INFANT EXPOSURE TO POLYBROMINATED DIPHENYL ETHERS (PBDEs) VIA CONSUMPTION OF HOMEMADE BABY FOOD IN KOREA

Yunsun Jeong[1]*, Sunggyu Lee[1], Sunmi Kim[2], Sung-Deuk Choi[3], Jeongim Park[4], Kyungho Choi[2], Sungkyoon Kim[2], Hyo-Bang Moon[1]

[1]Department of Marine Sciences and Convergent Technology, Hanyang University, Ansan 426-791, Republic of Korea; [2]School of Public Health, Seoul National University, Seoul, Republic of Korea; [3]School of Urban and Environmental Engineering, Ulsan National Institute of Science and Technology, Ulsan, Republic of Korea; [4]Colleage of Natural Sciences, Soonchunhyang University, Asan, Republic of Korea

Introduction

Polybrominated diphenyl ethers (PBDEs) are primarily used for brominated flame retardants (BFRs), which are used in a variety of consumer products such as textiles, electronic and electrical equipment, and other materials [1,2]. Due to the persistence, bioaccumulation, and adverse health effects of PBDEs, these contaminants have received considerable concerns during the past two decades. The primary exposure pathways of PBDEs for the general population are diet, dust ingestion and inhalation[3,4,5,6]. In particular, certain subgroups of populations may have higher risks from exposure to PBDEs than the general population[6,7]. Infants are simultaneously exposed to PBDEs through breast milk, house dust and diet such as baby food[7]. Korean women have traditionally used homemade baby food to supplement breast[9]. A previous study reported that organochlorine pesticides (OCPs) such as chlordanes and heptachlor epoxide posed potential health risks from simultaneous exposures by breast milk and homemade baby food to Korean infants[10]. Therefore, it is essential to determine PBDEs in baby food and to accurately assess the PBDE exposure with other exposure pathways, such as breast milk and house dust. However, the importance of baby food consumption to PBDE exposure for infants has not been well characterized. Homemade baby food could be a major source of PBDEs exposure for Korean infants, because the breast milk consumption rate is lower for Korean infants than infants in North America and EU countries[9,11]. To date, no reports are available on the occurrence and exposure of PBDEs in baby food. The objectives of this study were to investigate the occurrence, concentrations and compositional profiles of PBDEs in homemade baby food and to assess the daily intake and potential health risks of these contaminants for Korean infants. In addition, the contribution of baby food to the total exposure of Korean infants to PBDEs was assessed with multiple sources of PBDEs such as breast milk and dust ingestion.

Materials and methods

Sample collection

The homemade baby food samples (*n*=147) were collected from participating mothers (*n*=97) in the Children's Health and Environmental Chemicals in Korea Panel, or CHECK Panel (Kim et al., 2013; Lee et al., 2013b, c). CHECK Panel is comprised of pregnant women-fetus pairs recruited from four cities since 2011 in Korea. In this study, the baby food samples were collected from residential (Seoul and Pyeongchon), industrial (Ansan), and rural (Jeju) regions in Korea, during the period of 2012 to 2013. All of the food items were pooled and homogenized with an ultra-disperser to obtain one sample from each household or subject group. The homogenized baby food samples were stored at –25°C until analysis. The homemade baby food samples were classified as five subject groups according to infant age; (1) 6-month-old (*n*=21), (2) 9-month-old (*n*=29), (3) 12-month-old (*n*=37), (4) 15-month-old (*n*=12), and (5) from 24 to 27-month-old (*n*=48).

Chemical and instrumental analysis

Analyses of the PBDEs in the baby food samples were performed following methods described elsewhere[6.12]. In brief, approximately 15–20 g of the baby food samples were homogenized with anhydrous Na_2SO_4 and then extracted with mixed solvents of DCM:hexane (3:1) using a Soxhlet apparatus for 16 h. Prior to extraction, 100 ng of the surrogate standards (PCBs 103, 198, and 209) were spiked into the samples. The extract was concentrated to 11 mL, and an aliquot (1 mL) was used for gravimetric determination of the lipid content. The remaining aliquot was spiked with internal standards of ^{13}C-labeled PBDEs and then cleaned by a multi-layered silica gel column with 150 mL of 15% DCM in hexane. The eluted fractions were concentrated to 1 mL and were dissolved in 100 µL of nonane for instrumental analysis. Twenty-four PBDE congeners were measured using a high resolution gas chromatography interfaced with high resolution mass spectrometry. The recoveries of the surrogate standards ranged from 85% to 92%. The recoveries of the ^{13}C-labelled BDEs ranged from 86 to 113%.

Statistical analysis
Analysis of variance (ANOVA) was used to determine significant differences in the concentrations of the PBDEs and the estimated daily intakes (EDIs) between the subject groups. The statistical significance was based on the level of $p < 0.05$. All of the statistical analyses were performed using the Statistical Package for the Social Sciences (SPSS) Version 18 for Windows (SPSS, Chicago, IL, USA).

Results and discussion
Occurrence and concentrations of PBDEs
The occurrence and concentrations of PBDE congeners in the homemade baby food samples from Korea are summarized in Table 1. The concentrations of ΣPBDE in the baby food samples ranged from 24.5 to 6000 (mean: 263) pg/g fresh weight. Considerable variation was found in the concentrations of PBDEs among the baby food samples, due to the diverse ingredients of homemade baby food samples depending on the age of subject group. Among the PBDE congeners analyzed, BDE 209 showed the highest concentrations, ranging from <10.0 to 5940 pg/g fresh weight, with a mean value of 234 pg/g fresh weight. The concentrations of BDE 47 ranged from <0.1 to 55.5 pg/g fresh weight, with a mean value of 5.92 pg/g fresh weight. The highest contribution of BDE 209 reflected in the consumption pattern of PBDE technical mixtures in Korea.

Table 1. Concentrations (pg/g fresh weight) and detection rates (%) of the PBDEs in the homemade baby food samples collected in Korea between 2012 and 2013

	Mean ± SD[a]	50th	95th	Range	Detection rate (%)
BDE 28	0.55 ± 1.32	0.07	3.33	<0.1–8.25	26
BDE 47	5.92 ± 9.77	2.05	22.14	<0.1–55.5	80
BDE 71	0.10 ± 0.21	0.07	0.07	<0.1–2.48	5.4
BDE 99	1.94 ± 5.11	0.07	11.1	<0.1–34.0	31
BDE 100	0.54 ± 2.33	0.07	1.61	<0.1–18.7	16
BDE 153	0.51 ± 1.23	0.35	0.35	<0.5–13.8	4.1
BDE 154	0.56 ± 1.63	0.35	0.35	<0.5–17.5	5.4
BDE 183	0.59 ± 1.32	0.35	0.35	<0.5–10.5	6.8
BDE 207	5.27 ± 12.3	3.54	3.54	<5.0–134	12
BDE 209	234 ± 697	10.2	1120	<10.0–5940	59
ΣPBDE[b]	263 ± 702	45.3	1160	24.5–6000	89

[a]SD= standard deviation; [b]Sum of PBDE congeners detected.

Comparison of PBDEs in baby food among infant age groups
The concentrations of BDE 47, BDE 209, and ΣPBDE were compared among each subject group (Figure 1). The mean concentrations of ΣPBDE for each subject group were 93.9, 311, 184, 108, and 407 pg/g fresh weight for 6-, 9-, 12-, 15-, and 24 to 27-month-old infant groups, respectively. Our findings seem to be due to the difference in the ingredients in baby food with an increase in the infant ages. In fact, increasing lipid contents were found in our samples with an increase in the age of the subject group.
The overall concentrations of BDE 47 in the homemade baby food samples showed a gradual increasing trend with an increase in the infant age. In particular, the concentrations of BDE 47 in the baby food for 24 to 27-month-old infants were significantly higher (ANOVA, $p < 0.05$) than those found for the other subject groups. However, no significant differences were found in the concentrations of BDE 209 and ΣPBDE, despite the highest levels of PBDEs in the oldest infant group. The detection rates of BDEs 47 and 209 in the baby food samples in the older infant ages support our findings. The detection rates of BDE 47 gradually increased with an increase in the age of the subject group, whereas BDE 209 had no increasing trend for the detection rates. Our results imply that BDE 209 has diverse contamination sources for homemade baby food.

Estimation of the daily intake of PBDEs and health risk assessment
The estimated daily intakes (EDIs) of BDE 47, BDE 209, and ΣPBDE for 6, 9, 12, 15, and 24 to 27-month-old infant groups via the consumption of homemade baby food are summarized in Table 2. The EDIs of BDE 209

for all of the subject groups contributed over 76% to the EDIs of ΣPBDE. In particular, the contribution of BDE 209 for the high exposure group was >90% of the ΣPBDE exposure. This result suggests that BDE 209 is a major PBDE congener for infant exposure via baby food consumption in Korea. The EDIs of BDEs 47, BDE 209, and ΣPBDE for the 24 to 27-month-old infants were an order of magnitude higher than the EDIs found for the 6- to 15-month-old infants, associated with a change in the ingredients in the baby food. Our results indicate that Korean infants have higher PBDE exposure via the consumption of baby food with an increase in age.

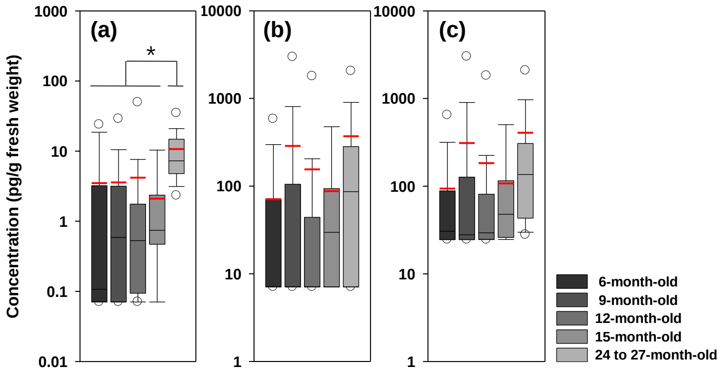

Figure 1. Comparison of concentrations of (a) BDE 47, (b) BDE 209, and (c) total PBDEs in homemade baby food samples for the infant subject groups. The box plot represents the 10th, 25th, 50th and 90th percentiles with error bars. The circles are the 5th percentile (lower) and 95th percentile (upper). The arithmetic mean concentrations are given as red bars. *Significant differences at $p < 0.05$.

Simultaneous exposure assessment of PBDEs with multiple sources

Infants are exposed to PBDEs from multiple sources including breast milk, dust ingestion, and diet, such as baby food[7]. Therefore, the simultaneous consideration of multiple sources of PBDE exposure is essential to accurately assess the contribution from each source and to establish efficient strategies for reducing PBDE exposure for infants. Together with the three exposure pathways of PBDEs to Korean infants, the total EDIs of ΣPBDE was 25.5 ng/kg bw/day. The relative contribution of the total PBDE exposure was 55%, 41%, and 4.0% for breast milk, dust and baby food, respectively. Our results suggest that baby food is an important exposure pathway for PBDEs for over 24-month-old infants. Further studies are required to investigate additional sources of PBDE exposure, such as inhalation and contact with products containing PBDEs, for more accurate exposure estimation.

Table 2. Estimated daily intakes (EDIs, ng/kg body weight/day) of BDE 47, BDE 209, and ΣPBDE and its contribution to the total EDIs via the consumption of homemade baby food in Korea

	BDE 47 (% contribution)	BDE 209 (% contribution)	ΣPBDE[a]
	Average		
6-month-old	0.04 (3.8)	0.80 (76)	1.06
9-month-old	0.06 (1.1)	4.88 (92)	5.28
12-month-old	0.09 (2.2)	3.45 (85)	4.08
15-month-old	0.06 (1.9)	2.52 (81)	3.10
24 to 27-month-old	0.58 (2.7)	20.3 (92)	22.3
	95th percentile		
6-month-old	0.23 (5.4)	3.80 (89)	4.00
9-month-old	0.20 (0.4)	47.9 (99)	48.3
12-month-old	0.54 (3.9)	12.4 (91)	12.8
15-month-old	0.24 (2.0)	11.0 (93)	11.7
24 to 27-month-old	1.39 (1.7)	75.7 (95)	77.0

[a]Sum of PBDE congeners detected.

Acknowledgments
We would like to express our gratitude to all participating mothers in the CHECK (Children's Health and Environmental Chemicals in Korea) Panel for their great efforts in collecting homemade baby food samples. This study was supported by the Korea Food and Drug Administration (KFDA, 12162KFDA732) and the Ministry of Environment of Korea as the "Environmental Health Action Program (ARQ201303083)".

References

1. La Guardia MJ, Hale RC, Harvey E. (2006); *Environ Sci Technol.* 40: 6247–54
2. Schecter A, Päpke O, Harris TR, Tung KC, Musumba A, Olson J, Birnbaum L. (2006); *Environ Health Perspect.* 114: 1515–20
3. Van Leeuwen SPJ, Van Velzen MJM, Swart CP, Van der Veen I, Traag WA, De Boer J. (2009); *Environ Sci Technol*. 43: 4009–15
4. D'Hollander W, Roosens L, Covaci A, Cornelis C, Reynders H, Van Campenhout K, De Voogt P, Bervoet L. (2010); *Chemosphere*. 81: 478–87
5. Besis A, Samara C. (2012); *Environ Pollut.* 169: 217–29
6. Lee S, Kannan K, Moon H-B. (2013); *Sci Total Environ.* 443: 24–30
7. Jones-Otazo HA, Clarke JP, Diamond ML, Archbold JA, Ferguson G, Harner T, Richardson GM, Ryan JJ, Wilford B. (2005); *Environ Sci Technol.* 39: 5121–30.
9. KIHASA (Korea Institute for Health and Social Affairs), 2006. Research on national status of family health and welfare (in Korean)
10. Jeong Y, Lee S, Kim S, Choi SD, Park J, Kim HJ, Lee JJ, Choi G, Choi S, Kim S, Kim SY, Kim YD, Cho G, Suh E, Kim SK, Eun SH, Eom S, Kim S, Kim GH, Choi K, Kim S, Moon H-B. (2014); *Sci Total Environ.* 470-1: 137–5
11. WHO (World Health Organization), 2001. Guiding principles for complementary feeding of the breastfed chlid; 2001.
12. Moon H-B, Kannan K, Choi M, Choi HG. (2007); *Mar Pollut Bull.* 54: 1402–12.