



Sediment Records of Polybrominated Diphenyl Ethers (PBDEs) from the Anhui Province Section of Yangtze River, China

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Abstract

This study investigated the temporal changes of PBDEs in the sediment cores from the Anhui Province section of Yangtze River (YR), China. The dramatic increase of $\sum\text{Br}_3\text{-Br}_9\text{-BDEs}$ and BDE-209 concentrations in cores in the period of ca. 1990–2008 was consistent with the booming of regional and national production and consumption of household appliances and electronics. Following declines in BDE-209 and $\sum\text{Br}_3\text{-Br}_9\text{-BDEs}$ concentrations can be associated with the decreasing use of legacy commercial PBDE mixtures in recent years. Compared with the different regions around the world, sediments in the YR were moderately polluted by PBDEs. The levels and the detection rates of BDE-209 were highest among the detected PBDEs single congener. The composition analysis indicated that Deca-BDE, Octa-BDE, and Penta-BDE were the dominant forms in the YR. The increasing trends of both BDE-209 and $\sum\text{Br}_3\text{-Br}_9\text{-BDEs}$ in the YR during 1990s and 2000s largely reflected the time periods for transferring PBDE sources from the developed countries to China. TOC and finer particles were strongly correlated with distributions of PBDEs in sediments.

Keywords Polybrominated diphenyl ethers · Sedimentation · Source identification · Yangtze River

Polybrominated diphenyl ethers (PBDEs) have been extensively used in various plastics, textiles, building materials, foam-upholstered furniture and electronic circuitry as flame retardants for more than five decades (Abbasi et al. 2015). PBDEs entered into the environment during production, use and disposal of PBDE-containing products (Drage et al. 2015). Existing production and usage inventory indicated that the cumulative global production of PBDE technical

products was ca. 1.5–1.7 million tons since the 1960s and reached a peak in 2003 (~ 75,000 tons) (Da et al. 2019). Commercial PBDEs comprise three main products: Penta-BDE (containing 70% of BDE-47 and BDE-99), Octa-BDE (containing 40% of BDE-183), and Deca-BDE (containing 98% of BDE-209) (Cetin et al. 2019). Although forbidden by the Stockholm Convention, Deca-BDE mixtures continue to be used in many countries at high production and usage levels, especially in China (Li et al. 2016). BDE-209, the dominant component of Deca-BDE, can degrade by debromination with appropriate light and microorganisms, changing from highly brominated diphenyl ethers to lowly brominated diphenyl ethers. The lowly brominated diphenyl ethers are more toxic than the higher brominated congeners (Lee and Kim 2015).

PBDEs can be transported to aquatic environments in different ways, including atmospheric deposition, surface runoff, and random discharges (Chen et al. 2018). Previous studies showed that PBDEs have been detected in sediments, atmosphere, soils, plants, bird eggs, human milk, marine animals and blood (Da et al. 2019). Sediments with anoxic and light-protected characteristics are important sinks for PBDEs due to slow anaerobic debromination (Abbasi et al. 2015). A well-dated sediment core could be used to reconstruct

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the historical records of PBDEs in aquatic environments. Investigations of the sedimentary records of PBDEs in China mainly focused on the coastal locations (i.e., Bohai Sea, Yellow Sea, East Sea, and Pearl River estuary), whereas less attention has been paid to the sedimentary records of PBDEs in inland river (Gao et al. 2013; Hong et al. 2010; Yao et al. 2016; Wu et al. 2017).

The metropolitan area of the Yangtze River (YR) has been an industrial heartland and a prime economic mover in China (Yao et al. 2016). With the rapid growth of economic and industrial development, the YR has been exposed to many different types of pollution. The excessive industrial waste of plastics, textiles and electronic products thrown in the YR could lead to massive accumulation of pollutants in the sediments (Sun et al. 2018). As of 2018, the cumulative production of PBDE technical products was estimated to be 3500 tons in Anhui Province section of the YR (Da et al. 2019). Previous investigations of PBDE, PCB, OCP, and PAH contamination has been conducted in the surface sediments in the YR (Chen et al. 2018; Sun et al. 2016). Building on these findings, this study further investigated the past changes in PBDEs in sediment cores collected from the Anhui Province section of YR. The specific objectives included investigating historical PBDE input and contamination pathways, apportioning PBDE sources, and investigating their environmental processes.

Materials and Methods

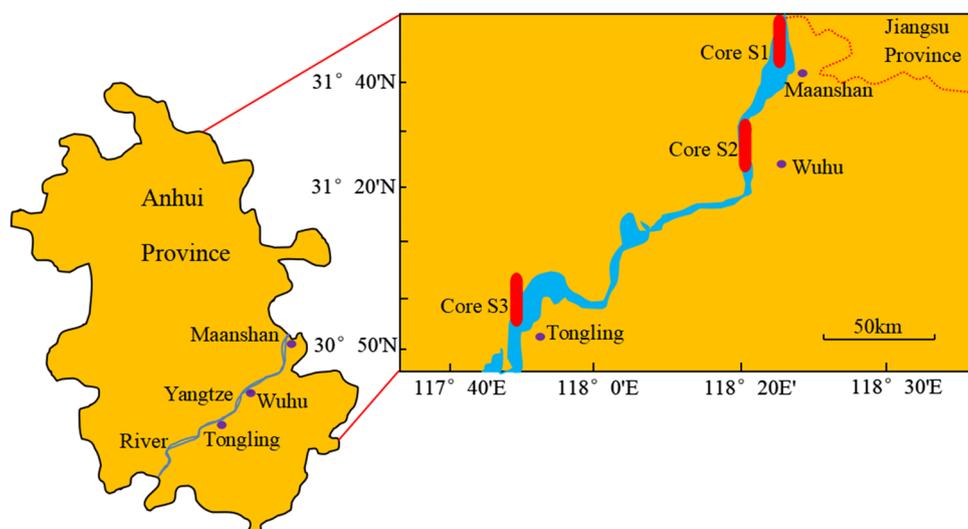
Core S1 (31° 44' 7.01" N, 118° 27' 46.64" E) was collected from the YR adjacent to Ma'anshan City in July of 2018 using a stainless steel static gravity corer of inner diameter 80 mm with length of 37 cm. Core S2 (31° 22' 55.94" N, 118° 20' 43.67" E) was collected from the YR adjacent

to Wuhu City in July of 2018 using a gravity corer with length of 35 cm. Core S3 (30° 51' 13.83" N, 117° 43' 48.84" E) was collected from the YR adjacent to Tongling City in July of 2018 with length of 39 cm. All three cities in the collection sites are industrial bases in Eastern China. A large number of electronic products (over 1000 tons) were produced from the three cities (<http://tjj.ah.gov.cn/>). It was noted that the location of core S2 and S3 was adjacent to the Yangtze River Bridge; the location of core S1 was adjacent to Ma'anshan Port (Fig. 1). There was almost no water at the three sampling locations, close to the beach. The three locations have high sediment deposition and low current velocity. The cores were cut into 1.0 cm intervals using an electric stainless steel cutter, then wrapped with aluminum foil baked at 450 °C, and transported back to the laboratory. The sediment samples were pulverized to < 200 mesh after freeze-drying, and stored in a brown glass sample bottle at - 20 °C until analysis.

BDE-28, BDE-37, BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209 were measured in the three sediment cores. The standard solutions of PBDEs used for quantification were obtained from AccuStandard (New Haven, USA). The surrogate standards ($^{13}\text{C}_{12}$ -BDE-138 and $^{13}\text{C}_{12}$ -BDE-153) were obtained from Cambridge Isotope Laboratories Inc (Andover, MA USA). Neutral silica gel (Qingdao Haiyang Chemical Co., Shandong, China) and alumina (Qingdao, China) were activated at 180 °C for 16 h prior to use and then deactivated with 3% ultrapure water (97% silica gel and alumina). Anhydrous sodium sulfate (Sinopharm Chemical Reagent Co., China) was baked at 400 °C for 8 h before use. All solvents used were of pesticide residue grade.

The samples were analyzed for ^{210}Pb activities with γ -spectrometry using Canberra ultra-low-background germanium detectors. The dating methods were detailed

Fig. 1 Sampling locations of the sediment cores in the Yangtze River, China



in the published literature (Da et al. 2019). Briefly, sediment samples were placed in sealed containers for 3 weeks to achieve equilibration of ^{226}Ra and its daughter isotope ^{210}Pb . Every ground sample (10 g) was homogenized, then loaded into a Petri dish (50 mm i.d.×9 mm). The relative errors for this method were better than $\pm 2.3\%$. The sediment samples represented 57 ± 2.34 years (from 1961 to 2018) in core S1, 53 ± 3.32 years (from 1965 to 2018) in core S2 and 60 ± 3.31 years (from 1958 to 2018) in core S3. The ^{210}Pb dated sedimentary rates for cores S1, S2 and S3 were 0.61, 0.59 and 0.54 cm/year, respectively.

A total of 111 sediment samples were analyzed for PBDEs in the three cores. Extraction and cleanup of PBDEs were based on the methods reported by Cheng and Ko (2018). Extraction of target compounds or congeners was performed with accelerated solvent extraction (Dionex ASE-200). Prior to analysis, the samples were spiked with surrogate standards (^{13}C -BDE-138 and BDE-153), and then extracted with 50 ml of acetone and n-hexane (v:v = 1:1). After extraction, the extracts were purified on a silica gel column (i.d.=1.0 cm) packed with 6 cm neutral silica (3% H₂O deactivated), 12 cm sulfuric acid silica (44% sulfuric acid, w/w) and 2 cm anhydrous sodium sulfate from bottom to top. The eluents were evaporated, to dryness with slight warming under nitrogen gas flow, exchanged to two ml of hexane, and an amount of internal standard (BDE-77) was added. The extraction solution was concentrated to 1 ml before instrumental analysis.

PBDEs were measured by GC-MS (Agilent 7890 coupled with an Agilent 5975C mass spectrometer) in EI mode. Separation was performed with two fused-silica capillary columns: 30 m×0.25 mm×0.1 μm for Br₃-Br₉-BDE, 15 m×0.25 mm×0.1 μm for BDE-209. Ultra-high purity helium was used as the carrier gas at a constant flow of 0.9 ml/min. The oven temperature program for Br₃-Br₉-BDE was as follows: The column oven temperature was set to 80 °C and held for 1 min, then ramped at 20 °C/min to 200 °C and held for 10 min, increased at 20 °C/min to 310 °C, and held for 8 min. The temperature program for BDE-209 was as follows: initial 100 °C and maintained for 1 min, increased at 20 °C/min to 280 °C for 10 min. The injector and transfer line temperatures were held at 280 °C and 300 °C, respectively.

The total organic carbon (TOC) percentage content of the sediment was measured by the combustion oxidation and nondispersive infrared absorption method (Yuan et al. 2016). Weighed 0.05 g of sample, put it into quartz cup, and added phosphoric acid solution slowly to remove interference until there was no bubble in sample. Put quartz cup into total organic carbon analyzer to measure response value. The calculation formula was as follows:

$$W = \frac{(A - A_0 - a)}{b \times m_1 \times 1000} \times 1000$$

where W is the content of organic carbon (%); A is response value; A₀ is response value of blank sample; a is intercept of standard curve; b is slope of standard curve; m₁ is the mass of dry matter (g).

The particle size distribution of sediment was measured by sieving on mesh sizes, with particle sizes classified as: > 0.3 mm, 0.1–0.3 mm and < 0.1 mm.

All glass containers were cleaned with distilled water and acetone and heated to 450 °C for 6 h to remove any organic contamination. One procedural blank (in which a filter paper identical to that used to wrap the sediments was solvent extracted and performed in the same way as the samples) was included in each batch of 10 samples to check for laboratory contamination, none of the compounds were detected in the blanks. A matrix-spiked (^{13}C labeled BDE-138) sample was used for calculation of matrix-spiked recovery. One duplicate sample was analyzed with every five samples to measure repeatability of the analysis. The recoveries for spiked samples ranged from 89.1% to 103.1%. The relative standard deviation (RSD) of the duplicate samples ranged from 0.8% to 7.9%. 10 ng/g for each standard solution was put into the blank samples, and their average recoveries were listed in Table S1.

Results and Discussion

The detection rate of $\Sigma_{10}\text{PBDE}$ was 89.2% in the core S1, 82.9% in the core S2 and 71.8% in the core S3 (Table 1 and Table S2), suggesting that PBDEs were widespread in the study region. The residual levels of $\Sigma\text{Br}_3\text{-Br}_9\text{-BDE}$ (sum of BDE-28, 37, 47, 85, 99, 100, 153, 154, 183) ranged from 0.02 to 86.7 ng/g dry weight (dw) (mean value = 24.7 ng/g dw) in core S1, 0.02 to 59.7 ng/g dw (mean value = 17.8 ng/g dw) in core S2, and 0.01 to 60.3 ng/g dw (mean value = 14.4 ng/g dw) in core S3. BDE-209 was much more abundant than the other single congeners, with concentrations varying from 0.02 to 2.6 ng/g dw (mean = 6.0 ng/g dw) in core S1, 0.01 to 19.7 ng/g dw (mean = 5.4 ng/g dw) in core S2, and 0.54 to 35.8 ng/g dw (mean = 6.0 ng/g dw) in core S3 (Table 1 and Table S2).

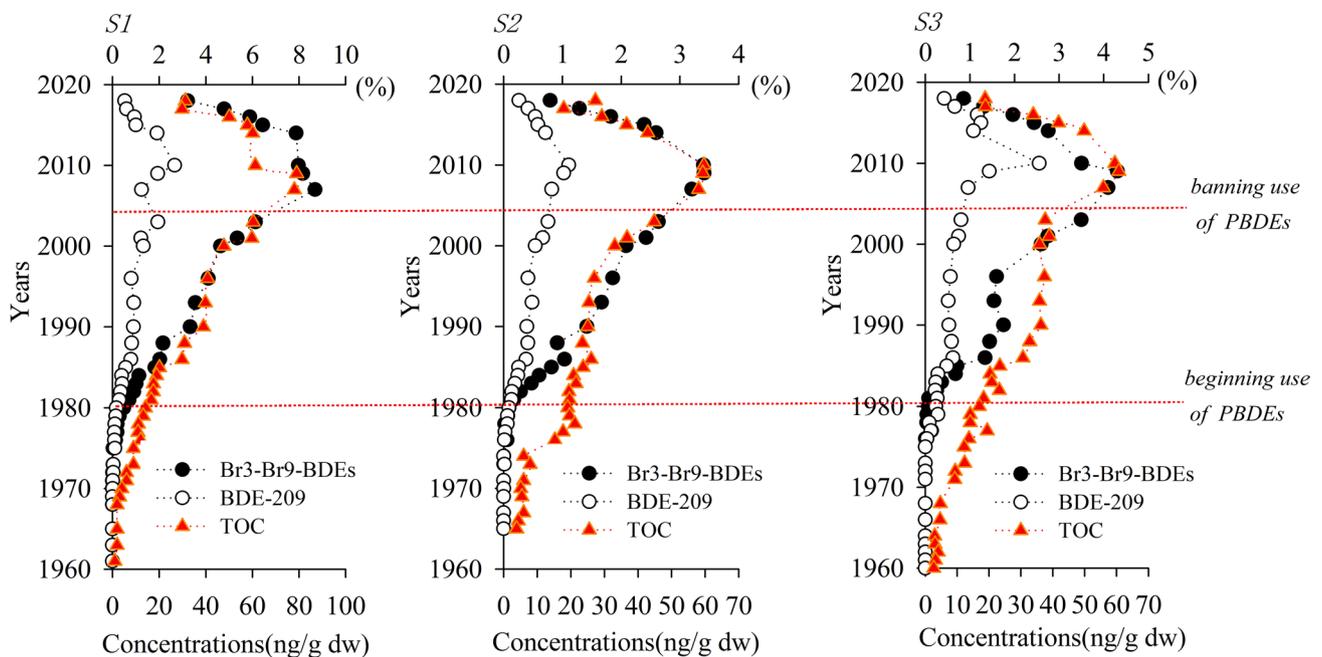
Compared with the previous studies (Table S3), the detected levels of $\Sigma\text{Br}_3\text{-Br}_9\text{-BDEs}$ from the YR were higher than those from most regions in China, except for those from East Sea of China (Li et al. 2016). The concentrations of BDE-209 from the YR were higher than those from most regions, except for those of East China Sea (Li et al. 2016) and Nansha mangrove in South of China (Wu et al. 2017). In general, the levels of PBDEs in this study area are relatively high.

Table 1 Concentrations of PBDEs (ng g⁻¹dw) in the three sediment cores

PBDEs	Concentrations of PBDE Congeners (ng g ⁻¹ dw)								
	Core S1			Core S2			Core S3		
	Mean	Range	Detection rate (%)	Mean	Range	Detection rate (%)	Mean	Range	Detection rate (%)
BDE-28(Tri-BDEs)	4.2	0.01–17.1	54.05	1.93	0.05–7.59	57.14	2.11	0.01–12.7	51.28
BDE-37 (Tri-BDEs)	4.2	0.09–16.1	75.68	2.19	0.18–7.11	68.57	3.10	0.02–10.4	69.23
BDE-47(Tetra-BDEs)	4.0	0.03–13.9	78.38	3.31	0.01–12.33	71.42	2.36	0.01–9.4	56.41
BDE-85(Penta-BDEs)	0.18	0.01–0.89	70.27	0.12	0.01–0.51	71.42	0.09	0.04–0.49	46.15
BDE-99(Penta-BDEs)	0.10	0.01–0.57	51.35	0.11	0.01–0.45	57.14	0.11	0.01–0.56	51.28
BDE-100 (Penta-BDEs)	0.17	0.01–0.56	59.46	0.12	0.01–0.41	57.14	0.09	0.01–0.45	53.85
BDE-153 (Hexa-BDEs)	3.0	0.01–13.5	64.86	2.62	0.01–10.99	65.71	2.17	0.01–9.6	56.41
BDE-154 (Hexa-BDEs)	3.7	0.02–16.8	67.57	2.89	0.02–10.99	60.00	2.04	0.01–9.9	53.85
BDE-183 (Hepta-BDEs)	5.2	0.01–17.4	72.97	4.48	0.01–13.45	68.57	2.33	0.1–8.2	56.41
ΣBr ₃ -Br ₉ -BDEs	24.7	0.02–86.9	81.08	17.77	0.02–59.71	80.00	14.41	0.01–60.3	71.79
BDE-209 (Deca-BDE)	6.0	0.02–26.6	86.49	5.35	0.01–19.37	80.00	6.04	0.54–35.8	66.67
Σ ₁₀ PBDEs	30.7	0.02–106.4	89.19	23.12	0.01–77.60	82.86	20.45	0.01–84.8	71.79

BDE congeners excluding BDE-209 (denoted as ΣBr₃-Br₉-BDEs) were detected at low levels in the deep sediment layers dated back to the mid-1960s (ca.1964–1966) (Table S2). The initial detectable time of BDE-209 was ca.10 years later (starting from 1973 to 1976) than those of ΣBr₃-Br₉-BDEs, coinciding with beginning of production, usage, and disposal of commercial Penta-, Octa-, and Deca-BDE products in the world (Abbasi et al. 2015). The ΣBr₃-Br₉-BDEs concentrations across the cores were in low

level during the mid-1960 to mid-1970s, followed by slow increases and of comparable magnitude from mid-1970s to post-1980s (Fig. 2). Rapid increases in residue levels started from 1970s to 2010s, and peaked in 2010s. Both ΣBr₃-Br₉-BDEs and BDE-209 concentrations declined after 2010s in the cores. Many global processing factories for electronic and telecommunication equipment were built in China due to cheap labor costs since the early 1980s (He et al. 2013). Therefore, the production and consumption in household


Fig. 2 Time trends of PBDE concentrations in the sediment cores

appliances and computers had increased rapidly with the rapid growth of the economy until the late 2010s in China (Guo et al. 2015). The Chinese government banned production and usage of commercial penta-BDEs and octa-BDEs in 2004, and promulgated some strict environmental protection regulations in recent years (Wang et al. 2016). Thus the historical accumulations of PBDEs in the cores could be affected by local PBDEs historical input. Interestingly, the decreasing trend of PBDEs did not occur before the 2010s although the restrictions on the production and usage of commercial Penta-BDEs and Octa-BDEs were implemented in 2004. The surface components of sediments often settle and get resuspended many times before they are actually buried by deposition of heavier loads of newer materials. In the process, mixing occurs which makes it impossible to strictly correlate depositional layers to an actual discrete year. Thus, the PBDE levels in the cores from the YR would be impossible to reflect any decline quickly from 2004 to 2010. Although the commercial Penta-BDEs and Octa-BDEs were forbidden in the factories in 2004, their legacy-products were still used and recycled (Zhang et al. 2014), which would possibly lead to the continuous release of PBDEs. A gradual decreasing trend in PBDEs pollution has been reported since the early 1990s in some developed countries and regions, including America, Japan and Europe (Yun et al. 2008; Minh et al. 2007). Nonetheless, the annual global consumption of brominated flame retardants showed an increasing trend in the past several years, particularly for the commercially available deca-BDE-containing products (Gao et al. 2013). The time (after the 1990s) when PBDEs gradually increased in the three cores from the YR, the residual levels decreased in the developed countries, possibly reflecting the off-shoring of their use from the developed regions to the YR (Abbasi et al. 2019; Chen et al. 2007).

Variation in levels of Σ_{10} PBDE in the cores was observed, likely resulting from the different geographic locations and surrounding anthropogenic activities. Residuals of PBDEs

in the upstream (S3) and midstream (S2) flowed with water to the downstream and accumulated in the site (S1). In addition, site S1 was adjacent to Jiangsu Province where the industrial economy was more developed than that of Anhui Province. The industrial PBDEs from the adjacent Jiangsu Province could migrate to the site S1, resulting in higher levels of PBDEs at this site.

BDE-209 was dominant in the three cores (Fig. 3), indicating industrial Deca-BDE mixtures (over 97% BDE-209) (Cheng and Ko 2018) were the major source of PBDEs in the YR. Industrial Deca-BDE mixtures were a large portion of the PBDE mixtures used in China. It was estimated that the total amount of Deca-BDE mixtures was over 2000 tons (Zhang et al. 2014). Moreover, BDE-209 was easily affiliated with fine-grained particles and settlement in sediments due to a high octanol–water partition coefficient (Li et al. 2017). BDE-209 concentrations after the 1980s were higher than those before the 1980s in the cores, attributing to high usage of the commercial Deca-BDE containing products after the 1980s in China (Chen et al. 2015). Compared with other ages, the highest proportion (over 50%) of BDE-209 in the cores was found in the 1960s. In the 1960s, polybrominated diphenyl ethers other than BDE-209 were rarely detected. BDE-209 accounted for a large proportion of the concentration detected in the 1960s, which also indicates that BDE-209 was more persistent than other polybrominated diphenyl ethers in use at the time. As shown in Fig. 2, the maximum concentration of BDE-209 in the cores appeared in 2010. However, the concentration ratio of BDE-209 was lower compared with that in the 1960s, likely due to the fact that the other polybrominated diphenyl ethers also began to be more widely used after the 1960s (Li et al. 2017). BDE-183 contributed 17%, 19% and 11% to Σ_{10} PBDEs in the cores S1, S2 and S3, respectively, implying the extensive use of commercial Octa-BDE products. The wide distribution of BDE-183 was also observed in the cores from the Huaihe River, Bohai Sea, Chaohu Lake, Deep Bay of South China

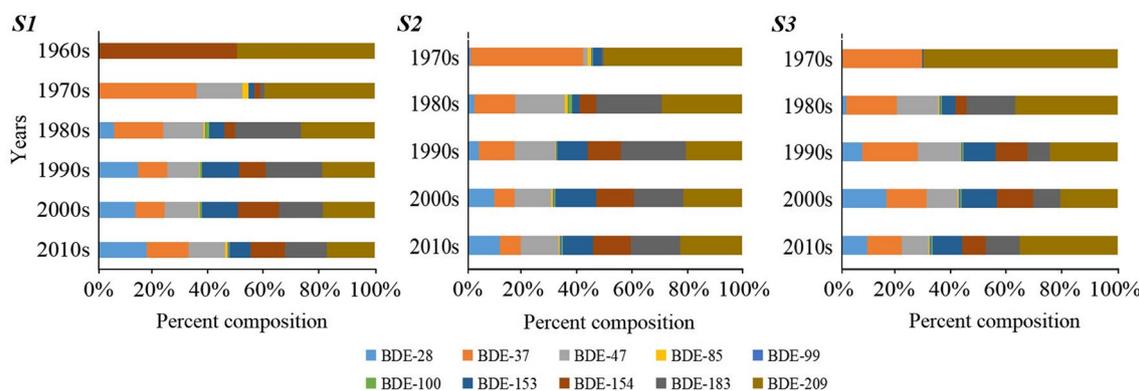


Fig. 3 Compositional patterns of PBDEs in the sediment cores

(Qiu et al. 2010). BDE-47 was also prevalent in the cores. BDE-47 was the major component in the commercial Penta-BDE. The wide distribution of BDE-47 in the study area suggests widespread use of industrial Penta-BDE substance in the YR. It was reported that BDE-99 was also the primary compound in the industrial Penta-BDE (Wu et al. 2019). The levels of BDE-47 exceeded BDE-99 in almost all the years. This was consistent with the usage records of commercial BDE mixtures, consisting of 80% of BDE-47, BDE-99, and BDE-153 for Penta-BDEs, 30% of BDE-153 and BDE-183 for Octa-BDEs, respectively (Qiu et al. 2010). Compared with BDE-47, BDE-99 has a lower water solubility and higher octanol-water partition coefficient (Cetin et al. 2019) and was relatively more unstable than BDE-47 (Liu et al. 2017). BDE-28 was also observed, indicating the debromination and degradation of higher brominated congeners to the lower brominated congeners.

TOC contents in the sediment samples varied from 0.1% to 7.9% with an average value of 2.68% in core S1, from 0.21% to 3.41% with an average value of 1.34% in core S2, and from 0.12% to 4.34% with an average value of 1.62% in core S3 (Table S4). In addition, the percentages of finer particles (particles size < 1 mm) varied from 0.01% to 87.13% with an average value of 23.56% in core S1, from 0.45% to 74.11% with an average value of 21.62% in core S2, and from 0.01% to 79.78% with an average value of 25.80% in core S3. Pearson correlation coefficients indicated both TOC ($R^2 > 0.7$, $p < 0.05$) and the percentages of finer particles ($R^2 > 0.8$, $p < 0.05$) were significantly correlated with PBDE concentrations in the core samples (Table S5). As shown in Fig. 2, a similar relationship between PBDEs and TOC was observed in the cores, where concentrations gradually increased at first and then decreased from the bottom to the sediment cores. Previous studies have also reported that the TOC content in sediment cores was influenced by the discharge of untreated domestic sewage and the hydraulic and mineralogical particulate fractions (need a reference to the previous studies). Moreover, linear regression analysis revealed a significant positive ($R^2 > 0.8$, $p < 0.01$) relationship between the finer particles and PBDE concentrations (Fig. 4 and Fig. S2), which possibly be attributed to the high K_{ow} of PBDEs congeners and the strong ability of fine particles and TOC to absorb the organic compounds (Chai et al. 2019).

The ecological risk of PBDEs to subaqueous organisms in the YR was evaluated according to the environmental quality guidelines (EQG) established by Canada (Environment Canada 2013). The EQG values for Tri-BDE, Tetra-BDE, Penta-BDE, Hexa-BDE and Deca-BDE in sediment were 44, 39, 0.4, 440, and 19 ng/g dw, respectively. Table S2 shows that Tri-BDEs, Tetra-BDEs and Hexa-BDEs levels were all lower than the EQGs. In addition, 48.8%, 42.9%, and 25.6% of the subsamples from cores S1, S2, and S3, respectively,

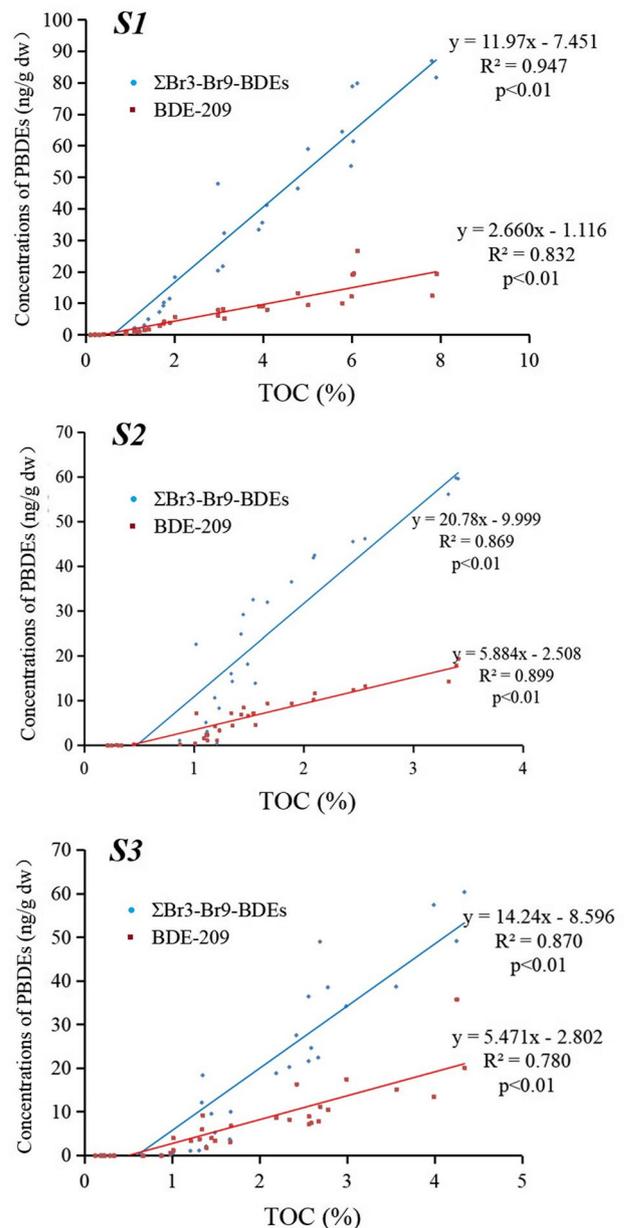


Fig. 4 Regression analyses of PBDE concentrations and TOC

contained Penta-BDE levels higher than the EQGs, and 10.8%, 2.9%, and 5.1% of the subsamples from cores S1, S2, and S3, respectively, had the Deca-BDEs at levels higher than the EQGs. These results indicated that there was a moderate hazard to the organisms in the study areas.

Temporal changes in the risk profiles are shown Fig. S3. The risk levels of Penta-BDEs and Deca-BDEs rapidly increased during the 1980s, 1990s, and early 2000s, followed by significant declines after circum-2010.

The dramatic increase of $\Sigma\text{Br}_3\text{-Br}_9\text{-BDEs}$ and BDE-209 concentrations in cores in the period of ca. 1990–2008 was consistent with the booming of regional and national

production and consumption of household appliances and electronics. Following this period, declines in BDE-209 and $\Sigma\text{Br}_3\text{-Br}_9\text{-BDEs}$ concentrations may be associated with reduced use of legacy commercial PBDE mixtures. Compared with the different regions around the world, sediments in the YR were moderately polluted with PBDEs. The levels and the detection rates of BDE-209 were highest among the detected PBDEs congeners. The composition analysis indicated that Deca-BDE, Octa-BDE, and Penta-BDE were the dominant forms in the YR. The increasing trends of both BDE-209 and $\Sigma\text{Br}_3\text{-Br}_9\text{-BDEs}$ in the YR during the 1990s and 2000s largely reflected the time periods for transferring PBDE sources from developed countries to China. TOC and finer particles were strongly correlated with distributions of PBDEs in sediments.

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Compliance with Ethical Standards

Conflict of interest The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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