yunker MB. Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. Sci Total Environ 2000;254:93-234.

Nicholson MD, Fryer RJ, Larsen RJ. A robust method for analysing contaminant trend monitoring data. Techniques in marine environmental sciencesH. C. Andersens-Boulevard, Copenhagen V: International Council for the Exploitation of the Sea; 1998. p. 44–6. vol. 20.

Ólafsdóttir K, Petersen Æ, Magnúsdóttir EV, Björnsson T, Jóhannesson T. Temporal trends of organochlorine contamination in Black Guillemots in Iceland from 1976 to 1996. Environ Pollut 2005;133:509–15.

Pol SSW, Becker PR, Kucklick JR, Pugh RS, Roseneau DG, Simac KS. Persistent organic pollutants in Alaskan murre (*Uria* spp.) eggs: geographical, species, and temporal comparisons. Environ Sci Technol 2004;38:1305–12.

Rigét F, Vorkamp K, Dietz R, Rastogi SC. Temporal trend studies on polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in ringed seals from East Greenland. J Environ Monit 2006;8:1000–5.

Rigét F, Vorkamp K, Dietz R, Muir DCG. Levels and temporal trends of HCH isomers in ringed seals from West and East Greenland. J Environ Monit 2008;10:935–40.

Ryan MJ, Stern GA, Diamon M, Croft MV, Roach P, Kidd K. Temporal trends of organochlorine contaminants in burbot and lake trout from three selected Yukon lakes. Sci Tot Environ 2005;351–352:501–22.

Sinkkonen S, Paasivirta J. Polychlorinated organic compounds in the Arctic cod liver: trends and profiles. Chemosphere 2000;40:619–26.

Van Oostdam J, Donaldson SG, Feeley M, Arnold D, Ayotte P, Bondy G, Chan L, Dewaily É, Furgal CM, Kuhnlein H, Loring E, Muckle G, Myles E, Receveur O, Tracy B, Gill U, Kalhok S. Human health implications of environmental contaminants in Arctic Canada: a review. Sci Total Environ 2005;351–352:165–246.

Vorkamp K, Rigét FF, Glasius M, Muir DCG, Dietz R. Levels and trends of persistent organic pollutants in ringed seals (*Phoca hispida*) from Central West Greenland, with particular focus on polybrominated diphenyl ethers (PBDEs). Environ Int 2008;34:499–508.

Verreault J, Bustnes JO, Gabrielsen GW. Organochlorine contaminants in glaucous gulls from Svalbard — temporal changes during the last 10 years. Tromsø, Norway: Environmental monitoring in Svalbard and Jan Mayen (MOSJ); 2006. p. 13. http://mosj.npolar/filearchive/PolarmaakeTolkning(2006).pdf.

Verreault J, Gabrielsen GW. Contaminants in polar bears: temporal and geographical trends. Tromsø, Norway: Environmental Monitoring in Svalbard and Jan Mayen (MOSJ); 2006. p. 12. http://mosj.npolar/filearchieve/IsbjornTolkning(2006).pdf.

Willet KL, Urlrich EM, Hites RA. Differential toxicity and environmental fates of hexachlorocyclohexane isomers. Environ Sci Technol 1998;32:2197–207.

Zhulidov AV, Robarts RD, Headley JV, Liber K, Zhulidov DA, Zhulidova OV, Pavlov DF. Levels of DDT and hexachlorocyclohexane in burbot (*Lota lota* L.) from Russian Arctic rivers. Sci Tot Environ 2002;292:231–46.