Atmospheric concentrations in air and deposition fluxes of POPs at Råö and Pallas, trends and seasonal and spatial variations

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1 Introduction

Persistent organic pollutants (POPs) are subjected to long-distance transport, are likely to bioaccumulate and may thus pose a risk to humans and wildlife in aquatic and terrestrial ecosystems both far from and close to source areas (UNEP 1998). Many POPs are semivolatile, and as such they can be transported in the atmosphere either in the gas- or in the particulate phase. The distribution between the phases affects their transport in the atmosphere, which for several POPs is characterised by exchange between the atmosphere and environmental surfaces in a way that has been described as the 'grasshopper effect' (Wania and Mackay 1996). The importance of atmospheric transport and deposition as a pathway for POPs to the Nordic environment as well as to the Arctic areas has been shown both by measurement activities and by model exercises (Stern et al., 1997, Harner et al., 1998, Hung et al., 2001, Kallenborn et al., 2002, Shatalov & Malachinev, 2000, UNEP 2002).

Several of the substances included in the POP group have been used as industrial chemicals and are today banned or have a restricted use. However, due to their persistence they are still present in the society and may be emitted via diffuse sources. Some of the POPs are also unintentionally formed e.g. via combustion. The chemicals are included or considered in the UN-ECE POP protocol, in the Stockholm convention (UNEP) and in the marine conventions; the Oslo and Paris Commission (OSPAR) and the Helsinki Commission (HELCOM). Several POPs are "priority substances" in the EU water framework directive (WFD) and atmospheric deposition has shown to be an important source for the occurrence of POPs in aquatic environments. Thus atmospheric transport processes also are relevant for the implementation of the WFD.

In order to assess the importance of atmospheric transport and deposition of POPs and to quantify the regional atmospheric cycling, measurements are carried out. A further aim with measurements is to obtain information in order to develop a policy to reduce this pollution (emission control). The results could be used to support international strategies and protocols. Data from these measurements are also reported and used within EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) and AMAP (Arctic Monitoring Assessment Programme).

In Sweden, measurements of selected POPs were included in the Swedish Monitoring Programme for Air Pollutants during 1995 at one station at the Swedish West Coast, Råö. In 1996, a monitoring programme for POPs started at one station in a sub-arctic area in Finland Pallas.

The monitoring programme includes different classes of substances, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and selected chlorinated pesticides (e.g. HCH). Measurements of polybrominated diphenyl ethers (PBDEs) were added to the monitoring programme at the Swedish West Coast in 2001 and in Pallas in 2003. All the above mentioned substances are frequently present in the atmosphere and are of varying origin, i.e. they are emitted to the environment via industrial processes, combustion, end-use of products and/or agricultural use.

In addition to the regular monitoring programme, screening studies are carried out in Sweden. The aim of these studies is to investigate the presence and concentration levels of selected chemicals

and to provide information about important transport pathways of the substance in the environment, e.g. atmospheric transport. Measurements of air concentration and deposition fluxes of new chemicals within the screening programmes are often coordinated with the monitoring programme. The results of a screening may lead to the inclusion of new chemicals in the regular monitoring programmes. Recently, a screening study of endosulfan and mirex was carried out and results are partly presented here (Palm et al., 2005).

In this report, the results from the measurements of POPs in air and deposition carried out between 1996 and 2005 will be presented. Comparison is made with previously published data from the Swedish West Coast in 1989-1994 (Brorström-Lundén, 1995). Seasonal and temporal trends will be discussed, as well as spatial variation in a European perspective. Results from screening studies will be presented. The data summarised and presented here can also be found in the Swedish EPA's database for air pollutants hosted by IVL (www.ivl.se).

2 Methods

2.1 Sampling and analyses

The measurements of POPs have been carried out at two sites: Råö, which is an EMEP station at the Swedish west coast (previously Rörvik) and Pallas, a monitoring station in northern Finland within AMAP. The sampling at Pallas is operated by the Finnish Meteorological Institute (FMI) and the measurements are performed in co-operation between Sweden and Finland.

The sampling program includes parallel sampling of POPs in air and deposition with a frequency of one week per month and the sampling is undertaken simultaneously at the two sites. In order to adopt the recommendations from EMEP, the sampling programme at Råö was extended in 2001 to weekly sampling in air and monthly sampling of deposition.

POPs in air are collected using a high volume air sampler (HVS). A glass fibre filter is used for trapping the particles followed by an adsorbent of polyurethane foam (PUF) for collecting compounds in the gas phase, Figure 1. Both wet and dry deposition is collected using an open sampler (bulk sampler). This sampler consists of a 1 m^2 Teflon coated surface with 10cm high edges. The bottom declines slightly to a central opening where a cassette with an adsorbent of PUF is attached, Figure 1. The deposition sample includes both compounds in the precipitation and compounds deposited to the collection surface of the sampler (PUF, filters and ethanol). Both the precipitation and the deposited particles are included in the analysis. At Pallas, a modified sampler is used for collection of POPs in the snow.

At the laboratory, the samples (filters and PUFs) were extracted in a Soxhlet apparatus with acetone during 24 hours (± 2 hours). After the Soxhlet extraction, internal standards were added to the samples and the organic compounds were extracted to an organic phase by liquid/liquid extraction, 25 % of the total samples were used for PAH analysis and 75 % for PCBs, PBDEs and pesticides.

Prior to analysis of persistent chlorinated and brominated compounds, the samples were treated with concentrated sulphuric acid. Pre-treatment procedures, such as fractionation of the organic compounds on silica (PAH fraction) and aluminium columns (PCB fraction) were also performed

as additional "clean-up" procedures. Laboratory blanks as well as field blanks followed the same procedures in the analytical work.

The PAHs were analysed using a high performance liquid chromatograph (HPLC) with a fluorescence detector. The PCBs, pesticides and PBDEs were analysed on a gas chromatograph (GC) equipped with an electron capture detector (ECD) and a capillary column with non-polar bonded phase. For more details see the 'Manual for monitoring work' that is available via the Swedish EPA (www.naturvardsverket.se).

Figure 1. High volume air sampler and deposition sampler.

3 Meteorology

Information about meteorological conditions is important for evaluation of transport processes of POPs, e.g. deposition and re-emission.

In Figure 2, monthly precipitation and mean air temperature for the years 2001- 2005 at Råö are shown. The temperature data has been taken from a weather station located in Göteborg, approximately 70 km north of Råö, while the precipitation data originate from the Råö station.

The average ambient temperature and the amount of precipitation at Pallas were provided by FMI and have been summarised for the different sampling occasions (1 week per month; Figure 3).

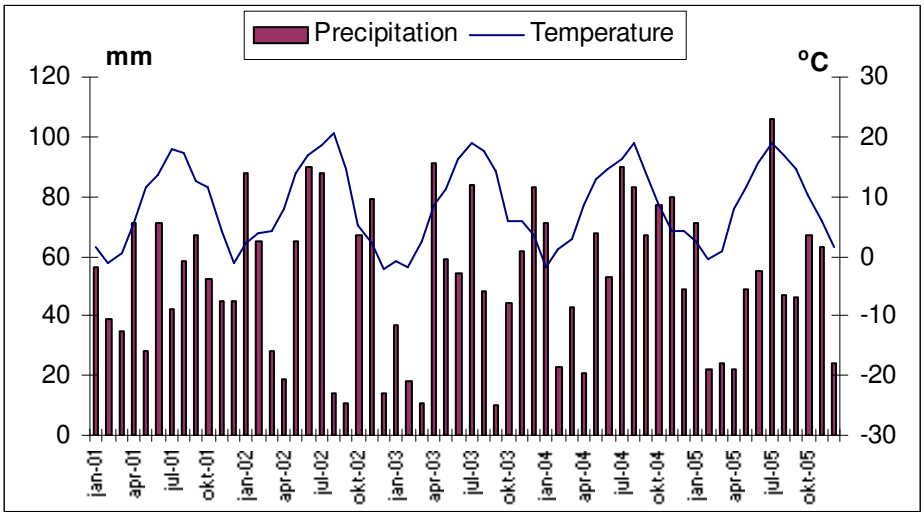


Figure 2. Average air temperature (Göteborg) and monthly precipitation at Råö (www.miljo.goteborg.se/luftnet and www.ivl.se).

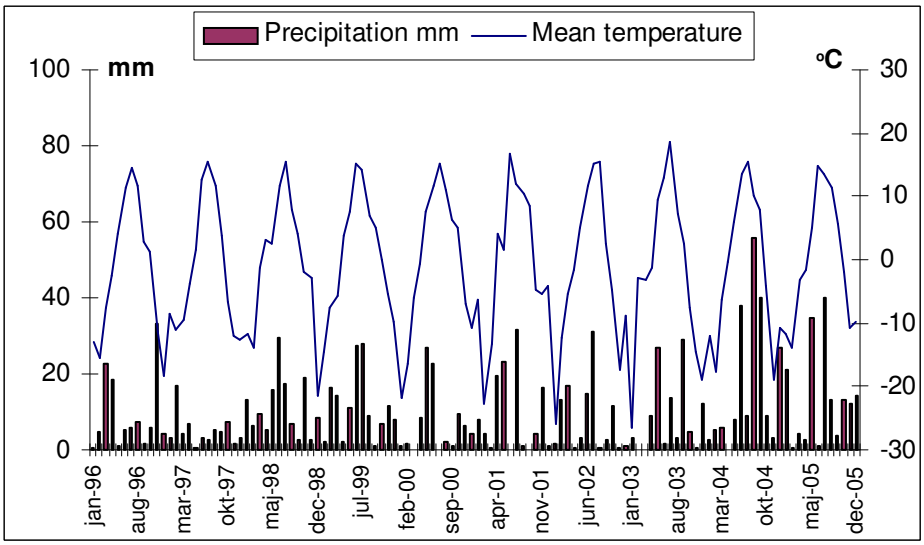


Figure 3. Average temperature and precipitation at Pallas (1996-2005) (data from FMI, 2005).

The average air temperature during the summer was generally 3-5°C lower in Pallas compared to Råö. The annual precipitation (Figure 4) at Råö during the last 10 years of measurements has usually been 100-200 mm higher than at Pallas. However, the last two years, the precipitation rates were at the same levels at both stations.

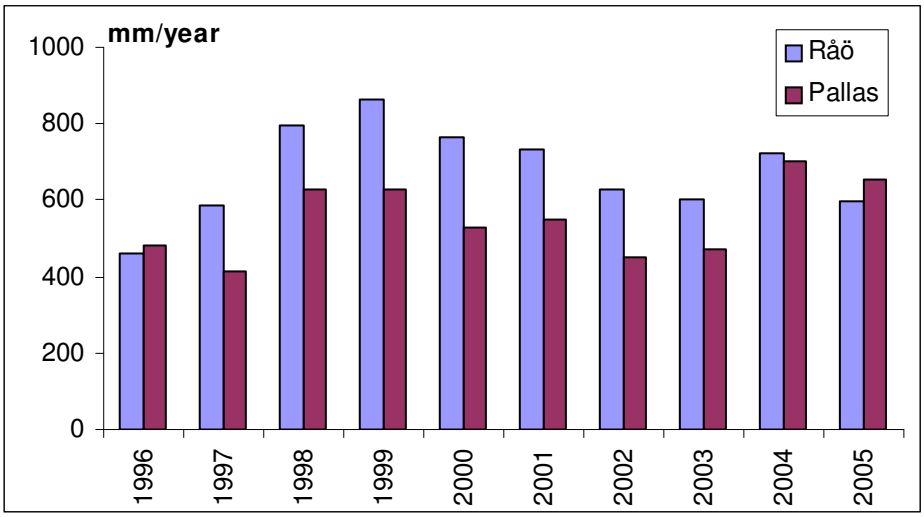


Figure 4. Annual precipitation at Råö and Pallas.

4 Results and Discussion

4.1 Atmospheric concentrations

The yearly averages (1996-2005) of PAHs, PCBs, HCHs, chlordanes, DDTs and PBDEs in air measured at Råö and Pallas are presented in Appendix 1 and 3. The data from Råö 1994-1995 are not included in the figures in this report. These data has earlier been discussed in the "Status report" (Brorström-Lundén et al., 2003).

4.1.1 PAH and PCB

The measured air concentrations of PAHs (sum12) and PCBs (sum 7) at Råö and Pallas during the sampling occasions 1996-2005 are summarised in Figure 5. The data are illustrated using "box whisker plots". The boundary of the box indicates the 25th and 75th percentiles and the line within the box marks the median. Whiskers (error bars) above and below the box indicate the 90th and 10th percentiles. In addition, the average (red line) and outlying points are shown.

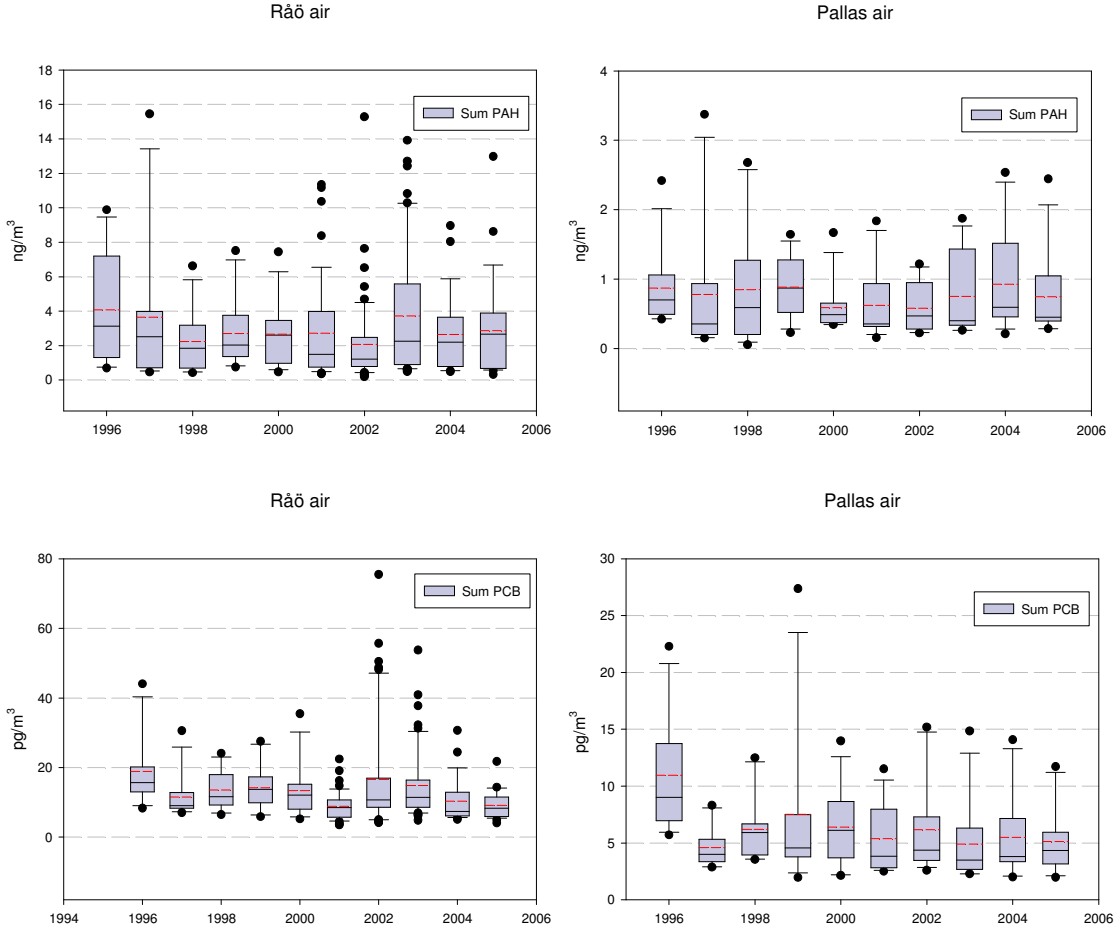


Figure 5. The atmospheric concentrations of PAHs and PCBs at Råö and Pallas (different scales).

Brorström-Lundén (1994) showed that the atmospheric concentrations of PAHs and PCBs at the Swedish West Coast declined between 1989 and 1994. Since 1996 the concentrations of PAHs and PCBs at both Råö and Pallas seem to have levelled off and now remain on a fairly constant level (Figure 5). The PAH as well as the PCB concentrations were lower in the north of Finland compared to the Swedish West Coast, e.g. the average concentrations of PCBs at Råö were about a factor of two higher compared to Pallas. The variability in concentrations between different sampling occasions was more pronounced for PAHs than for PCBs, especially at Råö. Two substances, benzo(a)pyrene and PCB153 were selected to illustrate this variability during 2005 at the two sites, see Figure 6

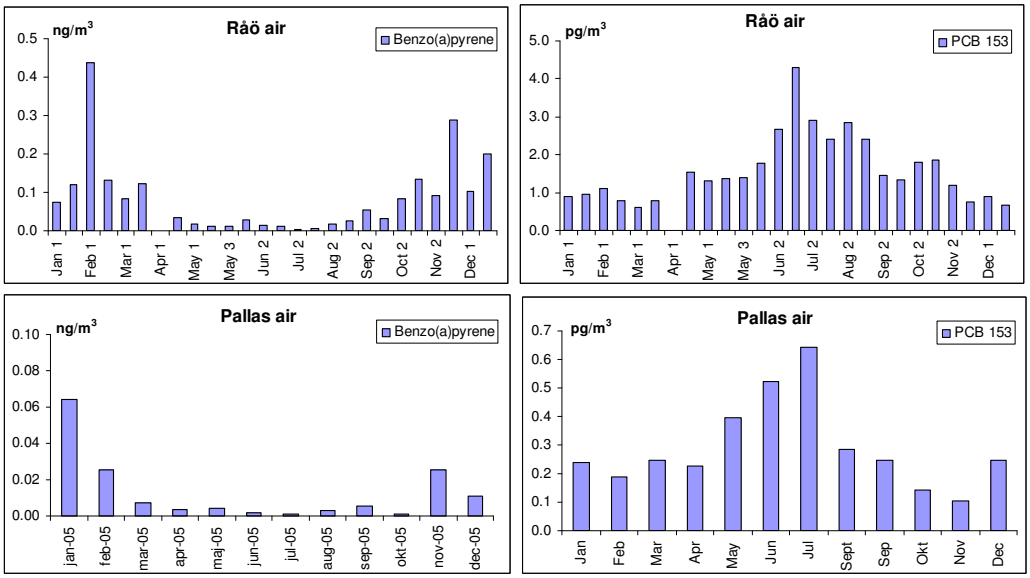


Figure 6. Atmospheric concentrations of Benzo(a)pyrene and PCB 153 at Råö and Pallas, 2005.

A seasonal variation in the atmospheric concentrations of benzo(a)pyrene was observed at both sampling sites, with the highest concentrations during the winter. The elevated concentration of benzo(a)pyrene in February (0.44 μg/m^3), was measured during a period (a few days) when air parcels originated from Eastern Europe. It is possible that also local sources contributed to the observed concentration, as increased combustion of garden waste has been observed, as a result of the strong storm (Gudrun) in January 2005.

The atmospheric concentrations of PCBs also varied with season. The increased atmospheric concentrations of PCBs during the warmer periods indicate that re-emission of PCBs back to the atmosphere takes place. This observation supports the theory of global fractionation of POPs. . The yearly variation in the ambient air temperatures is given in Figure 2 and Figure 3.

Profiles, e.g. the distribution of individual PAHs and PCBs in air during 2005 are presented in Figure 7. No clear difference between the two sampling stations was observed for the PAH profiles. For the PCBs, the distribution of the compounds was different at the Swedish West Coast than in northern Finland. A larger share of the high-chlorinated (non-volatile) PCBs was detected at Råö compared to Pallas. At Pallas, 83% of the PCBs (sum 7) in air consisted of the more volatile low-chlorinated components PCB 28, 52 and 101.

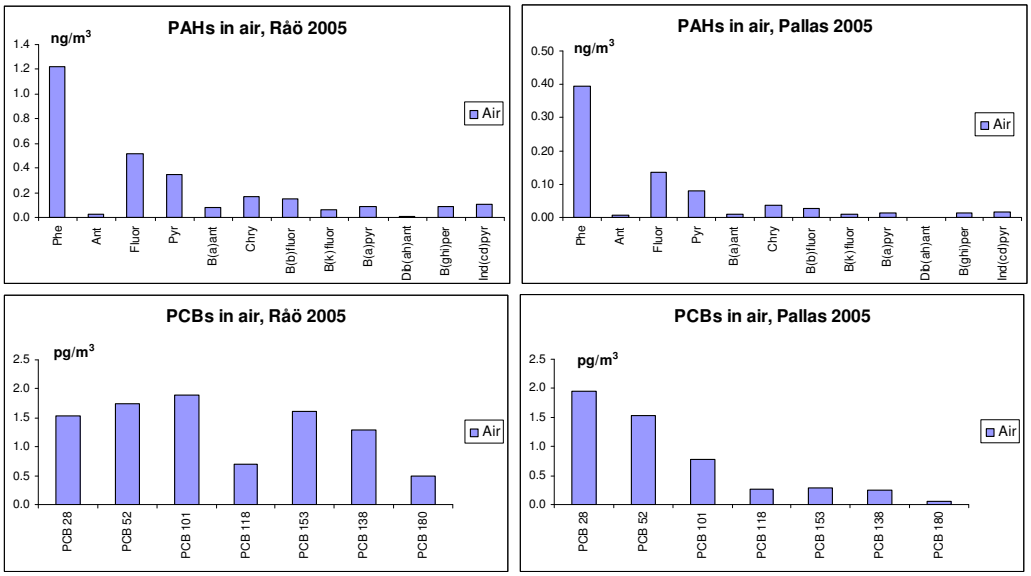


Figure 7. PAH and PCB profiles at Råö and Pallas 2005 (different scales in PAH figures)

4.1.2 Pesticides

The yearly average atmospheric concentrations of hexachlorocyclohexanes (HCHs) between 1996-2005 at Råö and Pallas are shown in Figure 8.

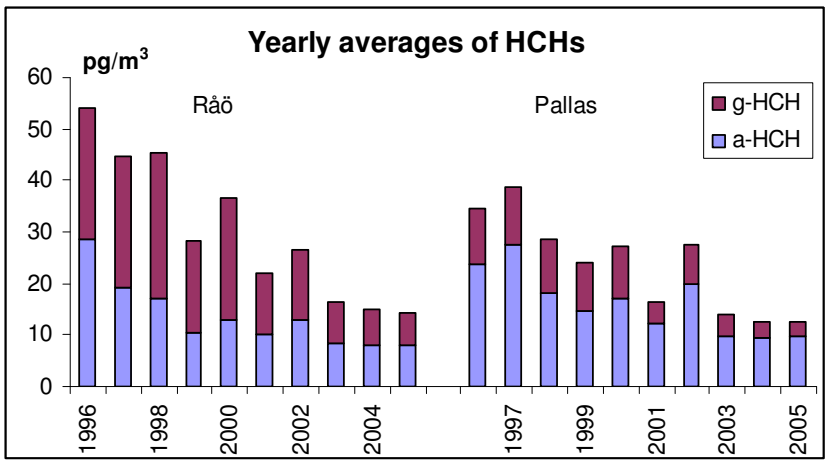


Figure 8. Atmospheric concentrations of HCHs at Råö and Pallas (annual average concentrations).

The atmospheric concentrations of both alpha- and gamma-HCH have decreased significantly at the Swedish West Coast since 1989-1990 when the concentrations of these compounds varied between 14-550 pg/m^3 (alpha-HCH) and 9-1100 pg/m^3 (gamma-HCH) (Brorström-Lundén et al., 1994). The concentrations have also decreased in the time period 1996-2005 (Figure 8). The concentrations of especially gamma-HCH were higher at the Swedish West Coast than in northern Finland while alpha-HCH was found in the same levels at Råö and Pallas.

Like for PCBs, there was an increase in the air concentrations during the spring and summer periods also for the HCHs, see Appendix 1 and 3. This is likely to be due to re-emission or connected to consumption of the pesticides in southern countries, reaching Scandinavia via long-range atmospheric transport.

In Figure 9 the yearly atmospheric average concentrations of DDD, DDT and DDE at Råö and Pallas are presented. The concentrations of ΣDDTs were considerably higher at Råö compared to Pallas. DDE was the dominating substance at both Råö and Pallas, with the exception of 1996, when DDD dominated at Råö. No clear time trend was observed for the last 10 years of measurements. The concentration of ΣDDTs varied between 3 and 6 pg/m^3 at the Swedish West Coast and between 1 and 2 pg/m^3 in northern Finland.

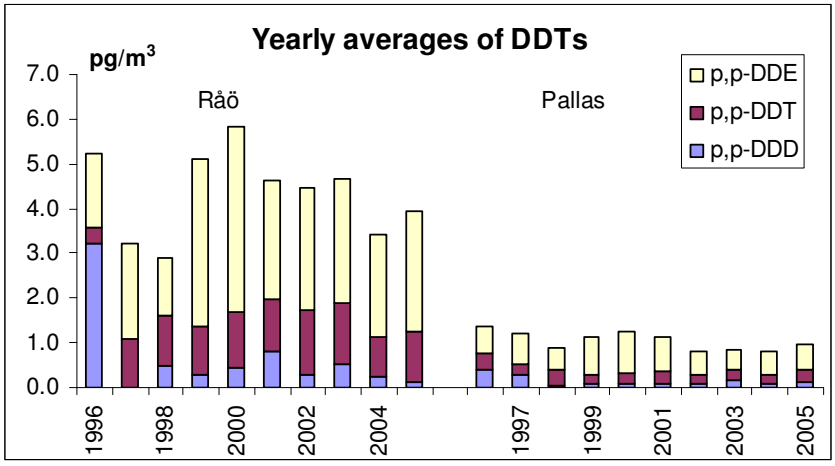


Figure 9. The yearly averages of DDTs in air at Råö and Pallas.

The yearly concentrations of gamma-chlordane, alpha-chlordane and transnonachlor at Råö and Pallas are summarised in Figure 10. Like alpha-HCH there were no significant differences in atmospheric concentrations of chlordanes between the Swedish West Coast and northern Finland and, except for 1996 at Råö. The chlordane levels have remained in the same order of magnitude during the last 10 years of measurements.

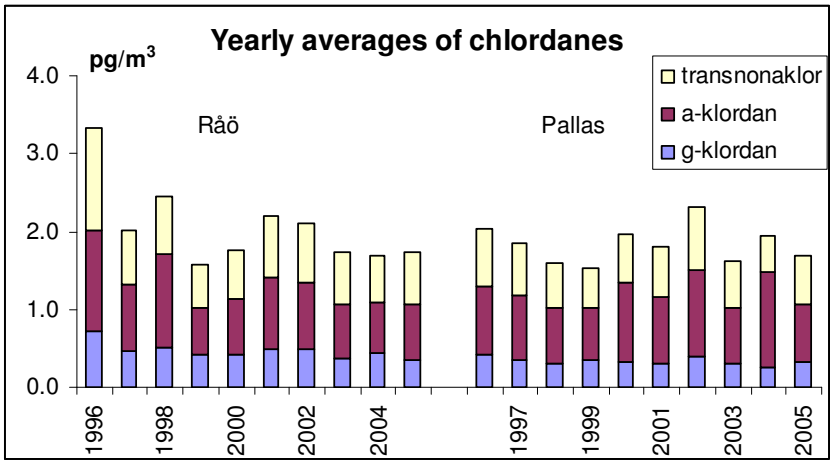


Figure 10. The annual average atmospheric concentrations of chlordanes at Råö and Pallas.

4.1.3 PBDE

The yearly atmospheric concentrations of PBDE-47, 99 and 100 at Råö and Pallas are shown in Figure 11 and the concentration found for the different sampling occasions are given in Figure 12.

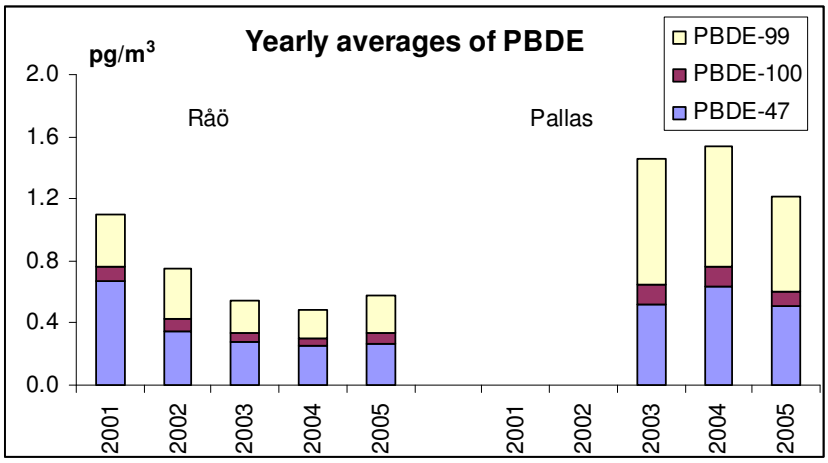
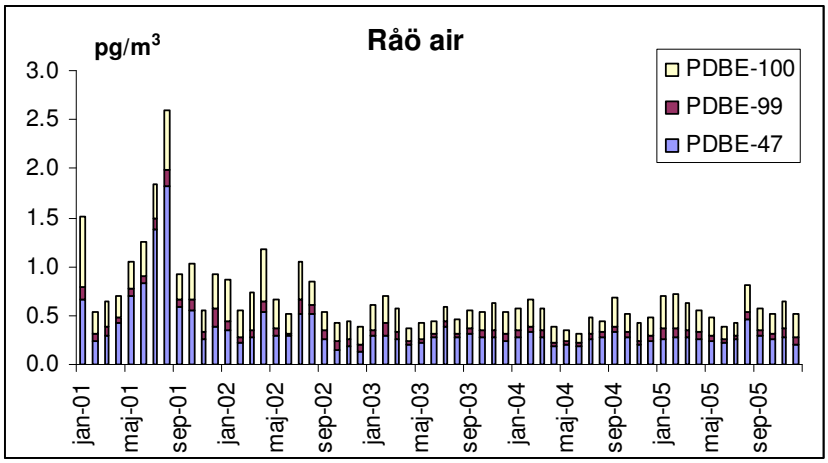


Figure 11. Atmospheric concentrations of PBDEs at Råö and Pallas.

The highest atmospheric concentrations of PBDEs at the Swedish West Coast occurred during 2001, the fist year of measurement, when also a seasonal variation was found. The measurements during 2001 were performed at the Rörvik station, where construction of buildings etc took place in the vicinity of the station, which may have affected the PBDE concentrations. This may also reflect a shift in use pattern of PBDEs, as pentaBDE was identified as a phase-out substance, and later banned in Aug 2004. The concentrations of PBDEs, from 2002 to 2005 varied between 0.6 and 0.8 pg/m^3 and no seasonal variation occurred.

The atmospheric concentrations of PBDEs were higher at Pallas compared to Råö and also in contrast to Råö a seasonal variation was observed (Figure 12), with highest concentrations in the summer. This was more obvious in 2004 and 2005 than in 2003. During the three years of measurements at Pallas, the concentration of PBDEs in ambient air varied between 1.2 and 1.6 pg/m^3.



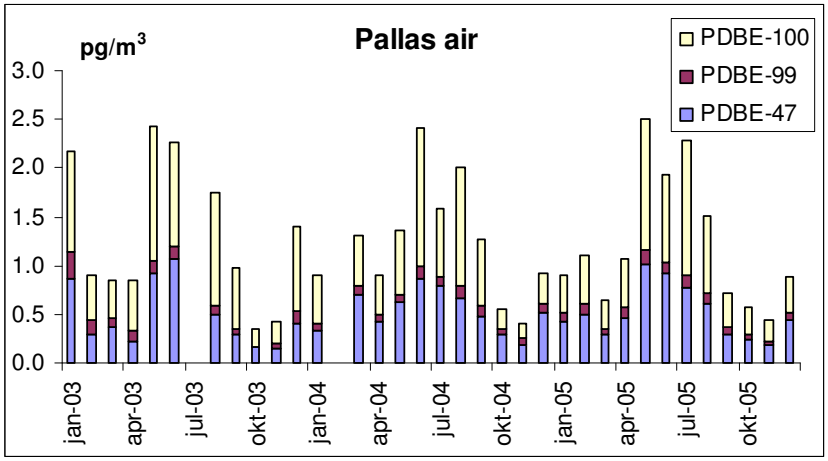


Figure 12. Seasonal variation of atmospheric concentration of BDE at Råö and Pallas

The weekly sampling periods make it difficult to use trajectories to explain increased concentrations. Thus no clear correlation between the transport of air parcels and high concentration of POPs in air at Pallas could be found. On a number of occasions, increased concentrations of PBDEs were found to coincide with air parcels originating from the Eastern Europe.

4.1.4 Endosulfan

During 2004, endosulfan was included in the Swedish screening programme and air was sampled both at Råö and Pallas (Palm Cousins et al., 2005). The results of these screening measurements are given here, in order to illustrate the importance of co-ordination of these measurements in the monitoring work. The concentration of endosulfan in air at Råö and Pallas is shown in Figure 13.

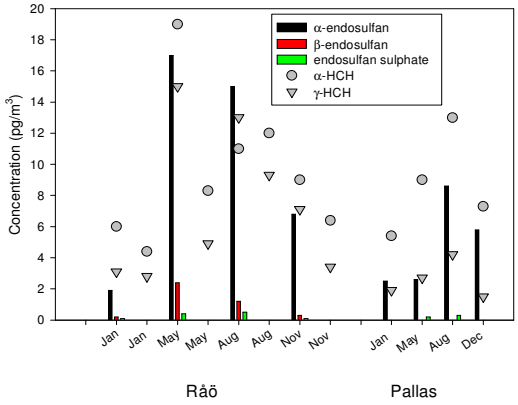


Figure 13. Concentration of α-endosulfan, β-endosulfan and endosulfan sulphate,as compared to the HCH-levels at Råö and Pallas. At Råö, the endosulfan samples were taken over a whole month, whereas the HCH samples are two-week-samples, thus the graph shows two values for HCHs each month, but only one value for endosulfan (Figure from Palm Cousins et al., 2005).

The concentrations of α-endosulfan at Råö were in the same range as both α-HCH and γ-HCH and showed a similar pattern of seasonal variation with higher concentration during the spring - summer, which may reflect the use of pesticides (endosulfan) in southern Europe (Figure 13). The concentrations of α-endosulfan at Pallas were lower compared to Råö, especially during the spring and summer periods, but very similar to the measured concentrations of γ-HCH (Palm Cousins et al., 2005).

4.1.5 Atmospheric concentrations in a European perspective

Benzo(a)pyrene, PCB (Sum 7) and HCHs have been selected to illustrate the regional differences in atmospheric concentrations when comparing the results from Råö and Pallas with other European sampling sites (www.EMEP.int).

For B(a)P and PCB, Košetice in the Czech Republic in central Europe stands out, showing atmospheric concentrations about 5-10 times higher than observed levels at Scandinavian and Arctic sites. For HCH, the difference is less pronounced, emphasizing the strong influence of atmospheric long-range transport of these compounds. Among the northerly sampling stations in Scandinavia and the Arctic, no obvious south to north gradient was found for ΣPCB or ΣHCH during 2004. PAHs were only measured at some of the stations.

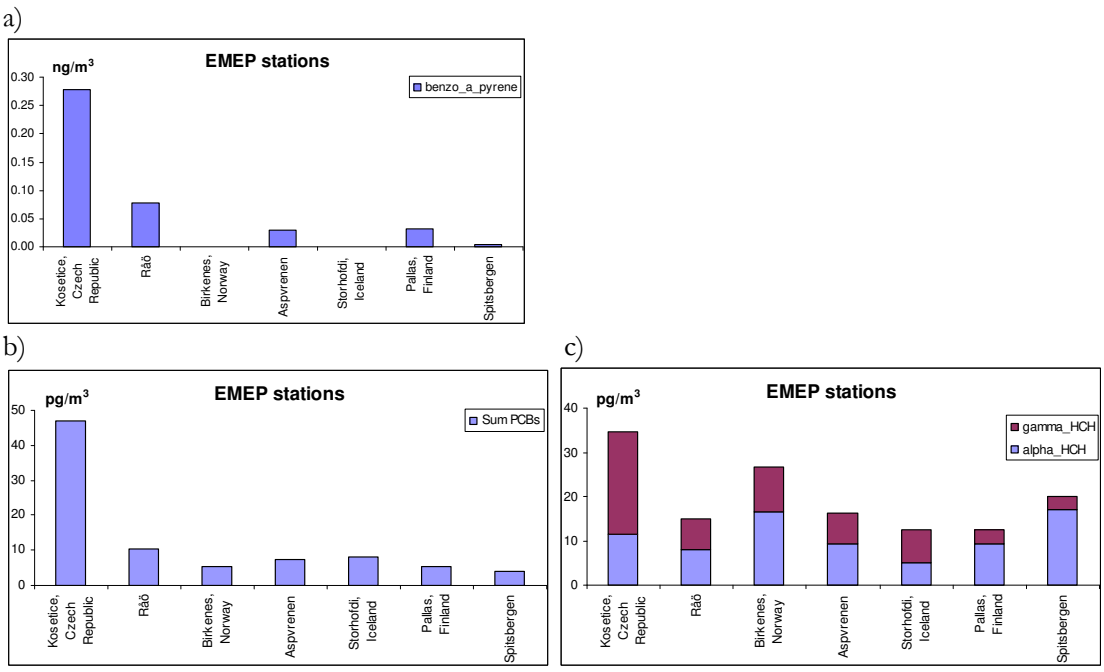


Figure 14. Concentration of a) benzo(a)pyrene, b) PCBs and c) HCHs in air at some EMEP stations as a yearly average for 2004 (www.emep.int).

4.2 Deposition fluxes

The yearly average deposition fluxes (1996-2005) of PAHs, PCBs, HCHs, chlordanes, DDTs and PBDEs measured at Råö and Pallas are presented in Appendix 2 and 4.

4.2.1 PAH and PCB

The deposition fluxes of sum (12) PAHs and sum (7) PCBs at Råö and Pallas are summarised and presented in Figure 15 using box-whisker plots. For explanations of the figures, see chapter 4.1.

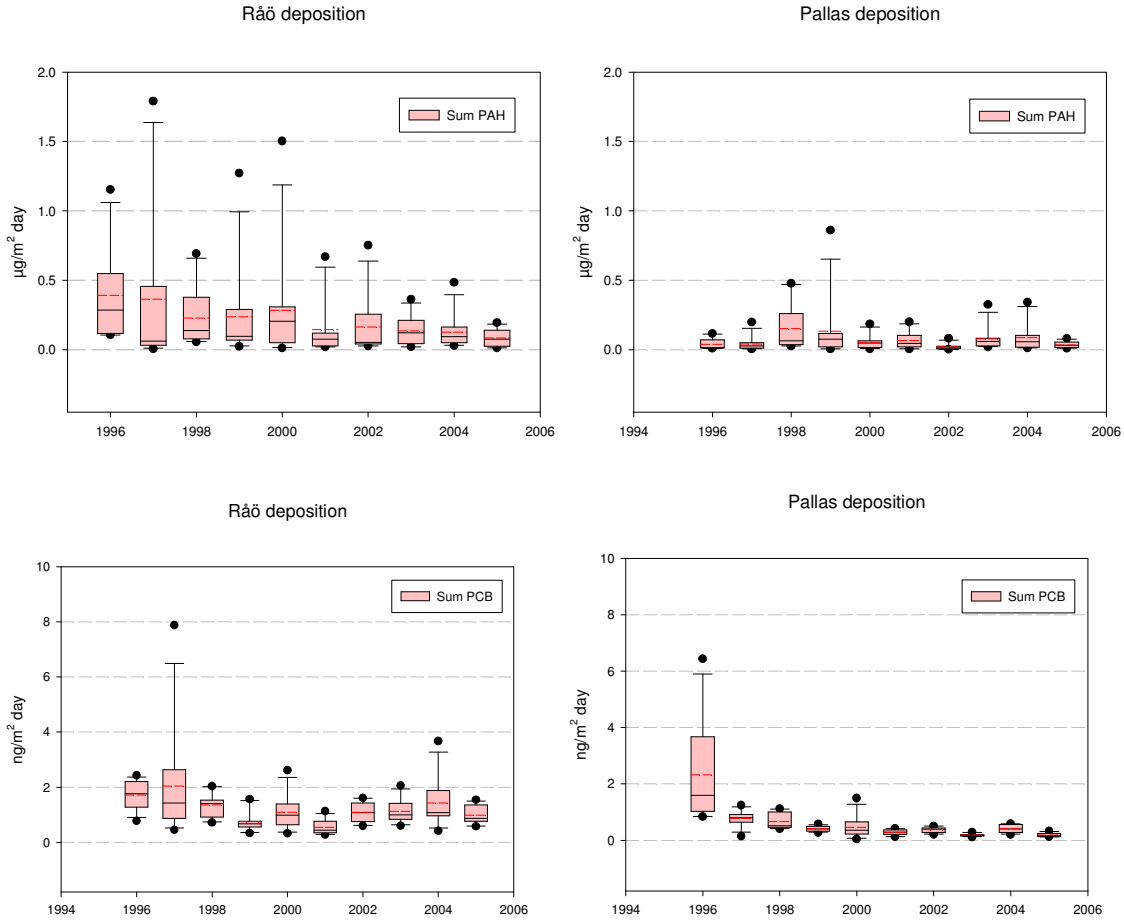


Figure 15. Deposition fluxes of PAHs and PCBs at Råö and Pallas between 1996 and 2005.

The deposition fluxes of PAH at Råö were generally slightly higher or in the same order of magnitude as in Pallas. The variation in measured PAH fluxes was larger at Råö than at Pallas, especially between 1996 and 2002. Also the deposition fluxes of PCBs and their variation was larger at Råö than Pallas. The yearly amounts of precipitation, which affect the deposition fluxes, are given in Figure 2 and Figure 3 for both Råö and Pallas.

The importance of atmospheric long-range transport of PAHs and PCBs to remote sites is evidenced by the occurrence of these pollutants in deposition samples also in Pallas.

In Figure 16, PAH and PCB profiles in deposition at Råö and Pallas 2005 are given. As in air (see Figure 7) there was no significant difference in the distribution of the specific PAH compounds between the two sampling stations. For the PCBs, the distribution of the compounds was different at Råö compared to Pallas. At Råö, there was higher percentage of the more high-chlorinated

compounds (74% PCB138-180), while the low-chlorinated compounds (PCB 52-118) constituted 55% of the total amount of PCBs in deposition at Pallas.

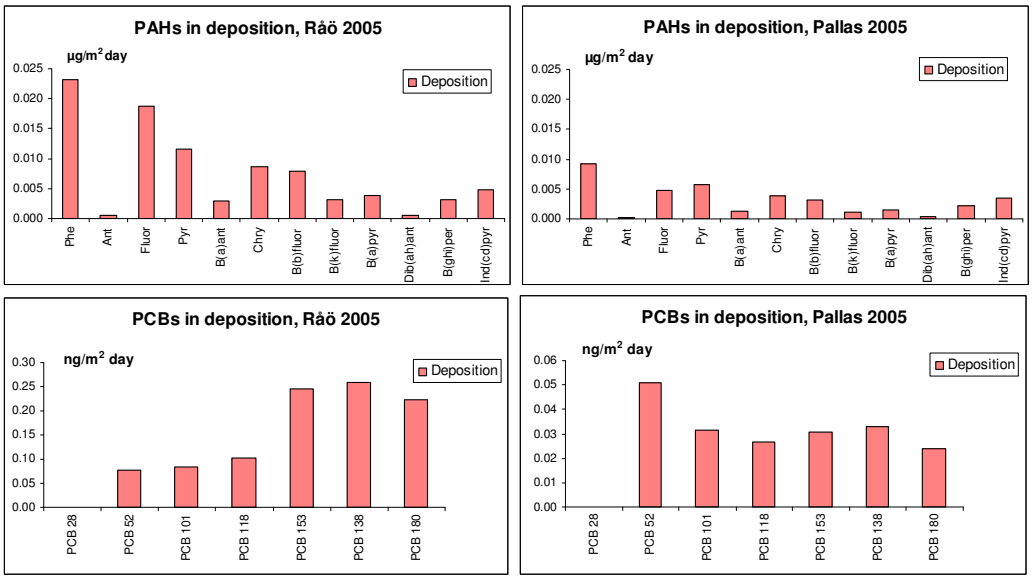


Figure 16. PAH and PCB profiles in deposition at Råö and Pallas 2005 (different scales in PCB figures).

A larger share of the more non-volatile PAHs and PCBs occurred in the deposition compared to the air (figure 7), which emphasizes the importance of particles for the deposition process of these POPs. There was no clear relationship between the concentration in air and the amounts in deposition, thus the seasonal patterns that were observed in concentration data were not observed for deposition. The highest deposition fluxes of PAHs and PCBs were observed in periods with high precipitation. Conclusively, most important for the deposition of PAHs and PCBs is particle deposition in connection with precipitation. This is in agreement with previous findings (Brorström-Lundén 1995).

4.2.2 Pesticides

The deposition fluxes of γ-HCH were higher at Råö compared to Pallas, whereas the deposition fluxes of α-HCH were similar in the south and north (Figure 17). At Råö, a slight decrease in deposition fluxes of HCHs was observed. It has not been stated whether this trend is statistically significant. The pattern is not obvious at Pallas, where the variation for the last seven years of measurements was higher than at Råö.

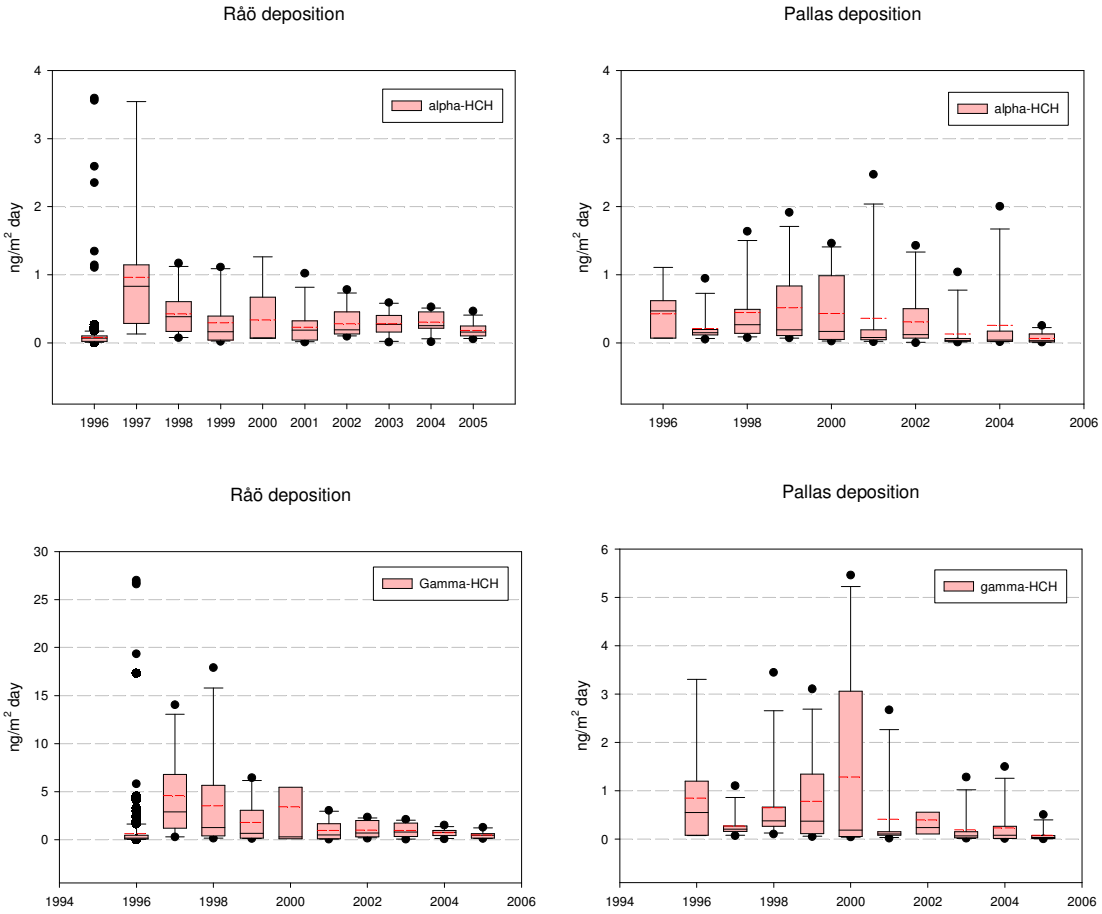


Figure 17. Deposition fluxes of HCHs, Råö and Pallas 1996-2005 (different scales for γ-HCH)

4.2.3 PBDE

The deposition of PBDEs was substantially higher in Pallas compared to Råö (Figure 18). No correlation between the deposition fluxes and the precipitation was found (see chapter 3).

The highest fluxes at the Swedish West Coast were measured during 2001, when the measurements were performed at the Rörvik station. During 2004 and 2005 the deposition fluxes of PBDEs at Råö were fairly constant (0.2-0.3 ng/m² day), with some higher peeks during the winter months. The highest deposition fluxes at Pallas were measured in the beginning of 2004 (4-4.5 ng/m² day).

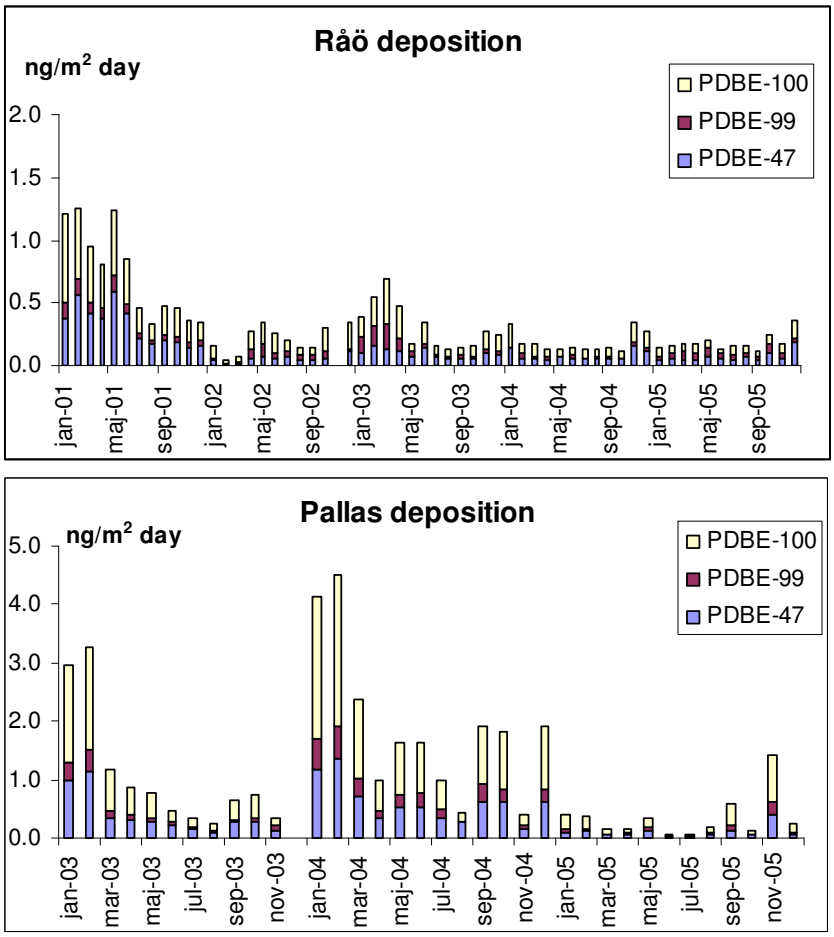


Figure 18. Deposition fluxes of PBDEs at Råö and Pallas

4.2.4 Annual deposition and impact on the aquatic environment

The importance of atmospheric deposition as a source of contaminants to water bodies is of interest, in particular within the work of the EU Water Frame Directive, where sources and their various impacts on water quality are central. In order to investigate the charge of PAHs, PCBs gamma-HCH and PBDEs to national water bodies, annual deposition fluxes for PAHs, PCBs, HCHs, chlordanes, DDTs and PBDEs at Råö and Pallas (1994-2005) were estimated and are summarised in Table 1. Adopting the annual deposition fluxes for 2005 and assuming that the fluxes measured at Råö are relevant for the entire western water district (Västerhavet; surface area 73 988 km^2), and that the Pallas fluxes are relevant for the northern water district (Bottenviken; surface area 154 702 km^2), this would yield total annual depositions according to Table 2.

Table 1. The annual deposition fluxes of POP measured at Råö and Pallas

Station	Year	PAH (sum11) ug/m^2 year	PCB (sum 7) ng/m^2 year	a-HCH ng/m^2 year	g-HCH ng/m^2 year	Chlordanes ng/m^2 year	DDTs ng/m^2 year	PBDE (47,99,100) ng/m^2 year
Råö	1994	110	840	560	1900			
Råö	1995	103	480	220	1200			
Råö	1996	140	630	490	3500	29	220	
Råö	1997	130	730	350	1700	14	210	
Råö	1998	84	470	260	1500	48	210	
Råö	1999	88	270	106	650	68	140	
Råö	2000	103	400	320	1200	64	240	
Råö	2001	53	200	84	500	20	120	
Råö	2002	60	400	101	370	8.7	110	70
Råö	2003	50	409	102	350	11	150	110
Råö	2004	45	520	110	260	14	108	76
Råö	2005	33	360	70	190	9.2	99	61
Pallas	1996	15	840	220	350	22	100	
Pallas	1997	15	280	77	99	32	45	
Pallas	1998	55	250	160	240	33	140	
Pallas	1999	49	150	190	280	17	63	
Pallas	2000	20	170	160	470	30	33	
Pallas	2001	24	104	130	150	31	14	
Pallas	2002	7.7	140	93	97	22	31	
Pallas	2003	29	69	47	70	12	34	890
Pallas	2004	47	170	86	78	15	44	700
Pallas	2005	13	72	24	28	6.5	17	130

Table 2. Estimated annual deposition (kg) of various POPs in the Northern and western water districts, based on data from the year 2005

Water District	PAH (sum11)	PCB (sum 7)	α-HCH	γ-HCH	Chlordanes	ΣDDTs	PBDE (47,99,100)
Western (Västerhavet)	2400	27	5.2	14	0.68	7.3	4.5
Northern (Bottenviken)	2000	11	3.7	4.3	1.0	2.6	20

As evident from Table 2, the annual amounts of PAH, PCB, HCH, chlordanes, DDTs and PBDEs deposited to national drainage basins are estimated to be in the size of kilograms to tonnes per year, indicating that the atmosphere can be regarded as a potentially important source of these contaminants to the aquatic environment.

5 Conclusions

The atmospheric concentrations of PAHs, PCBs and HCHs at Råö and Pallas seem to have levelled off since about 1996 and remain on a fairly constant level.

The concentrations of PAH and PCBs were higher in the south of Scandinavia than in the north, whereas the concentrations of α-HCH were of similar order of magnitude at the two sites. The atmospheric concentrations of PBDE were higher at Pallas compared to Råö.

The deposition measurements showed that all the analysed POPs occurred in the deposition both at Råö and Pallas.

The importance of long-range transport for the occurrence of POPs at remote sites, Pallas, is evidenced by both by the measurements in air and deposition.

Estimates of annual deposition in different water districts show that atmospheric deposition may be important source of these contaminants to water bodies.

The data in this report will be further evaluated.

6 Acknowledgements

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Appendix 1. Yearly averages (1994-2005) at Råö in air

Rörvik/Råö												
Year	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**
Air												
PAH	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**	**ng/m^3**
Phenanthrene	1.2	2.5	1.6	1.5	0.88	1.3	1.2	1.2	0.91	1.5	1.2	1.2
Anthracene	0.044	0.18	0.040	0.068	0.023	0.025	0.026	0.034	0.016	0.024	0.020	0.023
Fluoranthene	0.76	1.0	0.82	0.76	0.44	0.46	0.43	0.51	0.34	0.65	0.47	0.52
Pyrene	0.21	1.2	0.48	0.46	0.28	0.30	0.31	0.33	0.23	0.41	0.32	0.35
Benso(a)anthracer	0.065	0.14	0.085	0.10	0.098	0.060	0.12	0.075	0.052	0.11	0.070	0.080
Chrysene	0.15	0.30	0.19	0.20	0.12	0.13	0.14	0.15	0.11	0.21	0.15	0.17
Benso(b)fluoranthe	0.17	0.21	0.32	0.19	0.12	0.11	0.14	0.12	0.077	0.20	0.11	0.16
Benso(k)fluoranthe	0.074	0.077	0.13	0.089	0.056	0.055	0.069	0.054	0.039	0.083	0.051	0.066
Benso(a)pyrene	0.072	0.14	0.09	0.10	0.069	0.074	0.11	0.072	0.063	0.12	0.078	0.085
Dibenso(ah)antracene						0.017	0.038	0.010	0.011	0.023	0.015	0.011
Benso(ghi)perylen	0.079	0.18	0.072	0.15	0.092	0.085	0.13	0.077	0.069	0.13	0.082	0.085
Indeno(cd)pyrene	0.075	0.22	0.21	0.017	0.11	0.11	0.17	0.10	0.078	0.12	0.086	0.109
Sum	**2.9**	**6.1**	**4.1**	**3.6**	**2.2**	**2.7**	**2.7**	**2.7**	**2.0**	**3.6**	**2.6**	**2.8**
PCB	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**
PCB 28	5.7	5.7	5.3	2.3	2.5	3.1	2.5	1.4	2.5	1.8	1.5	1.5
PCB 52	4.1	5.7	4.2	2.7	3.5	3.6	3.5	2.1	3.4	3.5	1.9	1.7
PCB 101	5.7	5.4	3.5	2.3	2.8	2.9	2.8	1.8	3.2	3.1	2.2	1.9
PCB 118	1.8	1.7	1.1	0.79	0.98	0.96	0.90	0.59	1.1	1.0	0.72	0.70
PCB 153	5.8	4.4	2.1	1.5	1.6	1.6	1.7	1.3	2.9	2.5	1.8	1.6
PCB 138	5.7	4.0	2.0	1.4	1.5	1.5	1.5	1.2	2.6	2.2	1.6	1.3
PCB180	2.4	1.8	0.77	0.66	0.57	0.54	0.56	0.47	1.0	0.85	0.61	0.50
Sum	**31**	**29**	**19**	**12**	**14**	**14**	**13**	**8.9**	**17**	**15**	**10**	**9.3**
Pesticides and Bi	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**	**pg/m^3**
a-HCH	32	25	29	19	17	10	13	10	13	8.3	8.0	7.9
g-HCH	51	25	26	25	28	18	24	12	14	8.0	7.1	6.4
Sum	**84**	**50**	**54**	**45**	**45**	**28**	**38**	**22**	**27**	**16**	**15**	**14**
g-klordan			0.72	0.45	0.51	0.41	0.41	0.48	0.50	0.37	0.43	0.36
a-klordan			1.3	0.87	1.2	0.61	0.72	0.93	0.84	0.69	0.65	0.71
transnonaklor			1.3	0.69	0.72	0.55	0.62	0.79	0.8	0.67	0.60	0.66
Sum			**3.3**	**2.0**	**2.4**	**1.6**	**1.7**	**2.2**	**2.1**	**1.7**	**1.7**	**1.7**
p,p-DDD			3.2	<0.13	0.50	0.28	0.43	0.82	0.29	0.54	0.25	0.13
p,p-DDT			0.36	1.1	1.1	1.1	1.3	1.1	1.4	1.4	0.89	1.1
p,p-DDE			1.7	2.1	1.3	3.7	4.1	2.7	2.8	2.8	2.3	2.7
Sum			**5.0**	**3.2**	**2.6**	**5.0**	**5.5**	**4.5**	**4.4**	**4.7**	**3.4**	**3.9**
PBDE-47								0.67	0.34	0.28	0.25	0.27
PBDE-100								0.10	0.090	0.06	0.056	0.068
PBDE-99								0.33	0.32	0.20	0.18	0.24
Sum								**1.1**	**0.76**	**0.54**	**0.48**	**0.58**

Appendix 2. Yearly averages (1994-2005) at Råö in deposition

Rörvik/Råö												
Year	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**
Deposition												
PAH	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day	ug/m^{2}day
Phenanthrene	0.048	0.041	0.097	0.058	0.043	0.040	0.034	0.022	0.024	0.024	0.021	0.023
Anthracene	0.004	0.003	0.031	0.005	0.003	0.002	0.003	0.002	0.001	0.001	0.001	0.001
Fluoranthene	0.071	0.059	0.11	0.11	0.049	0.049	0.048	0.027	0.033	0.029	0.029	0.019
Pyrene	0.038	0.047	0.053	0.048	0.027	0.035	0.037	0.018	0.022	0.019	0.018	0.012
Benso(a)anthracene	0.011	0.013	0.015	0.024	0.025	0.016	0.024	0.006	0.009	0.005	0.006	0.003
Chrysene	0.035	0.035	0.024	0.041	0.020	0.025	0.032	0.019	0.018	0.014	0.014	0.009
Benso(b)fluoranthene	0.035	0.027	0.021	0.042	0.018	0.023	0.036	0.017	0.017	0.014	0.010	0.008
Benso(k)fluoranthene	0.013	0.009	0.009	0.019	0.009	0.012	0.018	0.007	0.007	0.006	0.004	0.003
Benso(a)pyrene	0.013	0.011	0.011	0.019	0.010	0.012	0.022	0.007	0.009	0.007	0.006	0.004
Dibenso(ah)antracene						0.005	0.006	0.002	0.002	0.001	0.001	0.001
Benso(ghi)perylen	0.022	0.020	0.017	0.024	0.022	0.019	0.032	0.008	0.009	0.008	0.008	0.003
Indeno(cd)pyrene	0.036	0.021	0.024	0.043	0.019	0.031	0.041	0.012	0.012	0.010	0.007	0.005
Sum	**0.30**	**0.25**	**0.39**	**0.36**	**0.23**	**0.24**	**0.28**	**0.14**	**0.16**	**0.14**	**0.13**	**0.089**
PCB	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day
PCB 28	0.35	0.30	0.46	0.18	0.24	0.059	0.17	0.047	0.12	<0.01	<0.01	<0.01
PCB 52	0.35	0.19	0.27	0.45	0.19	0.14	0.11	0.066	0.11	<0.01	0.17	0.076
PCB 101	0.37	0.15	0.16	0.33	0.21	0.10	0.14	0.069	0.11	0.15	0.11	0.085
PCB 118	0.12	0.10	0.090	0.25	0.11	0.079	0.10	0.039	0.080	0.115	0.10	0.103
PCB 153	0.49	0.25	0.28	0.40	0.26	0.19	0.26	0.12	0.22	0.30	0.37	0.24
PCB 138	0.51	0.31	0.29	0.49	0.29	0.19	0.23	0.12	0.25	0.33	0.41	0.26
PCB180	0.35	0.23	0.23	0.32	0.21	0.16	0.15	0.086	0.19	0.23	0.28	0.22
Sum	**2.3**	**1.3**	**1.7**	**2.0**	**1.4**	**0.75**	**1.1**	**0.54**	**1.1**	**1.1**	**1.4**	**0.98**
Pesticides and BFR		ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day	ng/m^{2}day
a-HCH		0.60	1.3	0.96	0.42	0.29	0.87	0.23	0.28	0.28	0.30	0.19
g-HCH		3.3	9.5	4.6	3.5	1.8	3.4	0.97	0.999	0.96	0.72	0.53
Sum		**3.9**	**11**	**5.4**	**4.2**	**2.1**	**3.7**	**1.2**	**1.3**	**1.2**	**1.0**	**0.72**
g-klordan			0.041	0.044	0.071	0.125	0.064	0.012	0.016	0.020	0.032	0.008
a-klordan			0.048	0.031	0.059	0.124	0.043	0.037	0.009	<0.01	<0.01	0.011
transnonaklor			0.040	<0.03	0.050	0.053	0.13	0.011	0.012	0.013	0.008	0.007
Sum			**0.080**	**0.038**	**0.13**	**0.19**	**0.18**	**0.056**	**0.024**	**0.031**	**0.040**	**0.026**
p,p-DDD			0.23	0.093	0.24	0.249	0.25	0.071	0.030	0.044	0.03	0.014
p,p-DDT			0.18	0.47	0.38	0.22	0.46	0.14	0.13	0.13	0.14	0.12
p,p-DDE			0.31	0.26	0.14		0.40	0.19	0.15	0.24	0.13	0.15
Sum			**0.61**	**0.58**	**0.6**	**0.39**	**0.67**	**0.33**	**0.31**	**0.41**	**0.30**	**0.28**
PBDE-47								0.32	0.05	0.10	0.077	0.059
PBDE-100								0.070	0.041	0.066	0.047	0.046
PBDE-99								0.34	0.11	0.15	0.090	0.061
Sum								**0.73**	**0.21**	**0.31**	**0.21**	**0.17**

Appendix 3. Yearly averages (1996-2005) at Pallas in air

Pallas										
Year	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**
Air										
PAH	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**	**ng/m³**
Phenanthrene	0.43	0.35	0.37	0.45	0.32	0.28	0.27	0.35	0.47	0.40
Anthracene	0.006	0.005	0.018	0.005	0.005	0.003	0.004	0.006	0.006	0.005
Fluoranthene	0.17	0.15	0.16	0.15	0.10	0.11	0.09	0.13	0.16	0.14
Pyrene	0.10	0.080	0.074	0.076	0.049	0.064	0.052	0.075	0.085	0.080
Benso(a)anthracene	0.010	0.026	0.087	0.041	0.046	0.025	0.037	0.033	0.032	0.011
Chrysene	0.037	0.062	0.038	0.043	0.028	0.042	0.056	0.052	0.08	0.037
Benso(b)fluoranthene	0.038	0.047	0.049	0.041	0.018	0.033	0.019	0.028	0.040	0.026
Benso(k)fluoranthene	0.013	0.019	0.018	0.021	0.005	0.012	0.007	0.010	0.015	0.010
Benso(a)pyrene	0.015	0.026	0.020	0.014	0.007	0.019	0.025	0.023	0.032	0.013
Dibenso(ah)antracene				0.003	0.003	0.003	0.004	0.003	0.003	0.001
Benso(ghi)perylen	0.021	0.029	0.028	0.024	0.017	0.013	0.014	0.017	0.025	0.014
Indeno(cd)pyrene	0.036	0.020	0.045	0.030	0.021	0.026	0.022	0.026	0.011	0.018
Sum	**0.87**	**0.78**	**0.85**	**0.88**	**0.59**	**0.62**	**0.58**	**0.75**	**1.0**	**0.75**
PCB	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**
PCB 28	3.2	1.5	1.8	2.5	2.0	1.7	2.1	1.5	1.9	2.0
PCB 52	1.9	1.4	1.9	2.4	2.3	1.9	2.3	1.7	1.9	1.5
PCB 101	1.5	0.74	1.1	1.1	0.96	0.85	0.87	0.81	0.74	0.77
PCB 118	0.52	0.28	0.40	0.50	0.31	0.25	0.21	0.20	0.24	0.27
PCB 153	1.5	0.31	0.49	0.46	0.39	0.31	0.36	0.37	0.33	0.29
PCB 138	1.4	0.32	0.43	0.41	0.36	0.28	0.27	0.24	0.27	0.24
PCB180	0.87	0.11	0.14	0.14	0.10	0.074	0.21	0.074	0.060	0.061
Sum	**11**	**4.6**	**6.2**	**7.5**	**6.4**	**5.4**	**6.2**	**4.9**	**5.5**	**5.1**
Pesticides and BFR	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**	**pg/m³**
a-HCH	24	28	18	15	17	12	20	9.8	9.5	10
g-HCH	11	11	10	10	10	4.5	7.6	4.0	3.0	2.7
Sum	**34**	**39**	**29**	**24**	**27**	**17**	**28**	**14**	**12**	**13**
g-klordan	0.42	0.35	0.31	0.35	0.32	0.29	0.40	0.30	0.24	0.31
a-klordan	0.87	0.83	0.71	0.68	1.0	0.87	1.1	0.71	1.2	0.74
transnonaklor	0.73	0.67	0.58	0.51	0.62	0.65	0.82	0.60	0.46	0.63
Sum	**2.0**	**1.8**	**1.6**	**1.5**	**2.0**	**1.8**	**2.3**	**1.6**	**2.0**	**1.7**
p,p-DDD	0.42	0.29	0.048	0.066	0.074	0.077	0.080	0.17	0.091	0.137
p,p-DDT	0.35	0.22	0.36	0.23	0.26	0.30	0.20	0.22	0.19	0.25
p,p-DDE	0.58	0.68	0.47	0.84	0.93	0.76	0.54	0.47	0.53	0.58
Sum	**1.2**	**0.89**	**0.86**	**1.1**	**1.2**	**1.0**	**0.65**	**0.85**	**0.79**	**0.93**
PBDE 47								0.52	0.63	0.51
PBDE-100								0.12	0.13	0.09
PBDE-99								0.82	0.78	0.61
Sum								**1.5**	**1.5**	**1.2**

Appendix 4. Yearly averages (1996-2005) at Pallas in deposition

Pallas										
Year	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**
Deposition										
PAH	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**	**ug/m^{2}day**
Phenanthrene	0.008	0.010	0.041	0.018	0.011	0.012	0.004	0.016	0.019	0.009
Anthracene	0.001	0.001	0.014	0.009	0.002	0.001	0.0002	0.002	0.001	0.0002
Fluoranthene	0.015	0.016	0.028	0.035	0.012	0.009	0.005	0.019	0.026	0.005
Pyrene	0.006	0.005	0.016	0.022	0.007	0.007	0.004	0.013	0.019	0.006
Benso(a)anthracene	0.003	0.004	0.028	0.015	0.014	0.012	0.002	0.004	0.005	0.001
Chrysene	0.005	0.007	0.009	0.015	0.004	0.011	0.003	0.007	0.012	0.004
Benso(b)fluoranthene	0.004	0.007	0.015	0.018	0.007	0.006	0.002	0.005	0.013	0.003
Benso(k)fluoranthene	0.004	0.003	0.008	0.009	0.003	0.004	0.002	0.002	0.005	0.001
Benso(a)pyrene	0.005	0.003	0.011	0.013	0.004	0.006	0.003	0.002	0.007	0.001
Dibenso(ah)antracene				0.004	0.002	0.001	0.000	0.001	0.001	0.000
Benso(ghi)perylen	0.005	0.004	0.011	0.012	0.007	0.004	0.004	0.003	0.010	0.002
Indeno(cd)pyrene	0.007	0.006	0.022	0.024	0.012	0.008	0.005	0.007	0.009	0.003
Sum	**0.039**	**0.041**	**0.15**	**0.13**	**0.053**	**0.067**	**0.021**	**0.079**	**0.13**	**0.035**
PCB	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**
PCB 28	0.40	0.090	0.13	0.048	0.14	0.049	0.087	<0.01	<0.01	<0.01
PCB 52	0.22	0.23	0.21	0.073	0.17	0.098	0.10	<0.01	0.073	0.051
PCB 101	0.28	0.10	0.092	0.045	0.055	0.034	0.070	0.050	0.080	0.032
PCB 118	0.13	0.058	0.049	0.049	0.024	0.019	0.031	0.038	0.070	0.027
PCB 153	0.46	0.13	0.14	0.071	0.066	0.033	0.047	0.065	0.138	0.031
PCB 138	0.48	0.15	0.13	0.075	0.050	0.035	0.034	0.037	0.074	0.033
PCB180	0.37	0.10	0.11	0.052	0.028	0.021	0.028	0.033	0.049	0.024
Sum	**2.3**	**0.78**	**0.67**	**0.41**	**0.46**	**0.28**	**0.38**	**0.19**	**0.46**	**0.20**
Pesticides and BFR	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**	**ng/m^{2}day**
a-HCH	0.60	0.21	0.45	0.52	0.43	0.36	0.26	0.13	0.24	0.065
g-HCH	0.95	0.27	0.65	0.78	1.3	0.41	0.27	0.19	0.21	0.078
Sum	**1.4**	**0.48**	**1.1**	**1.3**	**1.7**	**0.77**	**0.52**	**0.32**	**0.45**	**0.14**
g-klordan	0.032	0.050	0.042	0.026	0.067	0.026	0.013	0.021	0.032	0.005
a-klordan	0.033	0.049	0.033	0.024	0.030	0.075	0.007	0.029	0.011	0.009
transnonaklor	0.036	0.047	0.037	0.028	0.019	0.026	0.042	0.018	0.012	0.004
Sum klord	**0.060**	**0.089**	**0.091**	**0.046**	**0.082**	**0.086**	**0.061**	**0.034**	**0.042**	**0.018**
p,p-DDD	0.069	0.071	0.24	0.038	0.030	0.061	0.017	0.030	0.024	0.017
p,p-DDT	0.22	0.083	0.13	0.10	0.040	0.17	0.05	0.028	0.044	0.009
p,p-DDE	0.14	0.11	0.11	0.076	0.066	0.040	0.031	0.041	0.064	0.025
Sum	**0.28**	**0.13**	**0.40**	**0.18**	**0.090**	**0.040**	**0.086**	**0.094**	**0.12**	**0.047**
PBDE 47								0.75	0.60	0.10
PBDE-100								0.32	0.27	0.05
PBDE-99								1.4	1.05	0.19
Sum								**2.4**	**1.9**	**0.34**