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Occurrence of polychlorinated biphenyls (PCBs) together with sediment properties in the surface sediments of the Bering Sea, Chukchi Sea and Canada Basin

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HIGHLIGHTS

- ▶ We present PCB content in Bering Sea, Chukchi Sea and firstly open Canada Basin.
- ▶ The $\sum$PCBs were at the low end for marine sediments in the Arctic regions.
- ▶ The PCBs came mainly from the atmospheric and the ocean current transport.
- ▶ Poor relationships between sediment and PCB indicated a effect of multi-factors.

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ABSTRACT

The spatial distribution and potential source of polychlorinated biphenyls (PCBs) in surface sediments from Bering Sea, Chukchi Sea, and Canada Basin and the relationship between PCBs and sedimentary properties including grain size, water content, loss on ignition, total organic carbon, and black carbon were explored. ΣPCBs (the sum of the detected PCB congeners) concentrations fluctuated in the study area, ranging from 22–150, 60–640 and 24–600 pg g^{-1} dry weight for the Bering Sea, Chukchi Sea, and Canada Basin. A similar homologue pattern was observed at different locations, with *tri*-chlorinated PCBs being the dominant homologue, implying that the PCBs came mainly from the atmospheric transportation and deposition and ocean current transportation. No apparent co-relationships between PCB concentrations and sediment properties were obtained, indicating that the distribution of PCBs was not only controlled by their source, but also by the multi-factors such as atmospheric transport and depositing, mixing, partitioning and sorption in the water column and sediments.

1. Introduction

Polychlorinated biphenyls (PCBs) are a class of synthetic semi-volatile organic compounds with highly toxic and carcinogenic effects (AMAP, 2009; Gentleman, 2010; Hornbuckle and Robertson, 2010). Therefore, they were classified as one of the legacy persistent organic pollutants (POPs) at the Stockholm Convention in 2001 (UNEP, 2001). They are hydrophobic and bind readily to the particle fraction, and therefore, deposit to surface sediment via sedimentation, which makes estuarine and coastal marine sediments their ultimate sinks. In addition, the physical–chemical properties of PCBs such as the long half-life (60 d–27 years in water, 3–38 years in sediment), high affinity to organic carbon and black carbon (BC) (Schmidt and Noack, 2000; Hung et al., 2006; Shrestha et al., 2010), and resistance to transformation makes them a representative POP and an ideal marker for global re-distribution of POPs (Sinkkonen and Paasivirta, 2000; Ockenden et al., 2003).

POPs such as PCBs are assumed to be released in the low and temperate latitude and transported to the Arctic via atmospheric transport and deposition, ocean currents, riverine and biotic transport (AMAP, 2004; Carrizo and Gustafsson, 2011). The low ambient temperatures in the Arctic environment may decrease their degradation rates and volatilization, which might further prolong their half-life in the cold area. As a result, they tend to accumulate in

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the Arctic (Wania and Mackay, 1993). The Arctic, which was thought to be pristine, has been demonstrated to be polluted by POPs both in the biota and the abiotic compartments (AMAP, 2004, 2009; de March et al., 1998; Ma et al., 2011). Under global climate change, the Arctic, which has been a sink for global emissions of some POPs may reversely become an important source of POPs to the water and eventually the atmosphere (AMAP, 2009; Ma et al., 2011).

Marine sediments serve as a significant reservoir for PCBs in the ecosystem (Savinov et al., 2003) and have long been recognized as an available record of the atmospheric deposition (Sapota et al., 2009). Until now, studies on PCB pollution in the Arctic have mainly concerned their long range atmospheric transport and their accumulation in biota, especially in the European and Canadian Arctic (AMAP, 2004, 2009). Few studies of PCBs pollution concerning the influence of sediment properties in the Arctic marine sediments have been carried out, particularly in the western Arctic (including the Northern Bering Sea). During the 3rd Chinese Arctic Research Expedition from July to September, 2008, we collected sediments from the Bering Sea (BS), Chukchi Sea (CS) and Canada Basin (CB). PCBs and sedimentary properties such as grain size, water content, organic matter content (Loss on Ignition, LOI), total organic carbon (TOC), and black carbon (BC) content were determined. The study was carried out to: (1) determine PCB spatial distribution in the surface sediments of the BS, CS, and CB; (2) trace the potential sources of PCBs; and (3) explore the relationships between PCBs levels and the sediment characteristics.

2. Materials and methods

2.1. Sample collection

The sampling area covered the northern part of the BS, the CS, and the CB. The detailed locations of 24 surface sediments (seven in the northern BS, nine in the CS, and eight in the CB, shown in Fig. 1) were collected with a stainless steel box sampler. The 0–2 cm layer of sediments was collected using a pre-cleaned stainless steel scoop and placed into burned aluminum foil. Samples were preserved at -20 °C until chemical analysis.

2.2. Analysis of PCBs

2.2.1. Sample preparation

Details on the sample preparation are shown elsewhere (Cai et al., in press) and here, only a brief description is given. After being freeze-dried and homogenized, approximately 50 g sediment was spiked with surrogate standards (PCB30, PCB65 and PCB204) and Soxhlet extracted with a mixed solvent of acetone and hexane (1:1 v/v) for 48 h. Before extraction, activated copper flake was added to remove the sulfur from the sediments. The Soxhlet extract solutions were then exchanged into hexane after being concentrated with a rotary evaporator. A multi-layer silica-aluminum column (10 mm i.d., from bottom to top filled with neutral alumina (6 cm), neutral silica gel (2 cm), 25% sodium hydroxide silica (5 cm), neutral silica gel (2 cm) 50% sulfuric acid silica (8 cm) and de-hydro Na_2SO_4) was used for cleaning and fractionation. The mixture was then eluted with 80 mL of hexane: dichloromethane (1:1 v/v), and concentrated to 50 μL under a gentle N_2 stream. A known amount of internal standard (PCB24, PCB82 and PCB198) was added before instrumental analysis.

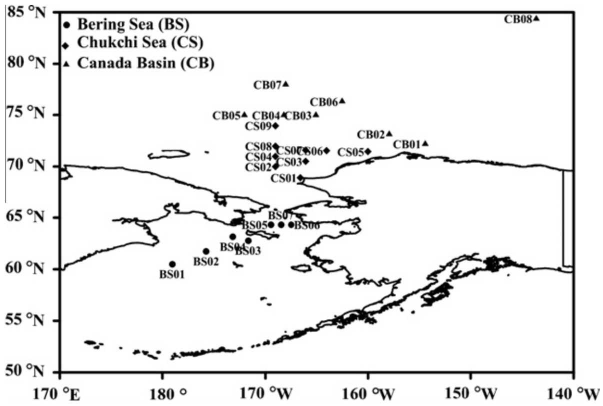

Fig. 1. Locations of sediment sampling stations in the study area.

2.2.2. Instrumental analysis

Identification and quantification of PCBs was performed on an Agilent 6890 GC-5975B Series MS system in the selective ion monitoring mode. Detailed information regarding the m/z values of the PCBs monitored for quantification and quality control are shown in the SI. Manual injection of the 1-μL samples was conducted in the splitless mode with a purge time of 1.0 min. Separation of PCB congeners was carried out with a DB-5 (60 m × 0.25 mm × 0.25 μm) capillary column. The oven temperature was programmed from 120 to 180 °C at a rate of 6 °C min^{-1}, to 240 °C at 1 °C min^{-1} and to 290 °C at 6 °C min^{-1} and finally hold on for 15 min. The temperature of both the injector and detector was 260 °C and He was used as the carrier gas.

2.2.3. Quality assurance and quality control

For each of eight field samples, a procedural blank (solvent with a filter paper), a spiked blank (targeted PCB congeners spiked into the solvent with the filter paper), a spiked matrix sample (targeted PCB congeners spiked into pre-extracted sediment), and a spiked matrix duplicate were processed. Only PCB170 and PCB209 were detected in the procedural blank and were subtracted in the reported concentrations. The limit of detection (LOD) defined as three times the ration of signal to noise for analytes which were not detected in the blanks and three times of the standard deviation of the values in the blanks. The LODs for the analytes were ranged from 0.054 to 0.56 pg g^{-1} dry weight (d.w.) based on the sample size of 50 g. The recoveries of the surrogates PCB30, PCB65, and PCB204 were 77–104%, 90–118%, and 87–103%, respectively, and recoveries of the targeted PCB congeners were 88.4–116.0% in three spiked blank duplicates, while they ranged from 83.6% to 119.1% in three spiked matrix duplicates. Three sample duplicates were analyzed and their relative standard deviations were <5%.

2.3. Determination of sediment properties

Details of the determination of the environmental parameters (including grain size, TOC and BC) are described elsewhere (Cai et al., in press) and only a brief description is shown here. After removal of inorganic carbon by HCl, the sample was dried for the determination of TOC and the inorganic carbon free subsamples were further oxidized for BC at 375 °C. Both the TOC and BC were measured with Vario EL III Elementar (Germany). Grain size was determined directly from the samples by Mastersizer 2000 (Malvern Co. UK). The final parameter loss on ignition (LOI) was measured by heating at 450 °C for 4 h after the removal of water.

Table 1
Properties of surface sediments in the Bering Sea, Chukchi Sea and Canada Basin.

Station	Clay (%)	Silt (%)	Gravel (%)	Sand (%)	TOC (%)	BC (%)	Water content (%)	Ignition loss (%)
BS01	1.75	14.84	9.78	73.62	0.40	0.07	26.47	2.11
BS02	10.07	81.00	2.29	6.64	1.80	0.24	59.40	6.71
BS03	8.28	79.15	12.40	0.14	0.25	0.04	50.73	5.72
BS04	1.16	10.10	83.40	5.37	0.20	0.05	27.64	1.66
BS05	2.25	12.20	78.60	6.93	1.30	0.13	21.34	1.17
BS06	2.96	22.32	25.80	48.88	0.21	0.03	29.18	2.01
BS07	2.32	16.64	70.10	10.93	0.21	0.05	23.49	1.65
CS01	10.62	60.87	19.50	9.01	1.39	0.17	33.60	6.10
CS02	13.77	58.25	11.50	16.49	1.63	0.17	21.57	4.10
CS03	8.37	66.24	25.40	<0.01	0.86	0.16	42.34	4.48
CS04	15.51	71.87	11.20	1.39	1.19	0.14	42.60	5.41
CS05	5.18	45.77	48.10	0.91	0.52	0.07	38.31	2.84
CS06	15.69	79.58	4.49	0.24	1.91	0.20	51.20	7.52
CS07	8.37	84.95	6.69	<0.01	1.17	0.15	63.68	7.99
CS08	7.71	81.21	9.82	1.25	1.59	0.40	52.35	6.26
CS09	30.91	65.76	3.32	<0.01	1.13	0.19	58.83	6.62
CB01	18.55	80.53	0.92	<0.01	1.51	0.15	55.35	6.08
CB02	29.66	70.05	0.29	<0.01	1.17	0.15	60.23	7.08
CB03	36.73	63.15	0.12	<0.01	0.64	0.19	53.03	6.00
CB04	23.46	64.20	6.85	5.50	1.02	0.16	31.40	5.07
CB05	27.50	71.59	0.91	<0.01	0.94	0.18	59.81	7.08
CB06	36.09	61.08	2.40	0.42	0.78	0.18	66.99	8.51
CB07	34.98	63.44	1.53	0.05	0.17	0.05	36.30	7.67
CB08	17.66	46.60	18.90	16.86	0.23	0.02	29.51	5.38

3. Results and discussion

3.1. Sediment properties

Table 1 displays the sediment properties in the study area. The components were divided into four parts based on grain size $\varphi = -\log_2\Phi$ (Φ is the diameter, mm): clay ($\varphi > 8$), silt ($4 < \varphi < 8$), gravel ($2 < \varphi < 4$), and sand ($\varphi < 2$). The contents lower than the limit of determination (LOD = 0.01%) were replaced with < 0.01%. Sediments in BS consisted mainly of gravel and sand (except for Station BS02 and BS03) while those in the CS and CB were mainly fine grained clay and silt. In general, the contributions of the fine grained component (including the sum of clay and silt) from most stations in the BS (averaged in 17.3%, not including BS02 and BS03) were lower than that in the CS (averaged in 81.2%) and CB (averaged in 93.2%).

The TOC and BC contents were in the range 0.17–1.91% and 0.02–0.40%, respectively. The BC content was generally less than 0.20% in all stations except CS08 (0.40%) and BS02 (0.24%). Similar to the grain size distribution, both of the TOC and BC decreased in the order: CS > CB > BS. A measurement of LOI was also determined in the range 1.17–8.51%. LOI in the CB were slightly greater that than those in the CS, both of which were higher than those in the BS. There was a strong positive correlation between LOI and the sum of clay and silt, corresponding to the relationship between organic matter and grain size distribution in the sediments (Thomas, 1969; Bergamaschi et al., 1997). A similar positive relationship was also observed between TOC and the sum of clay and silt although their correlation was not statistically significant.

3.2. Geographic distribution of PCBs

Only 14 congeners (PCB 18, 20, 22, 28, 95, 99, 101, 110, 118, 138, 149, 153,170, and 209) were detected in the study areas. Range, mean and median concentrations of individual congeners and $\sum$PCBs (sum of the 14 detected PCB congeners) are summarized in Table 2. Concentrations of $\sum$PCBs were in the range 22–150, 60–640 and 24–600 pg g^{-1} dry weight (d.w.), together with averages of 71 ($n = 7$), 190 ($n = 9$) and 150 ($n = 8$) pg g^{-1} d.w. in the BS, CS, and CB, respectively. Extremely high concentrations were observed in stations CS01 (640 pg g^{-1}) near the Alaskan coast and CB04 (600 pg g^{-1}) farther from the land, and a relatively higher concentration of $\sum$PCBs could be seen in CB01 (280 pg g^{-1}) near the Barrow Point, Alaska. Relatively lower concentrations of $\sum$PCBs were located in the far CB station CB08 (24 pg g^{-1}) and BS station BS07 (22 pg g^{-1}) in the Bering Strait. The lower concentration in CB08 might have been due to the distance from the land and a low sedimentation rate while that in BS07 could have resulted from the higher proportion of coarse sediment components with the low adsorption capacity. Although the $\sum$PCBs fluctuated in the three areas, however, without consideration of the stations with extremely high concentrations (relative to the uniform concentration ranging from 22 to 280 pg/g of the whole study area), the average concentration decreased in the order: CS ~ CB > BS. In general, the concentrations of $\sum$PCBs here were at the low end of the range for marine sediments found in previous AMAP POPs assessments (de March et al., 1998; AMAP, 2004).

Various studies have been conducted in the remote Arctic coastal sediments. In comparison, the concentrations in our study areas were one or two magnitudes lower than that in the marine sediments of the mid- and western Russian Arctic (Sericano et al., 2001; Savinov et al., 2003), Norwegian Arctic (Evenset et al., 2007; Jiao et al., 2009; Sapota et al., 2009), Saglek Bay, Labrador, Canada (Kuzyk et al., 2005) and Gulf of Alaska (Iwata et al., 1994a), where mineral exploration, military activities and harbors were more active (Bright et al., 1995; de March et al., 1998; Michelutti et al., 2009). When compared with a study in the BS and CS (Iwata et al., 1994a), $\sum$PCB concentrations showed either no change or only a slight decrease. Compared with 1993 samples (Strachan et al., 2001), these concentrations were about two magnitudes lower in our study. This seemed to illustrate the lag in the use history of PCBs, where usage increased before the bans in many countries in the 1970–1990s and then decreased progressively thereafter (de March et al., 1998). However, due to the lack of TOC and BC, and the difference in the grain size in other studies shown here, the influences of sedimentary properties on the PCB content in our study can not be compared with other reports.

3.3. Homologue pattern of PCBs and their possible source

Although PCBs are now ubiquitous in the environment, they result from industrial production without any known natural source.

Table 2
Summary of PCB congeners concentrations (pg g^{-1} d.w.) in surface sediments of the Bering Sea, Chukchi Sea and Canada Basin.

	BS			CS			CB		
	Range	Mean	Median	Range	Mean	Median	Range	Mean	Median
PCB18	1.1–7.0	3.2	2.4	ND-6.0	2.9	3.3	ND-5.0	1.8	0.79
PCB20	5.1–40	21	20	13–530	120	83	5.5–540	110	42
PCB22	1.5–11	5.3	4.1	1.7–10	5.1	4.7	ND-20	6.0	6.0
PCB28	3.8–23	11	8.2	11–31	19	19	4.6–21	12	8.9
PCB95	0.86–11	3.a9	1.7	3.8–11	6.7	7.3	ND-10	2.8	0.79
PCB99	ND-4.4	1.7	1.1	ND-5.3	2.5	2.2	3.3–101	1.3	1.2
PCB101	1.1–14	4.5	1.8	3.7–13	7.8	6.8	ND-11	2.9	2.2
PCB110	ND-7.5	2.8	1.6	2.9–7.2	5.5	6.0	ND-4.7	1.7	0.78
PCB118	ND-9.9	3.2	1.3	ND-8.4	4.7	4.6	ND-9.5	1.9	ND
PCB138	ND-6.5	2.9	3.7	ND-6.5	3.4	3.6	ND-12	3.3	2.3
PCB149	ND-9.3	3.2	1.9	ND-8.5	4.7	4.9	ND-7.0	1.6	ND
PCB153	ND-9.0	3.1	2.0	ND-9.0	5.3	5.0	ND-8.0	2.6	0.50
PCB170	0.90–5.9	2.8	2.2	1.5–11	4.7	4.8	0.5–5.9	2.4	2.1
PCB209	0.67–4.3	2.2	2.0	ND-15	3.4	2.5	1.0–7.1	2.7	1.9
∑PCBs	22–150	71	47	60–640	190	170	24–600	150	76

ND: not detected

Table 3
Pearson relationship between sediment properties and PCBs.

	Tri-PCBs	*Penta*-PCBs	*Hexa*-PCBs	*Hepta*-PCBs	*Deca*-PCBs	∑PCBs	Clay	Silt	Gravel	Sand	TOC	BC	Water Content	LOI
Tri-PCBs	1													
Penta-PCBs	0.279	1												
Hexa-PCBs	0.298	0.895**	1											
Hepta-PCBs	0.358	0.523**	0.363	1										
Deca-PCBs	0.845**	0.408*	0.393	0.271	1									
∑PCBs	0.990**	0.410*	0.421*	0.418*	0.864**	1								
Clay	0.057	−0.205	−0.195	−0.200	−0.047	0.021	1							
Silt	0.258	0.666**	0.626**	0.340	0.197	0.340	0.474*	1						
Gravel	−0.198	−0.342	−0.326	−0.174	−0.118	−0.236	−0.638**	−0.777**	1					
Sand	−0.112	−0.293	−0.267	−0.086	−0.071	−0.147	−0.415*	−0.588**	0.067	1				
TOC	0.322	0.670**	0.585**	0.546**	0.215	0.402	0.064	0.559**	−0.347	−0.319	1			
BC	0.171	0.539**	0.414*	0.128	0.094	0.232	0.214	0.576**	−0.406*	−0.359	0.779**	1		
Water content	−0.090	0.428*	0.478*	−0.091	−0.043	−0.022	0.528**	0.759**	−0.631**	−0.500*	0.358	0.510*	1	
LOI	0.168	0.387	0.359	0.082	0.172	0.213	0.709**	0.869**	−0.804**	−0.526**	0.399	0.472*	0.812**	1

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

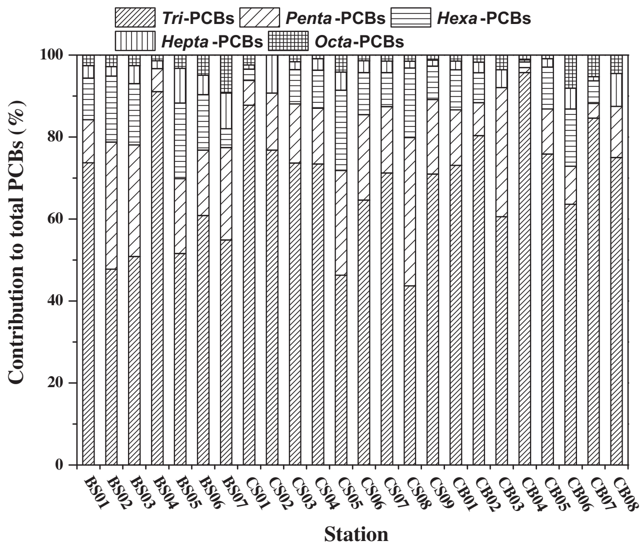

Fig. 2. Percentage contributions of different chlorinated congeners to the ∑PCBs.

Atmospheric transport and deposition, current transport, riverine input, sea-ice transport, and biotic transport are considered to be the main sources in the areas studied (de March et al., 1998; Macdonald et al., 2000; AMAP, 2004). In order to probe further the sources of PCBs in our area, the PCB homologue pattern (i.e. the proportion of contributions by different chlorinated congener groups to the ∑PCBs) is presented in Fig. 2. Overall, it was apparent that there were few if any difference between the three areas and that lower chlorinated congeners were dominant in the study areas, with *tri*-chlorinated congeners (mainly PCB20 and 28) contributing 44–96% (mean 69%) of the ∑PCBs followed by *penta*-chlorinated congeners (1.3–36%, mean 16%) and *hexa*-chlorinated congeners (0–20%, mean 8.7%). This is identical to the PCB patterns found in mixed layer in this area, where *tri*-chlorinated congeners make up a dominant fraction of the ∑PCBs (Carrizo and Gustafsson, 2011). In general, the similarities between the three areas indicated that there was a common source for all regions – atmospheric transport and deposition being the most obvious suspect with some contribution from ocean current transport (Carrizo and Gustafsson, 2011).

The enrichment of lower chlorinated congeners in our study corresponded with many reports that reveal the enrichment of low molecular weight PCB congeners in marine sediments and waters from high latitudes (Gustafsson et al., 2001; Sericano et al., 2001; Strachan et al., 2001; Savinov et al., 2003; Ishaq et al., 2009). This enrichment seems to be explained by their more efficient atmospheric transport resulting from their relatively higher vapor pressure from sources in the industrialized northern temperate latitudes between 30–60°N, where around 86% of global ∑PCBs consumption occurs (Wania and Mackay, 1993; AMAP, 2004), and the higher fluxes of air-seawater exchange in the lower ambient temperature area (Iwata et al., 1993; Iwata et al., 1994a; Iwata et al., 1994b).

3.4. Relationships between PCB distribution and sediment properties

Due to their high lipophilicity, POPs tend to bind easily to the sediments, especially organic matter enriched sediments. Therefore, sediment properties were thought to be one of the most influential factors for POPs concentrations in sediments. Studies show that grain size, LOI, TOC, and BC always play an important role in controlling POPs levels in sediments (Thomas, 1969; Persson et al., 2005; Hung et al., 2006; Koelmans et al., 2006; Nam et al., 2008; Chen et al., 2009; Hung et al., 2010). Kobayashi et al. (2001) point out that sediments consisting of fine sand have lower TOC and that the ∑PCBs are one or two orders of magnitude lower than at other stations in the Tokyo Bay. Therefore, in order to primarily evaluate the influence of sediment properties on the PCB contents in our study area, Pearson correlation relationships among the sediment properties and PCB content were explored using SPSS@16.0 for Windows (Table 3).

No significant relationships among the sediment properties and content of ∑PCBs were obtained, which was probably due to the low co-relationship with the dominant homologue – the *tri*-chlorinated congeners. This differs from the suggestion that within a given region in the Arctic surface sediments, the distribution of organic pollutants are linked with the TOC content (Gustafsson et al., 2001). On the contrary, in our studies, slightly positive relationships were obtained between the *penta*- and *hexa*-PCBs and silt, TOC, BC, water content, and LOI. This might be due to different solubility of congeners, which leads the higher chlorinated congeners to bind to organic carbon and fine grains. In general, the low co-relationships between PCBs and sediment properties may not only have been due to the adsorption capacity of different PCB congeners but also associate with atmospheric transport and deposition and other processes including mixing, partitioning and sorption of PCBs in the water and sediment column.

4. Conclusion

We evaluated the spatial distributions, potential source of PCBs and the potential influence of sediment properties in the surface sediments of the Northern BS, CS, and CB. In these areas, ∑PCBs concentrations were low relative to previous studies in Arctic regions and they did not show any apparent spatial distribution, except that the highest concentrations were occurred along the Alaska coast. In terms of the distribution of different chlorinated PCBs, *tri*-chlorinated PCBs (mainly PCB 20 and 28) were dominant. Furthermore, concentrations of ∑PCBs did not tend to be related to any sediment properties, implying a combined effect of the source and the processes from the source to the sediment sink. Therefore, in future, more studies of the processes which take place from the PCB source to the sediment sink should be carried out in order to have a clearer understanding of the fate of PCBs in the Arctic.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.chemosphere.2012.05.033.

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