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A review of polybrominated diphenyl ethers and alternative brominated flame retardants in wildlife from China: Levels, trends, and bioaccumulation characteristics

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Abstract

Accelerated industrialization and urbanization, and unregulated disposal of waste of electric and electronic equipment (e-waste) in China have caused environmental pollution of brominated flame retardants (BFRs). This review summarized the levels, trends, and bioaccumulation characteristics of polybrominated diphenyl ethers (PBDEs) and other potential alternative BFRs including hexabromocyclododecanes (HBCDs), 1,2-bis(2,4,6-dibromophenoxy) ethane (BTBPE) and decabromodiphenylethane (DBDPE) in wildlife from China. PBDE levels in wildlife from China were generally higher than those from other parts in Asia, and were comparable to those from Europe but were lower than those from North America. However, wildlife from the e-waste recycling sites in South China and East China contained much higher PBDEs compared to other reports around the world, suggesting the heavy contamination of PBDEs in these regions. The alternative BFRs were also detected in wildlife, revealing that the animals are exposed to these chemicals, in addition to PBDEs. Temporal trends indicated by levels in marine mammals from South China suggested that PBDE levels increased from the beginning of 1990s to 2000s, but decreased from the middle of 2000s, followed by relatively steady levels. In contrast, HBCDs were found to be continuously increasing from 1997 to 2007, indicating the increasing usage of HBCDs in China in recent years. Compared to PBDE profiles found in other parts, aquatic species and birds from China contained relatively higher contributions of BDE-28 and 209, respectively, suggesting the possible different usage pattern of PBDEs. Future works including keeping monitoring at a reasonable scale and frequency to make sure levels near urban centers indicative of population do not increase are needed. Additionally, focus effort on e-waste recycling regions to look for impacts and to determine if regulation/controls are resulting in lower environmental contamination, and incorporation of sentinel species in monitoring efforts are recommended.

Key words: brominated flame retardants; polybrominated diphenyl ethers; wildlife; electronic waste; China

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Introduction

Polybrominated diphenyl ethers (PBDEs) are abundantly used as additives in a variety of manufactured products, e.g., plastics, rubbers, textiles, and electronic and electric devices, to prevent or retard the initial phase of a developing fire (de Wit, 2002; Alaee et al., 2003). Three commercial PBDE mixtures, i.e., the PentaBDEs, OctaBDEs, and DecaBDEs have been generally used in the manufacturing process, with each mixture made up of congeners with varying degrees of bromination (Alaee et al., 2003). The PentaBDEs and OctaBDEs have been included in the Stockholm Convention on Persistent Organic Pollutants (POPs) in May 2009, because they share many properties to traditional POPs (UNEP, 2010). The DecaBDEs has also been restricted in Europe in 2008, and is now being phased out in the U.S. by the end of 2013 (USEPA, 2010). With the bans or phase-outs of PBDEs, some non-PBDE brominated flame retardants (BFRs), such as hexabromocyclododecanes (HBCDs), 1,2-bis(2,4,6-dibromophenoxy) ethane (BTBPE) and decabromodiphenylethane (DBDPE), have prompted to be alternatives to the discontinued PBDEs in some applications (Covaci et al., 2011). Given that these alternatives share physiochemical properties similar to those of PBDEs, similar environmental fate, e.g., bioaccumulation of these compounds is expected (Wu et al., 2011).

China is now one of the world's largest manufactures and consumers of textiles, plastics, and electronics and appliances, in which large amount of BFRs are incorporated to meet the rigorous fire regulations. It is estimated that the annual demand of BFRs in these products is 7.0

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$\times10^7$–8.7 $\times10^7$ kg in 2005–2010, with an increasing rate of 7%–8% per year (Jiang, 2006; Mai et al., 2005). Aside from domestic production, the largest portion of obsolete electronic products (e-waste) generated worldwide, which contained large burdens of BFRs, has been transported to China for recycling (Wong et al., 2007). For instance, approximately 145 million tons imported e-waste were recycled in Guangdong Province, South China in 2002, containing up to 2.61 $\times10^8$ kg PBDEs (Martin et al., 2004). The BFRs contained in these products may be released to the environment during use or recycling processes, and be accumulated in wildlife and humans.

Wildlife, especially the sentimental species are the bioindicator for environmental pollution status and trends for certain regions. In this article, we review all the available data on the levels, trends and bioaccumulation characteristics of BFRs, including PBDEs, HBCDs, BTBPE, DBDPE, and other non-PBDE BFRs in wildlife from China. The BFRs data acquired and classified in this review included all available literatures published until June 2011 in peer-reviewed scientific journals. BFRs in the farmed species and food items from the market in China were not covered in this review. The aim of the present study is to summarize the current state of knowledge about these BFRs in wildlife from China. The current existing knowledge gaps and needs for further research are also discussed.

1 Levels of PBDEs

Figure 1 shows PBDE levels in wildlife including aquatic invertebrates, fish, birds, and some marine mammal species from China. As will be discussed in detail further below, PBDE concentrations are generally rising along the food chain and are also elevated in the vicinity of point sources such as e-waste recycling sites. Please note that throughout this review, we use the total PBDE levels (∑PBDEs) for comparison, although the congener numbers determined in different studies may be different.

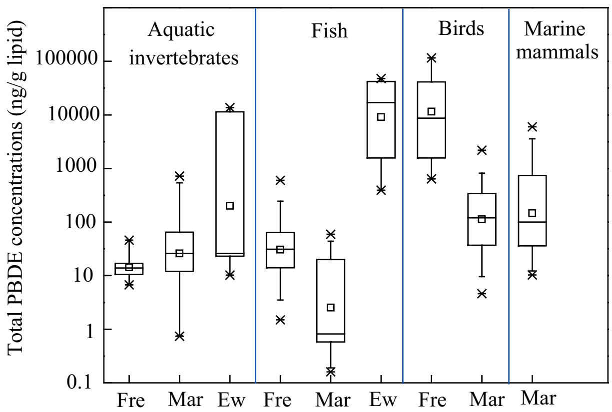

Fig. 1 Distribution of concentrations of total PBDEs in wildlife from China. Legend to the figure: center line, median; square symbol, mean; box plot edges, the 25th and 75th percentiles; whiskers, range of data values. Fre, freshwater; Mar, marine; Ew, e-waste recycling sites.

1.1 Fish

PBDEs were examined in freshwater fish species from rivers or lakes in North China, East China, South China, and Taiwan. Wang et al. (2007b) reported PBDE levels in four fish species from a small lake receiving effluent discharged from a large sewage treatment plant in Beijing, North China, finding them to occur at average ∑PBDEs of 93.8–401 ng/g lipid weight (lw). PBDEs were also detected in 8 fish species collected from Baiyangdian Lake, North China, with mean ∑PBDEs of 0.09–1.10 ng/g wet weight (ww) (4.1–114 ng/g lw) (Hu et al., 2010). Sixteen fish species were collected from the lower reaches of the Yangtze River, East China within two studies investigating the PBDE contamination status in this area in 2004–2007 (Gao et al., 2009; Xian et al., 2008). Concentrations of ∑PBDEs in these fishes ranged from 3.5 to 1100 ng/g lw. In fishes from the freshwater environment surrounding a PBDE manufacturing plant in East China (Xu et al., 2009), ∑PBDEs up to 130, 252, and 33.3 ng/g lw were found in muscle, liver, and eggs of the fish, respectively. Reports on PBDEs in wild freshwater fishes from South China are mostly from the e-waste recycling sites, e.g., Qingyuan City and Guiyu Town, both located in Guangdong Province, South China (Wu et al., 2008; Zhang et al., 2010b; Luo et al., 2007a, 2007b; Qin et al., 2009). These fishes contained very high PBDE burdens, at levels of ∑PBDEs up to 830 ng/g ww (186,000 ng/g lw) (Wu et al., 2008). Only one report was available on PBDE levels in wild fish from Taiwan (Peng et al., 2007), in which ∑PBDEs of 25.1–152 ng/g lw were reported. Overall, PBDE levels in freshwater fishes from North China, East China, and Taiwan were at low to moderate ranges of the values reported in fish around the world (de Wit, 2002; Law et al., 2003). They were comparable to or slightly higher than those from the other parts in Asia (Wang et al., 2007a; Law et al., 2008), Europe (Law et al., 2006b, 2008), and the Arctic areas (de Wit et al., 2006, 2010), but were generally lower than those from North America (Hale et al., 2003; de Wit, 2002). However, the levels of PBDEs in fishes from the e-waste recycling sites in South China were up to 3 orders of magnitude higher than those from the point sources in other parts of the world, suggesting the heavy contamination of PBDEs in these zones.

PBDEs were also detected in marine fishes from Bohai Bay and Liaodong Bay in North China, Xiamen offshore in Southeast China, Daya Bay and the Pearl River Estuary (PRE) in South China, several estuaries in Taiwan, and offshore waters and open seas of East China Sea, South China Sea, and Taiwan. Mean ∑PBDEs of 0.02–0.2 ng/g ww (0.56–6.31 ng/g lw) were found in 6 fish species from Bohai Bay (Wan et al., 2008). Slightly elevated concentrations (in averages of 1.2–24 ng/g lw) were examined in 6 fish species form Bohai Bay in a following study (Tian et al., 2010). Zhang et al. (2010a) investigated PBDEs in 8 fish species from Liaodong Bay, with mean PBDE levels of 3.86–44.9 ng/g lw. In 8 fishes collected from Xiamen offshore, the average levels of PBDEs were 0.33–0.74 ng/g lw (Li et al., 2010). Median levels of 1.9 ng/g ww (79.2 ng/g lw) and 0.1 ng/g ww (3.2 ng/g lw) of PBDEs were reported in 22 fish samples from Daya Bay and 19 fish samples from the PRE, respectively (Guo et al., 2008). Xiang et al. (2007) and Yu et al. (2009) reported ranges of 34.1–444 and 1.3–258 ng/g lw in fishes from the PRE, respectively. Fishes from 3 estuaries in Taiwan were collected to investigate the PBDE levels, with average ranges of 30.6–281 ng/g lw. A study investigating the global distribution of PBDEs showed that the skipjack tuna from offshore waters and open seas of the north part of East China Sea, the south part of East China Sea, South China Sea, and Taiwan contained 34, 23, 21, and 53 ng/g lw PBDEs, respectively (Ueno et al., 2004). Overall, PBDE levels in marine fish from China are generally lower than those from other bays or estuaries in Europe and North America (Law et al., 2006b, 2008; Hale et al., 2003; Shaw and Kannan, 2009). However, PBDE levels in Skipjack tuna from offshore waters around China were higher than those around other countries in Asia, e.g., Japan, Philippines, and Indonesia (Ueno et al., 2004).

1.2 Aquatic invertebrate organisms

Aquatic invertebrates especially mollusks are widely used as sentinel organisms for monitoring chemical contaminants in waters because they, being filter feeders and directly contacting with sediments, are especially susceptible to bioaccumulating contaminants.

PBDEs were examined in several freshwater invertebrates from Chinese environment, including river snail, swan-mussel, shrimp, and crab from Baiyangdian Lake in North China, shrimp and crab from the Yangtze River in East China, mud snail from an e-waste recycling site in East China, and Chinese mysterysnail, the Hydrobiidae, and prawn from an e-waste recycling site in South China. In the Baiyangdian Lake, swan-mussel contained the lowest levels of PBDEs (25.4–58.9 pg/g ww) of the 13 aquatic species sampled, while shrimp (116–218 pg/g ww, 5.6–18.1 ng/g lw), crab (106–173 pg/g ww, 7.2–14.3 ng/g lw), and river snail (353 pg/g ww, 46.0 ng/g lw) harbored PBDE levels even higher than most of the fish species, due to their directly contact with or ingestion of the sediment which contained high levels of PBDEs (Hu et al., 2010). Shrimp and crab from the Yangtze River have comparable PBDE levels to those from Baiyangdian Lake, with concentrations of 12.6–15.9 and 9.0–27.0 ng/g lw, respectively (Gao et al., 2009). Similar to fish, invertebrates from the e-waste recycling site (Wu et al., 2008; Zhang et al., 2010b) contained much higher PBDE levels than those from other parts in China. For example, PBDE concentrations reached up to 363 ng/g ww (18,200 ng/g lw) in shrimp, which are 3 orders of magnitude greater than those from Baiyangdian Lake (average of 11.2 ng/g lw) (Hu et al., 2010) and the Yangtze River (average of 13.9 ng/g lw) (Gao et al., 2009).

PBDEs were also investigated in several marine invertebrates from the bays, offshore areas, and estuaries in China, e.g., Bohai Bay, Jiaozhou Bay, Liaodong Bay, and Laizhou Bay in North China, Xiamen offshore in Southeast China, and the PRE in South China. Mean concentrations of 10.5, 13.1, 21.3, 78.2, and 11.3 pg/g ww (corresponding to 0.16, 0.33, 0.34, 1.09, and 0.19 ng/g lw and 0.11, 0.06, 0.14, 0.37, and 0.04 ng/g dry weight (dw), respectively) were found in the bay scallop, short-necked clam, veined rapa whelk, burrowing shrimp, and crab from Bohai Bay, North China, respectively (Wan et al., 2008). However, much higher PBDE concentrations were examined in invertebrates from this bay in a recent study, with mean concentrations of 27.1, 0.46, 56, 4.4, 3.3, 2.3 and 3.5 ng/g lw in mud snail, veined rapa whelk, ark shell, oyster, octopus, mantis shrimp and helice crab, respectively (Tian et al., 2010). In contrast, slightly higher PBDE levels (in average of 9.1 ng/g lw PBDEs) were found in mussels from Jiaozhou Bay, North China (Pan et al., 2007), and in short-necked clam (9.3 ng/g lw), Mactra quadrangularis (10.2 ng/g lw), rock shell (9.9 ng/g lw) and Chinese mitten crab (3.7 ng/g lw) from Liaodong Bay, North China (Zhang et al., 2010a). Elevated PBDE levels (in averages of 230–720 ng/g lw) were reported in four shellfish species collected from the neighborhood of a BFR manufacturing site in Laizhou Bay, North China (Jin et al., 2008). PBDE levels of (1.26 ± 0.14) and 0.72–1.43 ng/g lw were reported in the clam and crab, respectively, both collected from the Xiamen offshore, Southeast China (Li et al., 2010). Yu et al. (2009) reported 18–27, 14–29, and 6.4–13 ng/g lw of PBDEs in sand swimming crab, samoan crab, and ark shell from the PRE, South China, respectively, which are slightly higher than those reported in the marine invertebrates from North China. This is consistent with the result found in a study examining PBDE levels in mussel samples from the coastal waters of Asian countries, including several sites in North China, East China, and South China (Ramu et al., 2007). PBDE levels in mussel samples from the southern coast of China (25–130 ng/g lw), especially from Hong Kong coastal waters were much higher than those from the eastern coast of China (5.1–66 ng/g lw) and the northern coast of China (only one sampling site, 63 ng/g lw), possibly due to manufacturing operations as well as the e-waste recycling activities which are located along the southern coast of China (Ramu et al., 2007). It was also shown that PBDE levels in mussels from the coasts of China were comparable to or even higher than those from Philippines (69–140 ng/g lw), Japan (6.2–49 ng/g lw) and Korea (8.5–440 ng/g lw), but were much higher than those from other Asian countries (0.66–16 ng/g lw), e.g., Cambodia, India, Indonesia, Malaysia, and Vietnam (Ramu et al., 2007), suggesting that significant sources of PBDEs exist in and around China.

1.3 Amphibians and reptiles

Amphibians and reptiles have been used as important environmental stress bioindicators (Burkhart et al., 2000; De Solla et al., 2007). However, until now there is little information on PBDEs in the wild amphibians and reptiles from Chinese environment (Wu et al., 2008, 2009; Wang et al., 2007b) and in the world in general, despite the fact that these animals are sensitive to the environmental chemicals (Burkhart et al., 2000; Hale et al., 2002; Wu et al., 2009).

Only one study investigated PBDEs in wild amphibians from China. Wu et al. (2009) reported PBDE levels in frog

Rana limnocharis from an e-waste recycling site in South China, with levels of 0.63–11.6, 4.21–56.2, and 10.7–125 ng/g ww (92–1600, 240–3300, and 92–1600 ng/g lw) in the muscle, liver, and eggs, respectively. Levels of BDEs 47 and 99 in these frogs were 2–10 times higher than those reported in frog *Rana temporaria* collected from Sweden (Ter Schure et al., 2002), the only available data on PBDE levels in wild frogs for comparison.

For wild reptiles, PBDEs in water snake (*Enhydris chinensis*) and Chinese softshell turtle (*Chinemys reevesii*) from Chinese environment were examined (Wu et al., 2008; Wang et al., 2007b). Water snake collected from an e-waste recycling site contained up to 1700 ng/g ww (190,000 ng/g lw) of PBDEs (Wu et al., 2008), which was higher than those in the top predator fish species from the same sampling site. Levels of PBDEs in Chinese softshell turtle (average of 96.5 ng/g lw) from Gaobeidian Lake, North China were slightly lower than those in fish species collected in the same sampling site (Wang et al., 2007b).

1.4 Birds

Birds are widespread and sensitive to environmental changes, and generally occupy high trophic levels in the food chain. Therefore, birds have been used intensively as sentinel species for monitoring the levels and effects of environmental contaminants (Gauthier et al., 2008; Norstrom et al., 2002). Reports on PBDEs in birds inhabiting in China are limited, and all these species are from North China or South China.

PBDEs were investigated in tissues or eggs of both aquatic and terrestrial bird species from North China. PBDEs levels in muscle of seabird species herring gull (*Larus argentatus*) from Bohai Bay were (32.8 ± 5.1) ng/g lw (Wan et al., 2008), which were much lower compared to those in aquatic birds around the world (Chen and Hale, 2010). Gao et al. (2009) reported PBDE levels in eggs of six waterbird species, i.e., Saunders's gull (*Larus saundersi*), common tern (*Sterna hirundo*), kentish plover (*Charadrius alexandrinus*), black-winged stilt (*Himantopus himantopus*), oriental pratincole (*Glareola maldivarum*), and common coot (*Fulica atra*), and one terrestrial bird specie, ring-necked pheasant (*Phasianus colchicus*) from the Yellow River Delta, with median levels of 146, 78, 54, 30, 33, 11, and 90 ng/g lw, respectively. Levels of 206 and 174 ng/g lw were reported in black-headed gull (*Larus ridibundus*) and black-tailed gull (*Larus crassirostris*) from Liaodong Bay, respectively (Zhang et al., 2010a). These levels were comparable to the concentrations reported in seabirds and raptors around the world, and were not expected to have adverse reproductive implications when only PBDEs were considered (Gao et al., 2009). Chen et al. (2007a) investigated PBDE levels in muscle, liver, and kidney of eight terrestrial bird species, i.e., the common kestrel (*Falco tinnunculus*), sparrowhawk (*Accipiter nisus*), Japanese sparrowhawk (*Accipiter gularis*), scops (*Otus sunia*), long-eared (*Asio otus*) and little owl (*Athene noctua*), and common (*Buteo buteo*) and upland buzzard (*Buteo hemilasius*), from Beijing area. Mean levels of 140–12300, 15–12200, and 76–5330 ng/g lw of PBDEs were found in muscle, liver, and kidney of these birds, respectively, with large differences among the species. Even in the same species, PBDEs levels also varied largely, e.g., PBDE levels ranged 279–31,700 and 126–40,900 ng/g lw in muscle and liver of kestrel, respectively. Levels of PBDEs in the Chinese kestrels were several orders (generally 1–3) of magnitude higher than those reported for avian species from other countries, e.g., birds in Belgian and UK, and bird eggs from Greenland, Norwegian, Sweden, Japan, and North American (Chen et al., 2007a). The max PBDE levels detected in muscle (31,700 ng/g lw) and liver (40900 ng/g lw) of kestrel rivaled the highest reported in birds to date (Chen et al., 2007a). More recently, Yu et al. (2011) investigated PBDE levels in common kestrels (*Falco tinnunculus*) ant its prey, Eurasian tree sparrow (*Passer montanus*), with levels of 120–8500 ng/g and 100–2600 lw, respectively.

Levels of PBDEs were determined in the tissues, eggs, and sera of several waterbird species from South China. Luo et al. (2009) reported median PBDE levels of 37–2200 ng/g lw in muscles of five waterbird species, i.e., white-breasted waterhen (*Amaurornis phoenicurus*), slaty-breasted rail (*Gallirallus striatus*), ruddy-breasted crake (*Porzana fusca*), Chinese-pond heron (*Ardeola bacchus*), and common snipe (*Gallinago gallinago*), from an e-waste recycling site in South China. Among these species, Chinese-pond heron contained the highest PBDE levels (530–2500 ng/g lw), possibly due to their highest trophic levels. The concentrations of PBDEs in the five birds species were generally at the high end of the world-wide range, e.g., PBDEs in Chinese-pond heron were higher than those in muscles of fish-eating birds from Bohai Bay, North China (Wan et al., 2008), tissues of peregrine falcons sampled near urban areas of California, USA (Holden et al., 2009; Meng et al., 2009), and some bird species from Europe (Kunisue et al., 2008; Jaspers et al., 2006; Lundstedt-Enkel et al., 2005). Lam et al. (2007) collected eggs of two ardeid species, the little egret (*Egretta garzetta*) and the black-crowned night heron (*Nycticorax nycticorax*) from the coastal areas off three South China cities, i.e., Hong Kong, Xiamen, and Quanzhou, and another two ardeid species, cattle egret (*Bubulcus ibis*) and Chinese pond heron (*Ardeola bacchus*) from Xiamen, to assess the risk for potential effects of PBDEs on these waterbirds. Concentrations of PBDEs in ardeid eggs from Hong Kong, Xiamen, and Quanzhou ranged from 140–1000, 30–550, and 140–380 ng/g lw, respectively. These levels were comparable to or even higher than those in eggs of waterbirds from the Yellow River Delta, North China (Gao et al., 2009), and those from Europe (Karlsson et al., 2006). Spatial and interspecies variations of PBDEs were found, with highest levels in the little egret (mean concentration of 470 ng/g lw) and the black-crowned night heron (mean concentration of 400 ng/g lw) from Hong Kong. Concentrations of BDEs 99 and 209 in the eggs may have neurobehavioral effects and oxidative stress to the Ardeid populations, suggesting the potential risks (Lam et al., 2007). Liu et al. (2010) investigated PBDE levels in the sera of six aquatic bird species, i.e., white-breasted waterhens (*Amaurornis phoenicurus*), pintail snipes (*Gallinago stenura*), Chinese pond heron (*Ardeola bacchus*), lesser coucals (*Centropus bengalensis*), five spotted doves (*Streptopelia chinensis*), and Eurasian collared doves (*Streptopelia decaocto*), from an e-waste recycling site in South China. The total PBDE concentrations ranged from 0.64 ng/g lw in white-breasted waterhen to 580 ng/g lw in lesser coucal (*Centropus bengalensis*). The levels of PBDEs in sera of lesser coucal (*Centropus bengalensis*) from South China (two samples, 8.4 and 2.2 ng/g ww) were comparable to those in plasma of birds from North America (McKinney et al., 2006), but they were 2–9 times lower than those in glaucous gull's blood from the Norwegian Arctic (Verreault et al., 2005).

1.5 Aquatic mammals

Due to their high position in the food chain and the elevated exposure in the aquatic environment, freshwater and marine mammals often exhibit high residues of environmental contaminants.

Only one report existed on PBDEs in freshwater mammals from China environment. Yang et al. (2008) determined PBDEs in blubber, liver, kidney, stomach, small intestine, and brain tissues of five Yangtze finless porpoise (*Neophocaena phocaenoides asiaeorientalis*). This specie is the sole freshwater subspecies of finless porpoise, living only in the middle and the lower reaches of the Yangtze River, China, and its appended lakes. The calculated PBDE concentrations ranged from 8.9 to 267 ng/g lw, with levels of 12.2–72.8, 14.1–70.8, 10.2–60.0, 13.6–41.0, 5.32–17.9, and 7.4–24.6 ng/g lw in blubber, liver, kidney, stomach, small intestine, and brain, respectively. Relatively higher levels of PBDEs were found in the calf among the individuals, due to the maternal transfer of PBDEs during lactation. Levels of PBDEs in the Yangtze finless porpoise were comparable to or even lower than those reported in other cetaceans from various regions, with concentrations ranged from ng/g lw to μg/g lw levels (Johnson-Restrepo et al., 2005; Lam et al., 2009; Law et al., 2002; Ramu et al., 2005, 2006; Wolkers et al., 2004).

PBDE levels were investigated in several marine mammals from South China. Ramu et al. (2005, 2006) determined PBDEs in thirteen finless porpoises (*Neophocaena phocaenoides*) and seven Indo-Pacific humpback dolphins (*Sousa chinensis*) stranded in Hong Kong coastal waters, between 1997 and 2001. PBDEs ranged 84–980 and 280–6000 ng/g lw in blubber of finless porpoises and humpback dolphins, respectively, with higher levels in humpback dolphins. For both species, blubber contained the highest PBDEs compared to the other tissues. In a following study, Lam et al. (2009) reported PBDE levels in blubber samples of Indo-Pacific humpback dolphins (*Sousa chinensis*) and finless porpoises (*Neophocaena phocaenoides*) stranded in Hong Kong waters, between 2002 and 2007 and between 2003 and 2008, respectively. PBDE levels in humpback dolphins and finless porpoises ranged from 280 to 51,100 (mean of 3590) and from 103 to 6789 (mean of 1113) ng/g lw, respectively. PBDE residues in cetaceans from Hong Kong waters were apparently higher than those in marine mammals from other countries such as Japan, India, and Philippines, and the South Atlantic US coast (Kajiwara et al., 2004, 2006), and were comparable to or higher than harbor mammals from San Francisco Bay and British Columbia (Ikonomou et al., 2002; She et al., 2002), suggesting the heavy contamination of PBDEs in South China.

2 Levels of alternative BFRs

HBCDs and other alternative BFRs have been detected in the aquatic species, birds, and marine mammals from Chinese environment, most of which are those from South China, with sparse data from other regions in China.

2.1 HBCDs

Occurrence of HBCDs was investigated in aquatic species from the Yangtze River, several e-waste recycling sites, and nine coastal cities in China. Xian et al. (2008) reported HBCDs levels (sum of α-, β-, and γ-HBCDs) of 12–330, 24–91, and 11–240 ng/g lw in muscle, liver, and egg tissues, respectively, of 9 fish species collected from lower reaches of the Yangtze River. Zhang et al. (2009) determined HBCDs in three aquatic species from the streams in an e-waste recycling site in China, with levels ranged from 123 to 3530 ng/g lw. In the aquatic species from another e-waste recycling site in South China, HBCD levels ranged from 11 to 2370 ng/g lw (Wu et al., 2010). Concentration ranges of 0.62–8.7 and 0.57–10.1 ng/g lw were reported in large yellow croaker and silver pomfret from nine costal cities, respectively (Xia et al., 2011). HBCD levels in fish from the Yangtze River were higher than those in fish from several lakes in North America such as Lakes Winnipeg and Ontario, but were much lower than those in fish from sites confirmed as HBCDs hot spots in Europe (Covaci et al., 2006). However, aquatic species from the e-waste recycling sites contained comparable HBCDs levels to those from contaminated sites in Europe (Covaci et al., 2006), suggesting that the e-waste recycling site is likely to be another HBCD hot spot, in addition to HBCDs or HBCD-retarded material production facilities.

Only one report for HBCDs in birds from China existed. Birds from an e-waste recycling site in South China harbored very high levels of HBCDs, with concentrations of < 3.0–200 ng/g lw (He et al., 2010). The HBCDs are at the high end of the worldwide figures, suggesting the possible contamination of HBCDs in the e-waste recycling site.

Levels of HBCDs were investigated in two marine mammals from the costal waters of South China. Isobe et al. (2007) determined HBCDs in Indo-Pacific humpback dolphins (*Sousa chinensis*) collected between 1997 and 2001 and finless porpoises (*Neophocaena phocaenoides*) between 1990 and 2001 from Hong Kong waters, with levels of 4.7–55 and 31–380 ng/g lw, respectively. Lam et al. (2009) detected HBCDs in blubber of Indo-Pacific humpback dolphins and finless porpoises collected between 2002 and 2007 and between 2003 and 2008, respectively. HBCD levels ranged from 32 to 519 ng/g lw (mean of 168) and from 4.1 to 501 (mean of 55) ng/g lw in humpback dolphins and finless porpoises, respectively. Concentrations of HBCDs in these cetaceans were significantly correlated ($p < 0.05$) with those of PBDEs, suggesting similar uptake pathway/kinetics of these contaminants. HBCDs levels in marine mammals from South China were lower than those from European regions such as the United Kingdom (up to μg/g lw levels) (Law et al., 2006c, 2008; Zegers et al., 2005), and were comparable to those from Japan (Isobe et al., 2009), but were higher than those from North America such as the United States (Johnson-Restrepo et al., 2008; Stapleton et al., 2006). As Lam et al. (2009) pointed out, geographic differences in the use of HBCDs, and differences in bioaccumulation and metabolism among species might cause the observed different levels among Asia, Europe, and North America.

2.2 Other non-PBDE BFRs

Several studies investigated levels of BTBPE, DBDPE, and other non-PBDE BFRs in the fish and other aquatic organisms from an e-waste recycling region in South China. Shi et al. (2009) detected BTBPE in 6 of 10 fish samples collected from an e-waste recycling region, South China, with levels up to 0.15 ng/g lw. In the aquatic species from the same e-waste recycling region (different site), Zhang et al. (2010b) reported levels up to 40, 240, 1.8, and 37 ng/g lw for BTBPE, hexabromobenzene (HBB), pentabromotoluene (PBT), and pentabromoethylbenzene (PBEB), respectively. Wu et al. (2010) reported average ranges of 44.7–518, 14–338, 197–3100, 4–256, and 1.2–106 ng/g lw for BTBPE, DBDPE, HBB, PBEB, and PBT, respectively, in 6 aquatic species collected from a natural pond surrounded by e-waste recycling plants. Levels of these chemicals from the e-waste recycling site were much higher than those from other reports, e.g., levels of BTBPE and DBDPE were 1–4 orders of magnitude higher than the concentrations in fish from North America (Ismail et al., 2009; Law et al., 2006a), suggesting that the aquatic species in the e-waste recycling site in South China are exposing to high levels of these chemicals, in addition to PBDEs and HBCDs.

BTBPE and DBDPE were also reported in birds from China. BTBPE and DBDPE were 100% detectable in the muscle, liver, and kidney of birds (9 samples) from an e-waste recycling site, South China, with ranges of 0.12–2.41 and 12.1–124 ng/g lw for BTBPE and DBDPE, respectively (Shi et al., 2009). In the two following studies (Luo et al., 2009; Zhang et al., 2011), BTBPE, DBDPE, PBEB, and PBT were detected in muscle of the five aquatic bird species from the e-waste recycling region in South China, with median levels of < 0.26–3.3, 10–180, 1.6–33, and 0.1–13 ng/g lw, respectively. While in the eggs of five aquatic and one terrestrial bird species collected from North China, DBDPE had lower detection frequencies (54%), and lower levels (median levels of 0.1–1.7 ng/g lw) (Gao et al., 2009) compared to those in birds from the e-waste recycling site in South China. Levels of DBDPE in birds from the e-waste recycling region were compared to or higher than those reported in North America (Gauthier et al., 2009), but concentrations of these chemicals in bird eggs from North China were one order of magnitude lower than other reported values (Gauthier et al., 2009; Luo et al., 2009).

Only one study investigated BFRs other than PBDEs and HBCDs in marine mammals from China. Lam et al. (2009) reported levels of 2-ethylhexyl 2,3,4,5-tetrabromobenzoate (TBB), bis(2-ethylhexyl)-tetrabromophthalate (TBPH), and hexachlorocyclopentadienyldibromocyclooctane (HCDBCO), in blubber samples of Indo-Pacific humpback dolphins (*Sousa chinensis*) and finless porpoises (*Neophocaena phocaenoides*) from Hong Kong waters, South China. HCDBCO was below the detection limit (< 0.04 ng/g lw) in the two species, and TBB was only detected in finless porpoises, with mean levels of 5.6 ng/g lw. TBPH was detectable in both species, with mean levels of 0.51 and 342 ng/g lw in humpback dolphins and finless porpoises, respectively. Levels of TBPH were comparable to HBCDs in porpoise samples, suggesting marine mammals in South China waters may harbor high levels of these novel BFRs, in addition to PBDEs and HBCDs.

3 Temporal and spatial trends

3.1 Temporal trends

The temporal trends of BFRs levels in wildlife from China have been of serious social concern, because a trend of increasing levels of some BFRs such as PBDEs has been recognized in sediment cores from China (Chen et al., 2007b; Mai et al., 2005). There is therefore a serious question as to whether levels of BFRs in wildlife from Chinese environment might also continue to increase, causing adverse effects to these species and humans in the future. However, studies on temporal trends of BFRs in wildlife from China are limited, most of which are focusing on time trends of PBDEs and HBCDs in the marine fish and mammals from South China.

PBDE concentrations in finless porpoises increased significantly (six fold, $p < 0.01$) from 1990 to 2001 from Hong Kong waters, South China, suggesting an increasing trend of PBDEs in the marine mammals during this period (Ramu et al., 2006). Lam et al. (2009) investigated temporal trends of PBDEs in two marine mammals, Indo-Pacific humpback dolphins and finless porpoises collected from 1997 to 2008 from Hong Kong waters, South China. No significant time trends in PBDEs in the two species were observed from 1997 to 2008. However, the investigation suggested that in humpback dolphins, PBDEs increased from 1997 to 2003, and decreased from 2004 to 2006, followed by relatively steady levels after 2006. These results were generally consistent with the patterns observed in wildlife from other parts of the world, and possibly reflected the decreased use of PBDEs in consumer products in recent years because of the control on their production and usage (Lam et al., 2009).

Consistent with the observation in humpback dolphins from Hong Kong waters, PBDE levels in fish species from the PRE, South China seem to be decreased between 2004 and 2007. Levels of PBDEs in fish annually collected between 2005 and 2007 from the PRE were significantly lower than those in biota samples collected in the same sampling area in 2004 (Xiang et al., 2007), indicating that beginning of a decreasing trend for PBDE levels in biota from the PRE is possible (Yu et al., 2009). From 2005 to 2007, a decrease trend for PBDEs was also observed in several fish species from the PRE, although this trend was not statistically significant (Yu et al., 2009).

Different from PBDEs, HBCDs in humpback dolphins from Hong Kong waters significantly increased from 1997 to 2007, with an estimated annual rate of 5% (Lam et al., 2009). The authors suggested that HBCD levels in these dolphins will double from 1997 to 2017, assuming the rate of usage of HBCDs remains constant. This trend indicates that HBCDs may be used as alternatives for PBDEs in China and their levels in environment should be increasing due to the rapid economic development and industrial activities, as well as no control on production or use of these chemicals (Lam et al., 2009). Another study also found mean HBCDs levels in finless porpoises collected from Hong Kong waters increased (two fold) from 1990 to 2000 and 2001, although the difference was not statistically significant (Isobe et al., 2007). In contrast, no significant trend of HBCDs was found in finless porpoises sampled from waters from 2000 to 2008, possibly due to this species inhabited the eastern waters of Hong Kong, location being more oceanic-influenced and lightly contaminated by HBCDs compared to the habitat of humpback dolphins (Lam et al., 2009).

3.2 Spatial trends

It is difficult to compare data from different studies and conclude the spatial trends of BFRs in wildlife from Chinese environment because of the limited data available, the difference in species sampled and tissues examined, the number of PBDE congeners targeted, and the different expressions of the BFRs levels (e.g., lipid wt, wet wt, or dry wt). However, from the limited data available, two issues are evident (Fig. 2):

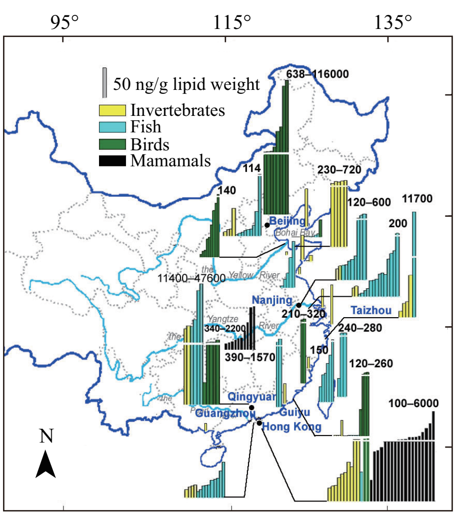

Fig. 2 Geographic distribution of PBDEs in wildlife from China.

(1) Elevated levels of BFRs were observed in wildlife from large urban centers such as Beijing, Guangzhou, Hong Kong, and Shanghai.

(2) High levels of PBDEs and HBCDs were generally found in wildlife from locations in the vicinity of HFRs production facilities (e.g., Jiangsu Province and Shandong Province, East China) and e-waste recycling sites (Taizhou City, Zhejiang Province, East China, and Shantou City and Qingyuan City, both located in Guangdong Province, South China). Concentrations of PBDEs and HBCDs in wildlife from these locations were often elevated by at least 1 order of magnitude compared to those from other parts in China.

4 Bioaccumulation characteristics

4.1 PBDE congener profiles

Consistent with the PBDE profiles observed in other studies around the world, aquatic organisms and terrestrial species from Chinese environment generally accumulated more fractions of lower brominated and higher brominated congeners, respectively; while amphibians (frogs) seem to show a intermediate PBDEs profile between those of aquatic and terrestrial species (Wu et al., 2009). For example, BDE 47 was the dominant congener, followed by BDE 99 in aquatic species (Guo et al., 2008; Hu et al., 2010; Luo et al., 2007a, 2007b; Qin et al., 2009; Wan et al., 2008; Wang et al., 2007b; Wu et al., 2008; Xiang et al., 2007; Yu et al., 2009; Zhang et al., 2010b), and the terrestrial species often preferentially accumulated BDE 153 over BDEs 99 and 47 (Chen et al., 2007a; Gao et al., 2009), whereas BDEs 99 and 153 were the largest contributors in frogs (Wu et al., 2009).

However, a different PBDE pattern in aquatic species from China was found, with relatively higher contributions of BDEs 15 and 28 (Gao et al., 2009; Meng et al., 2008; Xian et al., 2008). The elevated fractions of BDEs 15 and 28 was probably due to the usage of additional specific PBDE technical mixtures in China besides the well known commercial products such as the Penta-, Octa-, and Deca-BDEs mixtures, and/or the debromination of higher PBDEs in biota and in the environment (Gao et al., 2009; Meng et al., 2008; Xian et al., 2008). The relative abundance of these lower brominated congeners compared to the results in other reports was also found in environmental samples collected from China (Chen et al., 2006; Mai et al., 2005; Shen et al., 2006).

Another significant finding on the PBDE patterns was that bird (or bird eggs) from China contained remarkably elevated burdens of the higher brominated congeners such as nona- and deca-BDEs (BDE 209) (Lam et al., 2007; Chen et al., 2007a; Luo et al., 2009; Gao et al., 2009; Yu et al., 2011). In some bird species, BDE 209 was the dominant congener (Chen et al., 2007a; Luo et al., 2009; Gao et al., 2009; Yu et al., 2011). It can be anticipated that the contribution of BDE 209 may further increase in the near future, because large quantities of deca-BDE are still used in many products in China currently. BDE

209 may result in adverse effects to wildlife considering that this chemical is probably bioaccumulated in animals, and the fact that BDE 209 can be biotransformed to less brominated congeners which are more bioavailable and toxic (Law et al., 2006b).

4.2 Diastereoisomeric- and enantiomeric profiles of HBCDs

Only a few stereoisomer-specific studies have been performed on HBCDs in fish and marine mammals from China (Xian et al., 2008; Isobe et al., 2007; Lam et al., 2009; Zhang et al., 2009; Wu et al., 2010; He et al., 2010). The α-HBCD has been shown to be the dominant stereoisomer both in fish (55%–88%) and marine mammals (> 99%), which is consistent with other reports on HBCD diastereoisomeric patterns in fish (Covaci et al., 2006). In addition, Wu et al. (2010) observed a significant increase of fractions of α-HBCD, and a concurrently decrease of the fraction of γ-HBCD on ascending trophic levels of the sampled aquatic species, suggesting the diastereoisomeric patterns in aquatic species may associate with their trophic positions in the food web. The result was confirmed by the fraction of α-HBCD (> 99%) in marine mammals from the South China Sea, who occupied the highest trophic position in the marine food web (Isobe et al., 2007). Furthermore, the HBCD patterns were different in different tissues of the same individuals, e.g., fraction of the α-HBCD was relatively higher in liver (> 80%) than those in the muscle (ca. 60%) and eggs (ca. 40%) (Xian et al., 2008).

Wu et al. (2010) and Zhang et al. (2009) reported the selective accumulation of different HBCD enantiomers in aquatic species from China. The relative accumulation of (+)-α-HBCD was found in mud carp (Wu et al., 2010), crucian carp (Wu et al., 2010; Zhang et al., 2009), and loach (Zhang et al., 2009a). However, a reverse result was observed in winkle, with preferences of (-)-α-HBCD (Zhang et al., 2009). For β-HBCD, mud carp (Wu et al., 2010) and crucian carp (Wu et al., 2010; Zhang et al., 2009) selectively accumulate (-)-enantiomer. In the case of γ- HBCD, an enrichment of (+)-enantiomer was generally found in northern snakehead (Wu et al., 2010), crucian carp (Wu et al., 2010; Zhang et al., 2009), and loach (Zhang et al., 2009a). The results suggested the accumulation characteristics of HBCD enantiomers may be species-specific, and may be a complex combination of environmental and biological processes, such as aerobic and anaerobic microbial transformation in the water environment and biotransformation processes in the food web (Wu et al., 2010).

5 Summary and research directions

Presently, the data on BFRs levels in wildlife from China remains limited, especially for the non-PBDE BFRs, and these studies only limited to some locations such as e-waste recycling sites and the PRE. The limited data indicated that PBDE levels in wildlife from China were generally higher than those from other parts in Asia, and were comparable to those from Europe but were lower than those from North America. However, wildlife from the e-waste recycling sites in South China and East China contained much higher PBDEs compared to other reports around the world, suggesting the heavy contamination of PBDEs in the e-waste recycling sites in China. PBDE levels in marine mammals from South China seemed increased from the beginning of 1990s to 2000s, but decreased from the middle of 2000s, followed by relatively steady levels. Different from PBDEs, HBCDs were continuously increasing from 1997 to 2007, indicating the increasing usage of HBCDs following the restrictions or bans of the production and use of PentaBDEs and OctaBDEs. Compared to other reports around the world, aquatic species and bird from China contain relatively higher contributions of BDE 28 and BDE 209, respectively, suggesting a different PBDE usage pattern may have occurred in China.

Contamination of BDE 209 was thought to be heavier in China compared to European and North American countries. Studies indicated that BDE 209 had higher bioaccumulation potential in terrestrial animals compared to aquatic organisms. However, most of the studies investigating PBDE contamination in China and in the world in general, as above-mentioned were conducted in the aquatic ecosystems. More effects are needed to determine the environmental occurrence and behavior of BDE 209 in terrestrial animals.

For the monitoring of BFR levels near urban centers indicative of population, it should be keeping monitoring at a reasonable scale and frequency to make sure these levels do not increase. BFR levels near e-waste recycling sites are of great concern, focus effort on these areas should be performed to look for impacts and to determine if regulations/control are resulting in lower environmental contamination of BFRs. Additionally, bioaccumulation characteristic, e.g., PBDE congener profiles and HBCD distereoisomer ratios should be also examined to see whether they resemble those observed in regions without e-waste recycling issues or not.

Amphibians and birds are used as a sentimental species to indicate the environmental contamination. Data on PBDEs and other BFRs in amphibians and birds from China is limited. Nevertheless, large difference in BFR levels among species and individuals were found, even in the same sampling site. Incorporation of sentinel species within these groups (along with fish which are easy to do) in monitoring efforts is recommended.

Most of the current studies focused on PBDEs, few studies investigated the alternative BFRs in the wildlife in China. Our preliminary studies indicated that wildlife from South China harbored very high levels of HBCDs, BTBPE, DBDPE, and other non-PBDE BFRs. The levels, bioaccumulation characteristics, and trophic transfer of these chemicals should be performed, to better understand the environmental fate and risks of these pollutants.

The data base on BFRs levels and trends in wildlife from China needs to be expanded geographically, as little data is available outside of South China and North China.

The contamination status of BFRs in wildlife from other parts in China was not very clear. In addition, more work should be done on time trends of BFRs, especially for the alternative BFRs, for better understanding of their environmental fate, and aiding modelers in determining fluxes.

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References

Alaee M, Arias P, Sjödin A, Bergman Å, 2003. An overview of commercially used brominated flame retardants, their applications, their use patterns in different countries/regions and possible modes of release. *Environment International*, 29(6): 683–689.

Burkhart J G, Ankley G, Bell H, Carpenter H, Fort D, Gardiner D et al., 2000. Strategies for assessing the implications of malformed frogs for environmental health. *Environmental Health Perspectives*, 108(1): 83–90.

Chen D, Hale R C, 2010. A global review of polybrominated diphenyl ether flame retardant contamination in birds. *Environment International*, 36(7): 800–811.

Chen D, Mai B X, Song J, Sun Q H, Luo Y, Luo X J et al., 2007a. Polybrominated diphenyl ethers in birds of prey from Northern China. *Environmental Science and Technology*, 41(6): 1828–1833.

Chen S J, Gao X J, Mai B X, Chen Z M, Luo X J, Sheng G Y et al., 2006. Polybrominated diphenyl ethers in surface sediments of the Yangtze River Delta: Levels, distribution and potential hydrodynamic influence. *Environmental Pollution*, 144(3): 951–957.

Chen S J, Luo X J, Lin Z, Luo Y, Li K C, Peng X Z et al., 2007b. Time trends of polybrominated diphenyl ethers in sediment cores from the Pearl River Estuary, South China. *Environmental Science and Technology*, 41(16): 5595–5600.

Covaci A, Gerecke A C, Law R J, Voorspoels S, Kohler M, Heeb N V et al., 2006. Hexabromocyclododecanes (HBCDs) in the environment and humans: a review. *Environmental Science and Technology*, 40(12): 3679–3688.

Covaci A, Harrad S, Abdallah M A E, Ali N, Law R J, Herzke D et al., 2011. Novel brominated flame retardants: A review of their analysis, environmental fate and behaviour. *Environment International*, 37(2): 532–556.

De Solla S R, Fernie K J, Letcher R J, Chu S G, Drouillard K G, Shahmiri S, 2007. Snapping turtles (*Chelydra serpentina*) as bioindicators in Canadian areas of concern in the Great Lakes Basin. 1. Polybrominated diphenyl ethers, polychlorinated biphenyls, and organochlorine pesticides in eggs. *Environmental Science and Technology*, 41(21): 7252–7259.

de Wit C A, 2002. An overview of brominated flame retardants in the environment. *Chemosphere*, 46(5): 583–624.

de Wit C A, Alaee M, Muir D C G, 2006. Levels and trends of brominated flame retardants in the Arctic. *Chemosphere*, 64(2): 209–233.

de Wit C A, Herzke D, Vorkamp K, 2010. Brominated flame retardants in the Arctic environment – trends and new candidates. *Science of the Total Environment*, 408(15): 2885–2918.

Gao F, Luo X J, Yang Z F, Wang X M, Mai B X, 2009. Brominated flame retardants, polychlorinated biphenyls, and organochlorine pesticides in bird eggs from the Yellow River Delta, North China. *Environmental Science and Technology*, 43(18): 6956–6962.

Gauthier L T, Hebert C E, Weseloh D V C, Letcher R J, 2008. Dramatic changes in the temporal trends of polybrominated diphenyl ethers (PBDEs) in herring gull eggs from the Laurentian Great Lakes: 1982–2006. *Environmental Science and Technology*, 42(5): 1524–1530.

Gauthier L T, Potter D, Hebert C E, Letcher R J, 2009. Temporal trends and spatial distribution of non-polybrominated diphenyl ether flame retardants in the eggs of colonial populations of Great Lakes herring gulls. *Environmental Science and Technology*, 43(2): 312–317.

Guo L L, Qiu Y W, Zhang G, Zheng G J, Lam P K S, Li X D, 2008. Levels and bioaccumulation of organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) in fishes from the Pearl River Estuary and Daya Bay, South China. *Environmental Pollution*, 152(3): 604–611.

Hale R C, Alaee M, Manchester-Neesvig J B, Stapleton H M, Ikonomou M G, 2003. Polybrominated diphenyl ether flame retardants in the North American environment. *Environment International*, 29(6): 771–779.

Hale R C, La Guardia M J, Harvey E, Mainor T M, 2002. Potential role of fire retardant-treated polyurethane foam as a source of brominated diphenyl ethers to the US environment. *Chemosphere*, 46(5): 729–735.

He M J, Luo X J, Yu L H, Liu J, Zhang X L, Chen S J et al., 2010. Tetrabromobisphenol-A and hexabromocyclododecane in birds from an e-waste region in South China: influence of diet on diastereoisomer- and enantiomer-specific distribution and trophodynamics. *Environmental Science and Technology*, 44(15): 5748–5754.

Hu G C, Dai J Y, Xu Z C, Luo X J, Cao H, Wang J S et al., 2010. Bioaccumulation behavior of polybrominated diphenyl ethers (PBDEs) in the freshwater food chain of Baiyangdian Lake, North China. *Environment International*, 36(4): 309–315.

Ikonomou M G, Rayne S, Fischer M, Fernandez M P, Cretney W, 2002. Occurrence and congener profiles of polybrominated diphenyl ethers (PBDEs) in environmental samples from coastal British Columbia, Canada. *Chemosphere*, 46(5): 649–663.

Ismail N, Gewurtz S B, Pleskach K, Whittle D M, Helm P A, Marvin C H et al., 2009. Brominated and chlorinated flame retardants in Lake Ontario, Canada, lake trout (*Salvelinus namaycush*) between 1979 and 2004 and possible influences of food-web changes. *Environmental Toxicology and Chemistry*, 28(5): 910–920.

Isobe T, Ochi Y, Ramu K, Yamamoto T, Tajima Y, Yamada T K et al., 2009. Organohalogen contaminants in striped dolphins (*Stenella coeruleoalba*) from Japan: present contamination status, body distribution and temporal trends (1978–2003). *Marine Pollution Bulletin*, 58(3): 396–401.

Isobe T, Ramu K, Kajiwara N, Takahashi S, Lam P K S, Jefferson T A et al., 2007. Isomer specific determination of hexabromocyclododecanes (HBCDs) in small cetaceans from the South China Sea–Levels and temporal variation. *Marine Pollution Bulletin*, 54(8): 1139–1145.

Jaspers V L B, Covaci A, Voorspoels S, Dauwe T, Eens M, Schepens P, 2006. Brominated flame retardants and organochlorine pollutants in aquatic and terrestrial predatory birds of Belgium: levels, patterns, tissue distribution and condition factors. *Environmental Pollution*, 139(2): 340–352.

Jiang Y Q, 2006. Current situation and development of bromine retardant worldwide. *Chemical Techno-Economics*, 24(9): 14–19.

Jin J, Liu W Z, Wang Y, Tang X Y, 2008. Levels and distribution of polybrominated diphenyl ethers in plant, shellfish and sediment samples from Laizhou Bay in China. *Chemosphere*, 71(6): 1043–1050.

Johnson-Restrepo B, Adams D H, Kannan K, 2008. Tetrabromobisphenol A (TBBPA) and hexabromocyclododecanes (HBCDs) in tissues of humans, dolphins, and sharks from the United States. *Chemosphere*, 70(11): 1935–1944.

Johnson-Restrepo B, Kannan K, Addink R, Adams D H, 2005. Polybrominated diphenyl ethers and polychlorinated biphenyls in a marine foodweb of coastal Florida. *Environmental Science and Technology*, 39(21): 8243–8250.

Kajiwara N, Kamikawa S, Ramu K, Ueno D, Yamada T K, Subramanian A et al., 2006. Geographical distribution of polybrominated diphenyl ethers (PBDEs) and organochlorines in small cetaceans from Asian waters. *Chemosphere*, 64(2): 287–295.

Kajiwara N, Ueno D, Takahashi A, Baba N, Tanabe S, 2004. Polybrominated diphenyl ethers and organochlorines in archived northern fur seal samples from the Pacific coast of Japan, 1972–1998. *Environmental Science and Technology*, 38(14): 3804–3809.

Karlsson M, Ericson I, van Bavel B, Jensen J K, Dam M, 2006. Levels of brominated flame retardants in Northern Fulmar (*Fulmarus glacialis*) eggs from the Faroe Islands. *Science of the Total Environment*, 367(2-3): 840–846.

Kunisue T, Higaki Y, Isobe T, Takahashi S, Subramanian A, Tanabe S, 2008. Spatial trends of polybrominated diphenyl ethers in avian species: utilization of stored samples in the Environmental Specimen Bank of Ehime University (es-Bank). *Environmental Pollution*, 154(2): 272–282.

Lam J C, Kajiwara N, Ramu K, Tanabe S, Lam P K S, 2007. Assessment of polybrominated diphenyl ethers in eggs of waterbirds from South China. *Environmental Pollution*, 148(1): 258–267.

Lam J C W, Lau R K F, Murphy M B, Lam P K S, 2009. Temporal trends of hexabromocyclododecanes (HBCDs) and polybrominated diphenyl ethers (PBDEs) and detection of two novel flame retardants in marine mammals from Hong Kong, South China. *Environmental Science and Technology*, 43(18): 6944–6949.

Law K, Halldorson T, Danell R, Stern G, Gewurtz S, Alaee M et al., 2006a. Bioaccumulation and trophic transfer of some brominated flame retardants in a Lake Winnipeg (Canada) food web. *Environmental Toxicology and Chemistry*, 25(8): 2177–2186.

Law R J, Alaee M, Allchin C R, Boon J P, Lebeuf M, Lepom P et al., 2003. Levels and trends of polybrominated diphenylethers and other brominated flame retardants in wildlife. *Environment International*, 29(6): 757–770.

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(5): 673–681.

Law R J, Allchin C R, de Boer J, Covaci A, Herzke D, Lepom P et al., 2006b. Levels and trends of brominated flame retardants in the European environment. *Chemosphere*, 64(2): 187–208.

Law R J, Bersuder P, Allchin C R, Barry J, 2006c. Levels of the flame retardants hexabromocyclododecane and tetrabromobisphenol A in the blubber of harbor porpoises (*Phocoena phocoena*) stranded or bycaught in the U.K., with evidence for an increase in HBCD concentrations in recent years. *Environmental Science and Technology*, 40(7): 2177–2183.

Law R J, Herzke D, Harrad S, Morris S, Bersuder P, Allchin C R, 2008. Levels and trends of HBCD and BDEs in the European and Asian environments, with some information for other BFRs. *Chemosphere*, 73(2): 223–241.

Li Q Z, Yan C Z, Luo Z X, Zhang X, 2010. Occurrence and levels of polybrominated diphenyl ethers (PBDEs) in recent sediments and marine organisms from Xiamen offshore areas, China. *Marine Pollution Bulletin*, 60(3): 464–469.

Liu J, Luo X J, Yu L H, He M J, Chen S J, Mai B X, 2010. Polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyles (PCBs), hydroxylated and methoxylated-PBDEs, and methylsulfonyl-PCBs in bird serum from South China. *Archives of Environmental Contamination and Toxicology,* 59(3): 492–501.

Lundstedt-Enkel K, Johansson A K, Tysklind M, Asplund L, Nylund K, Olsson M et al., 2005. Multivariate data analyses of chlorinated and brominated contaminants and biological characteristics in adult guillemot (*Uria aalge*) from the Baltic Sea. *Environmental Science and Technology*, 39(22): 8630–8637.

Luo Q, Cai Z W, Wong M H, 2007a. Polybrominated diphenyl ethers in fish and sediment from river polluted by electronic waste. *Science of the Total Environment*, 383(1-3): 115–127.

Luo Q, Wong M H, Cai Z W, 2007b. Determination of polybrominated diphenyl ethers in freshwater fishes from a river polluted by e-wastes. *Talanta*, 72(5): 1644–1649.

Luo X J, Zhang X L, Liu J, Wu J P, Luo Y, Chen S J et al., 2009. Persistent halogenated compounds in waterbirds from an e-waste recycling region in South China. *Environmental Science and Technology*, 43(2): 306–311.

Mai B X, Chen S J, Luo X J, Chen L G, Yang Q S, Sheng G Y et al., 2005. Distribution of polybrominated diphenyl ethers in sediments of the Pearl River Delta and adjacent South China Sea. *Environmental Science and Technology*, 39(10): 3521–3527.

Martin M, Lam P K S, Richardson B J, 2004. An Asian quandary: where have all of the PBDEs gone? *Marine Pollution Bulletin*, 49(5-6): 375–382.

McKinney M A, Cesh L S, Elliott J E, Williams T D, Garcelon D K, Letcher R J, 2006. Brominated flame retardants and halogenated phenolic compounds in North American west coast bald eaglet (*Haliaeetus leucocephalus*) plasma. *Environmental Science and Technology*, 40(20): 6275–6281.

Meng X Z, Yu L P, Guo Y, Mai B X, Zeng E Y, 2008. Congener-specific distribution of polybrominated diphenyl ethers in fish of China: implication for input sources. *Environmental Toxicology and Chemistry*, 27(1): 67–72.

Norstrom R J, Simon M, Moisey J, Wakeford B, Weseloh D V C, 2002. Geographical distribution (2000) and temporal trends

(1981–2000) of brominated diphenyl ethers in Great Lakes herring gull eggs. *Environmental Science and Technology*, 36(22): 4783–4789.

Pan J, Yang Y L, Xu Q, Chen D Z, Xi D L, 2007. PCBs, PCNs and PBDEs in sediments and mussels from Qingdao coastal sea in the frame of current circulations and influence of sewage sludge. *Chemosphere*, 66(10): 1971–1982.

Peng J H, Huang C W, Weng Y M, Yak H K, 2007. Determination of polybrominated diphenyl ethers (PBDEs) in fish samples from rivers and estuaries in Taiwan. *Chemosphere*, 66(10): 1990–1997.

Qin X F, Xia X J, Li Y, Zhao Y X, Yang Z Z, Fu S et al., 2009. Ecotoxicological effects of mixed pollutants resulted from e-wastes recycling and bioaccumulation of polybrominated diphenyl ethers in Chinese loach (*Misgurnus anguillicaudatus*). *Journal of Environmental Sciences*, 21(12): 1695–1701.

Ramu K, Kajiwara N, Lam P K S, Jefferson T A, Zhou K Y, Tanabe S, 2006. Temporal variation and biomagnification of organohalogen compounds in finless porpoises (*Neophocaena phocaenoides*) from the South China Sea. *Environmental Pollution*, 144(2): 516–523.

Ramu K, Kajiwara N, Sudaryanto A, Isobe T, Takahashi S, Subramanian A et al., 2007. Asian Mussel Watch Program: contamination status of polybrominated diphenyl ethers and organochlorines in coastal waters of Asian countries. *Environmental Science and Technology*, 41(13): 4580–4586.

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. *Marine Pollution Bulletin*, 51(8-12): 669–676.

Shaw S D, Kannan K, 2009. Polybrominated diphenyl ethers in marine ecosystems of the American continents: Foresight from current knowledge. *Reviews on Environmental Health*, 24(3): 157–230.

She J W, Petreas M, Winkler J, Visita 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(5): 697–707.

Shen M, Yu Y J, Zheng G J, Yu H X, Lam P K S, Feng J F et al., 2006. Polychlorinated biphenyls and polybrominated diphenyl ethers in surface sediments from the Yangtze River Delta. *Marine Pollution Bulletin*, 52(10): 1299–1304.

Shi T, Chen S J, Luo X J, Zhang X L, Tang C M, Luo Y et al., 2009. Occurrence of brominated flame retardants other than polybrominated diphenyl ethers in environmental and biota samples from southern China. *Chemosphere*, 74(7): 910–916.

Stapleton H M, Dodder N G, Kucklick J R, Reddy C M, Schantz M M, Becker P R et al., 2006. Determination of HBCD, PBDEs and MeO-BDEs in California sea lions (*Zalophus californianus*) stranded between 1993 and 2003. *Marine Pollution Bulletin*, 52(5): 522–531.

Ter Schure A F H, Larsson P, Merilä J, Jonsson K I, 2002. Latitudinal fractionation of polybrominated diphenyl ethers and polychlorinated biphenyls in frogs (*Rana temporaria*). *Environmental Science and Technology*, 36(23): 5057–5061.

Tian S Y, Zhu L Y, Liu M, 2010. Bioaccumulation and distribution of polybrominated diphenyl ethers in marine species from Bohai Bay, China. *Environmental Toxicology and Chemistry*, 29(10): 2278–2285.

Ueno D, Kajiwara N, Tanaka H, Subramanian A, Fillmann G, Lam P K S et al., 2004. Global pollution monitoring of polybrominated diphenyl ethers using skipjack tuna as a bioindicator. *Environmental Science and Technology*, 38(8): 2312–2316.

UNEP, 2010. The 9 new POPs [EB/OL]. Available at: http://www.dioxinnz.com/pdf-X-stockholm/UNEP-POPS-NPOPS-OVERV.En.pdf. [accessed 13 June 2011].

USEPA, 2010. DecaBDE Phase-out Initiative [EB/OL]. Available at: http://www.epa.gov/oppt/existingchemicals/pubs/actionplans/deccadbe.html. [accessed 13 June 2011].

Verreault J, Gabrielsen G W, Chu S G, Muir D C G, Andersen M, Hamaed A et al., 2005. Flame retardants and methoxylated and hydroxylated polybrominated diphenyl ethers in two Norwegian Arctic top predators: glaucous gulls and polar bears. *Environmental Science and Technology*, 39(16): 6021–6028.

Wan Y, Hu J Y, Zhang K, An L H, 2008. Trophodynamics of polybrominated diphenyl ethers in the marine food web of Bohai Bay, North China. *Environmental Science and Technology*, 42(4): 1078–1083.

Wang Y W, Jiang G B, Lam P K S, Li A, 2007a. Polybrominated diphenyl ether in the East Asian environment: a critical review. *Environment International*, 33(7): 963–973.

Wang Y W, Li X M, Li A, Wang T, Zhang Q H, Wang P et al., 2007b. Effect of municipal sewage treatment plant effluent on bioaccumulation of polychlorinated biphenyls and polybrominated diphenyl ethers in the recipient water. *Environmental Science and Technology*, 41(17): 6026–6032.

Wolkers H, van Bavel B, Derocher A E, Wiig O, Kovacs K M, Lydersen C et al., 2004. Congener-specific accumulation and food chain transfer of polybrominated diphenyl ethers in two arctic food chains. *Environmental Science and Technology*, 38(6): 1667–1674.

Wong M H, Wu S C, Deng W J, Yu X Z, Luo Q, Leung A O W et al., 2007. Export of toxic chemicals – a review of the case of uncontrolled electronic-waste recycling. *Environmental Pollution*, 149(2): 131–140.

Wu J P, Guan Y T, Zhang Y, Luo X J, Zhi H, Chen S J et al., 2011. Several current-use, non-PBDE brominated flame retardants are highly bioaccumulative: evidence from field determined bioaccumulation factors. E*nvironment International*, 37(1): 210–215.

Wu J P, Guan Y T, Zhang Y, Luo X J, Zhi H, Chen S J et al., 2010. Trophodynamics of hexabromocyclododecanes and several other non-PBDE brominated flame retardants in a freshwater food web. *Environmental Science and Technology*, 44(14): 5490–5495.

Wu J P, Luo X J, Zhang Y, Chen S J, Mai B X, Guan Y T et al., 2009. Residues of polybrominated diphenyl ethers in frogs (*Rana limnocharis*) from a contaminated site, South China: tissue distribution, biomagnification, and maternal transfer. *Environmental Science and Technology*, 43(14): 5212–5217.

Wu J P, Luo X J, Zhang Y, Luo Y, Chen S J, Mai B X et al., 2008. Bioaccumulation of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in wild aquatic species from an electronic waste (e-waste) recycling site in South China. *Environment International*, 34(8): 1109–1113.

Xia C, Lam J C W, Wu X, Sun L, Xie Z, Lam P K S, 2011. Hexabromocyclododecanes (HBCDs) in marine fishes along the Chinese coastline. *Chemosphere*, 82: 1662–

1668.

Xian Q M, Ramu K, Isobe T, Sudaryanto A, Liu X H, Gao Z S et al., 2008. Levels and body distribution of polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) in freshwater fishes from the Yangtze River, China. *Chemosphere*, 71(2): 268–276.

Xiang C H, Luo X J, Chen S J, Yu M, Mai B X, Zeng E Y, 2007. Polybrominated diphenyl ethers in biota and sediments of the Pearl River Estuary, South China. *Environmental Toxicology and Chemistry*, 26(4): 616–623.

Xu J, Gao Z S, Xian Q M, Yu H X, Feng J F, 2009. Levels and distribution of polybrominated diphenyl ethers (PBDEs) in the freshwater environment surrounding a PBDE manufacturing plant in China. *Environmental Pollution*, 157(6): 1911–1916.

Yang F X, Zhang Q H, Xu Y, Jiang G B, Wang Y W, Wang D, 2008. Preliminary hazard assessment of polychlorinated biphenyls, polybrominated diphenyl ethers, and polychlorinated dibenzo-*p*-dioxins and dibenzofurans to Yangtze finless porpoise in Dongting Lake, China. *Environmental Toxicology and Chemistry*, 27(4): 991–996.

Yu L H, Luo X J, Wu J P, Liu L Y, Song J, Sun Q H et al., 2011. Biomagnification of higher brominated PBDE congeners in an urban terrestrial food web in North China based on field observation of prey deliveries. *Environmental Science and Technology*, 45(12): 5125–5131.

Yu M, Luo X J, Wu J P, Chen S J, Mai B X, 2009. Bioaccumulation and trophic transfer of polybrominated diphenyl ethers (PBDEs) in biota from the Pearl River Estuary, South China. *Environment International*, 35(7): 1090–1095.

Zegers B N, Mets A, Van Bommel R, Minkenberg C, Hamers T, Kamstra J H et al., 2005. Levels of hexabromocyclododecane in harbor porpoises and common dolphins from western European seas, with evidence for stereoisomer-specific biotransformation by cytochrome P450. *Environmental Science and Technology*, 39(7): 2095–2100.

Zhang K, Wan Y, An L H, Hu J Y, 2010a. Trophodynamics of polybrominated diphenyl ethers and methoxylated polybrominated diphenyl ethers in a marine food web. *Environmental Toxicology and Chemistry*, 29(12): 2792–2799.

Zhang X L, Yang F X, Luo C H, Wen S, Zhang X, Xu Y, 2009. Bioaccumulative characteristics of hexabromocyclododecanes in freshwater species from an electronic waste recycling area in China. *Chemosphere*, 76(11): 1572–1578.

Zhang X L, Luo X J, Liu H Y, Yu L H, Chen S J, Mai B X, 2011. Bioaccumulation of several brominated flame retardants and dechlorane plus in waterbirds from an e-waste recycling region in South China: associated with trophic level and diet sources. *Environmental Science and Technology*, 45(2): 400–405.

Zhang Y, Luo X J, Wu J P, Liu J, Wang J, Chen S J et al., 2010b. Contaminant pattern and bioaccumulation of legacy and emerging organhalogen pollutants in the aquatic biota from an e-waste recycling region in South China. *Environmental Toxicology and Chemistry*, 29(4): 852–859.