

Concentrations and origin of polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) in sediments of western Spitsbergen fjords (Kongsfjorden, Hornsund, and Adventfjorden)

Anna Pouch · Agata Zaborska · Ksenia Pazdro

Received: 20 September 2016 / Accepted: 15 February 2017 / Published online: 21 March 2017
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Abstract Contaminant profiles in sediment cores represent valuable natural archives of environmental contamination, by which contaminant sources and historical changes in contaminant input and cycling may be recognized. In the present study, we discuss the sedimentary profiles and historical trends of organic contaminants — polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) — in three fjords of the Svalbard archipelago differing in environmental conditions and anthropogenic impact. The obtained results revealed no significant differences between the fjords Hornsund and Kongsfjorden, in the average levels of the analyzed contaminants. Levels ranging from 0.05 to 1.47 ng/g d.w. for $\sum 7$ PCBs and from 37.3 to 1973 ng/g d.w. for $\sum 12$ PAHs were measured. The observed spatial and temporal differences in contaminant levels are rather related to local variations in the fjords associated with the location of sampling stations. Higher concentrations of the $\sum 7$ PCBs exceeding 1.00 ng/g d.w. were measured in sediment cores collected in the inner parts of both fjords, which remain under the influence of melting glacier outflows. Important concentrations of these contaminants were noticed in layers deposited

recently, suggesting intensive supply of these substances from secondary sources. The observed levels are generally low and well below known established no effect levels. Only the concentration of fluoranthene exceeded the threshold effect level at several sampling stations. Moreover, fluoranthene concentrations in almost all Adventfjorden sediment layer samples were above probable effect levels, which can indicate a risk of adverse effects in exposed benthic organisms. The fluoranthene/pyrene and phenanthrene/anthracene ratios, which are used for identification of hydrocarbon sources, suggest a dominance of PAHs of pyrolytic genesis in Kongsfjorden and Hornsund. In Adventfjorden, hydrocarbons of petrogenic origin were predominant. However, other sources like coal dust from stores on land are also possible at this location.

Keywords Polychlorinated biphenyls (PCBs) · Polycyclic aromatic hydrocarbon (PAHs) · Contamination · Sediments · Arctic · Spitsbergen

Introduction

Despite being far away from major sources of pollution, the Arctic has been recognized as a region contaminated by several harmful substances of anthropogenic origin (AMAP 2004; AMAP 2009). Contaminants like heavy metals, radionuclides, and organic compounds are detectable in sea ice, air, oceanic waters, sediments, and marine organisms, especially top predators (Macdonald et al. 2005). The sources of contaminants in the polar

Electronic supplementary material The online version of this article (doi:10.1007/s10661-017-5858-x) contains supplementary material, which is available to authorized users.

A. Pouch (✉) · A. Zaborska · K. Pazdro
Institute of Oceanology, Polish Academy of Sciences, Sopot,
Poland
e-mail: apouch@iopan.gda.pl

region can be divided into two groups: local and global. The local sources include coal mines, the metallurgic industry, tourism, coal-fired power stations, and sewage from settlements (AMAP 2009). The main transport pathways of contaminants from global sources include: long range transport by the atmosphere, sea currents, sea ice drift, and rivers (Iwata et al. 1993; Lohmann et al. 2007). Cold-condensation allows deposition of semi-volatile and volatile contaminants to the Arctic multiple times from the warm regions of the Earth. In relation to sea currents, the European part of the Arctic receives contaminants emitted in both western Europe and America by way of the Gulf Stream—the North Atlantic Current. This transport pathway is slow but particularly efficient for heavier contaminants sorbed on suspended matter and for contaminants dissolved in water (Nizzetto et al. 2008; Schulz-Bull et al. 1998). Moreover, contaminants may be transported to the Arctic by rivers. The largest rivers which discharge freshwater to the Arctic Ocean are the Ob, Yenisei, and Lena (Dahle et al. 2003; Macdonald et al. 2005). These rivers' catchment areas include many diffuse contaminant sources such as agricultural runoff and discharges of municipal and industrial sewage from regions of Asia which are heavily populated and rich in mining activity. In addition, contaminants are entrained by sea ice during the freezing process and are also encapsulated by the thickening ice. Sea ice transports pollutants from industrial Siberian areas to the Arctic (Pavlov et al. 2004; Pfirman et al. 1995; Pouch and Zaborska 2015).

A group of contaminants that poses several negative effects to the environment are persistent organic pollutants (POPs) which, among others, include organochlorine compounds like polychlorinated biphenyls (PCBs). PCBs, which were widely used as dielectric fluids in transformers and capacitors, are emitted to the environment only from anthropogenic sources. In the early 1970s both production and usage of these compounds was curtailed, as several compounds had been identified as hormone disruptors (Damstra et al. 2002; Vos et al. 2000). At present, PCBs enter the environment by evaporation, spills, elution from electronic transformers, during incineration, during disposal of industrial waste water and during storage of waste (Gioia et al. 2011). Although no longer in use, or banned under the Stockholm Convention on Persistent Organic Pollutants (<http://chm.pops.int/>), these compounds are ubiquitous in the Arctic environment due to their semi-

volatility, chemical persistence and strong tendency toward bioaccumulation (Macdonald et al. 2000; Pućko et al. 2015). Of 209 PCB congeners, dioxin-like compounds and indicator 7 PCBs are usually studied in a routine way. Another large group of contaminants detected in the Arctic environment are polycyclic aromatic hydrocarbons (PAHs). They are not classified as legacy POPs now included in the Stockholm Convention on Persistent Organic Pollutants 2011, but often exhibit similar environmental properties. Natural sources of PAHs include forest fires, volcanic eruptions and diagenesis (Wang et al. 2007), while anthropogenic sources include coal and wood burning and petrol and diesel oil combustion (Mostafa et al. 2009). Some PCBs and PAHs are carcinogens and toxic to all living organisms, and therefore pose a risk to the natural environment of the Arctic (Walker 1996; AMAP 2009).

Much research has been conducted to investigate PAHs and PCBs in different Arctic environmental samples. The presence of contaminants in all environmental components, their resistance to biodegradation, susceptibility to bioaccumulation and carcinogenicity mean that POPs are of great importance in environmental studies. Until now most studies have concerned mostly their long-term atmospheric transport to the Arctic and determination of their level in Arctic biota (Borga and Di Guardo 2005; Li and Macdonald 2005; Julshamn et al. 2013; Zhu et al. 2015; Evenset et al. 2016). Sediments are an important abiotic component of the marine ecosystem and serve as an important reservoir for POPs in the ecosystem (Hong et al. 2012). An extensive surface and porous structure allows sediments to be treated as a natural sorbent filtering out a variety of components. Contaminants like POPs have hydrophobic properties and thus have a tendency to bioaccumulate and/or to adsorb on organic matter and very fine mineral particles in suspension. Thus, a part of these contaminants is accumulated by marine organisms directly from the water column and the other part is deposited on the sea bottom. Deposited contaminants may be re-introduced to the water column and again be bioavailable to organisms. Contaminant profiles in sediment cores represent extremely valuable natural archives of environmental contamination, by which contaminant sources and historical changes in contaminant input and cycling may be recognized. For example, aquatic sediments have long been recognized as an available record of atmospheric deposition (Rose et al. 2004; Walker et al. 2009). Moreover, based on this

information, contaminant concentrations in surface sediments can be projected into the future (Evenset et al. 2007; Kuzyk et al. 2005). This is particularly important in the context of recent global climate changes, manifesting most strongly in the polar regions. Global climate change may alter contaminant transport pathways, contaminant fate, and therefore routes of exposure for Arctic wildlife and humans (Macdonald et al. 2005).

Despite this, very few studies on sedimentary profiles and the historical trend of PCBs and PAHs are yet available. So far, with a few exceptions (Yunker et al. 2015; Zaborska et al. 2011), most studies have focused on surface sediments of Svalbard fjords (Sapota et al. 2009; Evenset et al. 2009; Jiao et al. 2009; NGU/NVE, unpubl. data). The levels of PCB concentrations in marine sediments in Isfjorden, near to settlements (Pyramiden, Barentsburg, Longyearbyen), has been presented in the report "PCB on Svalbard" (Kallenborn et al. 2011).

The aim of this study was to investigate the historical trends of PCBs and PAHs in sediment cores collected from three fjords of the Svalbard archipelago differing in environmental conditions and anthropogenic impact. Svalbard is an archipelago that is a readily accessible Arctic research site, and its fjords are influenced by different water currents, melting sea ice, melting glaciers, and several sources of local pollution. In the present study the concentrations of certain PCBs (CB28, CB52, CB101, dICB118, CB138, CB153, CB180) and PAHs (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(g,h,i)perylene (BP), and indeno(1,2,3-c,d)pyrene (IND)) were studied in sediment cores collected along the fjords' axis (from the inner glacier bays to external basins). Hornsund (influenced by cold Arctic waters), with 12 melting glaciers and little human activity, and Kongsfjorden (influenced by warm Atlantic waters), with 9 glaciers, relatively high tourist activity and coal mine closed since 1963 (in Ny-Ålesund), were compared. Furthermore, to assess the local sources of organic pollutants, sediment cores from Adventfjorden near Longyearbyen town were collected. Simultaneous analysis of the sediment accumulation rate for the same stations (Zaborska, in prep.) allowed us to determine the history of POP accumulation. The relationships between the analyzed contaminant levels and selected

sediment characteristics were also explored. The FLT/PYR and PHE/ANT ratios were used for identification of hydrocarbon sources.

Materials and methods

The study area

The study area included fjords situated on the West Spitsbergen coast influenced by two water masses: relatively warm and saline Atlantic water (AW) and cold arctic water masses (ARW). AW is transported by the West Spitsbergen Current (WSC), an extension of the Norwegian Atlantic Current, while ARW is transported by the Sørkapp Current originating from the Barents Sea (Walczowski 2014; Piechura and Walczowski 2009). Specifically, the ARW influences the Hornsund fjord, which therefore presents cooler temperatures and lower salinity in comparison to Kongsfjord, where the fjord water is mainly influenced by the warmer Atlantic current. In Kongsfjorden, typical, undisturbed ARW is not observed and the fjord is filled with a mixture of ARW and AW (Svendsen et al. 2002; Cottier et al. 2005). Due to their topographic structure, both fjords may be divided into outer and inner parts. The outer parts are deeper and are more influenced by ocean currents. In the inner parts a strong influence of melting glacier outflows is observed (Wesławski et al. 1991; Svendsen et al. 2002). Sedimentation and accumulation processes in both fjords are highly impacted by the flux of (mainly mineral) material from the glacier outflows (Görlich et al. 1987; Svendsen et al. 2002). The coarsest sediment grains are deposited close to the glacier front while deeper fjord regions are dominated by silts and clays (Görlich et al. 1987; Zaborska et al. 2006). In central parts of the fjords the sediment accumulation rate is about half as low in Hornsund (0.23 ± 0.02 cm/year) as in Kongsfjorden (0.39 ± 0.03 cm/year) (Zaborska et al. 2016). The higher accumulation rate in Kongsfjorden is linked to large particulate matter flux from glacial meltwater discharge (Svendsen et al. 2002).

Hornsund (H) is the southernmost fjord in Spitsbergen. The length of the fjord is about 34 km, and the average depth is about 90 m (Błaszczuk et al. 2013). The fjord is strongly influenced by cold waters originating from Storfiorden or/and the Barents Sea. AW is mainly observed at the bottom of the outer part,

and the inflow of AW to the inner fjord is limited by the shallow sills. About 67% of the whole drainage basin area is covered by glaciers (Błaszczuk et al. 2013). The inner part (Brepollen) is under the impact of the huge melting glaciers Hornbreen and Svalisbreen. In Hornsund, the main basin is often covered with drift ice carried by the Arctic Sørkapp Current from the Barents Sea. Generally, the ice season in Hornsund lasts to May or June. Hornsund is a relatively pristine area (national park) characterized by little tourist activity. The Polish Polar Station is located near the entrance to Hornsund fjord, on its northern shore, at Isbjørnhamna.

The open fjord Kongsfjorden (K) is 20 km long and from 4 to 10 km wide, and is situated in the northwestern part of Svalbard (Hop et al. 2002; Svendsen et al. 2002). The fjord is divided into an inner basin with 50–60 m depth on average, and an outer part which is 200–300 m in depth on average. The Atlantic water influence makes this fjord sub-Arctic rather than Arctic. About 77% of the area of the drainage basin is covered by glaciers (Svendsen et al. 2002). The inner part of the fjord is under the impact of glaciers, the largest of which are Kongsvegen, Kronebreen, and Kongsbreen. In Kongsfjorden, fewer glaciers are present but they are more intensively retreating compared to the glaciers in Hornsund. In the inner part of the fjord, fluvio-glacial outflows are present (Svendsen et al. 2002). On the southern shore of Kongsfjorden, coal mining was carried out in the past, and since 1990 Ny-Ålesund, one of the world's northernmost human settlements, has developed. It is believed that human settlements has little impact to fjord environment but some residual contaminants exist in the vicinity of mining sites (Hop et al. 2001; van den Heuvel-Greve et al. 2016).

Adventfjorden (AD) is a southern branch of Isfjorden. The fjord is about 8 km long and about 3.5 km wide, and the depth of the fjord is up to 80 m. Two braided rivers (Longyear River and Advent River) are situated in the inner part of the fjord and transport melt waters from glaciers situated several kilometers from the coast (Wiedmann et al. 2016). The fjord water masses are influenced by warm, saline Atlantic waters that enter the fjord from the shelf. Longyearbyen, the largest settlement and the administrative centre of Svalbard, with a population of more than 2000, is situated on the shore of Adventfjorden. It had been previously impacted by intense mining activity, and it is still a transshipment dock for the coal extracted from Longyearbyen mine.

Field sampling

Sediment samples were collected from the r/v "Oceania" using a Gemax corer or Niemistö corer, at ten sampling stations in three fjords (Kongsfjorden (K), Hornsund (H) and Adventfjorden (AD)), in 2007 (H1, H2, H3, and H4 stations), 2013 (H5, K3, K4, AD stations), and 2014 (K1, K2 stations). Two sediment cores were collected at each station. The map of the study area and the research stations is presented in Fig. 1. K4 and H5 stations were located near glaciers and were chosen to represent regions of glacial runoff. Stations K2, K3, H2, and H4 were located in the central parts of the fjords, while stations H1 and K1 represent the outer parts of the fjords. Additionally, sediment samples were collected from Adventfjorden near Longyearbyen town to assess the significance of local pollution sources. Sediment cores were sliced at 1-cm intervals and frozen immediately. The samples were transported and analyzed in the laboratory of the Marine Chemistry Department, Institute of Oceanology, Polish Academy of Sciences.

Samples preparation and sediment characterization

The samples were freeze-dried (Christ Beta A apparatus), homogenized using a mortar and pestle, and split into two sub-samples. One of these sub-samples was devoted to determining sediment properties, and the other sub-sample was analyzed for concentration of PCBs and PAHs. Sediment characterization included water content, organic matter content, and particle size. Water content was calculated based on loss during the freeze-drying process. The organic matter content in sediment samples was measured by loss on ignition (LOI). Dry and homogenous samples of the sediments were weighed before and after heating for 5 h at 550 °C (Maher 1998). Pelite fraction content was determined by wet sieving (Guy 1969). Sediment dating, based on ^{210}Pb -analyses, was performed. The same sediment cores were analyzed for ^{210}Pb , and dating results will be presented in another study (Zaborska et al., in prep.). Indirect measurement of ^{210}Pb (^{210}Po) by alpha spectrometry was performed. For separation of ^{210}Po the method presented by Flynn (1968) was used. All analytical details are presented in Zaborska et al. (2007). Quality control was based on measurements of reference standards (IAEA-300 and IAEA-326).

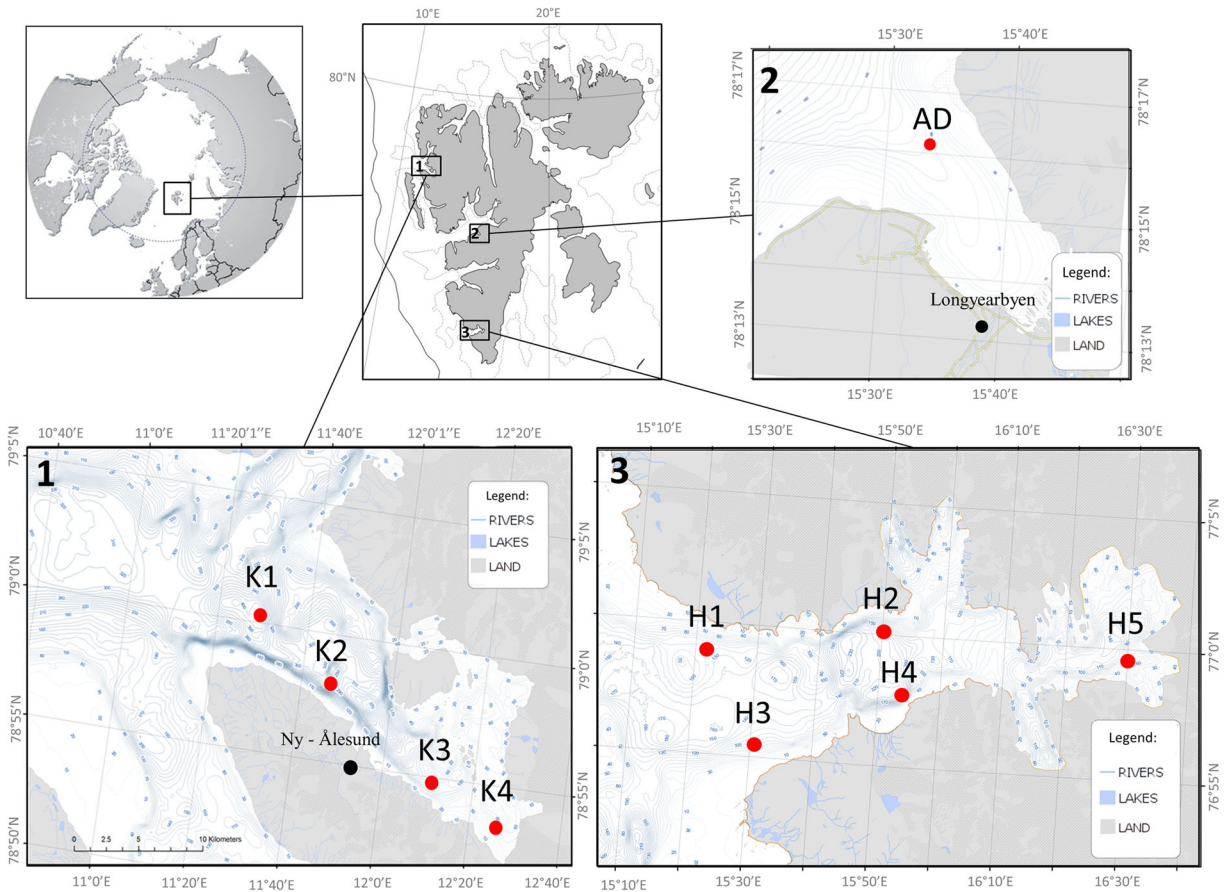


Fig. 1 Location of sampling stations in the Kongsfjorden (K1–K4), Hornsund (H1–H5), and the Adventfjorden (AD) (maps after iopan.gda.pl/ekologia/maps.html, modified)

PCB and PAH determination

Determination of PCBs and PAHs was performed according to procedures described elsewhere (Behar et al. 1989; Zaborska et al. 2011). Briefly, a sub-sample of 5 g was first treated by triplicate extraction with methylene chloride in an ultrasonic bath. An internal standard (octachloronaphthalene) was added to each sample prior to extraction to measure the recovery efficiency of the entire process. The extracts were combined and concentrated, and then elemental sulfur was removed from the extract using copper powder activated with hydrochloric acid. Further clean-up and fractionation were performed by absorption chromatography on silica gel and aluminum oxide (both activated with 5% water). Hexane was used to extract PCBs and a mixture of hexane:methylene chloride (9:1 v/v) to extract PAHs. The extracts were concentrated under a gentle flow of nitrogen. A gas

chromatograph (Shimadzu GC 17A) was used for qualitative and quantitative analysis of target analytes. The applied conditions of the chromatographic analysis were described in Pazdro (2007). A ZB-5 column (Phenomenex; 30 m × 0.25 mm id with 0.25 μm film thickness) was used. High purity (99.99999%) helium was used as a carrier gas with a flow rate of 1.2 ml/min for PAH analyses and a flow rate 1.5 ml/min for PCB analyses. The temperature of the injection port was 280 °C. The temperature programming was as follows: 50 °C held for 1 min, then 50 °C to 240 °C (6°/min), then 240 °C to 310 °C (20°/min) and 310 °C (25 min) for PAH analyses; and 100 °C held for 1.5 min, then 100 °C to 140 °C (6°/min), then 140 °C to 250 °C (2.5°/min), then 250 °C to 310 °C for PCB analyses. An electron capture detector (ECD) was applied for PCB detection and a flame ionization detector (FID) for PAH detection.

Identification of the 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 concentrations of the analytes in the samples were calculated by using external five-point calibration curves plotted for each compound in the linear range of the detector's response. A standard solution of PAHs containing 16 compounds in dichloromethane (each at 2000 $\mu\text{g/ml}$) was used, as was a standard solution containing 7 compounds from the PCB group in isooctane each at 10 $\mu\text{g/ml}$.

Quality assurance/quality control

A procedural blank analysis was performed with every 5 samples to monitor interferences. Sediment samples were heated at 550 $^{\circ}\text{C}$ for 5 h then subjected to the same extraction and clean-up procedures as real samples. In order to test the accuracy of the quantification method the entire analytical procedure was applied several times to a certified marine sediment (IAEA-383). The quantification limits, measured as average blanks plus six standard deviations, were for PAHs and PCBs in the range of 80–220 and 10–50 pg/g respectively. Recoveries of individual compounds ranged from 77 ± 5 to $104 \pm 5\%$ depending on the compound. Naphthalene, acenaphthene and acenaphthylene were excluded from the list of the PAHs studied due to low accuracy.

Statistics

The statistical analysis program, STATISTICA 10, was used for the calculation of means and standard deviations and to study correlations between concentrations of PCBs and PAHs and organic matter content and fraction $<63 \mu\text{m}$ content. Since results did not fulfill the assumptions of the Pearson correlation (normality of distribution), the non-parametric Spearman correlation was applied to the database of obtained results.

Results and discussion

Sediment properties

The highest content of water was measured in the surface sediment layers, and then the values decreased with

sediment depth. The concentration of organic matter ranged from 3.2 to 11.8% (Table 1). The high content of organic matter in sediments at stations H2 and H4 may be explained by the fact that these stations are located near a steep slope of the seabed (Drewnik et al. 2016). Areas located at the bottom of a slope accumulate sedimentary material, mainly fine grained sediments and organic matter (Zaborska et al. 2006). At stations H5 and K4, located near the glaciers, the outflow concentrations of organic matter were lower. Organic matter contents in sediments measured in this study generally agree with previously published data. Similar results have been found in Spitsbergen fjords by other researchers: Kongsfjorden (0.1–9.4%) (Zaborska et al. 2006), Woodfjorden (2.8–5.9%), Wijdenfjorden (1.6–5.0%), and Bockfjorden (3.2%) (Sapota et al. 2009). The content of fine sediment fraction ($<63 \mu\text{m}$) ranged from 81.6 to 99.5%. (Table 1). According to these results, the investigated sediments were classified as silts and clays. The accumulation rates differed between the sampling sites within each fjord and between particular fjords. Lower sediment accumulation rates ranging from 0.18 to 0.28 mm/year were determined at stations H1, H3, K1, and K2. At stations H2, H4, and K3, accumulation rates were 0.36, 0.40, and 0.47 mm/year respectively. The highest accumulation rate, reaching 0.69 mm/year , was measured at station H5 in Hornsund, close to the glacier (Zaborska et al. in prep.). Due to the very high accumulation rate near the Kongsbreen glacier in Kongsfjorden, it was impossible to date sediments at station K4. Generally, the accumulation rates were higher at stations in the inner parts of the fjords, close to the melting glaciers, due to the large influx of material from glaciers which are eroding bedrock. The sediment accumulation rates were typical for Spitsbergen fjords (Kuliński et al. 2014; Zaborska et al. 2006; Zajączkowski et al. 2004).

Spatial and temporal distribution of PCBs and PAHs in the sediment samples

In Hornsund $\sum 7$ PCB concentrations in sediments ranged, depending on location and sediments layer, from 0.05 ± 0.001 to $1.47 \pm 0.09 \text{ ng/g}$ dry weight (d.w.) The highest value of the $\sum 7$ PCB concentrations was observed at station H5, in the glacial bay (Fig. 2). In Kongsfjorden sediment concentrations of $\sum 7$ PCBs ranged from 0.27 ± 0.03 to $1.41 \pm 0.13 \text{ ng/g}$ d.w. (Fig. 3). Concentrations of $\sum 7$ PCBs higher than

Table 1 Sediment properties: water content, organic matter (M_{org}) content, and fine sediment (<63 μm) fraction, together with the mean year of sediment layer deposition. The age of sediment layers of the core collected at station K4 was not determined due to very high sediment accumulation rate

Station	Layer [cm]	Deposition year	Water content [%]	Morg content [%]	Fine fraction (< 63 μm) [%]
K1 79°00'03"N 11°26'17"E	0-1	2012	67.3	10.2	94.7
	1-2	2007	59.3	11.1	90.4
	2-3	2003	55.7	10.1	95.2
	3-4	1998	47.5	10.4	96.5
	4-5	1994	39.7	10.1	97.0
	10-12	1974	46.9	9.5	97.4
K2 78°57'58"N 11°43'11"E	18-20	1940	44.5	11.8	88.6
	0-1	2012	62.9	10.4	94.2
	1-2	2009	54.9	9.5	96.0
	2-3	2006	49.7	9.1	91.9
	3-4	2003	49.6	9.6	95.4
	4-5	2000	48.0	9.2	95.4
K3 78°55'03"N 12°08'31"E	10-12	1980	50.7	10.3	95.2
	24-26	1944	42.9	7.9	96.6
	0-1	2012	55.6	5.3	94.7
	1-2	2010	50.0	7.6	95.2
	2-3	2008	41.9	6.7	95.9
	3-4	2006	40.9	6.9	96.3
K4 78°53'02"N 12°27'07"E	4-5	2004	39.5	5.5	97.0
	10-12	1994	37.2	8.5	97.5
	26-28	1971	34.0	9.1	93.2
	0-1	-	64.2	6.3	98.2
	1-2	-	64.5	5.9	99.4
	2-3	-	56.4	7.8	98.9
AD 78°15'58"N 15°30'40"E	3-4	-	45.2	6.7	99.5
	4-5	-	50.1	5.4	99.1
	10-12	-	32.9	5.4	98.3
	38-40	-	29.7	5.2	97.5
	0-1	2012	53.6	7.2	93.9
	1-2	2009	44.9	6.6	88.9
H1 76°58'57"N 15°23'59"E	2-3	2005	40.0	7.0	91.6
	3-4	2002	38.0	7.0	88.3
	4-5	1999	34.8	6.8	84.2
	10-12	1981	31.7	6.2	81.6
	18-20	1960	28.6	6.0	85.4
	0-1	2004	55.5	7.2	96.4
H2 77°00'00"N 15°50'00"E	1-2	1999	53.9	7.0	96.6
	3-4	1988	53.5	7.6	94.6
	4-5	1982	50.1	7.2	92.4
	5-6	1976	48.4	5.9	94.4
	7-8	1965	45.9	4.5	93.4
H3 76°55'59"N 15°33'51"E	9-10	1954	43.1	7.0	92.6
	12-14	1935	39.2	6.0	91.2
	16-18	1913	36.5	6.3	94.7
	0-1	2006	58.5	9.5	96.0
	1-2	2001	54.6	9.1	95.6
	2-3	1997	53.1	9.0	98.7
	3-4	1992	53.0	10.6	96.6
	4-5	1988	50.5	8.9	95.4
	5-6	1978	49.7	8.2	94.8
	7-8	1969	47.9	7.4	98.3
H4 76°58'07"N 15°54'56"E	9-10	1957	45.6	9.0	95.6
	14-16	1944	43.9	11.0	97.0
	0-1	2004	63.5	7.8	96.5
	1-2	1999	62.4	7.6	96.0
	2-3	1993	60.3	7.1	96.2
	3-4	1988	57.3	7.4	95.8
	4-5	1982	56.9	5.8	95.7
	5-6	1976	54.5	7.0	95.7
	7-8	1965	52.7	7.7	95.5
	9-10	1954	50.9	7.0	95.3
H5 77°00'20"N 16°28'52"E	14-16	1924	47.8	7.1	95.0
	0-1	2006	65.9	9.4	83.8
	1-2	3002	63.4	9.5	96.0
	2-3	3001	62.9	9.0	95.2
	3-4	1998	60.0	8.4	93.7
	4-5	1996	57.8	7.9	96.2
	7-8	1988	56.1	5.2	97.4
	9-10	1983	54.1	4.4	93.9
	0-1	2012	64.0	6.2	93.1
	1-2	2001	63.7	6.5	93.8
	2-3	2009	58.9	7.0	93.7
	3-5	2007	45.2	8.9	93.7
	10-12	1999	39.7	6.5	93.8
	22-24	1986	34.9	3.2	93.9

Table 1 (continued)

1.0 ng/g d.w. were measured in two sediment layers of the core collected in the inner fjord (K4). The $\Sigma 7$ PCB concentrations in Adventfjorden were similar to those measured in Kongsfjorden and Hornsund, when the

inner stations in these fjords are excluded. The vertical profile of $\sum 7$ PCB concentrations at the station in Adventfjorden show a quite constant concentration of about 0.57 ± 0.04 to 0.83 ± 0.05 ng/g d.w. (Fig. 3). In general, the observed levels of sediment contamination by PCB compounds could be reported as low. The detailed results on individual PCBs are presented in the supplement material (Tab. S1). The concentrations of PCBs measured in the Arctic fjord sediments selected in this study were compared with PCB contents in Svalbard fjords and other Arctic marine sediments reported elsewhere (Table 2). The lowest concentration of PCBs in marine sediment samples was determined in the Bering and Chukchi Sea (0.04 ng/g d.w.). Similar concentrations to those measured in Adventfjorden were reported by the Norwegian Polar Institute in 2005 in Svalbard and Jan Mayen sediments (MOSJ 2016), where the $\sum 7$ PCBs ranged from 0.20 to 1.54 ng/g d.w. Evenset et al. (2009) report that $\sum 7$ PCB-levels in sediment from Billefjorden, near the Russian settlement of Pyramiden, ranged from 1.8 to 20.2 ng/g dw. Furthermore, the highest $\sum 7$ PCB concentrations in Woodfjorden (5.6 ng/g d.w.), Wijdefjorden (3.8 ng/g d.w.), and Bockfjorden (2.9 ng/g d.w.) (Sapota et al. 2009) were higher than the highest values measured in this study. The highest $\sum 7$ PCB values in this study (1.47 ± 0.09 ng/g d.w.) were similar to the highest concentrations of PCBs measured in surface sediments collected in Ny-Ålesund harbor (1.61 ng/g d.w.) (van den Heuvel-Greve et al. 2016).

In Hornsund sediments, the $\sum 12$ PAH values generally ranged from 33.52 ± 2.04 to 453.44 ± 22.79 ng/g d.w. apart from the station H5, located close to a glacier, where the concentrations reached even 1973.57 ± 111.34 ng/g (Fig. 4). In Kongsfjorden sediments, the $\sum 12$ PAH concentrations ranged from 51.93 ± 2.70 to 1482.13 ± 140.22 ng/g d.w. (Fig. 5). The concentrations were lowest at the inner station (K4; close to the glacier) where they ranged from 51.93 ± 2.70 to 707.16 ± 85.93 ng/g d.w., depending on the sediment layer. High concentrations of PAHs (up to 1580.01 ± 77.41 ng/g d.w.) were detected in sediments collected from Adventfjorden (Fig. 5). This may be caused by the fact that this fjord is strongly affected by human activity (storage of coal in the transshipment dock, marine traffic, coal exploitation and combustion at coal power station). Detailed results on individual analytes are presented in the supplement material (Tab. S2).

The concentrations of PAHs have been analyzed in Arctic sediments in different studies (Table 2). Significantly lower concentrations, ranging from 26.23 ± 13.12 to 132.02 ± 66.42 ng/g d.w., of these compounds were observed in sediment samples collected in the western Barents Sea (Zaborska et al. 2011). In surface sediment samples collected in Wijdefjorden, Woodfjorden, and Bockfjorden, the $\sum 16$ PAH concentrations were even lower, up to 50 ng/g d.w. (Sapota et al. 2009). Higher concentrations were measured in the Barents Sea sediments, where values varied from 39 to 4270 ng/g d. w. (Szczybelski et al. 2016). The highest sum of concentrations presented in this study ($\sum 12$ PAH = 1973.57 ± 111.34 ng/g d.w.) was similar to the average total PAH concentrations measured in southern areas of the Norwegian continental shelf (Boitsov et al. 2013). Moreover, the highest concentration presented in this study was similar to the lowest measured in sediments collected along the mainstream waters of the Vistula in the Gulf of Gdańsk (Baltic Sea) (Lubecki and Kowalewska 2010). According to Van den Heuvel-Greve et al. (2016), PAHs in surface sediments collected close to Ny-Ålesund exceed 2000 ng/g d.w., except for sediment collected from Thiisbukta. High levels of PAHs in some locations in Kongsfjorden may be related to old oil spills recorded in Ny-Ålesund, with major spills in 1986 and 1990 (Olsson et al. 1998; van den Heuvel-Greve et al. 2016). The sum of PAH concentrations varied along the sediment layers, and it was impossible to determine a constant PAH concentration that could be assumed as a natural background level of PAHs. There is no data available on the background level of PAHs in Arctic fjords. In the northern Barents Sea, east of Svalbard, total PAH concentrations below 30 ng/g d.w. are regarded as the natural PAH content (e.g., Zaborska et al. 2011).

Marine sediments are an archive of deposited contaminants, and thus in stable conditions, the history of contamination can be retrieved. PAH and PCB concentrations as a function of the sediment layer deposition depth (the middle of the sampled layer) are presented in Figs. 2, 3, 4, and 5. The historical sedimentary PCB concentrations do not exhibit a pattern of increasing concentrations until the 1980s, followed by decreasing concentrations in recent times, as would be expected knowing the history of PCB usage. Rather, both PCB and PAH concentrations increase recently as the highest concentrations of PCBs and PAHs were measured in the sediment layers dated 2011–2013 at stations K4 and H5.

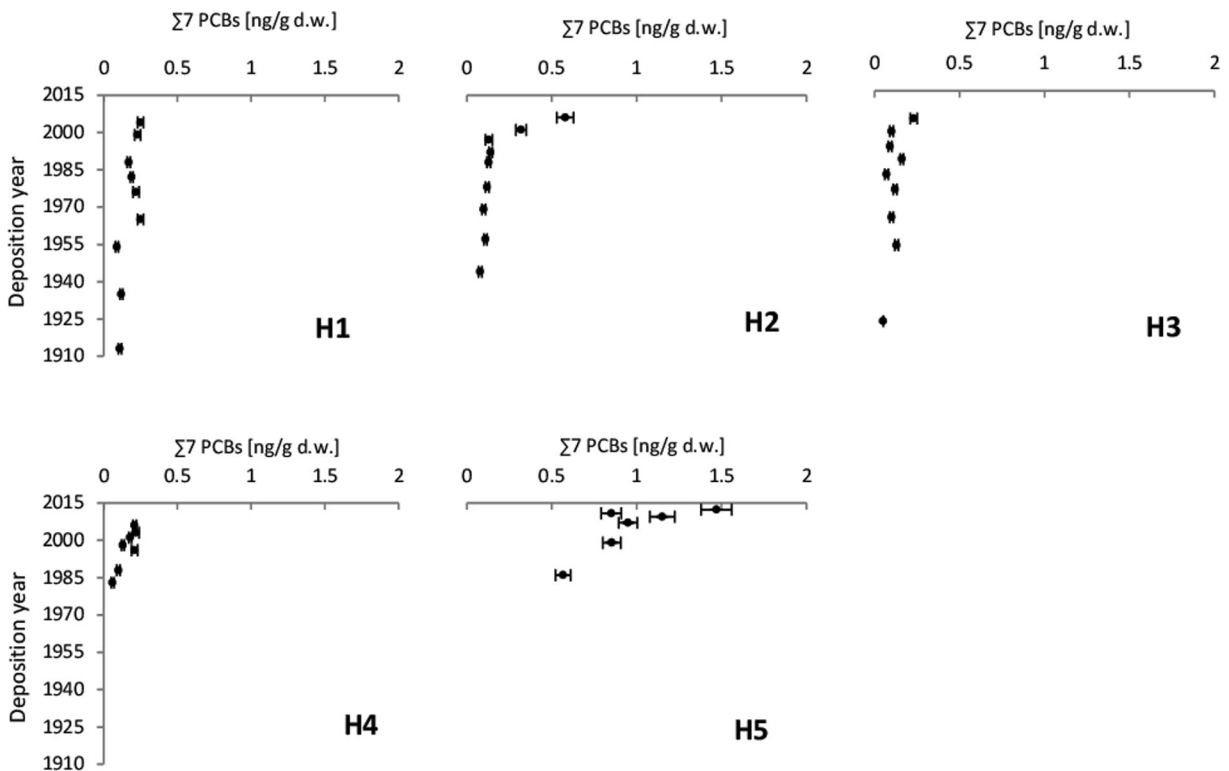


Fig. 2 Concentrations of $\Sigma 7$ PCBs [ng/g d.w.] in sediment cores collected from Hornsund fjord. The error bars show the standard deviation

This phenomenon may be caused by a complexity of processes governing a large number of contaminants in the Svalbard fjords systems. Fjords located in such a remote place are influenced by the disperse transport of contaminants occurring by several transport pathways. Apart from the atmosphere (Dommergue et al. 2010) and marine currents (Lu et al. 2013; Ardini et al. 2016), sea ice (Gustafsson et al. 2005;), and glaciers (Hermanson et al. 2005; Łokas et al. 2016) are recognized as important sources of contaminants to the Svalbard fjords. The flux of contaminants accumulated on the glaciers' surfaces and in permafrost will increase due to climate warming. For example, in Hornsund, about 47 km² of glaciers located at the sea margin retreated from 1986 to 2010 (Błaszczyk et al. 2013). As PAHs are also natural elements of the earth's crust, their elevated concentrations at some stations (e.g., H5) could be connected to natural weathering processes of hydrocarbon rich rocks or even accidental releases as, for example, dust from two large volcanic eruptions which took place in 2010 in Iceland (Karasiński et al. 2014). High PAH concentrations, not followed by PCB concentration increases, were measured in the down-core profile at station AD. This was probably caused by a local source of contamination, namely the activities

in the coal loading dock in Adventfjorden and intense shipping in this area. Although the significance of Svalbard coal extraction is decreasing, declining trends are not yet visible in marine sediments.

The other possible source of contaminants to the Spitsbergen fjords (particularly Hornsund) may be the drift sea ice transported from the Barents Sea (Błaszczyk 2013; Muckenhuber et al. 2016). The literature indicate that levels of $\Sigma 31$ PCBs in sea ice are up to around 30 pg/l (Gustaffson et al. 2005). Concentrations of PCBs of 100 and 3000 pg/l were measured in sea ice in the Laptev Sea (Dethleff et al. 1993). Near the Fram Strait, concentrations of PCB138 in drifting, porous sea ice ranged from 15,500 to 20,300 pg/l (Gaul 1989).

PCB composition and sources

The distribution of individual PCBs in the collected sediment samples is presented in Figs. 6 and 7. In Hornsund sediments, no dominant congener was noticed (Fig. 6). The CB28 congener was dominant in K1, K2, K3 (Kongsfjorden), and AD sediment cores. Light chlorinated biphenyls (CB28 and CB52) constituted from 78.61 to 97.49% at K1 station. In contrast, at station K4, in surface

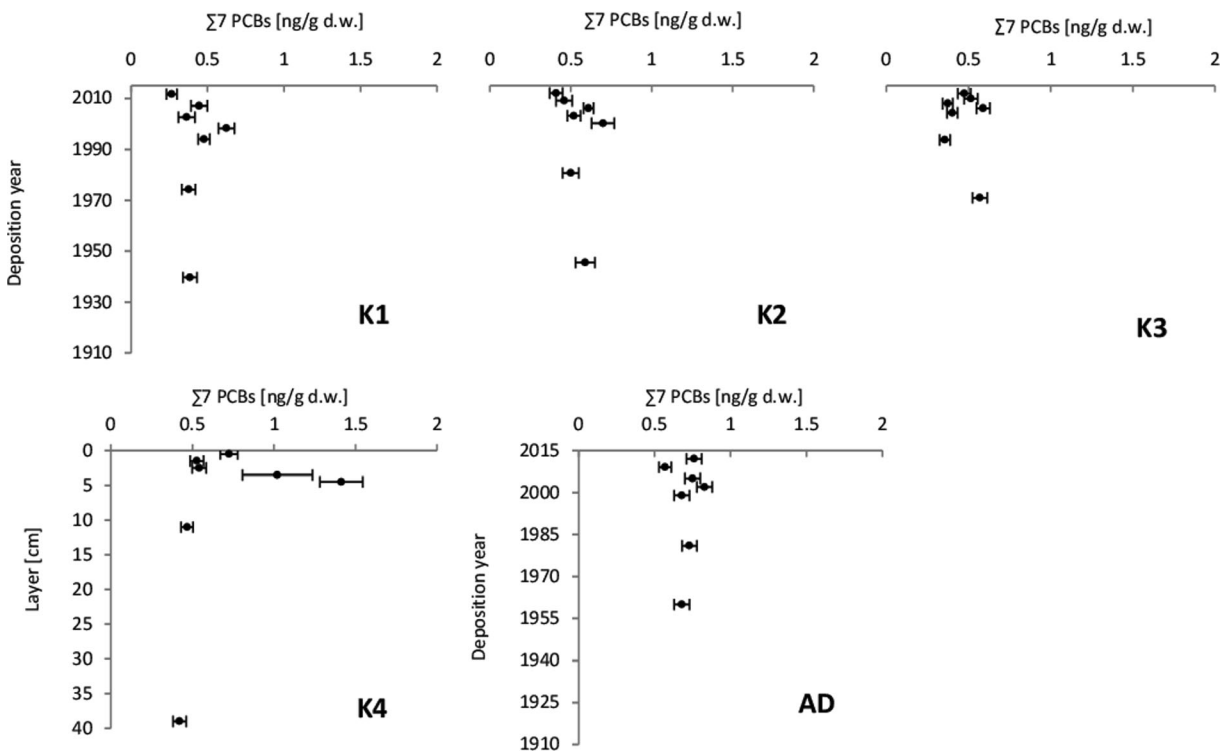


Fig. 3 Concentrations of $\Sigma 7$ PCBs [ng/g d. w.] in sediment cores collected from Kongsfjorden and Adventfjorden. The *error bars* show the standard deviation

sediment layers (<3 cm), CB 28 constituted only from 3.69 to 5.59% and CB153 and CB180 were the dominating congeners (Fig. 7). Generally, heavier PCBs (CB153

and CB138) dominate in the western Barents Sea (Zaborska et al. 2011). CB153 was stated as a dominant congener in sediments collected in Wijdefjorden and

Table 2 Comparison of PCB and PAH concentrations in different Arctic regions

Area	Sediment layer	PCBs (ng/g d.w.)	PAHs (ng/g d.w.)	Reference
Bering and Chukchi Sea	–	$\Sigma 7 = 0.04\text{--}0.08$	–	Iwata et al. 1994
Southwest Barents Sea	0–1	$\Sigma 11 = 1.06\text{--}37.9$	$\Sigma 27 = 151.0\text{--}3257.0$	Savinov et al. 2003
West Barents Sea	0–1	$\Sigma 7 = 0.73 \pm 0.29\text{--}1.54 \pm 0.61$	$\Sigma 12 = 35.29 \pm 17.76\text{--}132.02 \pm 66.42$	Zaborska et al. 2011
Wijdefjorden, Woodfjorden, Bockfjorden, Grøn fjorden	–	$\Sigma 7 < 0.9\text{--}5.6$	$\Sigma 16 < 36$	Sapota et al. 2009
Barentsburg, Pyramiden	–	$\Sigma 7 = 0.74\text{--}5.41$	–	Evenset et al. 2009
Billefjorden	–	$\Sigma 12 = 3.695$	–	NGU/NVE (unpubl. data)
Advents fjorden	–	$\Sigma 7 = 1.8\text{--}20.2$	–	Evenset et al. 2009
Kongsfjorden—reference	–	$\Sigma 7 = 0.25$	–	Evenset et al. 2009
Kongsfjorden (Ny-Ålesund harbor)	0–5	$\Sigma 7 < 0.568$	$\Sigma 16 = 1\text{--}26$	van den Heuvel-Greve et al. 2016
Kongsfjorden (Thiisbukta)	–	$\Sigma 7 < 1.613$	$\Sigma 16 = 476\text{--}2550$	
Kongsfjorden	–	$\Sigma 7 = 0.330$	$\Sigma 16 = 1787$	
Kongsfjorden	–	$\Sigma 7 = 0.00002\text{--}0.13$	$\Sigma 13 = 12\text{--}2315$	Szczybelski et al. 2016

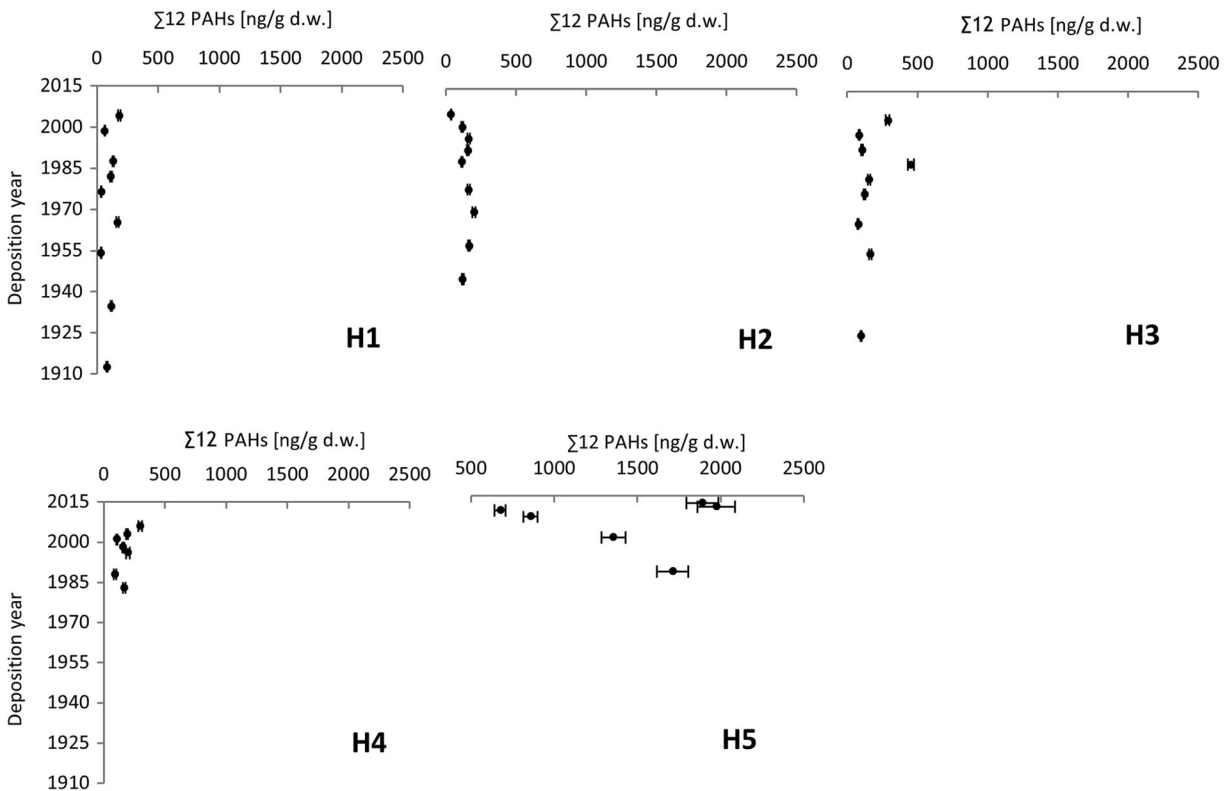


Fig. 4 Concentrations of $\Sigma 12$ PAHs [ng/g d.w.] in sediment cores collected from the Hornsund fjord. The error bars show the standard deviation

Woodfjorden (Sapota et al. 2009). Light PCB congeners (CB20, CB28) also prevail in surface sediments collected from the Bering Sea, the Chukchi Sea, and the Canada Basin (Hong et al. 2012).

During combustion of material containing chlorine atoms, most of the resulting PCB compounds contain a low number of chlorine atoms in the molecule. Such light PCBs are then transported by the atmosphere (Eckhardt et al. 2007); thus, high concentrations of light PCBs may be the effect of direct atmospheric deposition. Stations located in the inner and central parts of the fjords are influenced by an influx of water from melting of glaciers. The higher content of light PCBs at these stations may be caused by transport of highly volatile PCBs accumulated over many years over the glacier ice surface (Hermanson et al. 2005). The prevalence of heavier PCBs may suggest the influence of not only atmospheric but also marine (currents, sea ice) sources of PCBs. The concentrations of PCBs in marine waters were measured by Gioia et al. (2008). The concentrations of individual PCBs in suspended matter increased with increasing degree of chlorination.

PAH composition and sources

The hydrocarbon compounds were divided into two groups to determine the pattern of individual PAH dominance (low molecular PAHs and high molecular PAHs). The fingerprinting method, based on the relative concentration of individual PAHs (% of total PAHs), is widely used to determine the origin of PAHs (Dahle et al. 2003; Lubecki and Kowalewska 2012). PAHs with 2–3 rings are mostly found in the gas phase, not associated with airborne particles, which suggests petrogenic sources (Machado et al. 2009). Dominance of 3- or 4-ring compounds suggests emissions from a coal-fired power plant. Medium and high molecular weight PAHs are typical of transport by air masses from local industries and the by-products of coal and wood burning (Savinov et al. 2000). The 5–6-ring compounds are associated with gasoline combustion emissions. These compounds can be introduced by ships, boats and industrial traffic (Howsam and Jones 1998). In general, a higher proportion of high molecular PAHs is

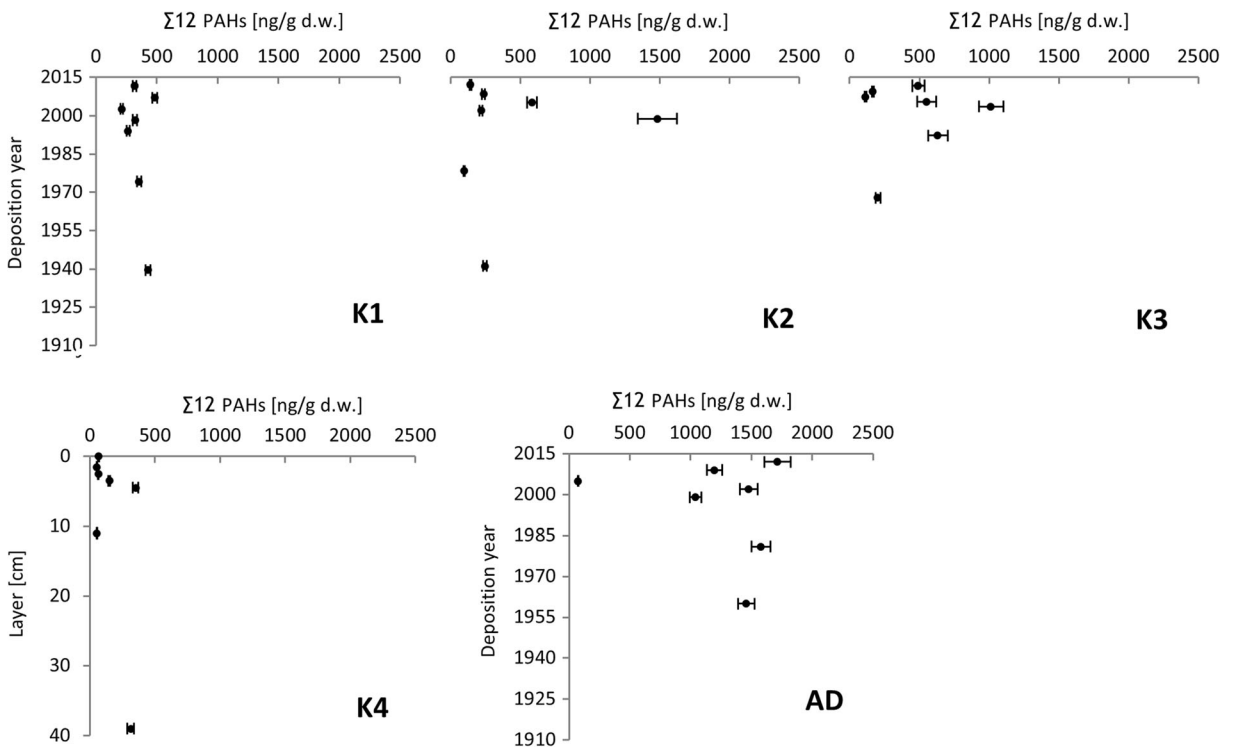


Fig. 5 Concentrations of $\Sigma 12$ PAHs [ng/g d.w.] in sediment cores collected from Kongsfjorden and Adventfjorden. The error bars show the standard deviation

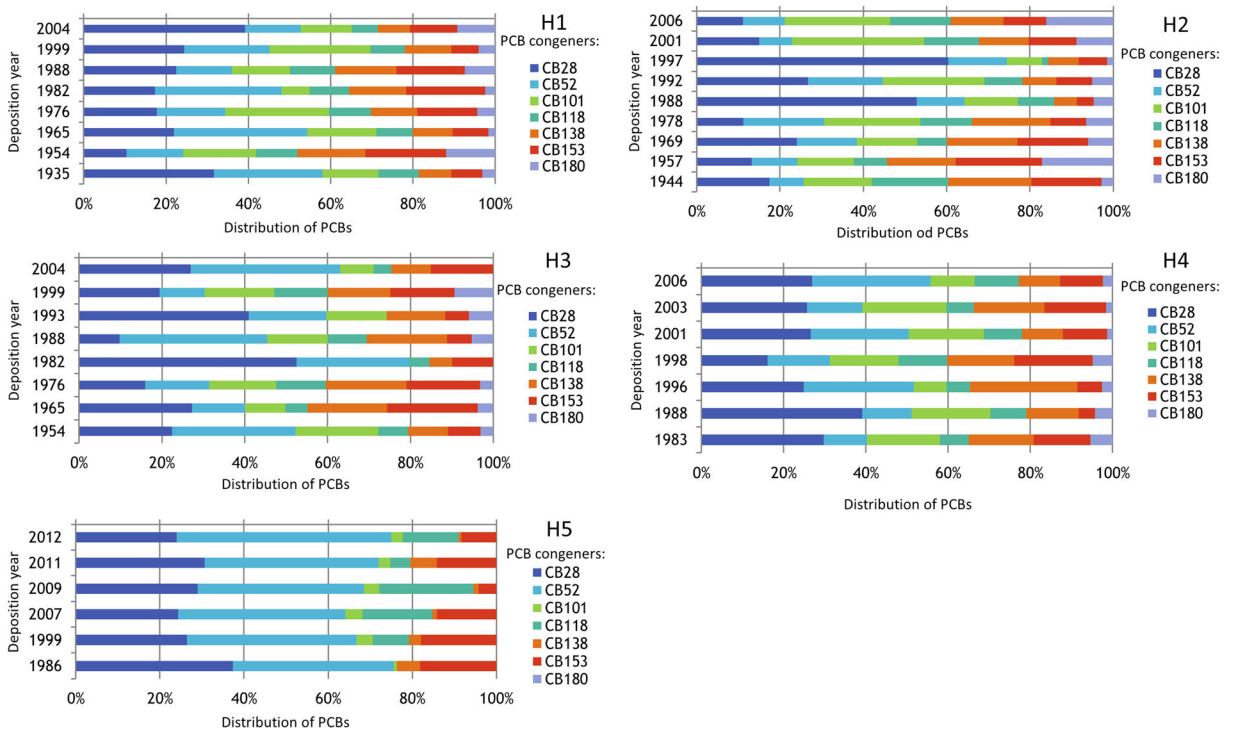


Fig. 6 Percentage content of individual PCBs in sediment samples collected from Homsund

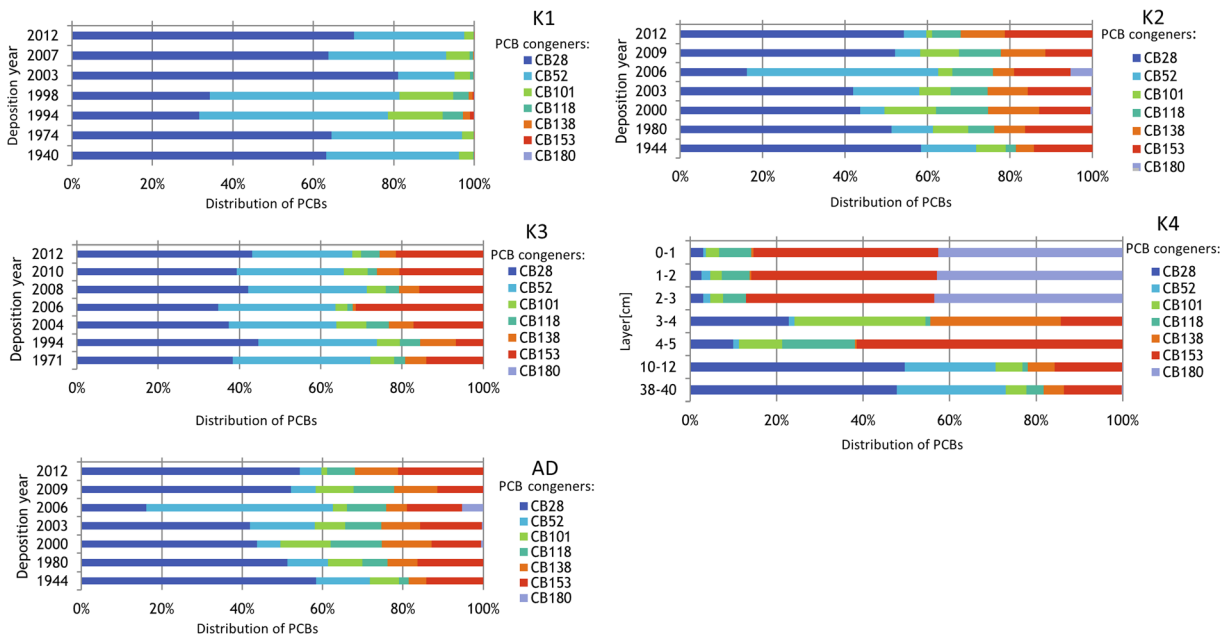


Fig. 7 Percentage content of individual PCBs in sediments samples collected from Kongsfjorden and Adventfjorden

considered to be an indicator of pyrogenic origin, while dominance of low molecular PAHs indicates importance of petrogenic sources. Generally, the results obtained in this study show high concentrations of the low molecular weight PAH group, consisting of FLU, ANT, PHE, FLA, PYR, BAA, and CHR

(Figs. 8 and 9). In Hornsund, at the different stations, the sediments tend to have similar average percentages of compounds with 2–3, 4, 5, and 6 rings in their structure. The PAHs with 2–3 rings (FLU, ANT, PHE) in the molecule were the most dominant in Hornsund sediments and constituted from 32.1 to

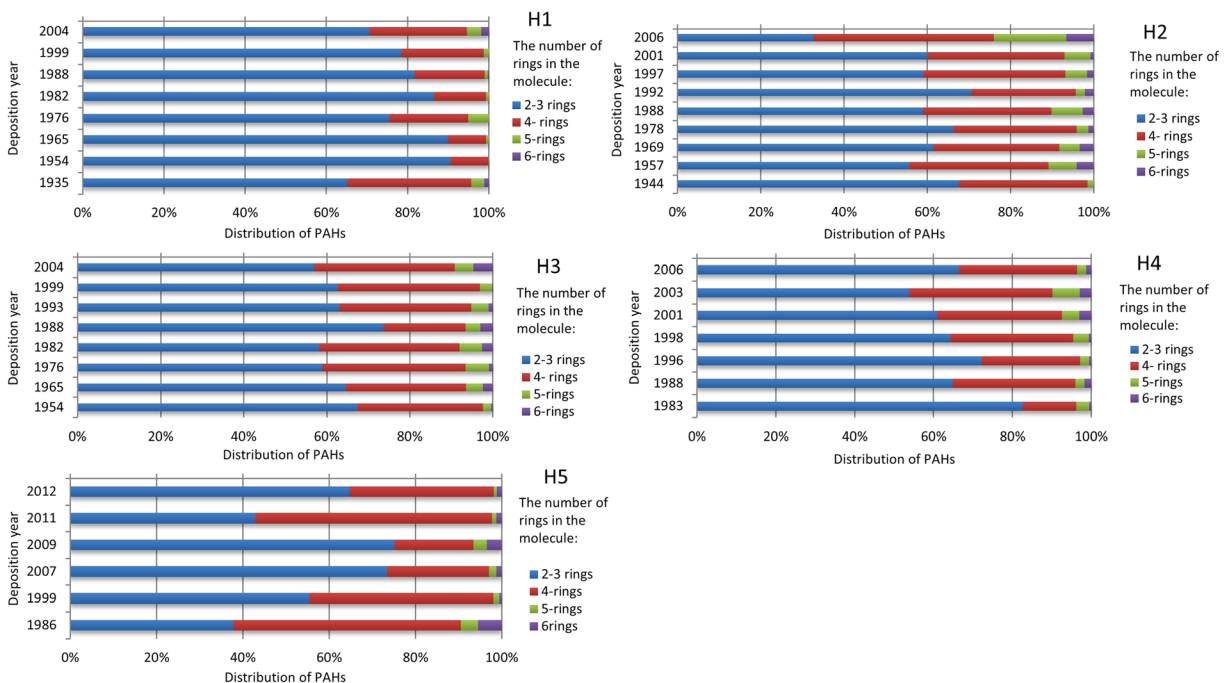


Fig. 8 Distribution of PAHs depending on the number of rings in the molecule in sediment cores collected from Hornsund

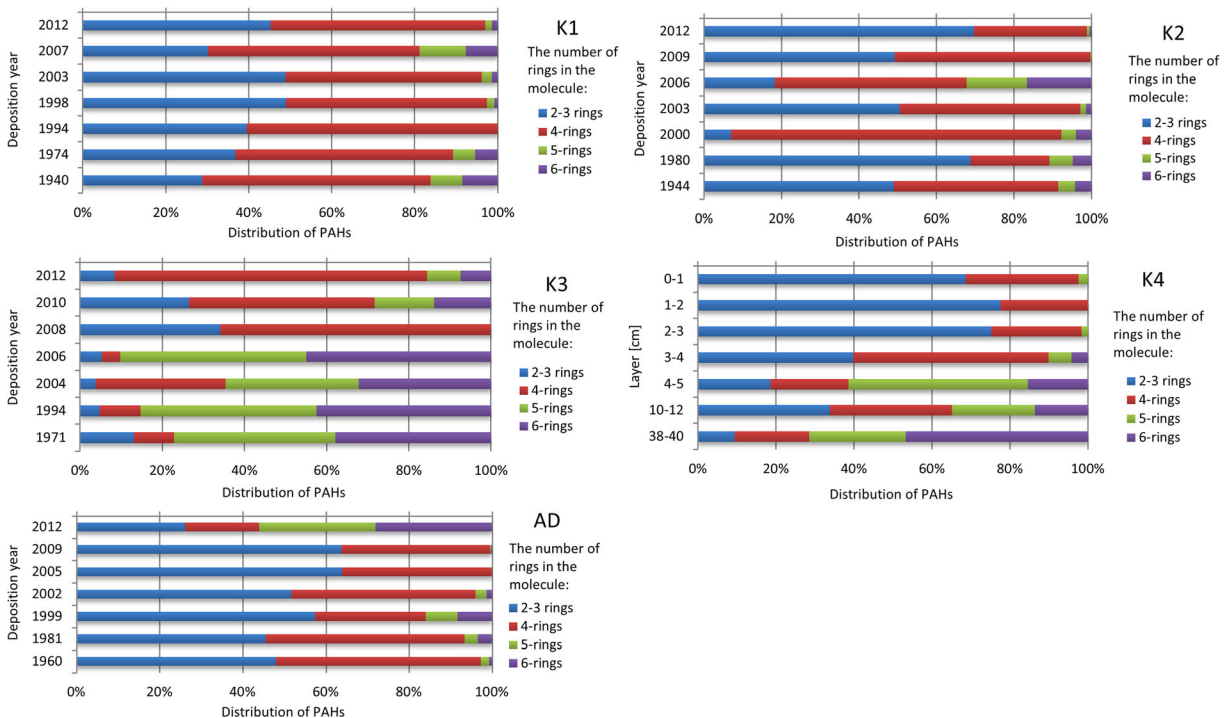


Fig. 9 Distribution of PAHs depending on the number of rings in the molecule in sediment cores collected from Kongsfjorden and Adventfjorden

90.5%, depending on the layer (Fig. 8). The 6-ring PAHs in all samples ranged from 0 to 11.8%. The dominant compound in Hornsund sediments was PHE (Tab. S2), except for layers 5–6 cm at station H3, 1–2 cm at station H1 and 4–5 and 7–8 cm at station H2, where the dominant compound was FLN. FLN is a product of combustion of organic matter and is obtained in fossil fuel products. The presence of PHE indicates petroleum, combustion and diagenetic origin (Savinov et al. 2003). In Kongsfjorden PAHs with 2–3 rings and 4 rings were dominant at the outer K1 and K2 stations (Fig. 9). At stations K1 and K2, and in surface layers at K4 station, PHE was the dominant compound (Tab. S2). PAHs with 4 rings predominated at the K3 station in surface layers (dated 2008–2012). PAHs with 5 and 6 rings prevailed in deeper layers (dated 1971–2006) at K3 station. At station K4, PAHs with 2–3 rings were dominant in surface layers (up to 3 cm). PAHs with 4 rings were dominant at 3–4 cm layers. 5-ring compounds prevailed in the 4–5 and 38–40 cm layers (Fig. 9).

At the station K4 BAP considered to be the most carcinogenic PAH, dominated in deeper layers of the sediment. BAA was predominant in the surface layer at

station K3, constituting 71.3% of $\sum 12$ PAHs. DBA was a dominant compound in deeper layers (3–4, 4–5, 10–12, 26–28 cm). These contaminants with 4–6 rings in the molecule have mutagenic and carcinogenic character and they are dangerous to organisms (Kanaly et al. 2002). In Adventfjorden sediments PAHs with 2–3 rings prevailed in surface samples (up to 5 cm) (Fig. 9). They constituted from 51.75 to 63.92% of $\sum 12$ PAHs. PAHs with 4 rings were predominant in deeper layers. In Adventfjorden sediments PHE was dominant (Tab. S2). PHE was also found to be one of the predominant PAHs in the western Barents Sea (Zaborska et al. 2011), at the Novaya Zemlia region (Yunker et al. 1996) and at Guba Pechenga (Savinov et al. 2003).

The acquirement of the ratios phenanthrene/anthracene (PHE/ANT) and fluoranthene/pyrene (FLT/PYR) allows identification of the source of hydrocarbon contamination (Budzinski et al. 1997; Raoux 1991; Mille et al. 1982). Phenanthrene is the thermodynamically most stable tri-aromatic isomer. As shown by thermodynamic calculations the PHE/ANT ratio is temperature-dependent. High temperature processes can be characterized by low PHE/ANT ratio values (4 to 10)

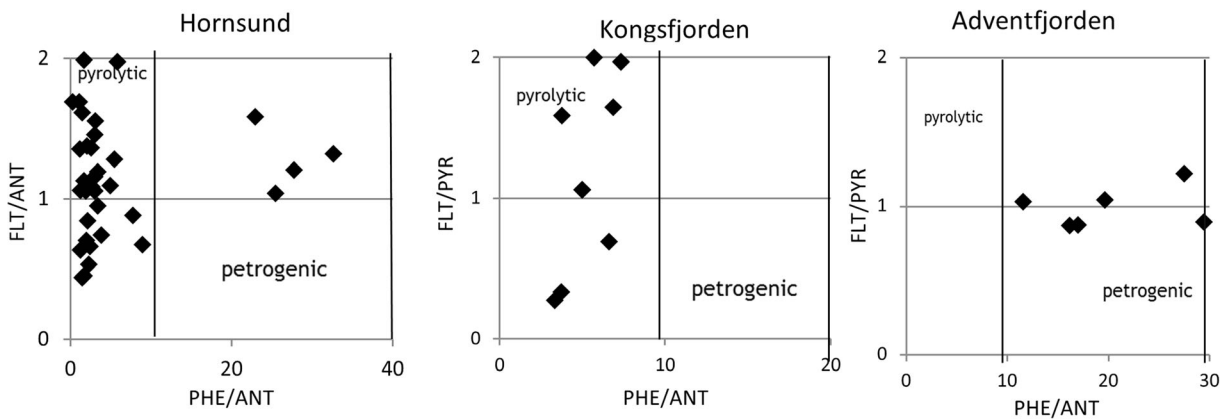


Fig. 10 FLN/PYR and PHE/ANT ratios in sediments at stations in Hornsund, Kongsfjorden, and Adventfjorden

(Colombo et al., 1989). Hence, a ratio of phenanthrene/anthracene <10 indicates that the contamination by PAHs is due to combustion processes (pyrolytic origin). Against that, slow thermal maturation of organic matter in petroleum leads to much higher PHE/ANT values (Budziński et al. 1997). The same considerations can be applied to the FLT/PYR ratio. Values greater than 1 are classically related to pyrolytic origins, namely to coal combustion (Sicre et al. 1987). When plotting FLT/PYR against PHE/ANT two zones can be defined. One is characteristic of a petrogenic origin for PAHs and the other is of a pyrolytic origin. The results are summarized in Fig. 8, which shows the relationship between isomer compounds in the studied fjord sediments. A PHE/ANT ratio lower than 10 is characteristic of pyrogenic matrices and a FLT/PYR ratio lower than 1 is indicative of a petrogenic origin (Budziński et al. 1997; Lubecki and Kowalewska 2012). These data indicate that PAHs in Kongsfjorden and Hornsund sediments seem to be of pyrolytic genesis (Fig. 10). At the station near Longyearbyen, the PHE/ANT and FLT/PYR ratios indicate that PAHs of petrogenic origin were predominant in most of the sediment layers (Fig. 10). However, other sources like coal dust from stores on land and combustion processes should also be considered.

Sediment properties are one of the most influential factors governing POP concentrations in sediments. Due to their hydrophobicity POPs tend to bind easily to sediments enriched in organic matter. Many authors suggest that concentrations of hydrophobic organic contaminants in sediments depend on organic matter content and fine sediment content (Lohse 1988; Razak et al. 1996; Hung et al. 2010; Gustafsson et al. 2001). Therefore, in order to evaluate the influence of fjord

sediment properties on the PCB and PAH concentrations, Spearman correlation relationships among the organic matter content and pelite fraction and contaminant contents were explored. There was no significant correlation between the sum of PCBs and PAHs and organic matter ($R^2 = 0.012$; $R^2 = 0.032$ respectively) or pelite fraction contents ($R^2 = 0.025$; $R^2 = 0.23$ respectively). A weak correlation was found only for one individual compound—CB153—and fine fraction content at Kongsfjorden stations. The fine fraction (<63 μm) content explained 58% of the variation of the CB153 concentration. No correlation was observed between the concentration of any other individual compound and the organic matter content/pelite fraction content. Thus, it may be suggested that in our study region contaminant concentrations do not depend so strongly on these sediment properties, but do depend on other environmental properties and/or processes, e.g., mixing, partitioning, and sorption of contaminants in the water and sediment column. This finding was also suggested by Hong et al. (2012), when investigating the occurrence of PCBs in surface sediments of the Bering Sea, Chukchi Sea, and Canada Basin. In their study, no significant relationships among the sediment properties (grain size, organic matter content, organic carbon content, black carbon content) and ΣPCB content were observed, while slightly positive relationships were obtained between penta- and hexa-PCBs and silt as well as organic matter content.

Ecotoxicological risk assessment

Organisms which have direct contact with sediments are exposed to organic contaminants. This can be

Table 3 The marine sediment quality guidelines for some PAH and $\Sigma 7$ PCB (CCME 2012)

	TEL [ng/g]	PEL [ng/g]
$\Sigma 7$ PCB	124.0	271.0
FLN	21.2	144.0
PHE	86.7	544.0
ANT	46.9	245
BAA	74.8	693.0
BAP	88.8	763.0
CHR	108.0	846.0
DBA	6.2	135.0
FLT	113	1490
PYR	153	1400

TEL threshold effect level, PEL probable effect level

dangerous, because some contaminants, e.g., PAHs, exhibit teratogenic and mutagenic effects. Sediment quality guidelines (SQGs) have been created to evaluate the ecotoxicological risk of anthropogenic contaminants in sediments and to protect aquatic biota (CCME 2012).

The concentrations of individual PAHs and $\Sigma 7$ PCBs presented in this study were compared to SQGs issued by the Canadian Council of Ministers of the Environment (CCME) (2012) and proposed by Macdonald et al. (1996). The guidelines include information about a threshold effect level (TEL) and a probable effect level (PEL) (Table 3). The TEL value represents the concentration below which a negative biological effect on benthic organisms is unlikely to occur. The PEL indicates the level of contaminant concentration above which negative biological effects are expected.

In our study, some PAH concentrations above TEL levels occurred, especially in sediment samples from Adventfjorden. The concentration of FLN at K2, H4, H5, and AD stations, and in most layers at H1, H2, and H3 stations, exceeded the TEL. Moreover, FLN concentrations in almost all Adventfjorden sediment samples were above the PEL, which indicates a high risk of adverse effects in exposed benthic organisms. Although the PAH concentrations are high in some sediment layers, some of the hydrocarbons (e.g., those originating from coal dust) are less mobile within sediments (Poot et al. 2014) and therefore less available for marine organisms (Szczybelski et al. 2016).

Concentrations of $\Sigma 7$ PCBs were below the TEL levels, thus a negative effect on sediment-dwelling organisms is not expected.

Conclusions

Sediments are important abiotic components of the ecosystem and they are habitat for benthic organisms. The data presented in this study provide a baseline record of contamination by PCBs and PAHs in Svalbard fjord sediments. In addition, a preliminary characterization of the spatial variability in POPs accumulated in present day and historic sediments (over the last 100 years) is presented. We observed differences in contaminant distribution between the inner and outer parts of the fjords. Generally, higher concentrations were measured in the inner parts, which was probably caused by limited sea water exchange and introduction of POPs from melting glaciers. In Kongsfjorden and Adventfjorden the light congener CB28 had the highest share in the majority of samples. Among the measured PAHs, in most samples, the most dominant were compounds containing 2–3 rings per molecule. Generally, the sums of PAH concentrations were similar in the investigated fjords, while PCB concentrations were lower in Hornsund. The historical profiles of PCB concentrations show an evident increase of contaminants in the surface sediments at some of the investigated stations, suggesting that although PCBs are banned in most countries there are still a lot of these compounds in the environment. On the basis of molecular diagnostic ratios used to determine the sources of PAH emissions, it was found that the origin of these compounds in Kongsfjorden and Hornsund is connected with pyrolytic processes. In Adventfjorden, PAHs of petrogenic origin were found. Important concentrations of these contaminants were noticed in layers deposited recently, suggesting intensive supply of these substances from secondary sources. The measured levels are generally low and well below known established “no effect” levels. Only the concentration of fluoranthene exceeded the threshold effect level at selected sampling stations. Moreover, fluoranthene concentrations in almost all Adventfjorden sediment layer samples were above probable effect levels, which can indicate a risk of adverse effects in exposed benthic organisms.

Acknowledgments This study was funded by the project of Ministry of Science and Higher Education (nr IPY/285/2006). Some sediment cores were sampled within the GAME (nr DET-2012/04/A/NZ8/00661) and DWARF (nr Pol-Nor/201992/93/2014) projects. The work was also partially financed from the funds of the Leading National Research Centre (KNOW), received

by the Centre for Polar Studies for the period 2014–2018, and was supported by the Institute of Oceanology of the Polish Academy of Sciences under statutory activity task no. II. 2.

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