



Polybrominated diphenyl ethers in water, sediment, soil, and biological samples from different industrial areas in Zhejiang, China

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) have been used extensively in electrical and electronic products, but little is known about their distribution in the environment surrounding the manufacturing factories. This study reports PBDE contamination in various matrices from the location (Liushi, Zhejiang province) that produces more than 70% of the low-voltage electrical appliances in China. Additionally, PBDE contamination was compared with other industries such as the e-waste recycling business (Fengjiang) in the same region. Specifically, we measured seven PBDE congeners (BDEs – 47, 99, 100, 153, 154, 183, and 209) in water, sediment, soil, plant, and animal tissues from four different areas in this region. The present study revealed elevated PBDE concentrations in all matrices collected from Liushi and Fengjiang in comparison with highly industrialized areas without significant PBDE contamination sources. In water samples, there were large variations of PBDE content and composition across different areas. In sediment/soil and biological samples, BDE-209 was the predominant congener and this could be due to the abundant usage of deca-BDE mixtures in China. Our findings provide the very first data on PBDE contamination in the local environments surrounding the electronics industry, and also reveal widespread PBDE contamination in highly industrialized coastal regions of China.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been widely used as flame retardants in many products such as plastics, electrical appliances, and furniture. The three major commercial PBDE mixtures are the penta-, octa-, and deca-BDEs. Because they are additives mixed into polymers and are not chemically bound to the plastic or textiles, they can be released into the environment easily [1]. In fact, PBDEs have been detected in various environmental matrices and human tissues, and an increasing trend was observed in recent years [2]. Their lipophilic, persistent, and bioaccumulative nature of PBDEs also renders them to be classified as one of the persistent organic pollutants (POPs). Their toxicity has also been widely studied in a variety of animal models (see [3] for a review), and significant concerns of toxicity on human health have

been raised, especially with the low-brominated PBDEs such as the penta-BDEs [4]. Correspondingly, the European Union banned the industrial penta- and octa-BDE products in 2004, and several states of the United States also joined this action in 2006 [2,5]. However, PBDEs are continuously released from previously produced materials treated with penta-BDE flame retardants and deca-BDE is still being widely used [5].

In China, the domestic production of brominated flame retardants (BFRs) was 10,000 tonnes in 2000, and deca-BDE is one of the most produced BFRs with continuous increases at an annual rate of 8% [6,7]. More recently, researchers in China started to focus on PBDE contamination in heavily polluted regions such as the notorious e-waste recycling site at Guiyu, Guangdong [8], the Pearl River Estuary in Guangdong province, south China [9], and the Yangtze River [10,11]. However, few studies examine the baseline PBDE levels in regular cities without any significant PBDE contamination source in comparison to cities with potential PBDE contamination in the same geographical region. In particular, no study has been conducted to examine PBDE distribution in various environmental matrices from areas that manufacture electrical and electronic products.

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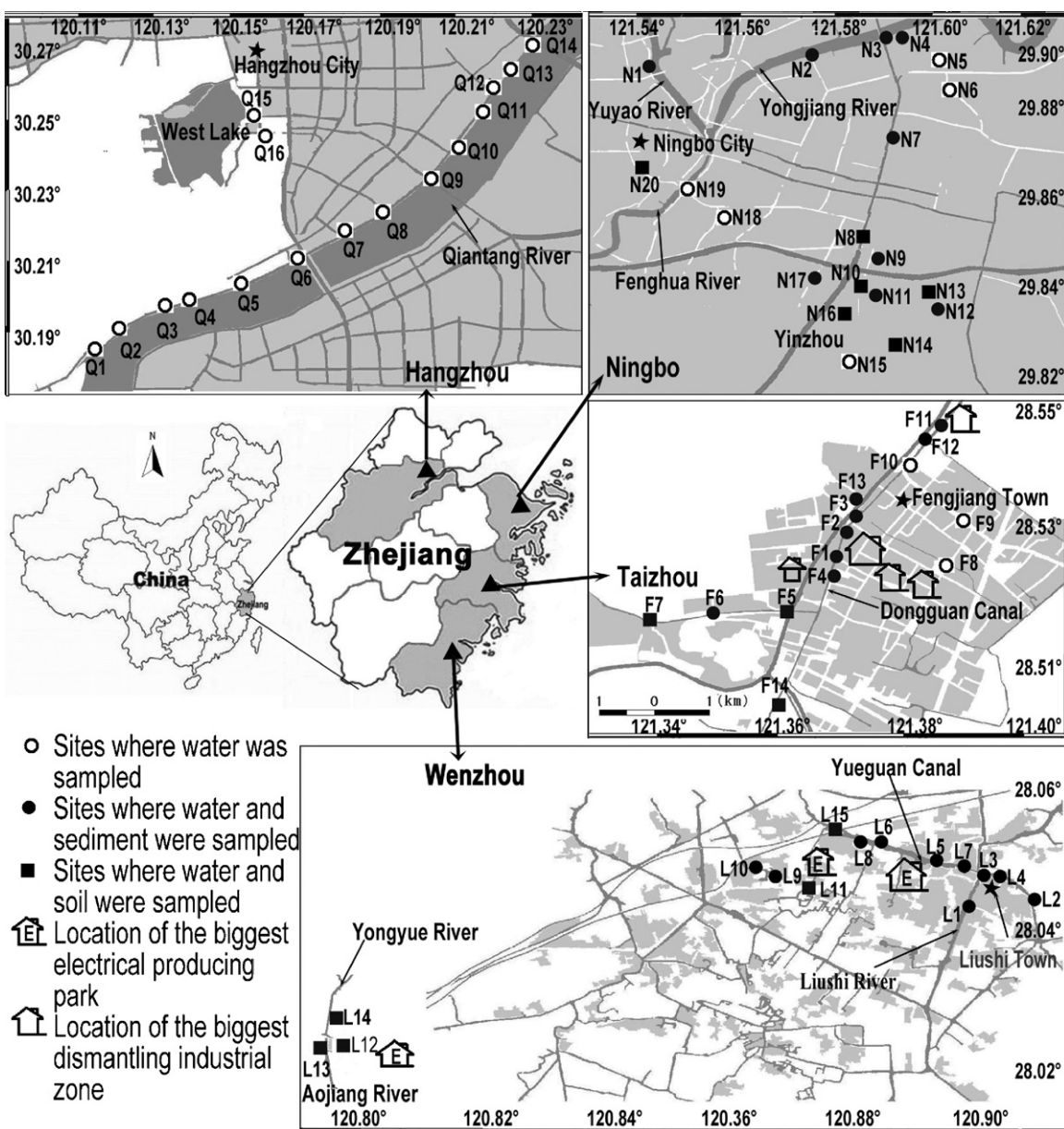


Fig. 1. Map of the study area and sampling sites.

The Zhejiang province is located on the southeast coast of China and is characterized by intensive industrial activities and urban development, where thousands of fast-growing electrical and electronic factories situated. The town of Liushi in Wenzhou district was renowned as the capital of low-voltage electronics in China, and it has a history of more than 30yr manufacturing various products. These products include electric capacitors, electric generators, cable wires and other PBDE-containing goods. This industry was expanded to a much larger scale in the past decade along with the rapid economic growth in China. Currently, there are approximately 1500 large scale manufactures with more than 200,000 workers employed therein. The present study was intended to investigate the PBDE levels from Liushi along with three other areas from Zhejiang province, and thus evaluate how the low-voltage electrical industry, with potential PBDE contamination sources, affect the local environment. Specifically, we quantified 7 BDE congeners in various environmental matrices such as surface water, sediment/soil, plants, and

animal tissues from four different industrial areas in Zhejiang province.

2. Materials and methods

2.1. Sampling sites

A total of 65 sampling sites from four areas with different industrial characteristics were established (Fig. 1). Besides Liushi (L1–L15), samples were also collected from Fengjiang (F1–F14), which is situated in Taizhou – the region that is currently predominated by e-waste recycling business, with a scale more or less similar to that of Guiyu in Guangdong province. Another two areas include Ningbo (N1–N20) and Hangzhou (Q1–Q16). Ningbo is the economic center of the Yangtze River Delta, and its Beilun Harbor is one of China's three biggest foreign trade ports. Hangzhou is the capital of Zhejiang province and is also a famous tourist city in China. It has undergone a rapid

Table 1
Descriptions of samples.

Sampling area	Water	Sediment	Soil	Biological samples			
				Plant	Alga	Snail	Crayfish
Liushi	15	10	4	8	4	10	6
Fengjiang	14	8	3	–	–	–	–
Ningbo	20	8	7	15	3	–	–
Hangzhou	16	–	–	–	–	–	–

“–”: no samples were collected.

industrialization and urbanization with its GDP tripling in the past decade.

2.2. Sample collection, preparation, and chemicals

Water, river sediment, surface soil, and biological samples were collected from all or some of the sites between November 2009 and June 2010 (Table 1). Water samples were collected from 0.5 m below the water surface using a 2 L sampler, and PBDEs were extracted with a solid-phase extraction (SPE) cartridge system (LC-C18) (Milwaukee, WI, USA). In brief, samples were first passed through 0.45 μm glass-fiber filters (GF/F) (Whatman, Clifton, NJ, USA) to get rid of the particulates, mixed with 40% methanol (v/v), and ultrasound sonicated for 30 min before being loading into the preconditioned SPE cartridges [12,13]. The cartridges were then dried and eluted with 3 mL methanol followed by 3 mL DCM and 3 mL n-hexane. The mixture of these extracts was collected in a 10 mL glass tube and used for further clean up. Water was removed from the extracts using processed anhydrous sodium sulfate (Na_2SO_4), the extracts were then concentrated, solvent-exchanged to hexane and further reduced to approximately 1 mL under gentle nitrogen gas. Further clean up was carried out in a 30 cm \times 10 mm i.d. glass column packed with, from bottom to top, 6 cm neutral alumina, 2 cm neutral silica, 5 cm alkalized silica, 2 cm neutral silica, 8 cm acidified silica, and 1 cm Na_2SO_4 . The PBDEs were eluted with 70 mL of 50% DCM in hexane, and the extract was concentrated with a rotary evaporator and evaporated to dryness under a gentle stream of nitrogen gas and re-dissolved in 0.6 mL iso-octane prior to analysis.

Sediment samples at the top 0–5 cm layer were collected with a stainless steel columnar sampler (K-BTM, Cole-Parmer, Vernon Hills, IL, USA). Surface soil (top 0–5 cm) samples were collected within 0.05 km of the water sampling site. PBDEs in these samples were extracted [14] using a microwave extraction unit equipped with Teflon extraction cells (Model Multiwave 3000 Anton Paar GmbH., Graz, Austria). In brief, sediment or soil samples were freeze-dried in the dark, sieved through a stainless steel mesh with 0.2-mm pore size, grounded, homogenized with anhydrous Na_2SO_4 , and dried. Samples of less than 1.0 g dry mass were resuspended in 20 mL of hexane/acetone (1:1, v/v), digested in the microwave extraction unit at 110 °C for 20 min, combined with activated copper granules, and sonicated for 30 min to remove sulfur. The extracts were filtered, concentrated, dissolved in 1 mL n-hexane, and cleaned up with sulfuric acid treatment followed by a Florisil column elution (Waters, Sep-Pak Milford, MA, USA). The PBDEs were subsequently eluted with 5.0 mL iso-octane, evaporated to dryness with nitrogen gas, and the residue was dissolved in 1 mL n-hexane prior to analysis.

Biological samples include plants of *Juncus effuses* L.; *Potamogeton crispus* L.; *Alternanthera philoxeroides* Griseb.; *Rumex japonicus* Houtt. and *Acorus calamus* L., algae (cyanobacteria), apple snails *Pomacea canaliculata* (Lamarck, 1819), and crayfish *Procambarus clarkia* (Girard, 1852), and were collected near or at water/soil sampling sites. Plants and algal samples were rinsed with distilled water and dried with filter paper before ground into a homogeneous

mixture by mortar and pestle. For animal samples, the edible parts, the soft tissue from the snail and the muscle from the crayfish, were collected and processed for PBDE analysis. These tissues were minced with a homogenizer. These samples of 1.0 g wet weight were mixed with Na_2SO_4 and subjected to microwave extraction as described above for soil and sediment samples.

Standards of BDEs – 47, 99, 100, 153, 154, 183, and 209 were purchased from AccuStandard (New Haven, CT, USA), and stock solutions were prepared with pesticide grade (re-distilled) iso-octane and stored at –20 °C. All chemicals used were of reagent grade with purity >99.8%. Except for iso-octane (Sigma, St. Louis, MO, USA), all other chemicals were purchased from Shanghai Chemical Company (Shanghai, China). Sodium sulfate was dried at 650 °C in a Muffle furnace for 8 h prior to use.

2.3. Sample analysis

PBDEs were analyzed with gas chromatograph 6890 coupled to a mass spectrometer (MS) 5975C (Agilent Technologies, Wilmington, DW, USA) using an electron impact (EI) ion source in the selective ion monitoring (SIM) mode. For GC analysis, a DB-5HT (15 m \times 0.25 mm i.d., 0.1 μm film thickness; J&W Scientific, Folsom, CA, USA) capillary column was used, and samples were injected in a splitless mode with a volume of 1 μL and 3 min solvent delay time. For determination of BDEs – 47, 99, 100, 153, 154, and 183, the column temperature started at 100 °C, held for 1 min, increased to 220 °C at a rate of 30 °C/min and held for 1 min, then increased to 250 °C at a rate of 3 °C/min and held for 1 min. Helium was used as the carrier gas at a flow rate of 0.8 mL/min with a port temperature of 280 °C. The ion source temperature was 230 °C and the interface temperature was 300 °C. For BDE-209, different temperature program was used to reduce the retention time, thus prevent it from degradation and increase its resolution. Specifically, the column temperature program was carried out as follows: started at 100 °C, increased to 325 °C at 30 °C/min and held for 3 min. Similarly, helium was used as the carrier gas at a flow rate of 1.2 mL/min with a port temperature of 300 °C. The ion source and interface temperatures were both set to 300 °C. We have also measured the recovery rate of BDE-183 under the temperature program used for BDE-209 and similar recoveries were obtained between these homologue groups. The MS was operated under positive EI condition at electron energy of 70 eV. The EI-MS conditions followed the methods published previously [15]. The full-scan operating mode was used to identify target ions for each BDE standard, and we then use these identified target ions to qualify BDEs in samples. Ion masses (corresponding to peaks at either M^+ or $\text{M}-2\text{Br}^+$) of 325.9, 485.7 and 483.7 were selected for BDE-47, masses of 405.8, 403.8 and 563.6 were selected for BDEs – 99 and 100, masses of 641.5, 483.7, 485.7 and 643.5 were selected for BDEs – 153 and 154; masses of 561.6, 563.6 and 721.5 were selected for BDE-183, and masses of 799.4 and 801.4 were selected for BDE-209.

Total organic carbon (TOC) was determined for water samples using a TOC analyzer (TOC-VcPH, SHIMADZU Corp., Kyoto, Japan) and expressed as ppm. TOC in sediment and soil was measured by a gravimetric method and expressed as a percentage [6,12]. Particle size (μm) of sediment and soil was determined with a laser particle size analyzer (Dandong Better Size Scientific Ltd., Dandong, China), and the median diameter (D50) was recorded for each sample.

2.4. Quality control and quality assurance (QA/QC)

To monitor methodological analyte losses, a fraction of samples from different matrices were spiked with surrogate standards of PCB 138 and 209, and the recoveries for PCB 138 ranged from 63.2% to 145.3% with an average of 96.7% ($n=41$), and for PCB 209

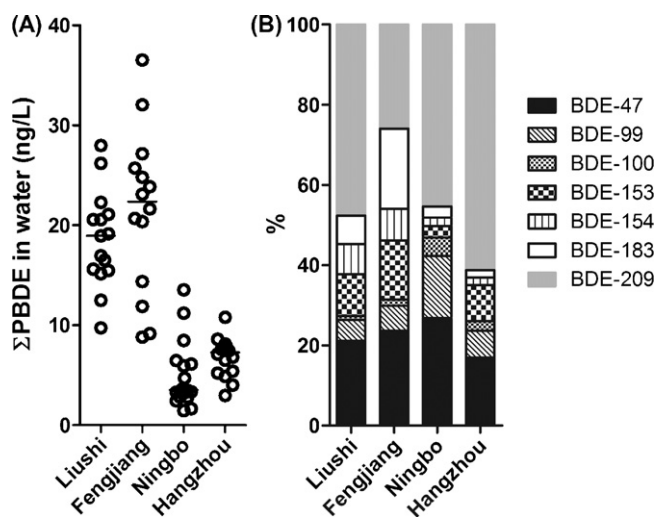


Fig. 2. Total PBDE concentrations (A) and the average composition of individual BDEs (B) in water samples from different sampling areas. Lines in the scatter plot represent the median values.

ranged from 52.8% to 129.5% with an average of 79.1% ($n=41$). For instrumental quality control, samples were spiked with a standard solution of PBDE containing 7 native congeners. Recoveries of these congeners were in the range of 75–110% for water samples, 94–120% for sediment samples and 80–110% for biological samples. The analyzed concentrations were not corrected for the recovery rates. Quantification was performed using the external calibration method based on a five-point calibration curve for individual component. The correlation coefficients of the calibration curves were greater than 0.999. The limit of detection (LOD) was defined as a signal/noise ratio (S/N) of 3. The method LODs on average ranged from 0.01 to 0.22 ng/g for BDEs - 47, 99, 100, 153, 154 and 183, and from 0.85 to 1.98 ng/g for BDE-209. A procedure blank was included for each set of 8 field samples, and all blank controls contained no detectable amounts of the target substances. Samples with concentrations below the detection limits were computed as zero. PBDE concentrations were expressed as ng/L in water samples, and ng/g of dry weight in sediment and soil, and ng/g of wet weight in biological samples.

3. Results and discussion

3.1. PBDEs in water samples

In water samples collected from the total of 64 sites, except for F8, all 7 BDEs were detected in every site (Figs. A1–A4), suggesting widespread PBDE contamination in waterway systems in Zhejiang Province. The median ΣPBDE concentration was highest in Fengjiang (22.4 ng/L), followed in descending order by Liushi (18.9 ng/L), Hangzhou (7.3 ng/L), and Ningbo (3.5 ng/L) (Fig. 2A). These data showed that the PBDE levels in water samples from the two urban areas (Hangzhou and Ningbo) are relatively low compared to those from the two rural areas that are characterized by the low-voltage electrical industry (Liushi) and e-waste recycling (Fengjiang). This finding indicates that the industries from the two rural areas could be responsible for the PBDE contamination in local environment. Previous studies have demonstrated that e-waste recycling is a significant source of PBDE contamination [16]. Moreover, the congener distribution pattern also differed among these four areas (Fig. 2B). Water samples from Liushi were dominated by BDEs - 209, 47, and 153 with BDE-209 accounting for approximately 50% of ΣPBDEs. BDEs - 209, 183, 153, and 47

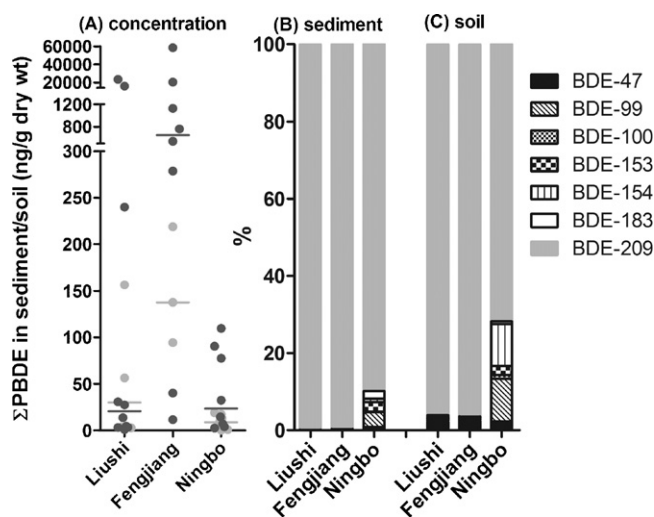


Fig. 3. Total PBDE concentrations (A) in sediment (dark gray circles) and soil (light gray circles); the average composition of individual BDEs in sediment (B) and soil samples (C) from different sampling areas. Lines in the scatter plot represent the median values.

all contributed similarly to ΣPBDEs in samples from Fengjiang. In Ningbo, BDEs - 209, 47, and 99 constituted the majority of ΣPBDE, while PBDE composition in Hangzhou was similar to that of Liushi. Correlations between PBDE concentration and TOC were not significant with the exception of BDE 47 ($r=0.30$, $P=0.015$), which could be due to their low water solubility in general.

3.2. PBDEs in sediment and soil samples

In sediment and soil samples, the highest ΣPBDE concentration in Liushi and Fengjiang was several hundred magnitudes higher than the highest ΣPBDE value in Ningbo (Fig. 3A), suggesting a historical PBDE contamination from both Liushi and Fengjiang. This is explainable because both the low-voltage electrical industry and the e-waste recycling have been established for more than two decades in these two areas. In comparison to water samples, most sediment and soil samples were predominated by BDE-209 (Fig. 3B and C), which is similar to studies in other regions of China [7,17,18]. All these could be linked to the predominant use of the deca-BDE mixture (e.g., Bromkal 82-ODE) in China [19]. The highest level of BDE-209 in our study was found at F1 (58,445 ng/g dry wt), which is very close to the e-waste dismantling industrial park. This value is comparable to the BDE-209 concentration found in the floor dust samples (5560–80,600 ng/g dw) collected from an e-waste dismantling workshop in this area [16], but is much higher than levels in other freshwater sediment such as the Spanish River Vero (12,500 ng/g dry wt [13]), and the Lake Maggiore basin in Europe (15 ng/g dry wt [20]), which could be due to the small dilution factor in river systems at Liushi or Fengjiang.

Correlations between PBDE concentration and TOC showed that all PBDE congeners except for BDE-47 and BDE-154 correlated significantly with TOC content. This explains the hydrophobic nature of PBDEs, which render them to strongly adsorb to particles with high organic carbon content [1,17]. Correlation between PBDE and particle size showed that only BDE-153 correlated significantly ($r=0.37$; $P=0.02$) with particle size. This indicates that BDE-153 was not preferentially enriched in small particles.

3.3. PBDEs in biological samples

The ΣPBDE concentrations in plant and cyanobacteria from Liushi were 10-fold higher than those from Ningbo (Table 2),

Table 2
Total PBDE concentrations from biological samples.

Area	\sum PBDE	Biological samples (ng/g wet weight)				
		Plant-leave	Plant-stem	Alga	Snail	Crayfish
Liushi	Range	22.0–222.7	10.8–84.3	22.4–58.5	6.7–78.1	21.0–105.2
	Median	75.1	31.3	31.3	26.0	38.2
Ningbo	Range	1.4–25.6	1.3–24.2	0.4–3.5	–	–
	Median	4.0	3.9	2.9	–	–

suggesting significant PBDE contribution from the low-voltage electrical industry. Analysis with plants showed higher \sum PBDE concentration in leaves than in the stems (Table 2), indicating significant contribution of PBDE contamination through air deposition [7]. The level of PBDEs in plants is expected to be considerably lower than in other solid or semi-solid matrices due to the larger water content and low lipid content. However, our study revealed similar PBDE contents in plant samples as those of aquatic animals of apple snail and crayfish in the same area (Liushi) (Fig. A1). The PBDE concentrations in plants of this study were lower than the leave samples of *Cinnamomum camphora* (0.46–399.93 ng/g dry wt) from Taizhou, Zhejiang [21] and grass samples (70–5900 ng/g ww) from Laizhou Bay, China [7].

Similar to sediment and soil samples, BDE-209 was the dominant congener in all biological samples in this study (Figs. A1 and A3). This differs from studies in other regions of the world, where BDE-209 was less abundant in biological samples but more prevalent in sediment and soil samples than other low-brominated congeners such as BDE-47 and BDE-99 [1,22,23]. One of the possible reasons could be due to the massive production and usage of deca-BDE mixtures in China [24]. However, other studies in China involving biological samples such as the *C. camphora* leaves [21] and the aquatic birds of Chinese-pond heron and ruddy-breasted crane [25] found that BDE-47 was the dominant congener. Therefore, variations of the congener profile in biological samples observed in different studies might be related to different species (thus different habitats, diets, and metabolism) and different routes of PBDE absorption (e.g., atmospheric, aquatic or terrestrial sources) [25]. In addition, the analytical difficulty of BDE-209 and subsequent exclusion of its analysis in many

studies could also lead to a bias view of BDE-209 in the literature [26].

4. Conclusion

Our study showed elevated PBDE concentrations in water, sediment/soil, and biological samples collected from areas with low-voltage electrical manufactures or e-waste recycling activities in comparison to highly industrialized areas within the same geographical region in South China. PBDE content and composition in water samples varied from one area to another, while PBDE composition in sediment/soil and biological samples was predominated by BDE-209. The detection of PBDE in plants, alga, and edible tissues from apple snail and crayfish raises significant concerns of human health risk associated with PBDE exposure.

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Appendix A.

Figs. A1–A4.

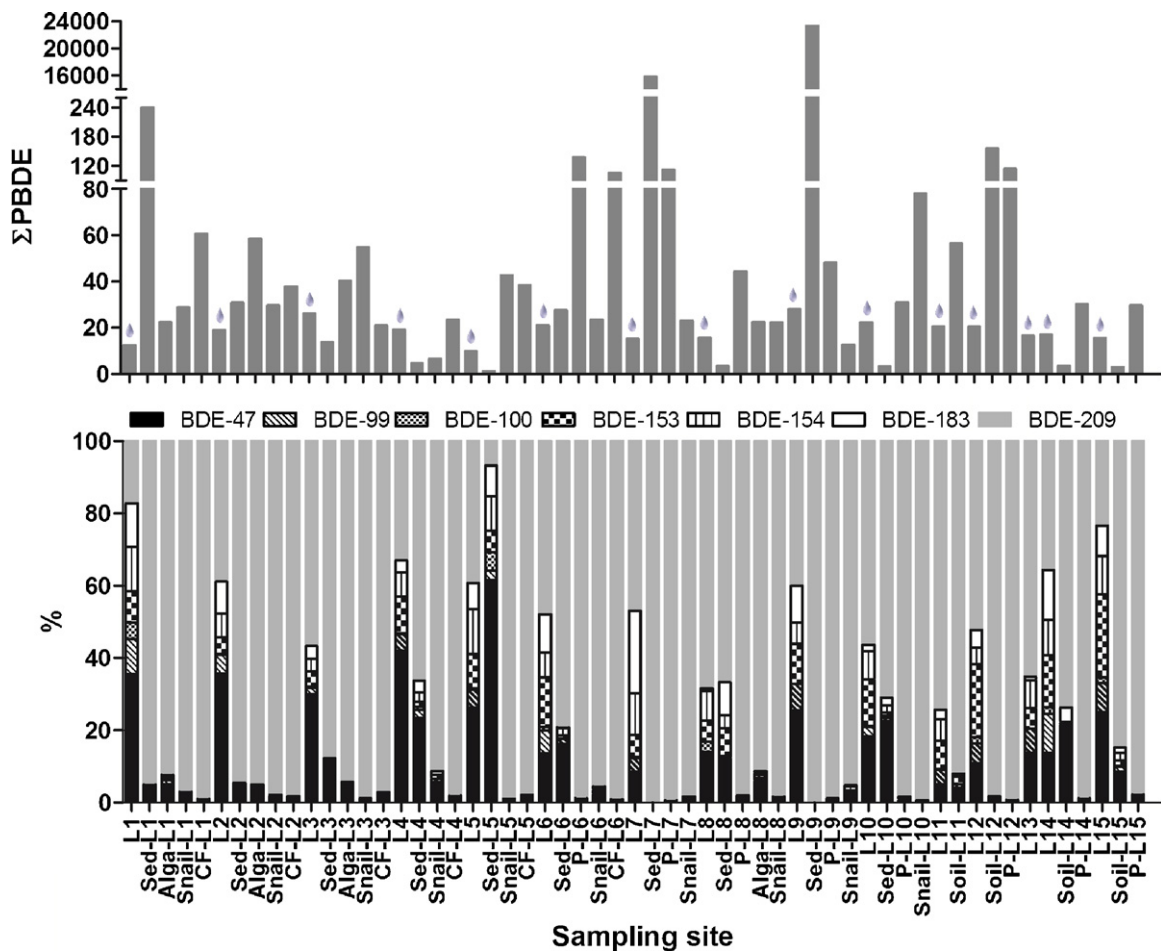


Fig. A1. Concentrations of total PBDE (Σ PBDE) and compositions (%) of individual BDEs in water samples (ng/L), sediment or soil (ng/g dry weight), plants, algae, apple snails and crayfish (ng/g wet weight) from Liushi Town (L1–L15). Site codes without prefix are water samples (♣). Sed: sediment; P: plant; CF: crayfish.

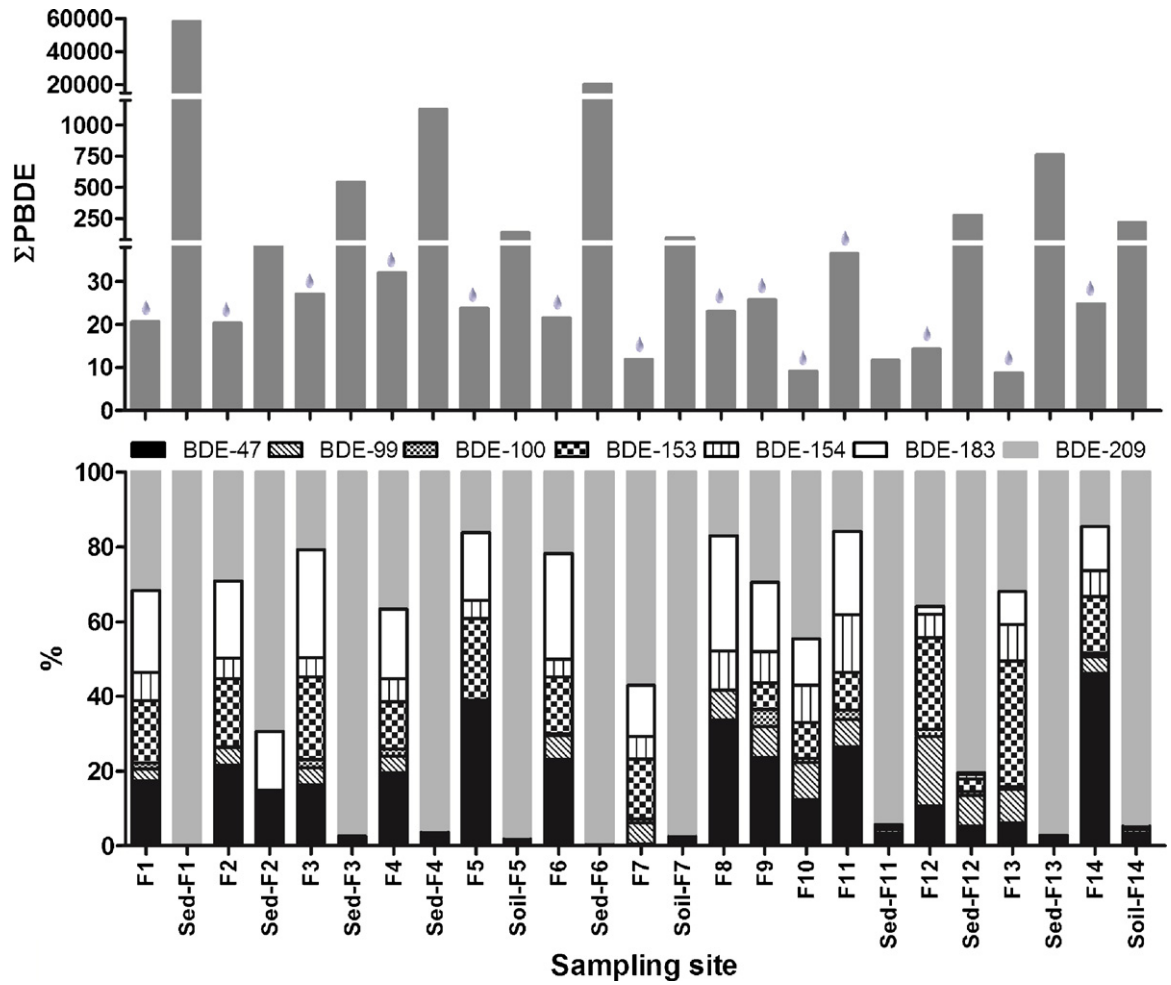


Fig. A2. Concentrations of total PBDE (\sum PBDE) and compositions (%) of individual BDEs in water samples (ng/L) and sediment or soil (ng/g dry weight) from Fengjiang Town (F1–F14). Site codes without prefix are water samples (♂). Sed: sediment.

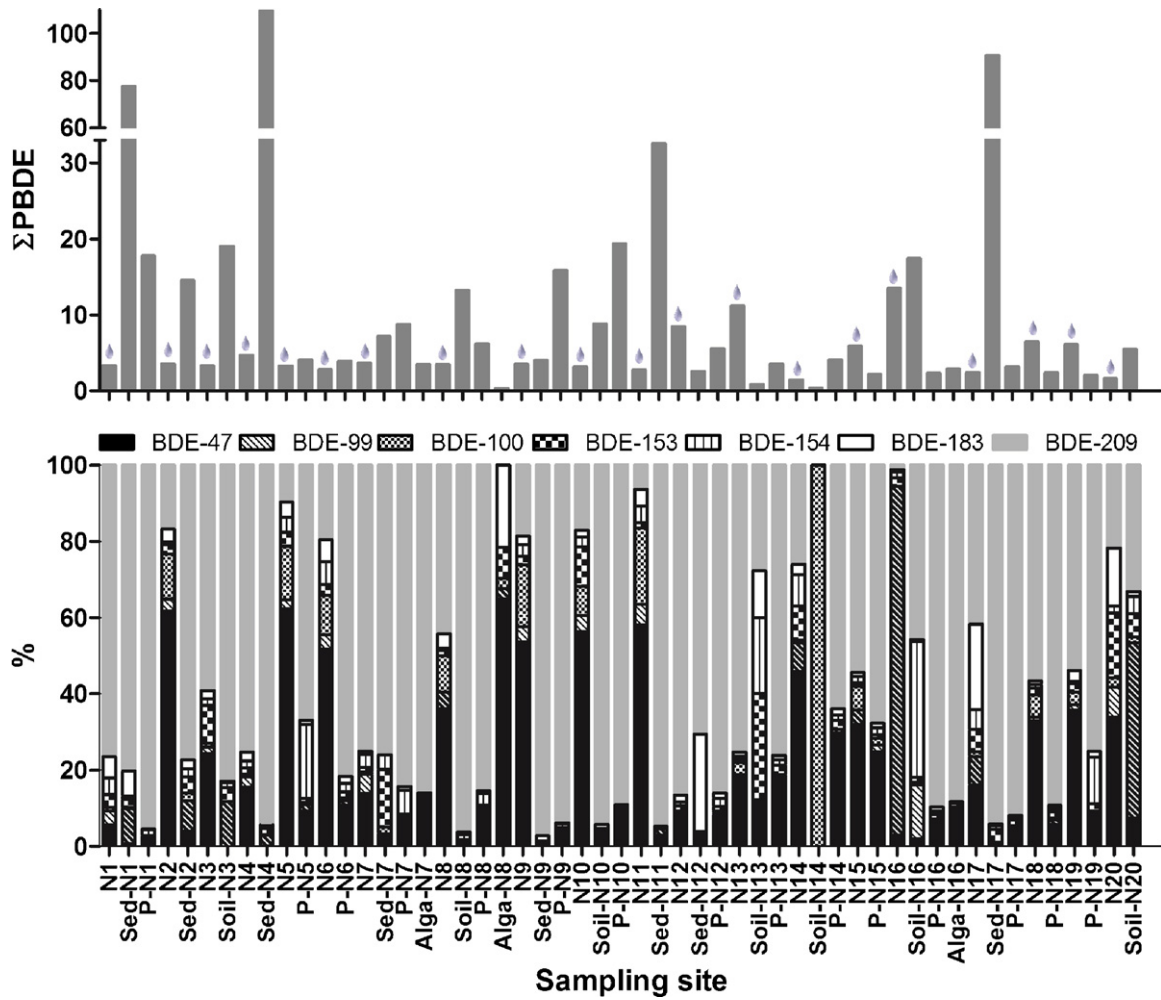


Fig. A3. Concentrations of total PBDE (Σ PBDE) and compositions (%) of individual BDEs in water samples (ng/L), sediment or soil (ng/g dry weight), and plants (ng/g wet weight) from Ningbo City (N1–N20). Site codes without prefix are water samples (♂). Sed: sediment and P: plant.

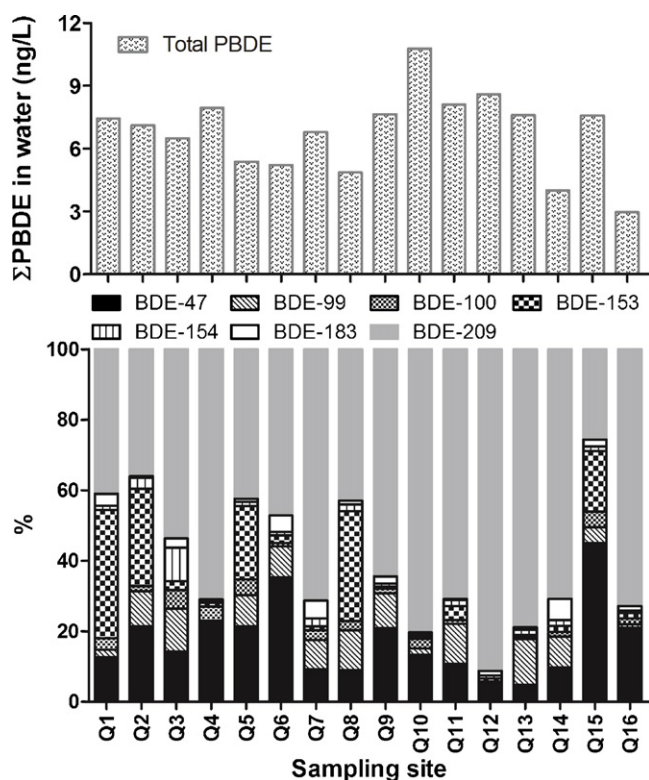


Fig. A4. Concentrations of total PBDE (\sum PBDE) and compositions (%) of individual BDEs in water samples from the Qiantang River (Q1–Q14) and the West Lake (Q15–Q16) in Hangzhou (Fig. 2).

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