Exposure to Organophosphorus Flame Retardants (PFRs) through Human Breast Milk Feeding

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Abstract

Organophosphorus flame retardants (PFRs) are added to a wide range of consumer products, including televisions, computers, electrical outlets and household textiles, to reduce the flammability. Since two major brominated flame retardants (BFRs), i.e., polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) were listed in the Stockholm Convention on persistent organic pollutants (POPs), the use of PFRs has been increasing. Therefore, environmental contamination and exposure for human are of public concern. However, limited information on human exposure to PFRs is available so far. In the present study, we investigated the levels of PFRs, PBDEs, and HBCDs in human breast milk collected from some Asian countries including Vietnam, the Philippines and Japan. PFRs and BFRs were detected in most of the analyzed breast milk samples from Asian countries. In Japan, the concentrations were in the order of PFRs ≈ HBCDs > PBDEs and levels of PFRs and HBCDs were higher than those of PCBs. As the result of temporal variation study, PFR and HBCD levels were higher in 2009-2014 than in 1999 while PCBs showed a decreasing trend. Since the estimated daily intake (EDI) values for some PFRs were close to the reference dose, adverse effects of these compounds should be investigated in future.

Conclusions

Accumulation level

- Philippines > Vietnam > Japan
- Exposure level could be affected by e-waste dismantling

E-waste dismantling is a major exposure source for PFRs

Temporal trend

- Levels of PFRs and HBCDs have increased during study period

Recent increase in production and use of PFRs and HBCDs

Exposure assessment

- Some EDI values for TBEP, TCEP and TDCIPP were close to RfDs

Health effect on breast feeding infant should be investigated

Results and Discussion

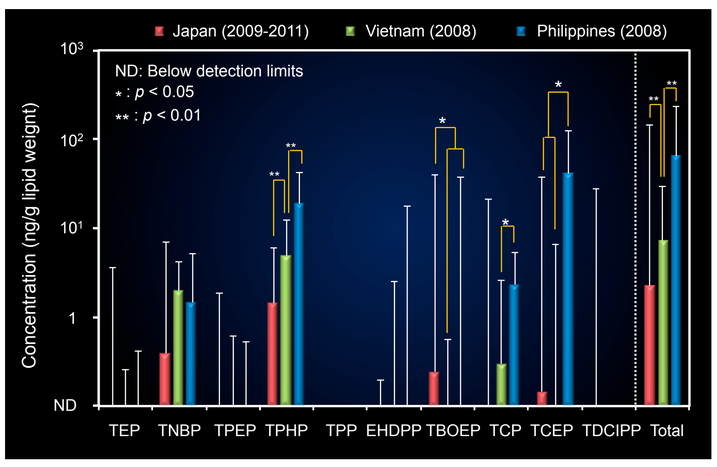


Fig. 1 Concentration of PFRs in breast milk from Japan, Vietnam and the Philippines.

Accumulation level

Total concentration was the highest in the Philippines, followed by Vietnam and Japan. Levels in the Philippines were even higher than those in Sweden. Among target compounds, TNBP and TPHP were detected in all the countries. In addition, TPHP and TCEP were higher in the Philippines compared to the other 2 countries. Unfortunately we couldn't get information on production in each country, amount and profile of PFRs use could be different among countries. In Vietnam, different profiles among locations were observed. TPHP was the highest PFR in Bui Dau and Hanoi whereas TNBP showed the highest level in Trang Minh. In Trang Minh, electronic parts, like printed circuit board, are dismantled and that is the major recycling activity. On the other hand, TV or PC casing plastics are dismantled in Bui Dau.

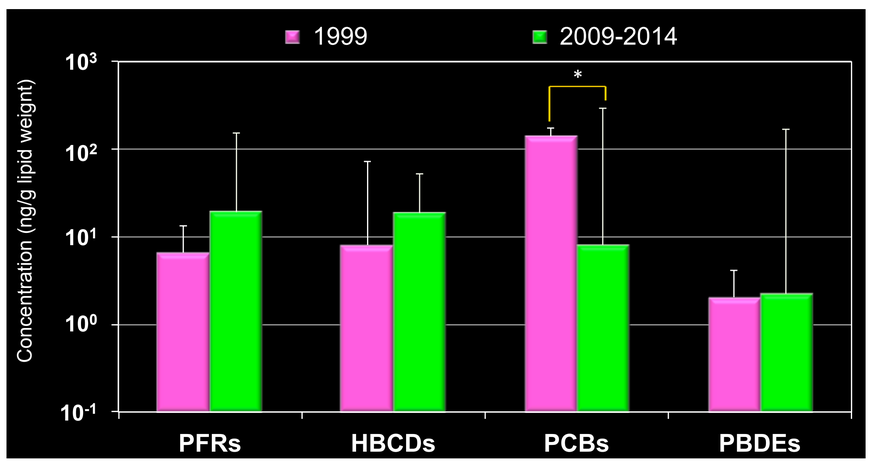
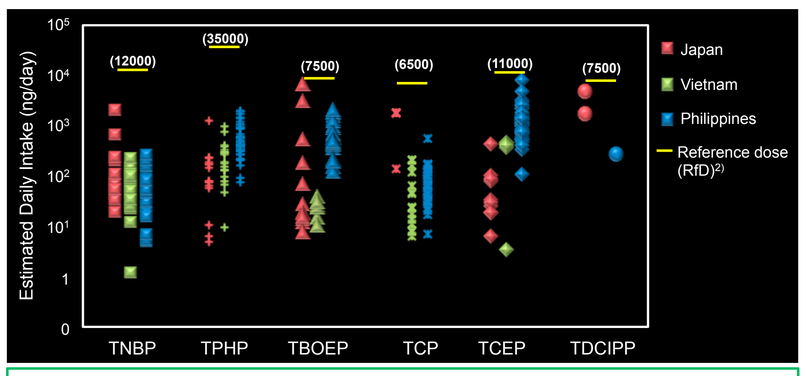


Fig. 2 Temporal variation of PFRs, HBCDs, PCBs and PBDEs in breast milk from Japan.

Temporal trend

We collected breast milk samples at 2 time points in Japan. In 1999, PCB levels were the highest among target compounds followed by PFRs and HBCDs with almost same levels and PBDEs ranked the last. Interestingly, PFRs and HBCDs were higher than PCBs in 2009-2014. PCBs level in 2009-2014 was significantly lower than that in the 1999. On the other hand, PFR and HBCD levels were higher in 2009-2014 than in 1999, though it was not statistically significant. The use of PFRs and HBCDs had not been regulated whereas PCBs and PBDEs were terminated few decades ago. This could be reflecting the regulations and consequent decrease in exposure levels in Japan. And the result implies that the exposure to HBCDs and PFRs for general Japanese population have been increasing since 1999. Therefore the increase in production and use of PFRs could result in increase in human exposure levels.



Estimated Daily Intake (EDI) =
PFR concentration x Daily breast milk intake
Daily breast milk intake[1] : **700 mL**

1) Oostdam *et al.*, 2005; 2) Van del Eede *et al.*, 2011

Fig. 3 Estimated daily intake (EDI) of PFRs through breast milk ingestion in Japan, Vietnam and the Philippines.

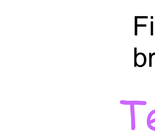
Exposure assessment

To estimate the daily intake of PFRs through breast milk feeding, EDI was calculated by the equation shown in the figure. In which, average detected PFR concentration in breast milk times average daily milk intake for an infant. Average daily breast milk intake was assumed as 700 mL that was cited from the reference. All the EDI values for each sample are plotted and compared with reference doses available in the literatures. As shown in the figure, most of the EDI values were less than RfD values, suggesting that PFR exposure through breast milk ingestion may not pose a serious health problem. However, EDIs for TBOEP, TCEP and TDCIPP were close to RfDs for some samples. Therefore, health effect of PFR exposure on breast feeding infant should be investigated in the future.

Materials and Methods

Targeted 10 PFRs Compounds

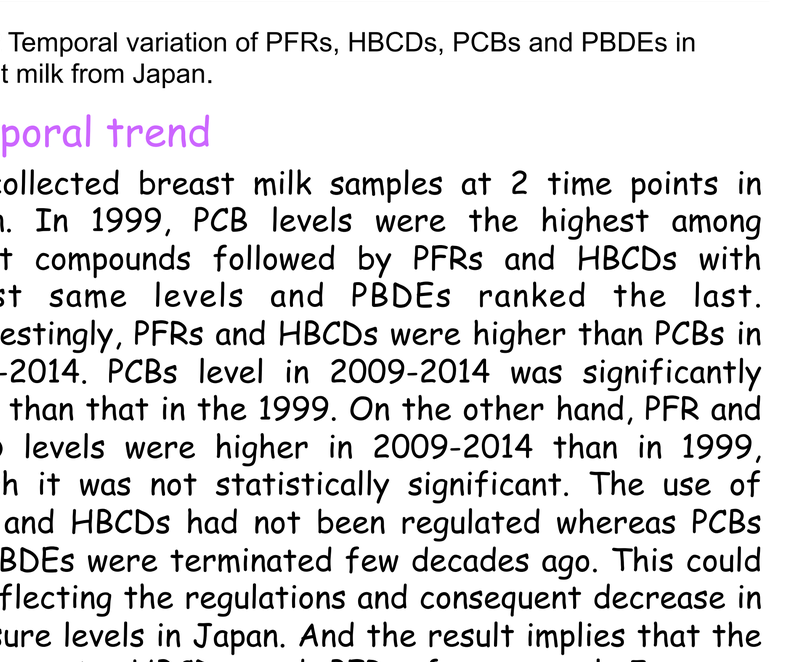
- TEP: Triethyl phosphate
- TNBP: Tri-*n*-buthyl phosphate
- TPEP: Tripentyl phosphate
- TPHP: Triphenyl phosphate
- TPRP: Tripropyl phosphate
- EHDPP: 2-Ethylhexyl diphenyl phosphate
- TBOEP: Tris-(2-butoxyethyl) phosphate
- TCP: Tricresyl phosphate
- TCEP: Tris-(2-chloroethyl) phosphate
- TDCIPP: Tris-(1,3-dichloro-2-propyl) phosphate



	TEP	TNBP	TPP	TPHP	TPRP	EHDPP	TBEP	TCP	TCEP	TDCIPP
Log K_{ow}	0.8	4.00	5.29	4.59	1.87	5.73	4.38	1.87	1.47	3.27
Substituent (R)	CH_3	CH_3	CH_3		CH_3		CH_3	CH_3	Cl	Cl Cl
FRs				○		○	○	○	○	○
Plasticizer	○	○		○		○	○		○	○
Lubricant		○		○				○		
Wax		○					○			
Paint		○		○		○	○	○	○	○
Antifoam		○					○			
Solvent	○	○						○	○	

Marklund *et al.* 2005

Analytical Method



Acknowledgements

Financial support was provided by Grants-in-Aid for Scientific Research (S: 26220103, A: 25257403, B: 25281008, Challenging Exploratory Research: 24651010) of the Japanese Ministry of Education, Science, Sports, Culture and Technology (MEXT) and Japan Society for the Promotion of Science (JSPS). This study was also partly supported by the Grant (No.143370) for Environmental Research Projects of Sumitomo Foundation, Japan

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