# Time trends of persistent organic pollutants (POPs) and Chemicals of Emerging Arctic Concern (CEAC) in Arctic air from 25 years of monitoring

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## HIGHLIGHTS

- Time trends of organic pollutants in air were analyzed at eight Arctic sites.
- Many Stockholm Convention-regulated chemicals are decreasing or not changing in air.
- Seasonal cycles of pollutants showed variability with location of the sampling site.
- Levels of emerging chemicals were scattered with non-changing or increasing trends.

## GRAPHICAL ABSTRACT
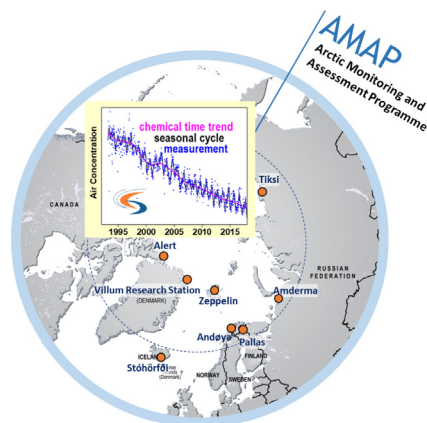


## ARTICLE INFO

## ABSTRACT

The long-term time trends of atmospheric pollutants at eight Arctic monitoring stations are reported. The work was conducted under the Arctic Monitoring and Assessment Programme (AMAP) of the Arctic Council. The monitoring stations were: Alert, Canada; Zeppelin, Svalbard; Stórhöfði, Iceland; Pallas, Finland; Andøya, Norway; Villum Research Station, Greenland; Tiksi and Amderma, Russia. Persistent organic pollutants (POPs) such as $\alpha$- and $\gamma$-hexachlorocyclohexane (HCH), polychlorinated biphenyls (PCBs), $\alpha$-endosulfan, chlordane, dichlorodiphenyltrichloroethane (DDT) and polybrominated diphenyl ethers (PBDEs) showed declining trends in air at all stations. However, hexachlorobenzene (HCB), one of the initial twelve POPs listed in the Stockholm Convention

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in 2004, showed either increasing or non-changing trends at the stations. Many POPs demonstrated seasonality but the patterns were not consistent among the chemicals and stations. Some chemicals showed winter minimum and summer maximum concentrations at one station but not another, and vice versa. The ratios of chlordane isomers and DDT species showed that they were aged residues. Time trends of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) were showing decreasing concentrations at Alert, Zeppelin and Andøya. The Chemicals of Emerging Arctic Concern (CEAC) were either showing stable or increasing trends. These include methoxychlor, perfluorohexane sulfonic acid (PFHxS), 6:2 fluorotelomer alcohol, and $C_9$-$C_{11}$ perfluorocarboxylic acids (PFCAs). We have demonstrated the importance of monitoring CEAC before they are being regulated because model calculations to predict their transport mechanisms and fate cannot be made due to the lack of emission inventories. We should maintain long-term monitoring programmes with consistent data quality in order to evaluate the effectiveness of chemical control efforts taken by countries worldwide.

## 1. Introduction

The Arctic Monitoring and Assessment Programme (AMAP) is an international programme for monitoring and assessing Arctic pollution. It was established in 1991 under the Arctic Environmental Protection Strategy (AEPS, 1991). AMAP is now a Working Group of the Arctic Council responsible for monitoring and assessing a range of pollution- and climate change-related issues. AMAP's main task is to coordinate Arctic monitoring programmes based on ongoing national monitoring and research activities of the Arctic countries.

For the purposes of this work, persistent organic pollutants (POPs) are chemicals listed to the Stockholm Convention (SC) due to their high persistency, toxicity, potential to bioaccumulate and ability to undergo long-range transport. Chemicals of Emerging Arctic Concern (CEAC) are chemicals that possess POP-like characteristics but not currently listed to the SC (AMAP, 2017). They began to be noticed due to knowledge about their usage or improved analytical techniques. Many of them are being considered for listing to the SC.

Monitoring of pollutants in Arctic air can provide insights for the long-range transport (LRT) and persistence of chemicals. It is the only way that information can be obtained for most of the pollutants as emission inventories are not available and thus "bottom-up" model calculations cannot be made. However, using air pollutant monitoring data in combination with "top-down" inverse modelling techniques, it is possible to estimate emission factors of pollutants in support of chemical control strategies (Hung et al., 2013; UNEP, 2017). Hence, the monitoring work is crucial to provide information for risk assessments in chemicals management and to evaluate the effectiveness of the SC and other chemical control measures, including but not limited to the United Nations Economic Commission for Europe (UNCEC) Convention on Long-range Transboundary Air Pollution (LRTAP) Protocol on POPs.

Air monitoring of POPs is one of the core component of AMAP. Efforts in monitoring began in the early 1990s, but required many years of data generation for the first time trends to be published. Hung et al. (2016) reported time trends of POPs at the four longest-running AMAP stations (i.e. Alert, Canada; Stórhöfði, Iceland; Pallas, Finland; Zeppelin, Svalbard) from the 1990s to 2012. Bossi et al. (2016) reported time trends of organochlorine pesticides (OCs), per- and polyfluoroalkyl substances (PFAS) and polybromodiphenyl ethers (PBDEs) in air at Villum Research Station, Greenland (2008–2013) and Anttila et al. (2016) analyzed trends of OCs and polychlorinated biphenyls (PCBs) in air at Pallas, Finland (1994–2011). The most recent time trend (2006–2014) was reported for PFAS in air at Alert, Canada; Zeppelin, Svalbard; and Andøya, Norway (Wong et al., 2018). A general finding of these time trend studies was that the initial twelve POPs, such as PCBs, chlordane and dichlorodiphenyltrichloroethane (DDT) were declining in Arctic air. However, time trends of other POPs were not clear. For example, PBDEs were decreasing in air at Pallas and Zeppelin, but not at Alert. Hung et al. (2016) suggested that PBDE at Alert might be influenced by local emission sources. Another example is hexachlorobenzene (HCB), one of the initial POPs listed on the SC since 2004.

Air concentrations of HCB at Zeppelin and Stórhöfði were increasing, while they were slowly declining at Alert. It has been suggested that these increasing trends may be due to unintentional release of HCB from industrial processing and re-volatilization of HCB from surrounding waters (Hung et al., 2016; Barber et al., 2005).

Here, we update the work of Hung et al. (2016) with an additional five years of data. We analyzed time trends of POPs at six AMAP monitoring stations, namely Alert, Zeppelin, Stórhöfði, Pallas, Andøya, and Villum Research Station. We also present baseline atmospheric POPs data for two stations in Russia, namely Tiksi and Amderma, to fill in a circumpolar spatial gap of POPs monitoring in air. In addition to the POPs that had already been investigated previously by Hung et al. (2016), we present long-term trend results on decaBDE, perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and CEAC, which includes methoxychlor, perfluorohexane sulfonic acid (PFHxS), long-chain ($C_9$-$C_{11}$) perfluorocarboxylic acids (PFCAs) and some PFAS precursor compounds such as the fluorotelomer alcohols.

## 2. Method and material

Fig. 1 shows the locations of the eight main AMAP monitoring stations and one subsidiary station. The main stations are: Alert, Canada;



**Fig. 1.** Location of air monitoring stations.

Zeppelin, Svalbard; Stórhöfði, Iceland; Pallas, Finland; Andøya, Norway; Villum Research Station, Greenland; Tiksi and Amderma, Russia. We focused on data gathered from the main AMAP stations. The subsidiary station is Little Fox Lake, Canada, in which passive flow through sampling has been deployed since 2011. Little Fox Lake is included here for reference. Table S1 shows the geographic coordinates of the stations.

Detailed information on sample collection, chemical analysis, data handling and quality assurance/quality control (QA/QC) for Alert, Stórhöfði, Amderma and Tiksi are given by Hung et al. (2010, 2016) and Wong et al. (2018), Pallas by Anttila et al. (2016), Villum Research Station by Bossi et al. (2016), Zeppelin and Andøya by Bohlin-Nizzetto et al. (2019). In general, air samples were collected at each station using a high-volume active air sampler equipped with a glass or quartz fiber filter to trap particles followed by one or two polyurethane foam plugs (PUFs) or XAD-2 resin in combination with PUF (so called PUF/XAD sandwich) to trap gas-phase chemicals.

Samples were collected over a few days to a week, resulting in average air volumes from 1000 to 13,000 $m^3$. The sampling strategies differ among stations because each station aims to achieve maximum results according to the goals of their national environmental monitoring programme, analytical capability, labour and financial resources. These factors may change over time and hence the sampling strategies are reviewed and may be adjusted accordingly to meet the priorities of the respective monitoring programme.

All participating laboratories regularly submit their data to the AMAP thematic data centre database hosted by the Norwegian Institute for Air Research (NILU), which are accessible through the EBAS database at: http://ebas.nilu.no/. For the present study, we extracted the data from the EBAS database. The total gas and particle phase concentrations were used to derive the time trends for most chemicals except that particle-phase concentration was used for PFAS at Zeppelin and Andøya. At Alert, concentrations below detection limits (BDL) were substituted with 2/3 of the DL. At Villum Research Station, BDL were replaced with 1/2 of the DL. At other sites, BDL were substituted with 1/2 of the DL prior to 2006. After 2006, BDL was substituted with DL. The influence of the different BDL treatments to the overall time trends was negligible.

The time trends were developed using the Digital Filtration technique. Kong et al. (2014) has demonstrated that Digital Filtration is a reliable approach to identify trends for time series longer than five years. Venier et al. (2012) has shown that Digital Filtration performed well in capturing seasonal variation and inter-annual variations of atmospheric chemicals in long-term trends. Detailed descriptions and applications of this method were presented in Hung et al. (2005). In brief, an approximate long-term time trend and an average seasonal cycle were iteratively fitted to the measurement data using a smoothing Reinsch-type cubic spline and Fourier components, respectively, until the fitted spline function becomes almost unchangeable. Outlier data points more than three standard errors away from the fitted curve were rejected after each iterative fit. For each chemical, an apparent first order half-life ($t_{1/2}$) was estimated by dividing ln 2 with the negative value of the linear regression slope of the trend line between the natural logarithm of air concentrations, ($C$, pg $m^{-3}$), and time (year, y). A positive half-life ($t_{1/2} > 0$) indicates the chemical is decreasing. A negative half-life ($t_{1/2} < 0$) indicates the chemical is increasing and it is also called a doubling time. The half-life or doubling time is the time required for the chemical concentration to decline to half or increase to double its original level in the beginning of the sampling period, respectively. Some chemicals do not necessarily decline linearly or consistently in the first order manner throughout the monitoring periods. The half-lives are the result of complex environmental processes which are driven by the chemical's physical and chemical properties, the environmental conditions, and emission sources. We advise the readers that the half-lives are only used to compare the relative rates of decline between stations. In the trend plots, we do not show the data that are lower than the lowest scales of the y-axis in order to focus on the temporal

and seasonal trends. For ease of discussion, $t_{1/2} = 0.01$ to 5.99 y (>11% per year) is considered as "fast to moderate" decline; $t_{1/2} = 6$ to 19.99 y (3.4 to 11% per year) is considered as "slow" decline, $t_{1/2} > 20$ y (<3.4% per year) or trends that were characterized by insignificant correlation coefficients are considered as "no trend" or "non-changing trend". In some cases, only the half-lives of the declining phase was determined. The peak year was identified by the Peak Analyzer function of OriginLab version 2019b. We round-off the peak year, which was determined as decimal year, to the nearest whole number in the discussion.

## 3. Quality control and quality assurance

Despite the differences in sampling and analytical procedures among the participating laboratories, all laboratories follow well-documented QA/QC protocols to ensure that the results are of high quality and comparable. Laboratories responsible for the analyses of AMAP air samples participate in relevant laboratory QA/QC programmes. For example, all laboratories took part in the annual NCP/AMAP Inter-laboratory Study in which the performance of laboratories is assessed based on their analysis of injection-ready standards and air extracts. The exercise covers a wide range of POPs and CEAC, including OCs, PCBs, PBDEs, hexabromocyclododecane (HBCDD), and PFAS (Cozzarin et al., 2019; Tkatcheva et al., 2013; Myers and Reiner, 2017a,b). The laboratories also participated in other international inter-laboratory studies such as the International Polar Year (IPY) multi-national inter-laboratory comparison study (Su and Hung, 2010), AMAP/EMEP/NCP air monitoring inter-laboratory study (Schlabach et al., 2012), QUASIMEME laboratory performance testing scheme (www.quasimeme.org), U. S. National Oceanic and Atmospheric Administration (NOAA)/National Institute of Standards and Technology (NIST)'s QA/QC programme, and UNEP's Biennial Global Interlaboratory Assessment on POPs study (Fiedler et al., 2017).

## 4. Results and discussions

The focus of this study is the time trends of POPs and CEAC in Arctic air at the AMAP monitoring stations. The time trends are assessed by the chemicals' apparent half-lives which are summarised in Tables S2 to S5. For reference, we present the air concentrations of the most frequently detected chemicals measured in recent years (i.e. 2015–2017) in Tables S6 and S7, and Figs. S1 to S3.

### 4.1. Organochlorine pesticides (OCs)

#### 4.1.1. Hexachlorocyclohexanes (HCHs)

Technical HCH contains a mixture of $\alpha$-, $\gamma$-, $\beta$-, and $\delta$- isomers in which $\alpha$-, and $\gamma$-isomers respectively accounted for ~60% and ~10% of the total mixture. Technical HCH was regulated in North America in the 1970s and in China, the former Soviet Union, and India in the mid-1980s to 1990s (Li and Bidleman, 2003). Only the $\gamma$-isomer has the insecticidal activity and it was later isolated and sold individually as lindane. Lindane was used in North America until the early 2000s (Vijgen et al., 2010). In 2009, $\alpha$-, $\gamma$- and $\beta$-HCHs were added to Annex A of the SC.

$\alpha$-HCH and $\gamma$-HCH showed continuous declining trends in Arctic air at all stations (Fig. 2a and b). Both isomers have similar half-lives, ranging from 4.6 to 8.9 y and from 4.4 to 10 y, for $\alpha$- and $\gamma$-HCH, respectively. $\gamma$-HCH at Villum Research Station showed an exceptionally long negative half-life, $t_{1/2} = -28$ y, that implied its concentration was not changing over time, which could be affected by the shorter monitoring period. The reduction in atmospheric concentrations of $\alpha$-HCH and $\gamma$-HCH in air at all Arctic stations is likely due to the declining global emission of technical HCHs and lindane. Our findings are consistent with previous time trends derived from the same monitoring stations from data up to 2012 (Hung et al., 2016) and 2016 (Kalina et al., 2019). Yu
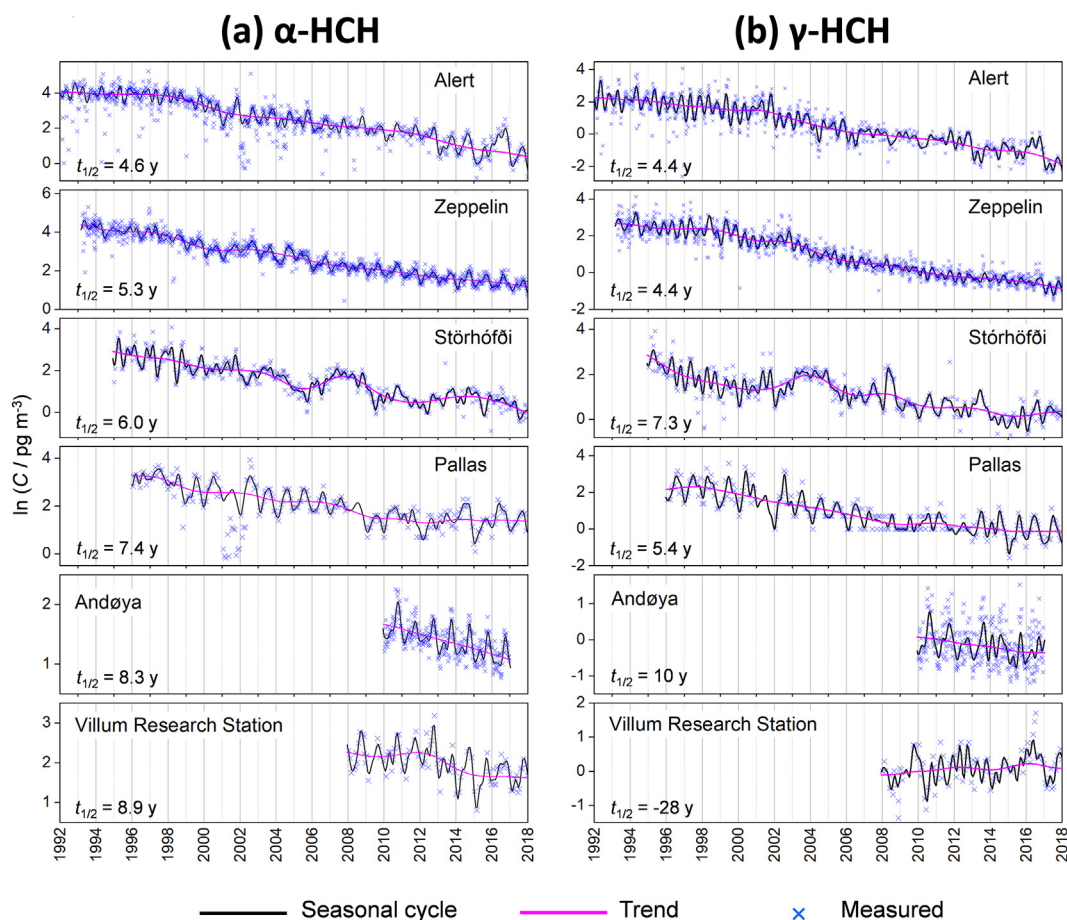
**Fig. 2.** Time trends, seasonal cycles and measurements of (a) α- and (b) γ-HCH in Arctic air.

et al. (2015) also reported decreasing α- and γ-HCH atmospheric concentrations at Little Fox Lake, Canada based on measurements collected in 2003, 2008 and 2013.

α-HCH showed strong seasonality at Zeppelin, Pallas, Andøya and Villum Research Station with summer maximum and winter minimum concentrations (Fig. 2a). At Villum Research Station, Bossi et al. (2016) reported that there was significant positive correlation between temperatures and α-HCH concentrations. This observation suggests that α-HCH at this site was substantially influence by secondary emission, such as volatilization from water. The Arctic Ocean was once a sink for α-HCH, but now it is a source, which sustains the atmospheric concentration of α-HCH via volatilization (Bidleman et al., 2015; Wöhrnschimmel et al., 2012). With warming temperature and sea ice disappearing at a rate of 3.2% per decade from 1979 to 2020 (NSIDC, 2020), we anticipate that there will be ongoing net flux of α-HCH from Arctic waters to the atmosphere. At Alert, α-HCH concentration in air showed no correlations with temperature. Hung et al. (2016) mentioned that Alert was ice-bound most of the year, and its ice condition could have prevented chemical volatilization from the surrounding water.

The seasonality of γ-HCH was not as strong as that of α-HCH at the Arctic stations (Fig. 2b). Bossi et al. (2016) reported that γ-HCH in air at Villum Research Station did not show temperature dependence, suggesting that direct atmospheric transport is likely a source.

### 4.1.2. Hexachlorobenzene (HCB)

HCB was used for industrial and agricultural purposes historically, and its production and emissions peaked in the late 1970s and early 1980s globally (Barber et al., 2005). It is one of the twelve initial POPs

listed to the SC in 2004. HCB, due to its high volatility, is known to be subjected to breakthrough in active air sampling using PUF. Some suggest that increased breakthrough in the sampler during warmer periods could lead to underestimation of summer concentrations (Melymuk et al., 2016; Kalina et al., 2019). Data presented here are semi-quantitative as most of our sites used PUF as the sorbent. It is noted that Villum Research Station is not affected by breakthrough because air samples were taken by a PUF-XAD-PUF sandwich that is supposed to capture all gas-phase chemicals and furthermore the sandwich is kept at constant temperature year round (Bossi et al., 2016).

Air concentrations of HCB at most stations did not show consistent trends. The half-lives of HCB varied considerably between stations, including Alert, $t_{1/2} = 35$ y, Störhöfði $t_{1/2} = 54$ y, Villum Research Station, $t_{1/2} = 44$ y, Andøya, $t_{1/2} = -33$ y, and Zeppelin $t_{1/2} < -100$ y. Given the long half-lives estimated at these sites, we considered that HCB levels remained stable over time (Fig. S4).

Pallas was the only site that showed a clear increasing trend for atmospheric HCB levels. It showed a moderate increasing trend with $t_{1/2} = -4.4$ y. Its concentration increased rapidly from 2008 to 2010. After 2010, the increasing rate appeared to have slowed down. Yu et al. (2015) reported that there was no difference in air concentration of HCB collected in 2003, 2008 and 2013 at Little Fox Lake. Our results are consistent with the fact that HCB is a by-product in combustion processes and in the manufacturing of chlorinated solvents and chemicals (Barber et al., 2005) and its emission will likely continue despite being subject to the SC.

Andøya and Pallas showed strong seasonal trends in the atmospheric concentration of HCB, with an elevated HCB concentration in winter (Fig. S4). Andøya and Pallas are closer to human settlements

than the other sites. As a result, the seasonal cycle at Andøya and Pallas may be a result of higher emissions from combustions during colder periods and thereby higher levels in wintertime.

### 4.1.3. Chlordane and related compounds

The U. S., Canada and Europe have banned the use of chlordane and related compounds since the 1980s. The air concentrations of trans- and cis-chlordane (TC, CC) showed decreasing trends at all Arctic stations, except that TC at Villum Research Station exhibited a non-changing trend. Excluding Villum Research Station, the half-lives of TC at all stations ranged from 3.2 to 13 y (Fig. 3a), and CC ranged from 7.9 to 25 y (Fig. 3b). Andøya has the shortest half-lives for TC and CC among all the stations but note that we used only 3 years of data (i.e. 2010–2012) to estimate these trends. The elevated levels of TC and CC at Stórhöfði from 2015 to 2017 are probably an artefact of replacing the non-detected data with the detection limit, which were higher than those in the early years. This is due to a change in sampling equipment and the issue has now been resolved. Oxychlordane, a degradation product of chlordane (Buser and Müller, 1992; Beeman and Matsumura, 1981), displayed a slow decreasing trend with $t_{1/2} = 19$ y at Alert (Fig. S5).

At Zeppelin and Andøya, we observed a strong seasonality in TC with maximum concentrations in winter and minimum concentrations in summer (Fig. 3a). Similar observations were made at Pallas and Alert, but the seasonality was less pronounced. The minimum concentrations of TC in summer may be due to its enhanced photo-degradation under 24 h of exposure to daylight in the Arctic (Bidleman et al., 2015). At Stórhöfði and Villum Research Station, TC did not show any seasonal cycle. This may indicate factors other than photo-degradation were influencing the concentrations of TC at these stations. CC did not show seasonality in air at any of the stations (Fig. 3b). This may be because CC is less susceptible to environmental degradation than TC.

We used the fraction of TC ($F_{TC}$) to the sum of TC and CC ($F_{TC} = TC / (TC + CC)$) to examine the age of the chlordane residues. The $F_{TC}$ of technical chlordane produced in China and the U.S. when corrected for their vapour pressures, were 0.43 and 0.61, respectively (Jantunen et al., 2000; Li et al., 2006). TC is more easily degraded than CC and thus $F_{TC}$ below the technical vapour values implies an aged chlordane residue. Fig. 3c shows that $F_{TC}$ at all stations were below the $F_{TC}$ of the technical chlordane, indicating input from aged chlordane. $F_{TC}$ shows a seasonal cycle with lower $F_{TC}$ in summer than in winter, implying more TC degradation in the summer. This is consistent with the findings of Bidleman et al. (2015) who reported a similar cycling pattern of enantiomer fractions (EF) of TC in air at Alert. The EF of TC was lower in summer than winter, which means TC in summer was more aged. The aged TC in summer is an indication of a biological source, such as emission of microbial processed TC in soil. On the contrary, there was less degradation in winter and hence higher $F_{TC}$. The low $F_{TC}$ in summer could also be explained by thermal effects. Su et al. (2008) revealed that the enthalpies of vaporization are slightly greater for CC than TC. Hence, the warmer temperatures in summer could have a greater effect on the vapour-phase concentration of CC than TC, thus a low $F_{TC}$. The $F_{TC}$ showed non-changing trends at all sites, which indicates that the proportion of TC and CC had been the same for the last 20 years, and hence the source as well.

The concentrations of trans-nonachlor (TN) exhibited long declining rates at all Arctic stations with half-lives ranging from 10 to 36 y (Fig. S6). Similarly, air concentration of cis-nonachlor (CN) was slowly declining with half-lives ranging from 8.5 to 12 y (Fig. S6). CN in air at Villum Research Station was increasing with $t_{1/2} = -6.7$ y. The increasing trend is probably driven by two measurements after 2016, and additional data are needed for confirmation. In general, the slow declines of TN and CN are likely a reflection of their persistence in the Arctic and tendency towards equilibrium with surface media. CN showed clear seasonal cycles at Alert, Zeppelin and Andøya with summer maximum and winter minimum. This is an indication of a secondary emission source.

### 4.1.4. The drins (dieldrin, aldrin and endrin), endosulfan, heptachlor and heptachlor epoxide

Fewer data are available for the drins. The complete list was only analyzed at Alert and Villum Research Station. For dieldrin, the half-life at
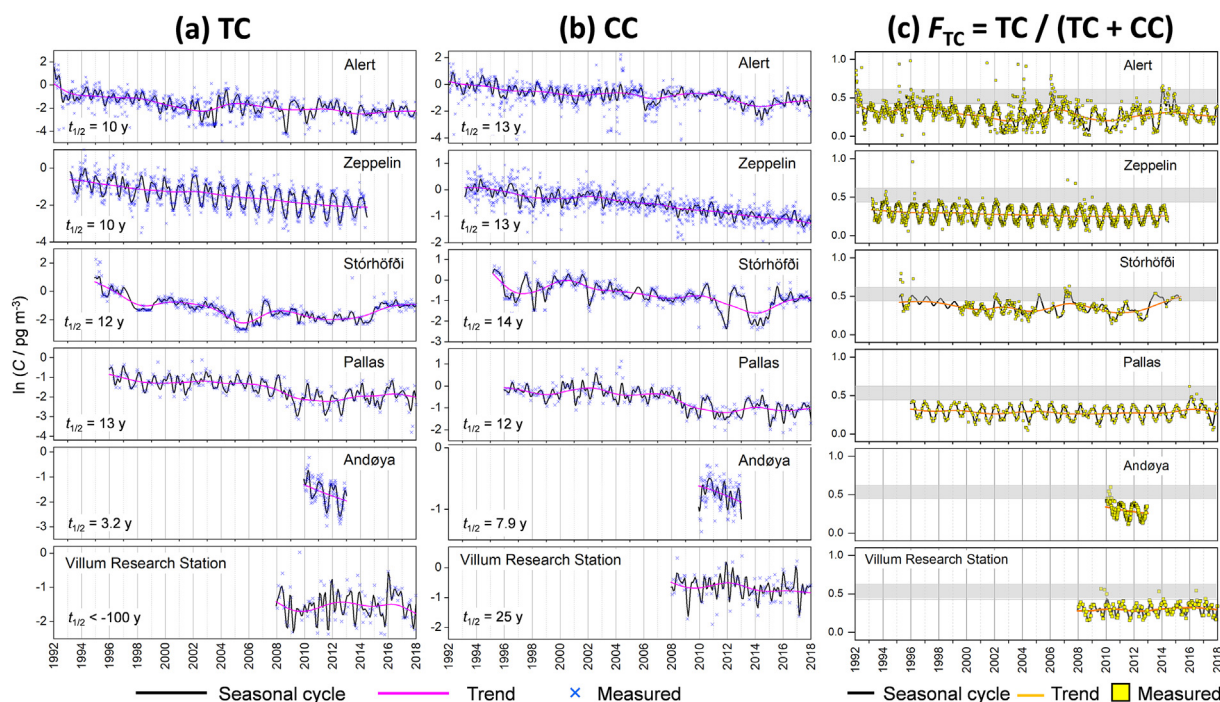


**Fig. 3.** Time trends, seasonal cycles, and measurements of (a) TC, (b) CC in Arctic air, and (c) $F_{TC}$ ($F_{TC} = TC / (TC + CC)$). The grey area is the boundary for the technical vapour $F_{TC}$ values of 0.43–0.61.

Alert was 17 y and at Stórhöfði was 12 y. At Villum Research Station, its half-life was extremely long (>100 y). The long half-lives of dieldrin at these stations indicate that its level will likely remain consistent for an extended time (Fig. S7). We did not derive a trend for aldrin as it was detected in less than 10% of the samples at Alert and Villum Research Station (Fig. S8a). Endrin was sporadically found in air at Alert and it showed a declining trend with $t_{1/2} = 11$ y. We noted that endrin at Alert was mostly non-detected since 2015. Endrin was rarely found at Villum Research Station, hence we did not derive its trend (Fig. S8b).

Air concentrations of $\alpha$-endosulfan at Alert, Pallas and Villum Research Station decreased rapidly since 2010/2011 (Fig. 4a to c). The half-lives of $\alpha$-endosulfan estimated from the peak year (shown in parentheses) were: Alert, $t_{1/2} = 3.1$ y (2010); Pallas, $t_{1/2} = 1.1$ y (2011); and Villum Research Station, $t_{1/2} = 1.7$ y (2011). At Alert, $\alpha$-endosulfan was decreasing at a faster rate now than previously reported for 1992–2012, when a half-life of 19 y had been calculated (Hung et al., 2016). Yu et al. (2015) reported a gradual decline of atmospheric $\alpha$-endosulfan at Little Fox Lake from 2003 to 2013. $\alpha$-Endosulfan also showed declining trends in air at the Greak Lakes Basin (Hites, 2019; Shunthirasingham et al., 2016). The enhanced decreasing rate of $\alpha$-endosulfan at Alert is likely related to the regulation of endosulfan in the U.S. in 2007 and in Canada in 2010 (Health Canada, 2020; USEPA, 2020). Endosulfan was added to the SC in 2011, which may also contribute to the decreasing trends, especially at the European Arctic sites. $\beta$-Endosulfan was mostly below detection limit at all stations and a time trend was therefore not derived.

Heptachlor and heptachlor epoxide at Alert and heptachlor epoxide at Villum Research Station showed non-changing trends with long half-lives $t_{1/2} > 20$ y (Fig. S9). At Alert, heptachlor was mostly non-detectable and hence Alert discontinued the measurements of this compound after 2012. Heptachlor at Villum Research Station appeared to be increasing since 2014 but its doubling time was long, with $t_{1/2} = -23$ y. Overall, atmospheric concentrations of heptachlor and heptachlor epoxide in the Arctic were not changing. This seems unexpected since heptachlor was among the first compounds regulated by the SC in 2004. This also demonstrates the persistence of such chemicals under the Arctic environment. Based on the lack of a decrease since 2004, the concentrations of heptachlor and heptachlor epoxide will probably remain steady in the future.

### 4.1.5. DDT and its metabolites

DDT is listed under Annex B of the SC, allowing restricted use for disease control purposes (UNEP, 2020a). In Arctic air, $p, p'$-DDE, which is the common degradation product of $p, p'$-DDT, is the dominant isomer in Arctic air. At Zeppelin, $p, p'$-DDE was declining with $t_{1/2} = 8.4$ y. Concentrations of $p, p'$-DDE at Alert, Andøya, Pallas and Stórhöfði were not changing with half-lives ranging from 21 to 42 y. At Villum Research Station, concentration of $p, p'$-DDE was also stable from 2008 to 2017, with $t_{1/2} = -29$ y. $o, p'$-DDE showed decreasing trends at Alert, Andøya and Zeppelin with half-lives ranging from 7.6 to 17 y (Fig. S10a). At Villum Research Station, $o, p'$-DDE discerned an increasing trend with $t_{1/2} = -1.9$ y. However, the increase in air concentration at Villum Research Station happened from 2009 to 2012 and the concentration has been stable since then.

$p, p'$-DDT is the major component of technical DDT. Air concentrations of $p, p'$-DDT at all stations were declining, except at Villum Research Station (Fig. 5b). Andøya and Zeppelin showed the shortest half-lives of 3.4 and 5.4 y, respectively. Other stations exhibited relatively long half-lives ranging from 10 to 20 y. $p, p'$-DDT at Villum Research Station was not changing with $t_{1/2} = -20$ y. $o, p'$-DDT was declining at Alert, Andøya and Zeppelin with half-lives ranged from 7.0 to 13 y. Stórhöfði had a $t_{1/2} > 100$ y for $o, p'$-DDT (Fig. S10b).

The degradation products, DDDs ($p, p'$-DDD and $o, p'$-DDD) were mostly undetected since the mid-2010s, therefore trends were only derived for selected stations (Fig. S11). At Zeppelin, both DDD isomers were declining with $t_{1/2}$ of about 9.0 y. At Andøya, $p, p'$-DDD was not changing but $o, p'$-DDD showed a declining trend with $t_{1/2} = 9.3$ y. At Villum Research Station, an increasing trend was estimated for $p, p'$-DDD. The overall trends of DDDs were erratic with no consistency among the DDT species and stations. It is noted that DDT (DDT, DDE and DDD isomers) at Stórhöfði during 2015–2017 were mostly non-detected and the increasing trend is probably due to a change in sampling equipment as mentioned in Section 4.1.3.

The Norwegian sites Zeppelin and Andøya showed clear seasonality in $p, p'$-DDE with winter maximum and summer minimum. Alert tended to follow the same pattern but it was not as pronounced as the Norwegian sites. The situation at Alert may be related to the Arctic Haze season during the wintertime in which there is enhanced transportation of particles to the Arctic and slow removal rates of the DDT in the winter (Hung et al., 2016). The lower temperature in winter may shift the partitioning towards more particle-bound DDT, which may enhance the effect of Arctic haze. To explore this hypothesis, we calculated the percent of DDT (i.e. $p, p'$-DDT, $o, p'$-DDT and $p, p'$-DDE) bound to particles. Fig. S12 shows the monthly average percent of particle-bound DDT and temperature at Alert during 1992–2017. We found that during the winter months (November to February), a large percentage of DDT was bound to particles. During the summer months,
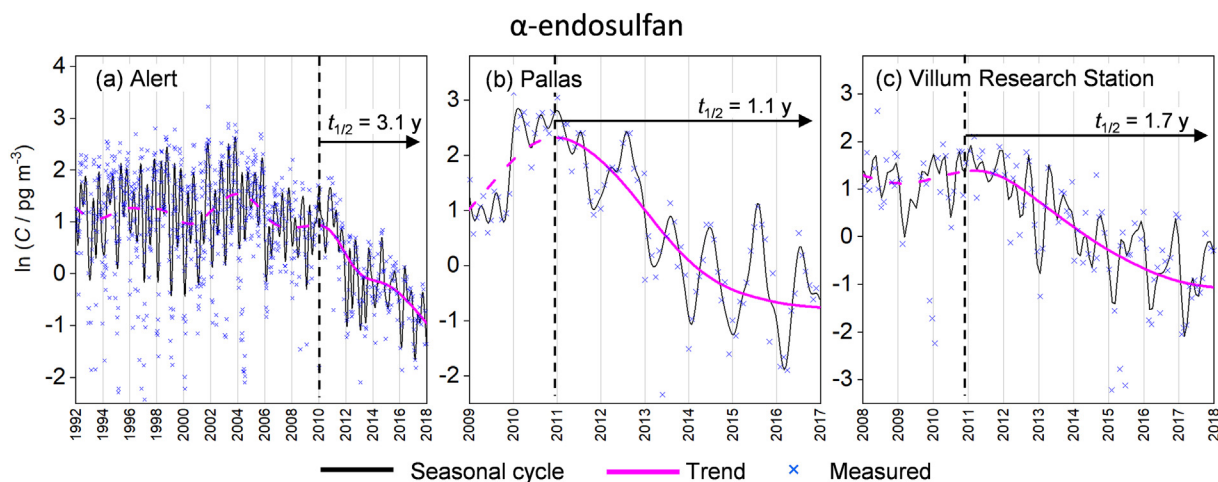


**Fig. 4.** Time trends, seasonal cycles, and measurements of $\alpha$-endosulfan in air at (a) Alert, (b) Pallas, and (c) Villum Research Station. The half-lives were derived from the solid trend line. The dash line indicates the trend prior to the declining phase.
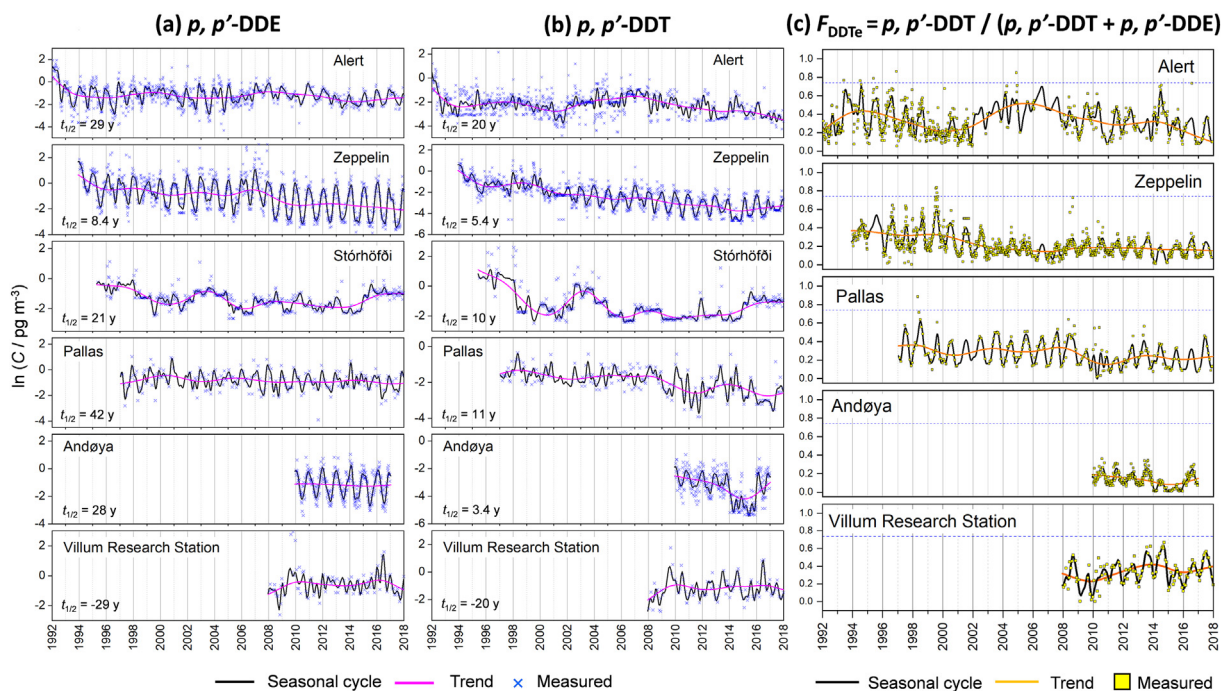
**Fig. 5.** Time trends, seasonal cycles and measurements of (a) $p, p'$-DDE, (b) $p, p'$-DDT, and (c) $F_{DDTe}$ in Arctic air. The dotted line represents $F_{DDTe}$ for technical DDT in vapour phase.

there were no $p, p'$-DDT and $o, p'$-DDT on the particles at all. Particle-bound $p, p'$-DDE was the lowest in the summer months.

To identify the source and age of DDT in Arctic air, we analyzed the relative amount of different DDT species. We expressed the DDT species as fractional values. $F_{DDTe} = p, p'$-DDT/($p, p'$-DDT + $p, p'$-DDE) and $F_{DDTo} = p, p'$-DDT/($p, p'$-DDT + $o, p'$-DDT). We performed the calculation only when both species in a fraction were above the detection limit. Assuming the composition for technical DDT reported by the World Health Organization (77%, $p, p'$-DDT, 15% $o, p'$-DDT, 4% $p, p'$-DDE; WHO, 1989), technical DDT has $F_{DDTe} = 0.95$, and $F_{DDTo} = 0.84$. The estimated technical vapour that was corrected for its subcooled liquid vapour pressure has $F_{DDTe} = 0.74$, and $F_{DDTo} = 0.58$ (Bidleman et al., 2013). We note that the $F_{DDTo}$ is confounded by dicofol, in which dicofol contains high proportion of $o, p'$-DDT as an impurity (Qiu et al., 2005). Technical dicofol vapour manufactured in China has $F_{DDTo} = 0.04$, which could be a marker for tracking "dicofol-type" DDT in air (Bidleman et al., 2013).

Fig. 5c shows the long-term trends of $F_{DDTe}$ at Alert, Zeppelin, Pallas, Andøya, and Villum Research Station. $F_{DDTe}$ were overall below the technical $F_{DDTe} = 0.74$, which is evidence of aged DDT in air at all stations, i.e. more $p, p'$-DDT converted to $p, p'$-DDE than would be the case in recent emissions. $F_{DDTe}$ decreased significantly at Zeppelin ($r^2 = 0.72$, $p < 0.001$) from 1994 to 2017 which implies that there was an increasing proportion of aged sources of DDT. At Alert, $F_{DDTe}$ fluctuated between 1992 and 2005, but it has been declining steadily since then. At Andøya, Villum Research Station and Pallas, there were no obvious changes in $F_{DDTe}$ over the monitoring period. The decreasing trends of $F_{DDTe}$ in Zeppelin and Alert indicate aged DDT residue was becoming the dominant DDT species in Arctic air. Regarding seasonality, we observe that there was a clear seasonal cycle in $F_{DDTe}$, in which it showed a winter minimum and summer maximum. This means $p, p'$-DDE dominates in winter and $p, p'$-DDT in summer. We will discuss the possible cause of elevated $p, p'$-DDE at Zeppelin in winter in Section 4.2. $F_{DDTo}$ in air at all stations were below the technical vapour $F_{DDTo}$ of 0.58, which indicates the DDT are aged and this is consistent with the $F_{DDTe}$ (Fig. S10c). Furthermore, $F_{DDTo}$ in air at all stations were well above the dicofol-$F_{DDTo}$ value. This implies that the DDT in Arctic air are mainly

associated with aged technical DDT rather than use of dicofol. However, others have reported that the proportion of $o, p'$-DDT to $p, p'$-DDT was increasing in air at the Great Lakes which suggest that dicofol may become a significant source of DDT in the future (Venier and Hites, 2014; Shunthirasingham et al., 2016).

*4.1.6. Pentachloroanisole (PCA), methoxychlor, hexachlorobutadiene (HCBD)*

PCA, the most common biotransformation product of pentachlorophenol (PCP), is often used as a proxy for PCP (UNEP, 2013). PCP was mainly used as a preservative in textiles and wood products, and as a pesticide in agricultural practices. PCP was listed in Annex A of the SC in 2015. PCA has been monitored in Alert air since the 1990s, but we only derived the trend from recent years due to analytical difficulties, i.e. the suspected co-elution of PCA with 1, 2, 4, 5-tetrachloro- 3, 6-dimethoxybenzene in samples that were analyzed in the early years (AMAP, 2017). At Alert, PCA showed a slowly declining trend from 2013 to 2017 with $t_{1/2} = 6.6$ y (Fig. S13). We caution that there have been reports that there are sources other than PCP contributing to PCA (Kylin et al., 2017); for instance, degradation of chlorinated hydrocarbons including PCB, HCB, HCH, and pentachloronitrobenzene (PCNB) could yield both PCP and PCA. Hence, PCA may not be an ideal proxy for PCP.

Methoxychlor is an OC designed to replace DDT. It is currently under review for listing under the SC. Methoxychlor was monitored in Alert air, but only detected in 9% of the samples. The mean concentration and range for methoxychlor in Alert air was 0.50 pg m$^{-3}$ (<0.0037 to 14 pg m$^{-3}$) over the last 25 years, i.e. 1992–2017. Methoxychlor in air at Villum Research Station was monitored but it was below detection limit.

HCBD was used for industrial purposes and now it is mainly a by-product in the manufacturing of chlorinated hydrocarbons (Balmer et al., 2019). The SC listed HCBD as a POP under Annex A in 2015 for elimination and under Annex C as an unintentional product in 2017. Alert began to monitor HCBD in 2002 but the chemical showed high breakthrough, in which its median ratio of HCBD found in the back PUF/front PUF was 0.81. It is because HCBD is very volatile with log

octanol-air partition coefficient (log $K_{OA}$) of 5.1 at 25 °C (Rauert et al., 2018a). We consider the data as semi-quantitative, probably underestimating the real concentrations of HCBD in Alert air. The data are presented in Fig. S14 and briefly describe here. Overall, the median concentration of HCBD at Alert of 2002–2017 was 1.8 pg m$^{-3}$. The yearly average of HCBD from 2002 to 2017 ranged from 1.2 to 13 pg m$^{-3}$. A previous report of HCBD in air at a super-site in Japan ranged from 330 from 2900 pg m$^{-3}$ (Takasuga et al., 2017), in Latin America and Caribbean region, HCBD ranged from <20 to 120 pg m$^{-3}$ (Rauert et al., 2018a). Similar concentrations have been reported in Sweden with air concentration of 160 pg m$^{-3}$ (Kaj and Palm, 2004) and Finland with air concentration of 120 to 150 pg m$^{-3}$ (AMAP, 2017).

### 4.2. Polychlorinated biphenyls (PCBs)

Table S3 gives the half-lives of individual PCB congeners at all stations. Air concentrations of PCB28, 52, 101, 118, 138, 153, 180 exhibited declining trends at most stations (Figs. 6a and S15). It is noted that the increasing levels of PCBs in air at Stórhöfði during 2015–2017 is due to the sampling equipment issue described previously (Section 4.1.3).

At Stórhöfði and Pallas, air concentrations of PCB28, 52, and 101 showed winter minimum and summer maximum concentrations. Summer maximum concentration implies that the chemical concentration was influenced by volatilization from ocean or land. The seasonal cycles of other PCB congeners at Stórhöfði and Pallas were not noticeable. This is likely because the high molecular weight PCBs are less volatile, their atmospheric concentrations are not influenced by the elevated temperature in summer time.

On the contrary, at Andøya, PCB28 and 52 showed winter maximum and summer minimum concentrations, so did PCB101 at Andøya and Zeppelin, and PCB28 at Alert (Fig. S15). Ubl et al. (2017) investigated the source of PCBs at Zeppelin by applying the Lagrangian Particle Dispersion Model FLEXPART. The authors suggested that the elevated PCB101 in winter was due to air arriving from the European continents

that is more contaminated. In contrast, air arriving from ocean to Zeppelin in summer had low concentrations of PCB101. The situation is different with PCB28 in which higher concentration was measured in summer when the clean air was coming from the ocean. Ubl et al. (2017) believed that local emission sources from Svalbard as well as long-range transport governed PCB28 levels in air at Zeppelin.

The situation of PCB101 may also explain the seasonal cycle of $p$, $p'$-DDE and TC in air at Zeppelin as discussed previously. Analogous to PCB101, the contaminated air arriving from European continents may carry a higher amount of $p$, $p'$-DDE and TC in winter than the summer oceanic air. Hence, a winter maximum and summer minimum concentration was observed (Fig. 6a).

### 4.3. Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD)

Fig. 6b shows the time trends of BDE47 and other congeners are shown in Fig. S16. Table S4 presents the half-lives of individual PBDE congeners. Air concentrations of PBDE congeners were declining at most of the stations. Exceptions were BDE99 and BDE100 at Stórhöfði as their concentrations were slowly rising. This is probably due to the change in sampling equipment in 2015 as mentioned in Section 4.1.3. In a previous report, PBDEs at Alert were stable from 2002 to 2012 (Hung et al., 2016). With an additional five years of data here, we found decreasing trends for most PBDE congeners. This may be explained by the reduction of emissions of technical penta- and octa-BDE mixtures in North America and Europe in mid-2000s, followed by their addition to the SC in 2009 (de Wit et al., 2010; USEPA, 2009, 2013). The declining trends of atmospheric PBDE at Alert are consistent with the previous report of Yu et al. (2015), who found that PBDE levels in air at Little Fox Lake dropped sharply from 2012 to 2014.

Alert showed a strong cyclical pattern in PBDE concentrations. We observed maximum summer and minimum winter concentrations for the relatively more volatile congeners, i.e. BDE47 (Fig. 6b), 99 and 100
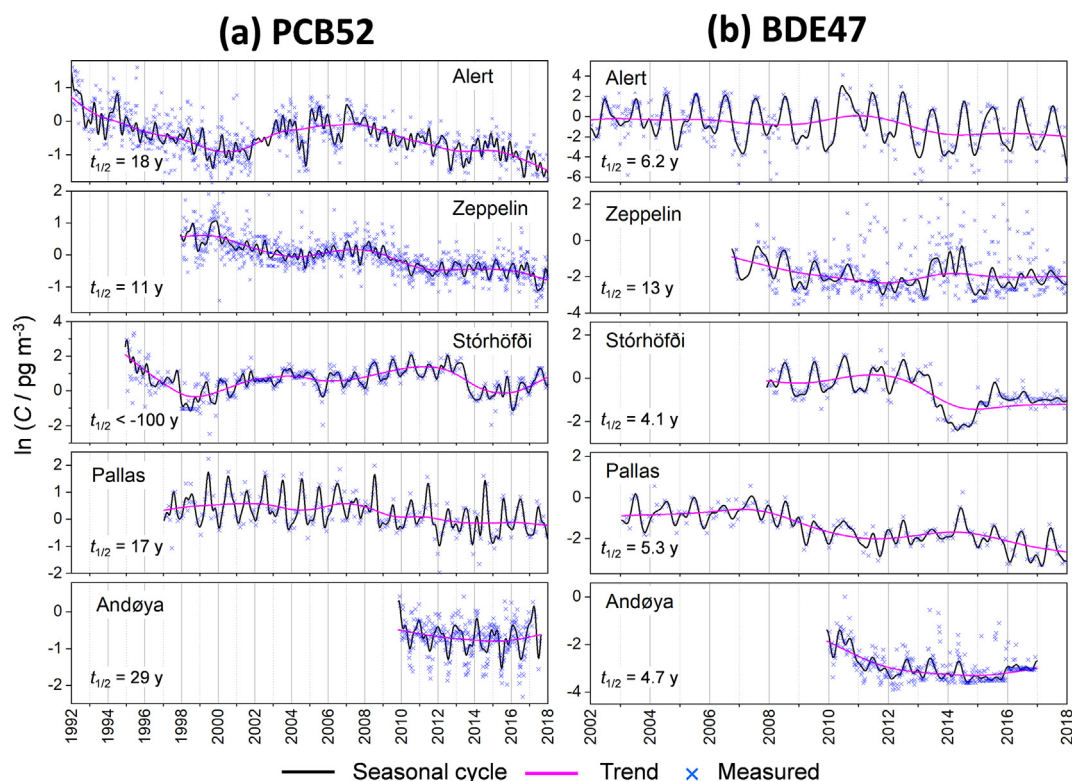


**Fig. 6.** Time trends, seasonal cycles, and measurements of (a) PCB52 (b) BDE47.

(Fig. S16). The same seasonal cycle is observed with BDE47 at Pallas. Hence, re-volatilization is likely controlling the atmospheric levels of these congeners. BDE138 did not show any seasonality because it is mostly particle-bound and episodic transport events are likely the cause in the fluctuation of its atmospheric concentration (Fig. S16).

At Alert, BDE209 (decaBDE) was moderately decreasing with $t_{1/2} =$ 4.8 y (Fig. S17a). This could be due to voluntary measures to reduce emissions initiated in mid-2000 in Europe in combination with the phase-out effort of BDE209 in Canada and U.S. in 2013 (de Wit et al., 2010; Commission Regulation (EU), 2017; USEPA, 2009, 2013; EC, 2013). BDE209 was added to the SC in 2017. The proportion of BDE209 to the ∑PBDEs at Alert (i.e. 47, 99, 100, 138, 209) was on average 22%, which remained constant compared to the previous AMAP assessment of the 2006–2012 data (AMAP, 2017) (Fig. S17b). In air from the Great Lakes, an increasing proportion of BDE209 was found from 2005 to 2014. That could be a result of the regulation of the penta- and octa-BDE in North America (Shunthirasingham et al., 2018).

At Zeppelin and Andøya, BDE209 showed slowly increasing and non-changing trends with $t_{1/2} = -14$ and $-25$ y, respectively (Fig. S17a). Compared with Alert, the percentage of BDE209 was much higher at these stations. At Zeppelin, the fraction of BDE209 ranged from 19 to 88%, during 2006 to 2016, and increased to 97% in 2017 (Fig. S17b). At Andøya, there was also an increase in proportion of BDE209 in the ∑PBDEs in 2017, from 13–57% during 2010 to 2016 to 74% in 2017. The elevated proportion of BDE209 could be due to the phase-out of the commercial penta-PBDE. This corresponds to the observation by Saini et al. (2020) that BDE209 in air in major cities around the world predominates the BDE profile. We caution that we found a high level of BDE209 in the field blanks in the Norwegian samples, which suggests possible contamination during sampling or transportation. On the other hand, the BDE209 levels in the high episodic samples

are up to 100 times higher than the levels in field blanks. No correlation to specific air masses is noticed for the high episodes. Often the air masses for the sampling periods come from west to south-west (e.g. Iceland or Greenland) (Bohlin-Nizzetto et al., 2019).

Zeppelin and Alert were the only stations that have HBCDD measurements and their concentrations were low (Fig. S18). At Zeppelin, the three isomers of HBCDD (α-, β- and γ-) were only detected in 14%, 3%, 9%, respectively, of the samples taken from 2006 to 2017. The most frequently detected isomer was α-HBCDD. At Alert, we report HBCDD as the sum of the three isomers, and the detection frequency was 29% from 2002 to 2017. Fig. S18 shows the seasonal cycle and measured data for Alert and Zeppelin. We did not develop time trends for HBCDD because there were many values below detection limits. However, visual inspection of the data suggests that levels of HBCDD at Zeppelin were decreasing but levels at Alert remained stable.

### 4.4. Per- and polyfluoroalkyl substances (PFAS)

Perfluorooctane sulfonic acid (PFOS) was listed in Annex B (restriction) of the SC in 2009 and perfluorooctanoic acid (PFOA) was listed in Annex A (elimination) in 2019 (UNEP, 2020b). Alert, Zeppelin and Andøya have monitored the ionic PFAS since 2006. Fig. 7 presents the time trends for (a) PFOA and (b) PFOS at these sites. At Alert, air concentrations of PFOA increased from 2006 to 2013. After 2013, its concentration has been steadily declining. Hence, we derived the half-life of PFOA for Alert from the peak concentration, instead of for the entire monitoring period and the half-life was 4.8 y from 2013 to 2017. At Zeppelin and Andøya, no peak concentrations were observed. PFOA showed a gradual decreasing trend over the entire monitoring period. The half-life of PFOA at Zeppelin was 3.5 y and at Andøya, was 2.3 y.
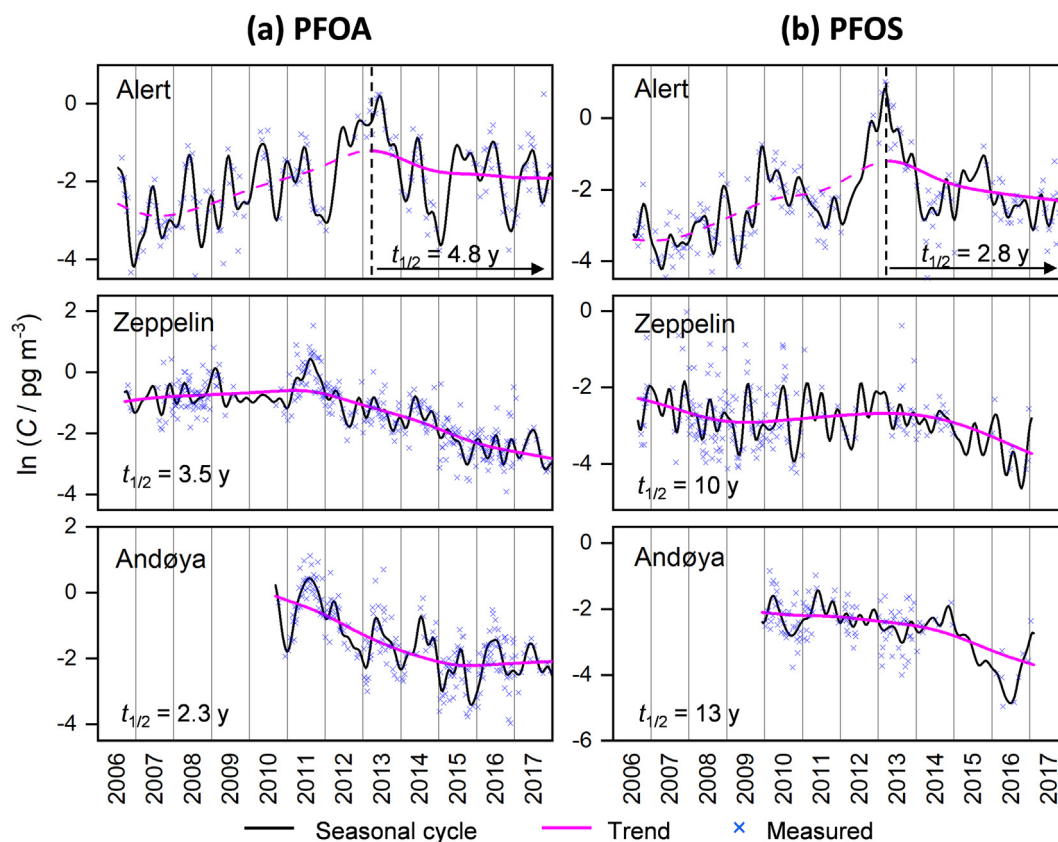


**Fig. 7.** Time trends and seasonal cycles of (a) PFOA and (b) PFOS at Alert, Zeppelin and Andøya. The half-lives were derived from the solid trend line. The dash line indicates the trend prior to the declining phase.

At Alert, PFOS showed a similar trend as PFOA for which we observed a peak in 2013. Its half-life from 2013 (i.e. the peak) to 2017 was 2.8 y which was considered moderate declining. Air concentrations of PFOS were slowly declining at Zeppelin ($t_{1/2}$ = 10 y) and Andøya ($t_{1/2}$ = 13 y).

We recognized that at Zeppelin, both PFOA and PFOS were decreasing at a faster rate in this study (i.e. up to 2017) than the rates in previous reports based on data up to 2014 (Wong et al., 2018). A similarly accelerated rate of decline was seen at Alert for these two chemicals. This may be due to the continuing global efforts in the phase-out of PFOS and PFOA and related substances since the 2000s (Paul et al., 2009; Wang et al., 2014).

Rauert et al. (2018b) reported elevated PFOS levels in air at Alert and Alaska in passive air samples collected in 2015 than those from 2009 and 2013. Here, we did not notice such an increasing trend at our Arctic stations. Muir et al. (2019) reported that perfluoroalkyl acids (PFAAs) in Arctic biota showed declining trends in the 2000s, but followed by increasing trends after 2010. The increasing trend in Arctic biota is probably more related to oceanic transport than to atmospheric transport due to the high water solubility of the PFAAs. Continued monitoring of these chemicals is necessary in order to provide insights to the trends in Arctic biota and other media.

We have analyzed trends of other short and long chain PFAAs. However, the analysis was hampered by their low detection frequencies and inconsistent blank values, which led to high method detection limits (MDL) in some sampling years. To be conservative, we have developed the trends only using data that were above the MDL. The trends of perfluorohexane sulfonic acid (PFHxS), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA) are presented in Fig. S19. At Alert, PFHxS appeared to decline from 2013 but this is probably driven by the few high measurements in 2013 and low measurements in 2017. Rauert et al. (2018b) reported that PFHxS in global passive air samples were increasing from samples collected in 2009, 2013 and 2015. At Alert, PFNA showed non-changing trend and PFDA and PFUnDA showed increasing trends. At Zeppelin and Andøya, these chemicals were not detectable.

Atmospheric levels of the PFAS precursor compounds, i.e. the long-chain fluorotelomer alcohols, decreased steadily at Alert and Villum Research Station (Fig. S20); except for 6:2 FTOH at the Villum Research Station which showed an increasing trend. At Alert, air concentrations of 8:2 and 10:2 FTOH were increasing previously from 2006 to 2012, followed by decreasing trends. We derived the half-lives for the declining period, i.e. 2012 to 2017 and the half-lives for 8:2 FTOH was 4.0 y and 10:2 FTOH was 3.0 y. At Villum Research Station, 8:2 and 10:2 FTOH were showing decreasing trends for the entire monitoring period (2008–2017). The half-lives for 8:2 FTOH and 10:2 FTOH were 10 y and 5.3 y, respectively. 6:2 FTOH at Alert was slowly decreasing with $t_{1/2}$ = 13 y but it was slowly increasing at Villum Research Station with $t_{1/2}$ = −14 y. Our results were consistent with the findings of Rauert et al. (2018b) that stable air concentrations of FTOHs were observed in passive air samplers deployed in Europe from 2009 to 2015. In global air, Gawor et al. (2014) reported that 8:2 and 10:2 FTOH showed a decreasing trend in XAD-based passive air sampling from 2006 to 2011.

8:2 and 10:2 FTOH showed seasonality at Alert and Villum Research Station with maximum summer and minimum winter concentrations. At Alert, it is interesting to see that the seasonal cycle of 8:2 and 10:2 FTOH became more apparent when the concentrations were in the declining phase (i.e. after 2011). This may indicate that after 2012, re-emission from secondary sources is the dominant controlling factor of their levels in the atmosphere. However, 6:2 FTOH showed an opposite trend at both sites in which the concentration was lowest in summer and highest in winter (Fig. S20). Piekarz et al. (2007) estimated the atmospheric residence times for the FTOHs using the Junge method and found that 6:2 FTOH had the shortest residence time of 50 d, compare to 8:2 FTOH of 80 d and 10:2 FTOH, of 70 d. There could be enhanced degradation of 6:2 FTOH in the summer and hence lower

concentrations. But from a smog chamber study, Ellis et al. (2003) reported similar atmospheric lifetimes of 20 d for the FTOHs. Continual monitoring of 6:2 FTOH is necessary because there have been concerns about its long persistency in humans from chronic dietary exposure. Rice et al. (2020) reported that 6:2 FTOH could be more toxic than its degradation products. The manufacturers of 6:2 FTOH have agreed to phase-out its use in the beginning of 2021 (FDA, 2020).

Perfluorooctane sulfonamidoethanol (FOSEs) and perfluorooctane sulfonamide (FOSAs) in air at the Villum Research Station showed different trends (Fig. S21). For example, ethyl (Et) -FOSA and EtFOSE were increasing ($t_{1/2}$ = −1.9 y, $t_{1/2}$ = 4.6 y respectively), methyl (Me) -FOSA was slowly declining ($t_{1/2}$ = 8.6 y) and MeFOSE was non-changing ($t_{1/2}$ < −100 y). Gawor et al. (2014) reported a global decline of FOSEs and FOSAs in air from 2006 to 2011 reported under the Global Atmospheric Passive Sampling programme which has not been observed in Arctic air here.

### 4.5. POPs monitoring in Russia

In 2015, two monitoring stations in Russia were established. These stations were located at Amderma and Tiksi. We did not calculate time trends for the Russian sites because only 3 years of data are available. The atmospheric concentrations at the two Russian sites were presented in Figs. S1 to S3. In brief, OCs and PCBs in air at Amderma seem to be higher and more variable than those found in Tiksi and other Arctic sites. This may be due to the differences in historical usage of POPs in Russia and elsewhere. It is interesting to observe that the tri- and tetra- PCBs (PCB28 and 31) in Russian air accounted for about 35% of the total PCBs, which was much lower than those in air at other Arctic sites (~60–80%) (Fig. S22). Breivik et al. (2002) reported that the technical PCB used in Russia, Sovol, contained about 24% of tri- and tetra-PCBs, which is consistent with the PCB composition in air from Amderma and Tiksi. Hung et al. (2010) similarly reported that the congener profiles in air at Valkarkai, Russia resembled those of technical PCBs used in Russia.

## 5. Conclusion

Temporal trends of most POPs in air at the Arctic monitoring stations showed declining trends and approaching detection limits. Many POPs exhibited seasonality with winter minimum and summer maximum concentrations or vice versa. However, not all chemicals at a specific station demonstrated the same seasonality; and the same chemical behaved differently at different stations. For example, PCB52 was high in winter and low in summer at Zeppelin but the reverse pattern, i.e. low in winter and high in summer at Pallas. Again, at Zeppelin, p, p-DDE was high in winter and low in summer, but the opposite seasonality was observed for α-HCH. This is because each station is unique in its physical environmental conditions, and has different environmental processes and sources, which governs the fate of chemicals at each station; hence, it is not surprising to observe such discrepancy in chemical fate and transport behaviour.

Our results confirm the effectiveness of chemical control measures by the SC to reduce POP contaminations in the atmospheric environment. However, it is known that many of the POPs began to decline well before the establishment of the SC. For example, α-HCH showed declines in the 2000s which is before they were added to the SC in 2009. Hence the declining trends of POPs were not solely due to the establishment of the SC, but together with industries' voluntary phase-out and national regulations.

On the contrary, POPs that showed stabilizing or increasing trends could be due to ongoing emission from landfills, waste or illegal stockpiles (Gewurtz et al., 2020; Vijgen et al., 2018). For POPs that are widely used in consumer products, such as PBDEs and PFOS, they have a long lifespan, thus remain in households or landfills many years after being regulated. Golnoush et al. (2019) pointed out that a significant amount of PBDEs could be recycled and reappear in plastic products. Another

plausible explanation is that there could be a lag time from a chemical being regulated to an observed decrease in air at remote sites. Shunthirasingham et al. (2018) analyzed long-term trends of PBDEs in air at Great Lakes found that the sites closer to urban settlements respond quicker to the withdrawal of PBDE technical mixtures from the market than the rural sites. Furthermore, there could be enhanced volatilization of chemicals from contaminated surfaces due to warming temperature driven by climate change.

Long-term monitoring programmes with consistent data quality are essential to evaluate the effectiveness of chemical control efforts. Under AMAP, we will continue to strive for good data comparability and consistency by participating in annual interlaboratory comparisons as additional POPs are being added to the SC and other national and international chemical control strategies.

We have demonstrated the importance of monitoring POPs or CEAC before they are being regulated especially because model calculations to predict their environment fate cannot be made without accurate emission inventories. For example, we were able to capture the peak concentrations of endosulfan in air at Alert, Pallas and Villum Research Station, and PFOS, PFOA, 6:2 and 8:2 FTOH in air at Alert and estimated their half-lives during their declining phase. This provided confidence in the rate estimation.

One particular challenge in studying the POPs and CEAC is that the Arctic monitoring stations are intended to act as remote sites that are far from global sources. However, many of the chemicals are found in consumer products (e.g. electronics, clothing, furniture, plastics) and in building materials. Hence, the detection of chemicals in the Arctic may not only due to long-range transport, but also to local sources such as waste disposal sites. We need to increase our efforts in characterizing chemical release from local Arctic communities, so that we can distinguish the contribution of chemicals from local vs. long-range transport.

Currently, most of the air monitoring stations are located in the Canadian and European Arctic region. Only two sites, Tiksi and Amderma are located in Russia where we have presented baseline data here. The Russian datasets were not sufficient to develop time trends and hence continual monitoring effort is essential. Increasing spatial coverage of the air sampling sites in the Russian Arctic region could improve the risk assessment of POPs.

## CRediT authorship contribution statement

**Fiona Wong:** Writing-original draft preparation, Laboratory and formal analysis, Visualization, Investigation. **Hayley Hung:** Conceptualization, Methodology, Validation, Supervision, Laboratory resources, Investigation, Writing – review & editing, Project Administration (Alert, Little Fox Lake), Funding Acquisition (Alert, Little Fox Lake). **Helena Dryfhout-Clark:** Data Curation, Validation, Writing - Review & Editing. **Wenche Aas:** Writing - Data Curation, Investigation, Review & Editing, Funding Acquisition (Zeppelin and Andøya). **Pernilla Bohlin-Nizzetto:** Investigation, Writing - Review & Editing, Project Administration (Zeppelin and Andøya). **Knut Breivik:** Investigation, Writing - Review & Editing. **Michelle Nerentorp Mastromonaco:** Investigation, Writing - Review & Editing, Project Administration (Pallas), Funding Acquisition (Pallas). **Eva Brorström Lundén:** Investigation, Writing - Review & Editing. **Kristín Ólafsdóttir:** Investigation, Writing - Review & Editing, Project Administration (Stórhöfði). **Árni Sigurðsson:** Field Sampling, Investigation, Writing - Review & Editing, Funding Acquisition (Stórhöfði). **Katrin Vorkamp:** Investigation, Writing - Review & Editing, Project Administration (Villum Research Station), Funding Acquisition (Villum Research Station). **Rossana Bossi:** Investigation, Writing - Review & Editing, Project Administration, Funding Acquisition. **Henrik Skov:** Investigation, Writing - Review & Editing, Project Administration (Villum Research Station), Funding Acquisition (Villum Research Station). **Hannele Hakola:** Investigation, Writing - Review & Editing, Funding Acquisition (Pallas). **Enzo Barresi:** Laboratory analysis, Writing - Review & Editing. **Ed Sverko:** Laboratory analysis, Writing - Review & Editing. **Phil Fellin:** Investigation. **Henrik Li:** Validation, Investigation. **Alexander Vlasenko:** Validation, Investigation. **Mikhail Zapevalov:** Investigation, Writing - Review & Editing, Project Administration (Amderma and Tiksi), Funding Acquisition (Amderma and Tiksi). **Dmitry Samsonov:** Investigation, Writing - Review & Editing, Project Administration (Amderma and Tiksi), Funding Acquisition (Amderma and Tiksi). **Simon Wilson:** Project Administration, Writing - Review & Editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.145109.

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