



## Polybrominated diphenyl ethers and decabromodiphenylethane in sediments from twelve lakes in China

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### ABSTRACT

Sediment cores from 12 Chinese lakes were analyzed to investigate the historical inputs of polybrominated diphenyl ethers (PBDEs) and decabromodiphenylethane (DBDPE) during the past few decades. Concentrations of  $\Sigma$ PBDE<sub>1</sub> (sum of tri- to hepta-BDEs),  $\Sigma$ PBDE<sub>2</sub> (sum of nona- to deca-BDEs) and DBDPE in the surface sediments were 0.02–0.29 ng g<sup>-1</sup>, 0.46–46.6 ng g<sup>-1</sup> and 1.02–3.64 ng g<sup>-1</sup>, respectively. The temporal trends of PBDEs and DBDPE followed a general increase from the bottom to the surface. The calculated fluxes for  $\Sigma$ PBDE<sub>1</sub>,  $\Sigma$ PBDE<sub>2</sub> and DBDPE were 0.001–0.09, 0.03–4.24, and 0.05–0.31 ng cm<sup>-2</sup> yr<sup>-1</sup>, and the inventories were 0.09–7.86, 0.91–461, and 3.83–24.6 ng cm<sup>-2</sup>, respectively. The urbanization and industrialization are highly related to the contamination of PBDEs and DBDPE in sediments. The DBDPE input in recent years was still lower than  $\Sigma$ PBDE<sub>2</sub> but the temporal trends indicated that the contamination would increase with the increasing usage of DBDPE in the future.

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### 1. Introduction

Since first being detected in soil and sludge in the United States in 1979 (DeCarlo, 1979), polybrominated diphenyl ethers (PBDEs) have become a great concern because of their ubiquitous presence and persistence in the environment and the mounting evidence of their adverse health effects for wildlife and humans (Darnerud et al., 2001; McDonald, 2002). Because of their strong hydrophobicity (Song et al., 2004), PBDEs can accumulate in aquatic sediments after entering the water column via direct and indirect urban domestic/industrial sewage discharges and atmospheric deposition. The concentrations and distribution of PBDEs in sediments have been reported in sediments in North America, Europe and some eastern Asian countries (Song et al., 2004, 2005a, 2005b; Allchin et al., 1999; Eljarrat et al., 2004, 2005; Watanabe et al., 1995).

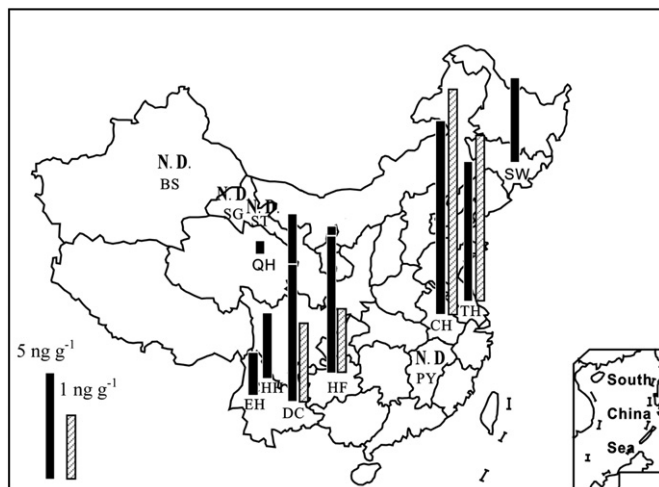
In China, the estimated annual production of the predominant commercial deca-BDE mixture increased from 10,000 to about 30,000 metric tons between 2000 and 2005 (Zhou, 2006). The legal and illegal import of E-wastes from overseas in recent years

(Brigden et al., 2005) is also believed to have aggravated PBDEs contamination and their effects on ecosystems and humans. The few published reports of PBDEs in sediment samples from China (Mai et al., 2005; Chen et al., 2007; Shi et al., 2009; Zhang et al., 2009) mainly involve locations in the Pearl River Delta (PRD) region. Contributed by being the largest manufacturing base for electronic and telecommunication equipment in China, up to 7340 and 94.7 ng g<sup>-1</sup> dry weight (dw) for BDE-209 and tri- to hepta-BDEs, respectively, were detected in this region. For other part of China, the median values of 2.29 and 0.16 ng g<sup>-1</sup> dw were reported for BDE-209 and tri- to hepta-BDEs in Bo Sea (Wang et al., 2009), 2.33 and 102.6 ng g<sup>-1</sup> dw in Fuhe River, and 0.78 and 10.4 ng g<sup>-1</sup> dw in Baiyangdian Lake (Hu et al., 2010).

Decabromodiphenylethane (DBDPE) was introduced in the early 1990s (Kierkegaard et al., 2004). It has the same applications as an additive flame retardant in different types of plastics and textiles as does BDE-209. With increasing regulation and phasing-out of commercial usage of PBDEs, DBDPE will soon be the major brominated flame retardant in production and use. Despite limited information about its environmental presence, the detection of DBDPE in various matrices, such as sewage sludge (Kierkegaard et al., 2004; Ricklund et al., 2008), house dust (Stapleton et al., 2008), air (Venier and Hites, 2008), and in the biota (Shi et al., 2009) has alerted us to a potential environmental pollutant. In China, DBDPE is reported to

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**Fig. 1.** Map of the 12 sampling lakes showing the detected surface concentrations of PBDEs (Black) and DBDPE (Shadow) (QH: Lake Qinghai; BS: Lake Bosten; SG: Lake Sagan; ST: Lake Shuangta; EH: Lake Erhai; CHH: Lake Chenghai; DC: Lake Dianchi; HF: Lake Hongfeng; PY: Lake Poyang; CH: Lake Chaohu; TH: Lake Taihu; and SW: Lake Sihailongwan maar).

have been produced and used in large amounts. In 2006, production of BDE-209 was 15,000 tons  $\text{yr}^{-1}$ , while production of DBDPE reached 11,000 tons  $\text{yr}^{-1}$  (Hu et al., 2010). Recently, DBDPE concentrations as high as 19–430  $\text{ng g}^{-1}$  dw were detected in surface sediments from the PRD region, south China (Zhang et al., 2009).

Lakes are very important water body; however, there have been few studies of the distribution and deposition of PBDEs and DBDPE in the sediments from Chinese lakes (Liu et al., 2009; Hu et al., 2010). This study was carried out to estimate the historical inputs of PBDEs and DBDPE into Chinese lakes. With this aim, sediment cores were collected from 12 lakes selected from five main lake regions of China (Fig. 1 and Table 1): Lake Sihailongwan maar from the northeastern lake region; Lakes Chaohu, Poyang and Taihu from the eastern lake region; Lake Qinghai from the Tibetan plateau lake region (Northwestern China); Lakes Bosten, Sagan and Shuangta from the Mengxin lake region (Northwestern China), and Lakes Chenghai, Dianchi, Erhai and Hongfeng from the Yungui plateau lake region (Southwestern China). The 12 lakes include the three major state lakes (Chaohu, Dianchi and Taihu), the largest freshwater lake (Poyang), and the largest saltwater lake (Qinghai). The

selected lakes have different pollution levels: some lakes like Chaohu, Dianchi and Taihu are severely polluted, and others (e.g., Sihailongwan maar) are located in remote areas. Although there are several reports of PBDEs in sediments (Mai et al., 2005; Chen et al., 2007; Shi et al., 2009; Zhang et al., 2009), information from remote areas of China is limited, and there are only a few studies of DBDPE in sediments globally (Kierkegaard et al., 2004; Hu et al., 2010; Lopez et al., 2008; Leonards et al., 2008). To expand knowledge of PBDEs and DBDPE pollution in China, 12 “new” lakes were selected in the current study. This is believed to be the first comprehensive study examining the long-term history pollution of PBDEs, especially of DBDPE in Chinese lakes and will help to understand their distribution, sources, transport and fate in the environment.

## 2. Experimental

### 2.1. Sampling

Sampling was conducted in 2006. Several sediment cores were collected from the central and deepest area in each lake with little anthropogenic processes. One core was used for PBDEs and DBDPE analyses and the others were used for sediment dating ( $^{137}\text{Cs}$  and Pu analyses) and other chemical analyses (Liao, 2007; Wu et al., 2010; Zheng et al., 2008b). The cores were sectioned into 1- or 2-cm intervals immediately after collection, wrapped and weighted in pre-cleaned (by heating at 450 °C) aluminum foil *in situ*, then transported on ice to the laboratory where they were stored at –20 °C until further treatment.

### 2.2. Chemicals

A mixed standard solution of 10 PBDE congeners, including BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183, as well as standard solutions of BDE-206, -207, -208, and -209, were acquired from Accustandards (New Haven, CT, USA). Standards of 2,2',4,4',5-pentachlorodiphenyl ether (CDE-99) and DBDPE were obtained from Wellington Laboratories (Guelph, ON, Canada).  $^{13}\text{C}$ -labeled PCB-141,  $^{13}\text{C}$ -labeled PCB-208 and  $^{13}\text{C}$ -labeled BDE-209 were purchased from Cambridge Isotope Laboratories Inc. (Andover, MA, USA). All solvents and reagents used in the extraction and cleanup procedures were AR grade and all organic solvents were re-distilled. Neutral silica gel (100–200 mesh) and alumina (80–100 mesh) were extracted with a mixture of dichloromethane and methanol (1:1 in volume) for 72 h, and heated at 180 °C and 250 °C for 12 h, respectively. Sodium sulfate was heated at 450 °C and stored in sealed containers.

### 2.3. Sample preparation

Sediment samples were freeze-dried, ground, homogenized and stored at –20 °C until extraction. The extraction procedure, cleanup, fractionation and analytical method have been applied and reported elsewhere (Mai et al., 2005; Chen et al., 2007; Shi et al., 2009; Zhang et al., 2009). Briefly, an aliquot (5–10 g dry weight) of each sample was spiked with surrogate standards (CDE-99,  $^{13}\text{C}$ -labeled PCB-141, and  $^{13}\text{C}$ -labeled BDE-209), and then Soxhlet extracted with a mixture of acetone and hexane (1:1 by volume) for 24 h. Activated copper granules were added

**Table 1**  
Characteristics of the 12 sampled lakes in China.<sup>a</sup>

Lake	Sampling site	Sampling date	Sedimentation rate (sediment accumulation rate)	Lake area (km <sup>2</sup> )	Maximum depth (m)	Catchment area (km <sup>2</sup> )
Dianchi	24°50'N, 102°43'E	May-06	0.28 (0.04)	306.3	10.4	2920
Hongfeng	26°39'N, 106°25'E	May-06	0.81 (0.307)	57.2	45	1596
Chenghai	26°34'N, 100°40'E	May-06	0.78 (0.427)	77.2	36.9	318
Erhai	25°49'N, 100°11'E	May-06	0.21 (0.045)	249.8	21.5	2470
Chaohu	31°31'N, 117°33'E	May-06	0.27 (n.a.) <sup>b</sup>	800	3	9258
Taihu	31°16'N, 120°07'E	Oct-06	0.10 (0.046)	2338	3	36,500
Poyang	29°12'N, 116°16'E	Oct-06	n.a.	3914	29.2	162,215
Sihailongwan maar	126°36'N, 42°17'E	Oct-06	0.07 (0.015)	0.06	50	n.a.
Qinghai	36°57'N, 100°05'E	Aug-06	0.07 (0.007)	4256	28	29,661
Shuangta	39°50'N, 97°36'E	Jul-06	0.79 (0.641)	40	26.8	12,000
Sagan	38°52'N, 93°52'E	Jul-06	0.19 (0.059)	103.7	6	7000
Bosten	41°59'N, 87°11'E	Sep-06	0.28 (0.083)	972	16.5	22,000

n.a.: not available.

<sup>a</sup> Obtained from China Lake Scientific database (Nanjing Institute of Geography and Limnology, 2010).

<sup>b</sup> Sedimentation rates are in  $\text{cm yr}^{-1}$  and  $\text{g cm}^{-2} \text{yr}^{-1}$  (in parentheses). The sedimentation rate in the Lake Chaohu sediment core is from a previous investigation (Yao et al., 2006).

for desulfurization. The extracts were concentrated to about 10 mL by rotary evaporation and solvent-exchanged to hexane. The solutions were then cleaned and fractionated on a silica/alumina column (30 cm long  $\times$  1 cm i.d.) packed, from the bottom to top, with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), 50% sulfuric acid silica (8 cm), and anhydrous sodium sulfate (1 cm). The target chemical mixture was eluted with 30 mL of hexane and 60 mL of hexane:methylene chloride (1:1), and the eluant was again concentrated by rotary evaporation to about 5 mL. The final extract volume was reduced to 100  $\mu$ L under a nitrogen stream.  $^{13}\text{C}$ -labeled PCB-208 was added as an internal standard prior to instrumental analysis.

#### 2.4. Instrumental analyses

The sample pretreatment procedure, instrumental analyses and quality control and assurance used in the current study have been previously reported and described elsewhere (Mai et al., 2005; Chen et al., 2007; Shi et al., 2009; Zhang et al., 2009). The determination of PBDEs and DBDPE was performed with a Shimadzu Model 2010 gas chromatograph (GC) coupled with a Model QP2010 MS using an electron capture negative ionization (ECNI) ion source in the selective ion-monitoring mode. The separation of tri- to hepta-BDE congeners (including BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, and -183) was performed on a DB-XLB (30 m  $\times$  0.25 mm  $\times$  0.25  $\mu$ m) capillary column. The same procedure as in our previous work was followed for the GC temperature programs used and ions monitored (Mai et al., 2005). The separation of nona- to deca-BDE congeners (BDE-206, -207, -208, and -209) and DBDPE was carried out on a DB-5HT (15 m  $\times$  0.25 mm  $\times$  0.1  $\mu$ m) capillary column. Quantification was conducted using an internal calibration method, with the ranges of the calibration standards being 0.2–200  $\mu\text{g L}^{-1}$  for tri- to hepta-BDEs, 20–500  $\mu\text{g L}^{-1}$  for nona-BDEs 20–5000  $\mu\text{g L}^{-1}$  for BDE-209, and 100–2000  $\mu\text{g L}^{-1}$  for DBDPE.

#### 2.5. Quality control and quality assurance

A mixed standard solution containing all the target analytes and surrogate standards was analyzed daily to ensure the validity of calibration curves. Surrogate standards were used for quantification of the concentrations of all compounds by use of mean relative response factors determined from standard calibration runs.  $^{13}\text{C}$ -labeled BDE-209 was used as the surrogate standard for nona- to deca-BDE congeners (BDE-206, -207, -208, and -209) and DBDPE. Isotope-labeled PBDEs cannot be used as the surrogate standards for tri- to hepta-BDE congeners (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, and -183), since GC-ENCI-MS was used in the analysis. Thus in the current study, tri- to hepta-BDE congeners were quantified relative to  $^{13}\text{C}$ -labeled PCB-141 and CDE-99, which have been successfully applied as the surrogate standards for tri- to hepta-BDE congeners (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, and -183) in previous studies (Mai et al., 2005; Chen et al., 2007; Zhang et al., 2009; Hu et al., 2010). The surrogate recoveries were  $86.2 \pm 16.2\%$  for CDE-99,  $87.6 \pm 16.0\%$  for  $^{13}\text{C}$ -labeled PCB-141 and  $96.3 \pm 15.5\%$  for  $^{13}\text{C}$ -labeled BDE-209. For each sediment core, a method blank, a spiked blank (target analyte standards spiked into the solvent) and a spiked matrix (target analyte standards spiked into the matrix) were processed. The average recoveries of 14 BDE congeners were 86.8–127.1% and 94.6–126.3% in the spiked blanks and spiked matrices, respectively, which were similar to the previous report (Wan et al., 2009). The limits of quantification, defined as 10 times the ratio of signal to noise of each congener, were 2–5  $\text{pg g}^{-1}$  for tri- to hepta-BDEs and 20–50  $\text{pg g}^{-1}$  for nona- to

deca-BDEs, and 0.2  $\text{ng g}^{-1}$  for DBDPE (all normalized to dry weight except where indicated). Reported concentrations were blank subtracted and recoveries corrected.

#### 2.6. Dating of the sediment cores

Sediment samples were analyzed for parameters such as organic contents, organic carbon, porosity, bulk density, particle density, and percent solids. Sediment cores were dated using  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  methods (Liao, 2007; Wu et al., 2010; Zheng et al., 2008a, 2008b; Xiong et al., 2010). The detailed experimental procedures and results have been described elsewhere (Wu et al., 2010; Zheng et al., 2008b; Xiong et al., 2010). Briefly,  $^{137}\text{Cs}$  activities were determined using gamma-spectrometry on a Canberra S-100 multi-channel spectrometer with a GC5019 HP-Ge coaxial detector (efficiency 50%), and the determination of Pu activities and its isotopes was carried out using a sector field ICP-MS instrument (Finnigan Element 2, Bremen, Germany) combined with an APEX-Q high efficiency sample introduction system (Elemental Scientific Inc., Omaha, NE, USA) (Wu et al., 2010; Zheng et al., 2008b; Xiong et al., 2010). All measurements were made in the self-aspirating mode to reduce the risk of contamination by the peristaltic pump tubing. For method validation, at least one segment in each sediment core was prepared and dated in three replicates. The average relative standard deviation (RSD) of replicated samples was less than 10%. Our previous study (Zheng et al., 2008b) reported that the year of annual maximum deposition of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$ , resulting from the global fallout in 1964, was in good agreement with the  $^{210}\text{Pb}_{\text{ex}}$  dating results, further validating the method applied in the current study. The sedimentation rate or sediment accumulation rate was estimated by using the year 1964, corresponding to the global fallout peak of artificial radionuclides, as the dating marker for the  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  activity peak profiles in sediments. The results are summarized in Table 1.

#### 2.7. Data analyses

PBDEs and DBDPE concentrations are reported on a dry weight basis. The inventory of PBDEs and DBDPE, in  $\text{ng cm}^{-2}$ , was the total integrated mass of PBDEs and DBDPE per unit area in the sediment core, and was calculated using the equation:  $\text{inventory} = \sum C_i \rho_i d_i$ . The flux, in  $\text{ng cm}^{-2} \text{yr}^{-1}$ , was calculated using the equation:  $\text{flux}_i = C_i \rho_i r_i$ . In the two equations,  $C_i$  ( $\text{ng g}^{-1}$ ) is the concentration in sediment segment  $i$ ,  $\rho_i$  ( $\text{g cm}^{-3}$ ) is the dry mass density of sediment segment  $i$ ,  $d_i$  (cm) is the thickness of segment  $i$ , and  $r_i$  ( $\text{cm yr}^{-1}$ ) is the sedimentation rate. Their total burden was obtained by multiplying the inventory and lake surface area. Sample preparation and dating of the sediment cores are provided in the Supporting Information.

### 3. Results and discussion

#### 3.1. PBDEs in sediment cores

The data from 14 BDE congeners (BDE-28, -47, -66, -100, -99, -85, -154, -153, -138, -183, -206, -207, -208, and -209), as  $\Sigma\text{PBDE}_1$  (sum of tri- to hepta-BDEs),  $\Sigma\text{PBDE}_2$  (sum of nona- to deca-BDE) and  $\Sigma\text{PBDEs}$  (sum of 14 PBDE congeners), are summarized in Table 2. The highest concentration of  $\Sigma\text{PBDEs}$  in surface sediments was found in Lake Dianchi (46.7  $\text{ng g}^{-1}$ ), followed by Lakes

**Table 2**  
Surface concentrations ( $\text{ng g}^{-1}$ ), surface fluxes ( $\text{ng cm}^{-2} \text{yr}^{-1}$ ), and inventories ( $\text{ng cm}^{-2}$ ) of PBDEs and DBDPE in sediments from 12 Chinese lakes.

		Dianchi	Hongfeng	Chenghai	Erhai	Chaohu	Taihu	Sihailongwan maar	Qinghai
Surface concn.	$\Sigma\text{PBDE}_1^a$	0.1	0.29	0.18	0.27	0.3	0.02	0.13	0.12
	BDE-206	2.56	1.43	0.17	n.d.	0.53	n.d.	0.88	0.06
	BDE-207	2.72	2.98	0.99	0.19	0.96	n.d.	2.06	0.19
	BDE-208	1.25	1.03	0.34	0.06	0.40	n.d.	0.59	0.10
	BDE-209	40.1	8.36	1.43	1.45	7.01	6.49	0.36	0.11
	$\Sigma\text{PBDE}_2^b$	46.6	13.8	2.94	1.7	8.9	6.49	3.89	0.46
	$\Sigma\text{PBDEs}^c$	46.7	14.1	3.12	1.97	9.2	6.51	4.01	0.58
Surface flux	DBDPE	1.26	1.02	n.d.	n.d.	3.64	2.67	n.d.	n.d.
	$\Sigma\text{PBDE}_1^a$	0.004	0.09	0.08	0.012	n.a.	0.001	0.002	0.001
	$\Sigma\text{PBDE}_2^b$	1.86	4.24	1.26	0.08	n.a.	0.3	0.058	0.003
	DBDPE	0.05	0.31	n.a.	n.a.	n.a.	0.12	n.a.	n.a.
Inventory	$\Sigma\text{PBDE}_1^a$	1.83	7.86	1.17	0.94	2.82	0.09	0.46	0.18
	$\Sigma\text{PBDE}_2^b$	461	289	10.5	5.95	92.9	32.5	13.6	0.81
	DBDPE	11.8	3.83	n.a.	n.a.	24.6	7.79	n.a.	n.a.

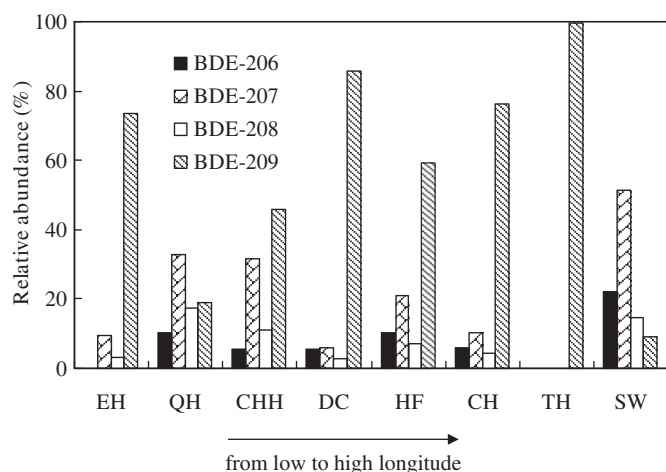
n.d.: not detectable.

n.a.: not available.

<sup>a</sup>  $\Sigma\text{PBDE}_1$ : sum of tri- to hepta-BDE congeners.

<sup>b</sup>  $\Sigma\text{PBDE}_2$ : sum of nona- to deca-BDE congeners.

<sup>c</sup>  $\Sigma\text{PBDEs}$ : sum of 14 BDE congeners.



**Fig. 2.** Nona- and deca-BDE profiles in lake surface sediments. Abbreviations for the Chinese lakes are as given in Fig. 1.

Hongfeng ( $14.1 \text{ ng g}^{-1}$ ), Chaohu ( $9.20 \text{ ng g}^{-1}$ ), and Taihu ( $5.61 \text{ ng g}^{-1}$ ). These findings are not surprising because these lakes, located in eastern or southwestern China, are adjacent to large cities and receiving significant local inputs of urban runoff and sewage discharge. The remote northwestern (Qinghai) and northeastern lake (Sihailongwan maar), where the watersheds are less urbanized and industrialized, have  $\Sigma\text{PBDEs}$  concentrations of  $0.56$  and  $4.02 \text{ ng g}^{-1}$  in the surface sediments, respectively. Because no direct pollution source was present in these remote areas, atmospheric deposition was probably the major source. Although similar spatial distribution was also observed for polycyclic aromatic hydrocarbons (PAHs) in 5 lakes from western China in our previous study (Guo et al., 2010), further investigation needs to be done in confirming atmospheric deposition being the major source to these lakes. Overall, PBDEs in surface sediments were dominated by  $\Sigma\text{PBDE}_2$ , which comprised 82.1–99.8% of the total PBDEs. This is consistent with the dominance of the  $\Sigma\text{PBDE}_2$  group of congeners in the deca-BDE technical mixture, which is the major brominated flame retardant used in China. It is interesting that the patterns of highly brominated BDEs were different among the lakes studied (Table 2 and Fig. 2). In Lakes Chaohu, Dianchi, Erhai and Taihu, BDE-209 was the dominant congener in the surface sediments, and

the patterns were similar to that of the deca-BDE technical mixture (La Guardia et al., 2007). This suggests that urban runoff and sewage discharge, in which the PBDE pattern in the technical mixture shows little change, is likely to be the main source of BDE-209 in these lakes. In the other lakes, the contributions of nona-BDE congeners (including BDE-206, BDE-207 and BDE-208) to  $\Sigma\text{PBDEs}$  were relatively high. Especially in the remote Lakes Qinghai and Sihailongwan maar, where the contributions of BDE-207 were significantly higher than those in Lakes Dianchi and Chaohu, BDE-207, rather than BDE-209, was the most abundant congener. MS analysis of the sample from Lake Sihailongwan maar revealed that the ratios between the fragment ions of BDE-207 were all within  $\pm 15\%$  of the theoretical values and the GC retention times matched those of the standard compound within  $\pm 0.3 \text{ min}$ , therefore qualitatively confirming its occurrence. This finding may be due to degradation and/or debromination of BDE-209 under sunlight and in the sediments (Söderström et al., 2004; Bezares-Cruz et al., 2004; Gerecke et al., 2005). As in many other studies (Song et al., 2004, 2005a, 2005b; Zhu and Hites, 2005; Mai et al., 2005), octa-BDE congeners were not included in the current study because of the facts that the usage of octa-BDE was officially banned in China in 2006 (Hu et al., 2010), and the contribution of octa-BDE congeners in technical deca-mixture relative to nona-BDE congeners could be neglected (La Guardia et al., 2007). Therefore, the fate and different sources contributed to the different patterns of highly brominated BDEs among the lakes studied.

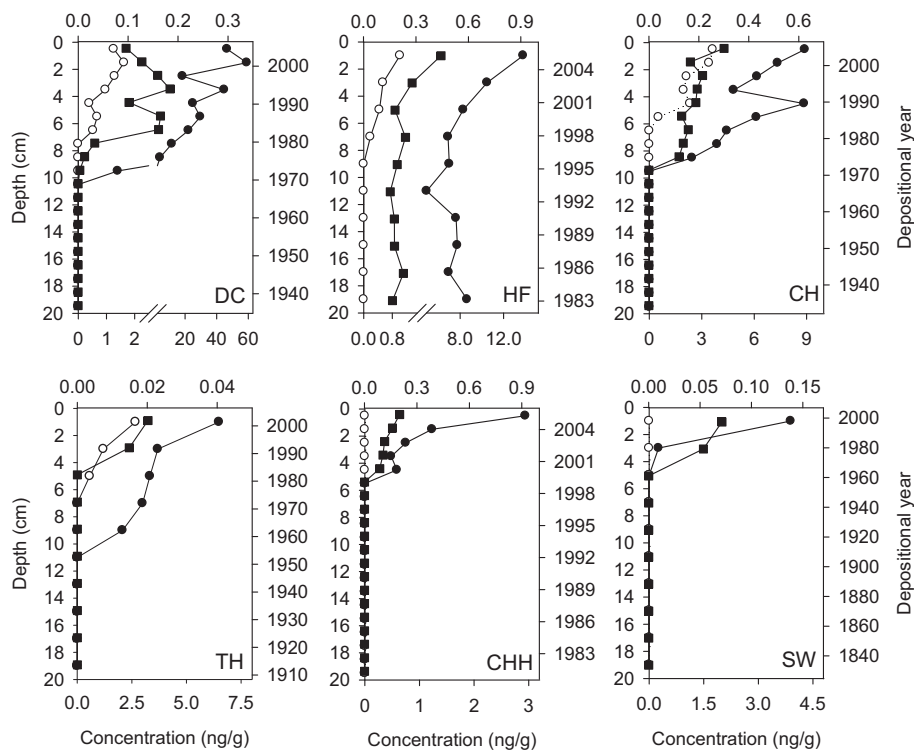
PBDEs have been extensively investigated in sediments worldwide. As shown in Table 3, less than  $50 \text{ ng g}^{-1}$  of  $\Sigma\text{PBDE}_1$  was reported in most studies, although up to  $1270.8 \text{ ng g}^{-1}$  for  $\Sigma\text{PBDE}_1$  was found in the area downstream from a manufacturing plant in the Tees Estuary in UK (Allchin et al., 1999), and up to  $352 \text{ ng g}^{-1}$  was detected in Osaka Bay in Japan (Ohta et al., 2002). High concentrations of BDE-209 were also found in sediment samples from Europe and Japan. The highest BDE-209 concentration reported was  $11,600 \text{ ng g}^{-1}$  in a river sediment sample in Japan (Watanabe et al., 1995), followed by  $7100 \text{ ng g}^{-1}$  in a river sample from Sweden (Sellström et al., 1998) and  $3190 \text{ ng g}^{-1}$  in a river sediment sample from UK (Allchin et al., 1999). High levels of PBDEs were also measured in the PRD region of China, where concentrations of  $\Sigma\text{PBDE}_1$  and BDE-209 were  $0.04$ – $94.7$  and  $0.4$ – $7340 \text{ ng g}^{-1}$ , respectively (Mai et al., 2005). By comparison, concentrations of  $\Sigma\text{PBDE}_1$  ( $0.02$ – $0.29 \text{ ng g}^{-1}$ ) and  $\Sigma\text{PBDE}_2$  ( $0.46$ – $46.6 \text{ ng g}^{-1}$ ) in the

**Table 3**

PBDE and DBDPE concentrations ( $\text{ng g}^{-1}$ ) in sediments of 12 Chinese lakes in comparison with those in the world.

Location	Tri- to hepta-BDE	BDE-209	Nona–deca-BDE	DBDPE
Great lakes, USA (Song et al., 2004, 2005a, 2005b)	0.49–6.33	4.3–242	n.a.	n.a.
Virginia watersheds, USA (Hale et al., 2001)	<0.5–52.3	n.a.	n.a.	n.a.
River, Sweden (Sellström et al., 1998)	8–50	68–7100	n.a.	n.a.
Rivers and estuaries, UK (Allchin et al., 1999)	1.3–1270.8	0.6–3190	n.a.	n.a.
Cinca River, Spain (Eljarrat et al., 2005)	0.24–3.94	2.46–132	n.a.	n.a.
River and Coast, Portugal (Lacorte et al., 2003)	0.5–20	n.a.	n.a.	n.a.
Rivers and estuaries, Netherlands (De Boer et al., 2003)	0.6–17.6	4–510	n.a.	n.a.
Osaka Bay, Japan (Ohta et al., 2002)	8–352	<25–11,600	n.a.	n.a.
Tokyo Bay, Japan (Minh et al., 2007)	0.05–3.6	0.89–85	n.a.	n.a.
Coast, South Korea (Moon et al., 2007)	0.05–0.90	0.22–493	n.a.	n.a.
Pearl River Delta, South China (Mai et al., 2005)	0.04–94.7	0.4–7341	n.a.	n.a.
Western Scheldt, Netherlands (Kierkegaard et al., 2004)	n.a.	n.a.	n.a.	24
Western Scheldt, Netherlands (Leonards et al., 2008)	n.a.	100,560	n.a.	1.1,10
Western Scheldt, Netherlands (Lopez et al., 2008)	n.a.	4.7–260	n.a.	0.65–9.8
Pearl River Delta, South China (Zhang et al., 2009)	0.7–7.6	n.a.	30–2300	19–430
E-waste area, South China (Shi et al., 2009)	n.a.	n.a.	n.a.	38.8–364
Fuhe River, North China (Hu et al., 2010)	0.13–6.39	11.8–292.7	n.a.	16.1–68.2
Lake Baiyangdian, North China (Hu et al., 2010)	0.05–5.03	4.35–19.3	n.a.	1.10–5.29
12 lakes, China (this study)	0.02–0.30	0.11–40.1	0.46–46.6	1.02–3.64

n.a.: not available.



**Fig. 3.** Vertical profile of PBDEs and DBDPE in the sediment cores (top axis:  $\Sigma$ PBDE<sub>1</sub>; bottom axis:  $\Sigma$ PBDE<sub>2</sub> and DBDPE; solid square:  $\Sigma$ PBDE<sub>1</sub>; empty circle: DBDPE; solid circle:  $\Sigma$ PBDE<sub>2</sub>). Abbreviations for the Chinese lakes are as given in Fig. 1.

current study were relatively low, which may indicate that PBDE contamination was still low in most parts of China up to 2006.

Detectable amounts of PBDEs were absent in Lakes Bosten, Poyang, Shuangta and Sugan and were only detected in the surface sediment in Lakes Erhai and Qinghai. Therefore, temporal trends of PBDEs are only available from 6 lakes. As shown in Fig. 3,  $\Sigma$ PBDE<sub>1</sub> were first observed in the 1970s for most lakes, and the concentrations generally showed an increasing trend to the surface. The increase was coincident with the production and use figures of penta-mixture in the world since the 1970s. This profile has also been observed in dated sediment cores from the Great Lakes (Song et al., 2004, 2005a, 2005b) and the Baltic Sea (Nylund et al., 1992). Notably,  $\Sigma$ PBDE<sub>1</sub> concentrations in Lake Dianchi decreased in upper sediments deposited after 1990, probably indicating reduced use of the penta-mixture in this area. The increasing  $\Sigma$ PBDE<sub>1</sub> trends in other lakes suggest the continuous inputs of  $\Sigma$ PBDE<sub>1</sub> despite the persistence and potential toxicity of the penta-BDE technical mixture. The concentrations of  $\Sigma$ PBDE<sub>2</sub> in sediments of the six cores also followed a general increasing trend from the bottom to middle sediment layers approximately corresponding to 1990. In sediments deposited after the first peak, the  $\Sigma$ PBDE<sub>2</sub> profiles were quite different. In Lakes Chaohu and Dianchi,  $\Sigma$ PBDE<sub>2</sub> concentrations fluctuate in a zigzag pattern in sediments deposited after 1990. These two lakes are very close to the large urbanized and industrialized areas of Kunming and Hefei Cities, respectively, which may be important PBDEs sources to the nearby watersheds. This temporal trend may reflect changes in the production and use of the deca-BDE technical mixture in these areas. In Chenghai, a mountain lake,  $\Sigma$ PBDE<sub>2</sub> concentrations occur first in sediment deposited after 1998, which is very late compared with the other lakes. It should be noted that separate cores were used for dating and chemical analysis in the current study, which has the potential to mismatch the chemical and  $^{137}\text{Cs}/^{239+240}\text{Pu}$  profiles. However, as discussed above for PBDEs and following for DBDPE, the time

trends are generally consistent with global production and usage of these chemicals (Song et al., 2004, 2005a, 2005b; Zhu and Hites, 2005), suggesting that any possible effect on this matching caused by the separate analysis is negligible.

Inventories of  $\Sigma$ PBDE<sub>1</sub> and  $\Sigma$ PBDE<sub>2</sub> in the lakes are presented in Table 2. The  $\Sigma$ PBDE<sub>1</sub> and  $\Sigma$ PBDE<sub>2</sub> inventories ranged from 0.09 to 7.86 ng cm<sup>-2</sup>, and 0.81 to 461 ng cm<sup>-2</sup>, respectively. Using the inventory values in Table 2, the total burden of  $\Sigma$ PBDE<sub>1</sub> and  $\Sigma$ PBDE<sub>2</sub> was estimated to be 5.6 and 1400 kg in Lake Dianchi, 21 and 760 kg in Lake Taihu, 22 and 740 kg in Lake Chaohu, and 4.5 and 160 kg in Lake Hongfeng, respectively. The total PBDE burden (0.008 kg) was obtained for Lake Sihailongwan maar, which has a small water surface area of 0.06 km<sup>2</sup>. However, the actual total inputs of PBDEs to these lakes were likely underestimated because the sampling sites were located in the central and deepest area in each lake, an area with little anthropogenic activity, especially for such polluted lakes as Chaohu, Dianchi and Taihu, which would have a much greater portion of PBDEs transported from their estuaries. With the rapidly developing industrial activity and increasing use of polymeric materials in construction and electronic equipment in recent years, the consumption of PBDEs has increased in China. However, this increase is geographically unbalanced, being most marked for coastal regions or areas of intensive manufacturing of electronic products such as the PRD region. Many western and northern inland lakes in China are not as contaminated by PBDEs as the PRD region due to less anthropogenic inputs.

The current fluxes of  $\Sigma$ PBDE<sub>1</sub> and  $\Sigma$ PBDE<sub>2</sub>, as observed at the sediment surface for these lakes sampled in 2006, are in the range of 0.001–0.09 ng cm<sup>-2</sup> yr<sup>-1</sup> and 0.003–4.24 ng cm<sup>-2</sup> yr<sup>-1</sup>, respectively. Using these values, the loading rates to these lakes are estimated to be from 0.00001 to 0.06 kg yr<sup>-1</sup> for  $\Sigma$ PBDE<sub>1</sub>, and from 0.0003 to 5.7 kg yr<sup>-1</sup> for  $\Sigma$ PBDE<sub>2</sub>. These estimates may be a fraction of atmospheric deposition and other sources. It should be noted that the fluxes presented in the current study are not normalized by

sediment focusing since the regional atmospheric inputs of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  are not available. Thus, the flux values may be affected by horizontal movement of sediment particles on the lake bottom.

### 3.2. DBDPE in sediment cores

DBDPE concentrations in the surface sediments collected from the three major state lakes (Lakes Chaohu, Dianchi and Taihu) and Lake Hongfeng ranged from 1.02 to 3.64 ng g<sup>-1</sup> (Table 2). High levels of BDE-209 were also found in sediments of the four lakes, but high ratios of DBDPE/BDE-209 were found in samples with low levels of BDE-209. DBDPE/BDE-209 ratios were close to 2.0 in Lake Chaohu and Taihu from eastern China, and were 0.03 and 0.12 in Lake Dianchi and Hongfeng from southwestern China, respectively. DBDPE was not detected in Lake Chenghai and Erhai from southwestern, Lake Qinghai from northwestern, and Lake Sihailongwan maar from northeastern China. The higher levels of DBDPE observed in Lake Chaohu and Taihu are not surprising because DBDPE production in China mainly occurs in the eastern part, especially in Shandong and Jiangsu provinces (Firete, 2010). However, the low ratios of DBDPE/BDE-209 in Lakes Dianchi and Hongfeng and the non-detection of DBDPE in the other studied lakes indicate the higher use of BDE-209 than DBDPE. To date, few data on the concentrations of DBDPE in global surface sediments are available. In 2004, Kierkegaard et al. (2004) first found 24 ng g<sup>-1</sup> dw of DBDPE in sediment from Western Scheldt in the Netherlands. In recent investigations (Lopez et al., 2008; Leonards et al., 2008) in the same river, Leonards and his colleagues detected DBDPE with a maximum concentration of 10 ng g<sup>-1</sup> dw. Most recently, as high as 38.8–364 ng g<sup>-1</sup> dw (Shi et al., 2009) and 19–430 ng g<sup>-1</sup> dw (Zhang et al., 2009) of DBDPE were reported in an e-waste area and in the PRD region (a large manufacture region of electronic products) in southern China, respectively. It was reported that DBDPE concentrations ranged from 1.10 to 5.29 ng g<sup>-1</sup> dw in Lake Baiyangdian from northern China, and they depended on the sampling sites, and were relatively high (up to 5.29 ng g<sup>-1</sup> dw) in the lake nearshore and river areas due to the effects of local point sources, and were much low (around 1.10 ng g<sup>-1</sup> dw) in the lake central areas (Hu et al., 2010). Thus, in comparison with Lake Baiyangdian, it seems that the DBDPE levels, especially in Lake Taihu (2.67 ng g<sup>-1</sup>) and Chaohu (3.64 ng g<sup>-1</sup>) in the current study are higher than those in and close to the central area, but are lower than those in the nearshore areas in Lake Baiyangdian. Therefore, it seems that DBDPE levels varied in different lakes in China, and also varied in different lake sampling areas. Further investigations need to understand the spatial distributions and sources of DBDPE in Chinese lakes.

Temporal trends for DBDPE were only found for sediment cores from the three major state lakes and Lake Hongfeng (Fig. 3). DBDPE is first present in sediment cores from the three major state lakes from the 1980s, and from ca. 1998 in the core from Lake Hongfeng, which is at least one decade later than that for PBDEs. As previously mentioned, DBDPE was introduced in the early 1990s (Kierkegaard et al., 2004), and the low concentrations of DBDPE in the sediment cores prior to the commercialization of DBDPE could be due to vertical mixing of the sediments. Similar to PBDEs, DBDPE concentrations increased steadily with decreasing sediment depth, which is consistent with the previous study in the PRD region (Zhang et al., 2009).

No previous studies appear to have reported on the inventories and fluxes of DBDPE in lake sediment cores. The DBDPE inventories in the current study were 24.6, 11.8, 3.83 and 7.79 ng cm<sup>-2</sup> in Lakes Chaohu, Dianchi, Hongfeng and Taihu, respectively. The total burden of DBDPE was calculated to be 2.2 kg in Chaohu, 200 kg in

Lake Dianchi, 36 kg in Hongfeng and 180 kg in Taihu. Calculated DBDPE fluxes were 0.05 ng cm<sup>-2</sup> yr<sup>-1</sup> for Lake Dianchi, 0.31 ng cm<sup>-2</sup> yr<sup>-1</sup> for Lake Hongfeng, and 0.12 ng cm<sup>-2</sup> yr<sup>-1</sup> for Lake Taihu. By comparison, both inventory and flux for DBDPE were less than those for  $\Sigma\text{PBDE}_2$  or BDE-209, especially in Lake Dianchi and Hongfeng, showing the higher use of deca-BDE technical mixture than DBDPE. Despite limited information about the adverse effects, the occurrence and fate of DBDPE in the environment requires more attention based on the alarmingly high levels of DBDPE in other environmental matrices and the rapidly increasing trend in their concentrations and fluxes around 2006 observed in the current study.

## 4. Conclusion

PBDEs and DBDPE were analyzed in twelve sediment cores across China. And the urbanization and industrialization are highly related to their contamination in sediments. The temporal trends of PBDEs and DBDPE concentrations generally increased from the bottom to the surface, although  $\Sigma\text{PBDE}_2$  concentrations fluctuate in a zigzag pattern in sediments deposited after 1990 in Lakes Chaohu and Dianchi. The fluxes and mass inventory indicate that the  $\Sigma\text{PBDE}_2$  input over the past three decades was much higher than that of  $\Sigma\text{PBDE}_1$ . The DBDPE input in recent years was still lower than  $\Sigma\text{PBDE}_2$  input but a rapid increasing trend of DBDPE in sediments was found in some lakes. The contamination of DBDPE is expected to increase with its increasing usage in the future. Therefore, widespread and continuous monitoring studies need to be done in the whole country to manage environmental risks.

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