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Twenty years of monitoring of persistent organic pollutants in Greenland biota. A review

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ABSTRACT

The Arctic Monitoring and Assessment Programme (AMAP) is a working group under the Arctic Council with the aim to monitor and assess temporal trends of contaminants in Arctic ecosystems. The Greenland AMAP Core programme was established to contribute to this effort. The Core programme includes three main components; routine monitoring, retrospective studies and new POP screening studies. The programme is based on an adaptive approach, which has led to changes throughout the years. An overview of the temporal trends during the last two to three decades is presently given together with selected examples of different characteristic trends of POPs. The results show how tissue banked samples and retrospective studies has helped in establishing time-series of compounds of emerging concern. Lastly, the statistical power of the Greenlandic time-series is discussed. The lesson learned is that trend monitoring improves with samples over time, and only pays off after decades of data are generated.
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1. Introduction

Monitoring of persistent organic pollutants (POPs) in the Arctic has a number of advantages and the Greenland region is ideal to study long-range contaminant transport, because of the limited local use in the Arctic, bioaccumulation in long food chains and in some cases adverse effects. These are criteria that serve to define what is meant by "POPs" identified and listed under the Stockholm Convention on POPs (De Wit et al., 2004). Monitoring of contaminants in Greenland marine biota was initiated in 1994; a few years after the establishment of the Arctic Monitoring and Assessment programme (AMAP). AMAP is a working group under the Arctic Council with the task to implement the Arctic Environmental Protection Strategy (AEPS) and to support international processes that work to reduce the global threats from contaminants and climate change. Besides UNEP's Stockholm Convention on POPs, these include, for example, the UN Framework Convention on Climate Change, the Minamata Convention on Mercury, and the Convention on Long-Range Transboundary Air Pollution (LRTAP) of the United Nation's Economic Commission for Europe (UN ECE). The Stockholm Convention on POPs was adopted on 22 May 2001 and entered into force on 17 May 2004. Initially, 12 POPs ("dirty dozen") were listed with the intended goal of minimizing risk through measures to reduce and/or eliminate their emissions or discharges. In 2009 nine extra POPs were added to the list, which was further extended in 2011 and in 2013 with the addition of endosulfan and hexabromocyclododecane (HBCDD), respectively. The POPs that are listed are categorised as pesticides, industrial chemicals and/or by-products. Some of the POPs fit into more than one category e.g. hexachlorobenzene (HCB), which was used as a pesticide but also is a by-product in the manufacture of certain industrial chemicals.

POP monitoring in Greenland is organised via the Greenland AMAP Core programme, with the primary objective being to study temporal trends. Temporal trends are useful in showing whether POPs accumulate in Arctic species, which might make them candidates for control on grounds of long-range transport and bioaccumulation, and in assessing the effectiveness of international agreements. The AMAP Core programme is based on ongoing biannual sample collection except for polar bears (*Ursus maritimus*) from East Greenland which are sampled annually. All samples are stored in a tissue bank. Several persistent organochlorine compounds have been included from the start, while others have been

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added later. The brominated flame retardant known as polybrominated diphenyl ethers (PBDEs) as well as per-/poly-fluoroalkyl substances (PFASs), for example, were included in the programme 10–15 years ago as a consequence of retrospective studies performed on samples from the tissue bank (Bossi et al., 2005; Rigét et al., 2006; Dietz et al., 2008; Vorkamp et al., 2008).

In recent years, screening studies of compounds of emerging concern have been an integrated part of the AMAP Core programme. The selection of new compounds for a screening study can be based on the scientific literature (Vorkamp and Rigét, 2014). There can also be interest from policy makers in specific compounds for which Arctic data is considered useful, for example, in the context of an assessment of their long-range transport and bioaccumulation potential.

Statistical power is an important consideration in relation to temporal trend monitoring (Fryer and Nicholson, 1993; Bignert et al., 2004). It can be interpreted as the probability to discover a true change in contaminant levels over time. If, for example, the power of the time-series to detect a certain change is rather low, it will take a long time to detect a statistically significant change with some confidence and thereby prolong the time period before answers can be given on the effectiveness of regulations or the trends of currently unregulated compounds. Monitoring programmes should aim to increase the statistical power of the time-series through the sample collection, chemical analysis and statistical analysis.

The aim of this review is to present an overview of the results and the experiences obtained through the monitoring of POPs in Greenland biota. Examples will be given for legacy POPs and those regulated more recently as well as for screening results of currently unregulated compounds. Emphasis will also be placed on questions of statistical power.

2. The monitoring programme of POPs in biota from Greenland

2.1. Samples

Samples included in the Greenland AMAP Core monitoring programme of POPs are landlocked Arctic char (*Salvelinus alpinus*) from a small lake at Isortoq, Southwest Greenland, ringed seals (*Pusa hispida*) from Qeqertarsuaq, West Greenland and from Ittoqqortoormiit, East Greenland, black guillemot eggs (*Cepphus grylle*) and glaucous gulls (*Larus hyperboreus*) from East Greenland (Fig. 1). Polar bears from Ittoqqortoormiit, East Greenland and from Avanersuaq, Northwest Greenland were included in the regular programme in 2006, but the latter was withdrawn a few years later because of logistical challenges. Shorthorn sculpin (*Myoxocephalus scorpius)* from Qeqertarsuaq, West Greenland and from Ittoqqortoormiit were included in the programme from the mid-1990s to 2004 but was withdrawn from the programme because of rather low levels of POPs. Samples have mainly been collected by local hunters (Rigét et al., 2010a).

When the sampling programme was initiated in 1994 it covered more locations in Greenland than it does now. The sampling frequency was originally designed to be every five years. However, after the second sampling in 1999 it became obvious that the statistical power to detect trends was rather poor and that more frequent sampling would be required to be able to statistically detect trends within a reasonable period of time (Riget et al., 2000). Since 2004 the sampling frequency has been every second year but that also implied that the number of locations had to be reduced to balance the costs. However, polar bears (Ittoqqortoormiit, East Greenland) are sampled every year because of the relatively low number, which is possible to sample in one year and the relatively high variability from year to year. The presented time-series include data derived from related studies and therefore the number of years with data varies between species and areas.

The sampling is standardized as much as possible with respect to time of the year, age and sex (in case of glaucous gulls) and a similar length range for Arctic char. However, for ringed seals and polar bears standardized sampling is not fully consistent as ages were first determined at a later stage. POPs are known to occur at lower levels in juvenile marine mammals than in adults, and they are lower in adult females than in adult males (e.g. Bernhoft et al., 1997; Krahn et al., 1997). On reason for this is that female marine mammals transfer some of their POP body burden to their offspring (e.g. Greig et al., 2007; Bytingsvik et al., 2012). Therefore, temporal trends are presently evaluated separately for juveniles, and adult females and adult males for ringed seals and polar bears. All samples were stored in a tissue bank at −20 °C, which allows for retrospective time-series (e.g. Riget et al., 2006) and to screen for new contaminants of emerging concern (e.g. Vorkamp et al., 2015a).

2.2. Chemical analyses and determinations of supporting parameters

Different laboratories have been involved in the POPs analyses during the programme. The laboratories at the Department of Environmental Science, Aarhus University, Denmark (ENVS) have performed the majority of the chemical analyses. The Organic Contaminants Research Laboratory (OCRL, Environment Canada) at Carleton University, Ottawa, Canada has performed the analyses of legacy POPs and PBDEs in polar bear samples. A minor portion (i.e. arctic char sampled in 1999, shorthorn sculpin sampled in 1999 and 2000, ringed seal sampled in 1999) of the samples were analysed at the National Laboratory for Environmental Testing (NLET) of Environment Canada, and in 1994 arctic char samples were analysed by DLO-Netherlands Institute for Fisheries Research (RIVO-DLO). The time-series of arctic char, shorthorn sculpin and ringed seal therefore include data generated by more than one laboratory. Based on results from inter-comparison studies, Asmund et al. (2004) compared the performance of these laboratories and found a proportional error in the range of 6–24% dependent on the type of substance and laboratory with the tendency of a larger proportional error being from NLET rather than from ENVS. However, we considered the influence from different laboratories to be minor on these time-series as the few data points derived by NLET are surrounded by data derived by ENVIS. In case of the ringed seals $\sum_{10}$-PCB and $\sum$DDT time-series (12 time-series) no changes in the statistical evaluations based on a 5% significance level were found by omitting the NLET data from the analyses.

Detailed descriptions of the chemical analyses have been published previously, including analyses of legacy POPs in polar bears (Dietz et al., 2013a; Letcher et al., 2015; McKinney et al., 2011) and the remaining animals (Vorkamp et al., 2004a, 2011; Rigét et al., 2013a). Descriptions of chemical analyses of PBDEs and HBCDD of ringed seals and glaucous gull are found in Vorkamp et al. (2004b, 2011, 2012) and elsewhere for polar bears (Dietz et al., 2013b; Letcher et al., 2015; McKinney et al., 2011). Chemical analyses of PFASs are described in detail in Rigét et al. (2013b).

The quality assurance and quality control (QA/QC) measures of the chemical analyses in the programme were described by Asmund et al. (2004). The ENVS laboratory performing the majority of the chemical analyses has been accredited according to ISO 17025 for the analyses of polychlorinated biphenyls (PCBs) in biota since 2005 and for PBDEs in biota since 2008. The laboratory has participated in the relevant QUASIMEME proficiency testing schemes since 2001. POP and PBDE analysis in polar bear fat tissues

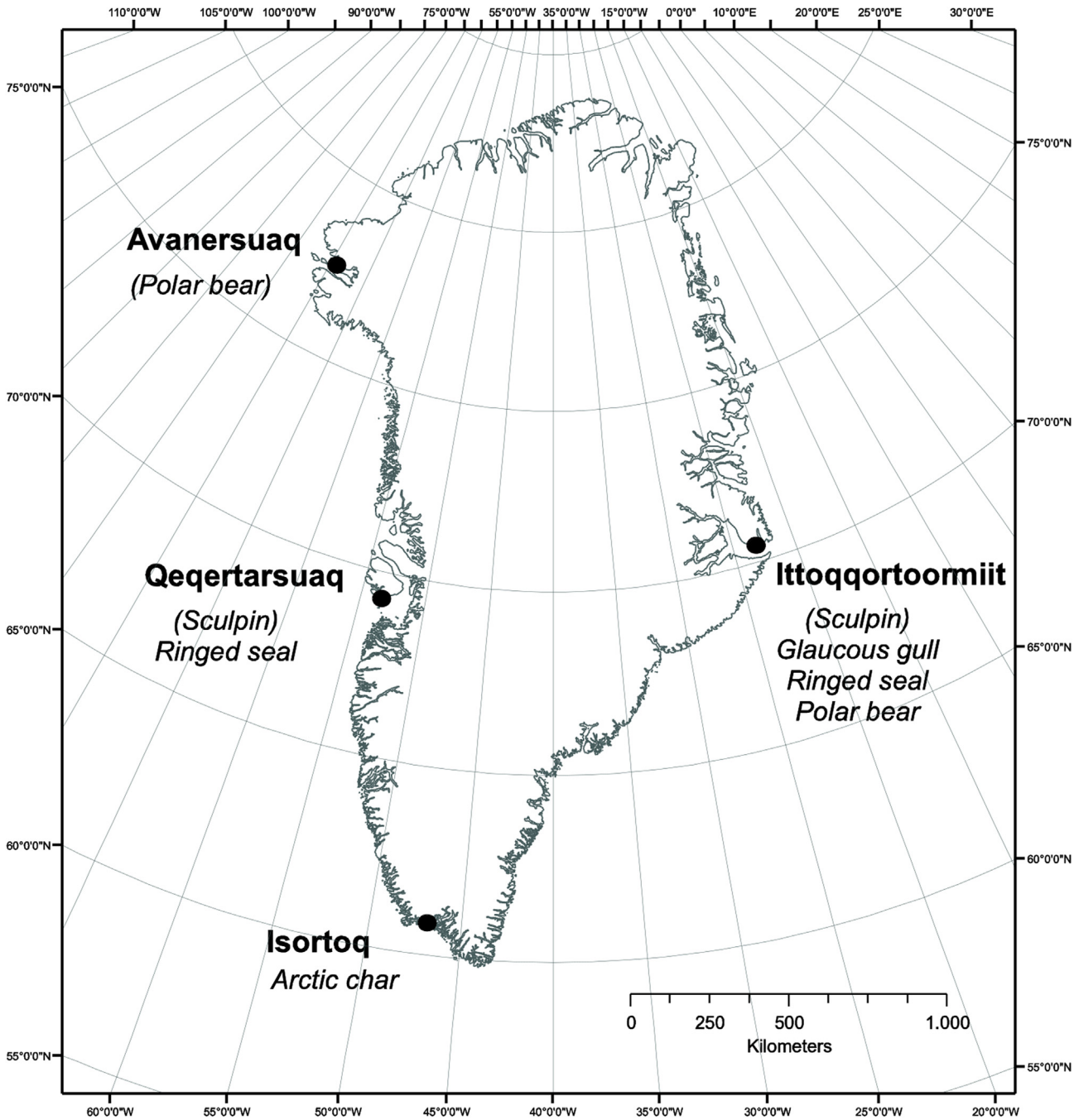

Fig. 1. Sampling locations and species in the Greenland AMAP Core monitoring programme for POPs.

carried out in the OCRL (Environment Canada, Ottawa), have been done in tandem with annual participation of the OCRL in the ILS-QA/QC of the Northern Contaminants Programme (NCP; Aboriginal Affairs and Northern Department Canada (AANDC)), where the OCRL consistently scores very high with respect to analytical compliancy (accuracy and precision) (McKinney et al., 2011; Letcher et al., 2015). Other analyses generally use the same QA/QC measures and criteria as applied to the accredited analyses, regarding acceptable blanks, recovery rates and deviation of duplicates, the use of control charts based on the analysis of in-house reference materials etc. With regard to new contaminants, new methods are developed and validated, based on descriptions in the literature and experiences from established analyses. The method used for novel brominated flame retardants was described by Vorkamp et al. (2015a).

The lipid content of the biota samples is usually analysed according to Smedes (1999) while the age of ringed seals and polar bears is determined according to Dietz et al. (1991). Data are presented on a lipid weight basis except for PFAS in all species and for all POPs in Arctic char. The lipid content of the Arctic char tissue was low, with annual means around 1% (Rigét et al., 2010b). The determination of these low lipid contents was therefore subject to some uncertainty and there is a risk of introducing noise in the time-series if using concentrations that have been lipid weight corrected.

2.3. Statistical analyses

The statistical analysis of the temporal trends is done by a robust regression-based method (Nicholson et al., 1998). Annual median concentrations are used as the yearly contaminant index. The total variation over the time period is divided into a linear and a non-linear component. Log-linear regression is applied to describe the linear component and a 3-point running mean smoother to describe the non-linear component. The two components are tested by means of analysis of variance (ANOVA). The residual error from this temporal trend test is considered as a random between-year variation and the power of the test is calculated as described by Fryer and Nicholson (1993). The trend analysis can have six different outcomes:

- ***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 clear increasing or decreasing tendency.
- ***No trend.*** The time-series did not exhibit a statistically significant trend.

3. Results and discussion

3.1. 'Legacy' POPs, the original 12 POPs listed in the Stockholm Convention

The original 12 POPs ("dirty dozen") listed in the Stockholm Convention on POPs are aldrin, chlordane, dichlorodiphenyltrichloroethane (DDT), dieldrin, endrin, heptachlor, hexachlorobenzene (HCB), mirex, toxaphene congeners, polychlorinated biphenyl (PCB) congeners, and polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/F) congeners. Time-series of these compounds in Greenland biota exist for all compounds except aldrin, endrin, and heptachlor.

The compounds of the original 12 POPs that have been studied in biota from both East and West Greenland (DDTs, HCB, toxaphene and PCBs) are found in highest concentrations in biota from East Greenland, which has been documented for several species over several years (Cleemann et al., 2000a, b, c; Rigét et al., 2006; Vorkamp et al., 2008, 2015b).

Most time-series of the original 12 POPs show significant decreasing trends, some of them also with a significant non-linear trend component (Table 1). In the case of $\sum$DDT and *p,p′*-DDE, twelve out of twenty time-series showed significant decreasing trends, some of them also with a significant non-linear trend component. The ratio between *p,p′*-DDE and $\sum$*p,p′*-DDTs may be a rough indicator of the age of DDT residues in the environment as *p,p′*-DDE is the major metabolite of *p,p′*-DDT (Kelce et al., 1995). In juvenile polar bears and ringed seals from East Greenland and ringed seals from West Greenland the ratio *p,p′*-DDE/$\sum$*p,p′*-DDT has increased significantly during the last thirty years (log-linear regression, all $p < 0.001$), indicating no ongoing or recent inputs of *p,p′*-DDT (Fig. 2). These results are consistent with findings in biota from other areas of the Arctic e.g. freshwater fish, seabird eggs and marine mammals from the Canadian Arctic (NCP, 2013); seabird eggs from northern Norway (Helgason et al., 2009); marine fish from Iceland (Sturlurdottir et al., 2014).

Decreasing concentrations of *p,p′*-DDT and its metabolites are in line with most time trends in air, animals and in humans reported from other areas (Salamova et al., 2013; Law, 2014; Fång et al., 2015). However, *p,p′*-DDT increases were recently observed in high mountain lakes in Tibet (Cheng et al., 2014), which the authors related to the release of DDT from melting glaciers. This process could also be of relevance for the environment of Greenland; however, as yet it has not led to changes in our biota time trends from Greenland.

Increasing concentrations of *o,p′*-DDT, possibly related to the use of the miticide dicofol, have been reported for the Great Lakes region (Venier and Hites, 2014). To study this in the Greenland time series, the ratio of *o,p′*-DDT to the sum of *o,p′*- and *p,p′*-DDT was analysed over time for four time-series (juvenile ringed seals from Qeqertarsuaq and Ittoqqortoormiit, black guillemot eggs from Ittoqqortoormiit and landlocked Arctic char from southwest Greenland). In all cases a log-linear regression shows an increase but in no cases the trend was significant ($p > 0.30$).

Time-series of dieldrin and mirex were only available for the polar bear population in East Greenland (juvenile, adult females and males). In case of dieldrin the juvenile and adult female polar bears showed a significant decrease together with a significant non-linear component, while for adult males only the non-linear component was significant. In case of mirex the juvenile and adult female polar bears showed a significant increasing trend; in case of adult female together with a significant non-linear component. The increasing trend was mainly caused by relatively high concentrations in recent years. Changes in feeding ecology as a consequence of a warming climate are a likely explanation and discussed in more detail in connection with the PCB trends below.

Significant decreasing trends of HCB were found for ringed seal from West Greenland and landlocked Arctic char from southwest Greenland. In East Greenland the trends among taxa were conflicting as young ringed seal showed a significant decreasing trend, while two time-series, black guillemot eggs and adult male polar bears showed a significant increasing trend or increasing trend together with a significant non-linear trend component. The annual change of the ten time-series of HCB was ranging from −3.8% to +5.7%. The observed significantly increasing trends in East Greenland are in contrast to the general trend of HCB found in Arctic biota. Rigét et al. (2010a, b) found no significant increasing trend among 40 Arctic biota time-series. The primary emissions to atmosphere peaked in the 1970s (Barber et al., 2005). However, HCB in air has shown an increasing trend or a very slow decline at some Arctic sites (Hung, in this issue). Bossi et al. (2013) found a weak correlation between Northeast Greenland air concentrations of HCB and temperature and no correlation with ice cover indicating that primary sources still play an important role rather than from re-emissions. Although HCB has been on the Stockholm Convention list since 2004, it is still being generated inadvertently as a by-product and/or impurity in several chemical processes (Barber et al., 2005) and thereby can still be released to the environment.

Six time-series of toxaphene (sum of 3, 5 or 6 congeners) exist and four of those show significant decreasing trends. Vorkamp et al. (2015b) found that the decreases of CHBs 40, 41 and 44 were generally steeper than those of CHBs 26, 50 and 62 in marine biota from Greenland. Decreasing trends of toxaphene were also found in the Canadian Arctic especially in freshwater fish (NCP, 2013) and in northern fur seal from Alaska (Kucklick et al., 2013). In a study on peregrine falcon eggs from Greenland, toxaphene was not found to decrease significantly (Vorkamp et al., 2014). This result might be influenced by the larger inter-sample variation in the peregrine falcon eggs compared with that of the monitoring programme.

Table 1
Number of POP time-series from the Greenland AMAP Core programme for each outcome of the temporal trend analyses.

	Decreasing trend	Decreasing + non-linear trend	Increasing trend	Increasing + non-linear trend	Non-linear trend	No trend	Total
∑CHL	4				1	5	10
∑DDT	6	1			1	2	10
DDE	4	1			1	4	10
Dieldrin		2			1		3
HCB	2	1	1	1	2	3	10
Mirex			1			2	3
$\sum_{10}$-PCB	4	1			1	4	10
PCB-153	4	1			2	3	10
α-HCH	8					1	9
β-HCH	5	2			1	1	9
γ-HCH	4					2	6
PentaBDE	2		3	2			7
Toxaphene	4					2	6
HBCDD			3	1	1		5
PFOS				2	1		3
Total	**47**	**9**	**8**	**6**	**12**	**29**	**111**

$\sum_{10}$-PCB sum of congener 28, 31, 52, 101, 105, 118, 138, 153, 156, 180.
∑DDT sum of *p,p′*-DDE, *p,p′*-DDD, *p,p′*-DDT.
∑CHL sum of *trans*- and *cis*-chordane, *trans*-and *cis*-nonachlor, oxychlordane.
Toxaphene sum of Parlar 26, 40, 41, 50, 62.

In general, most of the PCB time-series showed significantly decreasing trends (Table 1, Fig. 3). The steepest decrease was found for landlocked Arctic char: It was −12.9% as of 2008, and will be −10.5% including the most recent data from 2010 to 2012 (Rigét et al., 2010b). For ringed seals from East Greenland, ΣPCB concentrations were found to be relatively stable in the last 10–15 years (Vorkamp et al., 2011), which was also reported for cetaceans from UK waters (Law et al., 2012). Ongoing PCB emissions might occur from urban sources such as construction materials (Kohler et al., 2005; Diamond et al., 2010). In polar bears an unexpected increase of ΣPCBs has occurred in recent years between 2010 and 2013 (Fig. 3), resulting in a ΣPCB concentration in 2013 at the same level as in 1983. The reasons behind this increase are unknown, but might be related to climate related factors causing decreased ice-cover and thereby changes in dietary consumption and feeding (McKinney et al., 2013), which is being investigated in an ongoing study. This recent increase was not observed in ringed seals, which supports the explanation of polar bears shifting from predating on ringed seals towards predating on harp and hooded seals that are likely to hold higher contaminant loads (McKinney et al., 2013). Changes in trophic position have also been observed for seabird colonies in the Canadian Arctic, with the consequence of changes in contaminant exposure (Braune et al., 2015).

This example of recent unexpected increases in polar bear from East Greenland emphasizes the importance of continued monitoring of PCB and other legacy POPs in Arctic biota in combination with climate related changes.

PCDDs/Fs are not included in the regular Greenland monitoring programme. A retrospective study of PCDDs/Fs and non-*ortho* substituted PCBs (c-PCBs) in ringed seals from East Greenland covering the period 1986 to 2003 showed an annual decrease of WHO-TEQ concentrations of −5.2% and −5.3% for ∑PCDDs/Fs and c-PCBs, respectively (Riget et al., 2005). PCDDs/Fs and dioxin-like PCBs are still of high regional concern and more time trend knowledge has been requested for these regions (Law, 2014). As discussed for HCB, the unintentional formation of PCDDs/Fs might still be a significant source of these compounds, however, in contrast to HCB, our Arctic time series do not suggest the implication of increasing environmental concentrations.

3.2. 'New' contaminants, the POPs added to the Stockholm Convention since 2004

The following compounds have been added to the Stockholm Convention of POPs since 2004: chlordecone, hexachlorocyclohexanes (HCHs), hexabromobiphenyl (HBB), pentabromodiphenyl ether (PentaBDE), octabromodiphenyl ether (OctaBDE), pentachlorobenzene (PeCB), perfluorooctane sulfonate (PFOS; Perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride; added to Annex B of the POP convention), endosulfan, hexabromocyclododecane (HBCDD), polychlorinated naphthalenes (PCNs), hexachlorobutadiene and pentachlorophenol. Some of these compounds can also belong to more than one of the categories; pesticides, industrial compounds and by-product. Of this group of contaminants, time-series in Greenland biota are available for HCHs, PentaBDE, OctaBDE, PeCB, PFOS, endosulfan and HBCDD.

Most of the three isomers of HCH (α, β and γ) time-series showed significantly decreasing trends (Table 1). However, there was a marked difference in the rate of the decline among the isomers illustrated by the time-series of α- and β-HCH in black guillemot eggs (Fig. 4). The annual changes of nine time-series of α-HCH were considerably larger than for β-HCH, while γ-HCH was intermediate. Rigét et al. (2008) found similar levels of the three isomers of HCH in juvenile ringed seals from West and East Greenland, which differs from the general pattern of higher POP concentrations in biota from East than from West Greenland as discussed above. Technical HCH was gradually phased-out or banned during the 1990s and secondary emissions from soils and oceans rather than primary emissions are now probably controlling the air concentrations (Wöhrnschimmel et al., 2012). α-and γ-HCH can be more readily metabolized than β-HCH, which is recalcitrant in most animals (Willet et al., 1998) and is likely the reason for the slower decline of β-HCH. Our results underline that while many time trend studies address ΣHCH, it will be more informative to analyse the isomers separately as their different physical-chemical characteristics and persistence as well as previous uses of technical HCH vs. lindane might influence their temporal trends.

Polybrominated diphenyl ethers (PBDEs) in ringed seals from West and East Greenland were included in the regular monitoring programme in 2006 after a retrospective study had indicated increasing concentrations in West Greenland ringed seals (Vorkamp et al., 2008). Extending the time-series to 2012 of BDE-47, the only congener allowing a trend analysis for young ringed seal from West Greenland, an annual increase of +4% is observed. No trend was shown for young ringed seals from East Greenland in a time series starting in 1986, whereas a significant decreasing trend was reported for adult female and male ringed seals from East

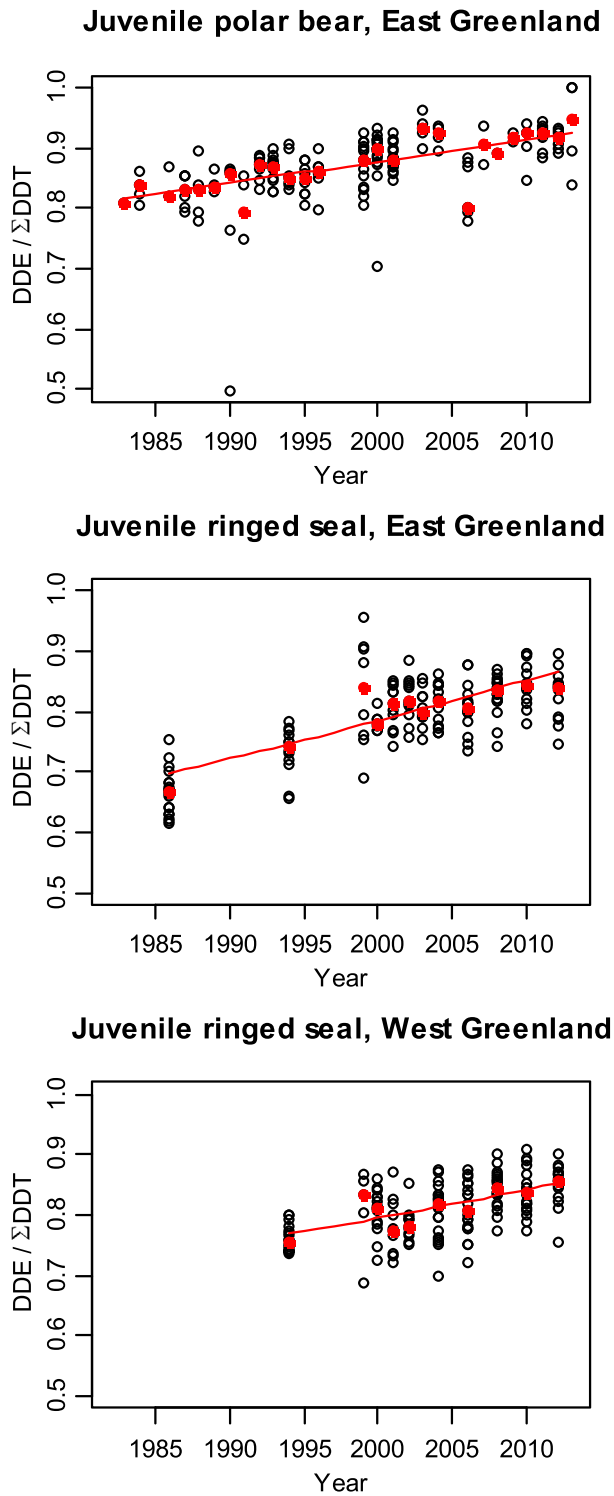

Fig. 2. Ratios of *p,p′*-DDE/∑*p,p′*-DDT in juvenile polar bears and ringed seals from East Greenland and juvenile ringed seals from West Greenland. Red points are annual medians, red lines are significant ($p < 0.05$) regression lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

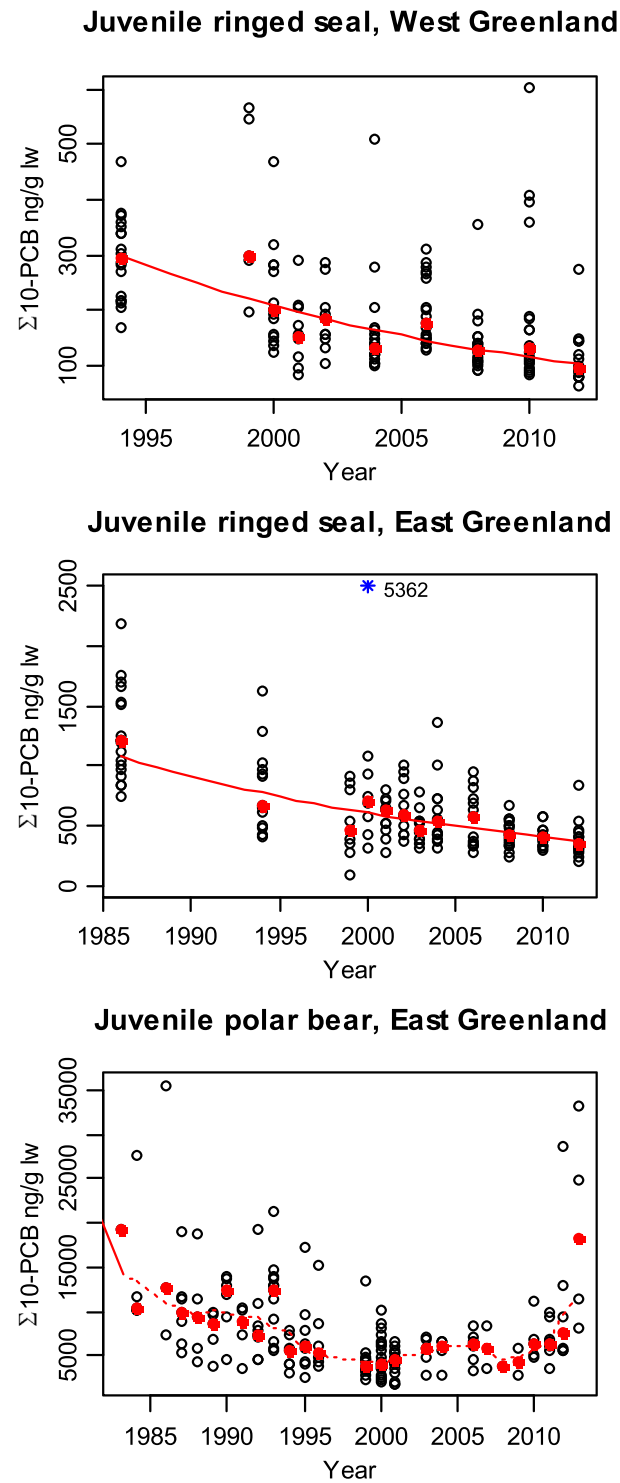

Fig. 3. $\sum_{10}$-PCB concentrations in juvenile ringed seal blubber from West and East Greenland and juvenile polar bear adipose tissues between 1983/1985 and 2013. Red points are annual medians, red lines are significant ($p < 0.05$) regression lines and a broken line is a significant three-years running smoother. Annual decreases for ringed seals are −5.9 and −4.1%, respectively. Blue star considered as individual outlier (updated from Rigét et al. (2013a), Vorkamp et al. (2011) and Dietz et al. (2013a)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Greenland (Vorkamp et al., 2011). However, the highest concentrations were found in the years around 2000, which may reflect a turning point as also proposed by Vorkamp et al. (2011). Polar bears were included in the regular programme in 2006. However, previous studies of POPs had provided time-series back to the middle of 1980s. In polar bears, ∑PBDE increased from the beginning of the 1980s to 2002–2004 where after it decreased

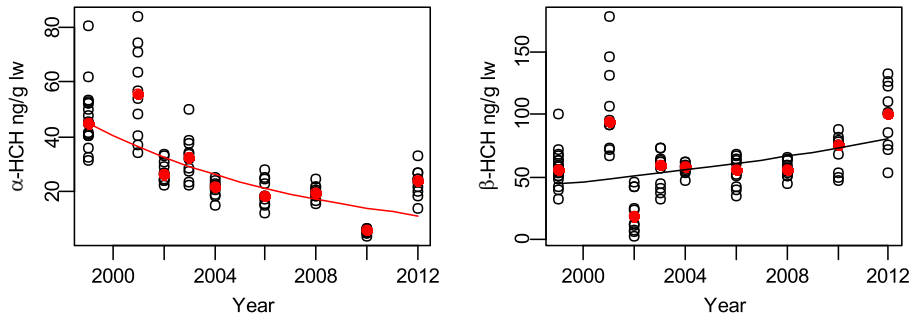

Fig. 4. α- and β-HCH in black guillemot eggs from East Greenland. The annual changes were −10.8% and +4.6%, respectively. Red points are annual medians, red (significant, $p < 0.05$) and black (not significant, $p \geq 0.05$) log-linear regression lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(Dietz et al., 2013b). However, PBDEs appear, as was the case with PCBs, to increase again in the most recent years (Fig. 5). In biota (e.g. seabirds, seals and polar bears) from the Canadian Arctic several time-series also show an increase of PBDEs until the early 2000s followed by a decline (NCP, 2013). Similarly Miller et al. (2014) found that ∑PBDE concentrations increased until approximately the year 2000 and then decreased in four species of seabirds from the Pacific coast of Canada. A similar trend was found for PBDEs in harbour porpoises *(Phocoena phocoena)* from the UK (Law et al., 2012). Hoguet at al. (2013), on the other hand, reported increasing trends of ∑PBDE in two populations of beluga (*Delphinapterus leucas*) from Alaska. However, the most recent years of sampling were in 2000 and 2006 for the two populations and may not be able to detect a decline in recent years if a turning point is in the early 2000s as seen in Greenland and Canada. In Brünnich's guillemot from Svalbard declining trends of ∑PBDE during the period 1993 to 2007 without any specific turning point were found (Bakke et al., 2008).

A time-series of PeCB in Greenland biota is only available for polar bears from East Greenland. In general PeCB has been declining since the early 1980s, however as seen for several other POPs the levels have increased in the most recent years. Chlorobenzenes are often reported as sum of PeCB, TeCB and HCB and only few time-series of PeCB alone in Arctic biota are reported. In the Great Lakes, PeCB in herring gull (*Larus argentatus*) eggs have been monitored annually since the 1970s and at all colonies the concentrations have dropped dramatically (Bailey et al., 2009). In harp seals from the White Sea, Muir et al. (2003) found a 50% decrease from 1992 to 1998.

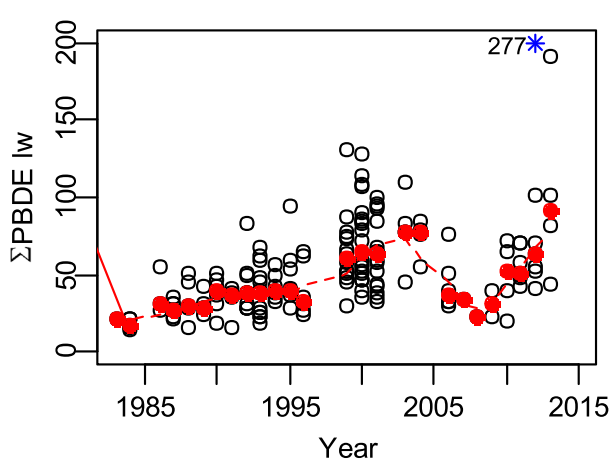

Fig. 5. ∑PBDE concentrations in adipose tissue of young polar bear from East Greenland. Red points are annual medians, broken line is significant 3-years running mean smoother. Blue star considered as individual outlier (updated from Dietz et al. (2013b)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

PFOS have shown increasing levels both in ringed seals (Bossi et al., 2005) and polar bears (Dietz et al., 2008) in initial retrospective studies. In subsequent analyses of the regular monitoring programme, PFOS concentrations peaked sharply around 2006 (Fig. 6) (Rigét et al., 2013b), likely because the largest producer, the 3M US company, voluntarily phased-out its production of PFOS in 2002. Similar patterns are seen for other PFASs, whereas the trend of long-chain perfluorinated carboxylic acids (PFCAs) is less clear (Rigét et al., 2013b). Also in other areas of the Arctic PFOS in biota has peaked. In ringed seal from the eastern Canadian Arctic PFOS peaked in 1998 and 2000 (Butt et al., 2007) and in northern sea otters (*Enhydra lutris kenyoni*) from Alaska the concentrations peaked in 2001 (Hart et al., 2009). In Europe PFOS peaked earlier than in the Arctic: PFOS in grey seal from the Baltic Sea peaked in 1998 (Kratzer et al., 2011) and in eggs of common guillemot (*Uria aalge*) also from the Baltic Sea the peak was in 1997 (Holmström et al., 2005). Considering the monitoring objects of analysing the effectiveness of international regulations, the time series of PFAS and PBDEs in particular have met this purpose as they have been able to demonstrate changes in environmental trends, which most likely are related to regulations or voluntary actions preceding the regulations.

In a retrospective time trend of endosulfan in ringed seals from East Greenland, only the metabolite endosulfan sulfate was consistently above detection limits. Endosulfan sulfate was found to decrease over the study period from 1986 to 2012. In Hudson Bay polar bear fat that was sampled in 2013 and 2014, trace levels of α-endosulfan were found, but β-endosulfan and endosulfan sulfate were not detectable (Letcher et al., 2015). There is a lack of reliable temporal trend data for endosulfan in Arctic biota mainly caused by the challenges of determining low levels of endosulfan isomers in biota (Weber et al., 2010).

HBCDD was included in the regular monitoring programme for three species (glaucous gull, ringed seal and polar bear) from East Greenland based on the results of initial retrospective studies. HBCDD showed increasing trends in ringed seals and polar bears (Vorkamp et al., 2011, 2012; Dietz et al., 2013b) while no trend was found for glaucous gull (Vorkamp et al., 2012). Updates of these time-series have not changed these trends. In biota from the Canadian Arctic (fish, seabirds, seals, and beluga) HBCDD appears to be increasing (NCP, 2013), whereas low levels of HBCDD reported in Hudson Bay polar bears from 2007 to 2012 were subsequently non-detectable in 2013 and 2014 (Letcher et al., 2015). Also in eggs from three seabird species from Northern Norway covering the period

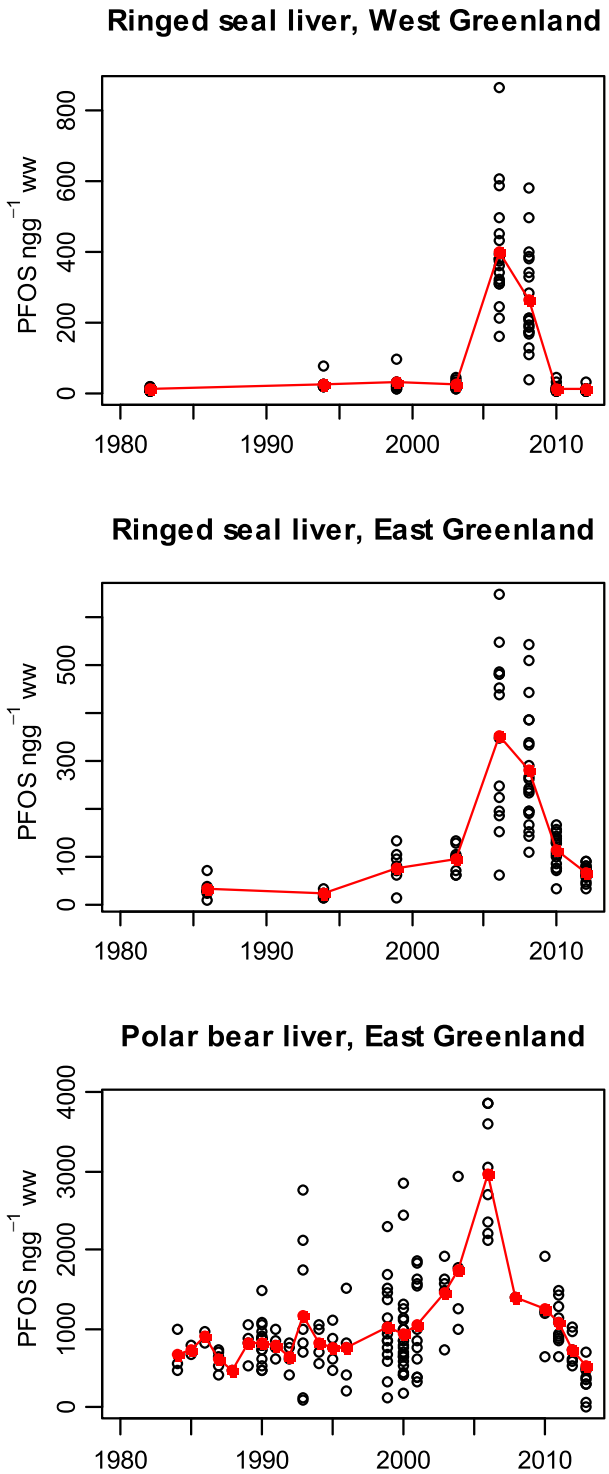

Fig. 6. PFOS concentrations in juvenile ringed seals from West and East Greenland and in juvenile polar bears from East Greenland. Red points are annual means. (Updated from Rigét et al. (2013b)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

1983–2003 and in four seabird species from British Columbia, Canada covering the period 1990–2011, HBCDD showed increasing trends (Helgason et al., 2009; Miller et al., 2014). In the more industrialised waters of the UK, HBCDD in harbour porpoises has been found to decline sharply since 2003 (Law et al., 2012).

3.3. Compounds proposed for listing under the Stockholm Convention and new compounds of emerging concerns

The following chemicals are proposed to be included on the Stockholm Convention list; decabromodiphenyl (decaBDE), dicofol and short-chained chlorinated paraffins (SCCPs). Beside these, concerns has been raised for several replacement BFRs, which substitute the flame retardants now being regulated (Muir and de Witt, 2010; Vorkamp and Rigét, 2014). Bis(2,4,6-tribromophenoxy) ethane (BTBPE) and ethyhexyl-tetrabromobenzoate (EH-TBB) are examples of such replacement compounds. BDE-209, the main component of decaBDE, has been included in all PBDE analyses of biota from Greenland. However, the concentrations were low and often below detection limits (Vorkamp et al., 2008) and no temporal trend could be derived.

Screening for new compounds of emergent concerns has been an integrated part of the monitoring programme in recent years. Typically four-five individuals of selected marine top predators such as ringed seals, black guillemot, glaucous gull and polar bears are analysed and based on the results more effort is allocated to increase the knowledge of their occurrence in the Greenland environment. Vorkamp et al. (2015a) reported detection frequencies close to 100% for EH-TBB, 2,3-dibromopropyl-2,4,6-tribromophenyl ether (TBP-DBPE) and dechlorane plus (*syn*-DCC-CO and *anti*-DCC-CO) while for bis(2-ethylhexyl)-tetrabromophthalate (BEH-TEBP), BTBPE and decabromodiphenyl ethane (DBDPE) the detection frequencies were below 50% in these selected marine top predators. ΣSCCP were detected in the same species, and quantification is ongoing.

3.4. Statistical power of the Greenland time-series

An important consideration in temporal trend monitoring is the statistical power of the time-series, defined as the probability that the data set of interest is sufficiently sensitive to detect a trend of a specified magnitude. If the statistical power is low then important changes are unlikely to be detected and may lead to the wrong conclusion that no significant change has occurred. The power to detect a linear trend depends on: the magnitude of the trend, the number of years in the time-series, the number of samples per year, the residual variance, the significance level of the log-linear regression and whether the test is one- or two-sided. The 111 Greenlandic time-series established in the programme so far have a mean power of 54% with SD of 33% to detect a 5% annual change with a 5% significance level given the present number of years. The lowest power is in general found for the time-series with the lowest number of years and the highest power for time-series with the highest number of years. A power of 80% or more is often considered as satisfactory and this is achieved for 31% of the time-series. This is considerably higher than found in the most recent AMAP POP assessment of temporal trends of legacy POPs in arctic biota including 316 time-series, where the corresponding percentage was only 8% (Rigét et al., 2010a).

A question often raised is "How many years should be monitored before sufficient statistical power is obtained?" Based on the residual standard deviation of the temporal trend analyses and assuming these are sufficiently accurately estimated, power curves can be constructed. Power curves can be quite different for different species and tissue combinations as seen in Fig. 7. The time-series of black guillemot eggs have in general the lowest residual standard deviation and the power curve reaches a target power value of e.g. 80% already after approximately seven years of monitoring, while time-series of polar bears and adult ringed seals do not reach this target before after 13–20 years. It is also evident that time-series of contaminants in young ringed seals in general are more powerful

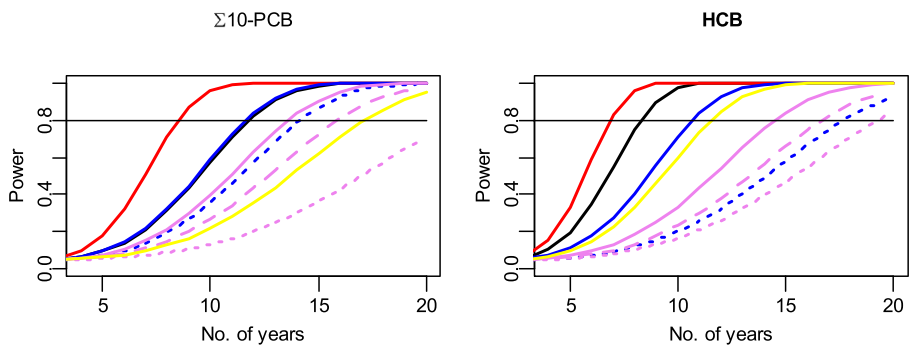

Fig. 7. Power curves for detecting a 5% annual change with a 5% significance level in case of selected $\sum_{10}$-PCB and HCB time-series. Black horizontal line at power = 80%. Red line – black guillemot eggs, East Greenland. Black line – juvenile ringed seals, West Greenland. Blue solid line – juvenile ringed seals, East Greenland. Broken blue line –adult female ringed seals, East Greenland. Solid violet line – juvenile polar bears, East Greenland. Broken violet line –adult female polar bears, East Greenland. Dotted violet line – adult male polar bears, East Greenland. Yellow line – landlocked arctic char. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

than for adult ringed seals, and similarly time-series of juvenile polar bears are more powerful than for the adult polar bears. The reason is that the number of specimens analysed each year were generally higher for young ringed seals and polar bears than for adults but also that contaminant levels were more variable in adults than in juvenile. These power considerations are in accordance with the conclusion in the AMAP POP Assessment that in most cases more than ten years of monitoring is required to obtain sufficient statistical power of the time-series (Rigét et al., 2010a).

4. Conclusions

The time-series of POPs in biota from Greenland obtained by the Greenland AMAP Core programme have constituted an important contribution to international actions to reduce emissions and releases of POPs to the environment such as in accordance with the Stockholm Convention on POPs. Together with time-series from other Arctic countries, a broad picture of the fate and trends of POPs in Arctic has emerged and been assessed under the coordination of the AMAP working group. The Greenland programme has documented decreasing trends of most of the legacy POPs but also that a compound group such as PCB has been relatively stable in the last ten to fifteen years despite production ceasing decades ago. Unexpected trends have also been detected such as an increasing trend in recent years in polar bears of several POPs, which most likely is connected to changes in feeding ecology as a consequence of global warming. The programme has also demonstrated the relatively rapid response of Arctic biota to international regulations as in case of PFOS and to some extent also for PBDEs.

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