Contamination of Polybrominated Diphenyl Ethers in Marine Environment and Humans in Korea

Hyo-Bang Moon[1], Kurunthachalam Kannan[2], Hee-Gu Choi[3], Yong-Rock An[3], Duk-Hee Lee[4]
[1]Department of Environmental Marine Sciences, Hanyang University, Ansan 426-791, Korea
[2]Wadsworth Center, NYS Department of Health and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, NY 12201-0509, USA
[3]National Fisheries Research and Development Institute (NFRDI), Busan 619-705, Korea
[4]Department of Preventive Medicine, School of Medicine, Kyungpook National University, Daegu 702-701, Korea

Introduction

Brominated flame retardants (BFRs) have not been produced in Korea, but were imported from various countries. The use of polybrominated diphenyl ethers (PBDEs) in Korea has been steadily increasing, by approximately 10% per year during last decade (Moon et al. 2007a). PBDEs released from various sources can be transported to coastal environment, and then bioaccumulate in fatty tissues and biomagnify in marine food web (Shaw and Kannan 2009). In May 2009, the Stockholm Convention included the penta- and octa-BDEs in the list of persistent organic pollutants (POPs). Despite the reports of extensive contamination by PBDEs worldwide and the extensive use of PBDEs in Korea, a little data have been reported concerning PBDEs in air and marine environment in Korea (Moon et al. 2007a, b, c). In this paper, we investigated the occurrence and distributions of PBDEs in air deposition, marine sediment and marine biota including fish, shellfish and marine mammals from Korean coastal waters. We also determined the concentrations and accumulation profiles of PBDEs in human adipose tissues from Korea.

Materials and Methods

Nationwide monitoring of PBDEs in marine environment has been performed using the samples of atmospheric deposition, seawater, sediment and bivalves such as mussels and oysters collected from 25 coastal locations in Korea. In order to characterize the contamination sources of PBDEs in Korean coastal waters, the intensive survey on PBDE distribution have been performed in industrialized bays, Korea. The edible tissues from 26 marine species including fish, shellfish, cephalopod and crustacean were collected from the largest fish market in Korea during 2005-2007. These marine species are the most commonly consumed species and commercially important species in Korea (Moon and Choi 2009). The blubber samples were collected from three cetacean species, minke whales (*Balaenoptera acutorostrata*), common dolphins (*Delphinus capensis*) and finless porpoises (*Neophocaena phocaenoides*), entangled in fishing nets along the Korean coast during 2003-2007. The adipose tissue samples were obtained from volunteer women during liposuction procedures on patients at Daegu city in Korea. Analyses of 23 PBDE congeners in abiotic and biotic samples from Korea were performed following the methods described elsewhere (Moon et al. 2007b, c).

Results and Discussion

Nationwide monitoring of PBDEs in marine environment

Total concentrations of PBDEs ranged from 0.45 to 494 ng/g dry weight in sediments, and from 0.38

to 9.2 ng/g dry weight in bivalves from 25 coastal locations in Korea (Figure 1). The highest concentrations were found at several locations from southeastern coastal areas such as Ulsan, Onsan, Busan, Jinhae Bays, which were characterized by close to industrial complexes and large harbors in Korea. PBDE concentrations measured in our study, excluding BDE 209 (deca-BDE), in sediment and bivalves were lower than those reported from other countries; whereas deca-BDE concentrations were comparable to those reported for other countries.

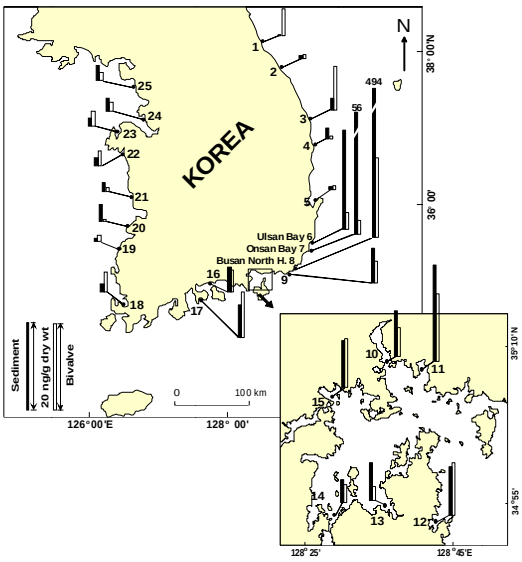

Figure 1. Concentrations of total PBDE concentrations in sediment and bivalves collected from 25 locations along the Korean coast.

Contamination sources of PBDEs in sediments from industrialized regions

Surface sediments were sampled at 146 locations representing highly industrialized bays (Ulsan, Busan and Jinhae Bays) and Shihwa Lake close to industrial complexes, to identify the contamination source of PBDEs in coastal environment of Korea. Total concentrations of PBDEs in sediments ranged from 2.0 to 2300 ng/g dry weight in industrialized bays, and from 1.7 to 18700 ng/g dry weight in Shihwa Lake. PBDE concentrations were higher at estuarine and inner part of the bay and lake close to industrial complexes and large harbors, indicating that the PBDE contamination is due to local discharge from industrial complexes and wastewater treatment plant (WWTP) (Moon et al. 2008). For Shihwa Lake, the highest concentrations of PBDEs were found at the sediments from rivers in industrial complexes (Figure 2). Multivariate statistical analyses and correlation analyses showed that deca-BDE technical mixture was the main source of PBDE contamination in Korean coastal waters, with minor contamination by octa-BDE product.

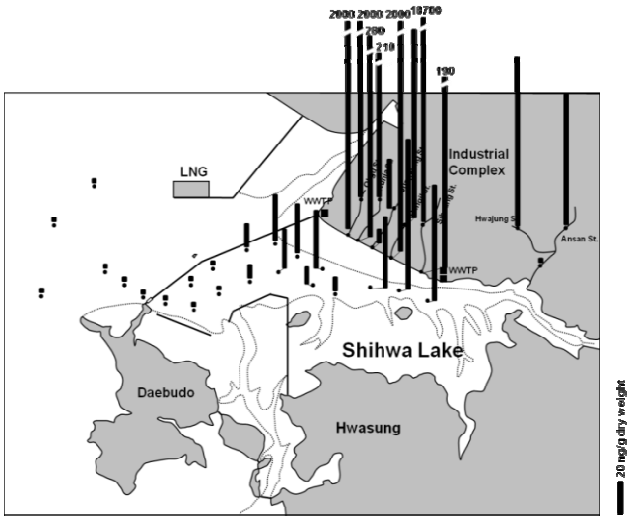

Figure 2. Distribution of PBDEs in sediments from Shihwa Lake, Korea.

Bioaccumulation of PBDEs in fish, marine mammals and humans

The total concentrations of PBDEs in 26 marine species (*n*=78) ranged from 2.2 to 180 ng/g lipid weight. The highest PBDE concentration in marine species was found for Alaska pollack, followed by flounder and rockfish. The long-lived predatory and larger fish with high lipid content showed relatively

high residues of PBDEs, compared with other species. The dietary intake of PBDEs through seafood consumption was calculated by multiplying the consumption rate of individual seafood with the corresponding PBDE concentrations. The estimated intakes of PBDEs by seafood consumption for the general population, males and females were 1.0, 1.2 and 0.9 ng/kg body weight/day, respectively. The PBDE intakes through seafood consumption according to age ranged from 0.6 to 1.4 ng/kg body weight/day. The age group with the highest exposure rate was 3-6 years old, followed by 50-64 years, <2 years and 30-49 years, which is consistent with a exposure pattern to PCDD/Fs and PCBs (Moon and Choi 2009).

The total concentrations of PBDEs in blubbers of three cetacean species from Korean coastal waters ranged from 30 to 4100 ng/g lipid weight. The overall concentrations of PBDEs in Korean cetaceans were one or two orders of magnitude lower than those of DDTs and PCBs, but were higher than those of HCHs, CHLs and HCB. This contamination pattern is similar to those reported for cetaceans from Asian coastal waters (Ramu et al. 2005; Kajiwara et al. 2006; Isobe et al. 2009). The contamination by organochlorines (OCs) such as DDTs and PCBs in Korean cetaceans were generally low, however the PBDE contamination was comparable to those reported for cetaceans in Asian coastal waters (Ramu et al. 2005; Kajiwara et al. 2006; Isobe et al. 2009). Significant inter-species difference in cetaceans was found for the PBDE concentrations, because of the difference of habitat and diet between cetacean species. Significant gender-specific difference was also found for the concentration and accumulation profiles of PBDEs in finless porpoises, due to maternal transfer and lactation from mature females to fetus.

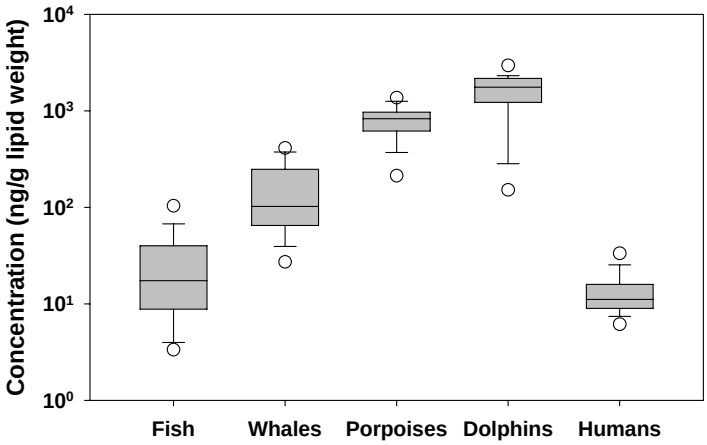

Figure 3. Comparison of total PBDE concentrations in fish (*n*=54), minke whales (*n*=27), finless porpoises (*n*=52) and common dolphins (*n*=22) from Korean coastal waters and humans (*n*=53) in Korea.

The PBDE residues in human adipose tissues from Korea were firstly determined in this study. The total PBDE concentrations in woman adipose tissues (*n*=53) from Korea ranged from 5.3 to 150 ng/g lipid weight. Similar to cetaceans, the PBDE residues in humans were lower than those of OCs such as PCBs, DDTs and CHLs. Among biota samples investigated in our study, human adipose tissues showed the lowest level of PBDEs (Figure 3). Significant correlations were found between OCs such as PCDD/Fs, PCBs and pesticides in human adipose tissues, but PBDEs showed relatively moderate correlations with legacy POPs. Our finding suggests that humans have a different exposure route to PBDEs such as inhalation associated with fine particles.

Congener profiles of PBDEs in abiotic and biotic samples

The congener profiles of PBDEs in atmospheric deposition, seawater and sediment were dominated by

deca-BDE (Figure 4), which is consistent with a high consumption of deca-BDE for the BFR market in Korea. Our result suggests that deca-BDE is already predominant in the air and then transported and distributed to coastal areas in Korea. Next to BDE 209, nona-BDEs such as BDEs 206 and 207 revealed high proportions of PBDEs to total PBDEs in abiotic samples.

The congener profiles of PBDEs for fish, marine mammals and humans were different (Figure 4). BDE 47 was a predominant congener for fish and cetaceans, however BDE 209 was a predominant congener in human adipose tissues. In particular, human adipose tissues were characterized by higher proportions for higher PBDEs than lower PBDEs. For cetaceans, the BDEs 49 and 66 comprised of 4-16% to total PBDEs in finless porpoises, which seems to be associated with debromination of higher BDEs such as octa- and deca-BDEs in marine mammals (Shaw et al. 2009).

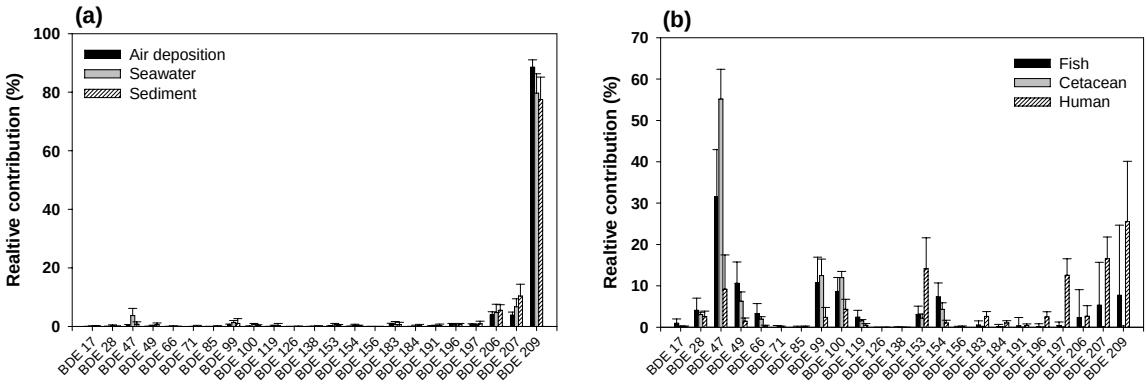

Figure 3 Representative congener profiles of PBDEs in (a) abiotic (air deposition, seawater and sediments) and (b) biotic (fish, cetaceans and humans) samples collected from Korea.

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