Original Article

Assessment of Exposure to Polybrominated Dipheny Ethers via Inhalation and Diet in China*

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Abstract

Objective This paper is to assess the current status of polybrominated diphenyl ethers (PBDEs) contamination in the environment in China and estimate the exposure to PBDEs in non-occupational populations.

Methods A total of 80 research papers published from January 2001 to October 2013 were selected. Geographic information system (GIS) was used in mapping PBDE concentrations and distributions in environmental media. Ni's model was applied to calculate ∑PBDE-intake via the intakes of contaminated food, water and air in the Pearl River Delta and Yangtze River Delta.

Results BDE-209 was found to be the major PBDE congener in the environmental media and food in China. PBDE concentrations varied among different areas, among which the contamination in Guangdong Province was most serious. Daily intake of ∑PBDEs was 225.1-446.0 ng/d for adults in the Pearl River Delta, which was higher than the intake for those living in the Yangtze River Delta (148.9-369.8 ng/d).

Conclusion PBDEs are ubiquitous in the environment of China. The estimated PBDEs daily dietary intake is comparable with that in European countries.

Key words: PBDEs; Exposure assessment; GIS; PBDE-distribution; Daily intake

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INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a group of brominated flame retardants (BFRs) commonly used in consumer products such as electronics, textiles and polyurethane foams[1]. PBDEs have been marketed as three commercial mixtures: pentabromodiphenyl ether (penta-BDE), octabromodiphenyl ether (octa-BDE), and decabromodiphenyl ether (deca-BDE). The global demand for PBDEs was estimated to be 67,490 tons in 2001 (11%, 6%, and 83% were produced as penta-, octa-, and deca-BDE, respectively), of which 49% was used in North America, 37% in Asia, and 12% in Europe[2]. Among the PBDEs used in China, BDE-47, -99, -209 are the most common congeners. The multiple exposure sources and wide use of PBDE have aroused great concern on PBDE-exposure on people's health, especially the exposures to BDE-47, -99, -209, in China.

PBDEs have been shown to have toxicological effects, including developmental neurotoxicity and

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Biographical note of the first author: CHEN Li, female, born in 1987, master, with major in health-based risk assessment.

interference with thyroid hormone homeostasis, in laboratory animals[3-5]. Due to these effects, penta- and octa-BDE have been banned in the European Union since 2004, and were classified as persistent organic pollutants (POPs) due to their bioaccumulation, toxicity, and persistence, and have been banned under a Stockholm convention[6]. Since these compounds are continued to be found in the environment, reservoir sources may continue to contaminate air, water, soil, sediment, and biotic organisms[7-8], all of which are believed to contribute substantially to the human intake of these compounds. Hites[9] found that the PBDE concentrations in people had increased exponentially by a factor of approximately 100 during the last 30 years.

Although there is a wealth of data on PBDE distributions in environmental media, including surface water, air, soil, and sediment, and in the food chain, until now no nationwide systematic monitoring and tracking system for PBDE distribution has been established and no nationwide assessment of PBDE-exposure has been conducted in China. Given that dozens of papers on regional PBDE concentrations have been published, the assessment of non-occupational exposure to PBDEs in humans has become possible in this study.

Geographic information system (GIS) is a powerful tool in the exposure assessment of environmental contaminants, which is not only feasible but also can provide a visualization of contamination in the environment[10]. The combination of GIS use and statistical analysis allows for better understanding of the distribution of the contaminants in the environment of study population.

GIS was used to visualize the published data reviewed in this study in order to evaluate PBDE concentrations and distributions in environmental media. Ni's equations[11] were used to determine the major routes of PBDE-exposure, and to identify the geographical areas with high PBDE exposure levels. The results might facilitate further health-based risk assessments of PBDEs and might be helpful for the decision making on the management and control of PBDEs in China.

METHODS

Data Sources and Research Methods

A literature retrieval was carried out by using MEDLINE. The MeSH terms 'PBDEs', 'environmental exposure', 'air', 'water', 'soil', 'sediment', 'food', 'geographic information system', and 'China' were used. We also retrieved the references listed in the related published papers and reviews included in PubMed and China National Knowledge Infrastructures (CNKI). For this analysis, we selected papers which: a) were original ecological studies, b) were published in English or Chinese, c) defined all or subgroups of PBDEs as the target chemicals, and d) studied the environmental or human exposure status by using measured concentrations of PBDEs. Moreover, we retrieved conference proceedings in the ISI Web of Science for abstracts of other unpublished studies, using the same MeSH terms mentioned above, but we did not find any other related study.

A total of 184 papers were selected for this analysis, but only 80 papers qualified were used in this assessment. These papers were published from January 2001 to October 2013, and reported PBDE concentrations in environmental media and food in China.

GIS Mapping of the Data

For all the 80 studies, the sample sites could be indicated on the map precisely by using GOOGLE EARTH® via their longitudes and latitudes. By using the ArcView GIS 3.2 software, the sampling sites were geocoded. Boundaries of municipalities and provinces were obtained from the State Bureau of Surveying and Mapping. All the data were entered into ArcView GIS 3.2 operated on a PC workstation, and the data were incorporated into several maps to indicate the PBDE distributions in different sampling sites. PBDE concentration data in different environmental media were shown as separate map layers, with levels of PBDE concentrations being color coded.

Regional Exposure Assessment of PBDEs in China

Given the availability of the environmental and food concentrations of PBDEs in the Yangtze River Delta and Pearl River Delta, the exposure levels in these two regions can be estimated. According to Ni et al.[11], the daily intake levels of PBDEs in humans were estimated according to the collective environmental and food data by using the following formula: $I=\sum(C_i \times IR_i)$.

Where I is the total intake of PBDEs (ng/d); C_i is the concentration of PBDEs in the environmental medium and in various foods (ng/m^3 for air; ng/L for drinking water; ng/g for foods); and IR_i is the intake rate from the environmental medium and various foods (m^3/d for air; L/d for drinking water; g/d for foods).

Data Analysis

The normality of the distribution was tested by using a nonparametric test (Kolmogorov-Smironov Z). Geometric mean (GM) was used to reflect the average PBDE-concentration if log-transformed data was normal, otherwise a median value was used. Statistical analysis was conducted with the software SAS 9.1.

RESULTS

Geographic Distributions and Environmental Concentrations of PBDEs Contamination in China

PBDEs exist in both gaseous and solid phases in air. Due to frequent contact of brominated fire retardants in daily life, the important pathway of human exposure to PBDEs is through inhalation of air. Figure 1 shows BDE-47, -99, -209 (particle plus gas phase) concentrations in the 20 sampling sites. The concentration of BDE-47 ranged from 0.04 to 4,446.5 pg/m^3 with the GM of 49.37 pg/m^3. Higher BDE-47 contamination in air was found in Guiyu in Guangdong Province. The concentration of BDE-99 in Guiyu was also in a high level (3,283.85 pg/m^3). The GM of BDE-99 concentrations in air was 45.36 pg/m^3. Air concentrations of BDE-209 were higher than other PBDEs in China, ranging from 37.1 to 7,161.5 pg/m^3 and with the GM of 544.25 pg/m^3.

Surface water has been sampled for PBDE-detection in China, and in total there were 42 sampling sites. PBDEs exist in both aqueous phase and suspended particle phases in water. During water treatment processes, most of the particles can be effectively removed. Therefore, PBDEs in aqueous phases was more focused in this assessment.

Figure 2 shows BDE-47, -99, -209 concentrations in different rivers, mainly in the Yangtze River and the Pearl River. The BDE-47 concentrations in surface water varied from 0.68 to 10,700 pg/L, with the GM of 55.11 pg/L. The concentrations of BDE-99 were lower than that of BDE-47, ranging from 0.01 to 3,220 pg/L) and with the GM of 28.66 pg/L. BDE-209 was found to have high concentrations in water, ranging from 0.36 to 18,000 pg/L and with the GM of 1,946.79 pg/L.

PBDEs are substances that hardly decompose and easily accumulate in soil, resulting in the high contamination of PBDEs in China (Figure 3). In the 44 sampling sites, the Yellow River Delta was found to have the lowest BDE-47 concentration of 0.003 ng/g. The concentration of BDE-47 in Hong Kong was high (2,287 ng/g). The GM of BDE-47 concentrations in soil was 1.30 ng/g. The lowest level of BDE-99 (0.002 ng/g) was detected in one sample site at the Yellow River Delta. The GM of BDE-99 concentrations was 1.14 ng/g, and an e-waste site in Hong Kong was reported to have the highest BDE-99 concentration (1,410 ng/g) in soil. BDE-209 concentrations ranged from 0.18 to 18,519.6 ng/g, with GM of 47.66 ng/g.

Studies on sediment PBDEs were productive, and a total of 195 sampling sites were found with sediment PBDE concentration data (Figure 4). The BDE-47 concentrations ranged from 0.004 to 3,054 ng/g (GM: 0.23 ng/g). And the BDE-99 concentrations ranged from 0.002 to 2,827 ng/g (GM: 0.25 ng/g). The concentrations of BDE-209 in sediment were also high, ranging from 0.02 to 7,341 ng/g and with the GM of 8.22 ng/g.

Estimated PBDEs Daily Dietary Intake

Due to the characteristics of lipophilicity, bioaccumulation and biomagnifications, PBDEs can accumulate in the food chain and in foodstuffs (i.e. fish from high trophic levels or with lipid-rich tissues)[11]. In this study, most of the data on PBDE concentrations in food came from eastern or southern China, such as Shanghai, Shenzhen, and Hong Kong, etc. In this study we used these data to assess the daily dietary exposure to PBDEs of residents living in areas of the Yangtze River Delta and the Pearl River Delta. Together with the Chinese Diet Guidelines from the Chinese Nutrition Society[12], PBDE-exposure via food in the areas mentioned above was calculated.

Results showed that PBDE concentrations varied with the type of food (Table 1). Both in the Pearl River Delta and the Yangtze River Delta, fish and seafood were shown to be the main dietary source for human exposure to PBDEs. In addition, lipid-rich foods such as meat, eggs, and dairy products contributed significantly to human exposure to PBDEs.

As shown in Table 1, in the Pearl River Delta, daily average PBDE-intakes from food were estimated to be 95.88 and 7.86 ng/d for BDE-47 and BDE-99 and estimated to be 41.26 ng/d for BDE-47, and 3.13 ng/d for BDE-99 in the Yangtze River Delta, respectively.

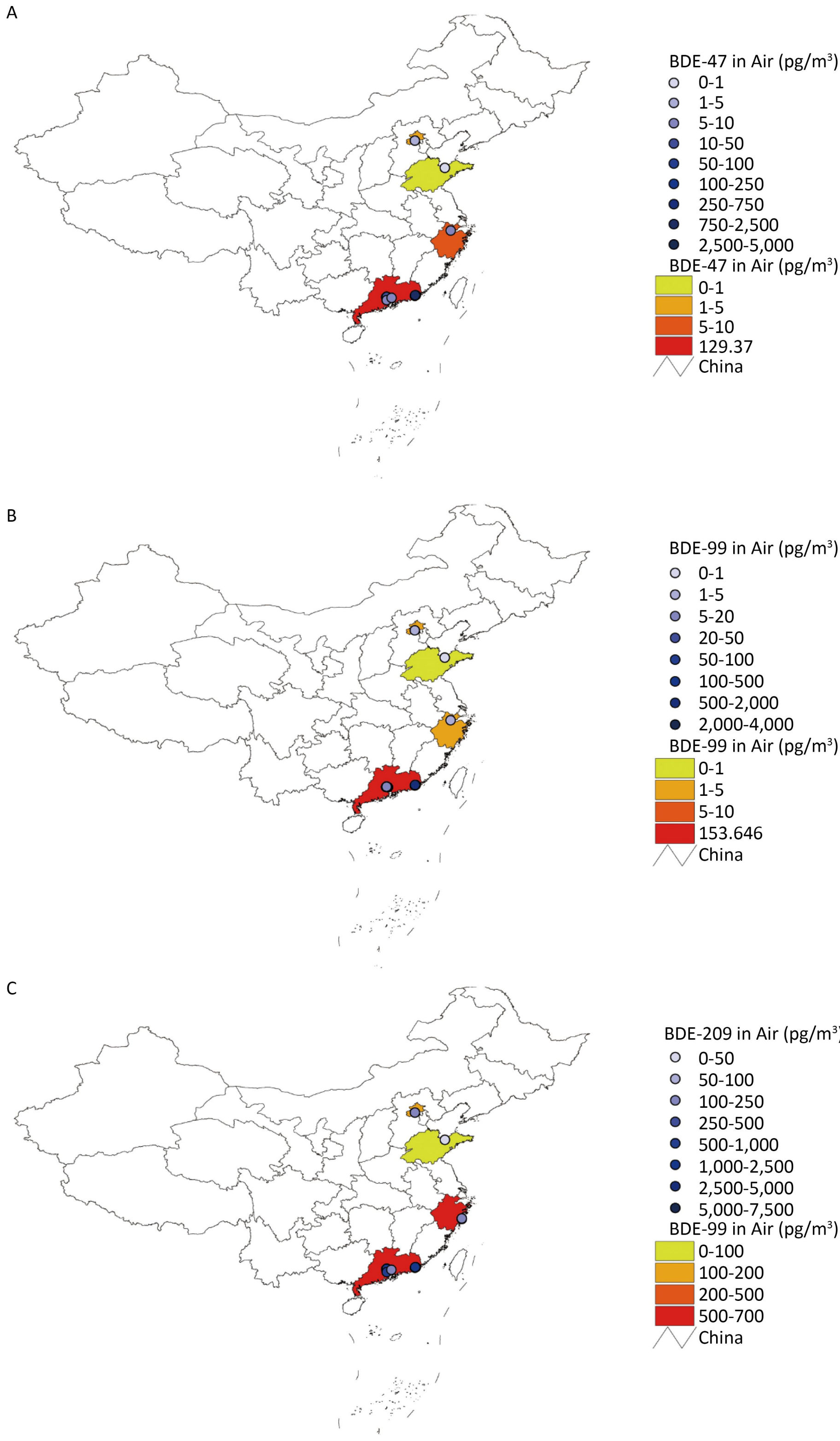

Figure 1. Geographic distributions and concentrations of PBDEs contamination in air. (A) BDE-47 in air; (B) BDE-99 in air; (C) BDE-209 in air.

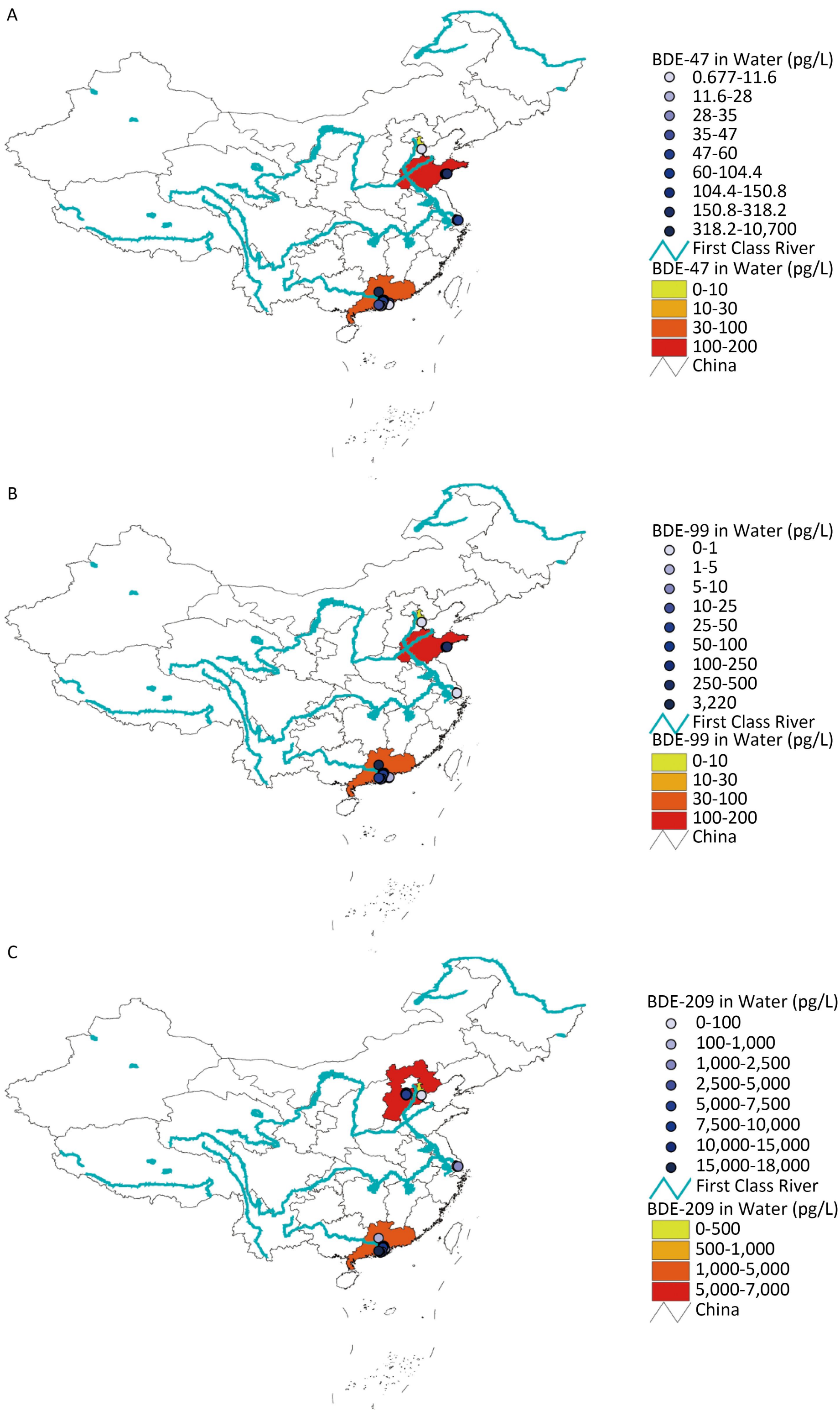

Figure 2. Geographic distributions and concentrations of PBDEs contamination in surface water. (A) BDE-47 in surface water; (B) BDE-99 in surface water; (C) BDE-209 in surface water.

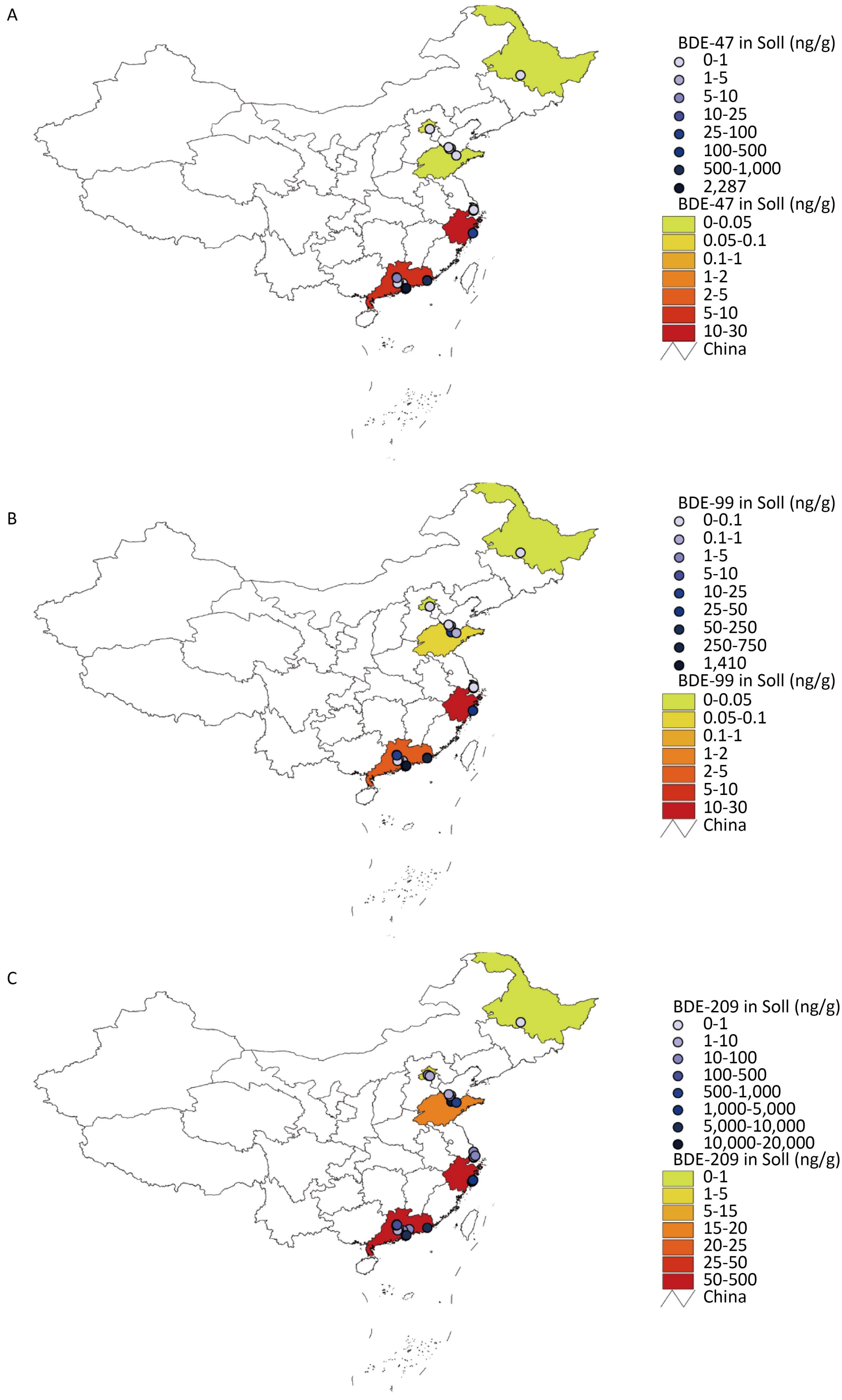

Figure 3. Geographic distributions and concentrations of PBDEs contamination in soil. (A) BDE-47 in soil; (B) BDE-99 in soil; (C) BDE-209 in soil.

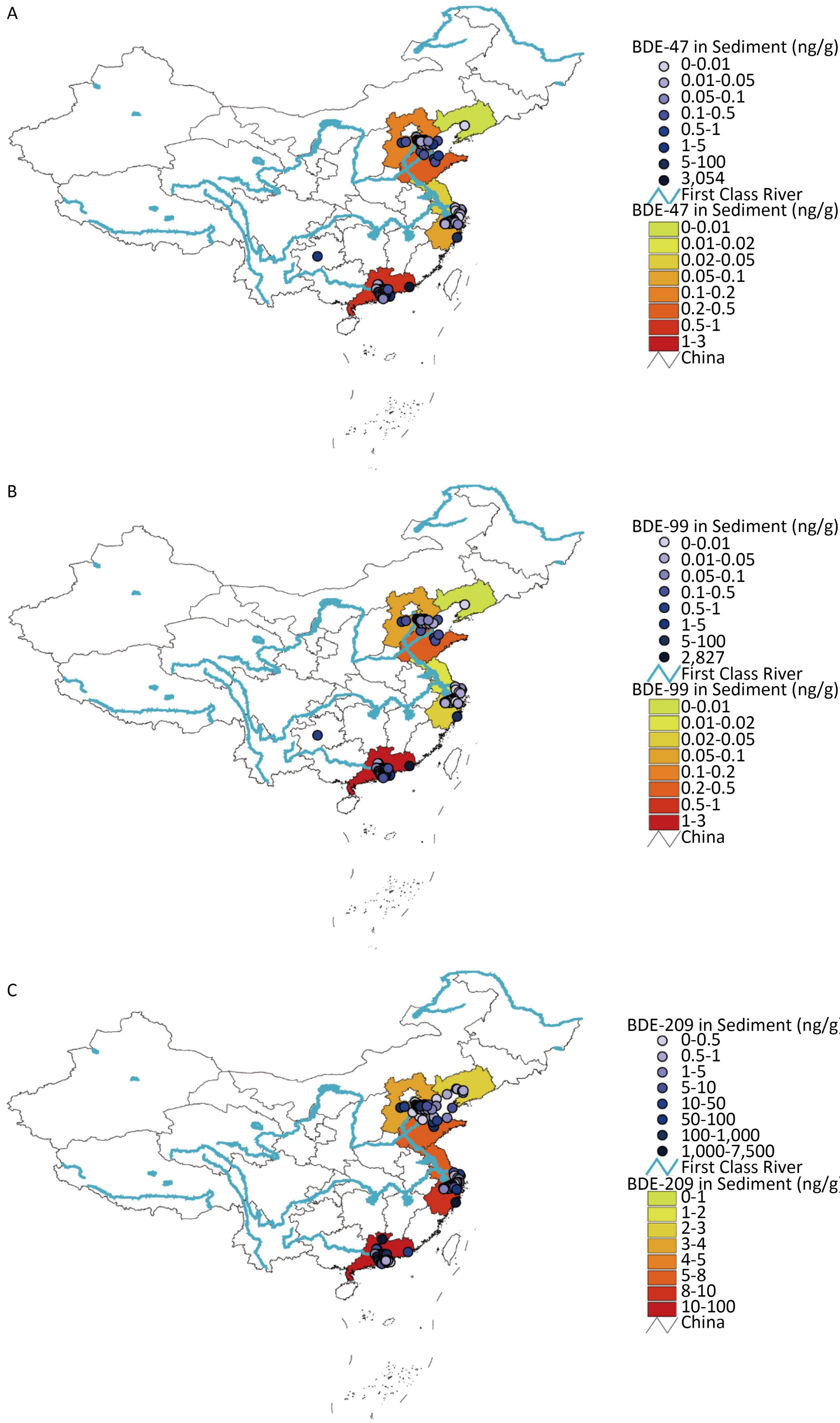

Figure 4. Geographic distributions and concentrations of PBDEs contamination in sediment. (A) BDE-47 in sediment; (B) BDE-99 in sediment; (C) BDE-209 in sediment.

In China, the concentration data on BDE-209 in food are limited, resulting in the failure to calculate dietary BDE-209 intake level in our study. To date, BDE-209 has only been measured in certain European food surveys. The UK Food Standards Agency estimated a daily BDE-209 intake of 318.6 ng/d (4.5 ng/kg BW/d, assuming a body weight of 70.8 kg[13]) for adults based on average consumption across their whole diet[14]. Moreover, Knutsen et al.[15] reported the range of BDE-209 intakes in a group of 184 Norwegians with a wide range of seafood consumption (4-455 g/d). Since we failed to figure out dietary BDE-209 intake for our Chinese samples, we used dietary BDE-209 intakes of 318.6 ng/d (UK) and 97.7 ng/g (1.38 ng/kg BW/d, assuming a body weight of 70.8 kg[13]) (Norway) for our calculations.

Assessment of Human Exposure to PBDEs

The exposure of PBDEs levels in the Pearl River Delta and the Yangtze River Delta were evaluated by using Ni's models. Table 2 shows the estimated levels of PBDE-exposure via different exposure routes in these two areas. Human exposure levels to the total PBDEs (∑PBDEs: the sum of congeners of BDE-47, BDE-99, and BDE-209) were 225.1-446.0 ng/d in the Pearl River Delta and 148.9-369.8 ng/d in the Yangtze River Delta. Considering the average body weight of Chinese adults (62.7 kg)[16], the intake levels of ∑PBDEs were estimated to be 3.6-7.1 ng/kg BW/d for adults living in the Pearl River Delta, and 2.4-5.9 ng/kg BW/d for adults living in the Yangtze River Delta, respectively. Dietary exposure was found to be the predominant exposure route in the Chinese adult population, accounting for >90% of total PBDE exposures. In the Pearl River Delta, the estimated exposure levels of BDE-47, BDE-99, and BDE-209 were 2.4-fold, 3.4-fold, and 1.2-fold higher than those in the Yangtze River Delta, respectively.

DISCUSSION

Human exposure to PBDEs has been of great concern, as it has become evident that PBDEs are ubiquitously found in human tissues[9,17-19]. There were several studies on human exposure to PBDEs worldwide. However, in China, most studies focused on specific exposure sources[20], and little is known about the PBDE-exposure through multiple pathways. This study provided comprehensive information on the current contamination status of PBDEs in China and discussed the potential sources of human exposure to PBDEs. Based on these sources, the first comprehensive assessment of PBDE-exposure in the non-occupational population of China was conducted.

Table 1. Estimated PBDEs Daily Dietary Intakes in the Pearl River Delta and Yangtze River Delta (ng/d)

PBDEs	Food	Concentrations (ng/g)		Intake Rate (g/d)	Intake of PBDEs (ng/d)	
		Pearl River Delta	Yangtze River Delta		Pearl River Delta	Yangtze River Delta
BDE-47	Oil	0.017	/	27.5	0.470	/
	Dairy	0.016	/	300	4.751	/
	Bean/Nut	0.030	0.042	40	1.180	1.685
	Meat	0.035	0.055	62.5	2.202	3.446
	Aquatic	1.121	0.160	75	84.072	11.982
	Egg	0.038	0.088	37.5	1.442	3.314
	Vegetable	0.001	0.008	700	0.798	5.662
	Grain	0.003	0.047	325	0.961	15.172
	Total	/	/	/	95.88	41.26
BDE-99	Oil	0.011	/	27.5	0.307	/
	Dairy	/	/	300	/	/
	Bean/Nut	0.0104	0.013	40	0.416	0.523
	Meat	0.011	0.008	62.5	0.711	0.476
	Aquatic	0.059	0.010	75	4.456	0.777
	Egg	0.041	/	37.5	1.519	/
	Vegetable	0.0004	0.002	700	0.277	1.354
	Grain	0.001	/	325	0.168	/
	Total	/	/	/	7.86	3.13
BDE-209	Total	/	/	/	97.7-318.6[14-15]	97.7-318.6[14-15]

In the environmental media contaminated with PBDEs, BDE-209 congener was found to have the highest concentration. The remarkable levels and dominance of BDE-209 may be related with the significant production, usage, or disposal of deca-containing products in China. BDE-209 is the only commercial PBDE mixture still permitted currently[21], which is contained in more than 95% of the commercial deca-BDE products[22]. Moreover, 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[23]. Our results were also consistent with those of the European Food Safety Authority (EFSA) Panel on Contaminants in the Food Chain, which revealed BDE-209 and BDE-47 to be the most common dietary contaminants[24].

Since dioxins' and similar persistent organic pollutants' (POPs) routes of entry into the general population is almost exclusively from food, it was also hypothesized that the predominant route of human exposure to PBDEs would be through dietary intake[25]. Consistent with existing literature, dietary exposure was found in our study to be a significant route of human PBDE-intake and was largely attributed to consumption of aquatic foods. This result was supported by studies conducted in the U.K. and Canada, where the dietary exposure accounted for 93% and 96% of the total exposures, respectively[26-27]. Studies in Finland, Sweden and Belgium also identified fish consumption to be the major route for human exposure to PBDEs[28-30].

Until now, the data concerning PBDE concentrations in food, as well as human exposure to PBDEs through the diet has been available. However, studies on PBDE-exposure via multiple routes are rare. Thus, we compared PBDE exposure data via ingestion in other countries to understand the PBDE-exposure in China. ∑PBDEs exposure level via ingestion in Spain was reported to be 97.3 ng/d[27], while in Japan and North American the ∑PBDEs exposure levels were 72 and 52-88 ng/d among the populations respectively[31-32]. Schuhmacher et al.[33] also reported the ∑PBDEs exposure levels in Spain, which were 72 ng/d for adults living in industrial area and 63 ng/d for adults in urban district. Taking into consideration of exposure via ingestion and inhalation, the ∑PBDEs exposure level in British people was 127 ng/d[28]. All of these data were lower than the estimated intake of ∑PBDEs in Chinese, especially for people living in the Pearl River Delta, which might be due to the high concentrations of PBDEs in food and soil in China.

The oral reference dose (RfD) for the deca-BDE homolog (e.g. BDE-209), tetrabromodiphenyl ether (tetra-BDE) homolog (e.g. BDE-47), and penta-BDE homolog (e.g. BDE-99) released by U.S. Environmental Protection Agency (EPA) are 7×10^{-3}, 1×10^{-4}, and 2×10^{-3} mg/kg BW/d, respectively[34-36]. In this study, the estimated intake levels of ∑PBDEs were 2.4-5.9 ng/kg BW/d in the Yangtze River Delta and 3.6-7.1 ng/kg BW/d in the Pearl River Delta,

Table 2. PBDE Exposure Levels in the Yangtze River Delta and Pearl River Delta

PBDEs	Environmental Media and Food	Intake Rate[a]	Pearl River Delta		Yangtze River Delta	
			C_i[b]	I[c]	C_i[b]	I[c]
BDE-47	Air	19[16]	0.129	2.45	0.001	0.02
	Water	1.7[16]	0.029	0.05	0.055	0.09
	Food	/	/	95.88[d]	/	41.26[d]
	Total	/	/	98.38	/	41.37
BDE-99	Air	19[16]	0.153	2.91	0.002	0.04
	Water	1.7[16]	0.024	0.04	0.031	0.05
	Food	/	/	7.86[d]	/	3.13[d]
	Total	/	/	10.81	/	3.22
BDE-209	Air	19[16]	0.685	13.02	0.327	6.21
	Water	1.7[16]	3.033	5.16	0.258	0.44
	Food	/	/	97.7-318.6[d]	/	97.7-318.6[d]
	Total	/	/	115.9-336.8	/	104.4-325.3
∑PBDEs	Total	/	/	225.1-446.0	/	148.9-369.8

Note. [a]Units of intake rate were m^3/d in air and L/d in water, respectively; [b]Units of PBDE concentrations were ng/m^3 in air and ng/L in water, respectively; [c]Unit of PBDEs intake is ng/d; [d]Intake of PBDEs via food consumption was showed in Table 1.

respectively, which were lower than the EPA RfD value, indicating that the environmental PBDE-exposure might not induce significant health hazards in Chinese adults.

While this study collected a wealth of PBDE concentration data in several environmental matrices, and conducted the first comprehensive assessment of PBDE exposure in human in China, there were still some limitations in this analysis. Firstly, the data used in our study were all from published papers. Though we contacted the original authors when necessary, there still might be some modified factors beyond our control, e.g. publication bias. Secondly, as published data on PBDE levels in food are very limited, there might be some uncertainties while assessing PBDE-intake in different areas.

CONCLUSION

PBDEs are ubiquitous in the environmental media in China. The PBDE concentrations varied in geographical patterns, and Guangdong Province was the most serious contaminated area. BDE-209 was the major PBDE congener in the environmental media and food, and BDE-47 also had a high level in food. Daily intake levels of ∑PBDEs were 148.9-369.8 and 225.1-446.0 ng/d for adults in the Yangtze River Delta and the Pearl River Delta, respectively. Moreover, dietary intake is the major route of PBDE-exposure. Decision-makers should take the potential health effects of PBDEs on people into consideration, especially on the people in areas with a high PBDE concentration, such as the Pearl River Delta. Furthermore, a comprehensive risk assessment for Chinese (including the sensitive population) exposed to PBDEs is needed in future research.

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