Geographical Distribution of Polybrominated Diphenyl Ethers and Organochlorine Compounds in Fish From Indonesia

Agus Sudaryanto[1], Natsuko Kajiwara[1], Hisato Iwata[1], Arif Dwi Santoso[2], Phillipus Hartono[3], Muawanah S.Pi[3], Yuichi Hayami[1], Koji Omori[1], Shinsuke Tanabe[1]

[1]Center for Marine Environmental Studies (CMES), Ehime University
[2]Agency for the Assessment and Application of Technology (BPPT)
[3]The National Seafarming Development Centre (NSDC)

Introduction

Currently, aquatic environment has been polluted by numerous contaminants including classical organochlorine compounds (OCs) and the newly identified and widely used flame-retardants known as polybrominated diphenylethers (PBDEs). PBDEs are used in a variety of commercial applications such as plastics, textiles, electronic circuits and other materials to prevent damage from fire [1]. PBDEs are structurally similar to other environmental pollutants, such as dioxins, polychlorinated biphenyls (PCBs) and polybrominated biphenyls (PBBs), and thus may have similar characteristics and toxic potency [2]. Their properties, such as lipophilicity, persistency and bioaccumulative nature, results in its bioaccumulation and biomagnification in the food chain involving a wide range of trophic levels [3]. As a consequence, they are often detected in wildlife including fish.

Fig. 1. Map showing sampling locations.

Because of the scarcity of reports, high Asian demand for products containing PBDEs [4,5], the present study investigated concentrations of PBDEs and OCs, including polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane and its metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), chlordane compounds (CHLs), hexachlorobenzene (HCB), and *tris* (4-chlorophenyl) methane (TCPMe) in fish from Indonesia. The paper aims at understanding the contamination status, the implications of congener specific profiles and elucidating potentially difference sources.

Materials and Methods

Twenty-three specimens comprising 9 species of fish were used in this study. Freshwater and coastal fish were collected from Ciliwung River (Bogor) and Jakarta Bay (Fig. 1), respectively during July 2003. The whole body tissues of the same species from each location were pooled, homogenized and analyzed for PBDEs and OCs.

Analytical methods for PBDEs and OCs were carried out following the procedure by Kajiwara *et al*[5]. Identifications and quantification of PCBs, DDTs, HCHs, CHLs and HCB was performed using a GC equipped with an ECD, while PBDEs and TCPMe were performed using a GC-MS in selective ion monitoring mode.

Results and Discussion

PBDEs (Table 1, Fig. 2) and OCs (Fig. 2) were detected in all the samples of the present study, indicating widespread contamination of these compounds in the environment of Indonesia. There were significant differences in the concentration of PBDEs in fish among locations (p<0.05), indicating localized sources of PBDEs. Similar patterns were also noticed for PCBs and DDTs. Among organohalogen compounds, PBDEs was the third most abundant pollutant class after DDTs and PCBs followed by CHLs, HCHs, HCB and TCPMe in freshwater fish, whereas PBDEs ranked forth (PCBs>DDTs>CHLs>PBDEs>HCB>HCHs>TCPMe) in marine fish from Jakarta Bay. Elevated concentration of PBDEs and DDTs in Bogor may be caused by its localized sources. Concentrations of PBDEs in fish found in the present study were much lower than those in North America [6,7,8], but relatively close to the fish from

European countries [9,10,11] and Australia [12].

Table 1. Concentrations of PBDEs (ng/g lipid wt.) in fish from two locations in Indonesia.

Fish	Fat (%)	BDE-3	BDE-15	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	SPBDEs
Jakarta Bay											
Mullet	7.3	<0.010	0.12	0.33	4.8	0.23	0.65	0.15	0.38	<0.010	6.7
Milkfish	3.9	<0.010	0.42	0.42	14	0.75	1.1	0.60	0.82	<0.010	18
Croaker	2.9	<0.010	<0.020	0.35	7.1	1.6	1.5	0.54	0.98	<0.010	12
Perch	4.4	<0.010	<0.020	0.16	2.7	1.0	0.57	0.39	0.23	<0.010	5.1
Mackerel	8.0	<0.010	0.07	0.28	2.4	1.5	0.50	0.25	0.34	<0.010	5.3
Average	5.3	nd	0.12	0.31	6.2	1.0	0.86	0.39	0.55	nd	9.4
Ciliwung River (Bogor)											
Tilapia	7.1	<0.010	0.12	0.56	8.7	0.21	1.6	0.52	2.9	<0.010	15
Carp	7.1	<0.010	0.31	0.99	19	0.05	2.0	0.31	1.4	<0.010	25
Gourami	5.3	<0.010	0.14	0.56	8.5	3.0	1.8	1.5	3.8	0.47	20
Catfish	11	<0.010	0.10	0.27	6.4	6.6	0.99	1.2	0.55	0.59	17
Average	7.6	nd	0.17	0.60	11	2.5	1.6	0.88	2.2	0.27	19

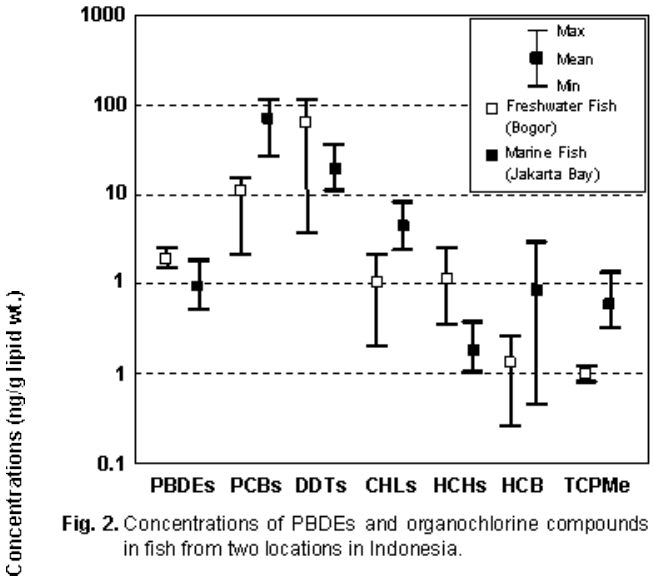

Fig. 2. Concentrations of PBDEs and organochlorine compounds in fish from two locations in Indonesia.

Among the PBDEs analyzed (Fig. 3), BDE-47 was predominant congener followed in order by BDE-99>BDE-154> BDE-100>BDE-153>BDE-28>BDE-15> BDE-183 in freshwater fish, and BDE-99>BDE-100>BDE-154>BDE-153>BDE-28>BDE-15 in marine fish. It is interesting to note that the composition and congeners pattern in freshwater fish was relatively different to those observed in coastal fish species (Fig. 3). The percentage contribution of higher brominated congeners such as BDE-154 and BDE-183 were greater in inland than coastal fish. This may probably be because of freshwater fish were in proximity to local anthropogenic sources. In fact, several benthic fish residing close to suspected PBDE point sources showed elevated concentrations of highly brominated congeners [6,7].

For some species (Fig. 3), the typical pattern of PBDEs shown above was not observed, probably because of species specific accumulation, metabolism and/or elimination. Among the fish in this study, carp (*Cyprinus carpio*) for

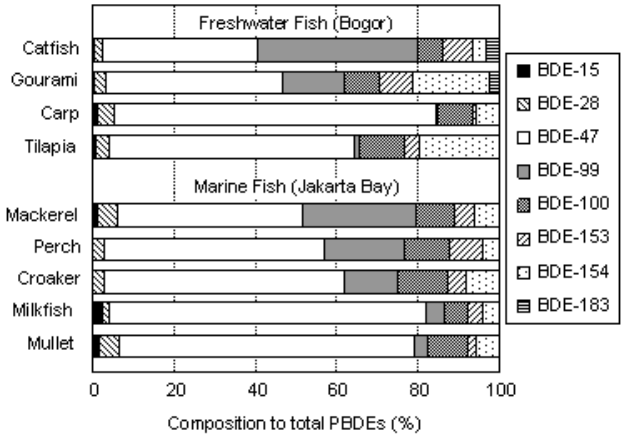

Fig. 3. PBDEs congener profiles in fish collected from two locations in Indonesia.

example, contained unusual pattern of PBDE congeners, in which BDE-99 and BDE-153 were significantly less relative to the tissues of other fish species from the same geographical area. This observation was similar to wild carp in Virginia freshwater[7] and may indicate that metabolization of these two congeners are well developed in this carp. In fact, these two congeners can be metabolized into BDE-47 by debromination processes1. Other explanation may be due to selective uptake of these two congeners. An *in vivo* study on common carp feeding pellets spike with PBDE mixtures (BDE-47, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-183) showed a low uptake of BDE-99, BDE-153 and BDE-183[13]. The lower BDE-99 and much higher percentage of BDE-47 in tilapia, mullet and milkfish of this study may also reflected species specific metabolic capacity and selective congener's accumulation. On the other hand, congener specific pattern in catfish represent an opposite pattern regarding the composition of BDE-47 and BDE-99, in which BDE-99 was higher than BDE-47. The overall pattern of PBDE congeners in catfish resembles the penta-product, indicating a local source that may have discharged PBDE with a congener profile similar to that of the penta-BDE technical product. Some fish from Ciliwung River also contained considerable proportion of higher congeners such as BDE-154 and BDE-183. This may indicate that the octa-BDE technical product (which contains BDE-153, BDE-154 and BDE-183) could have contributed to the profile observed.

Collectively, the results clearly indicate that PBDEs are ubiquitous contaminants present in the Indonesian environment. The higher contamination of PBDEs was found in the location near to the anthropogenic sources. Presences of PBDEs as well as classical OCs in fish are of further concern because fish is main diet for human. The increasing level of PBDEs contamination in various environmental matrices in recent years is also a matter of concern. Moreover, due to on going usage and high demand of PBDEs in the Asian region, contamination by these compounds may become a matter of attention in future. Continued investigation on contamination status and toxicokinetics of PBDEs are needed to evaluate their temporal trend, source and potential health risk. The present study provides a useful baseline for future research on the accumulation of PBDEs in the environment of Indonesia.

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) and •g21st Century COE Program•h 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. de Wit C.A. 2002. An overview of brominated flame retardants in the environment. Chemosphere. **46**: 583–624.

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. 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.

6. Dodder N.D., Strandberg B., Hites R.A. 2002. Concentration and spatial variations of polybrominated diphenyl ethers and several organochlorine compounds in fishes from northwestern United States. Environ. Sci. Technol. 36: 146-151.

7. Hale, R.C, La Guardia M.J., Harvey E.P., Matteson-Mainor T., Duff W.H., Gaylor M.O. 2001. Polybrominated diphenyl ether flame retardants in Virginia freshwater fishes. Environ. Sci. Technol. 35: 4585-4591.

8. Manchester-Neesvig J. B., Valters K., Sonzogni W.C. 2004. Comparison of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in lake Michigan salmonids. Environ. Sci. Technol. 35: 1072-1077.

9. Nylund K., Kierkegard A., Erikson U., Asplund L., Biegnert A., Olsson M. 2001. Spatial distribution of some polybrominated diphenyl ethers and hexabromomycyclododecane in Herring (Clupea harengus) along the Swedish Coast. Brominated Flame Retardant Workshop. Stockholm. 349-352.

10. Jacobs M., Covaci A., Schepens P. 2002. Investigation of selected persistent organic pollutants in farmed Atlantic Salmon (*Salmo salar*), Salmon aquaculture feed, and fish oil compartments of feed. Environ. Sci. Technol. 36: 2797-2805.

11. Paepke O. and Herrmann T. 2004. Polybrominated diphenyl ethers (PBDEs) in fish samples of various origin. Organohalogen Compounds. 66: 3971-3976.

12. Symons R., Burniston D., Piro N., Stevenson G., Yates A. 2004. A Study of the Presence of Brominated Flame Retardants in Australian Fauna. Organohalogen Compounds. 66: 3959-3964.

13. Stapleton H.M., Letcher R.B., Baker J.E. 2002. Uptake, Metabolism and Depuration of Polybrominated Diphenyl Ethers (PBDEs) by the Common Carp (*Cyprinus carpio*). Organohalogen Compounds. 58: 201-204.