Polybrominated diphenyl ethers in breast milk from Beijing, China

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Introduction

Polybrominated diphenyl ethers (PBDEs) are extensively used in electronic appliances, paints, textiles and furnishings as flame retardants. PBDEs are now ubiquitous pollutants due to large production and their persistence. The results of some research indicate that PBDEs might act as potential endocrine disrupters and interfere with brain development, thus they may pose potential environmental and human health problem. In the last decade human exposure to PBDEs has been documented worldwide particularly in developed countries such as Sweden (Fängström et al.2006), USA (Schecter et al. 2003), Canada (Ryan et al. 2006), Japan (Takasuga et al. 2006) and France (Antignac et al 2006).In China, due to the rapid developing of the manufacture industry, production and usage of brominated flame retardants (BFR) including PBDEs have increased considerably. However, until now, few data on levels of human exposure PBDEs in China was reported.

Usually, breast milk was used to assess the extent of human exposure to PBDEs. In this study, breast milk samples from Beijing were analyzed for 6 predominant PBDE congeners (BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183).

Materials and Methods

Sample collection and preparation

During January 2005, breast milk samples were collected from 127 healthy volunteers (20-30 per district) living in 6 different districts of Beijing including shijingshan district, tongzhou district, haidian district, daxing district, yanqing district and pinggu district as showed in Figure 1..Occupation as well as age, place and length of local residence, diet habit smoking habits and the information of infants such as birth weight and sex were recorded in a questionnaire when collecting samples. The questionnaire showed no occupationally exposure in these mothers. All samples were stored in a refrigerator at -20℃ until analysis. Before analysis, 8－10 individual samples from same district were mixed to one pooled sample . Finally, 15 pooled samples were analysis for PBDEs.

Chemicals

Six 13C-labeled internal standards (BDE47, BDE99, BDE100, BDE153, BDE154 and BDE183) and two 13C-labeled injection standards (PCB52 and PCB138) were purchase from Cambridge Isotope Laboratories. All solvent used in analysis were of pesticide grade.

Extraction and Clean-up

Pooled samples were dried in a freeze-dryer before blending with some anhydrous sodium sulfate. After spiking with known amounts of 6 13C-labeled internal standards samples were Soxhlet-extracted with a mixture n-hexane/acetone (1:1, 250ml). Gravimetric lipid determination was performed after solvent evaporation. Bulk lipid removal was achieved by shaking with acid-modified silica-gel and further clean-up was achieved using a Power Prep instrument (Fluid Management Systems, Waltham, MA, USA). The final extract was

concentrated to 50 ul. Before analysis the 13C-labeled injection standard were added.

Instruments analysis

The identification and quantification was performed by high resolution gas chromatograph coupling to high resolution mass spectrometry (MAT 95XP, ThermoFinnigan, Germany) with a DB-5MS capillary column (30m x 0.25mm i.d. x o.1um film thickness, Agilent, USA). The two most abundant masses were used for measurement and concentration of 6 PBDE congeners were quantified using isotope dilution method.

Quality control

Method blank samples were performed every 5 samples. The average amount in blank samples was subtracted from the amount in the samples. The 13C-labeled internal standard recoveries of all samples were in the criteria range established by the USEPA (EPA 1614 draft). The laboratory performance was validated by successfully participating in an inter-laboratory comparison study of PBDEs in breast milk organized by the Norway Institute of Public Health in 2006.

Results and Discussion

The concentrations of ∑PBDE in 15 pooled samples from 6 districts are listed in table 1. the range for the ∑PBDE varied from 683.77 to 3068 pg/g lipid, with a mean of 1175.0 pg/g lipid and a median of 1080.4 pg/g lipid. Table 1 further showed the average concentration of ∑PBDE for every district. The average value from pinggu district (752pg/g lipid) is the minimum among all the districts. The pinggu district is a rural area which is most far away from the center of the city and there is more volunteers whose occupation is farmer, which seem to be the reason for its low value. The maximum average value was from haidian district and the sample (BM7) with maximum value of ∑PBDE is in this district. The PBDEs profile of BM7 is apparently different from other samples, which will be discussed below.

The congener profiles of PBDEs in all samples are showed in figure 2. Among the all congeners BDE47 and BDE153 were the most abundant except BM7. The contribution of BDE47 to ∑PBDE varied from 37.3% to 58.0 with a mean of 45.8% for all sample except BM7 in which BDE47 accounted only for 27.7%. The contribution of BDE153 to ∑PBDE varied from 19.9% to 35.3% with a mean of 25.8%. The congener profile of sample BM7 from haidian district is obviously different from other samples. In BM7, BDE 183 accounted for 26.2% of ∑PBDE that almost is the same to BDE47. And contribution of BDE154 (11.7%) is more than that of BDE99 (6.7%) while other samples' percentages of BDE154 were very low varying from 1.6% to 4.1%. The high level of high brominated congeners implied that a certain mother or more in that pool seemed to expose to commercial product of PBDEs.

This is the first study of PBDEs in breast milk from Beijing, therefore the temporal trend can not be deduced. Generally, the concentrations of ∑PBDE from this study are less than that from Guangzhou, locating in the South of China, where the concentration of ∑PBDE containing 6 predominant congeners varied from 1440pg/g lipid to 6270pg/g lipid from 27 individual samples (Bi et al. 2006). The level of PBDEs from this study is much less than that in North America (Schecter et al. 2003; Ryan et al. 2006) and some European countries

(Fängström et al.2006; Antignac et al 2006). The PBDEs pattern of this study is similar to that of South China (Bi et al. 2006) and Sweden (Fängström et al. 2006). In these studies BDE 47 and BDE153 account to more than 60% of ∑PBDE while in the U.S. breast milk low brominated congeners BDE99 and BDE100 contributed much more than BDE 153, which may result from more usage of commercial product Penta-PBDE in U.S.

The level of PBDE in breast milk is low now in China. However, the domestic demand of BFR has increased at a rate of 8% per year. There is a potential for higher levels of contamination in the future. Therefore, extended and continual monitoring of the human exposure to PBDE is needed to assess the health risk from PBDE in China.

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Table 1 The concentration of ∑PBDE of pooled samples (pg/g lipid)

District	No.	Con.	Ave.	District	No.	Con.	Ave.
Shijingshan	BM1	1201.9	1232.3	Daxing	BM10	1200.3	1029.7
	BM2	1366.2			BM11	859.1	
	BM3	1129					
Tongzhou	BM4	870.7	959.4	Yanqing	BM12	1253.3	1134.6
	BM5	918.5			BM13	1015.3	
	BM6	1088.9					
Haidian	BM7	3068	1738.9	Pinggu	BM14	683.77	752
	BM8	1067			BM15	820.21	
	BM9	1080					

Figure 1 Districts in this study
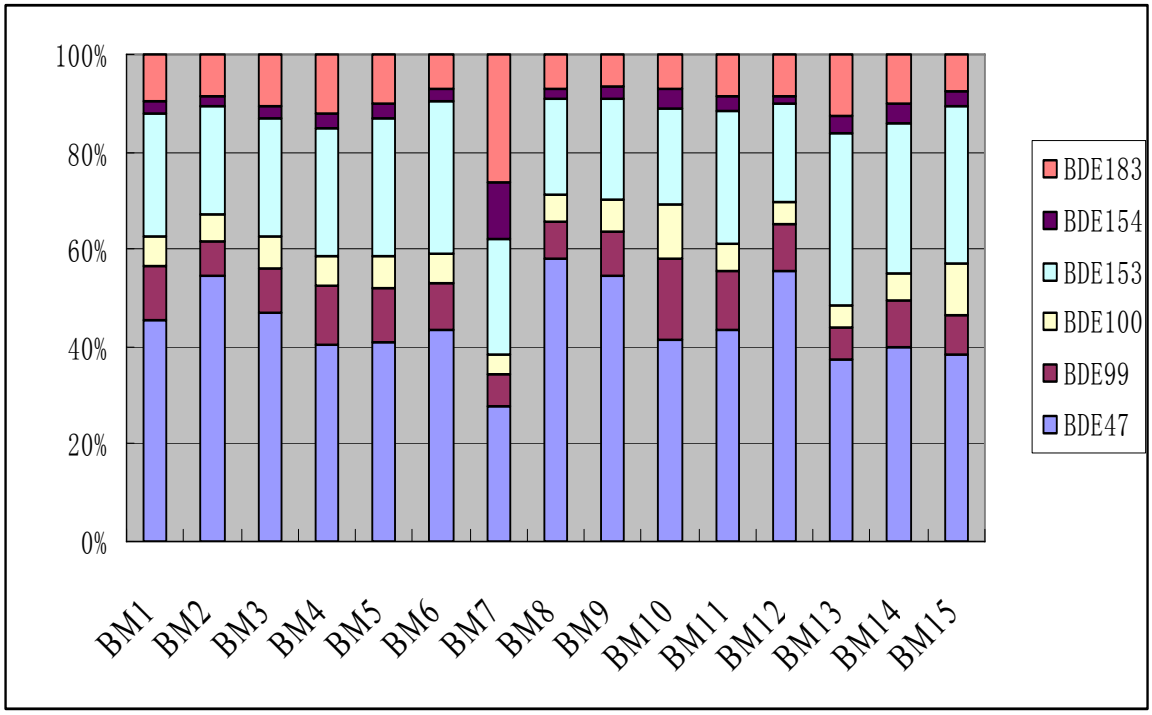
Figure 2 Congener profiles of samples