HUMAN EXPOSURE TO PBDES TROUGH SEAFOOD CONSUMPTION AND DUST INGESTION IN KOREA: PRELIMINARY SURVEY

Sunggyu Lee[1], Hye-Seon Kim[2], Minkyu Choi[2], Hee-Gu Choi[2], Hyo-Bang Moon[1]

[1]Department of Environmental Marine Sciences, Hanyang University, Ansan 426-791, Republic of Korea
[2]National Fisheries Research & Development Institute (NFRDI), 408-1, Sirang-ri, Gijang-eup, Gijang-gun, Busan 619-705, Republic of Korea

Introduction

Polybrominated diphenyl ethers (PBDEs) have been widely used as brominated flame retardants (BFRs) in many products such as electronic equipment, plastics, textiles, building materials, carpets and automobiles. Three different formulations of PBDEs are commercially available: pentabromodiphenyl ether (penta-BDE), octabromodiphenyl ether (octa-BDE) and decabromodiphenyl ether (deca-BDE). These three commercial mixtures have different uses and toxicity[1]. PBDEs are one of emerging persistent organic pollutants (POPs) and have been widely detected in various environmental compartments[2], due to their similar proprieties with legacy POPs such as PCBs and DDTs. General population are exposed to PBDEs through their daily life such as food supply[3], indoor and outdoor activities[4] and child may have additional exposure route to PBDEs such as the placenta and breast milk[5]. Seafood consumption and house dust are considered as major exposure contributors of PBDEs to human[4]. It is therefore important to investigate contamination levels and exposure status of PBDEs in seafood and house dust. Limited data are available concerning occurrence of PBDEs in seafood and dust as well as exposure assessment of these compounds in Korea[6,8]. In this study, we estimated intake of PBDEs through seafood consumption and house dust ingestion for general population and specific subpopulations according to sex and age in Korea. We also compared distribution profiles of PBDEs between species and matrix investigated.

Materials and methods

Twenty-six marine species (n=78) were collected annually for the same species during the period of 2005 to 2007 from the Busan cooperative fish market, which is the largest fish market in South Korea and the representative market covering the whole country. These organisms are the most commonly consumed species and commercially important species in Korea. Collected samples were stored in a cooler box with ice and immediately transported to the laboratory. After removing the skins of fish and cephalopods, the muscles tissues were homogenized with an ultra-disperser. The shells of shellfish and crustaceans were removed and the whole soft tissues were pooled and homogenized for analysis. Household vacuum cleaner bags were obtained from 46 residential houses located from Busan, Korea in 2007. After removing the hairs and other non-dust particles had been manually, the dust samples kept in a freezer at -4°C until chemical analysis was performed. Preparation and instrumental analyses of PBDEs in seafood and dust samples were performed following the methods described elsewhere[7].

Results and discussion

Concentrations and congener profiles of PBDEs in seafood

Concentrations of total PBDEs (ΣPBDE; the sum of 23 PBDEs) in individual seafood from Korea ranged from 0.06 to 21.3 ng/g dry weight. The highest concentration of ΣPBDE in marine species was found in herring (*Clupea pallasii*; 16.3 ± 4.42 ng/g dry weight), which is long-lived and an oily fish[8]. The overall ΣPBDE concentrations (0.01-6.94 ng/g wet weight or 0.06-21.3 ng/g dry weight) in seafood measured in the present study are similar to those reported from some studies in the Netherlands (0.01-4.81 ng/g wet weight)[8] and Japan (0.11-3.30 ng/g wet weight)[9]. The PBDE concentrations in fish and shellfish in Japan (0.02-1.72 ng/g wet weight)[6], Spain (0.02-2.02 ng/g wet weight)[10] were lower than those measured in the present study.

The relative contributions of the 23 PBDE congeners for fish (n = 54) and bivalves (n = 12) showed a similar distribution (Figure 1). BDE 47 was predominant congeners in fish and bivalves, accounting for 30% of ΣPBDE concentrations, consistent with some studies reported previously[8,10]. The next PBDE congeners in both species groups were BDEs 49, 99, 100, 154 and 209. PBDEs are known that can be occurred biotransformation via debromination within organisms[11,12]. Biotransformation and debromination of PBDEs in biota might be evidence of often enriched in tetra-, penta- and hexa-BDE in fish species, and that is consistent with accumulation of less brominated congeners within fish tissues[12]. In our study, BDE 209 accounted for 7.6% and 9.8% of the total PBDE concentrations in fish and bivalves, respectively. Similar results were found in previous studies[13,19].

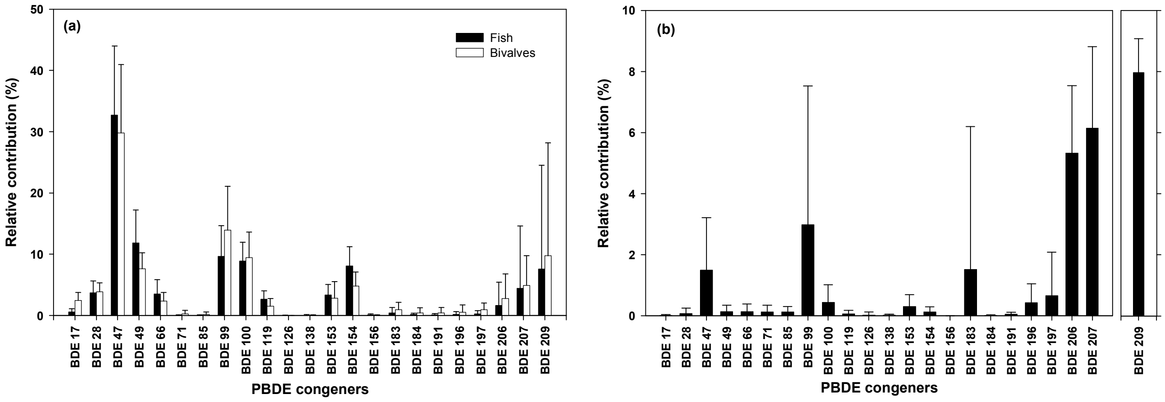

Figure 1. Relative contributions of 23 PBDE congeners (a) in fish (n=54) and bivalves (n=12) and (b) in house dust (n=46) from Korea. Whiskers on the bars represent standard deviations, for each PBDE congener.

Concentrations and congener profiles of PBDEs in house dust

The concentrations of ΣPBDE in house dust collected from Korea ranged from 80 to 16100 (median: 1400) ng/g dry weight. BDE 209 was the most abundant in all samples, with concentrations ranging from 43 to 11000 (median: 1200) ng/g dry weight. Higher brominated congeners of PBDEs such as BDE 207 (7%) and BDE 206 (5%) had comparatively higher proportions to the ΣPBDE concentrations. This contamination patterns were well consistent with high consumption of deca-BDE mixture as the BFR in Korea. Previous studies have reported the dominance of BDE 209 in house dust samples[14,18].

Total PBDE concentrations (80-16100 ng/g dry weight) in house dust measured in our study were similar to those reported for Singapore (110-13000 ng/g dry weight)[14], Australia (500-13000 ng/g dry weight)[15] and USA (920-17000 ng/g dry weight)[16]. The ΣPBDE concentrations in dust samples from Canada (170-170000 ng/g dry weight)[17], USA (780-30100 ng/g dry weight)[18] and UK (950-54000 ng/g dry weight)[15] were higher than those measured in the present study.

Intakes of PBDEs through seafood consumption and house dust

The intake of PBDEs through seafood consumption to Korean general population was estimated to be 55.4 ng/day. The highest contributor was mackerel (*Scomber japonicas*; 18.9 ng/day), accounting for 34% of total PBDE intakes. The estimated intakes of PBDEs through seafood consumption for men and women were 65.9 and 45.4 ng/day, respectively. The men had a higher intake than the women, because men (58.2 g/day) consume higher levels of seafood than women (43.5 g/day), consistent with other studies[19]. The estimated intakes of PBDEs via fish consumption were 20.8 ng/day in Spain[10] and 23.1 ng/day in Sweden[3], which were lower than those measured in the present study.

The estimated intakes of PBDEs through seafood consumption according to age ranged from 13.4 ng/day (< 2 years) to 73.4 ng/day (50 to 64 years) (Figure. 2). The overall body burden tends to increase with time because of the high consumption rate of seafood. In the present study, total body burden from intake of PBDEs increased with increasing age, with the exception of > 65 years old. Considering both factors of seafood consumption rate and body weight, the estimated intakes of PBDEs through seafood consumption ranged from 0.6 to 1.4 ng/kg body weight/day. The age group with the highest exposure rate was 3-6 years old, followed by 50-64 years, < 2 years and 30-49 years. In particular, children of 3-6 years and < 2 years relatively had high intakes compared with adult groups. Children could have higher potential risks from exposure by breast milk[5] and house dust[4,18].

The intakes of PBDEs through dust ingestion to adult (> 20 years old) and child (< 2 years) in Korea were estimated to be 0.83 and 10.3 ng/kg body weigh/day, respectively (Table 1). For high consumer, the estimated intakes of PBDEs via dust ingestion were 2.07 and 41.3 ng/kg body weight/day for adult and child in high scenario, respectively.

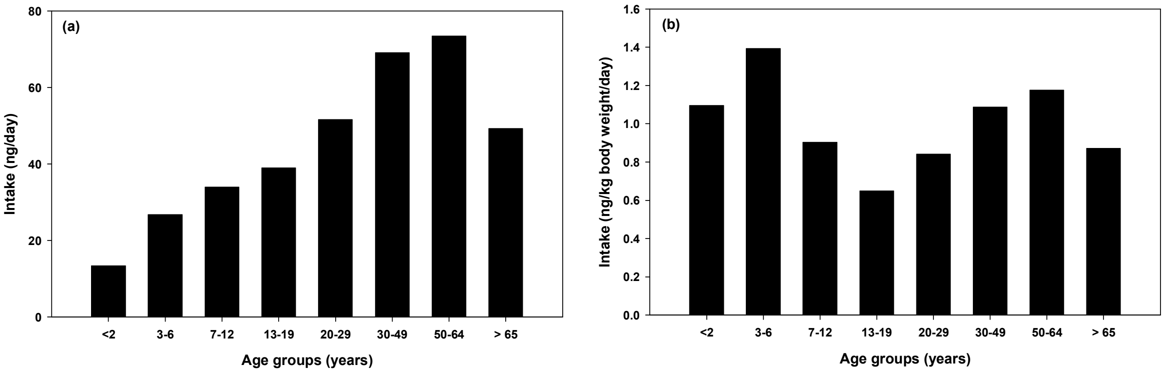

Figure 2. The estimated PBDE intakes through seafood consumption for eight age groups in Korea. (a) Intake (ng/day) is the total PBDE burden and (b) intake (ng kg body weight/day) is recalculated by body weight for each age group.

Table 1. Comparison of PBDEs exposure to adults and children through seafood consumption and dust ingestion in Korea.

	Adult[a]				Child[b]			
	average[c]	seafood-high[d]	dust-high[c]	high[d]	average[c]	seafood-high[d]	dust-high[c]	high[d]
Dust ingestion (mg/day)[e]	20	20	50	50	50	50	200	200
ng/day								
Seafood (% contribution)	60.3 (54)	94.4 (65)	60.3 (32)	94.4 (43)	13.4 (10)	22.3 (15)	13.4 (3)	22.3 (4)
Dust (% contribution)	50.4 (46)	50.4 (35)	126 (68)	126 (57)	126 (90)	126 (85)	504 (97)	504 (96)
Total intake	111	144	186	220	139	148	517	526
ng/kg bw/day								
Seafood	0.99	1.55	0.99	1.55	1.10	1.83	1.10	1.83
Dust	0.83	0.83	2.07	2.07	10.3	10.3	41.3	41.3
Total intake	1.82	2.38	3.06	3.62	11.4	12.1	42.4	43.1

[a]Assuming body weight of 60.95 kg for adult aged > 20 years; [b]Assuming body weight of 12.2 kg for child aged < 2 years; [c]PBDEs concentration in seafood at average concentration; [d]PBDEs concentration in seafood at 95th percentile concentration; [e]Assuming dust ingestion rate described by Jones-Otazo et al. (2005)[4]

Evaluation of exposure pathways of PBDEs through seafood consumption and dust ingestion

The estimated intakes of PBDEs for adults and children through seafood consumption and dust ingestion, with 4 exposure scenarios are summarized in Table 1. Considering average and high scenarios, the contribution of estimated intakes of PBDEs through seafood consumption and dust ingestion to adult was similar. However, the estimated PBDE intakes to Korean children showed that dust ingestion had higher contribution (> 90% to the total PBDE intakes) than seafood consumption for 4 scenarios. These results may be due to lower consumption rate of seafood and higher ingestion rate of house dust, compared to adults.

The US EPA IRIS database (2008)[20] as established an oral reference dose (RfD) of 100, 100, 200 and 7000 ng/kg body weight/day for BDEs 47, 99, 153 and 209, respectively. The risk assessment by these estimated PBDEs exposures were compared with the RfD values. The total PBDE intakes through seafood consumption and dust ingestion were 1.82 and 11.4 ng/kg body weight/day in average scenario for adults and children, respectively and 5.11 ng/kg body weight/day in high scenario for adults. These results were 2-50 times lower than the RfD for the only one congener such as BDEs 47 and 99. However, the other possible sources of PBDEs to humans were not considered. In particular, infant and toddler can be exposed to PBDEs and other contaminants through breast milk[5]. Therefore, systematic exposure assessments considering various potential routes should be needed to protect human health from POPs including PBDEs. The present preliminary survey will be useful for risk management for PBDEs from Korean seafood and house dust.

Acknowledgements
This study was funded by a grant from the National Fisheries Research and Development Institute (NFRDI), the Ministry of Land, Transport and Maritime Affairs (MLTM) and the National Institute Food and Drug Safety Evaluation (NFIDS), Korea.

References
1. Darnerud PO. (2003); *Environ Int.* 29: 841-53
2. de Wit CA, Alaee M, Muir DCG. (2006); *Chemosphere.* 64:209-33
3. Darnerud PO, Atuma S, Aune M, Bjerselius R, Glynn A, Petersson Grawé K, Becker W. (2006); *Food Chem Toxicol.* 44:1597-606
4. Jones-Otazo HA, Clarke JP, Diamond ML, Archbold JA, Ferguson G, Harner T, Richardson GM, Ryan JJ, Wildford B. (2005); *Environ Sci Technol.* 39:5121-30
5. Meironyte D and Norén K. (1999); *J Toxicol Environ Health Part A.* 58:329-41
6. Ohta S, Ishizuka D, Nishimura H, Nakao T, Aozasa O, Shimidzu Y, Ochiai F, Kida T, Nishi M, Miyata H. (2002); *Chemosphere.* 46:689-96
7. Moon HB, Kannan K, Lee SJ, Choi M. (2007); *Chemosphere.* 66:243-51
8. Bakker MI, de Winter-Sorkina R, de Mul A, Boon PE, van Donkersgoed G, van Klaveren JD, Baumann BA, ` Hijman WC, van Leeuwen SPJ, de Boer J, Zeilmaker MJ. (2008); *Mol Nutr Food Res.* 52:204-16
9. Akutsu K, Obana H, Okihashi M, Kitagawa M, Nakazawa H, Matsuki Y, Makino T, Oda H, Hori S. (2001); *Chemosphere.* 44:1325-33
10. Domingo JL Bocio A, Falcó G, Llobet JM. (2006); *Environ Sci Technol.* 40:4394-99
11. Tomy GT, Palace VP, Halldorson T, Braekevelt E, Danell R, Wautier K, Evans B, Brinkworth L, Fisk AT. (2004); *Environ Sci Technol.* 38:1496-504
12. Stapleton HM, Letcher RJ, Li J, Baker JE. (2004); *Environ Toxico Chem.* 23:1939-46
13. Gomara B, Herrero L, Gonzalez MJ. (2006); *Environ Sci Technol*. 40:7541-47
14. Tan J, Cheng SM, Loganath A, Chong YS, Obbard JP. (2007); *Chemosphere.* 66:985-92
15. Sjödin A, Päpke O, McGahee E, Focant J, Jones RS, Pless-Mulloli T, Toms LL, Herrmann T, Müller J, Needham LL, Patterson Jr. DG. (2008); *Chemosphere.* 73:S131-6
16. Harrad S, Ibarra C, Diamond M, Melymuk L, Robson M, Douwes J, Roosens L, Dirtu AC, Covaci A. (2008); *Environ Int.* 34:232-8
17. Wildford BH, Shoeib M, Harner T, Zhu J, Jones KC. (2005); *Environ Sci Technol.* 39:7027-35
18. Stapleton HM, Dodder NG, Offenberg JH, Schanta MM, Wise SA. (2005); *Environ Sci Technol.* 39:925-31
19. Schecter A, Päpke O, Harris TR, Tung KC, Musumba A, Olson J, Bimbaum L. (2006); *Environ Health Perspect.* 114:1515-20
20. US EPA's Integrated Risk Information System (IRIS) database. (2008); Available at URL: http://www. epa.gov/iris/index.html (assessed April 20 2011)