Global Contamination of PBDEs in Human Milk from Asia

Agus Sudaryanto[1], Natsuko Kajiwara[1], Oyuna Tsydenova[1], Hisato Iwata[1], Tussy A. Adibroto[2], Hongxia Yu[3], Kyu-Hyuck Chung[4], Annamalai Subramanian[1], Maricar Prudente[5], Touch Seang Tana[6], Shinsuke Tanabe[1]

[1]Center for Marine Environmental Studies (CMES), Ehime University
[2]Agency for the Assessment and Application of Technology (BPPT)
[3]School of the Environment, Nanjing University
[4]College of Pharmacy, SungKyunKwan University
[5]Science Education Department, De La Salle University
[6]Member of the Economic, Social and Cultural Observational Unit of the Council of Ministers

Introduction

Polybrominated diphenyl ethers (PBDEs), a family of chemicals widely used as flame-retardants are emerging contaminants appear to be ubiquitous in the environment [1,2] and magnify in the biological tissues as classical organochlorine compounds (OCs) [3]. These compounds have propensity to disrupt endocrine system in animal bodies [1]. Some recent studies have suggested that PBDEs may be transported to significant distances and their levels are increasing in remote areas and thus have to be included in the POPs (persistent organic pollutants) list [4]. Therefore, similar fate and behavior of PBDEs with OCs and its potential toxic effect to human and wildlife have been a matter of great concern.

Because humans occupy the top position in food chains, human tissue accumulates higher levels of persistent lipophilic contaminants. Monitoring studies using breast milk samples indicated that PBDEs concentrations varied widely[1,2,5,6], the levels in North American people being the highest [5,6]. Furthermore, contamination of human milk by PBDEs revealed increasing worldwide trends [6]. Despite a considerable number of published studies on environmental occurrences of PBDEs from Europe and North America [6], much less information is available in Asian region [4,7], even though Asia has a very high market demand for PBDEs [2]. The present study investigated concentrations of PBDEs in human milk from various countries in Asia in order to elucidate their contamination status, geographical variation, possible exposure source(s) and pathways, and to assess possible association with maternal characteristics in comparison with OCs. Finally, the potential health risk to the human was also evaluated.

Materials and Methods

Breast milk samples were collected from healthy women living in urban areas of various countries in Asia (Japan, Korea, China, Philippines, Vietnam, Cambodia, Indonesia, Malaysia and India) during the years 2000-2004 (Table 1).The milk was sampled during breastfeeding using a manual breast pump and/or a passive breast milk sampler. Before taking the milk, the donors completed informed consent form and an exposure assessment questionnaire. Breast milk samples were placed in analytical-grade glass containers with Teflon-lined caps, frozen immediately and kept at –20^{o}C in the laboratory until chemical analysis.

Analysis of PBDEs was performed following the method by Kajiwara *et al*. [7], with slight modification. Nine major congeners of PBDE (Table 1) were identified and quantified by GC-MS in selective ion mode.

Results and Discussion

PBDEs were detected in all the milk samples analyzed in this study (Table 1) with mean concentrations ranging from 0.60 to 3.8 ng/g lipid wt. These levels were one to three order magnitudes lower than PCBs and DDTs. The residue levels of halogenated compounds present in human milk were in the order of DDTs>PCBs>HCHs>CHLs>HCB> PBDEs. Unlike PCBs and DDTs, concentrations of PBDEs in breast milk of the present study did not correlate with

age of mother and parity (p>0.05), as also noticed in same other studies [5,8]. This may be because of the relatively short period of the usage of PBDEs and/or the different exposure pathways of PBDEs and OCs, which are yet to be fully determined. Except Japan, to our knowledge, this is the first study reporting the levels of PBDEs in breast milk from Asian countries. In Japan, the levels observed in the present study in the samples collected during 2000 (3.8 ng/g lipid wt.) was higher than in the years between 1973 (<0.01 ng/g lipid wt.) and 1988 (1.6 ng/g lipid wt.)[9]. The extent of contamination by PBDEs in human milk from Asia is comparable to those reported from Europe [6], but one or two order of magnitude lower than levels found in Noth America [5,6].

Table 2. Mean concentrations (ng/g lipid wt.) of PBDEs in human milk collected from Asian countries[a].

Country	Year of Sampling	Lipid (%)	Concentration (ng/g lipid wt.)									
			BDE-3	BDE-15	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	ΣPBDEs
Japan (n=10)	2000	2.8	<0.01	0.14	0.14	1.5	0.38	0.41	1.2	0.06	0.03	3.8
Korea (n=9)	2004	3.1	<0.01	0.17	0.13	0.81	0.20	0.24	0.79	0.06	0.08	2.6
China (n=9)	2004	1.8	<0.01	0.20	0.40	0.66	0.17	0.17	1.0	0.08	0.30	3.5
Indonesia (n=30)	2001-2003	2.3	<0.01	0.01	0.050	0.39	0.18	0.15	0.32	0.03	0.13	1.3
Malaysia (n=5)	2003	1.9	<0.01	0.04	0.12	1.4	0.42	0.52	0.65	0.06	0.24	3.5
India (n=5)	2000	2.9	<0.01	nd	0.010	0.27	0.11	0.06	0.10	0.02	0.02	0.60
Vietnam (n=5)	2000	2.6	<0.01	0.050	0.10	0.31	0.10	0.08	0.17	0.03	0.07	0.91
Cambodia (n=4)	2000	2.9	<0.01	0.030	0.040	0.83	0.22	0.17	0.18	0.03	0.04	1.5
Philippines (n=4)	2000	2.9	<0.01	0.070	0.18	1.1	0.47	0.30	0.39	0.09	0.08	2.6

[a] values indicated an arithmetic mean.

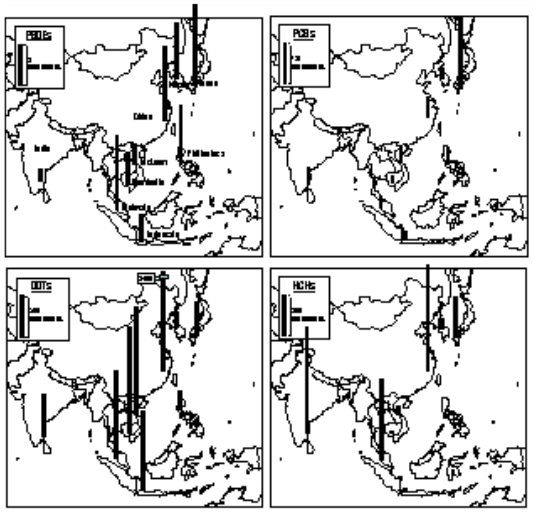

Fig. 1. Geographical distribution of PBDEs in human milk from Asia in comparison to OCs.

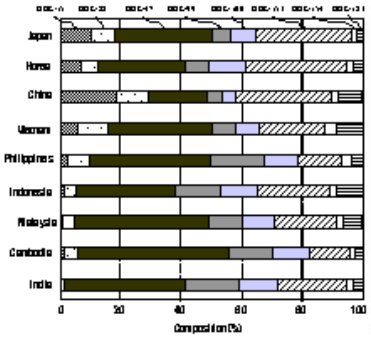

Fig. 2. PBDEs congener profiles in human milk from Asia.

Among the countries studied in the present work, relatively higher levels of PBDEs were found in the samples collected from Far Eastern countries, such as Japan, China and Korea (Table 1, Fig. 1). This may be due to larger usage of PBDEs in the northern hemisphere as in the case of OCs, such as PCBs [4]. However, human milk from Malaysia and Philippines also contained elevated levels of PBDEs, indicating some developing countries in South Asia may also be important sources of PBDEs. Relatively lower levels of PBDEs were detected in samples collected from India, Vietnam, Cambodia, and Indonesia. This geographical distribution pattern was different from that of PCBs and DDTs which showed specific regional contamination status, in which developed nations (Japan) were contaminated more by PCBs while developing countries in South Asia by DDTs or HCHs (Fig. 1).

In most of the donors from Asian countries (Fig. 2), BDE-47 was the predominant congener present in breast milk, similar to other studies on human matrices [5,6,8,10]. Apart from the fact that BDE-47 being the main component in penta BDE commercial mixture, the dominance of BDE-47 in biological samples may be due to its high stability and high bioaccumulative nature. However, an observation of individual PBDE patterns shows large inter-individual differences (Fig. 2), which may be due to influence to various exposure source(s) and pathways. In some donors, the proportions of BDE-153 or BDE-183 were equal to BDE-47. This study reveals the exposure of certain populations to higher brominated PBDEs. The exposure routes are different for BDE-47 and the higher brominated congener like BDE-183. For example, food is the major source for BDE-47, whereas airborne uptake seems to be more pronounced for BDE-183 [1]. Furthermore, the higher percentage contribution of lower congeners (BDE-15 and BDE-28) in samples from Far Eastern countries, such as China, Korea and Japan (Fig. 2) may indicate their relatively long history of usage when compared to South Asian nations and/or due to their temperate location. Ueno *et al*. [4] indicated a preferential atmospheric transportability of lower BDE congeners depending on latitude. Moreover, typically commercial penta mixtures (BDE-47, BDE-99 and BDE-100) were more prevalent in the samples collected from South Asian countries while the Far Eastern samples contained more octa mixture (BDE-183, BDE-153 and BDE-154) (Fig. 2). This pattern may be due to the different commercial mixtures of PBDEs used in these two

regions. However, due to lack of information on particular habits of milk donors it is rather difficult to identify the major source and pathways.

The presence of PBDEs in human milk is of great concern since these compounds can be transferred to an infant through lactation. Available evidence suggests that the PBDEs have the propensity to disrupt thyroid hormones, cause neurobehavioral deficits, some changes of fetal development and possibly cause cancer in laboratory animals [1]. However, it is unclear whether current concentrations of PBDEs in human tissues may be expected to adversely affect human health. The BDE-99 body burden (<1 mg/kg) found in infant of the present study is still much lower than the LOEL for penta BDE (0.4 mg/kg) associated with behavioral alterations in neonatal mice [1]. It must be emphasized that, investigation on PBDEs level in human matrices were conducted only in very few number of samples, and hence potential health impacts studies should include assessment at a large population level. Moreover, PBDEs is still widely used in huge amounts at present. Evidence on the increasing levels in the environment is mounting, and thus contamination by these compounds may become a matter of severe concern in future. Continued investigation on contamination status and toxicokinetics of PBDEs are needed to evaluate their temporal trend, source and potential health risk, particularly on critical target organs or tissues.

Acknowledgements

This study was supported by Grants-in-Aid for Scientific Research (A) (No. 16201014 and 14254004) from Japan Society for the Promotion of Science (JSPS), the Toyota Foundation, Tokyo, Japan (Project No. D03-B3-062) and 21st Century COE Program from the Japanese Ministry of Education, Culture, Sports, Science and Technology.

References

1. Darnerud P.O., Eriksen G.S., Jóhannesson T., Larsen P.B., Viluksela M. 2001. Polybrominated Diphenyl Ethers: Occurrence, Dietary Exposure, and Toxicology. Environ. Health Perspect. 109: 49-68.

2. Hites, R. A. 2004. Polybrominated Diphenyl Ethers in the Environment and in People: A Meta-Analysis of Concentrations. Environ. Sci. Technol. 38: 945-956.

3. Boon, J. P., Lewis, W. E., Tjoen-A-Choy, M. R., Allchin, C. R., Law, R. J., de Boer, J., ten Hallers-Tjabbes, C. C., Zegers, B. N. 2002. Levels of Polybrominated Diphenyl Ether (PBDE) Flame Retardants in Animals Representing Different Trophic Levels of the North Sea Food Web. Environ. Sci. Technol.36: 4025-4032.

4. Ueno, D., Kajiwara, N., Tanaka, H., Subramanian, A., Fillmann, G., Lam, P. K. S., Zheng, G. J., Muchtar, M., Razak, H., Prudente, M., Chung, K. H. and Tanabe, S. 2004. Global Pollution Monitoring of Polybrominated Diphenyl Ethers Using Skipjack Tuna as Bioindicator. Environ. Sci. Technol. 38: 2312-2316.

5. Schecter A, Pavuk M, Päpke O, Ryan JJ, Birnbaum L, Rosen R. 2003. Polybrominated diphenyl ethers (PBDEs) in U.S. mothers' milk. Environ Health Perspect. 111: 1723-1729

6. Ryan J.J. 2004. Polybrominated diphenyl ethers (PBDEs) in human milk: occurrence worldwide. The thirds international workshop on brominated flame retardants. June 6-9, 2004. Canada. 17-22.

7. Kajiwara, N., Ueno, D., Ramu, K., Kamikawa, S., Yamada, T., Chung, K.H., Tanabe, S. 2004. Geographical Distribution and Temporal Trends of Polybrominated Diphenyl Ethers (PBDEs) in Cetacean from Asian Waters. The thirds international workshop on brominated flame retardants. June 6-9, 2004. Canada. 187-190.

8. Mazdai A, Dodder NG, Abernathy MP, Hites RA, Bigsby RM. 2003. Polybrominated diphenyl ethers in maternal and fetal blood samples. Environ Health Perspect. 111: 1249-1252.

9. Akutsu, A., Kitagawa, M, Nakazawa, H, Makino, T, Iwazaki, K, Oda, H, Hori, S. 2003. Time-trend (1973-2000) of polybrominated diphenyl ethers in Japanese mother's milk. Chemosphere. 53: 645-654.

10. Covaci, A., de Boer, J., Ryan, J. J., Voorspoels, S. and Schepens, P. 2002. Distribution of Organobrominated and Organochlorinated Contaminants in Belgian Human Adipose Tissue. Environmental Research. 88: 210-218

11. Petreas, M., She, J., Brown, F.R., Winkler, J., Windham, G., Rogers, E. 2003. High body burdens of 2,2',4,4'-tetrabromodiphenyl ether (BDE-47) in California women. Environ Health Perspect. 111: 1175-1179.

12. Sjödin, A. Hagmar, L., Klasson-Wehler, E., Kronholm-Diab,K., Jakobsson, E., and Bergman, A. 1999. Flame Retardant Exposure: Polybrominated Diphenyl Ethers in Blood from Swedish Workers. Environ. Health Perspect. 107: 643-648.