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Temporal trends of Persistent Organic Pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP)☆

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ABSTRACT

Temporal trends of Persistent Organic Pollutants (POPs) measured in Arctic air are essential in understanding long-range transport to remote regions and to evaluate the effectiveness of national and international chemical control initiatives, such as the Stockholm Convention (SC) on POPs. Long-term air monitoring of POPs is conducted under the Arctic Monitoring and Assessment Programme (AMAP) at four Arctic stations: Alert, Canada; Stórhöfði, Iceland; Zeppelin, Svalbard; and Pallas, Finland, since the 1990s using high volume air samplers. Temporal trends observed for POPs in Arctic air are summarized in this study. Most POPs listed for control under the SC, e.g. polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethanes (DDTs) and chlordanes, are declining slowly in Arctic air, reflecting the reduction of primary emissions during the last two decades and increasing importance of secondary emissions. Slow declining trends also signifies their persistence and slow degradation under the Arctic environment, such that they are still detectable after being banned for decades in many countries. Some POPs, e.g. hexachlorobenzene (HCB) and lighter PCBs, showed increasing trends at specific locations, which may be attributable to warming in the region and continued primary emissions at source. Polybrominated diphenyl ethers (PBDEs) do not decline in air at Canada's Alert station but are declining in European Arctic air, which may be due to influence of local sources at Alert and the much higher historical usage of PBDEs in North America. Arctic air samples are screened for chemicals of emerging concern to provide information regarding their environmental persistence (P) and long-range transport potential (LRTP), which are important criteria for classification as a POP under SC. The AMAP network provides consistent and comparable air monitoring data of POPs for trend development and acts as a bridge between national monitoring programs and SC's Global Monitoring Plan (GMP).
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1. Introduction

Ever since the adoption of the Stockholm Convention (SC) on

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Persistent Organic Pollutants (POPs) by the United Nations Environment Programme (UNEP) in 2001, research and monitoring of POPs in the Arctic environment has played an important role in its continued development. In particular, Arctic monitoring data documented by the Arctic Council's Arctic Monitoring and Assessment Programme (AMAP) reflecting the environmental persistence and long-range transport potential (LRTP) of many chemicals were critical to the negotiations that resulted firstly in the agreement of the Aarhus Protocol on POPs to the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) and shortly thereafter the global agreement of the SC.

AMAP was established in 1991 to fulfill parts of the Arctic Environmental Protections Strategy concerned with monitoring and assessment of a number of identified priority 'pollution issues of concern'. In the late 1970s and early 1980s, studies found POPs to be present in unexpectedly high levels in the Arctic environment and biota. With little use of POPs within the Arctic and the remoteness of the region from main source/use areas, long-range transport (LRT) was the only possible explanation (e.g. Ottar, 1981; Wania and Mackay, 1993). One of the first major activities of AMAP was to establish a coordinated Arctic monitoring program based on ongoing national monitoring and research activities of the Arctic countries, harmonizing and extending these where necessary.

Hence, four Arctic air monitoring stations were established in the early 1990s: Alert [82° 30′ N, 62° 20′ W, 200 m above sea level (masl)], on Ellesmere Island, Canada, as part of Canada's Northern Contaminants Program (NCP); Stórhöfði (63° 24′ N, 20° 17′ W, 118 masl) on Iceland; the Zeppelin (78° 54′ N, 11° 53′ E, 474 masl) at Ny-Ålesund on Svalbard; and Pallas (68° 00′ N, 24° 15′ E, 340 masl) in Arctic Finland (Fig. 1). The Stórhöfði, Zeppelin and Pallas stations also contribute to the CLRTAP's European Monitoring and Evaluation Programme (EMEP) (Tørseth et al., 2012). Station Nord on Greenland (81° 36′ N, 16° 40′ W, 24 masl) was established in 1990. The name was changed to Villum Research Station, Station Nord in August 2015 and POP measurements started in 2009 (Fig. 1). Other POPs air monitoring sites (including Dunai Island, Valkarkai and Tiksi in Arctic Russia, and Tagish/Little Fox Lake and Kinngait in Canada), and Andøya in Norway were operated in past years, but lack long-term continuity in datasets to develop time trends (Hung et al., 2010; AMAP, 2014). POP concentrations in precipitation (not discussed in this paper) are also measured in parallel with air concentrations at Pallas and Stórhöfði which can be used to assess atmospheric loadings to the Arctic environment. Web links to the long-term stations, related programs and multilateral environmental agreements are given in Sections S1 and S2 in the Supporting Information (SI). Furthermore, atmospheric measurements of POPs have also been conducted during multiple cruise-based expeditions and surveillance studies in the Arctic and northern oceans by various scientific teams (e.g. Jantunen et al., 2015; Wu et al., 2015; Cai et al., 2012; Wong et al., 2011; Shoeib et al., 2010). Although not discussed in detail here, these studies provide invaluable information on the spatial distribution and transport processes of POPs which can be combined with land-based measurement data and temporal trends (such as those presented in this study) in environmental transport and fate models to assess LRT of POPs.

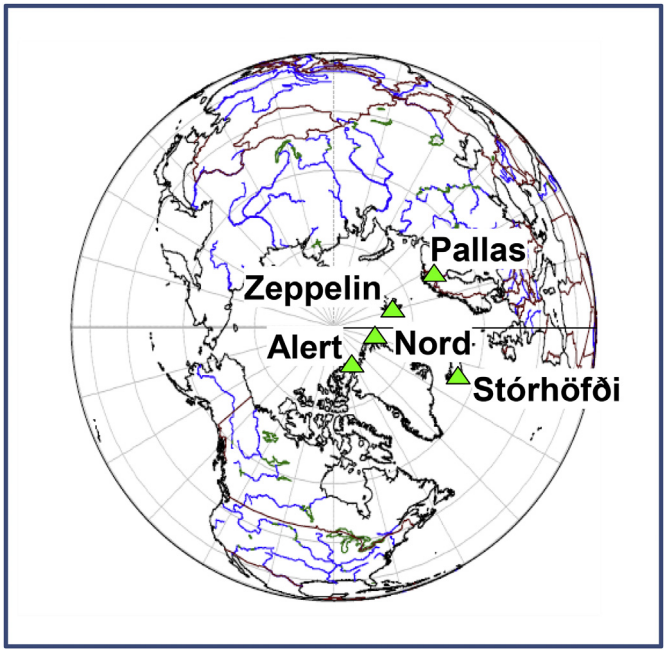

Fig. 1. AMAP station map with long-term observations of POPs in air.

The current study focuses on the evaluation of temporal trend results of POPs measured at the four longest-running stations (i.e. Alert, Stórhöfði, Zeppelin and Pallas) and the determination of their environmental dynamics in the Arctic atmosphere. Air monitoring data of POPs from Station Nord (2009–2013) are presented in Bossi et al. (2016). During the monitoring period (early 1990s to 2012), air samples have also been analyzed for chemicals of emerging concern and archived samples have been used to enhance the understanding of chemical profiles (e.g. isomer ratios and chiral signatures), which has provided insight into LRT and transformation processes, essential for interpreting trends. Lessons learned during the twenty years of monitoring will be discussed, challenges identified, and recommendations made for future circumpolar monitoring.

2. Material and methods

Air samples were collected at the four AMAP stations using high-volume air samplers equipped with a glass fiber filter (GFF) and polyurethane foam plugs (PUFs) to trap particle- and gas-phase chemicals, respectively. Detailed information on sample collection, chemical analysis, data handling and quality assurance/quality control (QA/QC) for the AMAP stations are given in Hung et al. (2010). In brief, weekly samples representing ~13,000 m^3 of air are taken at Alert. All samples were analyzed before 2008. In 2008–2009, samples were analysed every other week; and once every four weeks thereafter. At Zeppelin, weekly samples covering 48–72 h and ~1000–1800 m^3 of air are collected every week. At Pallas, seven-day integrated samples were collected with a volume of ~4000 m^3, and weekly samples are combined for analysis representing one month while at Stórhöfði 1000 m^3 of air was sampled every second week. While total (gas + particle) phase concentrations are reported at Zeppelin, Pallas and Stórhöfði, at Alert, the particle and gas phase compounds were analyzed separately before 2001 and after 2006. For ease of discussions and for comparison with other sites, total (gas + particle) phase concentrations are reported here for all stations.

Although the sampling frequency and time differ among sites, all four Arctic stations follow well-documented QA/QC protocols to ensure that resulting data are comparable (Hung et al., 2010). AMAP laboratories responsible for the analyses of air samples participate in relevant laboratory QA/QC programs, including the AMAP/NCP Inter-laboratory studies (Tkatcheva et al., 2013); the International Polar Year (IPY) multi-national interlaboratory comparison study (Su and Hung, 2010); AMAP/EMEP/NCP air

monitoring inter-laboratory study (Schlabach et al., 2012), the QUASIMEME laboratory performance testing scheme (www.quasimeme.org) and QA/QC programs run by the U. S. National Oceanic and Atmospheric Administration (NOAA)/National Institute of Standards and Technology (NIST). For Alert, there was a laboratory change during the monitoring period resulting in disruption of some datasets. Alert samples were analyzed by the Fresh Water Institute, Canada, between 1993 and 2001. Starting from 2002, samples are analyzed by the National Laboratory for Environmental Testing (NLET). Three rounds of rigorous inter-laboratory comparison studies were conducted to better characterize the discrepancies between the two laboratories to maintain the long-term time trends (Su et al., 2011). Air concentrations of the polychlorinated biphenyls (PCBs) were apparently affected by the cleanup procedure at NLET resulting in low detections of congeners in 2002. Here, the 2002 PCB data from Alert are invalidated and not used. The clean-up and fractionation procedures have since been modified as described in Su et al. (2011) and have successfully corrected for this artefact. Time trends are derived separately before and after 2002.

Air concentration data collected at all AMAP sites are regularly submitted to the AMAP thematic data centre (TDC) database hosted by the Norwegian Institute for Air Research (NILU) and are accessible through the EBAS database at: http://ebas.nilu.no/. For the present study, datasets were extracted from the EBAS database for statistical analysis.

Statistical analysis of time series were performed using the Digital Filtration (DF) Technique (see description in Section S3). DF fits seasonal cycles and inter-annual trends to time series with statistical techniques. This method was applied to derive long-term trends of POPs monitored under the AMAP network (Hung et al., 2005, 2010) and POPs datasets collected by other monitoring programs (Kong et al., 2014). By statistically "smoothing" the datasets, DF-fitted trends and seasonal cycles are statistically significant with 95% confidence.

An apparent first order half-life, $t_{1/2}$, is calculated by dividing ln 2 with the negative value of the linear regression slope of the trend between the natural log of air concentrations, ln C (pg/m^3), and time (year). Many POPs do not necessarily decline linearly or consistently in the first order manner throughout the monitoring periods. Half-lives presented here and summarized in Table S1 are only used for comparison of the relative decline rates among the 4 stations. Readers are advised to use the absolute values of $t_{1/2}$ with caution. When the trend is inconsistent or if half-life is infinitely long, it is labelled as ND (trend not determined) in Table S1.

3. Results and discussions

The fastest route of POPs transport to the Arctic is through the atmosphere. Ambient air responds to atmospheric emissions quickly and is a relatively well-mixed environmental compartment. It is also the starting point for contaminant delivery to the food webs via atmospheric deposition to land and water. With these considerations, air is designated as one of the two core media under the SC's Global Monitoring Plan (GMP) for effectiveness evaluation of the Convention. By making measurements in air in the remote Arctic over time, AMAP results can inform Article 16 of the SC on Effectiveness Evaluation by providing: i.) information on the LRT of POPs and ii.) monitoring data that reveal temporal trends. In addition, screening for emerging chemicals of concern in air samples collected at Arctic sites can provide evidence for chemicals that may be subject to LRT (which are by implication environmentally persistent), a key criterion for classifying a chemical as a POP.

3.1. Temporal trends and seasonal patterns of POPs in arctic air

Air monitoring for 'legacy' POPs, including the organochlorine pesticides (OCPs) and PCBs, started at the 4 AMAP stations in the 1990s, representing some of the longest atmospheric time trends of POPs worldwide. Although the hexachlorocyclohexanes (HCHs), endosulfan and polybrominated diphenyl ethers (PBDEs) have only been listed under the SC recently, air monitoring for these compounds started at the Arctic stations earlier. HCHs actually represent some of the longest POP time series monitored due to its listing under the Aarhus protocol on POPs before the SC. These 'pre-SC' datasets contributed significantly to the risk assessment process of these chemicals providing evidence for their LRTP which led to the SC POPs Review Committee's final decision to classify them as POPs.

Time trends and seasonal cycles derived for selected POPs measured at Alert, Zeppelin, Stórhöfði and Pallas, which have similar time span are presented in Figs. 2–4, S1–S6.

3.1.1. Organochlorine pesticides (OCPs)

Hexachlorocyclohexanes (HCHs): α- and γ-HCH declined in Arctic air at all four stations [Fig. 2a and S1b]. The half-lives of α-HCH range from 4.9 to 5.8 y; and those of γ-HCH were about 4 y for all sites, except at Stórhöfði where it was 7.3 y. β-HCH was also measured in air at Alert and Stórhöfði with declining concentrations in the late 90s and early 2000s with an estimated half-life of 3.7 y (r^2 = 0.88, 1993–2002) and 2.0 y (r^2 = 0.72, 1995–2002), respectively (Fig. 3). After 2004 at Alert, occasional episodically high concentrations were measured. At Stórhöfði, concentrations usually peaked in warmer months and the temporal trend remained more or less unchanged from 2002 to 2012. At Alert, the concentrations increased from 2003 to 2007 followed by a decline to 2012 with many samples below detection in recent years. In 2007, when the Arctic sea ice extent reached a record low [September Average Extent in 2007 was 4.3 million km^2 as compared to the 1981 to 2010 average of 6.52 million km^2 (NSIDC, 2015)], high concentration episodes of β-HCH were observed throughout the year at Alert. This observation is consistent with the hypotheses that the dominant transport pathway for β-HCH to the Arctic is via the ocean (Li et al., 2002) and its re-emission from oceans due to sea ice retreat sustains levels in Arctic air (Wöhrnschimmel et al., 2012). However, in 2011 and 2012 when the sea ice extent declined to 4.63 and 3.63 million km^2, respectively (NSIDC, 2015), almost all measured concentrations were below detection limit at Alert. At Stórhöfði, the summer peak air concentrations from 2002 to 2012 were similar, except for 2005 and 2006 which were lower. The difference in observed concentration patterns at Stórhöfði compared to Alert can be explained by the relative proximity of the former site to open ocean year round and the fact that they have different source areas (Gregor et al., 1998). While Alert is still ice-bound throughout the year and high concentration episodes observed are likely results of LRT, at Stórhöfði β-HCH peaked in summer due to increased volatilization from nearby oceans.

While the usage of technical HCH has declined significantly since the 1980s, lindane (~99% pure γ-HCH) continued to be used in Canada until 2004 (http://www.chemicalsubstanceschimiques.gc.ca/fact-fait/lindane-eng.php) and in the US until 2009 (http://www.epa.gov/oppfead1/cb/csb_page/updates/2006/lindane-order.htm). Atmospheric decline rates of lindane (γ-HCH) have accelerated in the Arctic with the half-life decreased from 6.3 years between 1993 and 2001–4.6 years between 2002 and 2012 (Fig. 2) after its use has been restricted in North America (Hung et al., 2010). At Villum Research Station in Greenland, γ-HCH measured in air (2008–2010) did not show significant temperature

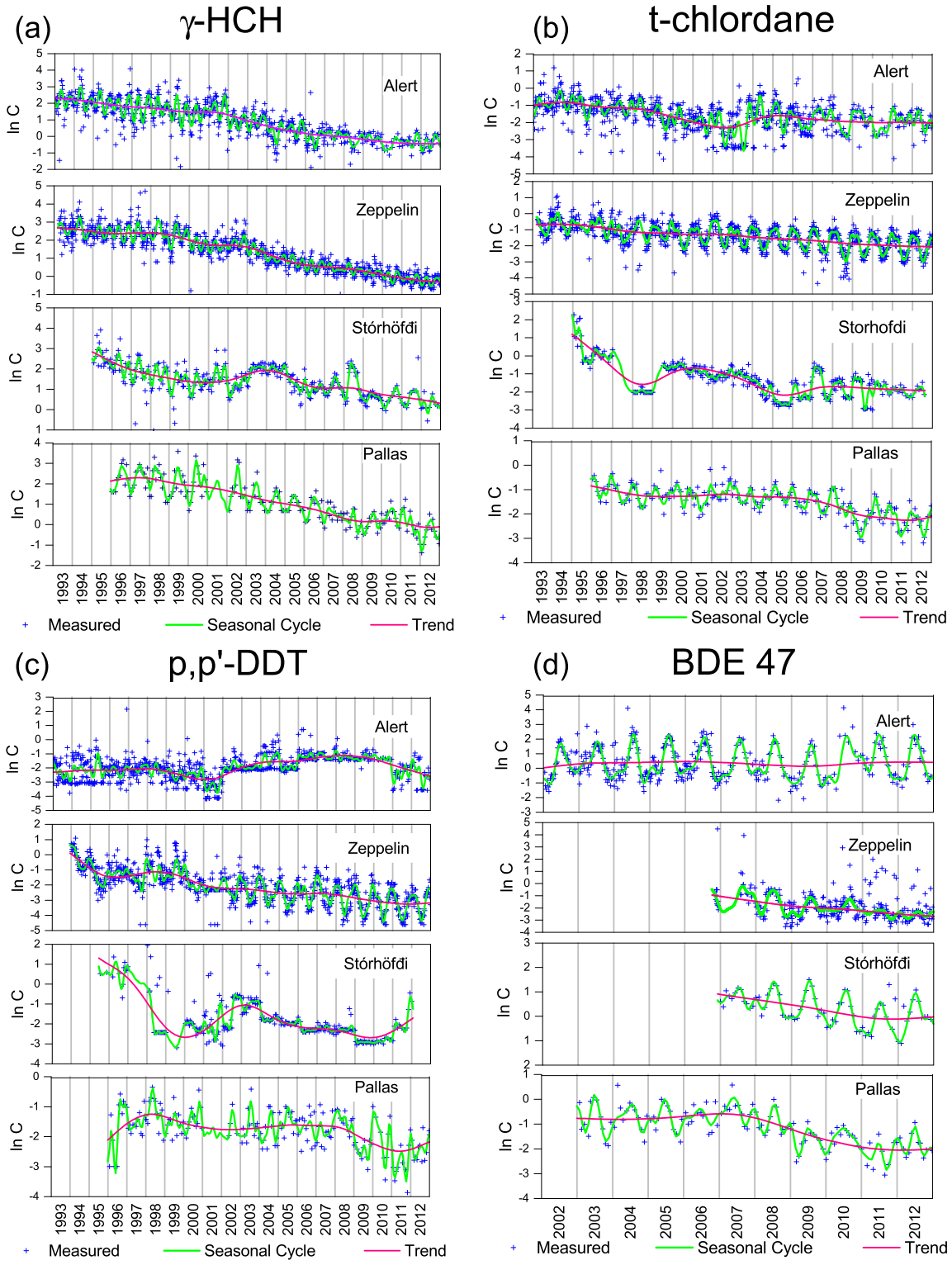

Fig. 2. Time trends of selected POPs: (a) γ-HCH, (b) *t*-chlordane, (c) *p,p′*-DDT, (d) BDE 47. Air concentrations are represented in natural log of concentration (ln C) on the y-axis. Air concentration data lower than the lowest scales of the y-axis are not shown to focus on the temporal trend and seasonal cycles.

dependence, implying that direct atmospheric transport from sources is more likely (Bossi et al., 2013).

The Drins (Aldrin, Dieldrin and Endrin), Chlordanes, Heptachlor and Heptachlor Epoxide: Air concentrations of aldrin and endrin measured at Alert did not show any consistent trends, with aldrin being mostly non-detectable (Fig. S2b). Alert and Stórhöfði reported time trends of dieldrin (Fig. S2a) since 1993 and 1995, respectively, and slow declining trends were found with half-lives

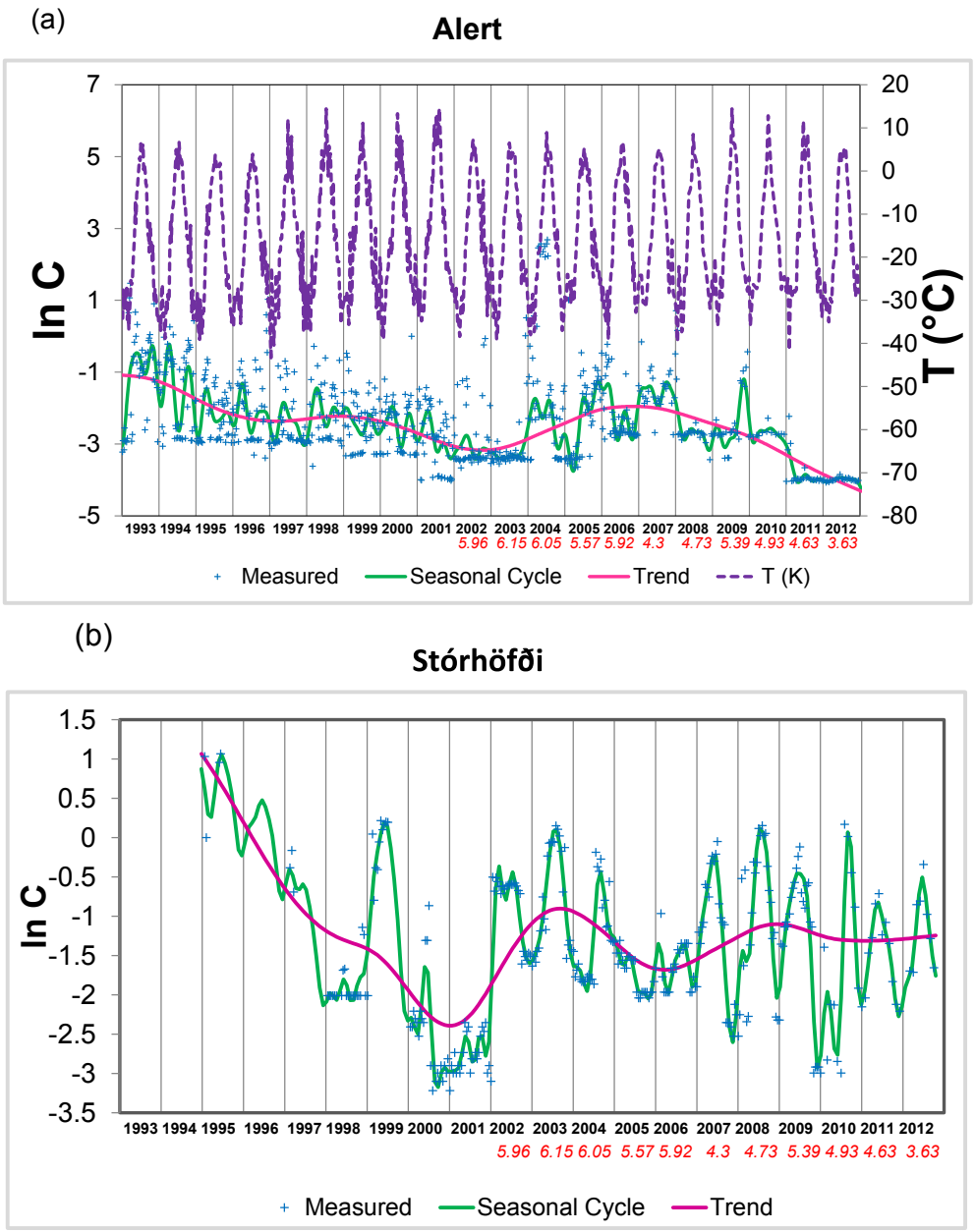

Fig. 3. Temporal trend of β-HCH in air at (a) Alert and (b) Stórhöfði. For Alert, when concentrations were below detection limits, the values are replaced with 2/3 instrument detection limit (IDL). Air temperatures are shown in purple dashed line at Alert. September Average Extents (2002–2012) in million km^2 estimated based on NSIDC Sea Ice Index are given in *italics red fonts* under each year on the x-axes (NSIDC, 2015). September is usually the month when sea ice reaches its annual minimum extent. For comparison, the 1981 to 2010 average is 6.52 million km^2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of 15 and 25 y, respectively. Declining trends of *t*- and *c*-chlordane were observed at most sites, with $t_{1/2}$ ranging from about 11 to 20 y for *c*-chlordane and 6.2–11 y for *t*-chlordane (Table S1). *t*- and c-nonachlor declines at $t_{1/2}$ of 17 and 10 y, respectively, at Zeppelin and 19 and 26 y, respectively, at Alert. Slow decline in the air with $t_{1/2}$ mostly longer than 10 years for all chlordane and nonachlor-related isomers reflect their persistence in the Arctic, although chlordane has been banned in western industrialized countries since the 1980s. Slow decline in trends also indicate tendency towards equilibrium with surface media. Heptachlor and heptachlor epoxide in air were reported for Alert only (Fig. S3). While heptachlor showed no discernible trend, heptachlor epoxide showed a very slow declining trend ($t_{1/2} = 26$ y). It is known that heptachlor degrades readily to heptachlor epoxide once it is released to the environment. Thus, the air concentrations of heptachlor measured at Alert were generally lower than those of heptachlor epoxide and appear to be more sporadic.

For seasonality, air concentrations of dieldrin tend to be lower in the colder months and higher in the warmer months at Alert. Bossi et al. (2013) also reported relatively low concentrations of dieldrin in air at Station Nord on Greenland with a statistically significant correlation with temperature. Slight decreases in the warmest months were observed for dieldrin in all years at Alert which may be related to greater photo-degradation during Arctic summer under 24-h sunlight.

By contrast, *t*-chlordane measured in Arctic air generally showed higher wintertime concentrations. This seasonality is especially apparent at Zeppelin (Fig. 2). Since *t*-chlordane is less

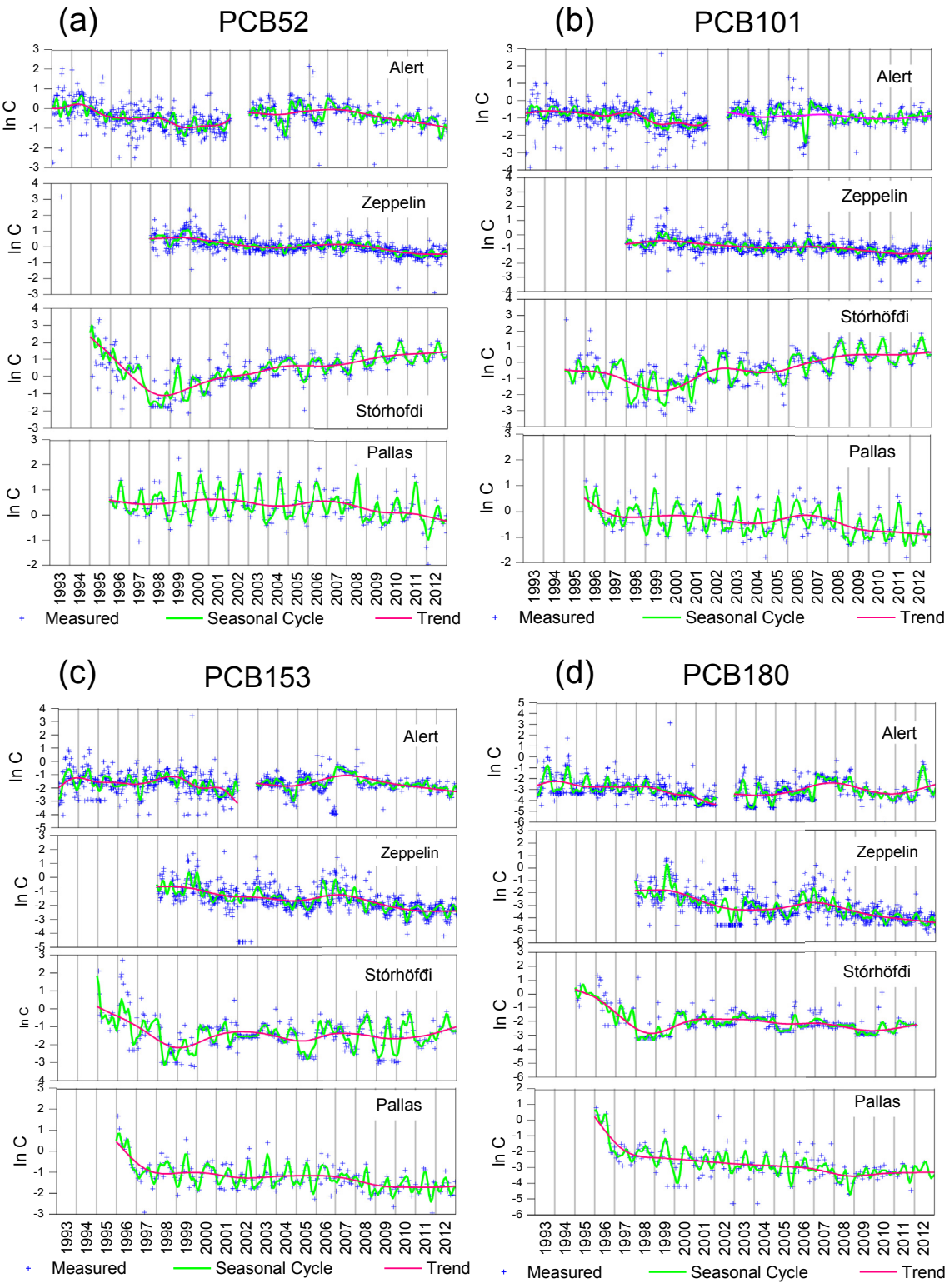

Fig. 4. Temporal trends of selected PCBs: (a) PCB 52, (b) PCB 101, (c) PCB 153 and (d) PCB 180. Air concentrations are represented in natural log of concentration (ln C) on the y-axis. Air concentration data lower than the lowest scales of the y-axis are not shown to focus on the temporal trend and seasonal cycles.

stable than *c*-chlordane, this seasonality may result from enhanced photo-degradation during the Arctic summer when there is 24-h daylight. No consistent seasonality was observed for *c*-chlordane in Arctic air (Fig. S1c), except at Alert where concentrations are slightly higher in the spring and fall. Due to the instability of *t*-chlordane, it was expected that the *t*- to *c*-chlordane ratios would decline over time. However, no consistent interannual trend was observed in this ratio at the four sites. Higher air concentrations of

heptachlor epoxide were observed at Alert during the warm period (possibly reflecting enhanced re-emissions) with a slight dip in concentration in the midst of summer which may be related to greater photo-degradation and enhanced scavenging during this time.

3.1.2. Dichlorodiphenyltrichloroethanes (DDTs: o,p′- and p,p′-DDT, o,p′- and p,p′-DDE, o,p′- and p,p′-DDD)

Among the four Arctic sites, Zeppelin is the only station where all DDT isomers show consistent declining trends (Table S1; Fig. 2c,S4,S5). For *p,p′*- and *o,p′*-DDT, $t_{1/2}$ were found to be 5.0 y and 8.5 y, respectively, at Zeppelin. Declines were also observable at Stórhöfði from the late 1990s to early 2000s for *p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD and at Pallas from 1999 to 2011 for *p,p′*-DDT. Air concentrations at Alert and for other isomers at other Arctic sites seem to have reached steady state and no discernible decreasing trends were found.

Air concentration maxima for all DDT isomers at Zeppelin and *p,p′*-DDE at Alert and Pallas generally occurred in winter. Halsall et al. (1998) reported that DDT-related compounds tend to associate with particles which show greater influx to the Arctic in the winter during the Arctic Haze season. Also, higher precipitation rates in summer may result in their lower summertime concentrations due to enhanced scavenging along the transport pathway to the Arctic (Su et al., 2008). Contrarily, Bossi et al. (2013) observed higher *p,p′*-DDT concentrations in the summer at Station Nord, Greenland, showing significant dependance on temperature and negative correlation with ice cover. However, temperature only accounted for about 27% of DDT variability in air at this site, meaning that LRT sources cannot be excluded. *p,p′*-DDE concentrations were not dependant on temperature, indicating a predominance of LRT rather than re-emission.

Hexachlorobenzene (HCB): HCB tends to break-through in air samples collected using polyurethane foam plugs (PUFs), for instance at the Alert site, HCB broke through in about 30% of all samples; thus air concentrations may be underestimated and results reported here are considered semi-quantitative (Melymuk et al., 2014). HCB in air are reported at three Arctic air monitoring stations showing increasing trends at Zeppelin and Stórhöfði over the last decade (Fig. S1a) and a very slow decline ($t_{1/2}$ = 26 y) at Alert. Air concentrations of HCB measured at Zeppelin were higher during the 1990s, but Alert and Zeppelin showed similar concentrations after 2000. Among the Arctic sites, the lowest HCB air concentrations were found at Stórhöfði in Iceland. At Villum Research Station in Greenland, where samples should not be affected by break-through as measurements were conducted with PUF-XAD-PUF sandwich to capture the gas phase, Bossi et al. (2013) found no seasonal variations in HCB air concentrations between 2008 and 2010, but a weak correlation with temperature, and no correlation with ice cover. These observations suggest that HCB concentrations were not substantially influenced by re-emission at this site in agreement with its high vapour pressure, and may still be controlled by primary sources. Unlike most other 'legacy' OCPs, (primary and secondary) emissions and releases of HCB continue. These are associated with by-production during manufacturing of chlorinated compounds, HCB presence as an impurity in other pesticides, and its industrial/combustion sources (Barber et al., 2005). Also, due to its relatively high vapour pressure, the increase in HCB in air has also been associated with reduction in sea ice coverage and volatilization from environmental sinks in a warming Arctic (Hung et al., 2010; Ma et al., 2011). Rigét et al. (2016) also found a significant increasing trend or increasing trend together with a significant non-linear trend component in East Greenland black guillemot eggs and adult male polar bears, which is in contrast to the general decreasing trend of HCB found in Arctic biota from other areas.

Endosulfans: Air concentrations of α-endosulfan measured at Alert (1993–2012) and Pallas (2009–2012) showed non-changing or slightly declining time trends ($t_{1/2}$ = 19 y at Alert) (Fig. S5), although a slight decrease in concentrations was observed in 2012 at Alert. β-endosulfan (not shown) was found to be mostly below detection limit and no time trend or consistent seasonality was observed. At Villum Research Station, α-endosulfan measured between 2008 and 2010 did not follow Clausius Clapeyrons equation indicating that reemission is of minor importance (Bossi et al., 2013).

Toxaphene: At Stórhöfði, Parlar 26, 50 and 62 are being measured in air since 2000. Parlar 62 was below detection limit for all samples. Fig. S3c shows the time trend of Parlar (26 + 50) with Parlar 26 being the dominant congener. From 2000 to 2004, the air concentration increased slightly, followed by a continuous decline after its inclusion into the SC with $t_{1/2}$ of 5.3 y (2000–2012).

3.2. Polychlorinated biphenyls (PCBs)

At Alert, Pallas and Zeppelin, the decline in PCB air concentrations seem to have slowed down in recent years as the concentrations become much lower resulting in longer half-lives (Fig. 4; Table S1). Negative $t_{1/2}$ for PCB 52 and 101 at Stórhöfði (−8.7 and −6.2 y, respectively) indicates steadily increasing trends, especially after 2000 (Fig. 4). Since Stórhöfði is a coastal site and is in close proximity to Icelandic ice caps, e.g. the Mýrdalsjökull and Eyjafjallajökull ice caps, de-glaciation of ice caps in a warming Arctic may result in re-emission of previously deposited PCBs from oceans and ice, rendering increasing concentrations of relatively lighter PCBs such as PCB 52 and 101. Summer maxima were apparent for PCBs measured at Stórhöfði and Pallas, but are not consistently observed at the more northerly stations of Alert and Zeppelin.

3.3. Polybrominated diphenyl ethers (PBDEs)

PBDEs were measured in air at all four stations (Fig. 2d,S6). BDE 47 and 99 were dominating congeners in Arctic air, reflecting influence from the penta-technical mixture. At Stórhöfði, BDE 47 was detectable in all samples while BDE 99 was only detected in 5 samples and BDE 100 was non-detectable. Concentrations of most BDE congeners were mostly unchanging in air at Alert (Canada) with apparent summer maxima corresponding to increased volatilization during warmer months. In contrast, the air concentrations at the European Arctic sites of Pallas, Stórhöfði and Zeppelin showed significant declining trends with half-lives of 2.6–4.4 y for detectable congeners. Also, the air concentrations at the European sites were generally much lower than those observed at Alert and showed no clear seasonality. Air concentrations at Alert were probably influenced by the nearby military base, which contains articles that were treated with PBDEs, and the generally much higher usage of these compounds in North America as compared to the rest of the world.

In summary, most of the 'legacy' POPs listed by the SC that have been regulated for extended periods of time (>20–30 years) in many countries, e.g. DDTs, the drins, PCBs and chlordanes, are now showing very slow declining rates in Arctic air. This observation likely reflects reductions in primary emissions during the study period, accompanied by an enhanced relative influence of secondary emissions (e.g. volatilization from soil, open oceans etc.). This also speaks to the persistence and slow degradation of POPs under the Arctic environment that they are still detectable in Arctic air decades after they were banned. In contrast, HCB and some PCBs are showing increasing trends in Arctic air at specific locations,

which may be related to enhanced re-emission from environmental sinks due to Arctic warming and continued primary emissions at source. PBDEs are significantly declining in air at Pallas and Zeppelin; possibly reflecting the banning of the penta- and octa-BDE mixtures in the European Union in 2004 (Betts, 2008). However, no decline was observed at Alert which may be related to influence of local sources and generally much higher historical usage in North America.

3.4. Analyzing air samples for 'new POPs' and chemicals of emerging concern

For chemicals with few sources in the Arctic, their presence in Arctic air is usually considered evidence of their environmental persistence (P) (i.e. the substance concerned has the ability to exist in the environment without degrading for a long enough time that it can be transported to the remote Arctic) and thus LRTP; these are important criteria for classifying a chemical as a POP and therefore its consideration for regulation under international agreements. Air samples collected at the AMAP sites are constantly being screened for chemicals that may cause Arctic contamination and may satisfy the P and LRTP criteria for consideration of inclusion under SC.

For the more recently listed POPs, such as hexabromocyclododecane (HBCD) and perfluorooctane sulfonic acid (PFOS), atmospheric long-term time trends are few; especially for PFOS which is more polar and tends to be transported via ocean currents rather than in air (Armitage et al., 2009). However, precursors of per- and polyfluoroalkyl substances (PFASs) may undergo LRT via the atmosphere (Shoeib et al., 2006, 2010; Ahrens et al., 2011) and are degraded to PFOS in the Arctic. At Zeppelin, PFOS was measured in airborne particles from mid-2006 to 2012 (Fig. 5a). The particle phase concentrations seem to remain relatively constant throughout this period and no consistent seasonality was observed (AMAP, 2014). PFOS precursors [e.g. methyl perfluorooctane sulfonamido ethanol (MeFOSE) (Fig. 5b)] are also measured in air at Alert and perfluorooctanoic acid (PFOA) at Zeppelin since late 2006 (AMAP, 2014). Also, PFASs are measured in air and snow at the Villum Research Station since 2008. Total HBCD was measured at Alert since 2002 and at Pallas since 2013 but were mostly non-detectable. Isomer-specific HBCDs (α-, β-, γ-HBCD) were measured at Zeppelin since 2006 and a declining tendency was observed (AMAP, 2014). This decline may be related to the listing of HBCD in the Annex XIV of the Registration, Evaluation, Authorisation and Restriction of Chemicals framework (REACH) of the European Union in 2011. Continuous measurements are needed to confirm long-term time trends for these POPs in Arctic air which would be useful in assessing effectiveness of the SC.

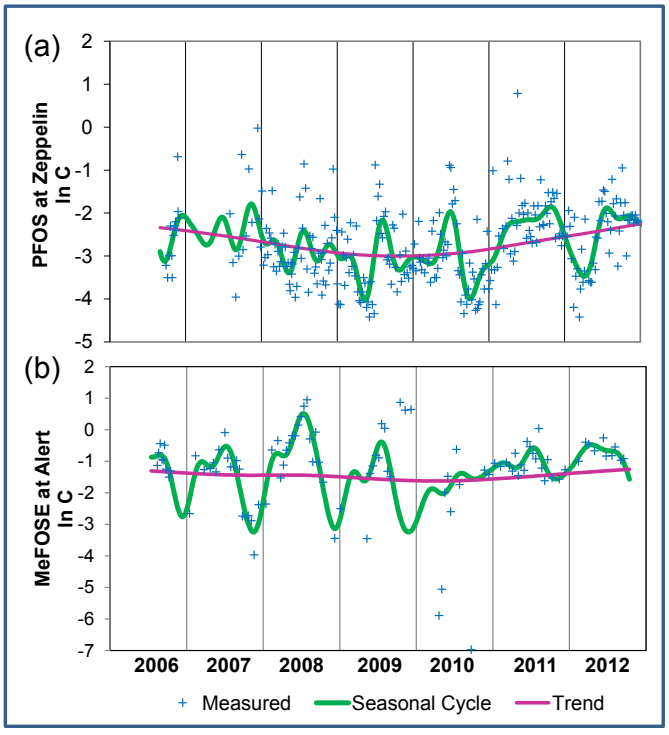

Fig. 5. (a) PFOS measured in air at Zeppelin and (b) MeFOSE measured in air at Alert.

Recently, Arctic air samples have been screened for many current-use pesticides (CUPs) and novel flame retardants (FRs) (Vorkamp et al., 2015), that are used as replacement products for the penta- and octa-technical mixtures of PBDEs. For instance, Xiao et al. (2011) reported air concentrations of FRs 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), 2-ethyl-1-hexyl 2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethyl-1-hexyl)tetrabromophthalate (TBPH) to have concentrations similar to those of the dominant BDE congeners at Alert. BDE 209, which is not listed under the SC, is also being measured at Alert (Xiao et al., 2011). Yu et al. (2015) has measured detectable air concentrations of various non-BDE FRs, including pentabromotoluene (PBT), dechlorane plus (DP), Dechlorane 602, 2,3-dibromopropyl-2,4,6-tribromophenyl ether (DPTE), hexabromobenzene (HBB), and EH-TBB at the sub-Arctic satellite station of Little Fox Lake, Canada. CUPs, such as dacthal, pentachloronitrobenzene, trifluralin and chlorpyrifos are being measured in air at Alert (Hung et al., 2013). Short-chain chlorinated paraffins (SCCPs) and cyclic volatile methyl siloxanes (cVMS) have been included in the measurement programme at Zeppelin since 2013.

Retrospective analysis of archived sample extracts may also provide required information on the occurrence and time trends of chemicals that may have LRTP. For instance, polychlorinated naphthalenes (PCNs), which have been used for decades but only recently listed under the SC as a POP (in May 2015), were analyzed retrospectively in air samples collected at Alert and the satellite stations of Dunai, Russia, and Tagish, Yukon, Canada (Hung et al., 2013). Archived samples from Alert were also screened for SCCPs in 2011 (Hung et al., 2015). Other chemicals of interest, e.g. cVMS, the pesticide dicofol; and organophosphate FRs and plasticizers (OPs) are currently being screened for in archived air samples.

Some new POPs and chemicals of emerging concern, such as the PFASs and cVMS, are more volatile than most 'legacy' POPs and may breakthrough the conventional filter/PUF sampling train used in the high volume air samplers. Various measures are implemented at the different sites to capture these compounds. As mentioned above, at the Villum Research Station, a system of filter/PUF-XAD-PUF is used. A separate high volume air sampler with filter/PUF-XAD-PUF setup is also used at Alert to capture PFASs and CUPs. At Zeppelin, a solid-phase-extraction active air sampling method (SPE-AAS) with ENV + resin (hydroxylated polystyrene–divinylbenzene copolymer) as sorbent is used to capture cVMS (Krogseth et al., 2013). It is, however, of note that precaution is being taken such that the implementation of new air sampling systems will not affect the consistency of routine air monitoring for POPs in order to maintain the long-term temporal trends without disruption.

4. Conclusions - lessons learned and future plans

Most legacy POPs are declining in Arctic air but have started to level off. Air monitoring of newly listed POPs and emerging chemicals of concern must be continued to establish trends and to

assess their environmental transport mechanisms and fate. POP trends measured in Arctic air provide evidence of progress achieved as a result of national and international control measures applied both before and since the establishment of the SC. In assessing trends, dataset consistency and comparability are essential, which speaks to the importance of program QA/QC, thorough documentation on site and in the laboratories, as well as sampling methods and laboratory intercomparisons. Regional programs, e.g. AMAP, which upholds strict QA/QC protocols and documentation, provide a bridge between national monitoring activities, e.g. Canada's NCP and the Europe's EMEP, and global monitoring activities, e.g. SC's GMP, with consistent and comparable datasets for developing trends.

Such trends are also sensitive to impacts of climate change, either directly (e.g. increased volatilization from both primary and secondary sources) or indirectly (e.g. change in Arctic land use and emission patterns, increase in mining/shipping in the North etc.) (UNEP/AMAP, 2011; Becker et al., 2012; Ma et al., 2011). To interpret observed trends and to evaluate SC's effectiveness, i.e. to determine if declining trends (or lack thereof) in air are due to primary emission reduction or other factors, better characterization and quantification of climate change influence and other drivers within and outside the Arctic that may affect trends (e.g. socio–economic transition) is necessary. This can be achieved through the use of environmental transport and exposure models coupled with emission data and observed environmental concentrations and trends of POPs. The unique and extensive multimedia monitoring datasets that AMAP has built, which now include a network of over 60 sites with POPs data in air, marine mammals, fish, invertebrates and birds for $\geq$20 years (Bossi et al., 2016; Riget et al., 2016; AMAP, 2014), is most suitable for improving the understanding necessary to link observations for POPs in air to the effectiveness of international regulation efforts.

Despite the extensive geographical coverage of the AMAP database, there are recognized gaps; for instance in the Russian Arctic sector, POPs monitoring has been performed in the past only on a campaign basis (covering 1–2 year periods at four locations) and temporal trends cannot be assessed. However, work is now underway to establish two Russian POPs air monitoring stations (Amderma and Tiksi) using high volume air samplers as part of AMAP to address this gap. The spatial resolution of monitoring could also be enhanced in an economical way by broader distribution of passive samplers (PASs). The Global Atmospheric Passive Sampling network (GAPS) deploys PASs at ~50 global sites including 7 arctic/sub-arctic stations. NCP has deployed PASs at 7 additional stations close to Arctic communities in 2014 and a flowthrough air sampler at Little Fox Lake (since August 2011) (Yu et al., 2015) to improve the spatial and, eventually, temporal resolution of atmospheric POPs monitoring in Canada's North. Additional regional campaigns are also carried out in Europe including the European Arctic, e.g. under EMEP (Halse et al., 2011) and under the MONET Europe project.

An important element under AMAP is the ongoing assessment of emerging contaminants in air with physicochemical properties that suggest LRTP or where there is evidence for Arctic contamination (e.g. observation of certain FRs in wildlife). Time trends of emerging contaminants may also be reconstructed by retrospective analysis of air samples. However, there are ongoing financial and technical challenges in analysing an ever-increasing target chemical list. To reduce costs, some programs have reduced the sampling/analytical frequency and/or the number of legacy POPs analysed, which are now close to detection limits. Many emerging contaminants are chemicals of commerce (e.g. FRs and PFOS) that may have local pollution sources in the Arctic (e.g. in homes and from landfills). To nominate a chemical to be added to the SC, risk profiles are compiled under Annex E to "evaluate whether the chemical is likely, as a result of its long-range environmental transport, to lead to significant adverse human health and/or environmental effects, such that global action is warranted". For future risk assessments of chemicals with potential local sources in the Arctic, increased spatial coverage of air sampling sites, including both remote sites (e.g. current AMAP stations) and stations within communities (e.g. the NCP PAS stations), combined with surveillance-type measurements of POPs in air over northern oceans less influenced by local sources and sinks, would allow for assessing the relative importance of LRT versus local emissions for these compounds.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.01.079.

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