

Determination of polybrominated diphenyl ethers in soils and sediment of Hanfeng Lake, Three Gorges

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As ubiquitous organic contaminants, polybrominated diphenyl ethers (PBDEs) were evaluated to explore the impacts of the water level fluctuating zone (WLFZ) on organic pollutant behavior and the retention mechanism of organic pollutants in the bank–WLFZ–water system of Hanfeng Lake in the Three Gorges region of China. The mean concentrations of total PBDEs were 103, 75.2, and 568 ng g⁻¹ dry wt for bank soils, WLFZ soils, and sediment samples, respectively. Except for sampling sites S1 and S2, the levels of PBDEs decreased in the order of sediment > WLFZ soil > bank soil, suggesting that PBDEs were transferred from bank soil to WLFZ soil and finally deposited in the sediment. Decabromodiphenyl ethers (deca-BDEs) were the predominant congener in the study area, comprising 93.8% to 98.3% of the total PBDEs. Greater photolytic degradation of deca-BDEs was suggested in bank soils based on a higher relative abundance of octa- and nona-BDEs than in WLFZ soils and sediment. This may have occurred because deca-BDEs in bank soils have relatively longer sunlight exposure than in WLFZ soils and sediment due to the annual alternation of water storage and drainage in the catchments of the Three Gorges Reservoir. More in-depth investigations of contaminants in bank–WLFZ–water systems are needed due to the large areas of WLFZ created by the construction of the Three Gorges Dam and their importance to the balance of aquatic ecosystems.

Keywords: Polybrominated diphenyl ethers, sediment, soil, water level fluctuating zone.

Introduction

The Three Gorges Dam (TGD) in the Yangtze River (the longest river in China and the third longest in the world) is the largest hydropower project in the world, with a width of 2335 m and a height of 185 m. The Three Gorges Reservoir (TGR) created by the TGD has been strictly regulated as a strategic freshwater resource by the government. However, the entire ecosystem structure and function of catchments of the TGR has been drastically altered, and a substantial area (up to 348.9 km²) comprised of a water level fluctuating zone (WLFZ) and riverine floodplains has formed in the area of the TGR since its completion in 2009. The WLFZ of Hanfeng Lake has an area of 45.2 km², making it the largest WLFZ in the area. This

WLFZ developed from the annual fluctuation of water level in the TGR. In recent years, the water quality of Hanfeng Lake has been severely threatened by increased fluxes of nutrients, metals, and organic contaminants originating from industrial, agricultural and residential activities (e.g., industrial production activities, waste water discharging, and littering) in the riparian land around the lake.

WLFZs are generally characterized by the interconnection of aquatic and terrestrial habitats. These areas act as buffers for flood water and filters of pollutants carried with river water and sediment from upstream. On a global scale, WLFZs occupy more than 2×10^6 km², or 1.3%, of the earth's land surface.^[1] Together with sediments in reservoirs and rivers, WLFZs have become an increasing environmental concern in recent years because they act as pollutant sinks as well as sources of organic and inorganic pollutants.^[2]

Several international efforts, such as the EU-funded AquaTerra project (2004–2009), have been launched. These studies have revealed that once organic compounds, such as persistent organic pollutants (POPs), are incorporated into the soil and sediment, their downward migration and degradation occur extremely slowly, with very long

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Received September 29, 2014.

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half-lives on the order of decades. However, flooding or periodical events such as freezing/thawing and wetting/drying cycles can greatly increase the release and downward migration of these POPs by physical re-mobilization. In addition, the alternating aerobic and anaerobic status in response to draining or inundation of WLFZ soil may increase the microbially-mediated degradation of some classes of POPs (e.g., polychlorinated biphenyls (PCBs)).^[2–5] As a result, it has become evident that WLFZ pollution severely impairs the integrity of freshwater and coastal ecosystems and poses serious threats to aquatic biota and food security.^[6,7]

As ubiquitous organic contaminants, polybrominated diphenyl ethers (PBDEs) have been reported in various environmental media, including air, sediment, soil, aquatic and terrestrial organisms, and the human body.^[8–10] Due to their environmental persistence, bioaccumulation, and potential toxicity to wildlife and humans, commercial penta- and octa-BDE formulations were banned in 2004 by the European Union and the United States.^[8,9] However, deca-BDE (BDE-209) has received increased interest in recent years because it remains the only brominated diphenyl ether flame retardant available and is therefore occurring in increasing environmental concentrations.^[11]

In China, deca-BDE is still commonly used, with more than 30,000 t being consumed in 2005, and its use has been increasing at a rate of approximately 8% per year.^[12] Because of their high hydrophobicity, PBDEs, especially deca-BDEs, tend to be absorbed by soil particulate matter in terrestrial environments or to accumulate in sediments once released into aquatic systems.^[13] In the WLFZ of the TGR, the anaerobic/aerobic cycles caused by water storage and drainage of the TGR may accelerate the degradation of deca-BDEs to lower brominated congeners (BDE 154, BDE 99, BDE 47, etc.),^[1,14] which have higher hydrophilicity and potential toxicity to aquatic organisms.^[11]

Therefore, a comprehensive survey on the occurrence and fate of PBDEs in different compartments of the bank–WLFZ–water systems is needed to ensure the safety of the water quality and the aquatic ecosystem. Some research

groups have documented the pollution status of POPs such as PCDD/Fs, PAHs, PCBs, and OCPs in areas of the TGR.^[15–19]

However, only one study reported a low contamination level of PBDEs (from nd to 502 pg g⁻¹ dry wt) in sediment from 28 sampling sites in the TGR region.^[20] In the present study, PBDEs were selected to evaluate the impacts of WLFZ on organic pollutant behavior in the bank–WLFZ–water system in Hanfeng Lake, Three Gorges. In addition, the concentration levels of PBDEs in the bank–WLFZ–water system are presented.

Materials and methods

Sample collection

Hanfeng Lake is an anthropogenic lake located in Kaixian County in the TGR area (Fig. 1). Due to the TGD, the water depth of Hanfeng Lake now rises to 175 m during the water storage period (May to September), and drops to 145 m during the water drainage period (October to April). In the present study, a total of 20 samples were collected from Hanfeng Lake in August 2011. Briefly, seven bank soil samples, six WLFZ soil samples, and seven sediment samples were included. Bank soil and WLFZ soil samples were collected using a stainless steel shovel, while sediment samples were obtained using a Van Veen stainless steel grab sampler. The samples were then put into previously prepared aluminum/polyethylene bags and transported to the laboratory on ice. All samples were freeze-dried, pre-cleaned, homogenized by sieving through a stainless steel 80-mesh sieve, and stored in brown glass bottles at –20°C until further analysis.

Sample extraction and cleanup

The detailed extraction and cleanup procedures applied to the PBDEs were described in our previous study.^[21] Briefly, each sediment and soil sample (10 g) was spiked

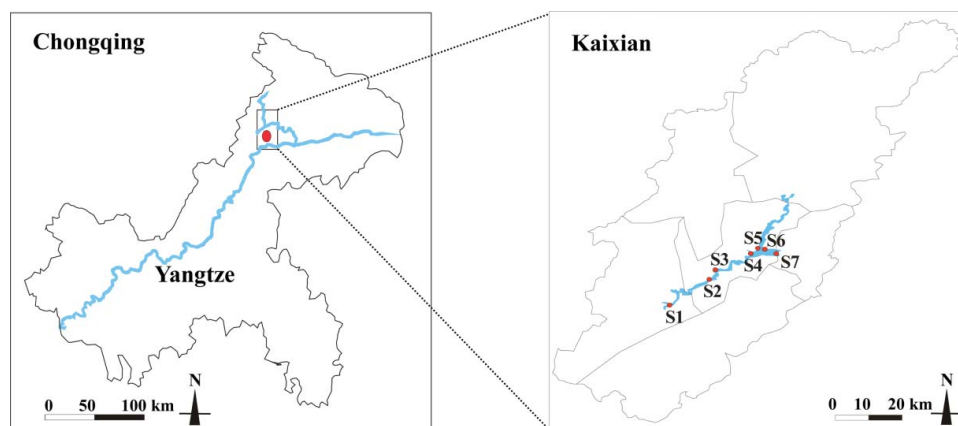


Fig. 1. Sampling sites at Hanfeng Lake, Three Gorges.

with PCB-209 and then Soxhlet extracted for 48 h with 200 mL acetone/hexane mixture (1:1, v/v). Activated copper granules were added to the extraction flasks during the extraction to remove elemental sulfur. The extracts were subsequently concentrated with a rotary evaporator and exchanged with hexane, after which they were cleaned and fractionated using a multilayer silica/alumina column.

The column was wet-loaded with 6 cm of aluminum, 2 cm of neutral silica gel, 5 cm of basic silica gel (3:1 silica gel: 1M NaOH, w/w), 2 cm of neutral silica gel, 8 cm of acidic silica gel (1:1 silica gel: sulfuric acid, w/w), and 2 cm of anhydrous Na₂SO₄, from the bottom to the top. The column was then eluted with 70 mL hexane/dichloromethane (1:1, v/v), after which the collected eluate was concentrated to near dryness under a gentle steam of nitrogen gas before being redissolved in 200 μ L hexane. Finally, 10 μ L of 400 μ g L⁻¹ ¹³C-labeled PCB-208 were added to each sample as an internal standard before instrumental analysis.

Instrumental analysis

All samples were analyzed with an Agilent 7890A gas chromatograph (GC) coupled to an Agilent 5975C mass spectrometer (MS) (Agilent Technologies, Palo Alto, CA, USA) in negative chemical ionization and selective ion monitor mode. The parameters of GC-MS for the PBDEs are listed below.

For the lower-brominated congeners (BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190), a 30 m HP-5 MS capillary column (250 μ m i.d., 0.25 μ m film thickness, J&W Scientific, Folsom, CA, USA) was used for qualitative analysis. The temperature program for the GC was as follows: initial temperature of 110°C (held for 1 min), followed by an increase to 180°C at 8°C min⁻¹ (held for 1 min), then to 240°C at 2°C min⁻¹ (held for 5 min), 280°C at 2°C min⁻¹ (held for 15 min), and finally 300°C at 10°C min⁻¹ (held for 12 min). The temperatures of the injection port, transfer line, ion source, and quadrupole rods were 280°C, 300°C, 200°C, and 150°C, respectively. The ions monitored were m/z 79 and 81 for the PBDEs and m/z 473.7 and 475.7 for the ¹³C-labeled PCB-208.

For the higher-brominated congeners (BDE-196, 197, 203, 206, 207, 208, 209), a 12.5 m DB-5-HT MS capillary column (250 μ m i.d., 0.10 μ m film thickness, J&W Scientific, Folsom, CA, USA) was used. The temperature program for the GC was as follows: initial temperature of 110°C (held for 5 min), followed by an increase to 200°C at 20°C min⁻¹ (held for 4.5 min), then to 300°C at 7.5°C/min (held for 16 min). The temperatures of the injection port, transfer line, ion source, and quadrupole rods were 260°C, 280°C, 250°C, and 150°C, respectively. The ions monitored were m/z 79 and 81 for the PBDEs, m/z 486.7 and 488.7 for BDE-209, m/z 498 and 500 for PCB-209, and m/z 473.7 and 475.7 for the ¹³C-labeled PCB-208.

Quality assurance / quality control (QA/QC)

For each batch of 12 samples, one procedural blank sample, one standard-spiked blank sample, one standard-spiked matrix sample, and one duplicate sample was analyzed for quality control. The surrogate recovery of PCB-209 in the samples ranged from 90.3% to 108.4%, while the PBDE recovery from the spiked matrices ranged from 83.4% to 98.7%. The limits of quantification, which were defined by a signal-to-noise ratio (S/N) = 10, ranged from 0.013 to 0.056 ng g⁻¹ dry wt for tri- to nona-BDEs and 0.085 ng g⁻¹ dry wt for BDE-209. Caution was used in the analysis of BDE-209 because of its sensitivity to high temperatures and higher susceptibility to degradation in the GC system. No detectable levels of PBDEs, with the exception of a small amount of BDE-209, were found in any of the control samples. No reported concentrations were blank or recovery corrected. The relative standard differences (RSD) of individual PBDE congeners between duplicate samples (n = 3) were all less than 10%.

Results and discussion

Concentration and spatial distribution of PBDEs

Twenty target PBDE congeners, BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 196, 197, 203, 206, 207, 208, and 209, were measured in all 20 of the samples collected from Hanfeng Lake in the Three Gorges region. BDE-17, BDE-85, and BDE-190 were not detected in any samples and are not included in further discussion. We defined \sum PBDEs as the total concentration of 16 congeners (except BDE-209). Table 1 summarizes the mean values and ranges of \sum PBDEs and BDE-209. PBDEs were detected in all 20 samples, indicating that they were ubiquitous contaminants in the Hanfeng Lake area.

The concentrations of \sum PBDEs ranged from 0.57 to 50.1 ng g⁻¹ dry wt with a mean value of 5.91 ng g⁻¹ dry wt, while the concentrations of BDE-209 ranged from 9.19 to 2261 ng g⁻¹ dry wt with a mean value of 252 ng g⁻¹ dry wt. Among the 17 detected PBDE congeners, BDE-209 was predominant, comprising 93.8% to 98.3% of the total PBDEs. In addition, octa- and nona-congeners, including BDE-196, 197, 203, 206, 207, and 208, accounted for 1.69% to 5.76% of the total PBDEs, whereas the lower-brominated congeners (tri- to hepta-BDEs) were detected in very small proportions, accounting for 0.03% to 1.25% of the total in all sediment and soil samples. These findings suggest that deca-BDEs were the predominant PBDE formulations used in the studied area. This observation was consistent with the fact that deca-BDEs account for 72% of the total PBDE commercial demand in the global market.^[21,22]

The mean values of BDE-209 were 101, 73.1 and 555 ng g⁻¹ dry wt for bank soils, WLFZ soils, and sediment

Table 1. The concentrations of polybrominated diphenyl ethers (PBDEs) (mean and range) in the soil and sediment samples from Hanfeng Lake, Three Gorges (ng g⁻¹, dry wt).

Congener	Bank soil (n ^a = 7)	Fluctuating zone soil (n = 6)	Surface sediment (n = 7)
BDE-28	0.02 (0.01–0.03)	0.04 (0.01–0.07)	0.10 (0.03–0.10)
BDE-47	0.02 (0.01–0.05)	0.05 (0.02–0.10)	0.20 (0.06–0.91)
BDE-66	0.00 (<LOD ^c -0.00)	0.01 (<LOD-0.01)	0.00 (<LOD-0.00)
BDE-71	0.01 (<LOD-0.01)	0.01 (<LOD-0.01)	ND ^b
BDE-99	0.01 (0.00–0.01)	0.02 (0.01–0.04)	0.07 (0.02–0.34)
BDE-100	ND	0.00 (<LOD-0.00)	0.01 (<LOD-0.03)
BDE-138	ND	0.00 (<LOD-0.00)	0.01 (0.00–0.02)
BDE-153	0.02 (0.01–0.02)	0.02 (0.01–0.05)	0.165 (0.03–0.79)
BDE-154	0.00 (<LOD-0.00)	0.01 (<LOD-0.02)	0.06 (0.00–0.30)
BDE-183	0.01 (0.01–0.03)	0.04 (0.02–0.05)	0.14 (0.03–0.64)
BDE-196	0.03 (0.01–0.07)	0.03 (0.02–0.04)	0.14 (0.03–0.48)
BDE-197	0.03 (0.01–0.06)	0.03 (0.01–0.04)	0.12 (0.03–0.39)
BDE-203	0.03 (0.01–0.07)	0.03 (0.02–0.03)	0.12 (0.04–0.39)
BDE-206	0.76 (0.15–2.03)	0.66 (0.33–0.87)	4.42 (1.04–18.2)
BDE-207	0.89 (0.19–2.47)	0.78 (0.48–0.98)	4.57 (1.10–17.4)
BDE-208	0.52 (0.12–1.19)	0.46 (0.25–0.60)	2.59 (0.68–9.75)
∑PBDEs ^d	2.34 (0.57–6.13)	2.14 (1.24–2.73)	12.7 (3.15–50.1)
BDE-209	101 (9.19–350)	73.1 (20.9–103)	555 (112–2261)

^an means sample number;

^bND means not detected;

^cLOD means limit of detection;

^d∑PBDEs = sum of the BDE-28, 47, 66, 71, 99, 100, 138, 153, 154, 183, 196, 197, 203, 206, 207 and 208.

samples, respectively, while the mean values of ∑PBDEs were 2.34, 2.14 and 12.7 ng g⁻¹ dry wt for bank soils, WLFZ soils, and sediment samples, respectively (Table 1). The mean levels of PBDEs in the sediment were much higher than those in the bank soils and WLFZ soils. These results are not surprising since sediment is generally thought to be the ultimate sink of all contaminants in the continuous bank soil–WLFZ soil–sediment system. During the water storage period, the bank soil and WLFZ soil in the area are easily eroded by rain and rising water; therefore, most PBDEs absorbed in the soils are likely gradually transferred to and accumulated in the sediment.

As shown in Figure 2, the levels of PBDEs decreased in the order of sediment > WLFZ soil > bank soil for sampling sites S3 to S7. This concentration pattern seems to confirm the transfer of PBDEs from the bank soil to the WLFZ soil and their final deposition in the sediment, as previously explained. However, a different trend was observed at sampling sites S1 and S2, with remarkably higher levels of PBDEs in the bank soil than the WLFZ soil. This difference may be explained by the presence of domestic and industrial waste containing PBDEs piled on the banks near S1 and S2 during the sampling period.

Notably, the highest PBDE concentration (2311 ng g⁻¹ dry wt) was found at sampling site S1, which was located upstream of the Hanfeng Lake. Similarly, a higher concentration of BDE-209 was also found in sediment samples from the River Calder in the UK (3190 ng g⁻¹)^[23] and the Viskan River in Sweden

(7100 ng g⁻¹)^[24] both of which were collected downstream of a sewage treatment plant. In addition, relatively higher PBDE levels were observed at sampling site S6. These results may be explained by the proximity of S6 to the former town of Kaixian, which had the largest population density and industrial activity among the seven sampling sites prior to completion of the TGD and therefore was the source of more domestic and industrial waste materials containing PBDEs than other areas during inundation.

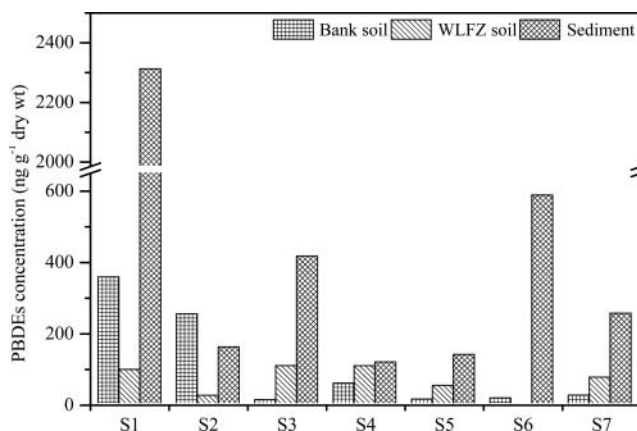


Fig. 2. Polybrominated diphenyl ethers (PBDEs) concentrations in all three sample types in the study areas. WLFZ = water level fluctuating zone; WLFZ soil sample of S6 is not available.

Comparison with other regions around the world

Although comparisons between different studies are difficult because of inconsistencies in the congeners analyzed, we conducted a simple comparison of concentrations observed in the present study with those reported for other areas around the world. Table 2 summarizes the mean concentrations of PBDEs in soils and sediments from different regions around the world. As shown in Table 2, the mean PBDE concentrations (12.7 and 555 ng g⁻¹ dry wt for \sum PBDEs and BDE-209, respectively) in the sediment samples in the present study were one order of magnitude lower than those observed in sediment at e-waste recycling sites in East China (191 and 1839 ng g⁻¹ dry wt, respectively),^[25] while they were comparable to those reported in sediment from the Scheldt estuary (19 and 713 ng g⁻¹ dry wt, respectively) in the Netherlands,^[26] Lake Michigan (2.6 and 315 ng g⁻¹ dry wt, respectively) in the United States,^[27] the Pearl River Delta (9.9 and 465 ng g⁻¹ dry wt, respectively) in China,^[28] and Osaka Bay (16.7 and 352 ng g⁻¹ dry wt, respectively) in Japan.^[29]

Additionally, the values were somewhat higher than those in sediment from Europe, North America, and

Asia.^[27,30–32] In comparison, the concentrations of \sum PBDEs and BDE-209 (2.25 and 88.1 ng g⁻¹ dry wt, respectively) in the bank soils and WLFZ soils of the present study were two orders of magnitude lower than those of soils at e-waste recycling sites in South China (580–1150 and 1539–2258 ng g⁻¹ dry wt, respectively),^[21,33] while they fell within the ranges reported for soils from Spain,^[34] the United States,^[22] China,^[21] and Turkey,^[35] and were somewhat higher than those of farmland soils or remote soils from a number of different countries.^[12,36–38]

The retention and fate of PBDEs in the soils and sediment of Hanfeng Lake

Figure 3 shows the congener profiles of higher-brominated BDEs (octa- to deca-) in the present study. As shown in Figure 3, the relative abundance of the higher-brominated BDEs in the bank soils, WLFZ soils and sediment were somewhat different. Specifically, an apparently higher relative abundance of octa- and nona-congeners was observed in bank soils than in the WLFZ soils and sediment, except for at S1 and S2.

Table 2. Mean concentrations of polybrominated diphenyl ethers (PBDEs) in soils and sediments from different regions around the world (ng g⁻¹, dry wt).

Location	Country	\sum PBDEs	BDE209	Reference
Sediment				
Rivers (n = 8)	Swedish	8–50 ^a	68–7100	[24]
Scheldt Estuary (n = 3)	Netherland	19	713	[26]
Rivers and Estuary (n = 29)	UK	1.3–1270	<0.6–3190	[23]
Coastal (n = 13)	Spain	0.24	23.4	[30]
Erie Lake (n = 1)	USA	1.1	39	[27]
Michigan Lake (n = 1)	USA	2.6	315	[27]
Niagara River (n = 11)	USA	18.5	NA ^b	[39]
Coastal (n = 25)	Korea	0.31	27.4	[32]
Osaka Bay (n = 6)	Japan	16.7	352	[29]
Yangtze River Delta (n = 32)	China	0.15	13.4	[31]
E-waste recycling site (n = 4)	China	191	1839	[25]
Pearl River Delta (n = 66)	China	9.9	465	[28]
Hanfeng Lake (n = 7)	China	12.7	555	The present study
Soil				
Woodland/Grassland (n = 54)	UK/Norway	0.97	NA	[36]
Sludge applied sites (n = 7)	Spain	13.8	334	[34]
Industrial areas sites (n = 8)	Turkey	1.73	26.4	[35]
Industrial areas sites (n = 3)	USA	29.9	NA	[22]
Floodplain soils (n = 26)	USA	1.61	5.28	[37]
Chongming Island (n = 9)	China	0.76	12	[38]
Farmland sites (n = 33)	China	1.02	13.8	[12]
Industrial areas sites (n = 32)	China	3.96	41.5	[21]
E-waste site soils (n = 10)	China	580	2258	[21]
E-waste road soils (n = 29)	China	1150	1539	[33]
Hanfeng Lake (n = 13)	China	2.25	88.1	The present study

^aRange of concentration (mean value is not given); ^bNA means not available.

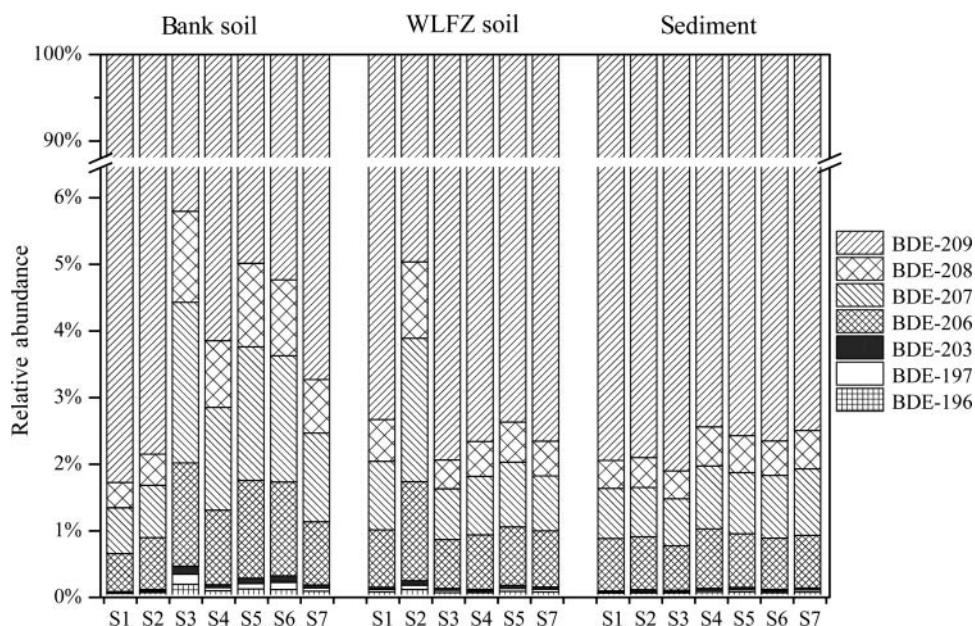


Fig. 3. The relative abundance of higher-brominated polybrominated diphenyl ethers (PBDEs) congeners. WLFZ=water level fluctuating zone; WLFZ soil sample of S6 is not available.

The photolytic degradation of deca-BDEs was believed to be responsible for these profile differences because deca-BDEs are photolytically labile and can be debrominated by exposure to natural sunlight. Bank soils are subjected to relatively longer periods of sunlight exposure than WLFZ soils and sediment due to the alternations in water storage and drainage in the catchments of the TGR. Watanabe and Tatsukawa^[40] found that DecaBDE was debrominated to lower brominated BDEs, while Eriksson et al.^[41] found that PBDEs with different degrees of bromination had different photolytic stability. Soderström et al.^[42] also reported that nona- and octa-BDEs readily formed in sediment and soil when technical deca-BDEs were added and exposed to either artificial UV light or natural sunlight conditions.

The continuous production of lower-brominated BDEs, such as BDEs 154 and 183, has also been observed. Although traces of octa- and nona-BDEs have been detected in deca-BDE technical products collected from European, North American, and Chinese markets,^[33,43] the distinct profiles and relative abundances of octa- and nona-congeners in soil and sediment samples collected in the present study suggest that the occurrence of these compounds in the present study could not be entirely explained by the technical mixtures. Nevertheless, it should be noted that the relative abundance of octa- and nona-congeners in bank soils from S1 and S2 were much lower than those of other bank soils. A possible explanation for this is that there was fresh input of deca-BDEs from domestic and industrial waste containing PBDEs piled on the bank before sampling was conducted. The remarkably higher concentration of PBDEs at these two sites (Fig. 2) also supported this assumption.

The congener profiles of the higher-brominated BDEs were generally similar for the WLFZ soils and the sediment samples. This similarity may have been associated with changes in hydrologic conditions and retention capacity for organic contaminants in the Hanfeng Lake area due to completion of the TGD. For the WLFZ soil, the wetting/drying cycles associated with the water storage and drainage of the TGR may considerably increase the release and downward migration of PBDEs adhering to soil particles, while also introducing high amounts of dissolved and particulate organic and inorganic matter.^[1]

Conversely, the rising water level of Hanfeng Lake will remobilize large amounts of sediment and soil particles and increase the mixing of lake water, sediments, and WLFZ soil, especially during the water storage period of the TGR.^[44] Notably, we also observed a remarkably higher relative abundance of octa- and nona-congeners in the WLFZ soil sample collected from S2. It is difficult to explain this specific observation because microbial mediated transformation processes and some abiotic transformation process, such as reductive debromination of BDE-209 catalyzed by zerovalent iron, can also lead to the formation of less brominated PBDEs.^[11,45]

Conclusion

The water level fluctuating zone acts as a buffer belt of interconnected aquatic and terrestrial habitats and plays an important role in the transfer of various contaminants between aquatic and terrestrial systems. The present study investigated the occurrence of PBDEs in bank soils,

WLFZ soils, and sediment from areas of Hanfeng Lake. The results demonstrated that the compounds are ubiquitous contaminants in the study area, and that deca-BDEs were the predominant formulations used in the region. The levels of PBDEs decreased in the order of sediment > WLFZ soil > bank soil, suggesting the transfer of PBDEs from bank soil to WLFZ soil and their final deposition in sediment. However, the variety of congener profiles may also reflect the complex interactions that occur as a result of hydrology, environmental transformation and pollutant levels in the dynamic bank–WLFZ–water systems. A multiyear survey and large scale spatial analysis of PBDE are needed to further elucidate the retention and fate of PBDEs in the bank–WLFZ–water system in Hanfeng Lake, Three Gorges.

Funding

This study was financially supported by the State Key Program of the National Natural Science Foundation of China (41130752) and the National Natural Science Funds for Distinguished Young Scholars (41225013).

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