

Using passive air samplers to assess local sources versus long range atmospheric transport of POPs.

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Abstract

Passive air samplers (PAS) are cost-efficient tools suitable for spatial mapping of atmospheric concentrations of persistent organic pollutants (POPs). The objective of this study was to use PAS (*i*) to determine atmospheric concentrations of selected POPs in Norwegian coastal zones with consumption advisories on seafood ($N=22$), and (*ii*) to evaluate a simple nested monitoring approach to assess the relative influence of local vs. long-range atmospheric transport (LRAT) at coastal sites. The latter was facilitated by comparison with data from a coordinated European-wide campaign which was using an identical sampling and analytical approach. Air concentrations were calculated based on the loss of performance reference compounds (PRCs), and results are presented for selected polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), dichlorodiphenyltrichloroethanes (DDTs) and chlordanes. Air concentrations of PCBs were generally highest at sites within larger cities and up to about an order of magnitude higher than anticipated on the basis of LRAT alone. The distribution of PAHs and HCB occasionally showed elevated concentrations at coastal sites with ongoing or former industrial activity, while an urban site was significantly influenced by banned insecticides (technical DDT and lindane). Coastal sites were also elevated in α -HCH beyond the anticipated LRAT contribution, which we attribute to volatilization from the sea. We conclude that a simple nested PAS monitoring approach provides useful information for screening efforts aiming to assess both atmospheric

burdens as well as the relative significance of local sources in controlling these burdens at sites in contaminated areas.

1. Introduction

Humans may be significantly exposed to various persistent organic pollutants (POPs) through consumption of fish and seafood from aquatic environments e.g. ^{1, 2}. Any attempts to mitigate further human exposure through this pathway without changing dietary habits would ultimately call for knowledge about sources and pathways of contaminants that are affecting concentrations in fish. Several studies have indicated that the atmosphere constitutes an important pathway for which POPs enter and exchange with aquatic ecosystems. Examples include PCBs in the Great Lakes ³ and the Inner Oslofjord ⁴ as well as HCHs ⁵ and dioxins (PCDD/Fs) ⁶⁻⁷ in the Baltic Sea. This calls for further studies aiming to identify the main sources affecting atmospheric concentrations close to contaminated aquatic environments where high levels of POPs of concern. Relevant cases in Norway are various coastal zones and fjords for which consumption advisories on fish and seafood exist ⁸ due to elevated concentrations of PCBs, PCDD/Fs and/or PAHs ⁹.

A key feature of POPs is their ability to undergo LRAT from global source regions to remote areas ¹⁰⁻¹¹. As the population density in Norway is relatively low compared to other parts of Europe, LRAT is therefore expected to exhibit a significant influence on concentrations of POPs in air for Norway as a whole. Thus, LRAT could also play a significant role in controlling atmospheric burdens along the Norwegian coast ¹²⁻¹⁴. Indeed, past studies on the occurrence of POPs in air within Norway have mainly been carried out in the context of LRAT and most data are available for background sites e.g. ^{15, 16}. However, the atmospheric burden in coastal zones with consumption advisories on seafood typically coincides with areas with a higher population density in Norway. Concentrations of POPs in air in these coastal zones may therefore additionally be influenced and potentially enhanced by atmospheric emissions of POPs from Local Sources (LS). Examples are emissions of PAHs and PCDD/Fs from e.g. industrial activities ¹⁷⁻¹⁸, and PCBs from e.g. building materials ¹⁹⁻²². Additionally, as our contaminated coastal zones are located within an interesting urban-remote transect along an expected pollution

gradient from more densely populated areas of Europe towards the Norwegian Arctic e.g. ^{15, 16}, this makes the study region an interesting area for studying LS vs. LRAT in controlling concentrations of POPs in air. Furthermore, specific topographic features in some coastal zones (e.g. narrow fjords surrounded by steep mountains) may cause local emissions to be “trapped” due to limited atmospheric circulation ²³, leading to locally elevated air concentrations.

Taken together, the atmospheric burden of POPs in contaminated coastal zones of Norway may thus in part be controlled by LS and in part by LRAT. So far, no study has been carried out to assess the relative importance of these two sources within these coastal zones. Yet, being able to discriminate between these two sources is of immediate relevance for the assessment of potential control strategies, including relevant monitoring efforts in support of the Stockholm Convention on POPs (SC). The relative importance of the two could furthermore be anticipated to differ, dependent on the contaminant in question. As the key goal of the SC is to protect human health and the environment from these substances by reducing or eliminating releases to the environment, it becomes important to carry out further studies to help assess whether POPs are actually becoming reduced or eliminated, or whether there are sources which remain active. The Global Monitoring Plan (GMP) for POPs specifically request comparable monitoring data to support evaluations whether the goal of the SC is reached, and highlights the use of passive air samplers for spatial and temporal trends assessment ²⁴.

The objective of this study was to carry out a passive air sampling campaign (PAS) to (i) determine atmospheric concentrations of selected POPs in Norwegian coastal zones and fjords where consumption advisories on seafood exists, and (ii) to gain insights into the relative importance of LRAT and LS for individual sites. The latter was made possible as this campaign was coordinated in time with a comprehensive PAS campaign across European background sites ²⁵, which in turn facilitates empirical estimates of the anticipated background concentration as attributed to LRAT alone. Finally, we end by discussing the merits and limitations of the PAS approach to assess LRAT versus LS in contaminated areas as well as remaining knowledge gaps.

2. Material and methods

2.1 Theory

The air samples were collected by passive air samplers (PAS) using polyurethane (PUF) disks. We refer to the literature for a detailed account of this method and its application e.g. ²⁶⁻³⁰. In brief, the PUF disk has a high ability to sorb semi-volatile organic compounds in air. During exposure, the chemicals will accumulate in the sampling material, and the amount on the PUF disk is equivalent to the rate of uptake minus the rate of loss. The uptake is airside controlled and a function of the planar area (A_{PUF}) to the disks, the mass transfer coefficient (k_A) and the concentration in air (C_A). The uptake is initially linear, but will reach equilibrium in time ²⁶. Dependent on the octanol-air partition coefficient (K_{OA}) of individual compounds, the duration of the linear phase will vary and the more volatile compounds (i.e. low K_{OA}) will reach equilibrium faster than the less volatile compounds ²⁹. The PUF disk is placed between two stainless steel metal domes ³¹ which protects the sampling media from precipitation, particle deposition, sunlight and shields against wind speed effects (e.g. Tuduri et al ³²).

2.2 Sampling

Air samples were collected using PAS deployed along the Norwegian coast for three months during late summer 2006, see Figure 1. The 22 coastal sites were selected on the basis of advisories on the consumption of seafood caused by elevated levels of POPs ⁸, see Table S1 in Supporting Information (SI). Additionally, two samplers were deployed in a few coastal sites (Kristiansand, Bergen, Trondheim, Tromsø, Narvik and Bergen) to evaluate spatial variability within contaminated coastal zones. All PAS samplers were deployed at least 1,5 meters above the ground in close vicinity to the sea. Areas close to major roads and industrial activity were avoided, while preference was given to parks and private properties in an attempt to aim for representative “background” concentrations in each coastal zone.

During the same period, a coordinated PAS campaign was carried out at 86 background sites across 34 European countries ²⁵. Site details for the Norwegian ($N=5$) and Swedish ($N=7$) background sites which were included in the European campaign and used to assess contribution from LRAT in this study (see 2.5) are additionally listed in Table S1 (see also Fig. 1).

2.3 Sample preparation, clean-up and analysis

Details involving solvents used, sample preparation, clean-up and analysis are previously described in Halse et al ²⁵, and only a brief summary is presented here. The PUF disks were pre-cleaned, spiked with performance reference compounds (PRCs) ³³, wrapped in double layer of alumina foil and stored in zip-lock bags prior to deployment. After exposure, the air samples together with field and method blanks were added a mixture of internal standards and Soxhlet extracted using *n*-hexane as a solvent. The air samples were analyzed for polychlorinated biphenyls (PCBs) (PCB-28,-52,-101,-118,-138,-153,-180), polycyclic aromatic hydrocarbons (PAHs) (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)pyrene, benz(a)anthracene, chrysene), hexachlorocyclohexanes (HCHs) (α ,- β - and γ -HCH), hexachlorobenzene (HCB), dichlorodiphenyltrichloroethanes (DDTs) (*p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *p,p'*-DDT) and chlordanes (trans-chlordane, cis-chlordane, trans-nonachlor, cis-nonachlor). The extracts were divided into two aliquots prior to the clean up procedures. One aliquot for the determination of the acid stable compounds (Σ_7 PCBs, Σ_3 HCHs, Σ_4 DDTs, HCB, Σ_4 chlordanes) was treated with sulphuric acid followed by a fractionation using a silica column. The other aliquot for the determination of the PAHs was cleaned using a deactivated silica column. After the clean-up procedures, both aliquots were reduced to ~50 μ l by a gentle stream of nitrogen and recovery standards were added. PCBs, HCHs, DDTs, HCB and PRCs (*d*₆ γ -HCH and selected PCBs, see SI) were analyzed using gas chromatography coupled to high resolution mass spectrometry in an electron impact (EI) mode (GC/HRMS). The PAHs were analyzed using gas chromatography coupled to low resolution mass spectrometry (GC/LRMS), while the chlordanes were analyzed using low resolution gas chromatography coupled to mass spectrometry in an electron capture negative ion mode (GC/ENCI-MS) ²⁵.

2.4 Deriving air concentrations

An estimate of the sampled volume of air is needed in order to back-calculate air concentrations for individual compounds. For these calculations, information on PUF characteristics, air temperature, measured loss of PRCs and their temperature-dependent K_{OA} values were used to

calculate site specific sampling rates e.g. ^{26, 28, 33-34}. The air temperatures used for these estimations were derived from the Norwegian Meteorological Institute ³⁵, except for Birkenes and Hurdal where temperatures are measured by NILU. A detailed description of the estimation of air concentrations is given by Halse et al ²⁵ and references therein. The site-specific sampling rates and number of PRCs characterized by a sufficient loss for each site (>40%) are presented in Table S1. For sites experiencing an insufficient loss of PRCs (<40%) ³⁴, the default sampling rate of 3.5 m³/day proposed by Harner et al ³⁶ was used for calculating the effective volume of air sampled. The estimated air concentration derived for these two sites should therefore be interpreted with caution.

2.5 Quality assurance and quality control (QA/QC)

For assessments of the quality assurance and quality control, the analytical procedures were monitored using NS/EN ISO/IEC 17025 accredited routines. For quantification, a calibration/quantification solution was injected for every fourth sample. A standard reference material (SRM 1588) from the National Institute of Standards and Technology (NIST), containing organic compounds in cod liver oil was analyzed routinely for PCBs, HCHs, DDTs, HCB and chlordanes. For PAHs, a dilution of a PAH mixture containing 16 compounds (NE 1378) from Promochem GmGH was analyzed for each run. To furthermore verify the quantification, the retention time for ¹²C-labeled compound should not be more than 3 seconds later than its corresponding ¹³C-labeled isomer. In addition, the 3:1 isotope ratio between ¹²C and ¹³C isomers must be within 20 % of the theoretical value for the two monitoring masses.

2.5.1. Field and method blanks

Field (*N*=5) and method blanks (*N*=5) consisting of pre-cleaned spiked PUF disks were extracted and analyzed in the same way as the exposed samples. Field blanks were prepared to discover possible contamination during general handling and transport, while the method blanks focus more on laboratory conditions (e.g. solvents, equipment, and adsorbents). The blank values were

converted into concentrations by use of averaged values for the sampling rate ($4.06 \text{ m}^3\text{day}^{-1}$), temperature ($14 \text{ }^\circ\text{C}$) and deployment time (92 days) from the deployed coastal samples.

2.5.2. Method detection limit (MDL)

The method detection limit (MDL) (Table 1) were calculated for the individual compounds as the average blank (field and method) concentration ($N=10$) plus 3 times the standard deviation (SD) (Table 1). The field and method blanks had similar values. When the target compound was not identified in the blanks, an instrumental detection limit (IDL, signal/noise. 3:1) divided by two, were used instead to estimate a MDL³⁴. A different approach was used for the PAHs, where the lowest measured range for air samples for the instrument was $0.01\text{-}0.02 \text{ ng/m}^3$. For individual PAHs only 0.01 ng/m^3 was used, while half of this measured value was used for samples below this value²⁵. When the target compound were absent in the exposed samples, $\frac{1}{2}$ MDL were used for statistical treatment. The concentrations were not blank corrected as the blanks had both low and stable values.

2.5.3. Recoveries

Recoveries were compiled for both the internal standards and for the PRCs, where the latter were provided from the field and method blanks as detailed in the SI (S1.1, S1.2) and Table S2. Quantification were based on the added ^{13}C or ^2D -labeled internal standards, hence the sample concentrations is automatically corrected for recovery.

2.5.4. Uncertainties

There are recognized uncertainties associated with procedures used to back-calculate air concentrations from PAS e.g.³⁷⁻³⁹ and as also discussed in Halse et al.²⁵. Different factors during the chemical analysis will furthermore influence the uncertainty in the air concentrations, e.g. sample clean-up, accuracy in the standards and instrumental conditions. The overall uncertainty associated with the chemical analysis was estimated to be $\pm 35 \%$ ²⁵. Internal and recovery

standards were used to compensate for possible loss during sample clean up; field blanks were furthermore included to evaluate possible contamination during transport and general handling. The method blanks were used to assess laboratory conditions. More information on the quantification of these uncertainties is provided in Halse et al. ²⁵.

2.6 Estimating the contribution from LS vs. LRAT

The availability of comparable and consistent data for European background air enables empirical estimates of the expected contribution attributed to LRAT alone. Specifically, the ratio (R) of observed concentrations at individual coastal sites divided by the anticipated “background” concentration attributed to LRAT alone provide insights into the significance of LS versus LRAT (Equation 1):

$$R = \frac{C(\text{coastal site})}{C(\text{background})} \quad 1)$$

Thus, $R > 1$ would indicate influence from LS; $R > 2$ would indicate predominance of LS, while $R \leq 1$ would indicate predominance of LRAT. The latter would also indicate that the LRAT contribution is overestimated for a given compound / compound group at a specific coastal site.

This approach thus requires an estimate of the background concentration attributed to LRAT alone. As both campaigns were carried out at the same time, temporal variability can be ignored. However, there is an expected and observed spatial variability in European background air ²⁵, which merit consideration. For this work, three scenarios for the background concentration were therefore explored: (i) a Norwegian background, defined as the median air concentration for the Norwegian background sites ($N=5$, Fig 1), (ii) a Nordic background, defined as the median air concentration for all background stations in two Nordic countries (Sweden and Norway) combined ($N=12$, Fig 1), and finally (iii) a European background, defined as the median air concentration from all European background sites ($N=86$) ²⁵. Consequently, three corresponding ratios were calculated: R_{NW} (Norwegian background), R_{ND} (Nordic background) and R_{EU} (Europe). Each of these background estimates have their own merits and limitations, which makes it interesting to explore the impact of different predicted LRAT contributions on the overall results. In brief, while the former two estimates have a more limited empirical basis ($N=5$

and 12) than the latter ($N=86$), the European estimate is considered less representative for the expected “true” background air concentrations in Norway. To further assess the spatial variability in estimated background air concentrations (LRAT contributions), we used the maximum (max) and minimum (min) observed background air concentration within both the Norwegian and Nordic data sets to derive an estimate of the uncertainty in R . This approach was not considered appropriate for the European dataset due to a much larger spatial variability²⁵, i.e. the higher concentrations observed across European background sites would typically be expected to significantly overestimate the true background in Norway. Finally, the analytical uncertainty previously estimated to be $\sim\pm 35\%$ ²⁵ was additionally incorporated by adding and subtracting 35% from the max and min background air concentrations for the three reference scenarios, respectively. Taken together, this resulted in nine different R s with median (default), max and min estimates for R_{NW} , R_{ND} and R_{EU} . Additional uncertainties associated with back-calculating air concentrations on the basis of PAS^{37, 39} were not considered as these errors were assumed to be of a more systematic rather than random character, e.g. as caused by uncertainties in K_{OA} . Finally, any data below MDL was omitted from this analysis.

3 Results and discussion

3.1 Introductory remarks

Table 1 shows the overall results in terms of calculated air concentrations for both exposed samples from the coastal sites and the blanks (average, median, range), while Tables S3a-S6a present results for individual compounds and compound groups at each coastal site. Three sites (Oslo, Ramsundet and Tromsø II) experienced an insufficient loss of all PRCs (<40%). The estimated air concentrations derived for these three sites critically rely on the default sampling rate and the results should therefore be interpreted with caution. Although the exact reason why these sites experienced an insufficient loss cannot be explained unequivocally, we note that the number of individual PRCs experiencing a sufficient loss tended to decrease towards the northern sites (Table S1), which could indicate that the volatility range of our selected PRCs were less suitable at sites experiencing colder temperatures. From theoretical considerations²⁶, we also caution that some of the more volatile substances (fluorene > HCB > α -HCH > phenanthrene \approx γ -HCH \approx anthracene > PCB-28 and to some extent PCB-52) may have started to

approach equilibrium during the 3-month deployment period as discussed in the SI (S 1.3), and as also can be seen from the effective air volumes derived for individual substances and compounds (Tables S3b-S6b).

PCB-28, 52 and 101, α - and γ -HCH, HCB, cis-Chlordane and trans-Nonachlor were detected in all samples. Among the PAHs, fluorene, phenanthrene, fluoranthene, pyrene together with chrysene were also detected in all samples. The spatial variability across the coastal sites is expressed by the ratio between maximum and minimum air concentrations (MMR) in Table 1. When the minimum concentration was below MDL, the MDL value was used when calculating the MMR, which implies that only a minimum estimate of spatial variability can be provided. The estimated influence of LS at each coastal site is explored by analyzing the ratios of observed concentrations at each coastal site, divided by the different scenarios for the estimated concentration attributed to LRAT alone (R_s ; Equation 1). Only results for selected substances and substance groups using the Norwegian background scenario (R_{NW}) are included in Figure 2. Additional results are included in the SI, Tables S7-S9, which compares and contrasts the predicted influence of LS for individual compounds and compound groups for each of the three background scenarios (R_{NW} (Table S7), R_{ND} (Table S8) and R_{EU} (Table S9)), including median (Tables S7a,S8a,S9a), maximum (Tables S7b,S8b,S9b) and minimum (Tables S7c,S8c,S9c) estimates. In Tables S7-S9, R_s based on concentrations at coastal sites below MDL are listed as NQ (not quantified) while R_s using background concentrations below MDL are tabulated as ND (not detected). Tables S10a-c likewise summarize the number of coastal sites for which the R_s are ≥ 2 , ≥ 1 and $>MDL$ for the 9 different background scenarios. An overview of the median background air concentrations for the three background scenarios is finally included in Table 2.

3.2 PCBs

The average concentration of Σ_7 PCBs was 21 pg/m^3 ($SD \pm 17 \text{ pg/m}^3$), with a range from 4.5 to 72 pg/m^3 (Table 1). The more abundant PCBs were PCB-52, -101, -28 and -153, which each contributed 29%, 24%, 22% and 12% to the average concentration of Σ_7 PCBs, respectively. MMRs for individual PCBs varied by about an order of magnitude or more, i.e. >9 (PCB-153 and PCB-180) to 21 for PCB-101. The highest concentrations of Σ_7 PCBs were found in Bergen I, Kristiansand II and Oslo with 72 , 59 and 51 pg/m^3 , respectively. These sites were all located

within major cities in Norway, while the lowest concentrations were observed in Narvik II, Ramsundet and Sunndalsøra with 5.8, 4.8 and 4.5 pg/m^3 , respectively (see Table S3a).

Figure 2a) shows the estimated contribution from LS (R_{NW}) for $\Sigma_7\text{PCBs}$ (see also Table S7a). R_{NW} falls between 1.5 and 13.6, which strongly suggests that LS likely contributes ($R_{\text{NW}} \geq 1$) or dominates ($R_{\text{NW}} \geq 2$) the observed air concentrations of PCBs at most coastal sites. The same pattern is also evident for R_{NWmax} (using maximum background concentrations) but the values are lower (0.6-5.3), indicating less influence from LS (Table S7b). Also note that R_{NWmin} is not included in Fig 2a) as a few congeners were below MDL at Norwegian background sites²⁵. The highest contribution from LS is estimated for sites located in larger cities, e.g. Bergen I (13.6: 5.3-ND), Kristiansand II (11.1; 4.3-ND) and Oslo (9.7: 3.7-ND) (Tables 7a-c).

It is noteworthy that $R_{\text{NW}} \geq 2$ at 21 out of 24, indicating that LS dominate the atmospheric burden (Table S10a). When considering R_{NWmax} (Table S7b), the number is reduced to 6 out of 24 for $R_{\text{NWmax}} > 2$ (Table S10b). Still, even when applying the more conservative approach with respect to possible influence from LS (Table S7b), as much as 15 sites experienced some influence from LS (i.e. $R_{\text{NWmax}} \geq 1$; Table S10b). When comparing results for the two parallels deployed at different locations, yet within the same coastal zone, such as Bergen, Tromsø and Kristiansand (Fig 1), it is also evident that our method and results are not applicable beyond a specific site alone. Thus, spatial variability in air concentrations of $\Sigma_7\text{PCBs}$ (and hence potential influence of LS) within a given coastal zone may differ significantly and any inferences made about the likely influence from LS in a specific coastal zone are restricted to the specific site where the sample was collected as well as when it was exposed.

When considering the Nordic background (R_{ND}) as our reference scenario (Table S8a-c), the overall results indicate a lesser influence from LS as the Nordic background concentrations are higher than the Norwegian background concentrations (Table 2). This ratio ranges from 0.9-8.2 and 0.2-1.5 for $\Sigma_7\text{PCBs}$ applying R_{ND} (Table S8a) and R_{NDmax} (Table S8b), respectively. Moreover, the number of sites which are predominantly controlled by LS ($R_{\text{ND}} \geq 2$) drops down to 13 out of 24 (Table S10a). In other words, only about half of the coastal sites are seen as elevated by LS when assessed in the context of Nordic background concentrations. However, when applying the European background scenario (Table 9a-c), R_{EU} falls below 1 for about half of the sites (Table S9a) and only five sites, Bergen I (4.7: 3.5-7.3), Kristiansand II (3.9: 2.9-5.9),

Oslo (3.4: 2.5-5.2), Tromsø I (2.9: 2.1-4.4) and Stavanger (2.7: 2.0-4.2) still stands out with $R_{EU} \geq 2$ (Table S10). Clearly, whenever R_{EU} is ≥ 2 (Table S9a-c), this strongly suggest that a significant influence of LS at a particular coastal site is likely. Hence, possible control strategies should emphasize local control measures if further reductions in atmospheric burdens of PCBs are to be achieved at these sites.

The median background air concentrations for Σ_7 PCBs for the Nordic and European scenarios are 1.7 and 2.9 times higher than the Norwegian background scenario, respectively (Table 2). The difference tends to increase with increasing chlorination, which is seen as a reflection of lighter PCBs being more prone to LRAT⁴⁰. Hence, the relative influence of LRAT at background sites may be higher for lighter PCBs compared with their heavier counterparts, which is in line with previous studies on the latitudinal distribution of PCBs in background air within Norway⁴¹. This may explain why coastal sites seem to be increasingly controlled by LS for heavier PCBs ($R_{NW} \geq 2$; Table S10a). Interestingly, the opposite trend across congeners is seen in comparison to the Nordic background (Table S10a), which may be seen as reflection of the Swedish background sites being more influenced by heavier PCBs (Table 2).

3.3 PAHs

The average concentration of Σ_8 PAHs was 13 ng/m³ (SD±16 ng/m³), and ranged from 1 to 84 ng/m³ (Table 1). The lighter PAHs, such as phenanthrene, fluorene and fluoranthene were more abundant and each contributed 49%, 31% and 11% on average, respectively. The MMR ranged from >1 (benzo(a)pyrene) to >108 (anthracene) which illustrates a marked spatial variability for some PAHs (Table 1). At decreasing temperatures, substances with a high K_{OA} like benzo(a)pyrene may be increasingly sorbed to atmospheric particles, for which the PAS method are not designed to capture. While the average air temperature at the coastal sites varied from 8 °C to 17 °C in our study (Table S1), Klanova et al.³⁷ previously predicted that benzo(a)pyrene will remain 80–90% particle-bound at 22 °C. In comparison, a calibration study in Toronto found that the particle-bound fraction of benzo(a)pyrene decreased from 77% at -4 C down to 23% at 16°C⁴². Sorption onto particles may thus help to explain why most samples were below detection limit for this substance (Table S4a). Sites with elevated air concentration of Σ_8 PAHs were found at Hommelvika, Fedafjorden and Sauda with 84, 33 and 19 ng/m³ respectively. Furthermore, Hommelvika had the highest concentrations of phenanthrene and fluorene with 41

and 32 ng/m³, respectively. The lowest concentrations of Σ_8 PAHs were found at Kragerø, Harstad, Ramsundet with 3.8, 3.5 and 1.2 ng/m³, in that order (Table S4a).

As the concentration of benzo(a)pyrene and sometimes also benz(a)anthracene often were below MDL, the following discussion will be restricted to Σ_6 PAHs. As shown in Fig 2b), R_{NW} for Σ_6 PAHs varied between 1.6 and 38.9. The highest influence from LS were found at Hommelvika, Fedafjorden and Sauda with 38.9 (5.9-ND), 15.3 (2.3-ND) and 8.9 (1.4-ND), respectively (Table S7a-c). Furthermore, 23 out of 26 sites had a $R_{NW} \geq 2$ (Table S10a), indicating LS to be more influential than LRAT at most sites. This number was reduced to 2 (out of 26) when instead applying R_{NWmax} (Table S10b). This implies that there are significant difficulties defining a reliable estimate of the atmospheric burden which may be attributed to LRAT alone for Σ_6 PAHs, attributed to spatial variability in background air concentrations. This, in turn, indicates that the “true” LRAT contribution may possibly overestimated using median background concentrations. Another interesting aspect is the striking variations within Σ_6 PAHs (Tables S7-S9) as also noted from the individual MMRs (Table 1). Thus, an assessment of the R_{NW} for individual PAHs (Table S7a) may provide clues about potential differences in sources. For example, both Fedafjorden and Saudafjorden have an industrial history and both exhibit elevated R_{NWS} for anthracene, fluoranthene, pyrene and chrysene in excess of R_{NW} for Σ_6 PAHs (Table S7a). This pattern is different to Hommelvika, a locality known to have been polluted by creosote in the past, which is seen as elevated by anthracene and fluorene in particular. For all these sites, PAHs are also among the pollutants which have caused measures on the consumption of seafood⁴³.

The median Nordic and European background air concentration for Σ_6 PAHs were 1.2 and 1.6 higher than the Norwegian median background (Table 2). Hence, the relative influence from LS does not decrease markedly dependent on the actual background reference. For R_{ND} , 23 out of 26 sites are still implicated as predominantly controlled by LS for Σ_6 PAHs ($R_{ND} \geq 2$), while this number is reduced to 17 for R_{EU} (Table S10a). Results from the coastal sites with duplicated PAS deployed indicate only a relatively minor difference in measured concentrations of Σ_6 PAHs, up to a factor of ~2 (Narvik I vs II).

3.4 HCHs

The average concentration for Σ_3 HCHs (α , β , γ) was 34 pg/m^3 ($\text{SD}\pm 22$ pg/m^3), and varied from 19 to 133 pg/m^3 (Table 1). The α - and γ -isomers were detected in all samples, and each contributed 46% and 53% on average, respectively. The β -isomer contributed with only $\sim 1\%$ on average. β -HCH is more water-soluble than the α - and γ -isomer⁴⁴, and hence more easily washed out from the atmosphere and thus considered less prone to LRAT. The spatial variability as expressed by MMR appears limited for Σ_3 HCHs, α -HCH and β -HCH with values of >7 , 2 and >4 , respectively. This is in striking contrast to γ -HCH which has a MMR of 26. Taken together, this indicates that there are coastal sites which possibly have been affected by past or contemporary lindane usage ($>99\%$ γ -HCH) as opposed to technical HCH which contains 55-80% α -HCH, 8-15% γ -HCH and 5-14% β -HCH⁴⁵. This result is somehow surprising given that lindane usage has been banned in Norway since 1992⁴⁵. The highest concentration of γ -HCH was found for Bergen I with 115 pg/m^3 . Other sites with elevated air concentrations of γ -HCH were Holmestrand and Stavanger with 37 pg/m^3 and 28 pg/m^3 , respectively (Table S5a).

Figure 2c) and 2d) display R_{NW} for individual HCHs (α , γ) evaluated against the Norwegian background (see also Tables S7a-c). R_{NW} for α -HCH reveals a relatively uniform pattern, with estimates ranging from 1.5-2.9 (Table S7a). The limited spatial variability in air concentrations of α -HCH in these coastal sites suggest that air concentrations are better explained by LRAT, rather than LS. At the same time, R_{NW} is ≥ 2 for 15 out of 27 sites, which in contrast suggest an influence from LS on the concentration of α -HCH measured at the coastal sites (Table S10a). Furthermore, R_{NWS} were ≥ 1 at all sites, indicating some influence of LS (Table S10a). When instead assessing R_{NWmax} (Table S7b), the number of sites ≥ 2 were reduced to zero (Table S10b). However, we believe this apparent inconsistency could have something to do with Norwegian background sites being located at some distance from the coast, which makes it appear that coastal sites have higher background air concentrations of α -HCH than inland stations. Dalla Valle et al⁴⁶ have previously discussed how net ocean-land transfers of POPs via the atmosphere may occur in some areas, because of sharp gradients in the storage capacities of terrestrial and aquatic environments. Thus, we hypothesize, given the fairly homogenous air concentrations observed at coastal sites, that our coastal air measurements to a large extent may reflect secondary re-emissions from the sea. Similar findings has previously been noted by e.g. Shen et al⁴⁷ in a study on HCHs in air across Northern America, whereby passive air sampling

combined with chiral analysis combined suggested that α -HCH was evaporating from the Labrador Current.

Figure 2d) similarly displays results for γ -HCH. R_{NW} for γ -HCH ranged from 1.0 to 26.6, and the highest influences from LS were found at Bergen I (26.6: 14.5-76.7), Holmestrand (8.5: 4.6-24.5) and Stavanger (6.6: 3.6-18.9) (Tables S7a-c). A predominant influence from LS was estimated for 20 out of 27 sites ($R_{NW} \geq 2$) (Table S10a). Furthermore, 10 out of 27 sites had $R_{NWmax} \geq 2$ and 21 out of 27 $R_{NWmax} \geq 1$ (Table S10b). These results clearly show some influence of LS, and more probably from lindane, rather than technical HCH. As γ -HCH appears less prone to re-volatilization compared to α -HCH⁵, it is not unlikely that the sample from Bergen I might have been affected by recent usage of lindane.

R_{NW} for the Σ_2 HCHs (α -HCH + γ -HCH) varied between 1.6-11.1. R_{NW} and R_{NWmax} were ≥ 2 for 20 out of 27 sites and 4 out of 27 sites, respectively (Tables S10a,b). The median Nordic and European background concentration for Σ_2 HCHs is 1.1 and 3.4 higher than the Norwegian background, respectively (Table 2). For R_{ND} , 17 out of 27 sites had ratios ≥ 2 for Σ_2 HCHs, and 27 out of 27 sites had ratios ≥ 1 , while only 1 out of 27 sites had a $R_{EU} \geq 2$ (Bergen I) (Table S10a).

3.5 HCB

This compound was detected in all samples and concentrations varied from 42 to 305 pg/m^3 (Table 1). The average concentration was 58 pg/m^3 with a relatively high SD ($\pm 50 \text{pg}/\text{m}^3$). At the same time, the low MMR (7) illustrates a limited spatial variability. HCB is a volatile compound with a high potential for long-range atmospheric transport⁴⁸. Fairly uniform concentrations were also seen in European background air²⁵ which in part is attributed to a significant influence of secondary emissions in controlling contemporary air concentrations⁴⁹. Still, elevated concentrations of HCB were found at Kristiansand II and, to a lesser extent Narvik II, with 305 pg/m^3 and 70 pg/m^3 , respectively (Table S5a). However, if Kristiansand II was excluded from the dataset, the MMR would be only 2.

R_{NW} varied from 1.0 to 7.3 (Table S7a). Only 1 out of 27 sites had a $R_{NW} \geq 2$ (Table S10a), indicating LS was dominating observed air concentration of HCB at Kristiansand II only (7.3: 4.8-12.0). In contrast, Kristiansand I experienced a R_{NW} of 1.2 only, which suggests that the

suspected source close to Kristiansand II did not significantly affect the atmospheric burden across the coastal zone as a whole. It is particularly noteworthy that Kristiansand II really stands out as primarily controlled by LS, even in a Nordic (Table S8; $R_{ND}=7.3$: 3.8-12.2) and European (Table S9; $R_{EU}=6.8$: 5.0-10.4) context. Further monitoring efforts to potentially identify or disconfirm the suspected hot-spot at Kristiansand II might therefore be desirable.

3.6 DDTs

The average concentration for Σ_4 DDTs was 6.5 pg/m^3 ($SD\pm 8.4 \text{ pg/m}^3$), and ranged from 1.8 pg/m^3 to 45 pg/m^3 (Table 1). p,p' -DDE was on average the major contributing isomer (44%), followed by p,p' -DDT (32%) and o,p' -DDT (22%) (Table 1). The technical mixture of DDTs contains up to 80-85 % of the p,p' -DDT isomer and only small amounts of the o,p' -DDT isomer (15-20 %) ⁵⁰. The MMRs for individual isomers varied from > 3 (p,p' -DDE) to > 86 (p,p' -DDT). The high variability in detected concentrations of DDT is noteworthy since usage of technical DDT has been severely restricted in Norway after 1980 and banned since 1989 ⁵¹. The highest concentration of Σ_4 DDTs was found at Bergen I, with 26 pg/m^3 , 11 pg/m^3 and 7.8 pg/m^3 for the p,p' -DDT, o,p' -DDT and p,p' -DDE isomers, respectively (Table S6a).

As median background air concentrations were below MDL for all four isomers in Norway and for two isomers in the Nordic region (Table 2), the discussion on relative influence from LS is restricted to Σ_2 DDTs (o,p' -DDT and p,p' -DDT) (Table S8a). For the sum of these two isomers, R_{ND} was ≥ 2 at 18 out of 22 sites (Table S10a). Sites being most influenced by LS were Bergen I (55.4: 5.1-N.D), Kristiansand II (8.7: 0.8-N.D) and Tønsberg (7.0: 0.6-N.D) (Table S8a-c). R_{ND} for individual isomers for Bergen I varied from 31 (3.6-N.D) for o,p' -DDT and 83.9 (6.3-ND) for p,p' -DDT. Also Kristiansand II had somewhat higher R_{ND} for the o,p' -DDT and p,p' -DDT isomers, with 7.4 (0.8-N.D.) and 10.3 (0.8-N.D.), respectively (Table S8a). In contrast, when applying R_{NDmax} , only Bergen I remained as mainly influenced by LS for Σ_2 DDTs (Table S8b). Thus, the large spatial variability in observed background air concentrations makes it difficult to firmly conclude whether most sites or merely one (Bergen I) is mainly controlled by LS. However, it is noteworthy that it is p,p' -DDT which appears elevated at Bergen I and some other sites, which may imply recent use of technical DDTs. Yet, a comparison of results from the two sites in Bergen reveals that only Bergen I is strongly elevated by this unknown local source(s) (Table S6a). Even when assessed in a European background context, the atmospheric

concentrations at Bergen I is seen as strikingly elevated. The corresponding R_{EU} remain ≥ 2 for Σ_2 DDTs (9.6: 7.1-14.7) *o,p'*-DDT (6.1: 4.6-9.5) and *p,p'*-DDT (12.6: 9.3-19.4) (Table S9a-c).

3.7 Chlordanes

The average concentration of Σ_4 Chlordanes was 3.8 pg/m^3 ($SD \pm 2.6 \text{ pg/m}^3$), and ranged from 1.1 pg/m^3 to 11 pg/m^3 (Table 1). Cis-Chlordane and trans-Nonachlor were the major components, each contributing 37% and 33% on average, respectively. The spatial variability expressed using MMRs varied from >91 for trans-chlordane to 8 for cis-Chlordane (Table 1).

Table S7a) shows R_{NW} for Σ_4 Chlordanes. R_{NW} varied from 0.5 (0.2-1.0) at Harstad to 5.7 (2.5-11.2) at Nøtterøy. 9 out of 22 sites had $R_{NW} \geq 2$ (predominantly influenced by LS) and 18 out of 22 sites had $R_{NW} \geq 1$ (some influence by LS) (Table S10a). When using R_{NWmax} , the number of sites ≥ 2 and ≥ 1 , is reduced to 2 and 5 sites, respectively (Table S10b) with only Nøtterøy (5.7: 2.5-11.2) and Kragerø (5.2: 2.2-10.2) still suspected of being mainly influenced by LS. The same overall conclusion applies when considering R_{ND} (Table S8) as the estimated Nordic and Norwegian background concentrations are virtually identical (Table 2).

When applying the European background scenario (Table S9a), the number of sites with $R_{EU} \geq 2$ were limited to 4 (Table S10a), i.e. Nøtterøy (3.8), Kragerø (3.5), Tønsberg (2.1) and Kristiansand II (2.0). Whilst all these sites are located in the south-eastern part of Norway (Fig 1) and thus closer to regions showing elevated air concentrations in mainland Europe²⁵, any inferences about LS should be interpreted with caution.

4 Summary and conclusions

This study has derived new data on air concentrations of selected POPs in contaminated coastal zones in Norway derived using passive air samplers. When compared and contrasted with different estimates of background air concentrations (as assumed attributed to LRAT alone), the results demonstrate that several legacy POPs are occasionally still elevated by local atmospheric emissions. However, the extent of influence from LS critically depends on the accuracy of the estimated contribution from LRAT alone. While the empirical basis is somehow limited for the Norwegian and Nordic background scenarios which makes it difficult to infer reliable

conclusions for substances exhibiting a high spatial variability in background air (e.g. PAHs), the European background estimate is considered more robust, due to the enhanced number of stations (N=86). While both the number of sites and extent of LS influence will vary dependent on the selection of actual background scenario, we believe a simple nested approach could be useful for screening exercises to assess potential influence from LS in other contaminated sites and areas.

It should also be kept in mind that the number of samplers deployed within each coastal zone was limited to one or two. Hence, more detailed follow-up studies in contaminated zones, e.g. by deploying a larger number of PAS may be warranted (*i*) to potentially confirm the presence of suspected hot-spots, and (*ii*) to identify more specific source areas and/or sources leading to locally elevated air concentrations, e.g. using a denser network of samplers, and (*iii*) to evaluate the impact of LS on contaminant burdens on the surroundings, including additional environmental surface media and contaminant exchange with the marine environment.

While the samples reported herein were all deployed during summer, we caution that there might be seasonal variability in both local emissions and influence from LRAT for some substances (e.g. PAHs from domestic heating). Hence, the relative influence of LS and LRAT may also vary seasonally. We finally caution that our method is not able to discriminate between primary anthropogenic emissions and secondary emissions from environmental compartments as contaminated in the past. An interesting finding in this context is the observation that coastal sites seem to be elevated in α -HCH in comparison to the estimated background concentrations, which we hypothesize could be due to volatilization from coastal waters.

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Figure 1: Map showing the locations for coastal sites (this study), and Norwegian and Nordic background sites ²⁵.



- 1) Oslo, 2) Holmestrand, 3) Tønsberg, 4) Nøtterøy, 5) Kragerø, 6) Arendal, 7a, b) Kristiansand I, II, 8) Farsund, 9) Fedafjorden, 10) Flekkefjord, 11) Stavanger, 12) Sauda, 13 a, b) Bergen I, II, 14) Sunndalsøra, 15 a, b) Trondhjem I, II, 16) Hommelvika, 17) Brønnøysund, 18) Mo i Rana, 19 a, b) Narvik I, II, 20) Ramsundet, 21) Harstad, 22 a, b) Tromsø I, II, I) Birkenes, II) Hurdal, III) Kårvatn, IV) Tustervatn, V) Karasjok, VI) Vavihill, VII) Råö, VIII) Hoburgen, IX) Aspvreten, X) Breckälän, XI) Videln, XII) Abisko

Figure 2(a-d): Estimated contribution from local sources, expressed as the ratio (R_{NW}) of measured air concentrations divided by the anticipated contribution from long-range atmospheric transport alone (Norwegian background scenario). The uncertainty in this ratio (R_{NWmax} , R_{NWmin}) is estimated as detailed in the text.

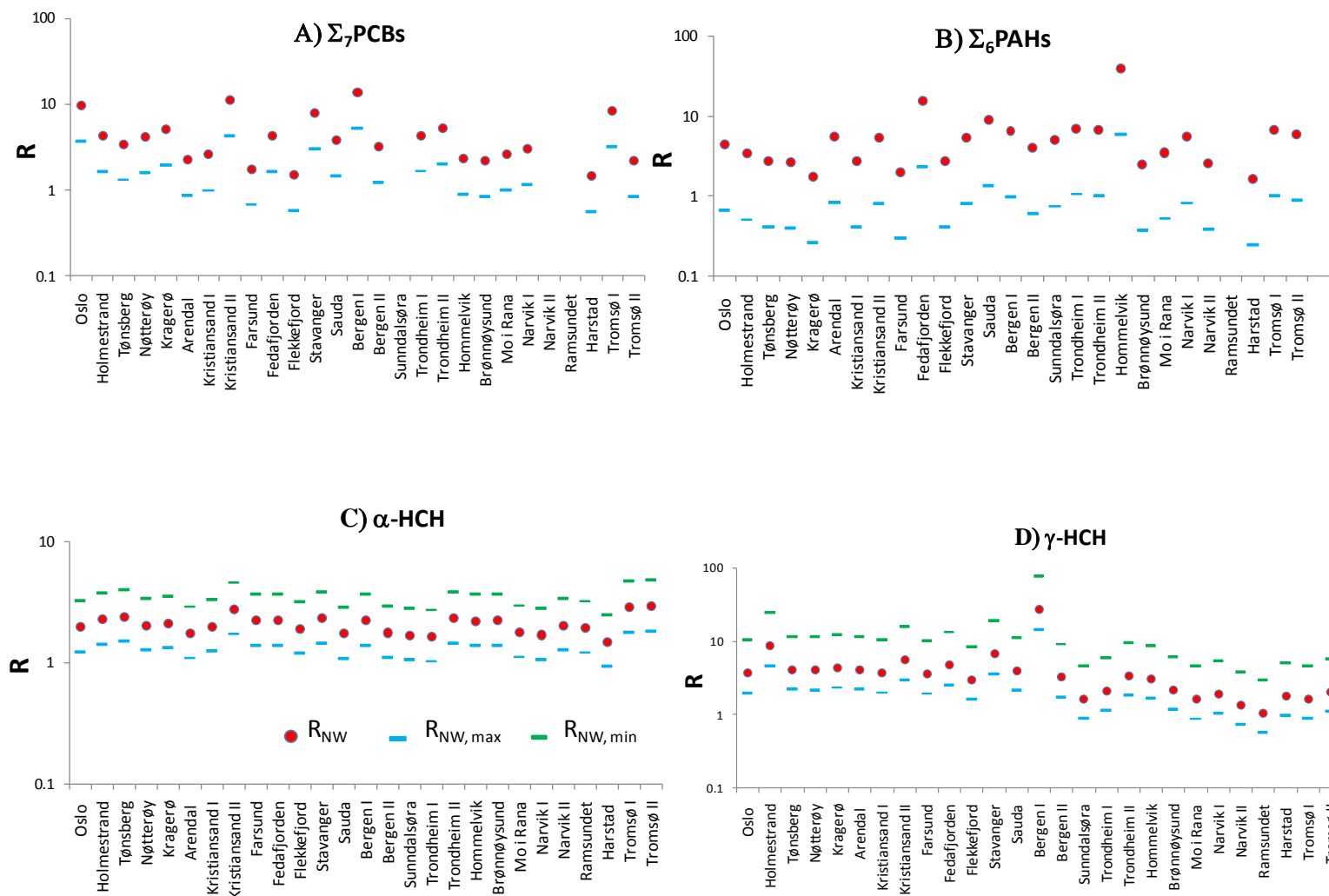


Table 1: Concentrations of selected PCBs, HCHs, DDTs, HCB, chlordanes (pg/m³) and selected PAHs (ng/m³) in air from coastal sites in Norway.

Compounds	Average± S.D	Median	Measured range		MDL	% above MDL	Max/min (MMR)	ratio
			Deployed samples	Blanks				
PCB-28	5±3	4	1.0-10	0.09-0.3	0.4	100	10	
PCB-52	6±5	5	1.2-21	0.07-0.5	0.6	100	18	
PCB-101	5±5	3	0.9-19	0.06-0.6	0.7	100	21	
PCB-118	1.7±1.8	1	0.2 ¹⁾ -7.3	0.04-0.3	0.4	93	>18	
PCB-138	1.7±1.4	1	0.3 ¹⁾ -6.2	0.05-0.5	0.5	93	>12	
PCB-153	2.5±2	2	0.5 ¹⁾ -8.7	0.08-0.8	0.9	88	>9	
PCB-180	0.5±0.4	0.4	0.09 ¹⁾ -1.6	0.02-0.1	0.2	93	>9	
Σ ₇ PCBs	21±17	17	4.5-72				>16	
Fluorene	4±6	2	0.4-32	0.006-0.03	0.04	100	74	
Phenanthrene	6±7	5	0.6-41	0.01-0.07	0.1	100	72	
Anthracene	0.2±0.3	0.1	0.007 ¹⁾ -1.5	0.001-0.008	0.01	96	>108	
Fluoranthene	1.4±1.6	0.9	0.1-6.5	0.003-0.02	0.03	100	54	
Pyrene	0.8±0.9	0.5	0.07-3.9	0.003-0.02	0.02	100	55	
Benz[a]anthracene	0.03±0.03	0.02	0.005 ¹⁾ -0.2	0.005 ³⁾	0.01	85	>15	
Chrysene	0.08±0.06	0.06	0.02-0.3	0.005 ³⁾	0.006	100	14	
Benzo[a]pyrene	0.006±0.002	0.005	0.005 ¹⁾ -0.01	0.005 ³⁾	0.01	11	>1	
Σ ₈ PAHs	13±16	9.5	1-84				>68	
α-HCH	16±3	15	11-22	0.1-0.9	1	100	2	
β-HCH	0.3±0.2	0.3	0.1 ¹⁾ -0.8	0.003 ⁴⁾ -0.2	0.2	70	>4	
γ-HCH	18±21	15	4.5-115	0.1-1.0	1	100	26	
Σ ₃ HCHs	34±22	31	19-133				>7	
HCB	58±50	47	42-305	0.4-1.1	1	100	7	
p,p'-DDE	3±2	1	1.3 ¹⁾ -7.8	0.1-2.4	3	48	>3	
p,p'-DDD	0.2±0.2	0.1	0.04 ²⁾ -1.0	0.001 ⁴⁾ -0.2	0.2	33	>4	
o,p'-DDT	1.4±2.04	0.9	0.2 ¹⁾ -11	0.002 ⁴⁾ -0.4 ⁵⁾	0.4	93	>28	
p,p'-DDT	2.1±4.8	1.0	0.2 ¹⁾ -26	0.005 ⁴⁾ -0.3	0.3	85	>86	

Σ₄DDTs	6.5±8.4	3.4	1.8-45				>26
trans-Chlordane	0.9±1.0	0.5	0.02 ¹⁾ -3.9	0.004 ⁴⁾ -0.03	0.04	96	>91
cis-Chlordane	1.4±0.8	1.2	0.5-4.0	⁶⁾ 0.006-0.1	0.1	100	8
trans-Nonachlor	1.3±0.7	1.1	0.4-3.4	0.002 ⁴⁾ -0.06	0.09	100	10
cis-Nonachlor	0.2±0.2	0.2	0.02 ¹⁾ -0.7	0.002 ⁴⁾ -0.02	0.03	96	>20
Σ₄Chlordanes	3.8±2.6	3.1	1.1-11				>11

¹⁾ The lower limit is ½ MDL

²⁾ The lower limit was influenced by interference

³⁾ The component was not detected in the blanks and ½ IDL (instrument detection limit) value was used

⁴⁾ The lower limit is ½ IDL

⁵⁾ The upper limit was influenced by interference

⁶⁾ Lower and upper values were below IDL

Table 2: Median background concentrations in air for the Norwegian, Nordic and European scenario for selected PCBs, HCHs, DDTs, HCB, Chlordanes (pg/m³) and selected PAHs (ng/m³). All data from Halse et al²⁵.

Compound/groups	Norwegian background		Nordic		European Median	Ratio (median values)	
	Median	Range (min-max)	Median	Range (min-max)		Nordic/Norwegian	European/Norwegian
PCB-28	1.5	0.9-2.1	1.9	0.9-4.7	3.8	1.3	2.5
PCB-52	1.6	1.1-2.3	2.1	1.1-5.7	4.2	1.3	2.6
PCB-101	1.0	0.6-1.9	1.7	0.6-8.7	2.5	1.8	2.7
PCB-118	0.3	0.2-0.6	0.5	0.2-3.3	1.0	1.6	3.0
PCB-138	0.3	0.2-1.1	0.9	0.2-4.7	1.2	2.8	3.9
PCB-153	0.5	0.3-1.6	1.4	0.3-6.8	2.0	2.9	4.2
PCB-180	0.1	0.1-0.6	0.3	0.1-2.1	0.6	2.9	5.5
Σ₇PCBs	5.3	3.4-10.2	8.7	3.4-32.4	15.3	1.7	2.9
Fluorene	0.7	0.3-4.5	0.7	0.3-4.5	1.0	1.06	1.3
Phenanthrene	1.2	0.6-5.0	1.5	0.5-5.0	1.9	1.3	1.6
Anthracene	0.01	0.01-0.1	0.01	N.D ¹⁾ -0.08	0.03	1.1	1.9
Fluoranthene	0.2	0.1-0.7	0.3	0.1-1.1	0.4	1.6	2.5
Pyrene	0.1	0.1-0.2	0.1	0.1-0.5	0.2	1.5	2.76
Benzo[a]pyrene	0.005	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾ -0.06	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾
Benz[a]anthracene	N.D ¹⁾	N.D ¹⁾ -0.04	N.D ¹⁾	N.D ¹⁾ -0.04	0.01	N.D ¹⁾	N.D ¹⁾
Chrysene	0.01	0.009-0.01	0.01	0.01-0.1	0.03	1.2	3.2
Σ₆PAH²⁾	2.2	1.1-10.5	2.6	1.0-11.2	3.6	1.2	1.6
α-HCH	7.6	7.1-8.9	8.6	6.8-12.7	20.6	1.13	2.7
β-HCH	N.D ¹⁾	N.D ¹⁾ -0.2	N.D ¹⁾	N.D ¹⁾ -1.1	0.7	N.D ¹⁾	N.D ¹⁾
γ-HCH	4.3	2.3-5.9	5.0	2.3-11.6	19.5	1.2	4.5
Σ₂HCHs³⁾	11.9	9.4-14.8	13.6	9.2-24.3	40.1	1.1	3.4
HCB	41.5	39.0-46.9	41.8	38.4-58.7	45.1	1.0	1.1
<i>p,p'</i>-DDE	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾ -8.0	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾
<i>p,p'</i>-DDD	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾ -0.2	N.D ¹⁾	N.D ¹⁾	N.D ¹⁾
<i>o,p'</i>-DDT	N.D ¹⁾	N.D ¹⁾ -0.6	0.4	N.D ¹⁾ -2.3	1.8	N.D ¹⁾	N.D ¹⁾
<i>p,p'</i>-DDT	N.D ¹⁾	N.D ¹⁾ -0.8	0.3	N.D ¹⁾ -3.0	2.0	N.D ¹⁾	N.D ¹⁾

Σ_2 DDTs ⁴⁾	N.D ¹⁾	N.D ¹⁾ -1.4	0.7	N.D ¹⁾ -5.3	3.8	N.D ¹⁾	N.D ¹⁾
trans-Chlordane	0.4	0.2-0.6	0.3	N.D ¹⁾ -0.6	0.4	1.0	1.2
cis-Chlordane	0.7	0.6-1.1	0.8	0.4-1.3	1.2	1.0	1.6
trans-Nonachlor	0.8	0.7-1.5	0.8	0.4-1.5	1.2	1.0	1.5
cis-Nonachlor	0.1	0.1-0.2	0.1	N.D ¹⁾ -0.3	0.1	0.9	1.4
Σ_4Chlordanes	2.0	1.6-3.4	2.0	0.9 ⁵⁾ -3.7	3.0	1.0	1.5

¹⁾ Concentration <MDL ²⁵

²⁾ Fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene

³⁾ α -HCH and γ -HCH

⁴⁾ *o,p'*-DDT and *p,p'*-DDT

⁵⁾ Lower range, trans-Chlordane and cis-Chlordane < MDL ²⁵