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Distribution of polybrominated diphenyl ethers and decabromodiphenylethane in surface sediments from Fuhe River and Baiyangdian Lake, North China

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Abstract

Nineteen surface sediment samples collected from Baiyangdian Lake and its inflowing river (Fuhe River) in North China were analyzed for polybrominated diphenyl ethers (PBDEs) and decabromodiphenylethane (DBDPE). The concentrations of PBDEs and DBDPE in sediments ranged from 5.5 to 300.7 ng/g dry weight (dw) and 1.1 to 68.2 ng/g dw, respectively. Their levels in sediments in Fuhe River were significantly higher than those in Baiyandian Lake. Compared to data from other regions, the PBDE levels in surface sediments from Baiyangdian Lake and Fuhe River were in the medium to lower range. Among the PBDE congeners, BDE209 was predominant, with contributions to the total PBDEs ranging from 79.4% to 97.3% in sediment samples. For the lowly brominated congeners (tri- to hepta-BDE), BDE47 and BDE99 were the most abundant, which contributed 52.1% and 44.1% to the sum of tri- to hepta-BDEs in the sediments from Baiyangdian Lake and Fuhe River, respectively. The compositional patterns of PBDEs in Baiyangdian Lake sediments indicated that technical deca-BDE mixture was the major pollutant sources with a minor contribution of penta-BDE mixture. The present study suggested that the importance of Fuhe River as a possibly potential sources of PBDEs contamination in Baiyangdian Lake.

Key words: polybrominated diphenyl ethers; decabromodiphenylethane; contamination; profile; Baiyangdian Lake; Fuhe River
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Introduction

Polybrominated diphenyl ethers (PBDEs) are flame retardants that are widely used in plastics, paints, electronics, textiles, foam, and rubber. As a result, they present in many commercial and household products, such as computers, televisions, furniture, carpets and toys (Hale et al., 2002, Chen et al., 2009). Three commercial PBDE mixtures have been produced: deca-BDE, octa-BDE, and penta-BDE (BSEF, 2003). The penta- and octa-products contain several BDE congeners, and the deca-product is composed of almost entire BDE209 (Hites, 2004). The worldwide demand for deca-BDE in 2001 was estimated to be almost 56,100 tons, corresponding to about 80% of the total demand for PBDEs worldwide (Hale et al., 2006). The domestic demand of BFRs has increased at a rate of 8% annually in China (Mai et al., 2005). In 2006, the productions of the technical deca-BDE mixture and decabromodiphenylethane (DBDPE) in China were 15,000 and 11,000 tons, respectively (http://coatren.cn/news/stuff/assist/20061215_10462.shtml).

In views of their bioaccumulation potential, environmental persistence, and potential human and wildlife toxicities, PBDEs are included amongst six environmentally hazardous substances in the European Union (EU) issued Directive 2002/95/EC regarding the restricted use of certain hazardous substances in electrical and electronic equipment (RoHS). The technical penta-BDE and octa-BDE products were banned in 2004 by the EU as well as by some states of the United States (Renner, 2004). Technical deca-BDE has received more interest, since it remains the only diphenyl-ether-based brominated flame retardant available. Widely usage, high persistence, lipid solubility, and bioaccumulation potential of some PBDE have led to the increasing contamination in the environment, wildlife, and humans (Renner, 2004; Qu et al., 2007; Luo et al., 2009; Zhang et al., 2009). Toxicological studies in animal models suggest that PBDEs can cause adverse physiological effects, including endocrine and reproductive disruption, interference with neurobehavioral development (Viberg et al., 2006; Darnerud, 2008).

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DBDPE was introduced as an alternative to deca-BDE in the early 1990s (Kierkegaard et al., 2004). With increasing regulation and phasing-out of production of the commercial usage of PBDEs, it is expected that the production and usage of DBDPE will be predominant in the coming future (Ricklund et al., 2008). Toxicological studies showed that oral toxicity of DBDPE in rats was low, due to poor bioavailability, large molecular weight and logP_{ow} (Hardy et al., 2002). It has been reported the presence of DBDPE in the environment. DBDPE has been detected in air near the Great Lakes (Venier and Hites, 2008), sediments from sewage treatment plant (Kierkegaard et al., 2004; Ricklund et al., 2008), and house dust from USA (Stapleton et al., 2008). DBDPE has also been found in benthic fish (Law et al., 2006), waterbird (Luo et al., 2009), terrestrial mammal (Hu et al., 2008) and tree bark from North American (Zhu and Hites, 2006). However, the potential sources and environmental behaviors of DBDPE were not clear.

It is generally accepted that sediments constitute a sink for hydrophobic compounds in aquatic environment. The contamination of sediments may pose an unacceptable risk to aquatic organisms, wildlife and humans (Eljarrat et al., 2004). The study of sediments is an important step in mapping possible pollution sources and exposure pathways that makes pollutants bioavailable to sediment-dwelling organisms. PBDEs and DBDPE, sharing similar structures, will very quickly bind to organic rich suspended particles in the water column and are transported to the bottom sediments. Sedimentation will play important roles in the process of contaminants transport.

Baiyangdian Lake is the largest natural freshwater body in the North China Plain. The lake consists of more than 100 small and shallow lakes linked to each other by thousands ditches with surface area of 366 km^2 and a catchment of 31,200 m^2. Fuhe River is a major inflowing river of Baiyangdian Lake with 63 km length, flowing through Baoding City. Baiyangdian Lake receives nutrient inputs from Fuhe River. Both the Baiyangdian Lake and Fuhe River are strongly influenced by human activities. In addition, Baiyangdian Lake is likely to play an important role in the South-to-North Water Division Project under construction because of its geographic location. Previous studies showed persistent organic pollutants (POPs) such as organochlorine pesticide (OCPs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbon (PAHs) were detected in the sediments (Dou and Zhao, 1998; Hu et al., 2010). However, there have been few studies of the distribution and deposition of PBDEs and DBDPE in the sediments from Baiyangdian Lake and Fuhe River.

In the present study, Baiyangdian Lake, a heavily industrialized plain area with extensively anthropogenic activities, was selected for the first time to study the environmental PBDEs and DBDPE contamination. The study detected PBDEs and DBDPE levels and assessed their spatial distribution in the surface sediments from Baiyangdian Lake and Fuhe River. In addition, the study primarily discussed the emission sources of PBDEs and DBDPE in the regions of Baiyangdian Lake.

1 Material and methods

1.1 Sample collection and material

Nineteen surface samples were collected, of which seven from Baiyangdian Lake and twelve from Fuhe River, in August 2007 and March 2008, using a stainless steel grab sampler (Fig. 1). The top 5-cm layer of sediments was scooped into solvent-rinsed aluminum containers using a precleaned stainless steel scoop. All samples were transported on ice to the laboratory, and were stored at –20°Cuntil further analysis.

PBDEs standard mixtures, containing BDE28, 47, 66, 85, 99, 100, 138, 153, 154, 183, 197, 203, 196, 205, 206, 207, 208, and 209, were obtained from AccuStandard (USA). ^{13}C-PCB141 and ^{13}C-PCB208 were obtained from Cambridge Isotope Laboratories Inc. (USA). DBDPE and 2,2',4,4',5-pentachlorodiphenyl ether (CDE99) were obtained from Wellington Laboratories (Canada). All solvents and reagents used in the extraction and cleanup procedures were AR grade and all organic solvents were re-distilled using glass system. Neutral silica gel (80–100 mesh) was extracted with a mixture of dichloromethane and methanol (1:1, *V/V*) for 72 hr using a Soxhlet extractor. Upon drying under room temperature, silica gel was baked at 180°C for 12 hr. Sodium sulfate was baked at 450°C and stored in sealed containers.

1.2 Sample extraction and cleanup

Sediment samples were freeze-dried, ground, and homogenized by sieving through a stainless steel 80 mesh (0.2 mm) sieve and stored in glass containers at –20°C until extraction. The extraction procedure was similar to that described for the determination of PBDEs in sediment

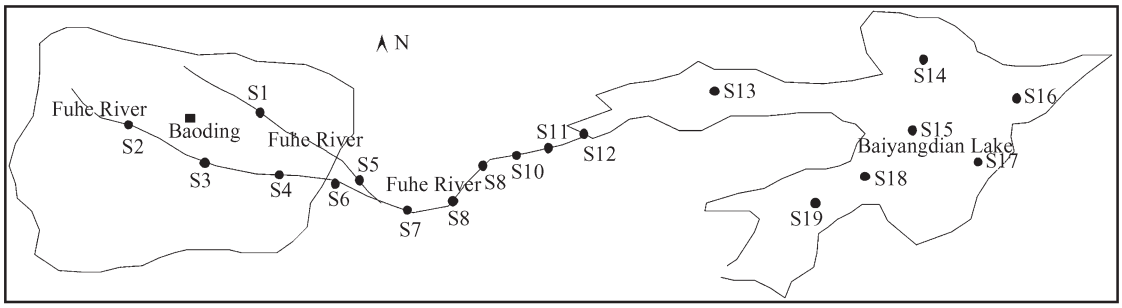

Fig. 1 Map of study area and sampling sites. S1–S12 from Fuhe River, S13-S19 from Baiyangdian Lake.

(Mai et al., 2005). After spike with ^{13}C-PCB141 and CDE99, samples were extracted with a mixture of acetone and hexane (1:1, *V/V*) for 48 hr with a Soxhlet extractor. Activated copper granules were added to the extraction flasks during the extraction to remove elemental sulfur. Concentrated extracts were cleaned on a 10 mm i.d. silica column packed with neutral silica (8 cm, 3% deactivated), 44% sulfuric acid silica (8 cm), and anhydrous sodium sulfate (1 cm). The column was eluted with 30 mL hexane:dichloromethane (1:1, *V/V*). BDE118 and BDE128 were added as internal standard for analysis.

1.3 Instrumental analysis

Sample analysis was performed with a Shimadzu Model 2010 gas chromatograph (GC) coupled with a Model QP2010 mass spectrometer (MS) (Shimadzu, Japan) using negative chemical ionization (NCI) in the selected ion monitoring (SIM) mode. A DB-XLB capillary column (30 m × 0.25 mm × 0.25 μm) was used for the separation of tri- to hepta-BDE congeners (BDE28, 47, 66, 85, 99, 100, 138, 153, 154, and 183). For octa- to deca-BDE congeners (BDE196, 197, 203, 205, 206, 207, 208, and 209) and DBDPE, a DE-5HT capillary column (15 m × 0.25 mm × 0.10 μm) was used. Details of the GC temperature program and monitored ions were given elsewhere (Mai et al., 2005; Hu et al., 2008). Splitless injection of 1 μL of sample was conducted with an autosampler. Methane was used as a chemical ionization moderating gas at an ion source pressure of 2.4×10^{-3} Pa and helium as the carrier gas at a flow rate of 1 mL/min. The ion source and interface temperatures were set to 200 and 280°C, respectively. Quantification was performed using an internal calibration method (five to nine concentration levels).

1.4 Quality control/quality assurance (QC/QA)

Instrumental quality control (QC) included regular injection of solvent blanks and standard solutions. The relative percent difference were less than 20% for all targets analyses. For methods quality assurance (QA), three procedural blanks, triplicate spiked blanks, and triplicate spiked matrices were analyzed. Only trace levels of BDE47 and BDE99 were detected in blanks and the mean concentrations were subtracted from those in samples. The recoveries of 11 PBDE congeners (BDE28, 47, 66, 85, 100, 99, 154, 153, 138, 183, and 209) ranged from 65.7% to 82.0% and 67.4% to 79.7% in spiked blanks and matrix spiked samples, respectively. The mean recoveries of DBDPE in spiked blanks and matrixes were 87.6% and 103.6%, respectively. The surrogate standard recoveries in 19 samples were 83.2%–103.8% for ^{13}C-PCB141, 86.1%–109.0% for CDE99, and 54.6%–117.4% for ^{13}C-BDE209. The limit of detection (LOD), defined as a signal/noise ratio (S/N) of 3, ranged from 0.1 to 2.2 pg/g dry weight (dw) for PBDEs and 2.9 to 4.4 pg/g dw for DBDPE, respectively.

1.5 Data analysis

For samples with concentration below LOD, zero was used for the calculations. All levels are presented as a dry weight basis. BDE66, 85, 138, and 196 were not detected in all the sediment samples. ∑PBDEs is defined as the sum of the 14 most frequently detected congeners (BDE28, 47, 99, 100, 153, 154, 183, 197, 203, 205, 206, 207, 208, and 209). The differences of contaminants in sediments between Baiyangdian Lake and Fuhe River were analyzed using Two-Sample Kolmogorov-Smirnov *T*-Test. Principal component analysis (PCA) was conducted to assess the compositions of PBDE congeners. PCA with varimax rotation was performed using SPSS 11.5 software (SPSS, USA). The level of significance was set at $\alpha = 0.05$ throughout this study.

2 Results and discussion

2.1 Concentration

Concentrations of PBDEs and DBDPE in the surface sediment samples are given in Table 1. ∑PBDE1 is defined as the sum of tri- to hepta-BDEs, including BDE28, 47, 99, 100, 153, 154, and 183. The concentrations of ∑PBDE1, octa-BDE, nona-BDE, and deca-BDE in sediments of Fuhe River were in the range of 0.13–6.39, 0.27–2.92, 5.07–34.9, and 11.8–292.7 ng/g dw, respectively. The concentrations of ∑PBDE1, octa-BDE, nona-BDE, and deca-BDE in sediments of Baiyangdian Lake were in the range of 0.05–5.03, 0.44–0.75, 2.10–4.19, and 4.35–19.3 ng/g dw, respectively. The levels of PBDEs in Fuhe River were significantly higher than those in Baiyangdian Lake ($P < 0.05$). PBDEs and DBDPE in Fuhe River and Baiyangdian Lake were likely originated from a combination of point and non-point sources and atmospheric deposition. Large amounts of industrial activities (such as dismantling electronic products, cables and wires, refinery oils, and chemical plants) were likely responsible for the high concentrations of PBDEs and DBDPE in sediments in Fuhe River. The primitive electronic wastes recycling activities were observed along the Fuhe River. Among

Table 1 PBDEs and DBDPE concentrations in sediments from Fuhe River and Baiyangdian Lake in 2008

	Concentration (ng/g dw)	
	Fuhe River	Baiyangdian Lake
BDE28	0.00–0.34 (0.09)[a]	0.00–0.07 (0.01)
BDE47	0.03–1.58 (0.58)	0.01–0.78 (0.13)
BDE99	0.02–1.92 (0.60)	0.01–2.68 (0.39)
BDE100	0.01–0.23 (0.07)	0.002–0.58 (0.09)
BDE153	0.02–1.03 (0.27)	0.01–0.47 (0.08)
BDE154	0.01–0.94 (0.18)	0.002–0.41 (0.06)
BDE183	0.04–1.82 (0.54)	0.01–0.04 (0.02)
Sum tri- to hepta-BDE	0.13–6.39 (2.33)	0.05–5.03 (0.78)
BDE197	0.07–1.57 (0.31)	0.17–0.23 (0.19)
BDE203	0.11–1.13 (0.34)	0.14–0.19 (0.16)
BDE205	0.10–0.26 (0.20)	0.14–0.38 (0.22)
Sum octa-BDE	0.27–2.92 (0.86)	0.44–0.75 (0.57)
BDE206	1.43–14.6 (4.51)	0.91–1.95 (1.34)
BDE207	2.69–17.1 (5.57)	0.85–1.75 (1.24)
BDE208	0.82–5.07 (2.17)	0.30–0.48 (0.37)
Sum nona-BDE	5.07–34.9 (12.3)	2.10–4.19 (2.95)
deca-BDE	11.8–292.7 (102.6)	4.35–19.3 (10.4)
DBDPE	16.1–68.2 (31.3)	1.10–5.29 (2.78)

[a] Mean value is presented in parenthesis.

the sediment samples from Baiyangdian Lake, the highest concentration of ∑PBDE1 (5.03 ng/g) was observed at S15 (Wangjiazhai), which possibly related to the large amount of anthropogenic activities (Fig. 2). This was not surprising, because Wangjiazhai was surrounded by the smaller lakes with tourism blooming.

Among PBDEs congeners, BDE209 was the most predominant. The average concentrations of BDE209 were 1–2 orders magnitude higher than those of ∑PBDE1 in sediments from Fuhe River and Baiyangdian Lake. The findings were consistent with previous reported results, which indicated that BDE209 was the dominant component of PBDEs detected in the sediments from Pearl River Delta, South China (Mai et al., 2005; Zhang et al., 2009).

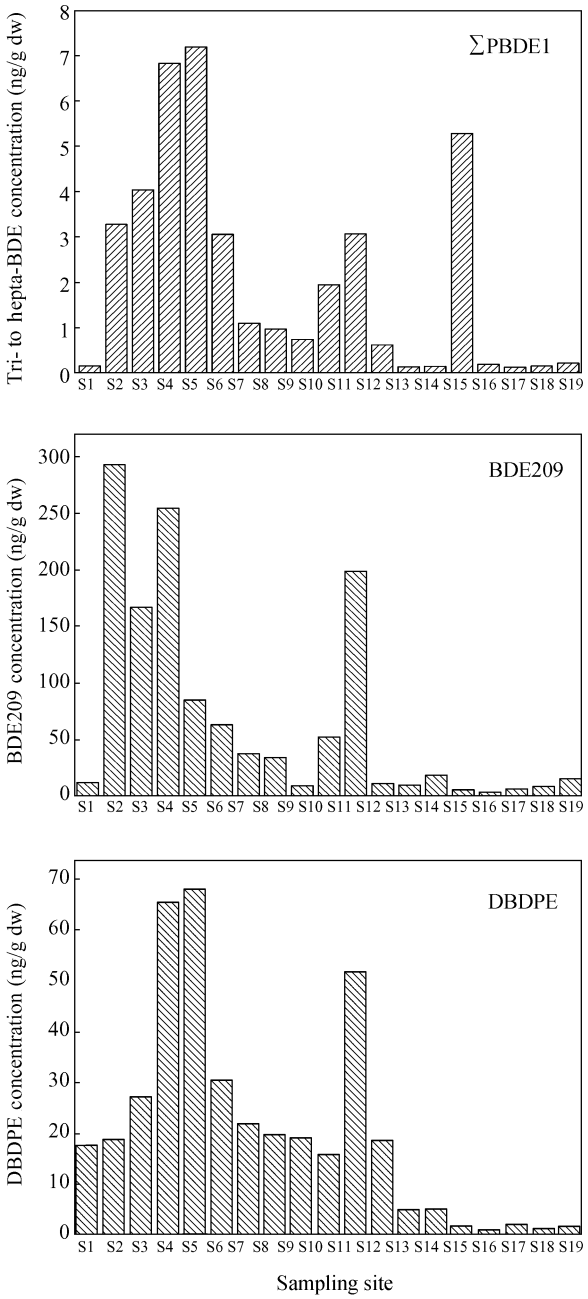

Fig. 2 Distribution of ∑PBDE1, BDE209, and DBDPE in surface sediments from Fuhe River and Baiyangdian Lake.

The highest concentration of total PBDEs in sediment was found at S2, Baoding City (307.2 ng/g dw), which is a larger industrial city comprising automobile, textile, building materials, electronic products manufacturing factories. The lower concentrations of PBDEs in sediments were found in Baiyangdian Lake, ranging from 7.4 to 23.7 ng/g dw, which possibly related to be far from industrial and urban activities.

DBDPE was found in all the sediment samples. The concentrations of DBDPE in sediments ranged from 16.1–68.2 ng/g dw for Fuhe River and 1.1–5.3 ng/g dw for Baiyangdian Lake. The levels of DBDPE in Fuhe River were significantly higher than those of Baiyangdian Lake ($P < 0.05$). It can be concluded that DBDPE was being used and emitted in North China. Generally, the DBDPE/BDE209 ratio can be used to assess the relative usage of the two brominated flame retardants (BFRs). In this study, the ratio of DBDPE/BDE209 was below 1 for most sediment samples, except for S1 and S12. Although DBDPE was marked as replacement for deca-BDE, deca-BDE (BDE209) still was the major brominated compounds in Baiyangdian Lake regions. The correlation analysis between DBDPE and BDE209 was conducted for all the samples (Fig. 3). Significant correlation between DBDPE and BDE209 was found in the present study ($P < 0.05$). The results indicated that the emission sources of DBDPE were similar to deca-BDE. DBDPE, as an alternative of BDE209, was used in applications similar to deca-BDE. These findings were consistent with those for sediment samples from South China (Zhang et al., 2009).

2.2 Geographical comparisons

PBDEs are predominantly incorporated into plastic polymers in electronic components (Bayen et al., 2003) and the presence of PBDEs in other locations around the world are generally thought to be derived from discharges of wastes associated with the production or dismantling of electronic equipment (Shi et al., 2009). PBDEs have been measured in river, lake, marine sediments from different regions of the world (Table 2). The concentrations of PBDEs in the present study were compared with those

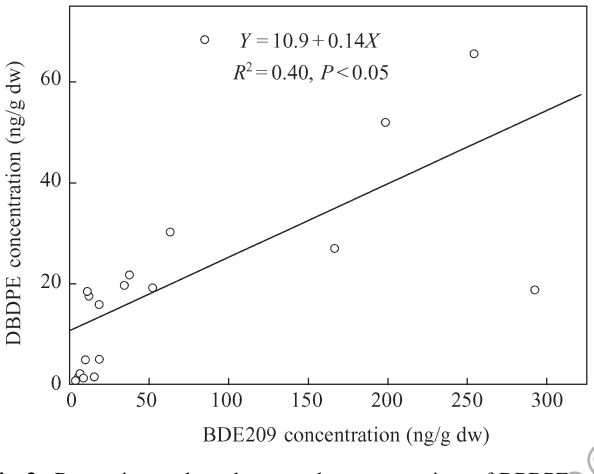

Fig. 3 Regression analyses between the concentrations of DBDPE and BDE209 in sediments from Fuhe River and Baiyangdian Lake.

Table 2 PBDEs and DBDPE concentrations (ng/g dw) in sediments from different regions worldwide

Location	Concentration (ng/g dw)			
	Tri- to hepta-BDE	BDE209	DBDPE	Reference
Qingdao near shore, China	0.1–5.5	–	–	Yang et al., 2003
Pearl River, South China	1.1–49.3	26.3–3580	–	Mai et al., 2005
Dongjiang River, South China	2.2–94.7	21.3–7340	–	Mai et al., 2005
Michigan Lake, USA	1.7–4.0	–	–	Song et al., 2005
Spanish coast, Spain	0.2–3.9	–	–	Eljarrat et al., 2005
Yangtze River Delta, China	2.1–3.9	–	–	Shen et al., 2006
Greifensee Lake, Switzerland	1.6	–	–	Kohler et al., 2008
Bohai Sea, North China	0.07–5.24	–	–	Wang et al., 2009
Western Scheldt, Netherlands	–	–	24	Kierkegaard et al., 2004
Sewage sludge, Canada	–	–	6–30	McCrindle et al., 2004
Sewage sludge, Germany	–	–	70–220	Ricklund et al., 2008
Pear River Delta, South China	–	–	19–430	Zhang et al., 2009
E-waste area, South China	–	–	38.8–364	Shi et al., 2009
Fuhe River, North China	0.13–6.39	11.8–292.7	16.1–68.2	Present study
Baiyangdian Lake, North China	0.05–5.03	4.35–19.3	1.10–5.29	Present study

"–" not available.

reported for several other regions. ∑PBDE1 concentrations in sediments measured in the present study were comparable to those reported for Michigan Lake (Song et al., 2005), Greifensee Lake (Kohler et al., 2008), Spanish coast (Eljarrat et al., 2005), Bohai Sea (Wang et al., 2009), Qingdao near shore (Yang et al., 2003), and Yangtze River Delta (Shen et al., 2006), however, lower than those of Pearl River and Dongjiang River (Mai et al., 2005). The concentrations of octa-BDE, nona-BDE, and deca-BDE in sediments from Fuhe River and Baiyangdian Lake were higher than those from Greifensee Lake (Kohler et al., 2008). The concentrations of BDE209 in sediments from Fuhe River and Baiyangdian Lake were lower than those from Pearl River and Dongjiang River (Mai et al., 2005). The high concentration of PBDEs in the Pearl River Delta may be due to the intensive manufacturing of electronic products in those areas.

Few publications are available for the concentrations of DBDPE in surface sediments. The concentrations of DBDPE in sediment from Fuhe River and Baiyangdian Lake were comparable to those of sediment from Western Scheldt (Kierkegaard et al., 2004), sewage sludge from Canadian (McCrindle et al., 2004), however, lower than those of sediments from Pear River Delta (Zhang et al., 2009), e-waste area of Southern China (Shi et al., 2009), and sewage sludge from Germany (Ricklund et al., 2008). High concentrations in sediments from China and sewage sludge from Germany may be attributed to the rapid development of electronic and automobile industry. The Pear River Delta was one of the larger manufacture regions of electronic products.

2.3 PBDE congeners pattern and PCA

The compositional pattern of sediments from Fuhe River was similar to that of Baiyangdian Lake. BDE209 was the most predominant among all the PBDE congeners in the study area (Fig. 4). The contributions of BDE209 to the total PBDEs ranged from 85.7% to 97.3% in sediments from Fuhe River, and 79.4% to 92.1% in Baiyangdian Lake with the exception of one sample (S15), in which, PBDEs had a relatively low abundance of BDE209 (35.6%). The

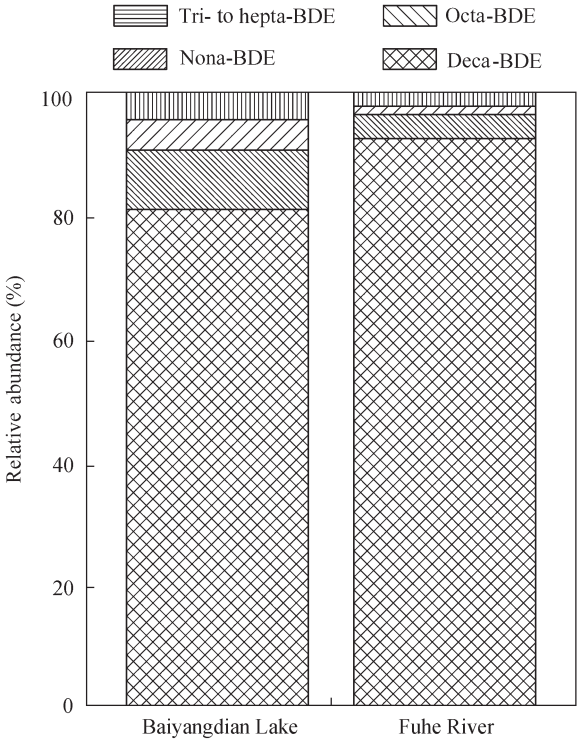

Fig. 4 PBDEs compositions pattern in sediments from Fuhe River and Baiyangdian Lake.

findings agree with the fact that deca-BDE mixture account for most of the total BFRs production in North China, which was similar to the reports in the other regions of China (Mai et al., 2005). The total contributions of octa-BDE and nona-BDE to the total PBDEs were 1.3% and 3.9% for Fuhe River, and 4.9% and 9.6% for Baiyangdian Lake, respectively. The regression analysis between deca-BDE and the sum of octa-BDE and nona-BDE was conducted for all the samples. Significant correlation was found in the present study ($P < 0.05$). The results indicated that deca-BDE mixture possibly could be the original source for the octa-BDE and nona-BDE in the study area.

The compositional pattern for low brominated flame retardants is illustrated in Fig. 5. Among the low brominated congeners (∑PBDE1), BDE47 and BDE99 were

the predominant compounds. The total contributions of BDE47 and BDE99 to the ∑PBDE1 ranged from 24.4% to 64.9% (average value, 52.1%), and 33.5% to 68.9% (average value, 44.1%) in sediments from Fuhe River and Baiyangdian Lake, respectively. BDE47 and BDE99 were the major congeners in the technical penta-BDE mixture, comprised 87% of ∑PBDE (La Guardia et al., 2006). The compositional patterns of Fuhe River and Baiyangdian Lake were similar to the compositions of major penta-BDE commercial mixtures (La Guardia et al., 2006). Clearly, penta-BDE commercial mixture was another major formula used in Baiyangdian Lake region in addition to the technical deca-BDE mixture. Although the usage of penta-BDE and octa-BDE was officially banned in 2006 in China, the findings indicated that Fuhe River and Baiyangdian Lake were possibly subjected to the contamination of penta-BDE mixture in the past years.

Principal component analysis (PCA) was used to further examine the potential sources of PBDEs in sediments from Fuhe River and Baiyangdian Lake (Fig. 6). The PCA classified the PBDE congeners into two distinct clusters, cluster I (low brominated congeners) and cluster II (high brominated congeners), accounting for 58.2% and 22.5% of variation in PBDE concentrations, respectively. It was noticeable that PBDE congeners were distributed to different principal components according to the molecular weights or bromine numbers of substitution. These results indicated that the PBDE congener profiles in sediment samples have been subject to complex environmental processes, and the different physical-chemical properties of the congeners may dictate the distribution of PBDEs in the present study.

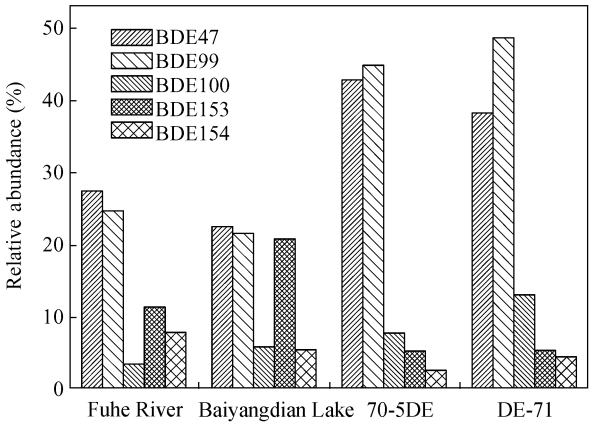

Fig. 5 penta-BDEs patterns in sediments from Fuhe River and Baiyangdian Lake compared with those of Bromkal 70-5DE and DE-71 technical mixture. Data of commercial formulations were drawn from the literature: Bromkal 70-5DE and DE-71 from (La Guardia et al., 2006).

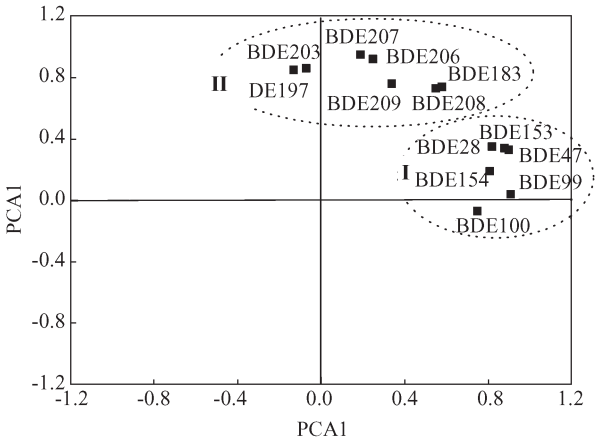

Fig. 6 Principle component analysis (PCA) score plot for sediments samples from Fuhe River and Baiyangdian Lake.

3 Conclusions

This study first provided the levels of PBDEs and DBDPE in the sediments from Fuhe River and Baiyangdian Lake, North China. PBDEs and DBDPE were detected in all the sediment samples, showing their ubiquity in the sediments in the study area. The high contamination of PBDEs and DBDPE found in sediment samples from Fuhe River could be attributed to the high industrial impact of this area. The levels of the contaminants appear to be comparable to the levels at contaminated sites worldwide. The congener patterns of PBDEs in sediments from Fuhe River and Baiyangdian Lake were similar, which indicated that BDE209 was the most predominant among the congeners. The studies for degradation of BDE209 are required to determine the origin of the contamination. The compositional patterns indicated that the major sources of PBDEs and DBDPE were probably wastes discharges from Baoding City, Hebei Province. Further study involving large number samples are needed to comprehensively investigate the contamination of organohalogen compounds in Fuhe River and Baiyangdian Lake, North China.

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