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Global Pollution Monitoring of Polybrominated Diphenyl Ethers Using Skipjack Tuna as a Bioindicator

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To elucidate the global distribution of polybrominated diphenyl ethers (PBDEs), these chemicals were determined in the muscle of skipjack tuna (*Katsuwonus pelamis*) collected from offshore waters of various regions in the world (Japan, Taiwan, Philippines, Indonesia, Seychelles, and Brazil, and the Japan Sea, East China Sea, South China Sea, Indian Ocean, and North Pacific Ocean). PBDEs were detected in almost all the skipjack tuna collected from the locations surveyed (from <0.1 to 53 ng/g of lipid), indicating widespread contamination by these compounds in the marine environment. Residue levels of PBDEs in these samples from the northern hemisphere seem to be higher than those from the southern hemisphere, which is plausibly due to larger usage of these compounds in the northern hemisphere. Higher concentrations of PBDEs were detected in the samples from waters around the East China Sea (up to 53 ng/g of lipid). Developing countries around the East China Sea are supposedly the "hot spots" releasing these chemicals into the marine environment. With regard to the composition of PBDE congeners, the percentage contribution by lower brominated congeners (BDE15, -28, and -47) showed an increasing trend with increasing latitude. On the other hand, higher brominated congeners (BDE153, -154, and -183) showed a reverse trend. These patterns suggest that lower brominated congeners of PBDEs (di-, tri-, and tetra-BDEs) were preferentially transported from pollution sources to northern colder regions through the atmosphere. PBDEs may have a high potency to cause global pollution like PCBs.

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Introduction

Polybrominated diphenyl ethers (PBDEs) are one of the brominated flame retardants (BFRs) widely used in plastics, textiles, paints, and electronic appliances including computers, televisions, and other electric household equipment (*1*, *2*). Contamination of PBDEs is widespread in the environment as evidenced by their detection in a wide range of environmental media and biota (*1*−*5*). Recent studies suggest that PBDEs underwent long-range atmospheric transport and deposited in the polar regions such as the Canadian Arctic (*6*, *7*). Although several investigators have monitored the PBDE pollution in localized areas, information on the global distribution of PBDEs which can explain their transport behavior and fate are still limited.

To elucidate the global distribution of PBDEs, the present study attempted to use skipjack tuna (*Katsuwonus pelamis*) as a bioindicator. Skipjack tuna is principally distributed from offshore waters to open seas in tropical and temperate regions almost all over the world such as the Pacific, Atlantic, and Indian Oceans (*8*). This species is an important commercial fish, and its ecology and biology have been well studied (*8*−*10*). Moreover, suitability of skipjack tuna for global monitoring of persistent organic pollutants (POPs) has been established in our previous report, indicating that the migration pattern, growth stage, and sex of these animals have no or little effect on the variations of POP residue levels in their bodies (*11*), because this species reflects POP pollution levels in seawater when and where they were collected, due to the rapid equilibrium partitioning of these compounds between seawater and body lipid in this species (*11*). These facts made skipjack tuna a suitable bioindicator for monitoring PBDE pollution also. The objectives of this study are to elucidate the global distribution of PBDEs in offshore waters and open seas, and to understand the transport behavior of these chemicals using skipjack tuna as a bioindicator.

Materials and Method

The specimens of skipjack tuna (*K. pelamis*) were collected from offshore waters of various Asian countries (Japan, Taiwan, Philippines, Indonesia, Seychelles, and Brazil, and the Japan Sea, East China Sea, South China Sea, Indian Ocean, and North Pacific Ocean) during the years 1996−2001 (Figure 1). Skipjack tuna were obtained from the fish market and fisher village after confirmation of the fishing areas. The samples from the North Pacific Ocean and Brazil were caught by fishing during a research cruise. Muscles taken from individual specimens were kept in polyethylene bags which were tested and found to contain no PBDEs. These samples were stored at −20 °C until chemical analysis. Pooled muscles of five fish from each location were employed for chemical analysis. Details of the samples are shown in Table 1.

Muscle samples were homogenized with anhydrous Na_2SO_4 and extracted using a Soxhlet apparatus with a mixture of diethyl ether and hexane. Lipid content was determined gravimetrically from an aliquot of the extract. An aliquot of the extract, after addition of 5 ng of internal standards ($^{13}C_{12}$-labeled BDE3, BDE15, BDE28, BDE47,

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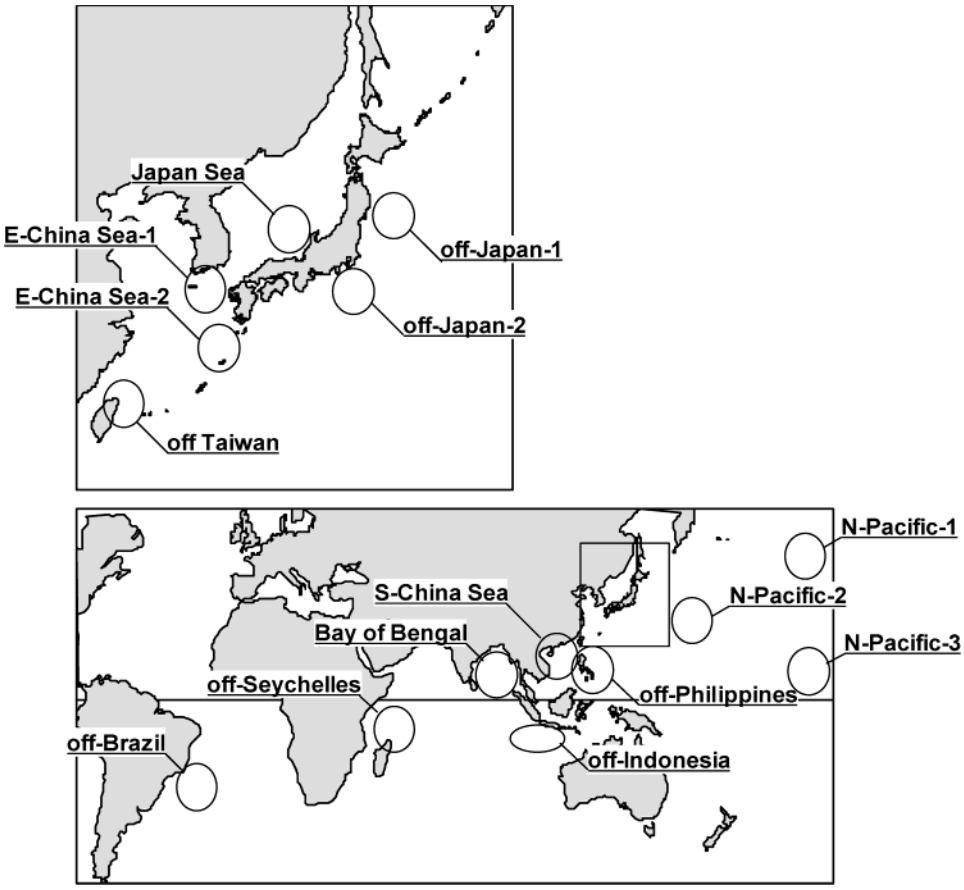

FIGURE 1. Map showing sampling locations of skipjack tuna.

TABLE 1. Biometric Data of Skipjack Tuna Collected from Asian offshore Waters, off-Seychelles, off-Brazil and Open Seas[a]

location	month/year	n	BL (cm)	BM (kg)
N-Pacific-1	08/1998	5	44 (43−45)	1.8 (1.7−2.0)
N-Pacific-2	05/1997	5	47 (43−54)	2.2 (1.7−3.5)
N-Pacific-3	03/1998	5	79 (75−80)	11 (9.0−12)
off-Japan-1	10/1997	5	49 (49−50)	2.6 (2.6−2.7)
off-Japan-2	10/1997	5	52 (49−55)	3.3 (3.1−3.7)
Japan Sea	10/1997	5	63 (62−65)	5.6 (5.5−5.8)
E-China Sea-1	10/1997	5	58 (55−60)	3.9 (3.4−4.5)
E-China Sea-2	10/1997	5	61 (60−63)	5.2 (4.9−5.4)
off-Taiwan	11/1998	5	61 (59−63)	4.9 (4.9−5.0)
S-China Sea	06/2001	1	31	0.5
off-Philippines	12/1997	5	42 (41−45)	1.5 (1.3−1.6)
Bay of Bengal	10/1998	5	47 (46−47)	2.0 (2.0−2.0)
off-Indonesia	01/1999	2	47 (46−48)	2.0 (2.0−2.0)
off-Seychelles	01/1999	3	55 (54−55)	3.6 (3.4−3.8)
off-Brazil	09/2000	4	55 (53−57)	3.7 (3.1−4.4)

[a] Figures in parentheses indicate the range. *n* = number of samples. BL = body length. BM = body mass. N-Pacific = the North Pacific. E-China Sea = the East China Sea. S-China Sea = the South China Sea.

BDE99, BDE153, BDE154, BDE183, and BDE209), was then subjected to gel permeation chromatography (GPC; Bio-Beads SX3, Bio-Rad Laboratories) for lipid removal. The GPC fraction containing organohalogens was concentrated and passed through a column of 1.5 g of activated silica gel S-1 (Wako Pure Chemical Industries Ltd., Japan) for cleanup and fractionation. The first fraction eluted with hexane contained PCBs and deca-BDE, and the second fraction eluted with 5% dichloromethane in hexane contained mono- to hepta-BDEs. $^{13}C_{12}$-labeled BDE139 was added to final solution prior to GC−MSD analysis as a recovery spike. The quantification was performed using a gas chromatograph (Agilent 6980N) equipped with a mass-selective detector (Agilent 5973) for mono- to hepta-BDEs, and a gas chromatograph (Agilent 6980N) coupled with a mass-selective detector (JEOL GC-Mate II) for deca-BDE, having an electron impact (EI). PBDEs were monitored in selective ion monitoring mode (EI-SIM) at masses of $[M]^+$ and $[M + 2]^+$ for mono- and di-BDEs, $[M + 2]^+$ and $[M + 4]^+$ for tri- and tetra-BDEs, $[M + 4]^+$ and $[M + 6]^+$ for penta- and hexa-BDEs, $[M + 6]^+$ and $[M + 8]^+$ for hepta-BDEs, and $[M - 2Br + 6]^+$ and $[M - 2Br + 8]^+$ for deca-BDEs. GC columns used for quantification were DB-1 fused silica capillaries (J&W Scientific Inc.) having 0.25 mm i.d. × 30 m × 0.25 μm film thickness for mono- to hepta-BDEs, and 0.25 mm i.d. × 15 m × 0.1 μm film thickness for deca-BDE. Eleven major congeners of PBDEs (BDE3, BDE15, BDE28, BDE47, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183, and BDE209) were quantified in this study. All the congeners were quantified using the isotope dilution method to the corresponding $^{13}C_{12}$-labeled congeners (BDE100 (penta-BDE) and BDE138 (hexa-BDE) were quantified using $^{13}C_{12}$-labeled BDE99 (penta-BDE) and BDE153 (hexa-BDE), respectively). Recovery of $^{13}C_{12}$-labeled BDE ranged between 60% and 120%. The detection limit was calculated as 3 times the procedural blank (0.02 ng/g of lipid for mono- to di-BDEs, 0.1 ng/g of lipid for tetra-BDE, 0.05 ng/g of lipid for tri- and penta- to hepta-BDEs, and 5 ng/g of lipid for deca-BDE).

Results and Discussion

Contamination Status. PBDE concentrations in skipjack tuna from Asian offshore waters, off-Seychelles, off-Brazil, and open seas are shown in Table 2. PBDEs were detected in almost all the skipjack tuna analyzed in this study, indicating widespread contamination of these compounds in the global marine environment. Concentrations of total PBDEs ranged

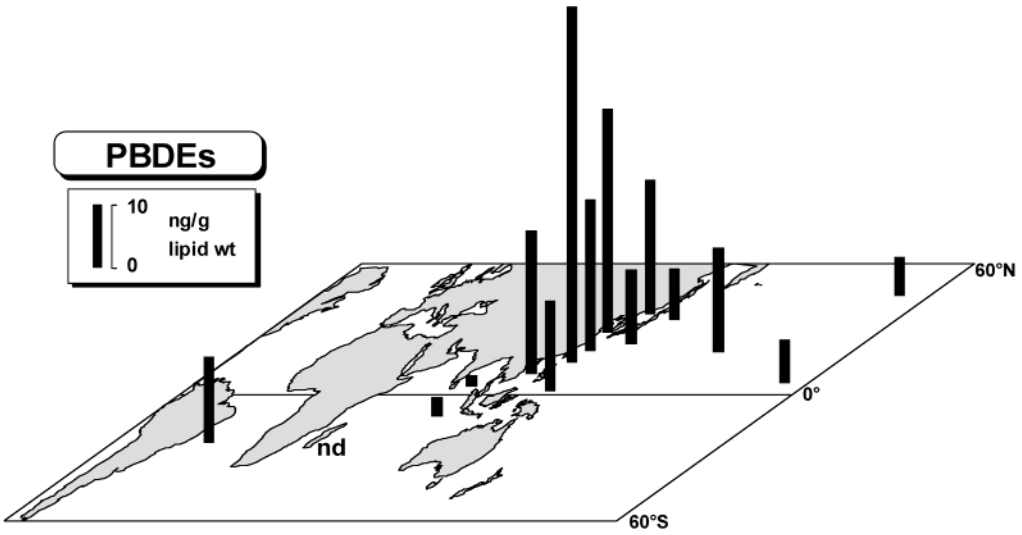

FIGURE 2. Geographical distribution of PBDE concentrations in the muscle of skipjack tuna.

TABLE 2. PBDE Concentrations (ng/g of lipid) in Pooled Muscle of Skipjack Tuna Collected from Asian Offshore Waters, off-Seychelles, off-Brazil, and in the Open Seas[a]

		concn (ng/g of lipid)											
location	lipid %	BDE3	BDE15	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE138	BDE183	BDE209	total PBDEs
N-Pacific-1	4.3	<0.02	0.030	0.48	3.1	0.56	0.43	0.92	0.24	<0.05	<0.05	<5.0	5.8
N-Pacific-2	1.3	<0.02	0.058	0.91	8.2	1.6	2.1	2.0	0.66	<0.05	0.21	<5.0	16
N-Pacific-3	4.4	<0.02	0.027	0.14	2.9	1.0	0.18	2.0	0.16	<0.05	<0.05	<5.0	6.4
off-Japan-1	5.8	<0.02	<0.02	0.35	3.6	0.62	1.7	0.98	0.39	<0.05	<0.05	<5.0	7.7
off-Japan-2	5.2	<0.02	0.028	0.67	5.3	1.2	1.1	2.2	0.62	<0.05	<0.05	<5.0	11
Japan Sea	4.0	<0.02	<0.02	0.53	8.0	2.9	2.0	5.1	1.5	<0.05	0.11	<5.0	20
E-China Sea-1	3.4	<0.02	<0.02	0.72	15	4.4	4.7	5.9	2.3	<0.05	0.085	<5.0	34
E-China Sea-2	1.4	<0.02	<0.02	0.53	9.0	3.4	2.4	5.6	1.7	<0.05	<0.05	<5.0	23
off-Taiwan	0.7	<0.02	0.17	0.82	18	9.2	4.7	16	4.2	<0.05	<0.05	<5.0	53
S-China Sea	1.1	<0.02	0.051	0.77	7.9	2.1	3.0	5.7	1.7	<0.05	0.13	<5.0	21
off-Philippines	0.6	<0.02	<0.02	0.36	5.9	1.5	2.1	2.4	0.90	<0.05	0.35	<5.0	14
Bay of Bengal	1.2	<0.02	<0.02	0.12	0.88	0.21	<0.05	0.32	0.25	<0.05	0.069	<5.0	1.8
off-Indonesia	0.7	<0.02	<0.02	<0.05	1.1	0.41	<0.05	1.1	0.43	<0.05	<0.05	<5.0	3.1
off-Seychelles	0.9	<0.02	<0.02	<0.05	<0.1	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<5.0	nd
off-Brazil	3.2	<0.02	<0.02	0.34	7.1	1.8	1.9	1.2	0.45	<0.05	<0.05	<5.0	13

[a] nd = less than the detection limit. N-Pacific = the North Pacific. E-China Sea = the East China Sea. S-China Sea = the South China Sea. Lipid % = lipid contents in pooled muscles.

from <0.1 to 53 ng/g of lipid (Table 2). These concentrations of PBDEs in skipjack tuna from offshore waters analyzed in this study were lower than those in fish collected from inland and coastal waters of Japan (*1*, *12*, *13*), European countries (*4*, *5*), and North America (*4*, *14*, *15*). This is due to the fact that samples for this study were collected from offshore waters and open seas far from anthropogenic activity.

Geographical Distribution of PBDE Concentrations. Geographical distribution of PBDE concentrations in skipjack tuna is shown in Figure 2. Although the sample number in the southern hemisphere is small (off-Indonesia, off-Seychelles, and off-Brazil), residue levels of PBDEs in skipjack tuna collected from the northern hemisphere seem to be higher than those from the southern hemisphere. This may be due to larger usage of PBDEs in the northern hemisphere as in the case of classic organochlorines such as PCBs, DDTs, etc. The highest concentration of PBDEs was detected in the sample from off-Taiwan (53 ng/g of lipid), and relatively higher concentrations of these compounds were observed around the East China Sea (E-China Sea-1 and E-China Sea-2) and the South China Sea (S-China Sea) (Table 2 and Figure 2). Lower residue levels of PBDEs were detected in samples off-Japan-1 and -2, off-Philippines, off-Indonesia, and Bay of Bengal. Ueno et al. (*11*) revealed that higher concentrations of PCBs were found in the same samples of skipjack tuna collected from offshore waters around Japan. It was suggested that PCBs have been discharged from Japan, which is a highly developed and industrialized country (*11*). However, the residue pattern of PBDEs was different from that of PCBs, with the concentrations in samples from offshore waters around Japan lower than those from the East China Sea (Figure 2). The global market demand of PBDEs (sum of penta-, octa-, and deca-BDE) in 2001 showed that Asian countries consumed about 40% of the total demand (*16*). Within the Asian demand, only 10% was used by Japan, and 90% by other Asian countries (*1*, *16*). It is known that many industrial manufacturers (computers, televisions, and other electric household equipment) are located in coastal areas of Asian developing countries and these manufactures produced and used huge amounts of plastics (*17*). Moreover, massive amounts of waste electric products (television, computer, etc.) used in developed nations such as the U.S., Japan, Canada, Korea, and Europe were exported as trash to Asian developing countries such as China, India, or Pakistan (*18*). This waste electric equipment was being hammered apart to extract some of the valuable metals, and plastic covers and casings were burned in open air (*18*). In this context, PBDEs may be released into the environment mainly from

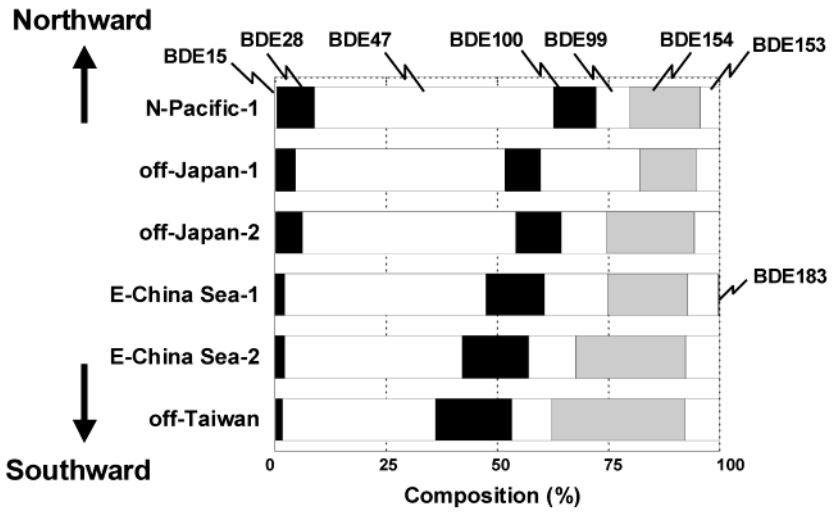

FIGURE 3. North−south profile of PBDE composition in skipjack tuna collected from Asian offshore waters.

the developing countries around the East China Sea. Results of this study indicate that some developing countries around the East China Sea are potentially the "hot spots" releasing these contaminants into the marine environment. Among the samples from the southern hemisphere, levels of PBDEs comparable with those of Asian regions were found in samples off-Brazil (Figure 2). Although several developed nations have banned or restricted the production and usage of PBDEs (*19*), relatively less restriction and monitoring study have so far been conducted in developing countries in the Asia-Pacific and the southern hemisphere. Further investigations of PBDE pollution in these regions are required.

Unexpectedly, relatively high concentrations of PBDEs were found in skipjack tuna collected from the mid North Pacific Ocean (N-Pacific-1 and N-Pacific-3), when compared with those in samples along offshore waters of Asian regions (Table 2 and Figure 2). Such a distribution pattern was also observed in PCB levels of skipjack tuna (*11*). It is known that PCBs have high transportability when compared with DDTs and PCDDs, and are causing global pollution (*20*). It has been assumed that PBDEs are less transportable than DDTs and PCDDs due to their lower vapor pressure and higher particle affinity. However, the present result suggests that PBDEs may have high transportability similar to that of PCBs, and thus have high potency to cause global pollution.

Despite the fact that the North Atlantic Ocean is considered as one of the hot spots of PBDEs because of the usage of these chemicals in this region, we could not collect skipjack tuna from this region for this study. To monitor global distribution, it is necessary to collect samples from this region also and analyze them for their PBDE levels in a further investigation.

Congener Profiles of PBDEs. Among the PBDEs analyzed, BDE47 was predominant (up to 18 ng/g of lipid), and the residue pattern of other congeners was in the order BDE154 > BDE100 > BDE99 (Table 2). The congeners of BDE3 (<0.02 ng/g of lipid) and BDE209 (<5 ng/g of lipid) were not detectable. To discuss the congener pattern of PBDEs, percentage contributions of individual congeners were compared among the locations from north to south along the Asia-Pacific region (Figure 3). It is interesting to note that the percentage contribution of lower brominated congeners (BDE15, -28, and -47) increased with increasing latitude, and the highest ratio was found in the sample from the northern colder region in N-Pacific-1. On the other hand, the proportion of higher brominated congeners (BDE153, -154, and -183) decreased with increasing latitude (Figure 3). There is a possibility that this profile may have reflected local pollution sources. However, it seems that the congener profile of PBDEs is unlikely attributed to less by local sources because the trend was clearly dependent on latitude and also the samples were collected from offshore waters and open seas far from anthropogenic activity. Therefore, the present profile suggests that lower brominated congeners (di-, tri-, and tetra-BDEs) are preferentially transported from pollution sources to northern colder regions. On the contrary, less transportable nature and highly localized characteristics may be suggested for higher brominated congeners (hexa- and hepta-BDEs). This north−south profile of PBDE compositions was similar to that of PCB compositions in skipjack tuna (*11*) and marine mammal (*21*). Moreover, PBDEs seem to have transportability similar to that of PCBs as mentioned in the section of geographical distribution. These results suggest that lower brominated congeners of PBDEs (di-, tri-, and tetra-BDEs) may have higher transportability similar to that of PCBs. Wania and Dugani (*7*) also indicated, using several models, that the lower brominated congeners of PBDEs have long-range transport potency (LRTP) comparable to that of PCBs. These results reveal that PBDEs, in particular, lower brominated congeners (di-, tri-, and tetra-BDEs), have a high potency to cause global pollution like PCBs.

Our findings indicate that PBDEs are widely transported and cause global pollution. Therefore, PBDEs should be designated as a new member to the list of POPs, and regulatory measures on the production and usage on global terms should be implemented. Some developing countries in the Asia-Pacific region are supposedly the hot spots releasing these compounds into the marine environment. Although several developed nations have banned or restricted the production and usage of these compounds (*19*), no restriction has so far been implemented in developing countries in the Asia-Pacific and the southern hemisphere. Further investigations of PBDE pollution are required in the developing countries around these regions.

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