ELSEVIER

Chemosphere 64 (2006) 287–295

CHEMOSPHERE

www.elsevier.com/locate/chemosphere

Geographical distribution of polybrominated diphenyl ethers (PBDEs) and organochlorines in small cetaceans from Asian waters

Natsuko Kajiwara [a], Satoko Kamikawa [a], Karri Ramu [a], Daisuke Ueno [a], Tadasu K. Yamada [b], Annamalai Subramanian [a], Paul K.S. Lam [c], Thomas A. Jefferson [d], Maricar Prudente [e], Kyu-Hyuck Chung [f], Shinsuke Tanabe [a,*]

[a] *Center for Marine Environmental Studies (CMES), Ehime University, 2-5 Bunkyo-cho, Matsuyama 790-8577, Japan*

[b] *National Science Museum, 3-23-1 Hyakunin-cho, Shinjuku-ku, Tokyo 169-0073, Japan*

[c] *Center for Coastal Pollution and Conservation, Department of Biology and Chemistry, City University of Hong Kong, 83 Tat Chee Avenue, Kowloon, HKSAR, China*

[d] *Southwest Fisheries Science Center, NMFS, NOAA, 8604 La Jolla Shores Drive, La Jolla, CA 92037-1508, USA*

[e] *Science Education Department, De La Salle University, 2401 Taft Avenue, 1004 Manila, Philippines*

[f] *Sungkyunkwan University, 300 Chunchun-dong, Jangan-gu, Suwon, Kyonggi-do, 440-746, Republic of Korea*

Available online 24 January 2006

Abstract

Polybrominated diphenyl ethers (PBDEs) are one of the flame retardants widely used in plastics, textiles, electronic appliances, and electrical household appliances. In this study, PBDEs and organochlorine compounds (OCs) were determined in the archived samples from the Environmental Specimen Bank for Global Monitoring (*es-BANK*) at Ehime University. The blubber of cetaceans found stranded along the coasts of Japan, Hong Kong, the Philippines and India during the period from 1990 to 2001 were employed for chemical analysis to understand the present status of contamination and the specific accumulation of PBDEs. PBDEs were detected in all the cetacean samples analyzed, and concentrations were one or two orders of magnitude lower than for PCBs and DDTs. Concentrations of PBDEs ranged from a low value of 6.0 ng/g lipid wt. in spinner dolphin (*Stenella longirostris*) from India to a high value of 6000 ng/g lipid wt. in Indo-Pacific humpback dolphin (*Sousa chinensis*) from Hong Kong. No difference in PBDE levels between coastal and offshore species from Japan was observed, implying the existence of pollution sources in this region other than Japan. Highest concentrations of PBDEs were found in animals from Hong Kong, followed by Japan, and much lower levels from the Philippines and India, suggesting that developing nations may also have pollution sources of PBDEs. Geographical distribution of PBDEs in Asian waters was different from PCBs but similar to DDTs.

Keywords: PBDEs; POPs; Organochlorines; Cetacean; Asia-Pacific; Japan

1. Introduction

Polybrominated diphenyl ethers (PBDEs), the popular flame retardants, are now a worldwide problem even in remote areas (Ikonomou et al., 2002a; Birnbaum and Staskal, 2004; Ueno et al., 2004). PBDEs are structurally similar to PCBs and DDT and, therefore, their chemical properties, persistence and distribution in the environment follow similar patterns. They have been found to bioaccumulate and there are concerns over the health effects in animals from exposure to PBDEs. They also have potential endocrine disrupting properties (Hallgren et al., 2001; McDonald, 2004). Another social concern is that PBDEs have the potential to form polybrominated dioxins and furans when burnt, and toxicity of the resultant compounds was estimated to be similar to chlorinated dioxins (Watanabe and Sakai, 2003).

* Corresponding author. Tel.:/fax: +81 89 927 8171.
E-mail address: shinsuke@agr.ehime-u.ac.jp (S. Tanabe).

0045-6535/$ - see front matter
doi:10.1016/j.chemosphere.2005.12.013

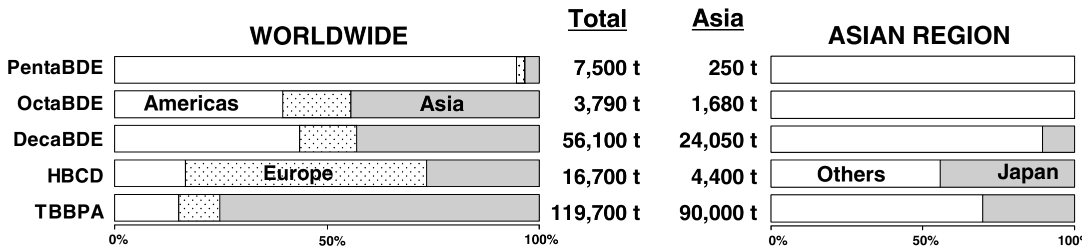

Fig. 1. Annual consumption of brominated flame retardants in the world in 2001. Data are from Watanabe and Sakai (2003) and BSEF (2004).

Growth in interest in PBDE has been as exponential as their apparent increase in the environment over the past 20–25 years in Europe (Norén and Meironyté, 2000; Thomsen et al., 2002) and North America (Ikonomou et al., 2002a; Norstrom et al., 2002; Rayne et al., 2003). However, there is still very little information on PBDE contamination in the Asia-Pacific region, compared to North America and Europe. On the Pacific coast of northern Japan, peak concentrations of PBDEs in fat tissue of northern fur seals (*Callorhinus ursinus*) occurred later than organochlorine compounds (OCs), and we concluded that it is essential to follow-up the patterns of PBDEs pollution, which may be of great concern in the future in this region also (Kajiwara et al., 2004a). The global market demand of brominated flame retardants in 2001 (Watanabe and Sakai, 2003; BSEF, 2004) showed that Asian countries consumed about 40% of the total PBDE demands. Within the Asian consumption, only 10% was used by Japan and 90% by other Asian countries (Fig. 1). It is plausible that there are some regions significantly contaminated by PBDEs in Asian developing countries apart from the developed countries including Japan (Tanabe, 2004).

Marine mammals including seals and whales are considered to be particularly sensitive to the effects of contaminant exposure because they have long life spans and feed high in the food chain. As a result, they are exposed to relatively high contaminant levels and can accumulate high concentrations in their tissues, and act as representative animals to illustrate global pollution. The present study attempts to elucidate the geographical distribution of PBDEs in cetaceans from Asian waters, especially from Japanese coastal waters, in comparison to classical OCs.

2. Materials and methods

2.1. Samples

In this study, PBDEs and OCs were determined in the archived samples from the Environmental Specimen Bank for Global Monitoring (*es-BANK*) at Ehime University. Fifty-six male specimens comprising eight cetacean species collected from Asian coastal waters during 1990 to 2001 were used (Fig. 2). The following samples collected from Japan during 1998 to 2001 were included: two coastal species including finless porpoise (*Neophocaena phocaenoides*) stranded along Seto-Inland Sea ($n = 5$) and the Pacific coast ($n = 2$), and harbour porpoise (*Phocoena phocoena*, $n = 3$), and four offshore species including Dall's porpoise (*Phocoenoides dalli*, $n = 10$), melon-headed whale (*Peponocephala electra*, $n = 5$), Pacific white-sided dolphin (*Lagenorhynchus obliquidens*, $n = 5$), and Stejneger's beaked whale (*Mesoplodon stejnegeri*, $n = 5$). Dall's porpoises are offshore species distributed around northern Japan and have two populations with distinct migration routes, *truei*-type in the Pacific Ocean, and *dalli*-type in the Japan Sea. We could obtain five samples from both populations to compare the contamination status between the Japan Sea and the Pacific. Six finless porpoises and seven Indo-Pacific humpback dolphins (*Sousa chinensis*) from Hong Kong waters found stranded in 1997–2001, two humpback dolphins and three spinner dolphins (*Stenella longirostris*) from India collected during 1990–1992, and three spinner dolphins from the Philippines in 1996 were also used in this study. Blubber samples were employed for chemical analysis. As for samples from Hong Kong, data were cited from our previous study reported by Ramu et al. (2005).

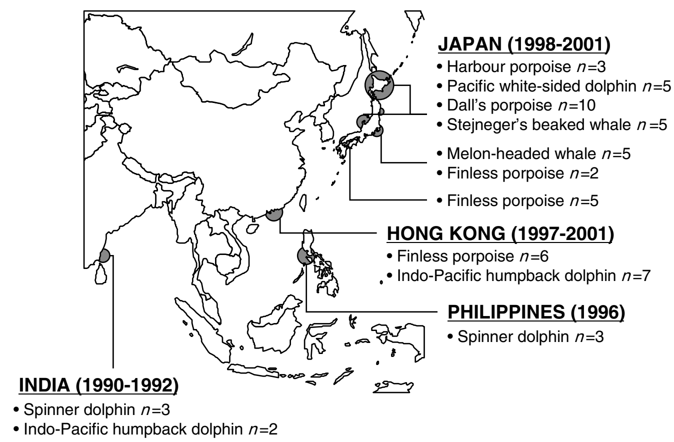

Fig. 2. Sampling locations of cetaceans from Asian waters.

2.2. Chemical analysis

Analysis of PBDEs was performed following the procedure described by Ueno et al. (2004) with slight modification. Briefly, approximately 5 g of blubber sample was ground with anhydrous sodium sulfate and extracted in a

Soxhlet apparatus with a mixture of diethyl ether and hexane for 7–8 h. An aliquot of the extract, after adding 5 ng of internal standards ($^{13}C_{12}$-labeled BDE-3, BDE-15, BDE-28, BDE-47, BDE-99, BDE-153, BDE-154, BDE-183 and BDE-209), was added to a gel permeation chromatography column (GPC; Bio-Beads S-X3, Bio-Rad Laboratories, CA, 2 cm i.d. and 50 cm length) for lipid removal. The GPC fraction containing organohalogens was concentrated and passed through 1.5 g of activated silica gel (Wakogel® S-1, Wako Pure Chemical Industries Ltd., Japan) column with 5% dichloromethane in hexane for clean up. $^{13}C_{12}$-labeled BDE-139 was added to the final solution prior to GC-MSD analysis. Quantification was performed using a GC (Agilent 6980N) equipped with MSD (Agilent 5973N) for mono- to hepta-BDEs, and GC (Agilent 6980N) coupled with MSD (JEOL GCmate II) for deca-BDE, having an electron impact with selective ion monitoring mode (EI-SIM). GC columns used for quantification were DB-1 fused silica capillary (J&W Scientific Inc.) having 30 m × 0.25 mm i.d. × 0.25 μm film thickness for mono- to hepta-BDEs, and 15 m × 0.25 mm i.d. × 0.1 μm film thickness for deca-BDE. Ten major congeners of PBDEs (BDE-3, BDE-15, BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) were quantified in this study. All the congeners were quantified using the isotope dilution method for the corresponding $^{13}C_{12}$-labeled congener. Recovery of $^{13}C_{12}$-labeled BDEs ranged between 60 and 120%.

OCs including PCBs, DDTs, HCHs, CHLs (chlordane related compounds) and HCB were analyzed following the methods described by Kajiwara et al. (2003). Another aliquot of the extract was subjected to GPC for lipid removal. The GPC fraction containing OCs was concentrated and passed through an activated Florisil column for clean up and fractionation. Quantification of PCBs and most of organochlorine pesticides was performed using a GC (Agilent 6980N) equipped with a micro-electron capture detector (micro-ECD) and an auto-injection system (Agilent 7683 Series Injector). The GC column used for OC analysis was a fused silica capillary (DB-1, 30 m × 0.25 mm i.d. × 0.25 μm film thickness, J&W Scientific Inc.). The concentration of individual OCs was quantified from the peak area in the sample to that of the corresponding external standard. The PCB standard used for quantification was a mixture of sixty-two PCB congeners (BP-MS) obtained from Wellington Laboratories Inc., Ontario, Canada. Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain total PCB concentrations.

Procedural blanks were analyzed simultaneously with every batch of five samples to check for interferences or contamination from solvents and glassware. Lipid contents were determined by measuring the total non-volatile solvent extractable material on sub-samples taken from the original extracts. The concentrations of organohalogens are expressed on a lipid weight basis unless otherwise specified.

For quality assurance and control, our laboratory participated in the Intercomparison Exercise for Persistent Organochlorine Contaminants in Marine Mammals Blubber, organized by the National Institute of Standards and Technology (Gaithersburg, MD) and the Marine Mammal Health and Stranding Response Program of the National Oceanic and Atmospheric Administration's National Marine Fisheries Service (Silver Spring, MD). Standard reference material (SRM 1945) was analyzed for selected PCB congeners and persistent OCs. Data from our laboratory were in good agreement with those for reference materials. The average percentage deviation from the certified values was 13% (range: 0.5–20%) for organochlorine pesticides and 28% (range: 1.3–57%) for PCB congeners.

Probability values less than 0.05 were considered as statistically significant using Mann–Whitney U test.

3. Results and discussion

PBDEs were detected in all the blubber samples of cetaceans from Asian waters (Table 1). A total of eight congeners of di- to hepta-BDE were detected in the samples. No BDE-3 (mono-BDE) or BDE-209 (deca-BDE) was found at the detection limits of the analysis, which were 0.01 and 0.5 ng/g lipid wt., respectively. Concentrations of PBDEs ranged from a low value of 6.0 ng/g lipid wt. in spinner dolphin from India to a high value of 6000 ng/g lipid wt. in Indo-Pacific humpback dolphin from Hong Kong (Table 1). As a general trend, concentrations of PCBs and DDTs were the highest, followed by CHLs, HCHs and HCB in that order, in Asian cetaceans. Levels of PBDEs were one or two orders of magnitude lower than that of PCBs and DDTs. PBDE residue levels observed in the present study were in the range reported in the North American studies and somewhat lower than in Europe (de Boer et al., 1998; Lindstrom et al., 1999; Covaci et al., 2002; Boon et al., 2002; Law et al., 2002). To our knowledge, this is the first comprehensive study reporting the accumulation of PBDEs in cetaceans from Asian waters.

3.1. PBDEs and OCs levels in Japanese cetaceans

3.1.1. Japan Sea vs. Pacific Ocean

To elucidate the contamination status of organohalogen contaminants around Japan, Dall's porpoise presents a unique opportunity because we could obtain samples for comparison from the Japan Sea and the Pacific Ocean on their migration routes in the year 2000. When residue levels in Dall's porpoises from different populations were compared, concentrations of PBDEs ($p < 0.01$), PCBs ($p < 0.05$), DDTs ($p < 0.01$), and HCHs ($p < 0.05$) detected in *truei*-type porpoise from the Pacific coast of northern Japan were significantly lower than *dalli*-type, which migrate inside the Japan Sea and the Sea of Okhotsk (Table 1, Figs. 3–5). Differences in PBDE concentrations were more prominent with the mean concentrations in

Table 1
Concentrations (ng/g lipid weight) of PBDEs and OCs in the blubber of cetaceans collected from Asian waters

Species	*n*	Lipid (%)	PBDEs	PCBs	DDTs	CHLs	HCHs	HCB
Japan								
1998–2000 Finless porpoise								
Seto Inland Sea	5	61 (46–70)	730 (410–1300)	120000 (44000–370000)	76000 (19000–270000)	21000 (9400–60000)	3300 (390–14000)	280 (110–870)
Pacific Coast	2	47 (42–52)	620 (590–640)	29000 (13000–45000)	32000 (11000–52000)	4800 (3800–5800)	950 (500–1400)	690 (650–720)
1999 Harbour porpoise								
	3	87 (–)	73 (24–100)	2200 (860–3000)	3300 (1100–5000)	1000 (400–1500)	730 (430–890)	710 (330–930)
2000 Dall's porpoise								
truei-type	5	87 (83–89)	57 (29–100)	9000 (6800–13000)	11000 (7700–18000)	4200 (2900–6800)	1200 (1000–1500)	860 (660–970)
dalli-type	5	86 (80–99)	530 (450–630)	18000 (12000–23000)	31000 (21000–41000)	5200 (2800–6800)	1900 (1500–3000)	580 (520–800)
2001 Melon-headed whale								
	5	68 (63–74)	320 (300–340)	24000 (15000–30000)	27000 (18000–33000)	4100 (3600–5400)	210 (170–250)	270 (220–340)
1999 Pacific white-sided dolphin								
	5	69 (46–84)	690 (100–1200)	8700 (1300–16000)	14000 (1500–26000)	3300 (350–6500)	900 (110–1700)	460 (34–1100)
2000/01 Stejneger's beaked whale								
	5	71 (49–92)	530 (390–650)	19000 (17000–20000)	110000 (83000–140000)	4500 (3600–4900)	2700 (1900–3500)	690 (530–1100)
Hong kong								
2000/01 Finless porpoise								
	6	46 (34–62)	600 (230–980)	13000 (1400–28000)	120000 (10000–260000)	740 (140–1900)	250 (32–860)	160 (75–280)
1997–2001 Indo-Pacific humpback dolphin								
	7	31 (15–56)	1900 (280–6000)	45000 (9400–83000)	190000 (51000–470000)	2200 (300–6000)	720 (110–2200)	280 (150–430)
India								
1992 Indo-Pacific humpback dolphin								
	2	57 (53–61)	11 (10–12)	2000 (1400–2600)	75000 (66000–84000)	160 (130–180)	110 (85–130)	16 (13–19)
1990–1992 Spinner dolphin								
	3	47 (37–60)	6.8 (6.0–8.0)	1600 (1100–2200)	48000 (32000–77000)	160 (110–220)	220 (130–340)	28 (16–42)
Phillippines								
1996 Spinner dolphin								
	3	38 (12–52)	36 (20–64)	3600 (2600–5400)	16000 (15000–17000)	540 (340–920)	110 (66–190)	220 (110–430)

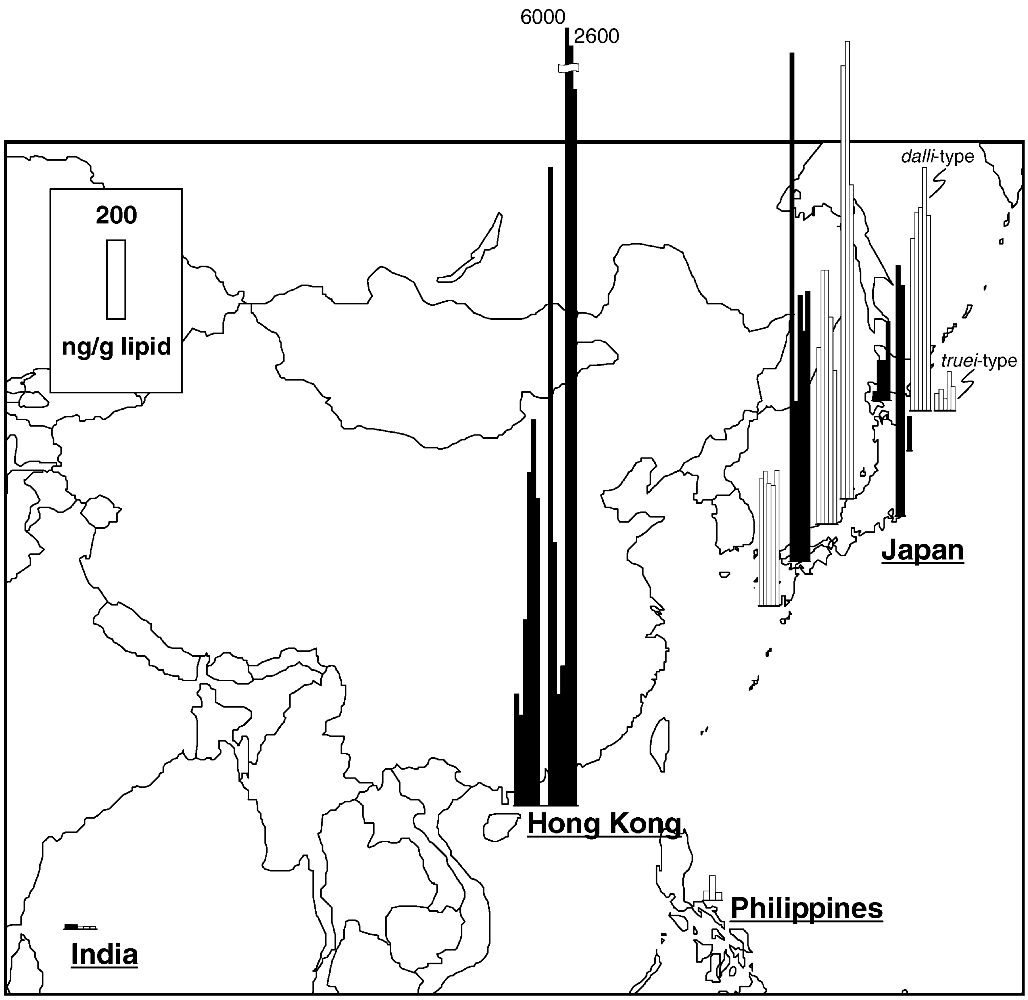

Fig. 3. Geographical distribution of PBDEs in Asian cetaceans. Black and white bars indicate coastal and offshore species, respectively.

dalli-type porpoises (530 ng/g) being tenfold higher than those in *truei*-type (57 ng/g). On the other hand, HCB residue levels were significantly higher in *truei*-type porpoises than *dalli*-type ($p < 0.05$). Our previous investigation, which focused on OC levels in Dall's porpoise collected in 1998/99 also showed significant differences in DDTs and HCHs levels between the two populations (Kajiwara et al., 2002). Moreover, residue levels of dioxin-related compounds in *dalli*-type porpoises were also found to be higher than in the specimens from the Pacific (Kajiwara et al., 2004b). These results indicate that the load of PBDEs and certain OCs in the Japan Sea is higher than in the Pacific, and the effluents from surrounding countries of the Japan Sea can be suggested as a possible source. Emissions from China and Southeast Asia are plausible contamination sources to the Japan Sea. Additionally, the closed ecosystem of the Japan Sea might have resulted in the accumulation of relatively less volatile compounds.

3.1.2. Coastal vs. Offshore species

Among Japanese cetaceans analyzed in this study, contamination levels of PBDEs were highest in finless porpoise followed by Pacific white-sided dolphin > *dalli*-type Dall's porpoise, Stejneger's beaked whale > melon-headed whale > harbour porpoise, *truei*-type Dall's porpoise (Table 1). Since finless porpoise in Japan usually inhabit coastal and semi-closed waters along the industrial zones, they might have been exposed to a variety of such chemicals and accumulate lipophilic compounds to high levels. Harbour porpoises, however, had lower PBDE and OC levels in this study even though they stay in shallow coastal waters without any wide migration. Although the reason for the low level of organohalogen contaminants in this species is still unclear, the fact that the areas they were found stranded do not have heavy human activities may partially account for this.

To elucidate if Japan plays a role as a pollution source of PBDEs and OCs, differences in residue levels between coastal and offshore species from Japan may give clues to some extent. As for PCBs, finless porpoise, which represents a coastal species in Japan, had apparently higher levels of these contaminants than offshore species (Fig. 4), reflecting continuous discharge of PCBs from Japan. PCB compositions in fur seals from the Pacific coast of

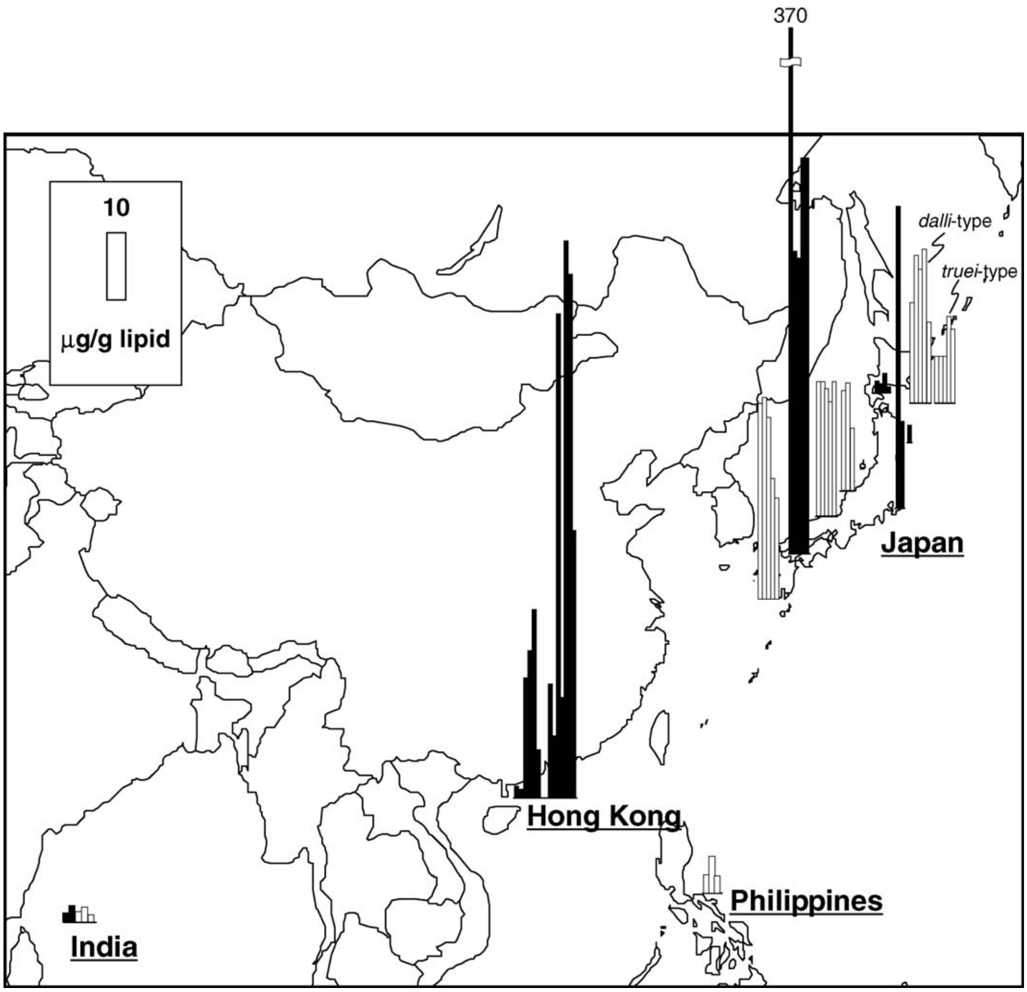

Fig. 4. Geographical distribution of PCBs in Asian cetaceans. Black and white bars indicate coastal and offshore species, respectively.

northern Japan showed no temporal variation between 1972 and 1998, suggesting a continuous input of PCBs into the marine environment in significant quantities (Kajiwara et al., 2004a). On the other hand, DDTs residue levels in Japanese offshore species seem to be comparable to or higher than coastal species (Fig. 5), implying that DDTs are not primarily released from Japan. As shown in Table 1, highest DDTs concentrations were detected in Stejneger's beaked whale stranded along the Japan Sea. Biological information on this species is very scarce and we only know that they inhabit at least the offshore area of the Japan Sea and feed on deep sea organisms. When this fact is coupled with the results on Dall's porpoise described earlier, it may be reasonable to consider that DDT levels in Stejneger's beaked whale reflect the heavy discharge of DDT from countries around the Japan Sea. As for PBDEs, interestingly, residue levels in this study showed almost comparable levels in finless porpoise and Japanese offshore species, except for *truei*-type Dall's porpoise (Fig. 3). Considering these facts, existence of pollution sources of PBDEs in the Asian region other than in Japan is suspected.

3.2. Geographical distribution of PBDEs and OCs in Asian waters

PBDE residue levels detected in Asian cetaceans were highest in specimens from Hong Kong followed by Japan (Table 1, Fig. 3). The levels in the animals near the Philippines and India were very much lower. When geographical distribution patterns of organohalogens were compared, it is much evident that for PCBs in this region, countries around Japan and Hong Kong are the main sources (Fig. 4), and for DDTs, South East Asian countries are the prime source (Fig. 5). However, the sources for the marine pollution by PBDEs in this region are obscure (Fig. 3). Perhaps, both Japan and Hong Kong are contributing equally. Our results indicate that developing nations may also have significant pollution sources for PBDEs. Our previous study to elucidate geographical distribution of PBDEs using skipjack tuna showed highest concentrations of PBDEs in samples from the East China Sea (Ueno et al., 2004). Although the reason for the high level of PBDEs in those samples is still unclear, this may be due to rapid industrialization in the recent past in some devel-

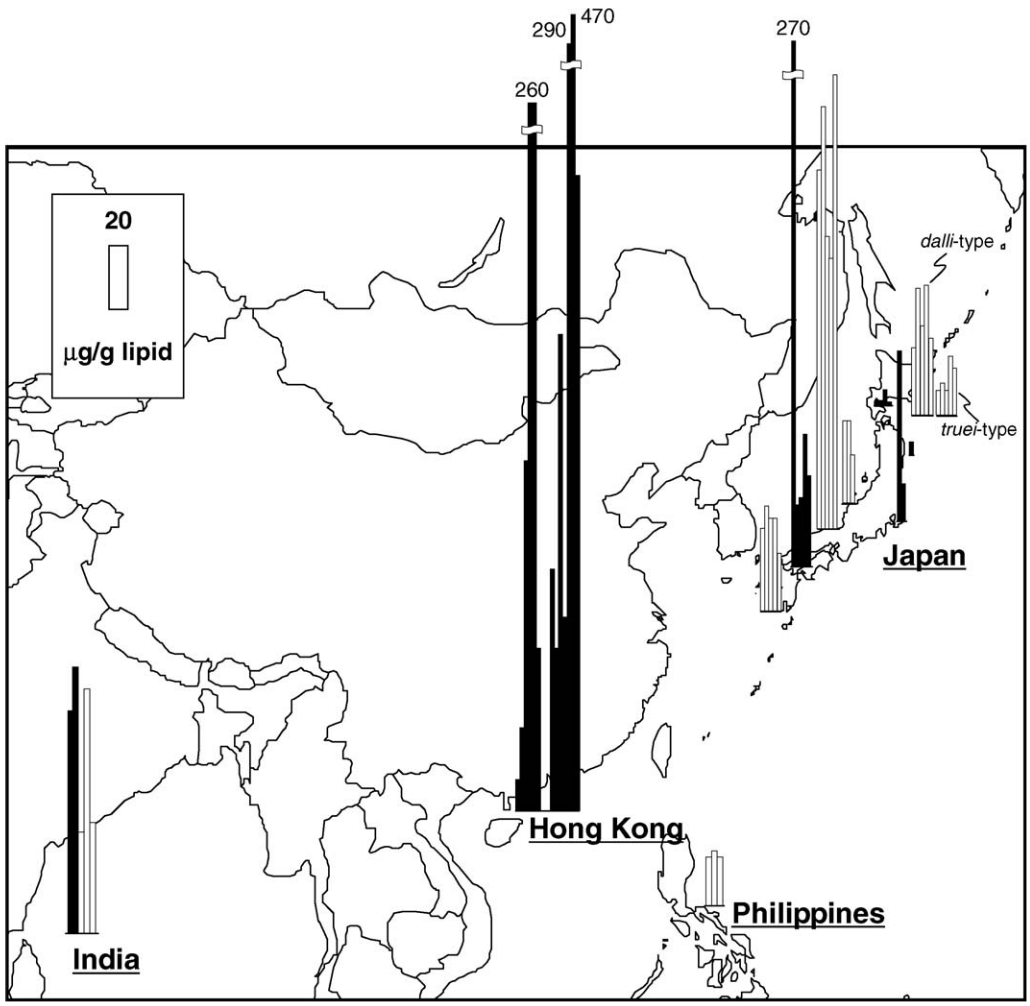

Fig. 5. Geographical distribution of DDTs in Asian cetaceans. Black and white bars indicate coastal and offshore species, respectively.

oping countries around the East China Sea (Tanabe, 2004). One of the main reasons for the high levels of PBDEs observed in Hong Kong specimens in this study may be the leaching of these compounds from the trash electrical equipments like computers, television sets, etc., imported into China from developed nations for extraction of reusable material (Schmidt, 2002). The released PBDEs might have spread to the nearby regions including Hong Kong waters, thus contaminating the cetaceans living there.

3.3. PBDE composition

BDE-47 was the predominant congener in almost all the cetaceans analyzed in this study (Fig. 6), although commercial PBDE mixtures contain BDE-47 not in such a large amount (Sjödin et al., 1998; Ikonomou et al., 2002b). Although about 80% of the PBDEs produced consist of Deca-BDE, predominantly lower brominated congeners accumulate in cetaceans possibly due to selective uptake into food chains or debromination of the Deca-BDE (Stapleton et al., 2004; Tomy et al., 2004). This is one of the paradoxes of PBDE accumulation that has not been clearly explained so far in mammals. Relative PBDE congener patterns in animals from different geographical locations may differ, mainly by the differences in contaminant sources, contaminant transport pathways, diet, and species.

The congener profiles observed in finless porpoise, Dall's porpoise and melon-headed whale around Japan having proportionately less BDE-47 and more hexa-BDEs, were clearly different from those in marine mammals reported from Europe and USA (Ikonomou et al., 2002a,b; Law et al., 2002; She et al., 2002). These studies reported PBDE accumulation patterns with BDE-47 comprising more than 60% of the total, followed by BDE-99 and -100, whereas the contribution by higher brominated congeners including BDE-153 and -154 was higher than penta-BDEs in the present study. The type of commercial PBDE mixture used in these regions might elicit these differences in the accumulation profile. In the finless porpoise specimens from Japan, the percentage of hexa-BDEs was higher than in the same species collected near Hong Kong (Fig. 6). This may be due to the different commercial mixtures of PBDEs used in these two countries. Pacific white-sided dolphins collected near northern Japan showed

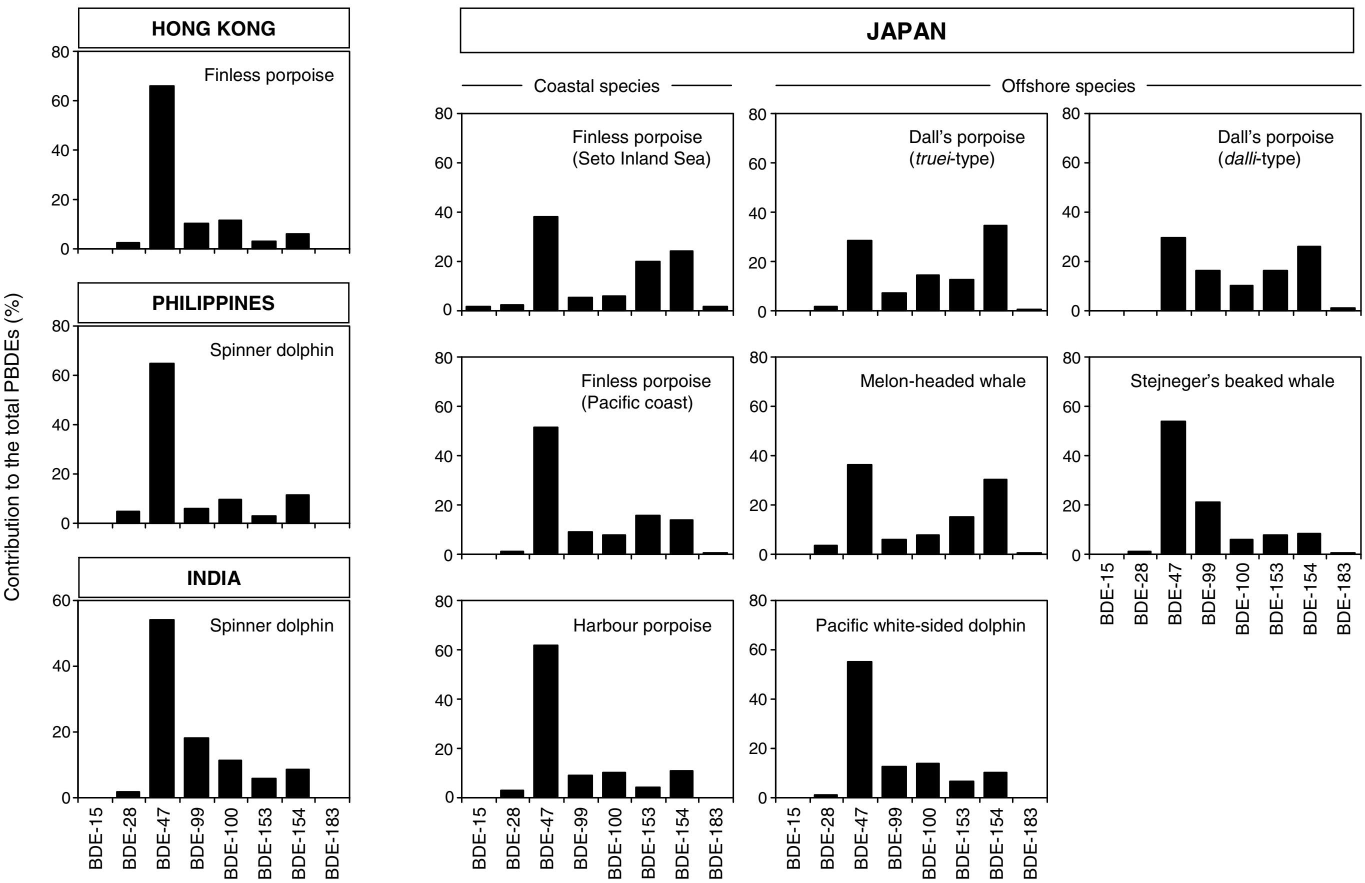

Fig. 6. PBDE congener profiles in cetaceans from Asian waters.

a different pattern with a lower percentage of hexa-BDEs than in the finless porpoise from southern Japan. This may be due to the different geographical ranges of these two species, or the species-specific metabolic capacities. Although PBDE residue levels in Dall's porpoise were clearly different among the populations as described earlier in this paper, PBDE profiles were very similar with a lower percentage of BDE-47 and higher hexa-BDEs residues in both populations. This result might suggest that PBDE profiles in cetaceans are species-specific and independent of exposure amount and/or body burdens, at least in Dall's porpoise. As previously noted, however, PBDE profiles of finless porpoises in Hong Kong and Japan were not similar and different contamination sources may thus be suspected. Further studies on specific accumulation of PBDEs in high trophic animals through food chains should be studied in detail.

4. Conclusions

PBDEs were detected in all the cetacean samples analyzed from Asian waters. Highest concentrations were found in animals from Hong Kong, followed by Japan, and much lower levels from the Philippines and India, suggesting that some developing nations may also have pollution sources of PBDEs. Considering these facts, PBDEs should be considered as an increasing pollution problem in the Asia-Pacific region, which may be of great concern in the future. It is also imperative to decipher potential sources of PBDEs and the toxicological risk for marine mammals, the top predators of the ecosystem.

Considering the widespread and increasing concentrations of PBDEs in wildlife coupled with their persistency and global transport, these compounds may be considered for inclusion in the list of persistent organic pollutants (POPs).

Acknowledgements

We are grateful to the numerous people and organizations in Japan, India, Hong Kong and the Philippines for collecting the stranded cetacean samples and carrying out detailed necropsies on these animals. The authors express their gratitude to Ms. Tamami Arakane (CMES) for help in chemical analysis.

This study was supported by Grants-in-Aid for Scientific Research (A) (No. 16201014) from Japan Society for the Promotion of Science (JSPS), and for Young Scientists (B) (Project No. 16780139) and "21st Century COE Program" from the Ministry of Education, Culture, Sports, Science and Technology, Japan, and "Material Cycles Modeling of Persistent Toxic Chemicals and its Policy

Research Applications for Recycling and Waste Management" from the Waste Management Research Grants and "Japan-Korea Co-operative Joint Research Program on Endocrine Disrupting Chemicals" from the Ministry of Environment, Japan. Financial support was also provided by the Association for the Environmental Conservation of the Seto Inland Sea, Japan. Funding for the Hong Kong stranding program has been provided by the Agriculture, Fisheries and Conservation Department of the Hong Kong Government. PKSL was supported by a grant from the Hong Kong Research Grants Council (CityU1103/03M). One of the authors (Dr. Ueno) thanks the Grant-in-Aid for JSPS Fellows from Japan Society for the Promotion of Science for the fellowship provided to him during the course of this study.

References

Birnbaum, L.S., Staskal, D.F., 2004. Brominated flame retardants: cause for concern? Environ. Health Perspect. 112, 9–17.

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.

BSEF, 2004. http://www.bsef.com/.

Covaci, A., Van de Vijver, K., DeCoen, W., Das, K., Bouquegneau, J.M., Blust, R., Schepens, P., 2002. Determination of organohalogenated contaminants in liver of harbour porpoises (*Phocoena phocoena*) stranded on the Belgian North Sea coast. Mar. Pollut. Bull. 44, 1152–1169.

de Boer, J., Wester, P.G., Klamer, H.J., Lewis, W.E., Boon, J.P., 1998. Do flame retardants threaten ocean life? Nature 394, 28–29.

Hallgren, S., Sinjari, T., Hakansson, H., Darnerud, P.O., 2001. Arch. Toxicol. 75, 200–208.

Ikonomou, M.G., Rayne, S., Addison, R.F, 2002a. Exponential increases of the brominated flame retardants, polybrominated diphenyl ethers, in the Canadian Arctic from 1981 to 2000. Environ. Sci. Technol. 36, 1886–1892.

Ikonomou, M.G., Rayne, S., Fischer, M., Fernandez, M.P., Cretney, W., 2002b. Occurrence and congener profiles of polybrominated diphenyl ethers (PBDEs) in environmental samples from coastal British Columbia, Canada. Chemosphere 46, 649–663.

Kajiwara, N., Watanabe, M., Tanabe, S., Nakamatsu, K., Amano, M., Miyazaki, N., 2002. Specific accumulation and temporal trends of organochlorine contaminants in Dall's porpoises (*Phocoenoides dalli*) from Japanese coastal waters. Mar. Pollut. Bull. 44, 1089–1099.

Kajiwara, N., Ueno, D., Monirith, I., Tanabe, S., Pourkazemi, M., Aubrey, D.G., 2003. Contamination by organochlorine compounds in sturgeons from Caspian Sea during 2001 and 2002. Mar. Pollut. Bull. 46, 741–747.

Kajiwara, N., Ueno, D., Takahashi, A., Baba, N., Tanabe, S., 2004a. Polybrominated diphenyl ethers (PBDEs) and organochlorines in archived northern fur seal samples from the Pacific coast of Japan, 1972–1998. Environ. Sci. Technol. 38, 3804–3809.

Kajiwara, N., Watanabe, M., Amano, M., Yamada, T., Tanabe, S., 2004b. Species-specific accumulation of dioxin related compounds in cetaceans collected from Japanese coastal waters. Organohalogen Compd. 66, 1554–1558.

Law, R.J., Allchin, C.R., Bennett, M.E., Morris, S., Rogan, E., 2002. Polybrominated diphenyl ethers in two species of marine top predators from England and Wales. Chemosphere 46, 673–681.

Lindstrom, G., Wingfors, H., Dam, M., van Bavel, B., 1999. Identification of 19 polybrominated diphenyl ethers (PBDEs) in long-finned pilot whale (*Globicephala melas*) from the Atlantic. Arch. Environ. Contam. Toxicol. 36, 355–363.

McDonald, T.A., 2004. A perspective on the potential health risks of PBDEs. Chemosphere 46, 745–755.

Norstrom, R.J., Simon, M., Moisey, J., Wakeford, B., Weseloh, D.V.C., 2002. Geographical distribution (2000) and temporal trends (1982–2000) of brominated diphenyl ethers in Great Lakes herring gull eggs. Environ. Sci. Technol. 36, 4783–4789.

Norén, K., Meironyté, D., 2000. Certain organochlorine and organobromine contaminants in Swedish human milk in perspective of past 20–30 years. Chemosphere 40, 1111–1123.

Ramu, K., Kajiwara, N., Tanabe, S., Lam, P.K.S., Jefferson, T.A., 2005. Polybrominated diphenyl ethers (PBDEs) and organochlorines in small cetaceans from Hong Kong waters: levels, profiles and distribution. Mar. Pollut. Bull. 51, 669–676.

Rayne, S., Ikonomou, M.G., Antcliffe, B., 2003. Rapidly increasing polybrominated diphenyl ether concentrations in the Columbia River system from 1992 to 2000. Environ. Sci. Technol. 37, 2847–2854.

Schmidt, C.W., 2002. e-Junk explosion. Environ. Hearth Perspect. 110, A188–A194.

She, J., Petreas, M., Winkler, J., Vista, P., McKinney, M., Kopec, D., 2002. PBDEs in the San Francisco Bay Area: measurements in harbor seal blubber and human breast adipose tissue. Chemosphere 46, 697–707.

Sjödin, A., Jakobsson, E., Kierkegaard, A., Marsh, G., Sellström, U., 1998. Gas chromatographic identification and quantification of polybrominated diphenyl ethers in a commercial product, Bromkal 70-5DE. J. Chromatogr. A 822, 83–89.

Stapleton, H.M., Alaee, M., Letcher, R.J., Baker, J.E., 2004. Debromination of the flame retardant decabromodiphenyl ether by juvenile carp (*Cyprinus carpio*) following dietary exposure. Environ. Sci. Technol. 38, 112–119.

Tanabe, S., 2004. PBDEs, an emerging group of persistent pollutants. Mar. Pollut. Bull. 49, 369–370.

Thomsen, C., Lundanes, E., Becher, G., 2002. Brominated flame retardants in archived serum samples form Norway: a study on temporal trends and the role of age. Environ. Sci. Technol. 36, 1414–1418.

Tomy, G.T., Palace, V.P., Halldorson, T., Braekevelt, E., Danell, R., Wautier, K., Evans, B., Brinkworth, L., Fisk, A.T., 2004. Bioaccumulation, biotransformation, and biochemical effects of brominated diphenyl ethers in juvenile lake trout (*Salvelinus namaycush*). Environ. Sci. Technol. 38, 1496–1504.

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., Tanabe, S., 2004. Global pollution monitoring of polybrominated diphenyl ethers using skipjack tuna as a bioindicator. Environ. Sci. Technol. 38, 2312–2316.

Watanabe, I., Sakai, S., 2003. Environmental release and behavior of brominated flame retardants. Environ. Int. 29, 665–682.