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Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006[☆]

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ABSTRACT

Continuous and comparable atmospheric monitoring programs to study the transport and occurrence of persistent organic pollutants (POPs) in the atmosphere of remote regions is essential to better understand the global movement of these chemicals and to evaluate the effectiveness of international control measures. Key results from four main Arctic research stations, Alert (Canada), Pallas (Finland), Storhofdi (Iceland) and Zeppelin (Svalbard/Norway), where long-term monitoring have been carried out since the early 1990s, are summarized. We have also included a discussion of main results from various Arctic satellite stations in Canada, Russia, US (Alaska) and Greenland which have been operational for shorter time periods. Using the Digital Filtration temporal trend development technique, it was found that while some POPs showed more or less consistent declines during the 1990s, this reduction is less apparent in recent years at some sites. In contrast, polybrominated diphenyl ethers (PBDEs) were still found to be increasing by 2005 at Alert with doubling times of 3.5 years in the case of deca-BDE. Levels and patterns of most POPs in Arctic air are also showing spatial variability, which is typically explained by differences in proximity to suspected key source regions and long-range atmospheric transport potentials. Furthermore, increase in worldwide usage of certain pesticides, e.g. chlorothalonil and quintozene, which are contaminated with hexachlorobenzene (HCB), may result in an increase in Arctic air concentration of HCB. The results combined also indicate that both temporal and spatial patterns of POPs in Arctic air may be affected by various processes driven by climate change, such as reduced ice cover, increasing seawater temperatures and an increase in biomass burning in boreal regions as exemplified by the data from the Zeppelin and Alert stations. Further research and continued air monitoring are needed to better understand these processes and its future impact on the Arctic environment.

1. Introduction

Atmospheric long-range transport (LRT) is the most rapid route for organic contaminants to reach the ecologically sensitive Arctic environment. Long-term measurements of persistent organic pollutants (POPs), including organochlorine pesticides (OCPs) in the Arctic atmosphere have been conducted at 4 stations under the Canadian Northern Contaminants Program (NCP), the national Norwegian pollutant monitoring programs (SFT), the Arctic Monitoring and Assessment Programme (AMAP), the Paris Convention (PARCOM-Comprehensive Atmospheric Monitoring Program — CAMP) and the European Monitoring and Evaluation Programme (UNECE-EMEP). The four stations representing the longest time series are Alert (Canada,

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[☆] This paper is a contribution to the AMAP POPs assessment.

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82° 30′ N, 62° 20′ W, 1992–2005), Zeppelin Mountain (Svalbard/ Norway, 78° 54′ N, 11° 53′ E, 1993–2006), Pallas (Matorova, Finland, 68° 00′ N, 24° 15′ E, 1996–2005) and Storhofdi (Vestmannaeyjar, Iceland, 63° 24′N, 20° 17′W, 1995–2005) (Fig. 1). In addition to these stations, there were 8 Arctic satellite stations under AMAP which were in operation at various times for periods ranging from 3 months to 2 years. These satellite stations include the three Canadian stations of Tagish, Little Fox Lake and Kinngait; the three Russian stations of Dunai, Amderma and Valkarkai; the Alaskan (U.S.) station of Barrow and the Nuuk station in southwest Greenland (Fig. 1). One common objective of the international monitoring programs within AMAP and UNECE-EMEP is to provide sound scientifically validated information on the presence (transport, transformation and distribution) of pollutants, although their geographical coverage in the Arctic is different. This information is required to support pollution regulations, as well as to give input to the development and further evaluation of international control strategies on emission, production and usage of organic chemicals, such as the Stockholm Convention on POPs. Long-term temporal trends of atmospheric POPs and other OCPs are required for assessing the effectiveness and efficiency of such international control strategies. In addition, new and emerging chemicals are occasionally included in the monitoring lists to provide air concentration and trend data in remote Arctic locations to facilitate the evaluation of potential emerging POPs. In this article, the current status of the atmospheric levels and long-term trends of organic pollutants at the abovementioned Arctic sites are evaluated and summarized. Potential factors that may influence the spatial differences and similarities in concentrations and temporal trends observed at the various stations are assessed. Measurements of atmospheric polybrominated diphenyl ethers (PBDEs) started at Alert in 2002 and an update of the dataset previously reported by Su et al. (2007) is presented here with a comparison to concentrations measured at Nuuk (Bossi et al., 2008).

2. Experimental procedure and data analysis

2.1. Sample collections and chemical analysis

Air sampling was conducted at all stations by high volume air samplers with glass fiber filter (GFF) and polyurethane foam plugs (PUFs) as sampling media. Sampling frequencies vary from site to site. At Alert, samples representing ~13,000 m^3 of air were taken each week of the year. For Zeppelin, 48-h sampling collected ~1000 m^3 of air every week. At Pallas, seven-day integrated samples were collected once a month with an approximate sample volume of 4000 m^3, while at Storhofdi 1000 m^3 of air was collected once every 2 weeks. When in operation, air sampling at all satellite stations, except Nuuk, follow the same sampling protocol as Alert. Sampling at Nuuk occurred over a period of about 14 days with a sample volume of 3500–4500 m^3 (Bossi et al., 2008). The sampling period at each station is given in Table 1. Detailed description of field sites, sampling processes, analytical methods and QA/QC procedures can be found in Hung

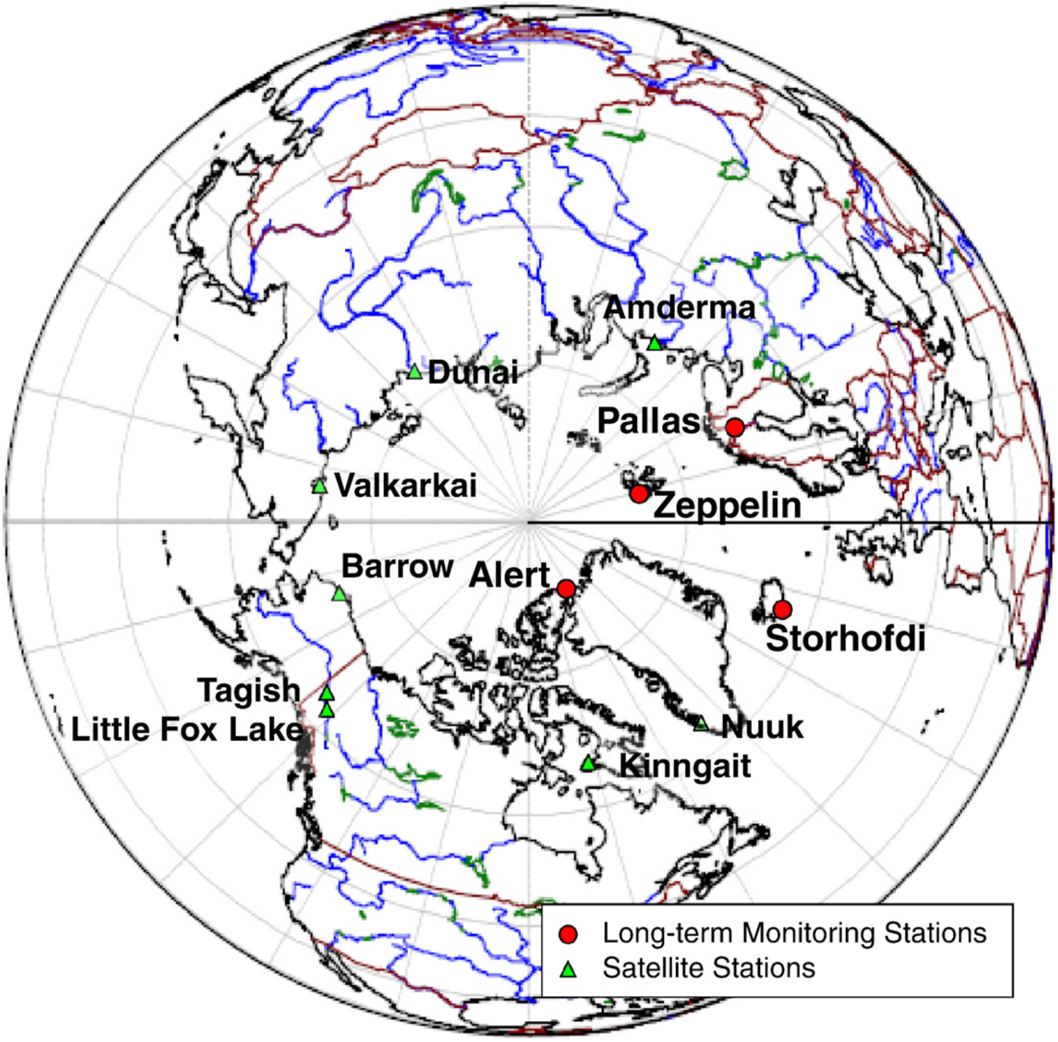

Fig. 1. Arctic air monitoring station map.

Table 1
AMAP air monitoring stations.

Station name	Country	Location	Elevation (masl)	Monitoring period
Long-term stations				
Alert	Canada	82° 30′ N 62° 20′ W	200	1992–2005
Pallas/ Matorova	Finland	68° 00′ N 24° 15′ E	340	1996–2005
Storhofdi	Iceland	63° 24′ N 20° 17′ W		1995–2005
Zeppelin mountain	Svalbard/ Norway	78° 54′ N 11° 53′ E	474	1993–2006
Satellite stations				
Tagish	Canada	60° 20′ N 134° 12′ W	~1000	1992/12/05–1995/03/30
Little Fox Lake	Canada	61° 21′ N 135° 38′ W	1128	2002/07/07–2003/07/06
Kinngait	Canada	64° 13′ N 76° 32′ W	119	I: 1994/03/15–1996/02/25 II: 2000/10/16–2002/09/06
Amderma	Russia	69° 43′ N 61° 37′ E	Sea level	1999/03/28-2001/04/02
Dunai	Russia	74° 6′ N 124° 30′ E	Sea level	1993/03/09–1995/04/31
Valkarkai	Russia	70° 5′ N 170° 56′ E	Sea level	2002/07/29–2002/09/30
Barrow	U.S.A.	71° 19′ N 156° 37′W	9	2002/03/19–2003/04/23
Nuuk	Greenland	64° 6′ N 51° 24′ W		2004–2005

et al. (2005), Bossi et al. (2008) and on the UNECE-EMEP website: http://www.emep.int. At Alert, the particle (on GFF) and vapour (on PUFs) phases were analyzed separately before 2001. After that, the two fractions of each weekly sample were combined for analysis to reduce cost. Total (gas + particle) phase concentrations were reported for all the other stations. For ease of discussions, air concentration data will be presented as total (gas + particle) phase in the current study.

Only selected compound groups are regularly monitored at the four atmospheric monitoring stations, these include priority POPs under the Stockholm Convention on POPs. Continuous monitoring data exists only for PCBs, dichlorodiphenyltrichloroethane isomers (DDTs) and transformation products (DDEs and DDDs); and α- and γ-hexachlorocyclohexane (α- and γ-HCH) for all 4 stations. Three of the stations (Alert, Zeppelin, Storhofdi) also reported air concentrations on hexachlorobenzene (HCB) and chlordanes (*trans*- and *cis*- isomers of chlordane and nonachlor). Alert also includes heptachlor, mirex, dieldrin, trifluralin and endrin in the analytical list but most of these compounds were found to be below or very close to detection limits in Arctic air, except for occasional high concentration episodes. Therefore, air concentrations of these chemicals were not used for assessments of long-term trends.

2.2. Data handling and quality assurance

Data comparability is a key goal of both AMAP and UNECE-EMEP. The EMEP stations (Storhofdi, Pallas and Zeppelin) were selected according to recommended EMEP site criteria guidelines. Participating institutions and laboratories are encouraged to use the recommended field and laboratory analytical methods by EMEP, but alternative methods which can be shown to be equally reliable may be employed.

The Alert air sampling station and all satellite stations (except Nuuk) operate according to the sampling and analytical protocols under the Canadian-operated NCP which is Canada's National Implementation Plan of AMAP. Under NCP, method detection limits (MDLs) (average field blank concentrations plus three standard deviations from the mean) are reported for each sampling year. Recovery and internal standards are included in each sample extract at all sampling sites. EPA standard reference materials are routinely included in the analytical sequence. The analytical laboratories regularly participate in NCP, AMAP and EMEP recommended interlaboratory comparison studies (e.g. Manø and Schaug, 2003). Data reported are not blank- or recovery-corrected. To retain the maximum amount of data, air concentrations below MDLs are included in all analysis. Samples that are found to be below instrument detection limit (IDL) were replaced with 2/3 IDL of the corresponding compound. The Research Data Management and Quality Control System (RDMQ), which is a SAS-based statistical database, with an extensive flagging system is used to track and calculate all air concentrations and quality control information.

The OCPs and PCBs air concentration data measured at Alert in 1992 did not meet quality control criteria as indicated in Stern et al. (1997). Therefore data from this year are not included in the current study. In 2002, as part of a decision to integrate analytical activities within Environment Canada, chemical analyses of samples from Alert have been moved from Fresh Water Institute (FWI), Winnipeg, which is a laboratory of the Department of Fisheries and Oceans, to the National Laboratory for Environmental Testing (NLET), an Environment Canada laboratory. NLET maintains the historical analytical procedures employed by FWI for consistency. Two rounds of rigorous interlaboratory comparison have been conducted between FWI and NLET, using samples collected at Alert in 2002 and 2004. Results from these comparison studies may be used to eliminate the discrepancies between the two laboratories in order to maintain the long-term trends as more data becomes available in the future. In the current study, the long-term trends have not been corrected to reduce the error involved in data interpretation.

For the Norwegian, Finnish and Icelandic sites, the same SRM certified material was added to the samples in the analytical sequence. Same as NCP, the Norwegian national monitoring program at the Zeppelin station (Svalbard) also reports annual MDLs for each compound. Quantification methods performed by the Norwegian Institute for Air Research are accredited and certified according to the international accreditation standards ISO-9001 and EN/ISO-17025. Note that local PCB contaminations before 1998 rendered the PCB concentrations measured in samples collected at Zeppelin unreliable. The sampling unit was temporarily moved in 1998 to Ny-Ålesund. In 1999, a new research facility was built at the Zeppelin mountain and the local PCB contamination source was eliminated. Therefore, only PCB air concentration data from 1998 have been included in the temporal analysis in Section 3.2.1.

The quantification of POPs in high volume air samples from Storhofdi, Pallas and Zeppelin are performed according to the analytical quality guidelines given by AMAP (1998). The sampling protocols as well as method descriptions are given in earlier publications for the Zeppelin data (Oehme et al., 1996; Berg et al., 2004; Eckhardt et al., 2007; Kallenborn et al., 2007) and briefly in recent annual EMEP reports for all three sites (e.g. Aas and Breivik, 2008). The quantification was performed using isotope dilution techniques with standard gas chromatography coupled to mass selective detectors. Only for Storhofdi, POPs were quantified in atmospheric samples using gas chromatography and electron capture detector (Aas and Breivik, 2008).

All air concentration data are submitted to the AMAP and EMEP databases in a specified format on a continuous basis along with necessary meta-data, such as information on methods used, detection limits, analytical precision and site information. Both databases are hosted by the Norwegian Institute for Air Research (NILU) and the EMEP data are available on the internet at: http://www.nilu.no/projects/ccc/onlinedata/pops/index.html. Data that are not available on this website are available upon request.

2.3. Temporal trends and apparent first order halflives/doubling times

For evaluation of long-term temporal trends, the Digital Filtration (DF) Technique was used. DF is a statistical fitting technique which extracts seasonal cycles and inter-annual trends from time series. This technique has been successfully applied to derive long-term trends of atmospheric trace chemicals, including PCBs and OCPs at Alert (Hung et al., 2005). Detailed description of this technique can be found in Hung et al. (2005). Long-term trends and seasonal cycles were developed using the DF technique for organic contaminants measured at the 4 long-term monitoring stations. A disruption of the Alert temporal trends has resulted due to the laboratory change in 2002. Here, long-term trends and seasonal cycles were developed between 1993 and 2001 and between 2002 and 2005 separately.

A decline in air concentrations over time is often quantified by an apparent first order halflife, $t_{1/2}$, which is estimated by dividing ln 2 with the negative value of the linear regression slope of the trend line between the natural log of air concentrations, C (pg/m^3), and time (year). Note that many compounds presented in this study do not necessarily decline linearly or consistently in the first order manner throughout the monitoring periods of the 4 long-term stations. The halflives presented here are only used to compare the relative rates of decline between the 4 stations. Readers are advised to use the absolute values of these halflives with caution. Results of the trend analyses are given in Section 3.2.

With 3 years of monitoring data at Alert, Su et al. (2007) have shown increasing tendencies of atmospheric PBDE concentrations from 2002 to 2004. In this study, the trends will be updated using data up to 2005. Doubling times, t_2, are calculated by dividing ln 2 with the positive value of the linear regression slope of the DF-derived trend line.

Concentration measurements at satellite stations were conducted for one or two years only (except for Valkarkai where measurements only occurred for 3 months in 2002) and, thus, are not continuous. Therefore, it is not possible to derive temporal trends for these locations based on the currently available data. However, if comparable atmospheric measurements were to be restarted at these locations/regions, it will be possible to estimate changes in atmospheric concentrations of POPs over time at a later stage. Two examples of this type of analysis are given in Section 3.3. Changes in OCPs and PCBs air concentrations measured at Kinngait from 1995 to 2001 are compared to those at Alert over the same period. A similar analysis was provided in Hung et al. (2005). However, at that time, samples from Alert in 2001 were still under analysis and were not available. Alert data from 1998–1999 were used for comparison instead. An updated analysis is given here. Another example is given to compare the changes in air concentrations measured in the western Canadian Arctic in the Yukon region. Tagish and Little Fox Lake are located in relatively close proximity and samples from these sites were both analyzed by the same laboratory, namely FWI, allowing for an analysis of atmospheric concentration change between 2002–2003 (Little Fox Lake) vs. 1993 (Tagish) in the Yukon.

2.4. Spatial comparisons of OCPs

Su et al. (2006, 2008) have summarized the spatial distribution of OCPs at 6 Arctic monitoring stations under AMAP between 2000 and 2003 in two publications. These two articles compared air concentrations, atmospheric compositions, temperature dependence and isomer ratios of OCPs measured at Alert, Zeppelin, Kinngait, Little Fox Lake, Barrow and Valkarkai, which were in operation simultaneously between 2000 and 2003. Brief summaries and major findings from these two articles will be given here under Section 3.1. Those interested in details of OCP spatial comparisons in Arctic air, please refer to the original articles (Su et al., 2006, 2008).

3. Results and discussions

3.1. Annual mean concentrations and ranges

Annual mean total (sum gas and particle phases) concentrations and ranges of sum of PCBs (ΣPCBs) of the long-term monitoring stations are given in Table 2. Those of the OCPs are given in Table 3. Air concentration data of PCBs and OCPs from the satellite stations are summarized in Table 4.

3.1.1. Polychlorinated biphenyls (PCBs)

Different stations report different numbers of individual and coeluting PCB congeners. Reported number of congeners are given in brackets as follows: Alert [FWI (1993-2001): 88; NLET (2002–2005): 106]; Pallas (7); Storhofdi (10); and Zeppelin (33). For comparison, Table 2 summarizes the ΣPCBs concentrations for the AMAP suite of 10 PCBs for Alert, Storhofdi and Zeppelin and sum of 7 reported PCBs for Pallas.

Since a smaller number of congeners are reported by Pallas, the following comparison does not include this site. Generally speaking, the air concentrations of PCBs were highest at Zeppelin from 1998 to 2001 and lowest at Alert, while Storhofdi showed similar concentrations. After 2001, the air concentrations at Zeppelin have declined to similar levels as Storhofdi. Atmospheric levels of PCBs at Alert remained to be the lowest and the air concentrations at this site were statistically significantly different (at a confidence level of 95%) from the other two stations between 1998 and 2005. This is probably because Alert is further away from global source regions of PCBs while Zeppelin and Storhofdi are affected by European PCB sources due to proximity.

Alert and Zeppelin reported a relatively larger suite of PCBs allowing for comparison of homologue distribution. Fig. 2 shows the homologue profiles at the two stations in terms of percentage of total reported PCBs. Tri-chlorinated CBs dominated the profiles at both stations. This corresponds to the fact that tri-CBs were the most important PCB homologue produced historically (Breivik et al., 2002). At Alert, there was a slight decline in relative contribution of lower chlorinated CBs from the 1990s to early 2000s with a slight increase in contribution of higher chlorinated homologues, e.g. tetra- to octa-CBs. Since production of PCBs has stopped in most industrialized countries during the 1970s and 80s, this shift in homologue profile may be considered as an indication that the air concentrations of lighter congeners are starting to level off as a result of environmental removal processes, e.g. photo- and bio-degradation. Increasing trends of equivalent black carbon (EBC) at Alert between 1998 and 2002 has been reported by Sharma et al. (2006). The increase in EBC may also enhance input of heavier PCB congeners which have higher tendencies to associate with particles. It was not possible to discern any temporal trends in homologue profile at the Zeppelin station as samples from the early 1990s were compromised by local PCB contamination.

Table 2
Atmospheric mean (standard deviation; range) of ΣPCBs concentrations (pg/m^3) at long-term monitoring stations.

Year/ location	ΣPCBs (pg/m^3)			
	Alert[a]	Pallas[b]	Storhofdi[a]	Zeppelin[a]
1993	5.8 (4.8; 0.18–26)	–	–	–
1994	6.1 (4.0; 1.2–26)	–	–	–
1995	5.1 (3.1; 2.2–22)	–	19 (14; 0.50–63)	–
1996	4.5 (2.5; 0.86–18)	11 (5.0; 5.8–22)	17 (23; n.d.–110)	–
1997	3.6 (1.6; 0.17–9.1)	4.6 (1.8; 2.9–8.3)	5.7 (6.7; n.d.–21)	–
1998	4.2 (1.4; 2.7–9.1)	6.2 (2.9; 3.5–13)	8.7 (14; 1.0–65)	34 (67; n.d.–330)
1999	5.8 (19; 0.17–137)	7.5 (7.0; 1.9–27)	6.7 (5.6; n.d.–20)	23 (19; n.d.–99)
2000	2.8 (1.6; 0.19–11)	6.4 (3.4; 2.1–14)	5.1 (2.5; 2.5–13)	14 (10; n.d.–58)
2001	2.2 (0.75; 0.70–4.8)	5.4 (3.0; 2.5–12)	11 (6.7; 4.7–39)	9.9 (5.8; 2.9–37)
2002	1.1 (1.9; 0.14–9.0)	6.2 (4.2; 2.6–15)	7.0 (2.4; 4.7–14)	7.6 (3.5; 2.8–16)
2003	3.9 (1.2; 2.4–8.2)	4.9 (3.7; 2.3–15)	12 (7.7; 4.5–42)	6.3 (2.4; 3.2–16)
2004	2.3 (1.3; 0.74–5.0)	5.5 (3.8; 1.8–14)	12 (2.4; 7.7–21)	5.7 (4.0; n.d.–29)
2005	4.9 (4.9; 2.0–38)	5.1 (3.0; 2.0–12)	7.8 (3.8; 1.9–17)	7.5 (2.6; 3.6–14)
2006	–	–	–	9.0 (4.2; 3.2–30)

[a] ΣAMAP 10 PCBs (PCB 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180).

[b] Sum of 7 PCBs (PCB 28, 52, 101, 118, 138, 153 and 180).

Table 3
Arithmetic mean (standard deviation; range) of total (GFF + PUF) air concentrations (pg/m^3) of selected OCPs.

(a) 1993–1999

	1993	1994	1995	1996	1997	1998	1999
Alert							
HCB	54 (18; 12–99)	51 (17; 1.8–96)	65 (26; 1.4–130)	62 (35; 0.33–240)	59 (25; 5.9–98)	70 (22; 28–130)	69 (19; 30–110)
α-HCH	57 (23; 12–130)	54 (26; 1.1–160)	56 (26; 2.1–140)	55 (31; 0.10–190)	47 (20; 12–96)	45 (13; 18–73)	34 (12; 1.5–68)
γ-HCH	13 (25; 2.0–170)	10 (7.4; 1.4–41)	8.4 (4.7; 0.89–25)	7.6 (8.5; 0.073–59)	6.5 (4.3; 0.99–17)	7.0 (4.9; 2.0–27)	5.9 (5.5; 1.0–30)
α/γ-HCH	6.6 (2.1; 0.13–11)	6.6 (6.3; 0.11–48)	6.7 (5.5; 2.3–5.7)	10 (10; 1.3–68)	9.2 (4.2; 3.8–24)	7.8 (2.9; 2.4–13)	8.3 (3.7; 0.72–18)
TC	0.42 (0.27; 0.088–1.5)	0.57 (0.33; 0.075–1.8)	1.0 (4.1; 0.024–29)	0.34 (0.19; 0.021–0.87)	0.36 (0.18; 0.025–1.1)	0.33 (0.16; 0.11–0.68)	0.23 (0.13; 0.052–0.60)
CC	0.92 (0.50; 0.22–2.6)	1.2 (0.62; 0.10–3.1)	0.76 (0.51; 0.024–1.9)	0.69 (0.45; 0.016–2.6)	0.57 (0.27; 0.029–1.4)	0.68 (0.32; 0.23–1.3)	0.60 (0.21; 0.21–1.2)
TC/CC	0.48 (0.15; 0.22–0.77)	0.47 (0.17; 0.23–0.88)	0.68 (0.33; 0.21–1.9)	0.61 (0.34; 0.030–1.6)	0.67 (0.21; 0.25–1.2)	0.52 (0.19; 0.27–1.0)	0.38 (0.16; 0.14–0.68)
TN	0.76 (0.50; 0.19–2.6)	1.1 (0.68; 0.11–3.2)	0.89 (0.50; 0.19–2.4)	0.70 (0.70; 0.017–5.1)	0.58 (0.27; 0.034–1.6)	0.76 (0.47; 0.15–2.5)	0.46 (0.23; 0.12–1.3)
OXY	0.76 (0.44; 0.16–1.9)	0.99 (0.48; 0.14–2.4)	0.66 (0.30; 0.037–1.4)	0.51 (0.39; 0.022–2.6)	0.40 (0.18; 0.040–1.0)	0.42 (0.18; 0.038–0.70)	0.34 (0.17; 0.063–0.83)
HEPX	1.0 (0.58; 0.23–2.5)	1.2 (0.67; 0.025–3.3)	1.2 (0.52; 0.19–2.5)	0.91 (0.61; 0.016–2.3)	0.71 (0.31; 0.030–1.4)	0.89 (0.37; 0.18–1.5)	1.0 (1.1; 0.19–6.3)
p,p′-DDT	0.12 (0.11; 0.023–0.54)	0.16 (0.13; 0.043–0.48)	0.17 (0.18; 0.043–0.98)	0.30 (1.2; 0.028–8.6)	0.14 (0.068; 0.043–0.40)	0.15 (0.12; 0.046–0.90)	0.11 (0.082; 0.031–0.45)
o,p′-DDT	0.20 (0.19; 0.023–1.0)	0.36 (0.30; 0.044–1.4)	0.36 (0.42; 0.044–2.2)	0.26 (0.16; 0.029–0.77)	0.28 (0.17; 0.043–0.86)	0.38 (0.39; 0.040–2.1)	0.21 (0.12; 0.032–0.46)
p,p′-DDE	0.31 (0.26; 0.027–1.0)	0.45 (0.33; 0.027–1.7)	0.43 (0.41; 0.026–1.7)	0.34 (0.43; 0.022–2.5)	0.37 (0.26; 0.049–1.1)	0.45 (0.35; 0.071–1.6)	0.47 (0.19; 0.16–0.92)
o,p′-DDE	0.11 (0.10; 0.019–0.43)	0.19 (0.16; 0.032–0.80)	0.16 (0.18; 0.035–0.83)	0.065 (0.077; 0.023–0.45)	0.13 (0.083; 0.030–0.44)	0.26 (0.34; 0.077–1.9)	0.12 (0.087; 0.026–0.44)
p,p′-DDD	0.041 (0.022; 0.017–0.16)	0.073 (0.11; 0.032–0.58)	0.091 (0.14; 0.031–0.88)	0.057 (0.71; 0.021–0.47)	0.069 (0.065; 0.030–0.41)	0.040 (0.021; 0.017–0.16)	0.042 (0.056; 0.023–0.42)
o,p′-DDD	0.048 (0.010; 0.021–0.074)	0.050 (0.010; 0.037–0.068)	0.057 (0.041; 0.037–0.25)	0.057 (0.065; 0.027–0.47)	0.048 (0.015; 0.035–0.13)	0.052 (0.031; 0.021–0.21)	0.051 (0.050; 0.029–0.33)
p,p′-DDT/*p,p′*-DDE	0.88 (0.96; 0.23–3.7)	0.91 (0.78; 0.082–3.2)	0.79 (0.79; 0.087–3.7)	0.86 (1.1; 0.053–6.3)	0.57 (0.40; 0.12–2.0)	0.51 (0.35; 0.11–1.4)	0.27 (0.15; 0.093–0.89)
PCA	2.5 (1.4; 0.10–5.6)	3.2 (1.4; 0.17–6.0)	2.6 (1.5; 0.31–7.6)	2.8 (2.3; 0.029–16)	1.3 (0.67; 0.31–3.0)	3.9 (2.2; 0.87–12)	4.4 (2.2; 0.30–8.9)
Diel	1.1 (0.68; 0.055–3.0)	1.4 (0.76; 0.18–3.4)	1.3 (0.64; 0.22–2.8)	1.1 (0.63; 0.079–3.9)	1.3 (0.70; 0.33–3.3)	1.2 (0.53; 0.29–2.8)	0.89 (0.58; 0.32–3.1)
OCS	0.52 (0.33; 0.093–1.6)	0.92 (0.32; 0.20–1.6)	0.83 (0.51; 0.10–2.3)	0.55 (0.25; 0.067–1.2)	0.33 (0.21; 0.10–1.4)	0.53 (0.13; 0.25–0.75)	0.47 (0.13; 0.15–0.75)
TeCV	0.82 (0.47; 0.16–1.8)	0.94 (1.4; 0.13–9.2)	1.6 (1.0; 0.15–3.7)	1.1 (0.96; 0.090–6.1)	0.67 (0.41; 0.14–1.8)	1.9 (1.1; 0.47–5.2)	1.7 (1.1; 0.15–4.3)
Endosulfan I	3.3 (2.7; 0.047–9.3)	4.6 (3.1; 0.076–12)	4.8 (3.0; 0.10–12)	4.0 (3.0; 0.044–15)	4.2 (3.4; 0.10–17)	5.7 (3.9; 0.21–14)	4.0 (3.5; 0.17–16)
Pallas							
α-HCH				24 (4.9; 16–31)	28 (6.9; 19–40)	18 (8.7; 3.0–35)	15 (7.2; 7.0–27)
γ-HCH				11 (6.7; 4.0–24)	11 (4.7; 5.0–19)	10 (7.3; 3.0–27)	9.7 (9.1; 4.0–36)
α/γ-HCH				3.1 (1.7; 0.95–5.8)	2.7 (0.92; 1.5–4.2)	2.2 (1.2; 1.0–4.3)	1.9 (0.76; 0.75–3.4)
p,p′-DDT				0.25 (0.18; 0.050–0.56)	0.22 (0.083; 0.14–0.44)	0.36 (0.17; 0.12–0.70)	0.23 (0.10; 0.12–0.50)
p,p′-DDE				0.57 (0.44; 0.16–1.4)	0.68 (0.74; 0.060–2.8)	0.47 (0.31; 0.090–0.98)	0.84 (0.59; 0.090–2.3)
p,p′-DDD				0.42 (0.47; 0.050–1.5)	0.21 (0.026; 0.20–0.26)	0.030 (0.024; 0.0050–0.070)	3.3 (1.7; 1.5–7.1)
p,p′-DDT/*p,p′*-DDE				0.47 (0.29; 0.13–1.0)	0.76 (0.90; 0.16–3.3)	1.5 (2.1; 0.34–7.8)	0.42 (0.38; 0.11–1.6)
Storhofdi							
HCB			7.4 (4.3; 1.7–16)	6.3 (1.7; 4.0–11)	10 (6.6; 2.1–27)	14 (16; 2.1–65)	6.0 (3.3; 2.8–17)
α-HCH			17 (11; n.d.–46)	16 (8.0; 7.7–39)	16 (9.1; 4.6–47)	15 (12; 3.7–58)	9.8 (4.2; 5.3–21)
γ-HCH			14 (11; n.d.–50)	9.6 (6.0; 3.6–32)	6.5 (4.0; n.d.–15)	5.2 (3.0; 0.33–1.0)	4.0 (2.7; n.d.–11)
α/γ-HCH			1.5 (0.84; 0.36–3.3)	2.0 (1.1; 0.33–5.7)	3.5 (3.0; 0.40–14)	2.8 (1.9; 0.69–8.3)	3.2 (3.3; 0.69–15)
TC			2.4 (2.8; n.d.–9.6)	0.95 (0.49; n.d.–1.6)	–	0.14 (0.014; 0.13–0.19)	0.28 (0.29; n.d.–0.72)
CC			0.98 (0.70; n.d.–2.0)	0.38 (0.14; n.d.–0.60)	0.53 (0.50; n.d.–1.4)	0.55 (0.35; n.d.–1.2)	0.88 (0.29; 0.41–1.5)
TC/CC			1.0 (0.95; 0.44–3.9)	2.9 (0.99; 2.0–6.5)	–	0.46 (0.42; 0.11–1.1)	0.52 (0.10; 0.35–0.75)
TN			–	0.59 (0.23; n.d.–0.90)	–	0.29 (0.14; 0.15–0.56)	0.53 (0.20; 0.14–0.82)
p,p′-DDT			0.15 (0.49; n.d.–1.8)	12 (34; n.d.–130)	–	1.8 (3.7; n.d.–0.70)	0.031 (0.14; n.d.–0.70)
o,p′-DDT			–	0.58 (0.65; n.d.–28)	–	0.20 (0.29; 0.070–1.2)	0.041 (0.11; n.d.–0.50)
p,p′-DDE			0.063 (0.21; n.d.–0.80)	0.63 (0.75; n.d.–3.1)	–	0.31 (0.30; 0.14–1.2)	0.090 (0.10; n.d.–0.36)
o,p′-DDE			–	0.25 (0.47; n.d.–1.3)	–	n.d.	–
p,p′-DDD			1.3 (2.2; n.d.–6.9)	0.52 (1.3; n.d.–6.4)	–	0.66 (1.1; 0.24–4.5)	n.d.
p,p′-DDT/*p,p′*-DDE			–	13 (16; 1.3–49)	–	4.1 (6.7; 0.20–28)	–
Diel			0.24 (1.2; n.d.–5.8)	0.75 (0.24; n.d.–2.6)	0.91 (0.66; n.d.–2.6)	1.1 (0.37; 0.47–1.8)	1.3 (0.46; 0.67–2.6)
Zeppelin							
HCB	92 (47; 23–310)	120 (69; 76–540)	99 (21; 41–210)	93 (29; 42–200)	110 (130; 0.18–760)	83 (17; 27–120)	88 (16; 62–160)
α-HCH	77 (34; 6.8–200)	61 (20; 16–110)	63 (18; 29–100)	60 (30; 33–190)	52 (17; 0.13–99)	82 (17; 27–120)	31 (7.4; 14–53)

γ-HCH	14 (7.6; 3.3–38)	16 (10; 5.3–62)	13 (6.2; 6.0–41)	13 (6.5; 4.9–39)	15 (19; 2.1–100)	9.9 (4.6; 2.2–2.3)	11 (5.0; 2.2–27)
α/γ-HCH	5.8 (2.0; 1.9–12)	4.5 (1.6; 1.2–7.2)	5.2 (1.6; 2.4–8.7)	5.1 (1.7; 2.2–9.4)	5.1 (2.1; 0.060–9.7)	11 (7.1; 1.2–36)	3.4 (1.7; 0.82–9.3)
TC	0.53 (0.27; 0.16–1.4)	0.70 (0.57; 0.21–3.0)	0.42 (0.24; 0.060–1.3)	0.60 (0.40; 0.18–2.0)	0.33 (0.21; 0.070–1.1)	0.35 (0.23; 0.029–0.92)	0.39 (0.21; 0.13–1.0)
CC	1.1 (0.37; 0.42–2.0)	1.2 (0.41; 0.55–2.4)	0.96 (0.30; 0.43–1.8)	1.2 (0.50; 0.010–2.6)	0.79 (0.45; 0.20–2.3)	0.76 (0.37; 0.088–1.9)	0.82 (0.20; 0.38–1.3)
TC/CC	0.49 (0.20; 0.20–0.89)	0.55 (0.25; 0.22–1.3)	0.44 (0.20; 0.061–0.78)	0.50 (0.22; 0.19–1.2)	0.44 (0.20; 0.16–0.86)	0.47 (0.22; 0.088–0.80)	0.47 (0.20; 0.18–0.94)
TN	0.85 (0.29; 0.35–1.6)	0.91 (0.34; 0.43–1.9)	0.67 (0.23; 0.29–1.3)	0.95 (0.41; 0.26–2.0)	0.53 (0.27; 0.10–1.8)	0.55 (0.29; 0.065–1.5)	0.65 (0.15; 0.30–0.95)
CN	0.18 (0.092; 0.031–0.39)	0.15 (0.085; 0.030–0.38)	0.14 (0.087; 0.040–0.55)	0.16 (0.10; 0.010–0.45)	0.11 (0.12; 0.024–0.68)	0.083 (0.062; 0.010–0.22)	0.079 (0.065; 0.011–0.40)
p,p′-DDT	–	0.87 (0.60; 0.25–3.0)	0.32 (0.25; 0.050–1.2)	0.38 (0.23; 0.11–1.4)	0.31 (0.43; 0.010–2.7)	0.54 (0.39; 0.13–1.6)	0.54 (0.56; 0.026–2.2)
o,p′-DDT	–	1.0 (0.75; 0.29–3.7)	0.50 (0.44; 0.070–1.9)	0.48 (0.28; 0.14–1.4)	0.40 (0.28; 0.070–1.3)	0.52 (0.28; 0.010–1.3)	0.43 (0.29; 0.13–1.6)
p,p′-DDE	–	1.9 (1.5; 0.41–6.5)	0.87 (0.80; 0.088–3.0)	1.5 (2.1; 0.15–11)	1.2 (1.6; 0.080–9.0)	1.0 (0.53; 0.21–2.0)	0.94 (1.4; 0.16–9.9)
o,p′-DDE	–	1.4 (1.0; 0.35–3.9)	0.22 (0.23; 0.020–0.81)	0.16 (0.15; 0.010–0.54)	0.16 (0.11; 0.020–0.45)	0.19 (0.11; 0.030–0.51)	0.11 (0.079; 0.020–0.30)
p,p′-DDD	–	0.090 (0.060; 0.030–0.39)	0.048 (0.030; 0.010–0.11)	0.11 (0.093; 0.020–0.51)	0.049 (0.067; 0.010–0.41)	0.064 (0.084; 0.020–0.49)	0.094 (0.14; 0.010–0.70)
o,p′-DDD	–	0.074 (0.054; 0.020 - 0.29)	0.048 (0.031; 0.010–0.15)	0.088 (0.059; 0.030–0.31)	0.055 (0.086; 0.010–0.52)	0.069 (0.090; 0.010–0.54)	0.063 (0.081; 0.010–0.48)
p,p′-DDT/*p,p′*-DDE	–	0.53 (0.17; 0.20–0.94)	0.52 (0.38; 0.13–2.1)	0.55 (0.44; 0.015–1.8)	0.43 (0.39; 0.020–1.8)	0.59 (0.36; 0.15–1.6)	0.99 (1.2; 0.0026–5.1)

(b) 2000–2006

	2000	2001	2002	2003	2004	2005	2006
Alert							
HCB	57 (19; 20–99)	69 (25; 24–130)	29 (20; 0.060–63)	49 (16; 15–78)	29 (21; 0.060–70)	52 (21; 0.060–120)	
α-HCH	25 (11; 3.7–45)	20 (13; 1.4–66)	11 (7.7; 0.018–26)	16 (8.1; 0.018–46)	11 (6.1; 0.11–27)	13 (5.7; 5.2–33)	
γ-HCH	4.4 (2.3; 0.92–10)	6.8 (3.9; 0.43–19)	2.5 (1.2; 0.11–5.5)	2.8 (1.6; 0.015–8.1)	1.4 (1.1; 0.014–4.9)	1.7 (0.68; 0.67–3.7)	
α/γ-HCH	6.4 (2.3; 1.4–11)	3.4 (2.0; 0.56–11)	4.5 (2.9; 0.23–10)	6.1 (1.8; 2.9–14)	8.0 (4.1; 1.4–22)	7.8 (2.2; 3.8–12)	
TC	0.19 (0.10; 0.047–0.43)	0.17 (0.085; 0.027–0.39)	0.13 (0.10; 0.025–0.43)	0.20 (0.22; 0.029–1.1)	0.29 (0.25; 0.031–1.2)	0.25 (0.17; 0.028–0.86)	
CC	0.51 (0.22; 0.089–0.97)	0.44 (0.21; 0.0094–0.94)	0.55 (0.23; 0.030–1.4)	0.47 (0.31; 0.028–1.2)	1.4 (2.6; 0.029–14)	0.64 (0.21; 0.35–1.3)	
TC/CC	0.38 (0.15; 0.14–0.64)	0.41 (0.17; 0.066–0.69)	0.38 (0.21; 0.10–0.80)	0.52 (0.22; 0.25–1.1)	0.37 (0.29; 0.049–1.6)	0.42 (0.21; 0.12–0.89)	
TN	0.40 (0.20; 0.044–0.86)	0.32 (0.16; 0.11–0.74)	0.43 (0.27; 0.016–1.0)	0.43 (0.16; 0.15–0.84)	0.36 (0.37; 0.020–2.5)	0.35 (0.13; 0.074–0.91)	
OXY	0.34 (0.17; 0.027–0.69)	0.28 (0.12; 0.066–0.54)	0.36 (0.21; 0.042–0.73)	0.44 (0.34; 0.038–2.1)	0.50 (0.37; 0.041–1.5)	0.52 (0.25; 0.043–1.4)	
HEPX	0.88 (1.8; 0.058–13)	0.51 (0.24; 0.12–1.2)	0.58 (0.24; 0.016–1.2)	0.66 (0.49; 0.016–3.0)	0.65 (0.30; 0.050–1.4)	0.84 (0.29; 0.40–1.9)	
p,p′-DDT	0.14 (0.16; 0.030–1.1)	0.044 (0.028; 0.015–0.14)	0.17 (0.092; 0.10–0.53)	0.14 (0.042; 0.11–0.34)	0.35 (0.27; 0.12–1.4)	0.14 (0.050; 0.10–0.41)	
o,p′-DDT	0.16 (0.14; 0.031–0.56)	0.13 (0.085; 0.019–0.41)	0.18 (0.13; 0.066–0.63)	0.085 (0.035; 0.063–0.30)	0.16 (0.10; 0.069–0.51)	0.22 (0.17; 0.071–0.86)	
p,p′-DDE	0.51 (0.54; 0.12–3.6)	0.31 (0.20; 0.051–0.91)	0.33 (0.28; 0.11–1.4)	0.34 (0.26; 0.11–1.2)	0.44 (0.33; 0.12–1.8)	0.30 (0.26; 0.10–1.2)	
o,p′-DDE	0.11 (0.074; 0.027 –0.36)	0.071 (0.046; 0.013–0.22)	0.050 (0.045; 0.025–0.24)	0.071 (0.067; 0.027–0.93)	0.18 (0.23; 0.027–0.93)	0.034 (0.021; 0.022–0.17)	
p,p′-DDD	0.035 (0.046; 0.022–0.32)	0.012 (0.0025; 0.010–0.023)	0.24 (0.058; 0.17–0.43)	0.23 (0.038; 0.19–0.40)	0.33 (0.21; 0.21–0.97)	0.23 (0.033; 0.17–0.35)	
o,p′-DDD	0.053 (0.074; 0.028–0.41)	0.021 (0.024; 0.013–0.18)	0.078 (0.053; 0.044–0.36)	0.059 (0.0066; 0.049–0.091)	0.10 (0.060; 0.053–0.27)	0.067 (0.030; 0.044–0.23)	
p,p′-DDT/*p,p′*-DDE	0.39 (0.34; 0.10–2.1)	0.23 (0.16; 0.056–0.83)	0.85 (0.68; 0.23–1.8)	0.67 (0.17; 0.47–0.78)	1.8 (1.5; 0.34–5.7)	0.99 (0.57; 0.26–1.6)	
PCA	3.5 (2.1; 0.49–11)	6.0 (5.2; 0.77–20)	12 (14; 0.013–55)	1.1 (0.68; 0.17–3.0)	8.5 (16; 0.11–62)	1.4 (5.0; 0.011–24)	
Diel	0.70 (0.35; 0.15–1.8)	0.49 (0.28; 0.085–1.5)	0.72 (0.40; 0.033–2.3)	0.86 (0.58; 0.033–3.4)	0.60 (0.33; 0.10–1.6)	0.81 (0.31; 0.28–1.6)	
OCS	0.43 (0.15; 0.18–1.0)	0.31 (0.13; 0.0076–0.73)	0.41 (0.13; 0.0076–0.57)	0.47 (0.15; 0.076–0.81)	0.33 (0.19; 0.0078–0.67)	0.58 (0.22; 0.0076–1.1)	
TeCV	1.1 (1.1; 0.10–5.5)	1.3 (1.5; 0.053–6.8)	1.4 (0.84; 0.050–4.3)	1.9 (1.2; 0.35–5.7)	1.6 (0.77; 0.052–3.1)	2.0 (1.1; 0.39–4.4)	
Endosulfan I	3.5 (2.6; 0.12–12)	5.1 (4.9; 0.31–25)	3.9 (2.9; 0.017–12)	6.5 (4.9; 0.021–18)	3.2 (2.7; 0.065–10)	5.6 (2.9; 1.4–15)	
Pallas							
α-HCH	17 (6.8; 9.0–28)	–	18 (15; 2.0–51)	9.9 (3.9; 5.0–15)	9.4 (4.1; 4.0–15)	9.9 (3.4; 6.0–15)	
γ-HCH	10 (8.1; 2.0–29)	–	7.3 (7.3; 1.0–24)	4.2 (3.1; 2.0–13)	3.1 (1.6; 1.0–6.0)	2.8 (0.87; 2.0–4.0)	
α/γ-HCH	2.4 (1.1; 0.69–4.5)	–	3.0 (1.8; 0.50–6.0)	2.8 (0.92; 1.2–4.0)	3.2 (0.70; 2.5–5.0)	3.6 (0.75; 2.3–5.0)	
p,p′-DDT	0.26 (0.14; 0.13–0.64)	–	0.078 (0.13; 0.0050–0.40)	0.22 (0.15; 0.11–0.66)	0.18 (0.11; 0.090–0.37)	0.25 (0.077; 0.13–0.37)	
p,p′-DDE	0.93 (0.76; 0.27–2.6)	–	0.55 (0.30; 0.11–1.2)	0.47 (0.25; 0.15–0.78)	0.52 (0.25; 0.17–1.0)	0.58 (0.35; 0.25–1.5)	
p,p′-DDD	0.017 (0.031; 0.0050–0.11)	–	0.010 (0.019; 0.0050–0.070)	0.17 (0.20; 0.060–0.76)	0.078 (0.055; 0.0050–0.19)	0.11 (0.083; 0.025–0.27)	
p,p′-DDT/*p,p′*-DDE	0.37 (0.21; 0.15-0.70)	–	0.22 (0.34; 0.0043–0.84)	0.58 (0.34; 0.18–1.0)	0.39 (0.22; 0.15–0.82)	0.54 (0.28; 0.16–1.0)	
Storhofdi							
HCB	5.8 (1.6; 3.6–11)	6.8 (3.1; 3.0–16)	3.7 (1.1; 1.7–6.6)	4.3 (1.0; 1.3–5.8)	2.8 (1.0; 1.1–5.6)	2.0 (0.69; 1.2–4.1)	
α-HCH	7.8 (1.8; 5.0–14)	10 (3.7; 4.8–20)	5.5 (1.7; 3.4–10)	7.4 (1.7; 3.9–12)	5.0 (1.5; 2.3–7.4)	2.7 (1.1; 0.99–5.9)	
γ-HCH	3.8 (1.5; 1.8–7.2)	6.3 (4.2; 2.4–19)	3.5 (1.2; 1.6–5.9)	7.5 (1.7; 3.1–10)	7.5 (1.5; 4.8–10)	3.8 (1.6; 1.2–7.3)	
α/γ-HCH	2.3 (0.89; 1.0–4.4)	1.9 (0.92; 0.63–4.7)	1.8 (0.84; 0.81–3.6)	1.0 (0.42; 0.53–2.4)	0.69 (0.25; 0.26–1.2)	0.73 (0.15; 0.48–1.1)	
TC	0.52 (0.12; 0.31–0.83)	0.42 (0.11; 0.22–0.59)	0.36 (0.10; 0.24–0.61)	0.34 (0.099; 0.18–0.62)	0.19 (0.066; 0.085–0.34)	0.082 (0.035; 0.060–0.23)	

(continued on next page)

Table 3 (*continued*)

(b) 2000–2006

	2000	2001	2002	2003	2004	2005	2006
CC	1.0 (0.21; 0.72–1.6)	0.98 (0.25; 0.62–1.6)	0.59 (0.23; 0.22–0.94)	0.67 (0.19; 0.41–1.0)	0.56 (0.15; 0.39–1.0)	0.11 (0.12; 0.060–0.56)	
TC/CC	0.52 (0.13; 0.34–0.73)	0.45 (0.13; 0.25–0.68)	0.68 (0.23; 0.36–1.1)	0.53 (0.17; 0.23–0.81)	0.35 (0.11; 0.16–0.56)	0.53 (0.31; 0.27–1.1)	
TN	0.58 (0.13; 0.38–0.83)	0.53 (0.16; 0.31–0.87)	0.34 (0.14; 0.16–0.75)	0.30 (0.088; 0.075–0.44)	0.28 (0.099; 0.095–0.49)	0.079 (0.037; 0.060–0.25)	
p,p′-DDT	0.13 (0.066; 0.075–0.42)	0.27 (0.43; 0.045–1.7)	0.29 (0.21; 0.045–0.59)	0.46 (0.49; 0.22–2.5)	0.24 (0.31; 0.14–1.7)	0.14 (0.015; 0.13–0.19)	
o,p′-DDT	0.12 (0.074; 0.040–0.22)	0.14 (0.21; 0.040–1.1)	0.090 (0.047; 0.050–0.27)	0.17 (0.21; 0.050–0.93)	0.13 (0.049; 0.075–0.22)	0.11 (0.039; 0.060–0.19)	
p,p′–DDE	0.23 (0.13; 0.090–0.65)	0.21 (0.19; 0.050–0.95)	0.19 (0.056; 0.11–0.31)	0.36 (0.16; 0.17–0.89)	0.18 (0.039; 0.14–0.34)	0.16 (0.075; 0.070–0.34)	
o,p′-DDE	–	–	–	–	–	–	
p,p′-DDD	0.10 (0.011; 0.075–0.12)	0.13 (0.034; 0.085–0.20)	0.16 (0.019; 0.085–0.18)	0.16 (0.057; 0.10–0.40)	0.18 (0.072; 0.14–0.52)	0.14 (0.015; 0.13–0.19)	
p,p′-DDT/*p,p′*-DDE	0.86 (0.83; 0.15–4.0)	1.4 (1.6; 0.16–6.7)	1.5 (0.89; 0.21–2.5)	1.3 (0.83; 0.48–4.1)	1.4 (1.8; 0.50–9.8)	1.1 (0.55; 0.38–2.1)	
Diel	1.1 (0.23; 0.66–1.5)	1.2 (0.51; 0.21–2.1)	0.94 (0.46; 0.27–2.2)	0.82 (0.25; 0.52–1.5)	0.57 (0.18; 0.22–0.96)	0.10 (0.092; 0.060–0.51)	
Zeppelin							
HCB	56 (10; 2.8–73)	55 (9.4; 36–87)	57 (34; 33–270)	54 (11; 14–75)	65 (6.2; 54–80)	67 (7.4; 48–83)	72 (18; 48–150)
α-HCH	21 (6.0; 0.28–35)	21 (7.4; 11–50)	27 (8.1; 16–61)	19 (5.0; 6.1–28)	17 (6.7; 9.2–32)	15 (3.8; 7.7–21)	11 (3.3; 6.6–19)
γ-HCH	5.8 (2.2; 0.43–11)	5.7 (2.6; 2.3–1.7)	7.9 (4.4; 3.4–23)	4.2 (1.4; 2.0–7.7)	2.8 (0.81; 1.3–6.4)	2.4 (0.75; 1.1–4.3)	1.9 (0.65; 1.1–4.0)
α/γ-HCH	3.8 (1.5; 0.66–7.7)	4.1 (1.2; 2.0–7.1)	4.0 (1.9; 1.3–9.8)	4.8 (1.5; 2.3–9.0)	6.6 (2.6; 2.6–14)	6.7 (2.0; 3.4–12)	6.2 (2.0; 2.5–11)
TC	0.26 (0.14; 0.053–0.62)	0.33 (0.18; 0.080–0.93)	0.38 (0.25; 0.090–1.0)	0.27 (0.14; 0.090–0.81)	0.25 (0.14; 0.070–0.62)	0.25 (0.14; 0.080–0.55)	0.22 (0.13; 0.050–0.72)
CC	0.65 (0.15; 0.22–0.98)	0.83 (0.28; 0.36–2.0)	0.83 (0.20; 0.47–1.3)	0.68 (0.21; 0.29–1.4)	0.66 (0.15; 0.40–1.1)	0.62 (0.12; 0.39–0.90)	0.61 (0.15; 0.37–1.1)
TC/CC	0.41 (0.20; 0.14–0.84)	0.41 (0.20; 0.11–0.95)	0.44 (0.23; 0.15–0.90)	0.39 (0.17; 0.14–0.66)	0.37 (0.17; 0.11–0.67)	0.40 (0.18; 0.13–0.71)	0.35 (0.16; 0.089–0.67)
TN	0.50 (0.14; 0.12–0.86)	0.70 (0.28; 0.26–1.6)	0.67 (0.18; 0.37–1.2)	0.67 (0.26; 0.21–1.6)	0.56 (0.14; 0.34–0.97)	0.55 (0.12; 0.34–0.88)	0.60 (0.38; 0.28–2.1)
CN	0.064 (0.032; 0.010–0.13)	0.082 (0.064; 0.010–0.36)	0.081 (0.040; 0.020–0.18)	0.075 (0.040; 0.010–0.15)	0.073 (0.030; 0.030–0.14)	0.056 (0.027; 0.020–0.12)	0.094 (0.094; 0.010–0.47)
p,p′-DDT	0.18 (0.19; 0.050–1.2)	0.13 (0.12; 0.010–0.90)	0.14 (0.14; 0.030–0.85)	0.15 (0.11; 0.012–0.45)	0.10 (0.074; 0.012–0.37)	0.090 (0.074; 0.017–0.40)	0.11 (0.10; 0.023–0.44)
o,p′-DDT	0.28 (0.25; 0.050–1.5)	0.25 (0.24; 0.010–1.6)	0.26 (0.19; 0.050–0.85)	0.23 (0.14; 0.049–0.70)	0.23 (0.16; 0.055–0.72)	0.19 (0.14; 0.047–0.64)	0.21 (0.23; 0.020–1.2)
p,p′-DDE	0.50 (0.46; 0.080–2.6)	0.81 (1.0; 0.10–5.6)	0.94 (2.2; 0.060–16)	0.90 (0.81; 0.12–3.3)	0.58 (0.55; 0.078–2.4)	0.59 (0.65; 0.049–3.0)	1.2 (1.3; 0.13–6.8)
o,p′-DDE	0.10 (0.074; 0.020–0.35)	0.12 (0.10; 0.010–0.34)	0.11 (0.10; 0.010–0.40)	0.12 (0.092; 0.016–0.37)	0.10 (0.085; 0.011–0.34)	0.086 (0.078; 0.013–0.33)	0.085 (0.074; 0.010–0.32)
p,p′-DDD	0.066 (0.055; 0.020–0.25)	0.041 (0.050; 0.010–0.20)	0.035 (0.060; n.d.–0.35)	0.073 (0.086; 0.010–0.40)	0.025 (0.032; 0.010–0.17)	0.029 (0.023; 0.010–0.15)	0.10 (0.17; 0.010–0.85)
o,p′-DDD	0.049 (0.042; 0.010–0.20)	0.032 (0.037; 0.080–0.14)	0.019 (0.019; 0.0050–0.10)	0.026 (0.017; 0.010–0.080)	0.019 (0.012; 0.010–0.072)	0.021 (0.012; 0.010–0.060)	0.042 (0.070; 0.010–0.34)
p,p′-DDT/*p,p′*–DDE	0.52 (0.33; 0.12–1.4)	0.34 (0.32; 0.032–1.0)	0.30 (0.17; 0.054–0.73)	0.19 (0.077; 0.060–0.38)	0.23 (0.12; 0.092–0.68)	0.21 (0.087; 0.079–0.47)	0.15 (0.13; 0.012–0.76)

Table 4
Arithmetic mean (range) of total atmospheric PCBs and OCPs concentrations (pg/m^3) at Satellite Stations (GFF+PUF).

	Kinngait I (1994/03/15–1996/02/25)		Kinngait II (2000/10/16–2002/09/06)	
	1994	1995	2001	2002[e]
HCB[a]	55 (16–120)	64 (28–120)	58 (7.3–110)	47 (4.4–86)
α-HCH	78 (19–190)	71 (32–110)	25 (2.7–66)	27 (8.2–55)
γ-HCH	14 (2.3–39)	9.4 (3.1–27)	4.7 (2.2–18)	3.1 (1.2–7.4)
Σchlordane[b]	3.6 (1.3–7.7)	2.3 (1.0–3.6)	1.2 (0.49–2.4)	0.83 (0.25–1.9)
Dieldrin	1.6 (0.60–4.0)	1.5 (0.53–2.2)	0.46 (0.13–1.1)	0.39 (0.030–0.93)
ΣDDT[c]	1.6 (0.64–3.3)	1.4 (0.62–2.8)	1.2 (0.57–2.8)	0.63 (0.15–1.8)
ΣPCB[d]	5.6 (2.9–8.8)	5.6 (1.9–15)	5.9 (2.4–19)	3.9 (0.82–17)
Endosulfan I	5.8 (2.1–13)	5.6 (1.9–12)	2.5 (0.25–7.1)	3.3 (0.28–2.9)

	Tagish		Little Fox Lake	Barrow
	1993	1994	2002/07/07–2003/07/06	2002/03/19–2003/04/23
HCB[a]	42 (10–140)	55 (0.34–900)	67 (13–120)	47 (13–130)
α-HCH	79 (24–160)	78 (0.25–760)	48 (17–87)	19 (6.0–37)
γ-HCH	11(3.3–25)	13 (0.18–140)	4.5 (1.7–10)	2.7 (0.89–5.8)
Σchlordane[b]	2.9 (1.1–6.6)	3.2 (0.26–31)	1.0 (0.51–1.5)	0.99 (0.29–2.6)
Dieldrin	0.93 (0.37–2.2)	1.3 (0.040–21)	0.31 (0.11–0.60)	1.0 (0.10–3.7)
ΣDDT[c]	1.4 (0.26–3.2)	2.0 (0.43–26)	0.93 (0.28–1.6)	1.2 (0.48–4.4)
ΣPCB[d]	3.7 (0.92–8.3)	4.5 (0.86–47)	4.1 (0.75–11)	17 (8.6–47)
Endosulfan I	5.8 (0.080–15)	8.3 (0.08–89)	8.3 (3.1–18)	2.8 (0.67–5.9)

	Amderma[f]		Dunai[f]		Valkarkai[f]
	1999/03/27–2000/03/28	2000/03/27–2001/04/02	1993/03/09–1994/03/14	1994/03/14–1995/04/31	2002/07/29–2002/09/30
HCB[a]	41 (15–97)	35 (12–74)	57 (19–170)	64 (31–100)	76 (27–100)
α-HCH	24 (5.3–78)	20 (2.3–40)	45 (0.63–100)	55 (4.4–140)	64 (60–75)
γ-HCH	8.1 (0.79–24)	7.2 (1.0–24)	10 (0.020–23)	10 (0.17–36)	7.1 (4.4–15)
Σchlordane[b]	1.0 (0.31–1.8)	0.83 (0.13–1.6)	2.3 (0.11–6.0)	2.1 (0.28–8.6)	1.8 (1.4–2.8)
Dieldrin	0.69 (0.060–1.4)	0.65 (0.020–1.9)	1.1 (0.020–2.0)	1.3 (0.20–5.3)	0.60 (0.26–0.89)
ΣDDT[c]	2.2 (0.83–5.9)	4.7 (0.45–53)	0.98 (0.24–2.0)	1.3 (0.45–5.3)	23 (5.5–77)
ΣPCB[d]	7.8 (2.9–110)	4.7 (1.4–10)	8.1 (0.54–23)	5.9 (2.1–18)	58 (31–120)
Endosulfan I	3.7 (0.04–14)	3.5 (0.05–12)	3.1 (0.05–7.9)	5.4 (0.06–38)	3.2 (1.6–4.7)

	Nuuk[g]
	2004–2005
α-HCH	20 (10–66)[h]
γ-HCH	5.1 (1.9–11)
c-chlordane	0.46 (0.11–0.87)
t-chlordane	0.40 (0.27–0.72)
Dieldrin	1.9 (N.D.–4.3)
p,p′–DDE	0.41 (N.D.–0.94)
o,p′-DDE	0.56 (N.D.–5.0)
Endosulfan I	4.8 (N.D.–10)

[a] HCB broke through in ~30% of all samples taken, mean concentrations may be underestimated.

[b] Σchlordane = sum of *c*-chlordane, *t*-chlordane, *c*-nonachlor and *t*-nonachlor.

[c] ΣDDT = sum of *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT and *p,p′*-DDT.

[d] ΣPCB = AMAP sum of 10 PCBs (PCB 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180).

[e] Only samples from 2001/1/15 to 2002/09/06 collected were included in deriving means and ranges.

[f] For the Russian stations of Amderma and Dunai, annual sampling cycles started and ended in March. Annual values were therefore derived from one full year of sampling from the start date. For Valkarkai, values were derived from the 5 PUF samples taken during the summer of 2002 due to relatively high GFF field blanks.

[g] Data adopted from Bossi et al. (2008). N.D. = not detected (≤ method detection limits).

[h] Samples collected in July–September 2004 and 2005 not included due to breakthrough problems.

ΣPCB air concentrations at the 7 satellite stations (Table 4) were similar to those at the 4 long-term stations (Table 2), except for Barrow and Valkarkai. Barrow, Alaska, showed slightly higher average air concentrations in 2002/2003 of 17 pg/m^3 comparing to 1–8 pg/m^3 measured at the long-term sites. Air concentrations of ΣPCB at Valkarkai, Russia, were much higher than those found at the other stations with an average value of 58 pg/m^3. Note that measurements at this location only represent the gas phase concentrations during the summer of 2002 since high blank values were observed on the filters (Su et al., 2008). The relatively high concentration could be the result of greater volatilization from surfaces during the summer time. While most countries have stopped production of PCB technical mixtures since the 1970s or 1980s, production in Russia continued up to 1993 (Breivik et al., 2002). The elevated air concentrations measured at this location may thus also be a reflection of more recent use in the region. Penta- and tetra-chlorinated congeners dominated the homologue profiles of the 5 samples taken at Valkarkai with average percentages of 42 and 25%, respectively, of the sum of total PCBs. This is consistent with the historical production and use of Sovol and Sovtol (90% Sovol + 10% trichlorobenzene), which resemble Aroclor 1254, in the former Soviet Union. Sovol contains 1.0%, 23%, 53% and 22% of tri-, tetra-, penta- and hexa-PCBs, respectively (Breivik et al., 2002).

3.1.2. Organochlorine pesticides (OCPs)

3.1.2.1. Hexachlorobenzene (HCB). HCB was previously used as a fungicide and is formed as a by-product during production of a large variety of chemicals, including solvents (Barber et al., 2005). As Su et al. (2006) have noted, the air concentrations of HCB were quite uniform in 2000–2003 at all stations and in all seasons with a nearly normal distribution at Alert, Kinngait, Barrow and Zeppelin. From Tables 3 and 4, air concentrations of HCB measured at Zeppelin were highest during the 1990s with average air concentrations ranging from 83 to 120 pg/m^3. After 2000, all stations showed similar air concentrations ranging from an average of 29 to 76 pg/m^3, except Storhofdi which showed the lowest air concentrations with averages ranging from 2 to 6.8 pg/m^3. However, an examination of the temporal trend shows increasing tendencies at Alert and Zeppelin in recent years. This will be further discussed in Section 3.2.

3.1.2.2. α-Hexachlorocyclohexane (α-HCH). α-HCH is a major constituent (~60–70%) of technical HCH (UNEP 2008). Most countries have banned the use of the technical mixture since the 1980s. Major users such as China have stopped using the technical formula since 1983 and India and the former Soviet Union have limited its use in agriculture in 1990. Its air concentrations were similar at all Arctic stations at any one time. Air concentrations measured in the early 1990s at Kinngait in the eastern Canadian Arctic were similar to those measured in the western Canadian Arctic at Tagish, and at Zeppelin, with average values of about 60 to 80 pg/m^3. Slightly lower air concentrations were measured at the Canadian High Arctic station of Alert and the Russian station of Dunai during this time with average values of about 54 to 57 and 45 to 55 pg/m^3, respectively. After 2000, the air concentrations have declined to about 20–30 pg/m^3 and below at all stations, except at Little Fox Lake and Valkarkai. Measurements at Storhofdi showed the lowest α-HCH air concentrations with average values of 2.7 to 17 pg/m^3. Su et al. (2006) have attributed the relatively higher air concentrations measured at Little Fox Lake to secondary emission of previously deposited technical HCH from soil; as a result of higher precipitation rate, which may enhance scavenging to surfaces when technical HCH was still in use, and higher air temperature, which facilitates evaporation, at this location compared to other Arctic sites. Samples collected at Valkarkai (July–September) only represent higher air concentrations during the summer period as summer maxima were observed at other stations as well (Su et al., 2006).

3.1.2.3. γ-Hexachlorocyclohexane (γ-HCH). Generally, air concentrations of γ-HCH were quite similar at all sites (~4–16 pg/m^3 in the 1990s and 1.4–10 pg/m^3 in the 2000s) but the seasonality varies more significantly than α-HCH. Air concentrations of γ-HCH in the Arctic are log-normally distributed, reflecting the use of lindane (>99% γ-HCH) which is a current-use pesticide. Bimodal seasonal cycles, referred to as “Spring Maximum Event” (SME), were observed at Alert, Zeppelin, Kinngait and Barrow (Su et al., 2006; Becker et al., 2008). SME may be caused by pesticide application in the spring and tilling of soils in the fall with accelerated summertime degradation. Air concentrations of γ-HCH measured at Tagish and Little Fox Lake showed stronger correlations with temperature (T) than at other stations; with steeper, statistically significant slopes of the natural logarithm of partial pressure, ln P, vs. $1/T$, indicating the importance of secondary emission (Su et al., 2006). Slightly higher air concentrations in the spring and fall coinciding with application and tilling were also observed at these locations. Lower air concentrations observed in the Arctic in recent years may indicate the recent bans or phasing out of lindane in Europe and Canada. Further discussion will follow in Section 3.2.

A relative enrichment of γ-HCH to the total (α+γ) HCH air concentrations at Zeppelin as compared to Alert has been attributed to the proximity of Zeppelin to major use region of lindane in Europe (Becker et al., 2008). Pallas and Storhofdi in the European Arctic showed similar average concentrations of γ-HCH as Zeppelin with slightly higher concentrations measured at Pallas after 2000 (average ranged from 2.8 to 10 pg/m^3 from 2000 to 2005); but lower average concentrations of α-HCH [Pallas (1996–2005): 9.4–28 pg/m^3; Storhofdi (1995–2005): 2.7–17 pg/m^3]. This resulted in much lower average α- to γ-HCH ratios at Pallas and Storhofdi [Pallas (1996–2005): 1.9–3.6; Storhofdi (1995–2005): 0.69–3.5] as compared to those at Alert and Zeppelin [Alert (1993–2005): 3.4–10; Zeppelin (1993–2006): 3.4–11] (Table 3).

3.1.2.4. Chlordanes. The technical formula of chlordane is a mixture of cyclodiene compounds, used for insect and termite control, contains mainly *trans*-chlordane (TC), *cis*-chlordane (CC) and *trans*-nonachlor (TN) with trace amounts of heptachlor (HEPT), *cis*-nonachlor (CN) and other species (Su et al., 2008). Alert and all satellite stations reported seven chlordane-related compounds, namely TC, CC, TN, CN, HEPT, oxychlordane (OXY) and heptachlor epoxide (HEPX), in which the latter two are degradation products under environmental conditions. Chlordane-related compounds were not reported at Pallas. Zeppelin reports air concentrations of TC, CC, TN and CN; while Storhofdi reported concentrations of TC, CC and TN. Seasonality of chlordane-related compounds in Arctic air was marked by lower concentrations of parent compound TC but higher concentrations of degradation products OXY and HEPX in the summer. This cycle could be the result of higher temperature and sunlight during the Arctic summer enhancing transformation processes (Su et al., 2008). More stable compounds of CC and TN showed weak seasonal cycles. Slightly higher concentrations were found for TC at Valkarkai ranging from 0.20 to 1.0 pg/m^3 in the summer of 2002; while measurements at Alert, Storhofdi and Zeppelin showed average concentrations ranging from 0.082 to 0.42 from 2001 to 2005. TC/CC ratio correlates with (TC + CC) indicating influence of technical chlordane sources at Valkarkai (Su et al., 2008). TC, CC, TN, OXY and HEPX were normally distributed indicating the homogeneous distribution of chlordane-related compounds in Arctic air (Su et al., 2008). High concentrations of atmospheric HEPT, which are usually found to be below detection limits as it readily transforms to HEPX, measured at Barrow coincided with enhanced TC/CC ratios, implying influence of technical HEPT containing some TC and CC (Su et al., 2008). Otherwise, generally uniform spatial and seasonal distributions of chlordane-related compounds at other sites reflect diminishing influence from primary sources (Su et al., 2008).

3.1.2.5. Dichlorodiphenyltrichloroethane isomers (DDTs) and transformation products. Current use of DDTs is only limited to public health purposes such as for malaria control. Usage of dicofol, containing relatively high levels of *o,p*′-DDT, can also be a source of DDTs in the environment (Qiu et al., 2004, 2005). Alert, Zeppelin and all satellite stations reported 6 isomers of DDTs (namely *p,p*′- and *o,p*′- DDT, DDD and DDE). Pallas only reported 3 *p,p*′-substituted isomers. Storhofdi reported 3 *p,p*′-substituted isomers, *o,p*′-DDT and DDE. DDDs were usually found below detection limits at all stations. Generally, no apparent seasonal profiles can be observed for DDT isomers but winter concentrations of *p,p*′-DDE were usually higher, except at Barrow. It was believed that DDT-related compounds have greater tendencies to associate with particles which show greater input in the winter due to the influx of Arctic Haze. Higher precipitation rates in the summer may account for their low summertime concentration due to enhanced scavenging (Su et al., 2008).

DDT air concentrations found at the Russian stations of Amderma and Valkarkai were generally higher than at other Arctic sites. However, the *p,p*′-DDT/*p,p*′-DDE ratios were much lower than that of air equilibrated with the technical mixture at 0 °C, which is 2.38, with annual average values of 0.51–0.81 at Amderma and range of 5 samples of 0.78–1.1 at Valkarkai. Similarly, this ratio at other Arctic stations reported here were mostly lower than that of the technical mixture (Table 3). This implies that atmospheric DDTs in the Arctic were influenced by weathered sources. Extremely low ratios of *p,p*′-/*o,p*′-DDT (~0.14; Qiu et al., 2004, 2005) were seldom observed in Arctic air, indicating negligible impact of dicofol on Arctic air (Su et al., 2008). Pooled data from Alert, Little Fox Lake, Barrow, Kinngait and Zeppelin in 2000–2003 do not fit the normal distribution, indicating that DDT-related compounds were not well-mixed in the Arctic atmosphere (Su et al., 2008). Spatial variations in DDT usage in tropical areas for vector control may be responsible for this observation in Arctic air.

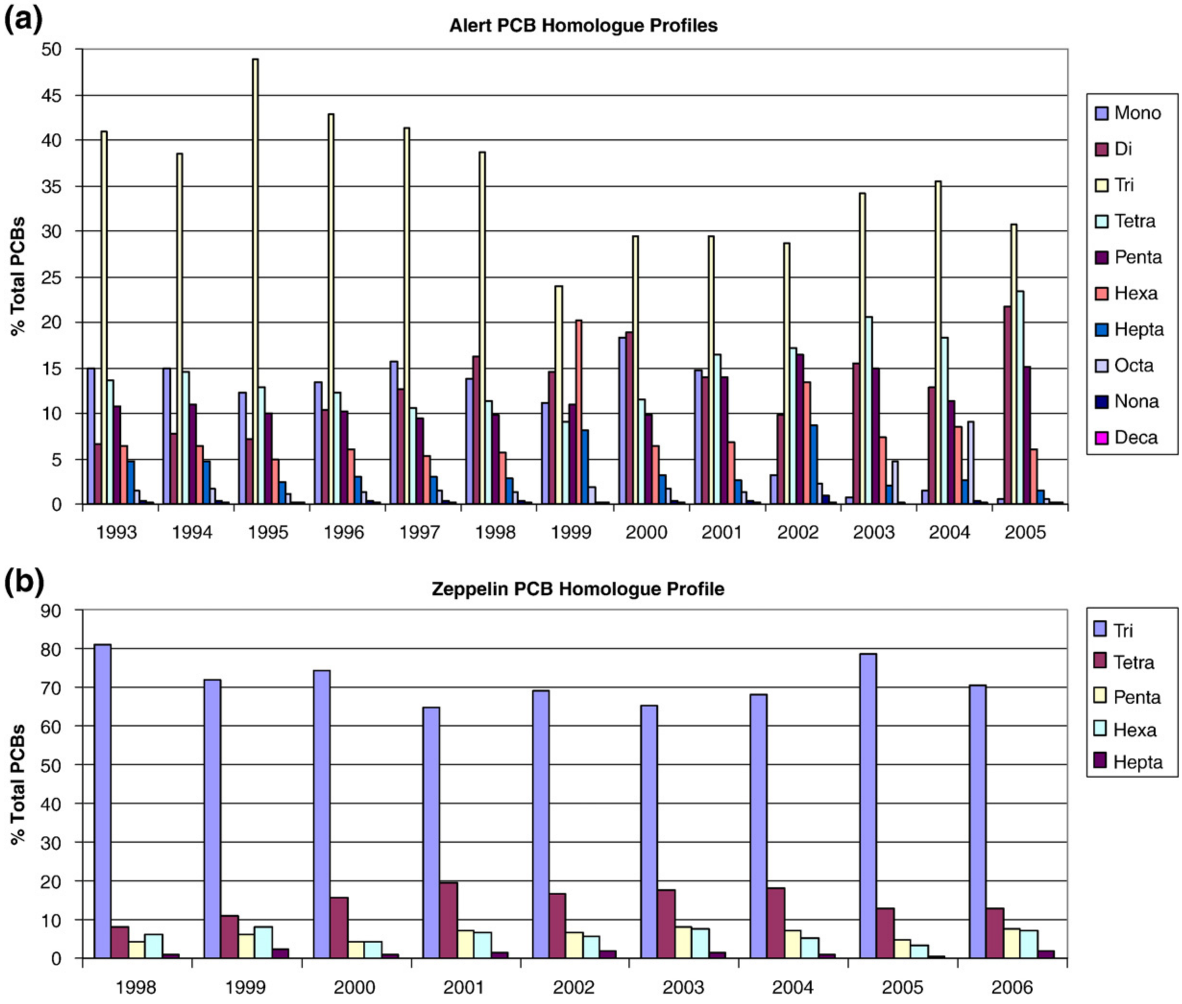

Fig. 2. PCB homologue profiles at (a) Alert and (b) Zeppelin.

3.1.2.6. Endosulfan I and II. Endosulfan is a current-use insecticide extensively applied on cotton, potatoes, tomatoes, and apples. Alert was the only long-term station that reported multi-year endosulfan I air concentrations (Table 3). Endosulfan I was reported at all satellite stations (Table 4). It was measured at Alert with average air concentrations ranging from 3.3 (in 1993) to 6.5 (in 2003) pg/m^3. Similar average concentrations were observed at the Greenland station of Nuuk (4.8 pg/m^3 in 2004–2005) and Russian Arctic stations of Amderma, Dunai and Valkarkai [ranging from 3.1 (in 1993–1994) to 5.4 (in 1994–1995) pg/m^3 at Dunai]. The highest averages were measured in the Yukon region at 8.3 pg/m^3 at Tagish in 1994 and, again, at Little Fox Lake in 2002–2003. As these two stations are located in the lower western North American Arctic, they may be subjected to air mass influence from Asia and western North America (Bailey et al., 2000) where endosulfan is commonly used.

Measurements of endosulfan II in air were only reported at Alert starting in 2002 (Table 3). The concentrations were found to be much lower than that observed for endosulfan I with mean values ranging from 0.10 to 0.17 pg/m^3 from 2002 to 2005. No particular seasonality was observed with the highest concentration detected during the week of April 1, 2003 at 1.8 pg/m^3. Endosulfan II was not detectable in samples collected at Nuuk.

3.1.3. Polybrominated diphenyl ethers (PBDEs)

Annual average concentrations of PBDE congeners measured at Alert and at Nuuk in 2005 are given in Table 5. At Alert, BDE 28/33, 47, 99, 100, 153, 154 and 209 accounted for, on average, 89% (32–97%) of the total of 14 BDE congeners. Dominant congeners were BDE 47 (average 29%, range 6.6–53%), BDE 99 (average 24%, range 6.3–56%) and 209 (average 26%, range 0.67–79%), reflecting the influence of penta-BDE and deca-BDE technical mixtures. Among 11 BDE congeners measured at Nuuk, the dominant congeners were also BDE 47 (average 40%) and BDE 99 (average 32%); followed by BDE 100, 153 and 28 indicating influence of penta-BDE technical mixture (Bossi et al., 2008). Despite the fact that Nuuk is a semi-urban location with a population of 13,500 while Alert is a military base with a crew of ~45, average air concentrations of BDE congeners measured at Nuuk were 2.5 to 5.9 times lower than those measured at Alert. PBDE air concentrations measured at Nuuk seems to reflect background levels in the remote Arctic while Alert may be influenced by the historical use of PBDEs at the military base and the Global Atmospheric Watch (GAW) laboratory facility where the air sampler is located. However, atmospheric trends at Alert (reported later in Section 3.2.3) should not be affected as they reflect relative air concentrations with respect to time. This will be discussed further in Section 3.2.3.

3.2. Temporal trends

The most rapid route of transport for persistent organic contaminants to the Arctic is via the atmosphere. Atmospheric LRT events have been identified annually moving polluted air masses within a few days from source regions into the central Arctic region (e.g. Eckhardt et al., 2007). Studying the long-term trends and seasonal cycles at different Arctic stations may reveal the influence of local and

Table 5
Annual mean (standard deviation; range) air concentrations of PBDEs (pg/m^3) at Alert and Nuuk.

	Alert				Nuuk[a]
	2002	2003	2004	2005	2005
BDE-17	0.055 (0.044; 0.022–0.20)	0.062 (0.068; 0.025–0.40)	0.087 (0.086; 0.027–0.35)	0.077 (0.079; 0.022–0.28)	0.03 (0.02; N.D.–0.06)
BDE-28/33	0.17 (0.19; 0.020–0.72)	0.17 (0.22; 0.021–1.2)	0.23 (0.35; 0.020–1.4)	0.26 (0.32; 0.019–1.1)	0.05[b] (0.04; N.D.–0.14)
BDE-47	2.2 (2.3; 0.21–11)	2.7 (3.0; 0.30–18)	2.8 (3.8; 0.21–16)	2.7 (2.7; 0.35–9.2)	0.46 (0.34; 0.08–1.40)
BDE-66	0.086 (0.093; 0.021–0.42)	0.099 (0.10; 0.024–0.56)	0.13 (0.19; 0.026–0.83)	0.12 (0.14; 0.021–0.45)	0.03 (0.02; N.D.–0.06)
BDE-85	0.11 (0.11; 0.045–0.71)	0.10 (0.15; 0.051–1.1)	0.13 (0.11; 0.055–0.60)	0.094 (0.056; 0.045–0.29)	0.02 (0.02; N.D.–0.05)
BDE-99	2.2 (2.7; 0.19–15)	2.3 (3.3; 0.26–22)	2.1 (2.4; 0.22–9.8)	2.0 (1.8; 0.31–6.9)	0.36 (0.30; 0.05–1.2)
BDE-100	0.40 (0.46; 0.031–2.1)	0.45 (0.47; 0.032–2.5)	0.47 (0.60; 0.032–2.4)	0.46 (0.46; 0.054–1.7)	0.08 (0.06; 0.01–0.22)
BDE-138	0.034 (0.011; 0.024–0.097)	0.042 (0.033; 0.027–0.21)	0.050 (0.036; 0.029–0.21)	0.034 (0.013; 0.024–0.12)	–
BDE-153	0.14 (0.19; 0.025–1.2)	0.16 (0.21; 0.023–1.4)	0.16 (0.22; 0.025–1.3)	0.13 (0.11; 0.023–0.61)	0.05 (0.03; N.D.–0.12)
BDE-154	0.16 (0.17; 0.015–0.81)	0.17 (0.18; 0.016–0.93)	0.16 (0.19; 0.016–0.88)	0.14 (0.13; 0.014–0.50)	0.03 (0.03; N.D.–0.11)
BDE-183	0.081 (0.10; 0.018–0.53)	0.21 (0.46; 0.020–2.7)	0.073 (0.051; 0.022–0.26)	0.067 (0.080; 0.018–0.52)	0.02 (0.02; N.D.–0.04)
BDE-190	0.046 (0.0080; 0.034–0.079)	0.048 (0.012; 0.038–0.11)	0.065 (0.041; 0.041–0.20)	0.44 (0.97; 0.043–6.7)	–
BDE-209	1.0 (0.70; 0.091–2.4)	2.1 (2.5; 0.098–9.8)	1.2 (1.1; 0.22–4.1)	1.8 (1.6; 0.21–7.0)	–
ΣBDE[c]	6.7 (6.3; 0.78–32)	8.6 (7.6; 1.4–47)	7.6 (7.2; 1.3–31)	8.4 (5.6; 1.5–21)	1.14 (0.81; 0.14–3.3)

[a] Adopted from Bossi et al. (2008).

[b] BDE 28 only, not coelution with BDE 33.

[c] ΣBDE was calculated with 14 measured BDE for Alert and 11 measured BDE for Nuuk.

seasonal factors, such as geomorphology, ambient temperature, elevation, humidity, precipitation, proximity to open or ice covered ocean surface and distance to potential sources, on the air concentrations of POPs at each location. This will give us information on how best to interpret the results of long-term air monitoring programs in assessing the effectiveness of control strategies.

3.2.1. Polychlorinated biphenyls (PCBs)

Fig. 3 shows the temporal trends of PCB 52, 101, 153 and 180 at the 4 long-term stations derived by DF. Apparent first order halflives are given in Table 6. From Fig. 3, it is obvious that the air concentrations of PCBs at most stations did not show continuous and consistent decline throughout the time series. This is probably why previous studies have shown no evidence of overall declines of PCBs in the Arctic region, e.g. Berg et al. (2004) and Gioia et al. (2008). Therefore, as mentioned above, these halflives should be interpreted with caution. For Alert, during the first year of the laboratory change, i.e. 2002, the atmospheric PCB data were apparently affected by certain artifact resulting in low detections of most congeners. Since NLET adopted the historical analytical method of FWI in order to maintain consistency in analysis, results in the first year might have been affected by unfamiliarity of the analytical procedures. From Fig. 3, it can be seen that after 2002, this problem did not persist and the long-term trends were reasonably maintained. Therefore, data reported for this station in 2002 should not be used to derive temporal trends. Halflives were only derived between 1993 and 2001, i.e. before the laboratory change, since it is not possible to derive meaningful halflives after 2002 with only 3 years of data.

It can be seen from Fig. 3 that air concentrations of PCBs showed a general decline at Pallas, Alert and Zeppelin. At Storhofdi, general reductions in air concentrations were observed for all reported congeners between 1995 and 1998. After 1998, the air concentrations of almost all congeners increased. Especially the lighter congeners, the air concentrations rose back to almost the same level as in 1995/1996, resulting in prolonged halflives of 19 and 22 years for PCBs 28 and 52, respectively, and no apparent decline for PCBs 31 and 101 (as represented by not determinable halflives in Table 6). Heavier congeners still showed overall decreasing trends with halflives ranging from 5.4 to 6.5 years. Nonetheless, the air concentrations of PCBs at this station after 2002 were almost constant, except for PCBs 105 and 138 which showed declining concentrations between 2002 and 2005. This may be an indication that the PCB air concentrations at Storhofdi are approaching some kind of steady state.

Despite the generally rapid decline for all congeners at Zeppelin ($t_{1/2}$ ranging from 3.9 to 9.1 y), it is interesting to note that most congeners increased in air concentrations at this location in 2005/2006. This increase is obvious for lighter congeners, such as PCBs 28 and 31, starting in 2005 and continued through 2006; but is particularly prominent for heavier congeners, e.g. PCBs 138, 153 and 180, in 2006. Some possible influencing factors will be discussed later in Section 3.2.2.

With 2 more years of data (2000 and 2001), the decline in atmospheric PCBs at Alert has shown an acceleration for most congeners as compared to that observed between 1993 and 1999 reported by Hung et al. (2005) (Table 6). The halflives now range from 3.6 to 20 years which are similar to those derived for Zeppelin for most congeners and to Pallas and Storhofdi for higher chlorinated congeners.

Eckhardt et al. (2007) have attributed high air concentrations of PCBs measured at Zeppelin in July 2004 and spring 2006 to boreal forest fires in Yukon/Alaska and agricultural fires in Eastern Europe, respectively. It was believed that biomass burning can enhance volatilization of previously deposited organic chemicals, such as PCBs, from soil. These high concentration episodes are also apparent in Fig. 3. However, similar increases in air concentrations of PCBs were not observed at the other 3 stations during the same period. This may be explained by the fact that the air sampling time was relatively shorter (48 h) at Zeppelin as compared to the other sampling stations (usually 7-day integrated). Samples with shorter sampling time would better facilitate the analysis of long-range transport episodes.

3.2.2. Organochlorine pesticides (OCPs)

Fig. 4 presents the temporal trends and seasonal cycles of 4 selected OCPs, namely HCB, c-chlordane, γ-HCH and *p,p′*-DDT, developed by DF for the four stations. HCB and c-chlordane were not measured at Pallas and trends are therefore not given in Fig. 4. Apparent first order halflives derived are given in Table 6.

For HCB, a past-used fungicide and industrial by-product, generally a downward trend can be expected. However, between 2003 and 2006, the air concentrations at the Zeppelin station have been increasing continuously. A similar increase in air concentration was also observed at Alert after 2002 despite the disruption in trend due to the laboratory change. Due to slightly increasing trends, halflives of HCB at Alert are not presented in Table 6. Note that breakthrough was observed in about 30% of all samples for HCB measured at Alert and therefore the air concentrations may be underestimated. Breakthrough is defined as (amount on back PUF)/(amount on front PUF)>0.333. However, breakthrough mainly affects higher concentration samples during warmer periods of time and should

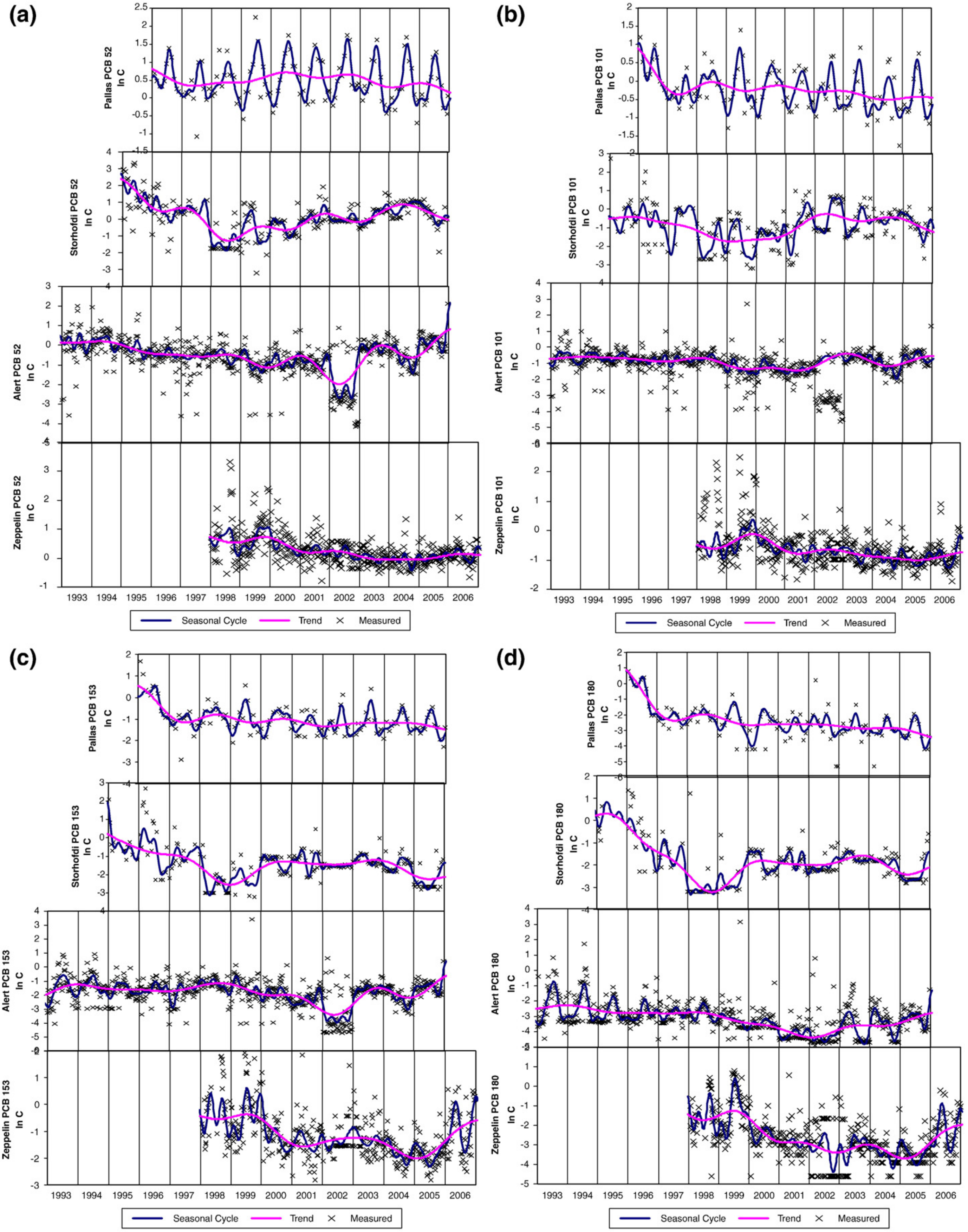

Fig. 3. Trends of PCB (a) 52, (b) 101, (c) 153 and (d) 180 at Pallas, Storhofdi, Alert and Zeppelin.

Table 6
First order apparent halflives of various POPs at the 4 long-term monitoring stations.

PCBs	Alert (93–01)[a]		Alert (93–99)[c]		Pallas (96–05)		Storhofdi (95–05)		Zeppelin (98–06)[d]	
	$t_{1/2}$ (y)[b]	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2
28	7.1	0.76	10	0.55	18	0.27	19	0.064	6.0	0.54
31	5.6	0.92	6.9	0.86	–	–	N.D.	–	5.6	0.56
52	4.6	0.81	4.0	0.86	47	0.09	22	0.017	8.3	0.70
101	6.9	0.77	7.3	0.53	11	0.51	N.D.	–	9.6	0.60
105	7.0	0.45	11	0.21	–	–	6.2	0.28	3.2	0.54
118	8.3	0.28	N.D.	–	8.3	0.61	6.5	0.21	4.8	0.60
138	20	0.17	N.D.	–	5.7	0.60	5.5	0.27	3.6	0.47
153	8.3	0.32	79	0.01	7.7	0.48	6.5	0.30	6.3	0.34
180	3.6	0.84	4.2	0.76	3.4	0.59	5.4	0.24	3.9	0.40

OCPs	Alert (93–01)[a]		Alert (02–05)		Pallas (96–05)		Storhofdi (95–05)		Zeppelin (93–06)	
	$t_{1/2}$ (y)[b]	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2	$t_{1/2}$ (y)	r^2
HCB	N.D.	–	N.D.	–	–	–	6.2	0.67	18	0.52
α-HCH	5.0	0.81	15	0.38	6.1	0.77	4.3	0.87	4.7	0.89
γ-HCH	7.3	0.71	3.7	0.92	4.6	0.89	10	0.26	4.6	0.88
t-chlordane	4.9	0.82	N.D.	–	–	–	3.5	0.51	9.3	0.86
c-chlordane	6.7	0.75	N.D.	–	–	–	N.D.[e]	–	16	0.70
t-nonachlor	5.1	0.89	13	0.44	–	–	N.D.[e]	–	26	0.40
c-nonachlor	4.4	0.71	2.6	0.86	–	–	–	–	94	0.77
Endosulfan I	62	0.024	N.D.	–	–	–	–	–	–	–
o,p′-DDD	N.D.	–	N.D.	–	–	–	–	–	6.3	0.66
o,p′-DDE	N.D.	–	N.D.	–	–	–	–	–	4.4	0.59
o,p′-DDT	N.D.	–	N.D.	–	–	–	–	–	5.7	0.84
p,p′-DDD	N.D.	–	N.D.	–	–	–	–	–	9.2	0.29
p,p′-DDE	N.D.	–	N.D.	–	–	–	–	–	11	0.36
p,p′-DDT	N.D.	–	N.D.	–	–	–	–	–	4.1	0.82

[a] Halflives are only reported for Alert between 1993 and 2001 before the laboratory change. See text for details.

[b] $t_{1/2}$ = halflife, calculated as ln 2/slope. Compounds that do not show a consistent declining trend (i.e. greatly fluctuating or steady or slightly increasing trends) are indicated as N. D. = not determinable.

[c] Hung et al. (2005).

[d] PCB concentrations were compromised by local contamination before 1998, data not included in half-life derivation.

[e] Air concentrations of *c*-chlordane and *t*-nonachlor measured at Storhofdi have been almost constant from 1995 to 2003 but decreased drastically in 2004 and 2005 with no apparent reason. It is therefore not possible to provide a meaningful halflife.

not influence the overall temporal trend. This increasing concentration may be due to the following potential causes:

1) *Reduction in sea ice coverage may result in increased volatilization of previously deposited chemicals from ocean*: There could be potential increased evaporation of previously deposited HCB from open surfaces along the western coast of Spitsbergen (Svalbard, Norway), where Zeppelin is located, which has been ice-free in the winters of 2003 to 2006. Due to short-term changes in the ocean current regime along the Fram-Strait, deep Atlantic waters were continuously up-welling onto the surface causing increased surface water temperatures resulting in ice-free winters. Fig. 5 shows icemaps in winter/spring of 2005, 2006 and 2007 which indicated very open drifted ice (green colour) and open water (white colour) along the west coast of Spitsbergen. As mentioned in the previous section, a slight increase in the atmospheric trend of PCB concentrations was also observed at Zeppelin in 2005 and 2006. This could be the result of the same cause. As the major source of organic pollutants in Arctic air is probably the result of long-range transport, the increase in air concentration of HCB at Alert may be attributed to the same hypothesis since Arctic sea ice coverage has reduced significantly in recent years in both winter and summer (Stroeve et al., 2008).

   However, one must also bear in mind that the air concentrations of OCPs, such as HCB, can be influenced by many other factors as well. For instance, changes in air and ocean current flow over time can potentially increase or decrease the relative capacity in the two media, subsequently reversing the direction of OC flux from the air to the ocean and *vice versa*. Despite the reduction in sea ice in recent years, it is unclear whether our first hypothesis was correct without direct measurements of HCB fluxes from the Arctic Ocean. Su et al. (2006) have estimated a fugacity ratio (fugacity in sea water over fugacity in air) of 0.21 at 0 °C in the Arctic, indicating deposition potential of HCB in this region. However, this ratio was calculated using average air and seawater concentrations which are not site specific. No sea water concentrations of HCB close to Svalbard is currently available. Gioia et al. (2008) have reported deposition dominates over volatilization for PCBs in the Arctic region, particularly near the marginal ice zone (78–79 °N) close to Svalbard, possibly due to ice melting releasing PCBs to the atmosphere. They have also noted the complexity of air–ocean exchange of PCBs as fluxes are influenced by water column characteristics, e.g. dissolved-particle phase partitioning, biological productivity, reactivity and salting; air–water concentration gradients and transport by molecular and turbulent motion. At the current state of limited knowledge on air–ocean–sea ice–snow interaction of pollutants, the hypothesis of retreating sea ice contributing to higher air concentration due to evaporation of previously deposited HCB cannot be validated.

2) *Increased and continuous use of pesticides containing HCB*: The use of HCB has been banned in most countries in the 1970s. However, usage of chlorinated solvents and pesticides containing HCB as a by-product continues. Gouin et al. (2008) have shown higher concentrations of HCB in air measured in the central valley agricultural area in Costa Rica as compared to that measured in the mountains far from farming activities. They have attributed this difference to past use of HCB and current use of chlorinated fungicides, such as chlorothalonil and quintozene, which contain HCB impurities (Ambrus et al., 2003) and were used in the central valley. Dore et al. (2007) reported that the use of chlorothalonil in the U.K. increased significantly in 2004 and remained high in 2005. This renders a significant increase in emission estimates of HCB

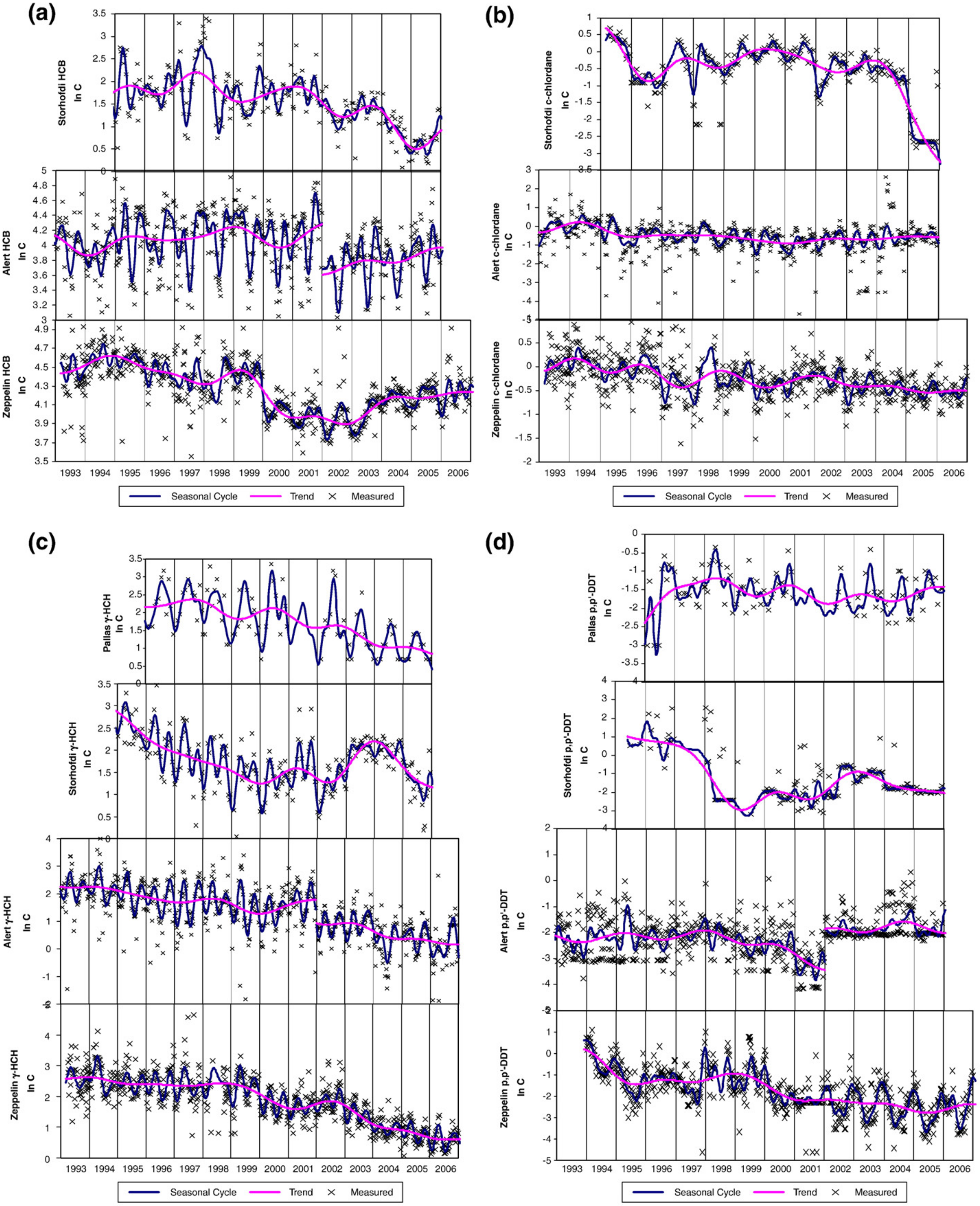

Fig. 4. Temporal trends of (a) HCB, (b) *c*-chlordane, (c) γ-HCH, and (d) *p,p*′-DDT.

from the U.K. as a result of pesticide use from 298 kg in 2003 to 822 kg and 825 kg in 2004 and 2005, respectively (Dore et al., 2007). This corresponds to the increase in air concentrations of HCB in 2004 and 2005 observed at Zeppelin (Fig. 4a), which is generally more affected by input from Europe as compared to Alert due to proximity. The increase of HCB observed in Arctic air could

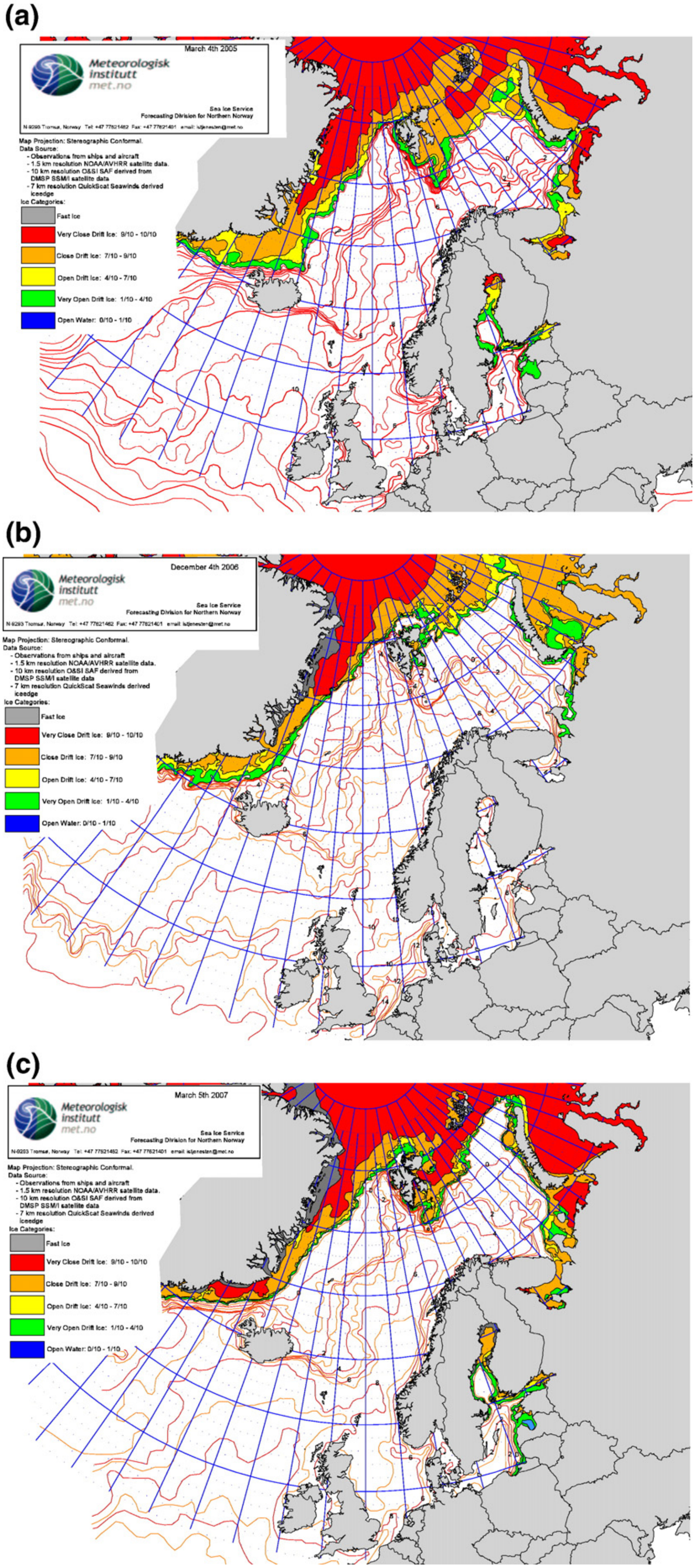

Fig. 5. Ice maps showing generally open water on the west coast of Svalbard on (a) March 4, 2005; (b) December 4, 2006 and (c) March 5, 2007.

be related to increased worldwide usage of HCB-contaminated pesticides in recent years. However, no other emission or usage data for HCB-contaminated pesticides is available to confirm this hypothesis.

For *c*-chlordane, slightly declining trends were observed at Alert and Zeppelin which is consistent with the fact that technical chlordane has been banned for almost 20–30 years in most western industrialized countries. At Alert, a relatively shorter halflife of 6.7 years was observed between 1993 and 2001 but the decline has more or less leveled off after 2002 with no apparent declining trend. At Zeppelin, an overall apparent first order halflife of 16 years was derived. Note that given the variations in the dataset, this is not inconsistent with the conclusion reached by Berg et al. (2004) that there is no clear trend for the chlordanes measured at Zeppelin. As Berg et al. (2004) have pointed out that the dataset has not gone through DF prior to final statistical analysis in their study, a direct comparison of results from the two studies is not applicable. At Storhofdi, the trend is not determinable (Table 6) as the concentrations were more or less constant from 1996 to 2003 ($t_{1/2,\ 96-03} = 64$ y), then declined significantly in 2004 and 2005 to mostly below detection limit. Less stable than *c*-chlordane, *t*-chlordane shows a more rapid decline at Alert, Storhofdi and Zeppelin with halflives of 4.9 years (93–01), 3.5 years and 9.3 years, respectively.

Fig. 4c shows the air concentration trends and seasonal cycles of γ-HCH. The pesticide lindane contains almost pure γ-HCH and has been widely used around the globe (Li et al., 1996). Generally declining trends of atmospheric γ-HCH was observed at Pallas, Alert and Zeppelin throughout the 1990s and early 2000s. For Storhofdi, declining trend was observed up to 2002, followed by an increase in concentration up to end of 2003 and a declining trend was resumed. Several countries have reduced/eliminated the use of lindane since it has been shown to persist in biota (Li et al., 2004). Global usage has declined significantly from the 1980s to the 1990s (Li et al., 2004). Canada, a major user of lindane in North America, has deregistered lindane for use on canola seeds in July 2001 and a ban was introduced in 2004 (Becker et al., 2008). Despite the disruption in trend at Alert in 2002, it is still apparent that the air concentration decline of lindane in Arctic air has accelerated between 2003 and 2005 ($t_{1/2,\ 02-05} = 3.7$ y) compared to between 1993 and 2001 ($t_{1/2,\ 93-01} = 7.4$ y) (Fig. 4c). This increase in decline rate is also clearly observed at the Zeppelin station after 2002. If halflives before and after 2002 were derived for γ-HCH air concentrations at Zeppelin, they would be $t_{1/2,\ 93-01} =$ 6.6 y and $t_{1/2,\ 02-05} = 2.4$ y, respectively; showing similar acceleration in decline rates as those observed at Alert. This reflects the fact that organic contaminants, such as lindane, can travel rapidly via the air to the remote Arctic and a decline in use in source regions will result in a decrease in Arctic air concentrations quickly. On the other hand, it is

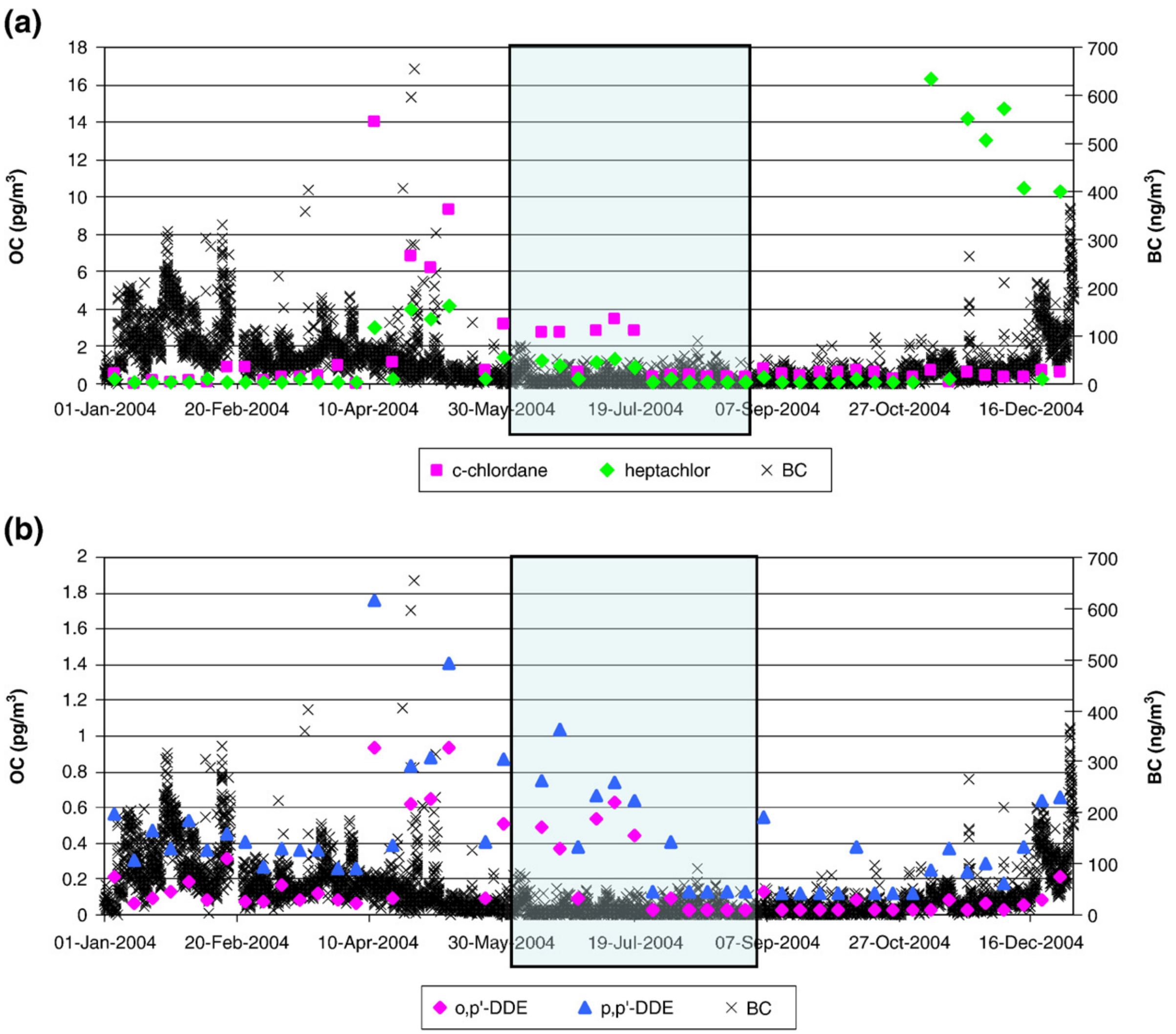

Fig. 6. Air concentrations of BC (ng/m^3) and (a) *c*-chlordane and heptachlor; and (b) *o,p′*- and *p,p′*-DDE in pg/m^3 measured at Alert in 2004. Blue rectangle indicates time period of Yukon/Alaska forest fire.

not apparent why the air concentrations of γ-HCH would increase between 2002 and 2003 at Storhofdi but the reduction in air concentration seems to have resumed in 2005.

DDT-related compounds were usually found at low levels at the 4 stations. Consistent long-term trends were generally not distinguishable in Arctic air (Fig. 4d for *p,p′*-DDT), except at Zeppelin, where declining trends for all 6 isomers were observed with halflives ranging from 4.1 to 11 years (Table 6).

Several high air concentration episodes of c-chlordane, *p,p′*- and *o,p′*-DDE were observed at Alert and Zeppelin in 2004. While no high air concentrations of PCBs were noted at Alert during the summer forest fire in Yukon/Alaska, elevated concentrations measured for *c*-chlordane and DDEs in summer 2004 could possibly be the result of biomass burning. Fig. 6 shows the air concentrations of black carbon (BC), *c*-chlordane and DDEs measured at Alert in 2004. Elevated air concentrations of all three OCPs were observed during the haze period from end of April to early May. This is consistent with the fact that *c*-chlordane and *p,p′*-DDE showed elevated particle-bound concentrations during Arctic Haze seasons (December to April); with up to 80–90% of the total concentrations found on the filter (associated with particles). During the Yukon/Alaska forest fire event from June to August 2004 (indicated on Fig. 6 by blue box), EBC air concentrations were relatively low compared to the haze season. Therefore, higher concentrations of *c*-chlordane and DDEs observed during this time may be the result of enhanced evaporation of previously deposited pollutants from forest soil during biomass burning. Higher concentrations of other chlordane- and DDT-related compounds were not observed during this period. This could be because *c*-chlordane is relatively more stable and may remain in soil for prolonged period of time [degradation halflife of chlordanes in soil is over 20 years (Mattina et al., 1999)] and subject to evaporation when temperature increased during a forest fire event. DDTs were used in the Yukon/Alaska region before its ban. Previously deposited compounds would have been degraded to DDEs and DDDs.

Alert is the only station that reported a temporal trend for endosulfan I. Very slow decline is observed in air at Alert with an apparent halflife of 62 years between 1993 and 2001 and remained more or less constant between 2002 and 2005 (Fig. 7). It is not

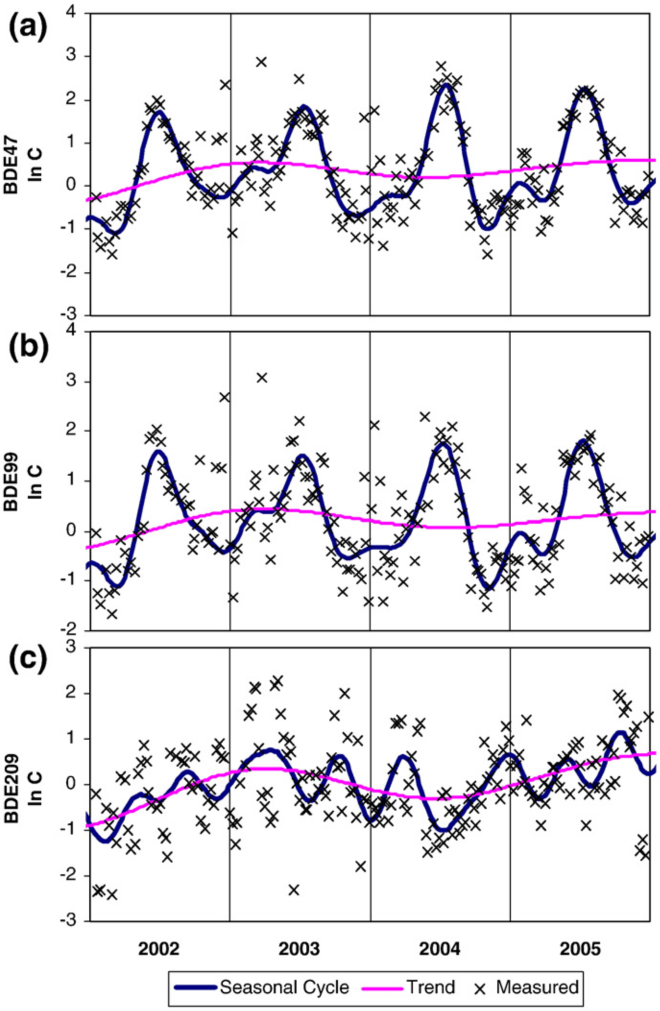

Fig. 8. PBDE temporal trends at Alert: (a) BDE 47, (b) BDE 99 and (c) BDE 209.

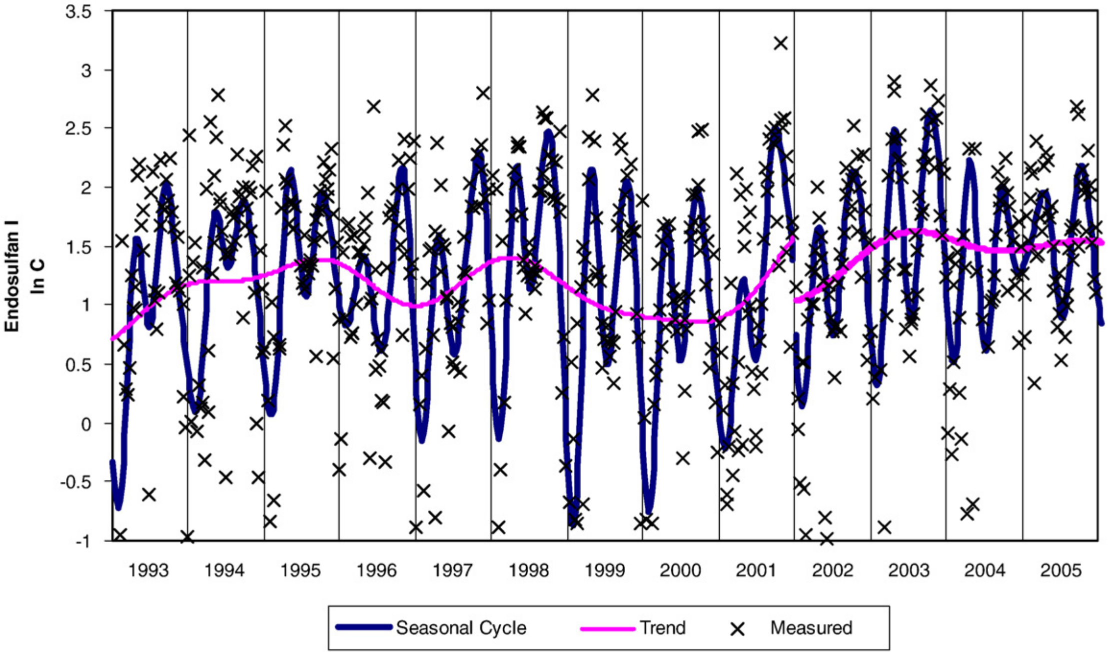

Fig. 7. Temporal trend of Endosulfan I at Alert.

Table 7
Trends of PBDEs at Alert.

BDE	t_2 (y)	r^2	p-value
17	11	0.95	<0.0001
28	7.3	0.58	<0.0001
47	6.4	0.34	<0.0001
66	11	0.91	<0.0001
85	17	0.44	<0.0001
99	12	0.13	<0.0001
100	8.3	0.16	<0.0001
153	28	0.03	<0.01
154	N.D.[a]	–	–
209	3.5	0.37	<0.0001

[a] N.D. = not determinable. No significant increasing trend.

possible to derive a temporal trend for endosulfan II at this location since only 4 years of data are available.

3.2.3. PBDEs

Su et al. (2007) have reported the first time series of PBDEs measured in air at Alert. An update of the temporal trends is given here. Fig. 8 shows the temporal trends of selected congeners. Doubling times, t_2, are given in Table 7, ranging from 3.5 years for BDE 209 to 28 years for BDE 153. The t_2 are longer than those reported by Su et al. (2007). This can be expected as the penta- and octa-BDE technical mixtures are being phased out or banned in many countries in North America and Europe recently. From Fig. 6, it can be seen that BDE 209 (deca-BDE) shows continuous increasing trend with a short doubling time of 3.5 years. This reflects the continuous use of deca-BDE technical mixture worldwide. While the air concentrations of lighter BDE congeners, such as BDE 47 and 99, followed the seasonal temperature cycle well, with higher concentrations during the summer and lower concentrations in winter, higher BDE congeners, such as BDE 153 and 209, do not show good correlations with temperatures. The difference in seasonality between lighter and higher congeners may be the results of a combination of higher input of heavier congeners bound to particles during the Arctic haze season, the difference in the ability of different congeners to bind to particles, the lower volatility and higher tendency to deposit for the higher congeners (Su et al., 2007).

As mentioned earlier, the air concentrations of PBDEs measured at Alert may be influenced by the emission of these compounds from consumer products impregnated with PBDEs used in the nearby military base and at the GAW station where the sampler is located. Therefore, the absolute concentrations measured may be an overestimation of the Arctic background level. This is apparent when comparing the Alert concentrations with those measured at Nuuk. However, this should not affect the temporal trends, which are the relative changes in air concentrations of PBDEs with respect to time, since the amount of consumer products at both the military base and the GAW laboratory do not change consistently with time. Due to the remoteness of the site, furniture and equipment rarely changed unless it is necessary. In other words, the increasing trends observed at this location should be a real reflection of that of the influencing source regions.

3.3. Changes in air concentrations at satellite stations

3.3.1. Kinngait — changes in air concentrations (2001 vs. 1995)

Atmospheric measurements at Kinngait were conducted in 1994–1995 and again in 2001–2002. The relative changes in POP air concentrations at this station can be compared with those observed at long-term measurement stations, such as Alert, as shown in Table 8.

It can be seen that the past-used chemical of HCB does not show a significant change in air concentration between 1995 and 2001 at either Kinngait or Alert. Both stations show significant declines in air concentrations of the HCH isomers, chlordanes and dieldrin between 1995 and 2001. The decline in air concentration of lindane (γ-HCH) was relatively faster at Kinngait than at Alert which may reflect the input of lindane from Eurasian sources sustaining air concentrations

Table 8
Comparisons of concentration change between (a) 1995 and 2001 at Kinngait and at Alert; and (b) 1993 and 2002/2003 in Yukon (Tagish and Little Fox Lake) and at Alert.

(a)						
	Kinngait			Alert		
	1995 (pg/m^3)	2001 (pg/m^3)	% changed[e]	1995 (pg/m^3)	2001 (pg/m^3)	% changed[e]
HCB[a]	63	58	*−8.8*	65	69	*5.8*
α-HCH	71	25	**−64**	56	20	**−64**
γ-HCH	9.4	4.7	−50	8.4	6.8	−19
Σchlordane[b]	2.3	1.2	**−48**	2.8	0.95	**−66**
Dieldrin	1.5	0.46	**−68**	1.3	0.49	**−62**
ΣDDT[c]	1.4	1.2	*−7.8*	1.3	0.58	**−54**
ΣPCB[d]	5.6	5.9	*5.0*	5.1	2.2	**−57**

(b)						
	Yukon			Alert		
	Tagish 1993 (pg/m^3)	Little Fox Lake 2002/2003 (pg/m^3)	% changed[e]	1993 (pg/m^3)	2003 (pg/m^3)	% changed[e]
HCB[a]	42	67	**21**	54	49	*−8.9*
α-HCH	79	48	**−39**	57	16	**−71**
γ-HCH	11	4.5	**−66**	13	2.8	**−79**
Σchlordane[b]	2.9	1.0	**−68**	2.2	1.2	*−45*
Dieldrin	0.93	0.31	**−76**	1.1	0.86	**−19**
ΣDDT[c]	1.4	0.93	**−54**	0.82	0.93	*13*
ΣPCB[d]	3.7	4.1	*−8.1*	5.8	3.9	**−33**

[a] HCB broke through in ~30% of all samples taken, mean concentrations may be underestimated.

[b] Σchlordane = sum of *c*-chlordane, *t*-chlordane, *c*-nonachlor and *t*-nonachlor.

[c] ΣDDT = sum of *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT and *p,p′*-DDT.

[d] ΣPCB = AMAP sum of 10 PCBs (PCB 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180).

[e] % change $= \frac{(\text{annual average concentration in 2001} - \text{annual average concentration in 1995})}{\text{annual average concentration in 1995}} \times 100\%$, a positive value indicates an increase while a negative value indicates a decrease in air concentrations. **Bold type** indicates statistically significant change with p-value<0.01. *Italics* indicate not statistically significant change with p-value>0.1. Normal fonts indicate statistically significant change with p-value<0.05.

at Alert. On the other hand, the atmospheric concentrations of ΣDDT and ΣPCB did not change significantly at Kinngait while those at Alert have declined 54% and 57%, respectively, from 1995.

3.3.2. Western Canadian Arctic — changes in air concentrations between Little Fox Lake (2003–2004) vs. Tagish 1993

Tagish and Little Fox Lake are both located in the Yukon region of Canada in relatively close proximity. Atmospheric measurements were conducted at Tagish in 1993–1994 and at Little Fox Lake in 2003–2004. Therefore, similar comparisons with concentration changes observed at Alert as that for Kinngait can be performed and results are summarized in Table 8. It can be seen that all OCPs showed reduced air concentrations in the Yukon region, except HCB. The air concentrations of α-HCH at Alert were lower than those observed in the Yukon in both 1993 and 2003 and seem to have declined at a faster rate. α-HCH is a major component of the banned technical mixture of HCH. Asian countries, e.g. India, China and the former Soviet Union, were major users of the technical mixture before its ban. Su et al. (2006) have attributed the higher air concentrations of α-HCH observed in Yukon to the potential influence of trans-Pacific transport from Asia when technical HCH was still in use and higher precipitation rate in the Yukon compared to other Arctic locations. There might be enhanced deposition of α-HCH to the Earth's surface following long-range transport from Asia in the Yukon region previously and volatilization from soil now becomes a source of α-HCH to the Yukon atmosphere. This is consistent with the slower rate of decline of this compound in the Yukon compared to Alert.

4. Conclusions and perspectives

Long-term atmospheric measurements of organic pollutants at the four AMAP Arctic sampling sites provide a unique opportunity to assess sources, distribution and variations in atmospheric contaminant levels. This is especially useful for evaluating the effectiveness of national and international production/emission/usage control strategies, such as the Stockholm Convention on POPs. In addition, the extensive Arctic air concentration dataset enables a comprehensive evaluation and refinement of environmental transport and fate models which are important for forecasting the future behaviour and distribution of pollutants.

Recently, the association of inter-annual variabilities in the atmospheric concentrations and spatial distribution of organic pollutants with potential climate change-related patterns is of great scientific interest. Climate change and variations will have significant implications on how organic pollutants will be transported and partitioned among environmental media (Macdonald et al., 2005). From the current dataset, it has been demonstrated that changes in local climate regime, such as that at Zeppelin (Svalbard), may already be affecting temporal trends. Increased frequencies of forest fire events due to climate change may result in enhanced input of pollutants to the Arctic; as seen from the current dataset for higher PCB and OCP concentrations measured at Zeppelin and Alert. To enable the comparison of POP levels over time and provide information on climate change influence on global and regional POP transport, sustaining the established operation of comparative atmospheric measurement programs is most important. Collaboration between established atmospheric monitoring programs and regional/global transport studies will provide the knowledge basis to interpret POP measurements under the influence of climate change. For this kind of comparison to be meaningful, data comparability between monitoring programs is of utmost importance. Regular participation in coordinated QA/QC comparison activities is recommended.

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