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Polybrominated diphenyl ethers in chicken tissues and eggs from an electronic waste recycling area in southeast China

Xiaofei Qin[1], Zhanfen Qin[1,*], Yan Li[1], Yaxian Zhao[1], Xijuan Xia[1], Shishuai Yan[1], Mi Tian[1], Xingru Zhao[2], Xiaobai Xu[1], Yongjian Yang[3]

1. State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China. E-mail: xiaofeiqin@163.com
2. Chinese Research Academy of Environmental Sciences, Beijing 100012, China
3. Taizhou No. 1 Middle School, Taizhou 318000, China

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Abstract

The levels and distributions of polybrominated diphenyl ethers (PBDEs) in chicken tissues from an electronic waste (e-waste) recycling area in southeast China were investigated. Human dietary intake by local residents via chicken muscle and eggs was estimated. The mean PBDEs concentrations in tissues ranged from 15.2 to 3138.1 ng/g lipid weight (lw) and in egg the concentration was 563.5 ng/g lw. The results showed that the level of total PBDEs (∑PBDEs) in the chicken tissue was 2–3 orders of magnitude higher than those reported in the literature. The large difference of ∑PBDEs concentrations between tissues confirmed that the distribution of PBDEs in tissues depend on tissue-specificity rather than the "lipid-compartment". BDE-209 was the predominant congener (82.5%–94.7% of ∑PBDEs) in all chicken tissues except in brain (34.7% of ∑PBDEs), which indicated that deca-BDE (the major commercial PBDE formulation comprising 65%–70% of total production) was major pollution source in this area and could be bioaccumulated in terrestrial animals. The dietary PBDEs intake of the local residents from chicken muscle and egg, assuming only local bred chickens and eggs were consumed, ranged from 2.2 to 22.5 ng/(day·kg body weight (bw)) with a mean value of 13.5 ng/(day·kg bw), which was one order of magnitude higher than the value reported in previous studies for consumption of all foodstuffs.

Key words: polybrominated diphenyl ethers; chicken; egg; bioaccumulation; tissue distribution; exposure risk

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Introduction

Rapid technology change has resulted in a fast-growing surplus of electronic waste (e-waste) around the globe. Recently, e-wastes are becoming a major environmental concern, particularly in developing countries where e-wastes are mainly imported from developed countries (Schwarzer et al., 2005; Leung et al., 2007). It was reported that 50%–80% of the global e-wastes are legally or illegally imported to Asia. In the process of primitive recycling treatment of e-wastes, the released hazardous chemicals resulted in serious pollution problem in local area and surrounding regions (Chan et al., 2007; Leung et al., 2008; Zheng et al., 2008). Among these pollutants, polybrominated diphenyl ethers (PBDEs), as additive flame retardants in electronic equipments, are one class pollutant of the most concerns due to persistence, bioaccumulation and potential toxicity (Darnerud and Sinjari, 1996; Eriksson et al., 1999, 2002).

Wenling is located in Taizhou City, southeast coast of China. As well as Guiyu of Guangdong Province, the region near Wenling is one of the largest e-wastes recycle process areas (Wang et al., 2005; Bi et al., 2007; Leung et al., 2007). In this area many small family-run workshops have devoted to processing discarded electronic waste, known as e-waste, for ten years. They disassembled and shattered e-waste into powder to select the usable materials. Although several reports presented high concentrations of PBDEs in soils, sediments and biotic samples from this PBDEs-polluted area (Liang et al., 2008; Yang et al., 2008, 2009), information on PBDEs pollution status in this area is scarce, and data on exposure risk of PBDEs for local residents are limited.

Birds have been applied as sentinel species for monitoring PBDEs pollution status on relative large geographic scales (Naert et al., 2007; Chen et al., 2007; Voorspoels et al., 2007a; Van den Steen et al., 2009). The increasing trend of PBDEs levels in bird tissues and eggs, in past decades, is coincident with that observed for other animals and human being (Nortstrom et al., 2002; Hites, 2004). There

* Corresponding author. E-mail: qinzhanfen@rcees.ac.cn

is potential exposure risk of PBDEs for local residents via poultry meat and eggs, which are common constituents of human diets.

The main objectives of the present study were to measure the levels of PBDEs in chickens and eggs from Wenling and to estimate dietary uptake of PBDEs by local residents via chicken and eggs. In addition, distribution and congener patterns of PBDEs in chicken tissues were also discussed.

1 Materials and methods

1.1 Chemicals

Polychlorinated biphenyl (PCB) 209 used as surrogate was obtained from Supelco (USA). A standard solution of PBDE congeners (EO-5278) was obtained from Cambridge Isotope Laboratories, Inc. (USA); *n*-hexane, methylene dichloride, and nonane at pesticide grade were from Tedia (USA); and anhydrous sodium sulfate, sulfuric acid, sodium hydroxide, and anhydrous ethanol at analytical grade were from Beijing Chemical Factory (China).

1.2 Study area and sampling

Wenling area is located in the south of Taizhou, east of Zhejiang Province, with population of about one million. In this area, family-run workshops for e-waste recycling process are very popular. For many families it is the main familiar income. In addition, traditional agriculture, such as rice growing, vegetables planting, fish farming, and poultry breed, also plays an important role in the local economic system. Sanhuang (SH) chicken, a popular Chinese indigenous breed, is among the most consumed meat in Chinese market. In this investigation SH chickens (n = 4, about 3–4 months old) and eggs (n = 15) were obtained from an e-wastes recycling village in Wenling area. Various tissues of chickens were excised and all the tissues were cleaned with deionized water, wrapped in aluminum foil, and sealed in plastic bags to minimize the possibility of contamination after sampling. Grain used to feed the chickens and soil samples from location where the chickens roamed were also collected. Then, tissue, grain and soil samples were transported to analysis laboratory in an ice box and stored in freezer at –20°C until analysis. Eggs were transported at room temperature and stored in refrigerator at 4°C until analysis.

1.3 Extraction and cleanup

The samples were freeze-dried and homogenized with anhydrous sodium sulfate, 1 ng PCB 209 were spiked into the sample as surrogate and equilibrated for 2 hr in desiccator. Then, the sample was ultrasonic extracted for 4 min with 30 mL of hexane/dichloromethane (1:1, *V/V*). After centrifuged (3000 $\times g$ for 3 min) the supernatant was transferred to a flask. The extraction process was repeated 3 times and the extracts were combined. The lipids content was determined gravimetrically after evaporating the extract to dryness. The sample was then reconstituted in 1 mL hexane and was cleaned up with multilayer silica gel column (0.2 m × 15-mm internal diameter) packed with 1 g activated silica gel, 3 g basic silica gel, 1 g activated silica gel, 4 g acid silica gel (44% concentrated sulfuric acid, *W/W*), 4 g acid silica gel (22% concentrated sulfuric acid, *W/W*), 1 g activated silica gel and 2 g anhydrous sodium sulfate. PBDE fraction was eluted from the column with 100 mL hexane. The eluent was concentrated to about 2 mL in a rotary evaporator. Then the sample solution was transferred to a vial and reduced to a volume of 20 μL under a gentle stream of N_2.

1.4 Analysis of PBDEs

The analysis of PBDEs was performed using an Agilent 6890 series gas chromatograph equipped with Agilent 5973 mass spectrometer (Agilent Technologies, USA) operated in electron capture negative ionization (ECNI) mode. The gas chromatography column was a 15 m × 0.25 mm i.d. DB-5 MS capillary column with a film thickness of 0.25 μm. The injector and interface temperatures were 290 and 300°C, respectively. A 1 μL aliquot of sample solution was injected with pulsed splitless injection mode. Helium was used as carrier gas at constant flow (1.0 mL/min) and methane was used as reagent gas. The ion source and quadrupole temperatures were 150°C. The oven temperature was programmed from 80°C (held for 1 min) to 200°C at a rate of 10°C/min and then from 200 to 300°C (held for 15 min) at a rate of 20°C/min. The mass spectrometer was operated in the selected ion monitoring (SIM) mode and ions of m/z 79 and 81 were monitored for BDE-47, 100, 99, 154, 153, 183 and m/z 486.7 and 488.7 for BDE-209.

Identification of analyte was based on comparison of retention time and mass spectrum with appropriate standards. Quantification was performed by external standard method with multi-level calibration curve spanning the range of anticipated analyte concentrations in the samples.

1.5 Quality assurance/quality control

To avoid the potential sample contamination and PBDE degradation, the proper handling was adopted from sample collection to chemical analysis. The method precision and recovery were determined by analyzing fish tissue that was spiked with PBDEs standard. The recoveries for target congeners ranged from 85% to 115% with relative standard deviation of 20%. One procedural blank was run for every batch of nine samples to check the potential contamination in analysis process. The recoveries of surrogate in all samples ranged from 85% to 110%. The limit of detection (LOD) was defined as the concentration of analyte in the sample producing a peak with the ratio of signal to noise of 3 (peak-to-peak). LODs ranged from 0.001 to 0.05 ng/g for BDE-47 to BDE-183 and 0.1 ng/g for BDE-209.

1.6 EDI of PBDEs through chicken and eggs consumption

The daily dietary intake of PBDEs was estimated depending on PBDEs concentration in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance of pollutants. Estimated daily intake (EDI) was calculated assuming that the local residents consume only local bred chickens and eggs. EDI was calculated as follows:

$$\text{EDI} = \frac{C \times \text{Cons}}{\text{BW}}$$

where, C (ng/g lw) is the concentration of PBDEs in contaminated chicken muscle and eggs, Cons stands for the daily average consumption of chicken muscle and eggs in this region, and BW represents the body weight. Average body weight of the adult residents in this area was 60 kg in the present study. Based on the dietary nutrition intake level survey by Zhong et al. (2006), poultry meat (chicken as the dominant) was one of the staple foods for daily consumption, and the adult residents in this area have an average daily intake of 23 g poultry meat and 17.6 g eggs per day.

1.7 Statistical analyses

All the statistical analysis was performed using SPSS software version 13.0 (SPSS Inc., Chicago, USA). Significant difference among the congener in the same tissue was analyzed using one-way analysis of variance. Significant difference concentration of total PBDEs (∑PBDEs) between tissues was analyzed using Mann-Whitney U test. A value of α = 0.05 was chosen to give a significant difference.

2 Results and discussion

2.1 PBDEs levels and tissue distribution

Table 1 and Fig. 1 show lipid-normalized concentrations (lipid weight, lw) of individual PBDE congeners and ∑PBDEs in various tissues of chickens, including blood, fat, intestine, liver, muscle, kidney, lung, testis and brain. The mean concentrations of ∑PBDEs in various tissues ranged from (15.23 ± 8.27) ng/g lw in brain to (3138.06 ± 2476.93) ng/g lw in fat. The concentrations of ∑PBDEs in chicken muscle (5.90 ± 2.54 ng/g wet weight (ww)) were much higher than those in chicken breast from markets of U.S. (0.3 ng/g ww) (Schecter et al., 2004) and Belgium (0.03 ng/g ww) (Voorspoels et al., 2007b). The mean concentrations of ∑PBDEs in the chicken muscle and liver samples (muscle: 1092.42 ng/g lw; liver: 1077.81 ng/g lw) were about 100 times higher than those reported in male chickens (muscle: 66 ng/g lw; liver: 41 ng/g lw) and male ducks (muscle: 6.6 ng/g lw; liver: 8.5 ng/g lw) collected from an e-waste recycling site in Guangdong Province, South China (Luo et al., 2009). Concentration of ∑PBDEs in eggs averaged (563.51 ± 339.82) ng/g lw and (38.31 ± 42.31 ng/g ww) in the present study. This PBDEs contamination level was two orders of magnitude higher than those in Belgian home-produced eggs (7.8 ng/g lw) (Covaci et al., 2009) and eggs (0.5 ng/g lw) from markets of Spain (Bocio et al., 2003).

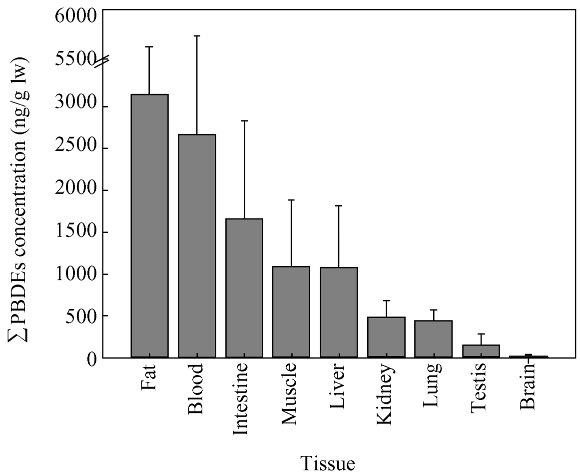

Fig. 1 Tissue distribution of total polybrominated diphenyl ethers.

The concentration of ∑PBDEs decreased in the following order in the present study: fat > blood > intestine > muscle ⩾ liver > kidney ⩾ lung > testis > brain (Fig. 1). Statistical analysis revealed that the concentration of ∑PBDEs in muscle and liver were not different. Also, no significant difference in the concentration of ∑PBDEs between kidney and lung was found. However, brain showed lower levels than the other tissues. Matthews and Dedrick (1984) suggested that lipophilic compounds, such as PCBs, distributed uniformly into the "lipid-compartment" in biota tissues. If so, concentrations of PBDEs, as a class of lipophilic compounds, on a lipid-weight basis might be similar in all the tissues. Our results showed that lipid-normalized concentrations of ∑PBDEs in some tissues were very different. This result is in consistent with other reports (Voorspoels et al., 2006; Liang et al., 2008).

Some researchers suggested that the difference distribution of ∑PBDEs in tissues resulted from different metabolic activities in different tissues. For example, Voorspoels et al. (2006) reported concentration of ∑PBDEs in muscle were higher than that in liver for several bird species and suggested that relatively higher metabolic activity in liver than in muscle could explain this phenomenon. Luo et al. (2009) also explained their results in chickens using this hypothesis. However, some studies

Table 1 Levels of individual PBDE congeners in various tissues of chickens (n = 4) from an e-wastes recycling area in Southeast China*

PBDE congener	Level of individual PBDE congeners (ng/g lipid weight)								
	Fat	Testis	Kidney	Lung	Muscle	Intestine	Liver	Blood	Brain
BDE-47	25.24 ± 11.83	6.84 ± 4.52	6.99 ± 3.83	7.34 ± 5.45	20.10 ± 28.84	37.85 ± 39.20	21.27 ± 15.04	25.98 ± 13.95	1.90 ± 1.28
BDE-100	9.05 ± 6.70	1.93 ± 1.77	3.19 ± 1.27	1.94 ± 0.72	3.32 ± 2.54	8.90 ± 6.93	7.69 ± 4.52	14.69 ± 6.22	0.67 ± 0.51
BDE-99	31.34 ± 17.87	5.88 ± 4.19	6.14 ± 2.42	6.80 ± 4.03	14.25 ± 17.06	41.83 ± 48.93	15.45 ± 11.60	26.63 ± 12.79	1.72 ± 1.23
BDE-154	7.60 ± 6.31	0.97 ± 0.84	1.45 ± 0.43	1.47 ± 0.75	2.10 ± 1.77	4.98 ± 3.96	2.04 ± 2.15	9.35 ± 4.74	0.43 ± 0.41
BDE-153	39.90 ± 27.61	5.09 ± 2.79	7.95 ± 3.83	6.53 ± 8.10	10.14 ± 8.43	28.72 ± 20.74	16.27 ± 13.38	29.83 ± 15.37	2.50 ± 1.43
BDE-183	54.57 ± 57.13	7.22 ± 5.38	14.52 ± 19.61	17.64 ± 19.31	8.66 ± 6.11	42.44 ± 59.24	35.72 ± 62.13	62.12 ± 73.07	2.72 ± 2.09
BDE-209	2971.65 ± 2454.27	131.22 ± 121.95	443.34 ± 161.50	402.67 ± 88.76	1033.86 ± 729.58	1491.31 ± 1120.27	979.38 ± 643.38	2491.16 ± 3019.06	5.29 ± 3.22

* Data are presented as mean ± SD (standard deviation).

reported that no significant difference of ∑PBDEs concentration was found between liver and muscle of terrestrial birds (Chen et al., 2007). In the present study, our results also showed that there was no significant difference of ∑PBDEs concentration found between liver and muscle. It seems that different metabolic activities in tissues can not completely explain this phenomenon.

It was reported that PBDEs and their metabolites like other lipophilic compounds could bind to some proteins or lipoproteins, such as thyroid hormone transport protein, retinol binding protein, fatty acid binding protein (Gómez-Catalán et al., 1991; Marsh et al., 1998; Cheek et al., 1999; Meerts et al., 2000). Maybe, these proteins or other proteins involve in the tissue distribution of PBDEs in different tissues.

In the present study, the concentrations of ∑PBDEs in brain were significantly lower than those in other tissues (e.g., muscle and liver) of chickens. This was also observed in birds of prey and wild rodents (Voorspoels et al., 2006, 2007a), suggesting that the efficiency of the blood-brain barrier was probably protecting the brain from accumulation of PBDEs (Bachour et al., 1998; Voorspoels et al., 2006). Considering potential developmental neurotoxicity of PBDEs (Branchi et al., 2003; Viberg et al., 2003), the presence of PBDEs in animal brains should be paid much attention. In addition, this study reported for the first time PBDEs bioaccumulation in testis of animals. Several studies showed that PBDEs have potential adverse effects on spermatogenesis, such as the decrease in sperm and spermatid counts (Kuriyama et al., 2005; Lilienthal et al., 2006). Although very low concentrations of pollutants might not cause any obvious damage, our results indicated that the potential damage of PBDEs to animal spermatogenesis should arouse much attention.

2.2 Congener patterns in chickens

The congener pattern of PBDEs detected in chicken tissues, grain and soil from this area is shown in Fig. 2. BDE-209 contributed up to 48.04% and 60.64% of ∑PBDEs in grain and soil, indicating that main PBDE pollution source in Taizhou region was deca-BDE mixture. BDE-209 was also the dominant congener among detected PBDE congeners in all chicken tissues (Table 1), and contributed up to 82.46%–94.70% of ∑PBDEs except for brain samples (Fig. 2). BDE-183 was the second abundant congener in chicken tissues. In early study, it was suggested that high brominated congeners could not be bioaccumulated in biotic tissues due to its large molecular size and high octanol-water partition coefficient (Boon et al., 2002). However, with increasing finding of BDE-209 in many biotic samples, it was shown that all PBDE congeners were bioavailable (Sjödin et al., 1999; Eljarrat et al., 2007). Our results also showed that high brominated congeners (e.g., BDE-209 and BDE-183) could be bioaccumulated in terrestrial animals. Generally, BDE-47 was the most abundant congener in birds while high brominated congeners were relatively less (Law et al., 2003; Jaspers et al., 2006; Naert et al., 2007). However, in the present study, BDE-47 only contributed up to 0.63%–13.66% of ∑PBDEs in chicken tissues (Fig. 2). It is well known that three technical PBDE formulations (penta, octa, and deca-BDE) were produced in large volumes in the world. Penta- and octa-BDE mixtures were the major commercial PBDE formulation before they were banned in some countries about ten years ago. However, deca-BDE mixture (comprising 65%–70% of total production) is still produced and used. This might result in that the pattern of PBDEs in some situation shifted to higher brominated congeners. We suggest that low levels of BDE-209 in terrestrial birds in previous studies (Voorspoels et al., 2006) might be due to low exposure of BDE-209 but not to poor accumulation of BDE-209.

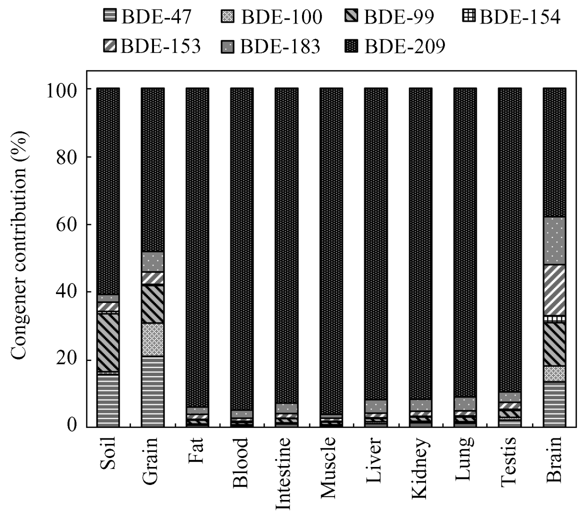

Fig. 2 Congener patterns of polybrominated diphenyl ethers in soil, grain as chicken food and various tissues of chickens (n = 4) from an e-wastes recycling area in Southeast China.

In brain samples, BDE-209 contributed up to 34.73% of the total PBDEs, which was lower than those (82.46%–94.70%) in other tissues. Correspondingly, the percentages of lower brominated congeners such as BDE-47 and BDE-99 in brain were slightly higher than those in other tissues. Therefore, we suggest that the brain might be protected more effectually from accumulation of the higher brominated congeners (BDE-209) than the lower brominated congeners, such as BDE-47 and BDE-99.

2.3 Dietary exposure risk of PBDEs

The EDI was calculated based on PBDEs levels in chicken muscle and egg samples (Table 2), which ranged from 2.21 to 22.48 ng/(day·kg body weight (bw)). The mean estimated daily intake (MEDI) value (13.51 ng/(day·kg bw)) in the present study was one order of magnitude higher than that (1.1 ng/(day·kg bw), summation of 16 PBDE congeners) from intakes of chicken and duck consumption in a recent survey for consumed free-range domestic fowl collected from an e-waste recycling site in Guangdong Province, South China (Luo et al., 2009). In a previous study on EDI of PBDEs, the calculated exposure levels ranged from 35 to 97 ng/day (0.6–1.6 ng/(day·kg bw)) based on the average daily food consumption in Belgium and varied geographically (Voorspoels et al.,

Table 2 Levels of individual PBDE congeners and total PBDEs (ng/g wet weight) in chicken muscle (n = 4) and eggs (n = 15) from an e-wastes recycling area in Southeast China and the mean estimated daily intake of PBDEs through muscle and egg consumption by a 60 kg body weight person

PBDE congener	Muscle		Egg		MEDI (ng/(day·kg bw))	MinI (ng/(day·kg bw))	MaxI (ng/(day·kg bw))
	Mean	SD	Mean	SD			
BDE-47	0.09	0.10	6.05	5.59	1.81	0.29	3.02
BDE-100	0.02	0.01	9.59	13.45	2.82	< 0.01	5.63
BDE-99	0.07	0.06	2.26	2.47	0.69	0.04	1.23
BDE-154	0.01	< 0.01	0.05	0.04	0.02	0.01	0.03
BDE-153	0.05	0.02	1.90	2.55	0.58	0.01	1.14
BDE-183	0.10	0.14	1.06	0.48	0.35	0.02	0.50
BDE-209	5.56	2.30	17.40	17.81	7.24	1.84	10.93
Total	5.90	2.54	38.31	42.31	13.50	2.21	22.48

SD: standard deviation.
MEDI: mean estimated daily intake, which is calculated from the mean concentrations of PBDEs in chicken muscle and eggs; MinI and MaxI: estimated minimum and maximum daily intake, which are calculated from the minimum and maximum concentrations, respectively, of PBDEs in chicken muscle and eggs.

2007b), which was also lower than that estimated in the present study.

It should be mentioned that in some previous studies the data calculated for dietary PBDE intake did not include the high brominated congeners (e.g., BDE-209), which might underestimate the total PBDEs exposure. In the present study, the MEDI value of BDE-209 from chicken muscle and eggs was 7.24 ng/(day·kg bw). The EDI maximum value of BDE-209 from chicken muscle and eggs was 10.93 ng/(day·kg bw) in this e-wastes recycling area. Therefore, the exposure risk of BDE-209 is of great concern for residents in this e-wastes recycling area.

3 Conclusions

The concentrations of PBDEs in chickens and eggs from this e-waste recycling process area were 2–3 orders of magnitude higher than those reported in the literature, suggesting PBDEs pollution was serious in this area. PBDEs daily dietary intake through chicken muscle and eggs consumption for residents in this area was one order of magnitude higher than the values reported in pervious studies. The large difference in PBDEs concentrations between different tissues suggested that distribution of PBDEs in tissues might depend on tissue-specificity rather than the “lipid-compartment”, which mean uniform distribution in lipid. BDE-209 was the predominant congener in all chicken tissues, which indicated that BDE-209 was major pollutant source in this area and can be bioaccumulated in terrestrial animals. In addition, low PBDEs levels in brain relative to other tissues suggest that the blood-brain barrier might protect the brain from accumulation of PBDEs, and the brain might be protected more effectually from accumulation of the higher brominated congeners than the lower brominated congeners.

Acknowledgments

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