

# Spatio-temporal patterns of PAHs, PCBs and HCB in sediments of the western Barents Sea

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## KEYWORDS

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## Abstract

We examine the composition and levels of organic contaminants (PAHs, PCB, HCB) in four sediment cores collected from the Barents Sea. We assess the influence of temporal variations in contaminant supplies and post-depositional reworking on contaminant distribution. Anthropogenic levels of  $\sum 12\text{PAH}$  reached  $95 \text{ ng g}^{-1}$ , higher inventories dominated by BKF were observed at southern stations, while

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northern stations exhibited lower inventories with PHE as the dominant compound. The PCB composition was similar at all stations dominated by CB 101, 138 and 153.  $\sum 7$ PCB concentrations were higher at northern stations. The observed composition and spatio-temporal pattern of organic contaminants is in accordance with long-range transport supplies.

## 1. Introduction

Long-range atmospheric transport and chemical condensation reactions are responsible for the widespread distribution of compounds such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) in the Arctic (e.g. Halsall et al. 2001, Wania & Su 2004). In the European sector of the Arctic, regional sources of pollutants such as metallurgical smelters and military installations operating along the Norwegian and Russian coasts add to supplies from global emissions sources (Savinov et al. 2003, Carroll et al. 2008a).

Marine sediments are the final sink for volatile persistent organic pollutants (POPs) entering the Arctic (Wania & Su 2004). Marine sediments acquire their contaminant composition through direct particle deposition (Ab Razak et al. 1996) and by transfer from seawater to the bottom surface sediments during downwelling events. For example, in the Norwegian Sea the PCB flux via settling particles was  $320 \text{ kg yr}^{-1}$  compared to a direct removal flux to surface sediment deposits of  $870 \text{ kg yr}^{-1}$  in the North Atlantic (Lohmann et al. 2006). Sea ice transport also facilitates the transfer of contaminants from industrialized areas of the Siberian coast to other locations in the Arctic (Pfirman et al. 1995, 1997, Pavlov et al. 2004). Estimates prepared by the Arctic Monitoring and Assessment Program (AMAP) report that approximately 45% of PCBs reaching Svalbard are by air transport, 30% by ocean currents and 25% by sea ice transport (AMAP 2004).

Sediment accumulation is an important process governing the storage of contaminant-laden sediments on the sea floor. However, contaminant distribution and composition are further affected by post-depositional processes. Sediment mixing may affect the down-core concentration and composition of contaminants, causing chemicals to spread further down the sediment column. In high energy and/or high benthic infauna density environments, resuspension events may result in contaminant reintroduction to the water column (Thibodeaux & Bierman 2003, Carroll & Lerche 2003). Moreover, polychlorinated biphenyls can be degraded by both anaerobic and aerobic bacteria. Under anaerobic conditions, highly chlorinated congeners may be converted into less chlorinated biphenyls by microbial groups. Under aerobic conditions, microorganisms break down less chlorinated

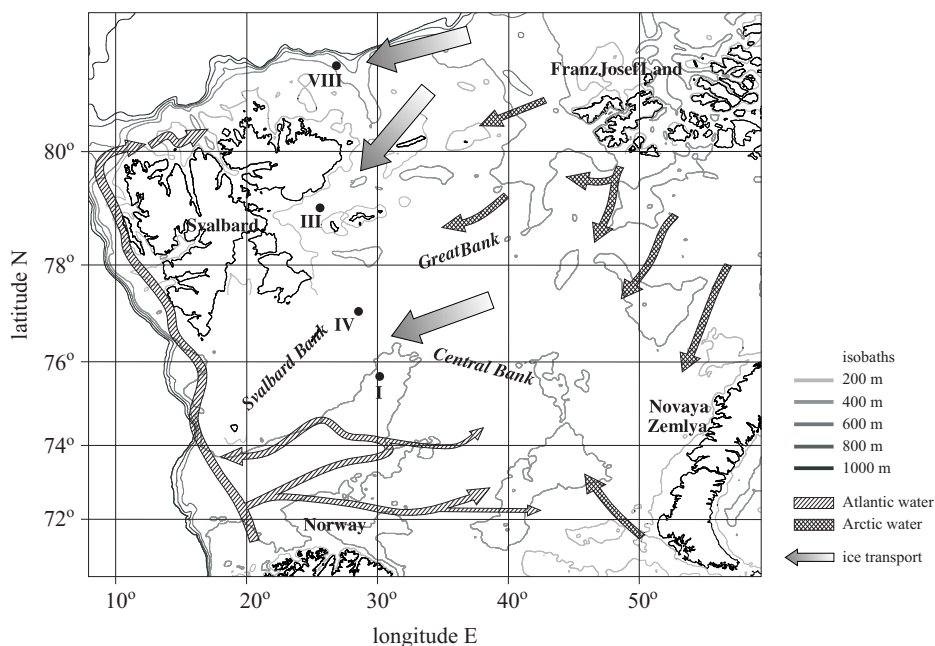
biphenyl rings to yield chlorinated benzoates and pentanoic acid derivatives (Rodrigues et al. 2006).

The spatial distribution pattern of POPs in surface sediments has been widely investigated in the Arctic (e.g. Valette-Silver et al. 1999, Savinov et al. 2000, 2003, Gustafsson et al. 2001, Strachan et al. 2001, Kuzyk et al. 2005), providing insight into linkages between sources and contamination patterns. The Barents Sea has been a focal point of investigation in the European Arctic both offshore (Yunker et al. 1996, Boitsov et al. 2009a, Dahle et al. 2009) and in adjacent coastal areas (Næs et al. 1995, Sericano et al. 2001, Dahle et al. 2003, Carroll et al. 2008a).

However, with the notable exception of Yunker et al. (1996) and Boitsov et al. (2009a,b), the majority of studies are limited to the investigation of surface sediments (down to ~1–2 cm). In the present study, we examine the contaminant record (~150 years) from four locations along a south-north latitudinal transect of the western Barents Sea using sediment cores dated by  $^{210}\text{Pb}$  geochronology. We identify potential contaminant sources based on interpretation of the congener proportions and overall sediment concentrations of the studied compounds. For PCBs and HCB we assess whether sediment contaminant levels reflect the decline in production associated with the regulatory ban on the usage of products containing these compounds. Finally, the study provides an opportunity to discuss the influence of burial and post-depositional sediment reworking processes on the interpretation of persistent organic contaminants detected in marine sediments.

## 2. Material and methods

Sediment cores were collected from four stations in the central and northern regions of the western Barents Sea using a 4-core multi-corer (Figure 1). At each station, two of the four retrieved sediment cores were sliced at 1 cm intervals, and 1 cm of the outside edge of each interval was discarded to eliminate down-core contamination. Sediments from similar depth intervals in each of the two cores were combined to obtain sufficient sample material for contaminant analyses. Sediment subsamples were stored in covered glass jars previously heated to 450°C. Sample jars were frozen at  $-20^\circ\text{C}$  until further processing in the laboratory. The remaining two sediment cores collected during each multi-corer cast were stored for the analysis of sediment properties and of radionuclide concentration measurements:  $^{234}\text{Th}$ ,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ . Sediments at all stations were composed mainly of fine material (45–98% pelite) with organic carbon contents ranging from 1.0–2.4%  $\text{C}_{\text{org}}$  (Carroll et al. 2008b). Profiles of both  $^{210}\text{Pb}$  and  $^{234}\text{Th}$  were used to determine sediment mixing rates (Carroll



**Figure 1.** Study area and location of sampling stations in the western Barents Sea (Atlantic and Arctic water mass circulation patterns are derived from Sundfjord et al. (2007)); the sea ice transport pathways were modelled by Hop & Pavlova (2007)

et al. 2008b), while sedimentation velocities were determined by  $^{210}\text{Pb}$  and validated with  $^{137}\text{Cs}$  (Zaborska et al. 2008). Sediment accumulation rates and mixing depths/rates are presented in Table 1.

Analyses of organic pollutants (polychlorinated biphenyls – PCBs, hexachlorobenzene – HCB and polycyclic aromatic hydrocarbons – PAHs) were performed on sediment samples from selected depth intervals. Individual samples were freeze-dried (Christ Beta A apparatus) and homogenized. Sub-samples of 15–20 g were treated by triplicate extraction with methylene chloride in an ultrasonic bath. Internal standards (octachloronaphthalene and hexamethylbenzene) were added to each sample prior to extraction in order to control the recovery efficiency of the entire process. The extracts were concentrated followed by clean-up procedures (Behar et al. 1989, Tronczyński et al. 2004, Pazdro 2004). Briefly, elemental sulphur was removed from an extract using copper powder activated with hydrochloric acid. Afterwards the extract was concentrated under a gentle flow of nitrogen, and a second clean-up and fractionation were performed by absorption chromatography on silica gel and aluminium oxide (both deactivated with 5% water). Solvent mixtures of increasing polarity were used

**Table 1.** Sampling stations in the western Barents Sea (WBS).  $^{210}\text{Pb}$  sediment profiles were modelled as a two-layered system with mixing and sediment accumulation in the surface layer and only mass sediment accumulation (MSA in  $\text{g m}^{-2} \text{yr}^{-1}$ ) below the mixed layer. Surface sediment layer mixing rates are in  $\text{cm}^2 \text{yr}^{-1}$

Station	Latitude [N] Longitude [E]	Location	Depth [m]	MSA	Mixing Rate/ Mixed Depth	Comments
I	75°40' 30°10'	Hopen Trench	345	340 ± 6%	0.1/1 cm	surface sediment resuspension and mixing associated with bottom currents
IV	77°01' 29°29'	Hopen Bank	222	320 ± 14%	0.1/2 cm	surface sediment resuspension and mixing associated with bottom currents
III	79°01' 25°46'	East Erik Eriksentretet	198	510 ± 2%	no mixing	profile provides an accurate reconstruction of contaminant deposition in the WBS
VIII	81°20' 27°19'	North Kvitøya Trench	503	600 ± 5%	0.4/2 cm	area of sediment focusing, serves as a conduit for sediments from the shelf to central Arctic basin

(F1 – 100% hexane, extracting HCB and PCBs; F2 – 90% hexane : 10% methylene chloride, extracting PAHs.) The purified sample fractions were evaporated and dissolved in isoctane prior to final quantitative and qualitative analysis.

Extracts were analysed by gas capillary chromatography. A Shimadzu GC 17 equipped with a split/splitless injector at 280°C and a DB 5 column (60 m × 0.25 mm i.d. × 0.25 μm film thickness) were used. A flame ionization detector (FID) and helium carrier gas were used for the PAH analyses at the following oven temperatures: 50°C held for 1 min, followed by a 5°C min<sup>-1</sup> increase to 150°C, followed by a 30°C min<sup>-1</sup> increase to 310°C, held for 25 min. PCBs and HCB were analysed by applying an electron capture detector (ECD), helium (carrier gas) and the following oven temperature programme: 100°C held for 1 min; 6°C min<sup>-1</sup> to 140°C; 2.5°C min<sup>-1</sup> to 250°C; 10°C min<sup>-1</sup> to 310°C, held for 20 min. Identification of the individual compounds was based on their retention time using internal and external standards (LG PROMOCHEM). The identification was checked by the analysis of selected extracts by GC-MS. The individual compounds were quantified by using external five-point calibration curves plotted for each compound in the linear range of the detector's response, and taking into account the concentration ranges of the compounds in the samples. Laboratory calibration solutions were prepared in isoctane by appropriate dilutions (by weight) of standard mixtures (LG PROMOCHEM).

The QA/QC procedures included procedural blanks (in each batch of samples), analyses of replicate samples and the use of internal recovery standards added to each sample prior to extraction in order to monitor the recovery efficiency of the entire process. Procedural blanks for each set of samples were prepared by extracting a sample of sediment devoid of organic matter (heated at 450°C for 12 h). These samples were processed in the same manner as real samples. The quantification limits, measured as average blanks plus six standard deviations of the average blanks) were 10–50 pg g<sup>-1</sup> d.w.<sup>-1</sup> for organochlorine compounds and 80–220 pg g<sup>-1</sup> d.w.<sup>-1</sup> for PAHs. Recoveries of individual compounds were in the 75–105% range, while relative standard deviations varied from 9 to 25% of average recoveries (triplicate analyses). Analyses of certified reference sediment material (IAEA-383) were routinely included in each batch of samples to monitor procedural accuracy. The low accuracy of naphthalene, acenaphthene and acenaphthylene mean that these analytes were excluded from the list of the PAHs studied. The following PAHs were measured: Fluorene (FLN), Phenanthrene (PHE), Anthracene (ANT), Fluoranthene (FLT), Pyrene (PYR), Benzo(a)anthracene (BAA), Chrysene (CHR), B(b+k)fluoranthene (BKF), Benzo(a)pyrene (BAP),

Dibenzo(a,h)anthracene (DBA), Benzo(ghi)perylene (BP) and Indeno(1,2,3-c,d)pyrene (IND). The PCBs included CB 28, CB 52, CB 101, CB 118, CB 138, CB 153 and CB 180. Individual component measurement uncertainty was calculated from 5 replicate analyses of compounds in certified reference material. The measurement uncertainties ranged from 10.75% (CB 180) to 23.26% (CB 28) for individual PCBs and from 7.43% (FLT) to 27.27% (DBA) for individual PAHs.

### 3. Results and discussion

Seafloor sediment dynamics modulate contaminant accumulation on continental shelves. The historical reconstruction of contaminant supplies to the western Barents Sea was obtained by converting sediment depth to time using  $^{210}\text{Pb}$  derived sedimentation velocities (Zaborska et al. 2008). This enabled an average age to be assigned to the individual sediment depth intervals in each core. The temporal pattern of POPs preserved in these sediment layers should reflect the dual influences of varied contaminant supplies over time and post-depositional sedimentary reworking and mineralization. Sediment mixing through physical and/or biological mechanisms was observed at three of the four stations sampled in this investigation (Table 1). Sediment disturbance was most pronounced at station VIII. This station is located in the Kvitøya Trench, which serves as a conduit of material to the central Arctic Basin (Vandieken et al. 2006, Carroll et al. 2008b). At both southern stations (I and IV), sediment mixing is pronounced in the upper 2 cm. This depth interval corresponds to a time period of approximately 40–60 years. The profile of organic contaminant concentrations with depth at station III provides an accurate historical record owing to the negligible influence of sediment mixing at this location.

#### 3.1. Polycyclic Aromatic Hydrocarbons

##### 3.1.1. Spatial and temporal patterns

PAH concentrations ( $\sum 12\text{PAH}$ ) measured in surface sediments ranged from  $35 \pm 18 \text{ ng g}^{-1} \text{ d.w}^{-1}$  to  $132 \pm 66 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (Table 2). Surface sediment concentrations were lowest at northern stations –  $35 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (III) and  $51 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (VIII) – compared to southern stations –  $132 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (I) and  $103 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (IV). Based on PAH levels detected in the deepest layers of the sediment cores ( $> 12 \text{ cm}$ ) and corresponding to sediment ages prior to 1850, natural background levels of  $\sum 12\text{PAH}$  were fairly constant throughout the western Barents Sea, ranging from 25 to  $37 \text{ ng g}^{-1}$  (mean  $30 \text{ ng g}^{-1} \text{ d.w}^{-1}$ ;  $n=7$ ). Our data are in

**Table 2.** Concentrations of  $\sum 12$  PAHs and  $\sum 7$  PCBs [ $\text{ng g}^{-1} \text{ d.w}^{-1}$ ] at four stations in the western Barents Sea. Compound concentrations below the quantification limit are marked 'b.d.'. Sediment layers where HCB was not measured are marked '–'

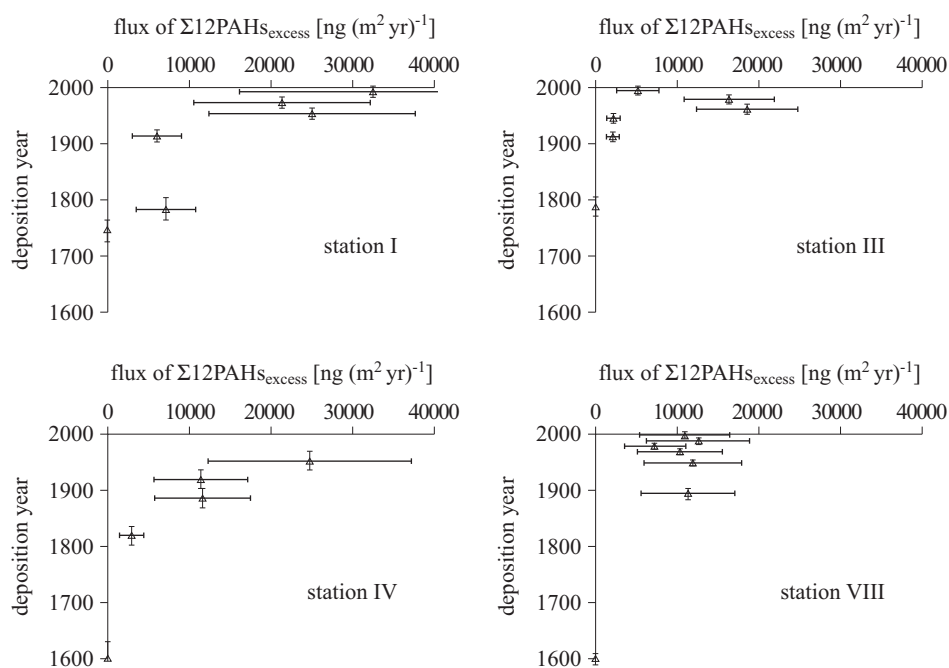
Station/layer [cm]	$\sum 12$ PAH [ $\text{ng g}^{-1} \text{ d.w}^{-1}$ ]	$\sum 7$ PCB [ $\text{ng g}^{-1} \text{ d.w}^{-1}$ ]	HCB [ $\text{ng g}^{-1} \text{ d.w}^{-1}$ ]
I/0–1	132.02 ± 66.42	0.79 ± 0.31	–
I/1–2	99.48 ± 50.05	0.69 ± 0.27	–
I/2–3	110.02 ± 55.35	0.88 ± 0.35	–
I/4–5	–	0.98 ± 0.39	–
I/5–6	54.20 ± 27.27	–	–
I/10–12	57.73 ± 29.05	b.d.	–
I/12–14	36.65 ± 18.44	b.d.	–
III/0–1	35.29 ± 17.76	0.73 ± 0.29	0.73 ± 0.15
III/1–2	57.19 ± 28.78	0.93 ± 0.37	–
III/2–3	61.45 ± 30.92	3.54 ± 1.40	0.47 ± 0.09
III/3–4	29.57 ± 14.88	1.57 ± 0.62	0.33 ± 0.07
III/5–6	29.28 ± 14.73	b.d.	b.d.
III/12–14	25.05 ± 12.60	b.d.	b.d.
IV/0–1	–	1.54 ± 0.61	0.52 ± 0.10
IV/1–2	103.27 ± 51.96	1.06 ± 0.42	0.76 ± 0.15
IV/2–3	62.11 ± 31.25	1.75 ± 0.69	0.61 ± 0.12
IV/3–4	62.74 ± 31.57	1.51 ± 0.60	0.59 ± 0.12
IV/5–6	35.34 ± 17.78	b.d.	0.43 ± 0.09
IV/10–12	–	b.d.	b.d.
IV/12–14	26.23 ± 13.20	b.d.	b.d.
VIII/0–1	51.21 ± 25.77	1.42 ± 0.56	2.01 ± 0.40
VIII/1–2	54.00 ± 27.17	1.80 ± 0.71	0.45 ± 0.09
VIII/2–3	45.23 ± 22.76	0.90 ± 0.36	0.34 ± 0.07
VIII/3–4	50.27 ± 25.29	0.72 ± 0.28	0.35 ± 0.07
VIII/5–6	52.75 ± 26.54	b.d.	0.38 ± 0.08
VIII/10–12	51.92 ± 26.12	b.d.	b.d.
VIII/18–20	32.95 ± 16.58	b.d.	b.d.

relatively good agreement with previously reported results for the region (Yunker et al. 1996, Sericano et al. 2001, Boitsov et al. 2009b). However, a detailed comparison of findings is problematic because of differences in the number of compounds investigated among these investigations. Boitsov et al. (2009b) conducted a large study of PAH concentrations in surface



sediments of the western Barents Sea ( $\Sigma 20$  PAH concentrations ranging from 20 to 1426 ng g<sup>-1</sup> d.w<sup>-1</sup> were reported from some stations in the vicinity of our stations I and IV). Yunker et al. (1998) reported  $\Sigma$ PAH<sub>178–278</sub> concentrations from 18 to 160 ng g<sup>-1</sup> d.w<sup>-1</sup> in sediment cores from the vicinity of Novaya Zemlya with higher concentrations (43–500 ng g<sup>-1</sup> d.w<sup>-1</sup>) detected in cores from the NW and SE Barents Sea. In another study, Sericano et al. (2001) reported 2,3-ring PAHs of  $\leq 110$  ng g<sup>-1</sup> d.w<sup>-1</sup> in the Kara Sea.

In the present investigation, mixing resulted in relatively uniform  $\Sigma 12$  PAH versus sediment depth profiles at the southern stations. At station VIII, where mixing also influences the contaminant profile, there is a general pattern of increasing PAH concentrations from pre-industrial background values to the present-day. Station III provides the least disturbed temporal pattern of sedimentary  $\Sigma 12$  PAH (Figure 2), exhibiting a pattern of increasing concentrations until the 1980s, followed by decreasing concentrations in recent times.



**Figure 2.** Flux of  $\Sigma 12$  PAHs excess (total – natural background) [ $\text{ng m}^{-2} \text{ yr}^{-1}$ ] plotted against deposition year (derived from  $^{210}\text{Pb}$ ) at four stations in the western Barents Sea

After correction for natural background, PAH inventories provide a relative measure of differences in the accumulated load of contaminants

among stations. As we measured  $\sum 12$  PAH at similar depth intervals in each core, the inventories among the four stations are comparable. The pattern that emerges is in agreement with our earlier conclusions regarding the concentration pattern observed in surface sediments alone, that is, inventories are higher at southern stations I ( $51 \pm 26$  ng cm<sup>-2</sup> d.w<sup>-1</sup>) and IV ( $70 \pm 36$  ng cm<sup>-2</sup> d.w<sup>-1</sup>) compared to northern stations III ( $22 \pm 11$  ng cm<sup>-2</sup> d.w<sup>-1</sup>) and VIII ( $21 \pm 11$  ng cm<sup>-2</sup> d.w<sup>-1</sup>).

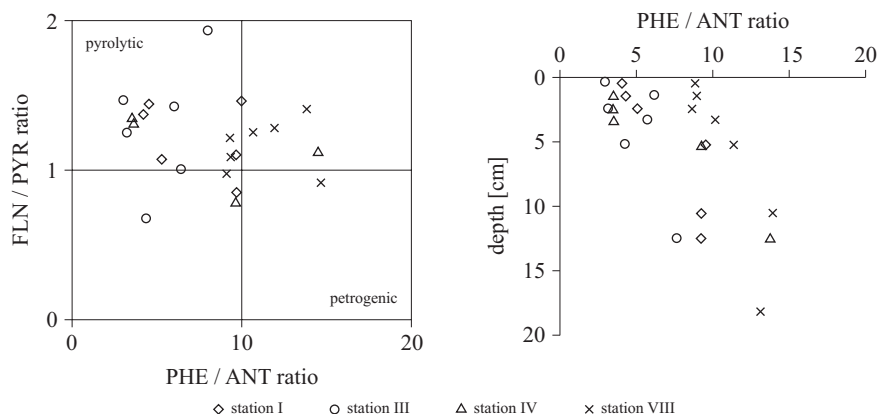
### 3.1.2. PAH composition and sources

At the southern stations (I and IV), BKF is the dominant compound, constituting respectively 15–30% and 28–42% of  $\sum 12$  PAH. Other dominant compounds at the southern stations are PHE (9–23%) and CHR (6–17%). In contrast, the dominant compound at stations III and VIII is PHE, representing respectively 12 to 38% and 12 to 45% of  $\sum 12$  PAH. In addition, CHR (4–21%) and BKF (7–21%) are compounds detected in relatively high concentrations at the more northerly stations. There is a predominance of medium and high molecular weight PAHs that is characteristic of atmospheric releases from local industries such as aluminium smelters and the by-products of coal and wood burning (Savinov et al. 2000). Both the present study and that of Yunker et al. (1996) identified BKF as a dominant PAH in Barents Sea sediment deposits (Table 3). This compound is not produced commercially on an industrial scale (Lide (ed.) 1991) but enters the environment as a by-product of the incomplete combustion of organic material. PHE, the predominant PAH at the northern stations (III and VIII), is also a combustion by-product. Hence, the PAH composition at all stations exhibits an anthropogenic signature consistent with known industrial activities in the region. In contrast, Boitsov et al. (2009b) reported a predominance of alkylated PAHs in sediments collected from the western Barents Sea: an indication of petrogenic PAHs. However, we are unable to compare their results with ours because in the present investigation we did not measure alkylated compounds.

To assess the origin of PAH contamination of sediments, we use individual component ratios as a diagnostic tool (Budzinski et al. 1997, Qiao et al. 2006). Since we were not able to measure lighter alkylated PAHs, only FLT/PYR, PHE/ANT and CHR/BAA indices are presented. Compound ratios of FLT/PYR > 1, PHE/ANT < 10 and CHR/BAA < 1 are characteristic of pyrolytic sources of PAH, while FLT/PYR < 1, PHE/ANT > 15 and CHR/BAA > 1 indicate PAHs of petrogenic origin (Dahle et al. 2003). At the four stations investigated, FLT/PYR > 1 and PHE/ANT < 10 (Figure 3) are consistent with the conclusion that PAHs are of pyrogenic origin, e.g. coal combustion. At station VIII, the PHE/ANT ratio (9–15) was relatively

**Table 3.** Comparison of  $\sum$ PAH concentrations [ $\text{ng g}^{-1} \text{d.w}^{-1}$ ] in different Arctic regions. If the cited articles listed the concentrations of the different compounds, the sum of the same 12 PAH as in the current investigation was calculated. In five cases the total sum of more than 12 PAHs was presented. Sericano et al. (2001) measured 4 more PAHs: Naphthalene, Acenaphthylene, Acenaphthene, Benzo(e)pyrene and Biphenyl. Yunker et al. (1996) measured PAHs of molecular weight 178–278. The sum of 20 PAHs is presented in Boitsov et al. (2009b), of 22 PAHs in Boitsov et al. (2009a) and of 23 PAHs in Stange & Klungsøyr (1997)

Region	$\sum$ PAH [ $\text{ng g}^{-1} \text{d.w}^{-1}$ ]	Dominants	Reference
<b>Open shelf areas</b>			
western Barents Sea	35.3–132.0	BKF, PHE	this paper
Barents Sea	1.0–2000	Alkyl PAHs	Stange & Klungsøyr (1997)
NW Barents Sea	44–500	BKF	Yunker et al. (1996)
SE Barents Sea	43–390	FLN, BKF, IND	Yunker et al. (1996)
SW Barents Sea	20–363	BKF, IND, PER, C2-NAP	Boitsov et al. (2009a)
southern and central Barents Sea	10.2–1426	Alkyl PAHs	Boitsov et al. (2009b)
Barents Sea Novaya Zemlia region	18–160	PHE, FLN, BKF	Yunker et al. (1996)
coastal Barents Sea	83.6–239.4	BKF, FLT	Savinov et al. (2003)
White Sea	10.1–151.6	BKF, BP, FLT	Savinov et al. (2000)
Kara Sea shelf	36.0	FLT, BKF, BP	Dahle et al. (2003)
Kara Sea	b.d. to 110	2, 3 ring PAHs	Sericano et al. (2001)
<b>Areas influenced by rivers</b>			
Gulf of Ob and Yenisei	41.2–99.5	FLT, BKF, BP, PHE	Dahle et al. (2003)
Rivers Ob and Yenisey	22–810	4, 5, 6 ring PAHs	Sericano et al. (2001)
Guba Pechenga	up to 2479	BKF, PHE	Savinov et al. (2003)
Beaufort Sea shelf influenced by the Mackenzie River	7 434	PHE, BP, BKF	Yunker & Macdonald (1995)
<b>Area influenced by industry</b>			
Norwegian fjords affected by aluminium and manganese-alloy smelters	784 296	BKF, CHR, IND	Næs & Oug (1998)



**Figure 3.** a) FLN/PYR and PHE/ANT ratios in sediments at four stations in the western Barents Sea; b) PHE/ANT ratio profiles plotted against sediment depth [cm]

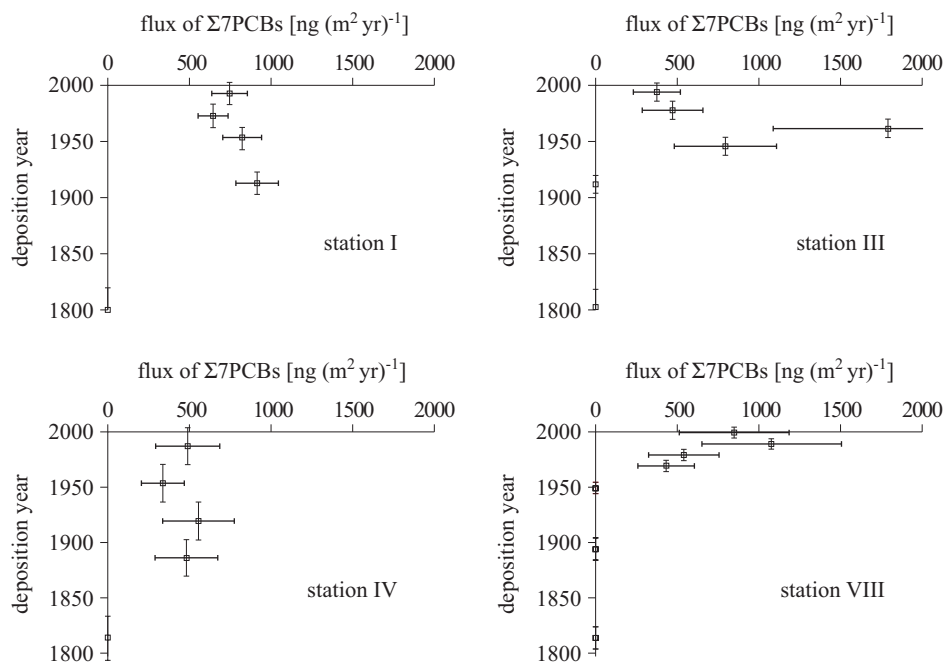
high compared to the other stations (3–10). This feature is explained as resulting from mixed pyrogenic and petrogenic origins, a finding that is consistent with the sediment mixing reported earlier. Boitsov et al. (2009b) report PHE/ANT ratios (from 9.4 to 113) for 69 stations in the western Barents Sea. As previously mentioned, these authors detected petrogenic PAHs with only minimal influence from anthropogenic sources. The difference between our conclusion and that of Boitsov et al. (2009b) regarding the origin of PAHs is most likely due to local differences in mixing regimes. The sediments collected for this study were mostly mixed in the surface intervals; hence, modern sediments were contaminated by the signatures laid down in previous decades.

There is a general pattern of increasing PHE/ANT ratios with sediment depth/deposition time (Figure 3). This pattern reflects the down-core transition from anthropogenic to natural hydrocarbon sources over time from the present day to the pre-industrial period.

### 3.2. Polychlorinated biphenyls and hexachlorobenzene

#### 3.2.1. Spatial and temporal patterns

Polychlorinated biphenyls were detected in sediments down to 4 cm depth. Concentrations of  $\sum 7\text{PCB}$  within this depth interval ranged from  $0.7 \pm 0.3 \text{ ng g}^{-1}$  to  $3.5 \pm 1.4 \text{ ng g}^{-1}$  (Table 2), with the highest concentration detected at station III and the lowest one at station I. 7PCB inventories in the uppermost sections of the cores (0–4 cm) were lower ( $1.0 \pm 0.4 \text{ ng cm}^{-2}$ – $1.2 \pm 0.5 \text{ ng cm}^{-2}$ ) at stations I and VIII and higher



**Figure 4.** Flux of  $\Sigma 7$ PCBs [ $\text{ng m}^{-2} \text{yr}^{-1}$ ] plotted against deposition year (derived from  $^{210}\text{Pb}$ ) at four stations in the western Barents Sea

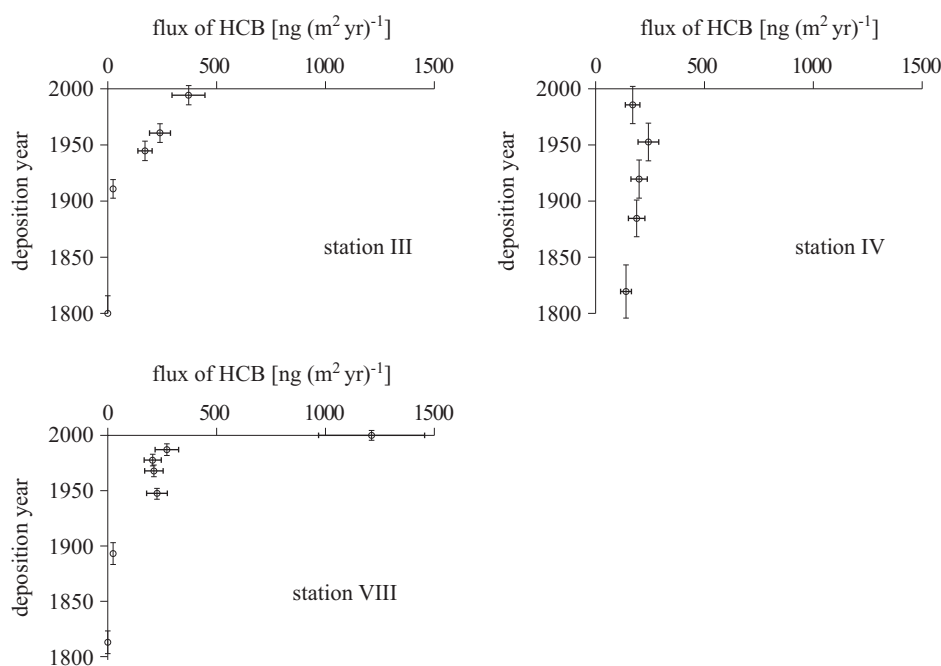
( $2.4 \pm 1.0 \text{ ng cm}^{-2}$ – $2.6 \pm 1.0 \text{ ng cm}^{-2}$ ) at stations III and IV. The down-core concentration pattern of  $\Sigma 7$ PCB is, however, similar to the one observed for  $\Sigma 12$ PAH. At station I, the  $\Sigma 7$ PCB content is relatively uniform throughout the length of the core. Station IV exhibits measurable  $\Sigma 7$ PCB concentrations in sediment layers deposited before biggest industry development (the beginning of the 19th century), suggesting that exchange of PCBs between surface contaminated layers and deeper pristine sediment layers has occurred at this location. The overall pattern observed for  $\Sigma 7$ PCB with sediment depth indicates that stations I, IV and VIII do not follow the historical global discharge pattern for PCBs. Surface sediment mixing at these stations (Carroll et al. 2008b) results in the homogenization of PCB concentrations within these sediment cores. The higher surface PCB concentrations at station VIII located in the trench system may have been caused by strong resuspension of sedimentary material from the surrounding slopes (Carroll et al. 2008b). The undisturbed sediment profile at station III exhibits a maximum measured  $\Sigma 7$ PCB concentration ( $3.54 \pm 1.4 \text{ ng d}^{-1} \text{ d.w}^{-1}$ ) corresponding to a deposition time of 1961 ( $\pm 8$  years) (Figure 4). After this date, the  $\Sigma 7$ PCB concentration at this station decreases to  $0.73 \pm 0.29 \text{ ng g}^{-1}$  at the sediment surface. This

agrees well with the ban on PCB production introduced in 1966 in Europe and North America (Figure 4). A similar pattern has been documented in sediments from the North Sea and Baltic Sea (Van Zoest & Van Eck 1993, Axelman et al. 1995).

The  $\sum 7$ PCB burial fluxes derived using sedimentation velocities (Figure 4) indicate that maximum  $\sum 7$ PCB fluxes are 2–5 times higher at the northern stations III (372–1806 ng m<sup>-2</sup> yr<sup>-1</sup>) and VIII (432–1079 ng m<sup>-2</sup> yr<sup>-1</sup>), compared to the southern stations I (235–334 ng m<sup>-2</sup> yr<sup>-1</sup>) and IV (340–559 ng m<sup>-2</sup> yr<sup>-1</sup>). Analyses of <sup>137</sup>Cs in the same sediment samples (Zaborska et al. 2008, 2010) showed that northern stations III and VIII are influenced by additional sources of sedimentary material. Inventories of <sup>137</sup>Cs at these locations were three times higher than at southern stations I and IV. We think that in the northern part of the Barents Sea, terrigenous material from sea ice melting or coastal erosion plays an important role. The high  $\sum 7$ PCB burial flux at station VIII may also have been caused by intense sediment focusing, since this station is located in the trench where sedimentary material is supplied from surrounding slopes (Carroll et al. 2008b). Analyses of <sup>210</sup>Pb, <sup>234</sup>Th and C<sub>org</sub> at this station indicate scavenging and focusing of organic carbon from non-local sources (Carroll et al. 2008b).  $\sum 7$ PCB concentrations and burial flux were the lowest at the southernmost station I. This region was found to be dominated by sediments of marine origin (C/N: 7–9). These sediments, containing organic carbon derived from marine primary production, exhibit a high burial rate of 7.1 gC m<sup>2</sup> yr<sup>-1</sup> (Carroll et al. 2008b). This may indicate that contaminants at this location are diluted by organic material associated with high rates of primary productivity in the region.

Studies of organic contaminants typically report on different congeners, making it difficult to compare results among different investigations. Thus, we adopt the strategy of Gustafsson et al. (2001) and evaluate CB 52 alone as an indicator of site-to-site differences in contaminant supplies. The CB 52 fluxes at our stations were 79–146 pg m<sup>-2</sup> d<sup>-1</sup> (station I), 62–304 pg m<sup>-2</sup> d<sup>-1</sup> (station IV), 138–853 pg m<sup>-2</sup> d<sup>-1</sup> (station III) and 33–341 pg m<sup>-2</sup> d<sup>-1</sup> (station VIII). In the Baltic Sea, CB 52 fluxes were ~400 pg m<sup>-2</sup> d<sup>-1</sup>, whereas in Baffin Bay, CB 52 fluxes were considerably lower, ranging from 19 to 56 pg m<sup>-2</sup> d<sup>-1</sup> (Savinov et al. 2000). Thus, CB 52 burial fluxes for the Barents Sea are generally higher than those at the Baffin Bay site in the Canadian Arctic and comparable to fluxes in the more heavily industrialized Baltic Sea area: this is quite an astonishing feature, considering the long distance between industrial sources and the study area.

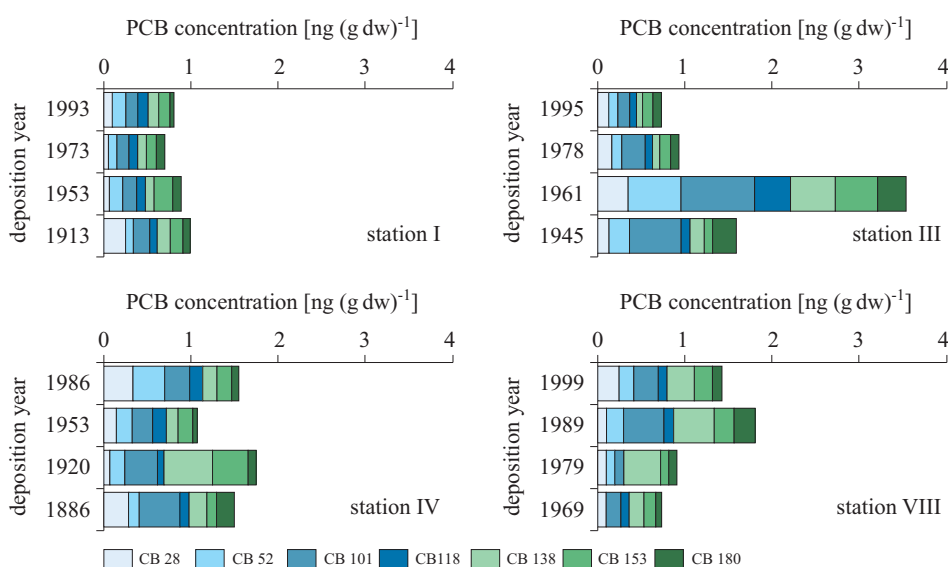
HCB concentrations in surface sediments (stations III, IV and VIII only) were 0.5–2.0 ng g<sup>-1</sup> d.w<sup>-1</sup> (Table 2). Previous measurements of HCB levels in sediments from Guba Pechenga (northern Russia) and the southern Barents Sea shelf ranged from 0.3 to 1.8 ng g<sup>-1</sup> d.w<sup>-1</sup> (Savinov et al. 2003). These sediment concentrations are higher than those reported for the Bering and Chukchi Seas (0.04 to 0.08 ng g<sup>-1</sup> d.w<sup>-1</sup>) (Iwata et al. 1994), while concentrations up to 6.7 ng g<sup>-1</sup> d.w<sup>-1</sup> have also been reported in some harbours of northern Norway (Dahle et al. 2000). At stations III and VIII the highest HCB burial fluxes (Figure 5) are observed at surface sediments and decrease down-core. Although the industrial, direct production of HCB in Europe and N. America ended in the early 1990s (no data from the former USSR is available), this recent contamination may have originated from the production of other chlorinated compounds, such as perchloroethylene, carbon tetrachloride and, to some extent, trichloroethylene, polychlorinated-p-dioxins and polychlorinated dibenzofurans (CEPA 1993). The pattern of HCB burial flux at station IV is constant and similar to the pattern observed for  $\sum 7$ PCB (Figure 5), which again provides confirmation of the strong sediment mixing there (Zaborska et al. 2008).



**Figure 5.** Flux of HCB [g m<sup>2</sup> yr<sup>-1</sup>] plotted against deposition year (derived from <sup>210</sup>Pb) at three stations in the western Barents Sea

### 3.2.2. PCB composition and sources

The dominant PCB congeners in the western Barents Sea are CB 101, CB 153 and CB 138 (Figure 6). However, the southernmost station (I) has a lower total PCB concentration than the other stations. Moreover, these sediments exhibit no dominant PCB congener. In contrast, CB 101 dominates the composition at station IV, accounting for 23–28%  $\sum 7$  PCB. At station III CB 101 is predominant (22–41%), particularly in the deeper sediment layers. In addition, the congeners CB 153 and CB 138 are important at station III. At station VIII, both CB 101 (23–57%) and CB 138 (22–47%) are the dominant congeners in the sediment layers.



**Figure 6.** PCB concentrations [ $\text{ng g}^{-1} \text{d.w}^{-1}$ ] and congener composition plotted against sediment deposition year (middle of sediment layer deposition year derived from  $^{210}\text{Pb}$ )

As the composition and relative proportion of PCB congeners are comparable among stations and throughout time, the flux of PCBs to the western Barents Sea has remained relatively constant over time. Skotvold & Savinov (2003) evaluated several chemicals as potential sources of PCBs to this region of the Arctic. These included Clophens produced in Germany, Aroclor (USA), Kanechlors (Japan) and Sovol (Russia). They found that the dominance of CB 101, CB 138 and CB 153, particularly at the northern stations, points to global emission sources, namely of Clophen A60 and Aroclor 1260, as the main sources.



#### 4. Conclusions

On the basis of contaminant burial fluxes, inventories and congener composition, we conclude that the western Barents Sea is a sink for PCBs supplied by long-range atmospheric transport from lower latitude sources (Breivik et al. 2002), but other sources may also be important, e.g. sea ice melting. For PAHs, proximity to combustion sources is the major factor controlling both the distribution and composition patterns in sediments. The levels of both groups of volatile organic compounds are relatively low compared to areas adjacent to industrial activities. For the heavily industrialized Guba Pechenga area of north-west Russia, Savinov et al. (2003) report  $\sum 11$  PCB levels in surface sediments reaching  $38 \text{ ng g}^{-1} \text{ d.w}^{-1}$  ( $\sum 11$  PCBs = congeners measured in the present study + CB 31, CB 105, CB 156 and CB 209). The PCB compositions in sediments from the SE Barents Sea and Kara Sea have also been shown to reflect the predominant influence of local PCB sources (Savinov et al. 2003, Skotvold & Savinov 2003).

The levels of PAHs in sediments reported in the present investigation are well below pollution levels that would raise concerns for marine organisms or human health. BKF, together with BAA, BAP, IND and DBA, are known carcinogenic compounds; the sum of these is designated as CPAH (Savinov et al. 2003). In the present study, maximum CPAH concentrations are  $61 \text{ ng g}^{-1} \text{ d.w}^{-1}$  (station I, layer 0–1 cm), a value consistent with surface sediment levels from other areas in the region, e.g. the White Sea ( $< 150 \text{ ng g}^{-1} \text{ d.w}^{-1}$  – Savinov et al. 2000), Kara Sea ( $< 110 \text{ ng g}^{-1} \text{ d.w}^{-1}$  – Dahle et al. 2003) and the coastal Barents Sea ( $< 240 \text{ ng g}^{-1} \text{ d.w}^{-1}$  Savinov et al. 2003). In contrast, CPAH levels in several Norwegian fjords affected by direct discharges from aluminium and manganese-alloy smelters are considerably higher ( $\sim 800 \times 10^3 \text{ ng g}^{-1} \text{ d.w}^{-1}$ ) (Næs & Oug 1998). Higher concentrations have also been detected in the vicinity of Guba Pechenga (up to  $\sim 2500 \text{ ng g}^{-1} \text{ d.w}^{-1}$ ) (Savinov et al. 2003). The combined influences of sediment mixing ( $< 0.1 \text{ cm}^2 \text{ yr}^{-1}$ ) and low sedimentation velocities ( $< 0.1 \text{ mm yr}^{-1}$ ) in the western Barents Sea (Carroll et al. 2008b, Zaborska et al. 2008), restrict the temporal resolution of down-core changes in contaminant concentrations. Mixed depth intervals of approximately 2 cm resulted in homogenization of contaminant concentrations over a time-scale of  $\sim 50$  years. The inclusion of sedimentation dynamics in this study provides an improved context for interpreting the temporal trends and the evaluation of spatial distribution patterns of contaminants supplied to the western Barents Sea.

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