Trophodynamics of Polybrominated Diphenyl Ethers in the Marine Food Web of Bohai Bay, North China

YI WAN, JIANYING HU,* KUN ZHANG, AND LIHUI AN

College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

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Polybrominated diphenyl ethers (PBDEs) are of great environmental concern due to the exponential increase of the concentrations in the environment, especially in high trophic level organisms, and the trophodynamics of these chemicals in aquatic food webs is an important criterion for assessing their ecological risk. This study analyzed 13 PBDEs in the zooplankton, five invertebrate species, six fish species, and one marine bird species collected from Bohai Bay. PBDE concentrations in organisms from Bohai Bay (ΣPBDEs: 0.15–32.8 ng/g lipid weight) were low compared with other marine organisms worldwide, and BDE-47 was the predominant compound in most samples, followed by BDE-28, BDE-99/BDE-100, and BDE-119. Correlation between lipid-normalized concentrations of PBDEs, and trophic levels determined by stable nitrogen isotope technologies confirmed that PBDEs were biomagnified in the marine food web. Significantly positive relationships were found for total PBDEs and four PBDE compounds (BDE-28, BDE-47, BDE-100, and BDE-119), and their trophic magnification factors (TMFs) were 3.53, 3.57, 7.24, 3.23, and 2.60, respectively. The concentration ratios between congeners (BDE-99/BDE-100 and BDE-99/BDE-47) were found to decrease with increasing trophic levels, suggesting that trophic-level-dependent concentrations ratios between BDE-99 and BDE-100 would be contributed by trophic level-dependent biotransformation between BDE-99 and BDE-47, and therefore resulting in the dominance of BDE-100 compared with BDE-99 and the relatively high trophic magnification of BDE-47 in the marine food web.

Introduction

A number of papers have highlighted the increasing global pollution of polybrominated diphenyl ethers (PBDEs), the most important class of flame retardants. Available toxicological evidence shows that PBDEs can disturb thyroid homeostasis, cause hepatomegaly and neurobehavioral deficits, and exhibit fetal and maternal toxicity after prolonged exposure in spite of their low acute toxicity (*1*, *2*). PBDEs have been shown to be ubiquitous in the environment as evidenced by their occurrence in various environmental medias (*3–9*), and their concentrations in most environmental compartments are exponentially increasing with doubling times of about 4–6 years (*5*). Global distribution studies suggest that PBDEs undergo long-range transport through the atmosphere, therefore causing global pollution (*10–12*).

* Corresponding author phone: 86-10-62765520; fax: 86-10-62765520; e-mail: hujy@urban.pku.edu.cn.

In addition to their toxicity, ubiquity, and long-range transport, many studies have been conducted on the analysis of biota samples to assess the bioaccumulation potential of these pollutants. Because of their properties of high lipophilicity (log K_{ow}: 5.9–10) (*3*, *13*) and resistance to metabolism (*14*), PBDEs are bioaccumulative in aquatic biota, which was supported by the residues in fish and invertebrates from both freshwater and marine environments (*4*, *15–18*). Besides the ubiquitous occurrence of PBDEs in fish and invertebrates, PBDEs have also been detected in high trophic marine organisms such as seabirds, whales, and seals (*19–22*). Compared with the studies of bioaccumulation for PBDEs, there is limited work about the trophic magnification of PBDEs in aquatic food webs, a vital criterion for assessing the ecological risk of chemicals. Qualitative studies have been conducted by comparing PBDE concentrations in organisms at different trophic levels (*24–27*), and it was found that the concentrations of PBDEs decreased in the following order: mammals, birds > fish > invertebrates, and PBDEs might have comparable trophic magnification potential with PCBs. As for quantitative studies, little comprehensive data is available about the trophodynamics of PBDEs in aquatic food webs (*28–30*). Burreau et al. analyzed PBDEs in the food web from the Baltic Sea (including zooplankton and three fish species) and the northern Atlantic Ocean (including two fish species) by using the nitrogen isotopes as trophic levels directly, showing that tri- to hepa-BDEs and PCBs all have similar biomagnification potentials in the short food web (*28*, *29*). Trophic magnification factors (TMFs) of PBDEs have only been reported for a freshwater food web from Lake Winnipeg (including zooplankton, one invertebrate, and six fish species) (*30*), and only BDE-209 and BDE-47 were reported to have significant TMF values. The TMF of BDE-47 (1.5) was found to be lower than that (3.6) of BDE-209, whereas BDE-209 was reported to be bioeliminated more rapidly than BDE-47 (*31*, *32*). These two works both excluded organisms at high trophic levels such as birds or mammals which are often included in the trophodynamic studies of chemicals (*33–35*).

Bohai Bay is an enclosed inner sea in North China, and its food web model has been established and successfully applied to analyze the characterization of trophic transfer for various pollutants such as dioxins, nonylphenol and nonylphenol ethoxylates, organotins, organochlorines, and PAHs (*36–39*). Therefore, the trophodynamics of new chemicals further studied in this ecosystem can be compared with those of the above pollutants. In this study, we analyzed 13 PBDEs in the same food web (including zooplankton, five invertebrate species, six fish species, and one marine bird species) to determine their trophodynamics in the food web. The relationships between concentration ratios between congeners (BDE-99/BDE-100 and BDE-99/BDE-47) and trophic levels were analyzed and compared to explore the reason for profile variations and the main factor influencing the biomagnification of PBDEs.

Materials and Methods

Sample Collection. The samples analyzed in this study were the same as those in our previous papers (*36–39*). The part of the marine food web included primary producers (zooplankton), five invertebrate species (crab (*Portunus trituberculatus*), short-necked clam (*Ruditapes philippinarum*), burrowing shrimp (*Upogebia* sp.), veined rapa whelk (*Rapana venosa*), and bay scallop (*Argopecten irradians*)), six fish species ((weever (*Lateolabras japonicus*), catfish (*Chaeturichthys sitgmatias*), bartail flathead (*Platycephalus indlcus*),

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wolffish (*Obontamblyopus rubicundus*), white flower croaker (*Nibea albiflora*) and mullet (*Liza so-iuy*)), and one seabird species (herring gull, (*Larus argentatus*)). Detailed information about sample collection is provided in the Supporting Information, and the whole bodies of zooplankton, soft tissues of invertebrates, and muscles of fish and seabird were used for PBDEs and isotope analysis.

Analysis of PBDEs. PBDE congeners were analyzed following methods that have been described and validated in previous investigations (*23, 40–42*). The whole bodies of zooplankton, the soft tissues of crabs, burrowing shrimps, short-necked clams, veined rapa whelks, and bay scallops and the muscles of fish and seabird were first freeze-dried. Then, about 1–10 g samples were spiked with surrogate standards (PCB-198 and PCB-209), and Soxhlet-extracted with a mixture of dichloromethane and hexane (3:1) for 24 h. Half of the extracts were stored, and the rest were concentrated and passed through an acidified silica packed glass column (1 g Na_2SO_4, 8 g acidified silica, 10 mm i.d.). To prepare the acidified silica, 50 g silica gel was heated at 120 °C overnight, and then mixed with 27 mL concentrated sulfuric acid. This column was eluted with 15 mL hexane and 10 mL dichloromethane. After the addition of PCB-204, the eluant was concentrated to 0.2 mL in hexane, and analyzed by gas chromatography-electron capture negative ionization mass spectrometry (GC-ENCI-MS) (Shimadzu QP 2010 plus, Japan). The information of chemicals, GC-MS condition, quantitation and quality assurance quality control (QA/QC), lipid content analysis, calculations of trophic magnification factor, and statistical analysis were provided in the Supporting Information.

Results and Discussion

PBDE Levels. Of the 13 PBDE congeners measured, nine compounds (BDE-28, BDE-71, BDE-47, BDE-66, BDE-100, BDE-119, BDE-99, BDE-154, and BDE-153) were detected in zooplankton, invertebrates, and fish from Bohai Bay (Table 1), and two high brominated congeners (BDE-138 and BDE-183) in addition to the above, nine compounds appeared in birds. The summed concentrations of PBDE congeners ranged from 10.45 (bay scallop) to 3320 pg/g ww (herring gull). When the concentrations were expressed on a lipid weight basis, the highest average concentrations were still detected in herring gull (32.78 ± 5.09 ng/g lw (lipid weight)), followed by fish (0.56–6.31 ng/g lw), invertebrates (0.15–1.09 ng/g lw), and zooplankton (1.00 ng/g lw). To compare the PBDE levels with other studies, total PBDE concentrations and concentrations of individual congener (BDE-47) calculated to lipid-based results were used. In this study, the summed PBDE concentration and the BDE-47 concentration in zooplankton were 1.00 ng/g lw and 0.03 ng/g lw, respectively, and the concentrations of BDE-47 are comparable to those of zooplankton in the Arctic food chain in Svalbard (BDE-47: 0.08 ng/g lw) (*25*), and lower than those reported in the Baltic Sea (BDE-47: 1.8–3.4 ng/g lw) (*29*). The PBDE concentrations in marine zooplankton were all much lower than those of freshwater zooplankton in Lake Winnipeg (ΣPBDEs: 36.42–90.42 ng/g lw; BDE-47: 6.38–15.94 ng/g lw) (*30*) and two Arctic lakes (BDE-47: <5–39.3 ng/g lw) (*43*). The PBDEs concentrations in invertebrates from Bohai Bay were lower than those from other locations (*16, 18, 23, 44*), and the concentrations in fish were close to those in fish from Svalbard and northern Atlantic Ocean (*25, 29*) (Table S1, Supporting Information). In birds (herring gulls), the summed PBDE concentrations and BDE-47 concentrations were 32.78 ± 5.09 and 16.13 ± 2.37 ng/g lw, respectively, which were much higher than those of other organisms in Bohai Bay, suggesting PBDEs would have higher biomagnification potential in homeotherms than in poikilotherms (*34*). But the PBDE concentrations in the present study were much

TABLE 1. Mean Biological Parameters and PBDEs Concentrations (pg/g wet weight) of 13 Marine Organisms Collected from Bohai Bay, North China[a]

species	ZK[b]	AI	RP	RV	US	PT	LS	OR	PI	CS	NA	LJ	LA
lipid content (%)	1.8	6.4	4.1	6.2	7.2	5.9	4.4	4.4	2.5	4.2	3.14	4.6	10.1
water content (%)	90	77	85	79	74	80	78	81	77	77	78	77	65
TL	2.00 ± 0.14	2.15 ± 0.12	2.17 ± 0.20	2.79 ± 0.12	3.16 ± 0.14	3.10 ± 0.17	3.01 ± 0.44	3.58 ± 0.11	3.65 ± 0.24	3.67 ± 0.04	3.65 ± 0.39	3.88 ± 0.49	3.84 ± 0.43
length (cm)							20(15.5–24.5)	27(20–30)	36(23.5–61)	20(17–24)	21(10.5–34)	35(22.5–48)	35(31–41)
weight (g)							93 (44–158)	36 (18–56)	357 (95–1119)	83 (56–120)	195 (10–538)	728 (180–1276)	211 (142–321)
number of individuals analyzed	-[b]	3	3	3	3	3	3	3	3	3	3	3	3
BDE-28	1.5	1.5 ± 1.4	0.7 ± 0.2	0.8 ± 0.8	12.2 ± 4.2	4.5 ± 2.6	3.7 ± 0.3	2.9 ± 0.9	6.1 ± 2.4	1.7 ± 1.2	15.8 ± 11.3	5.0 ± 1.4	196 ± 124
BDE-71	2.0	0.7 ± 0.6	0.6 ± 0.4	ND	4.8 ± 1.4	0.7 ± 0.5	1.8 ± 0.8	1.4 ± 0.6	4.2 ± 1.9	ND	9.1 ± 1.8	1.3 ± 0.6	54.2 ± 30.2
BDE-47	0.5	3.3 ± 2.7	5.1 ± 1.4	9.7 ± 3.0	29.3 ± 6.0	1.9 ± 0.7	16.7 ± 4.2	9.3 ± 2.9	55.1 ± 31.2	10.0 ± 4.4	119 ± 45.5	18.8 ± 1.4	1640 ± 240
BDE-66	4.0	1.5 ± 1.3	0.8 ± 0.2	1.8 ± 0.7	4.2 ± 0.6	0.8 ± 1.0	2.1 ± 1.0	1.9 ± 0.9	4.8 ± 1.9	1.3 ± 1.9	9.3 ± 7.9	2.4 ± 0.6	113 ± 60.2
BDE-100	2.7	0.8 ± 0.8	0.7 ± 0.2	2.3 ± 0.2	11.6 ± 3.1	0.8 ± 0.7	2.7 ± 0.9	2.1 ± 0.6	9.3 ± 5.9	2.9 ± 0.6	18.9 ± 2.7	3.7 ± 0.6	309 ± 136
BDE-119	2.8	0.8 ± 0.8	0.9 ± 0.4	2.2 ± 0.9	3.9 ± 0.2	0.9 ± 0.8	3.2 ± 1.9	2.1 ± 1.1	5.0 ± 1.4	2.8 ± 0.2	9.1 ± 2.6	3.5 ± 0.6	65.7 ± 15.2
BDE-99	4.8	1.0 ± 1.0	3.4 ± 0.9	3.4 ± 1.8	3.9 ± 0.6	0.8 ± 0.5	0.7 ± 0.6	2.7 ± 1.5	2.0 ± 1.8	2.1 ± 2.4	11.3 ± 3.7	0.8 ± 0.2	277 ± 156
BDE-154	0.4	ND	ND	ND	2.7 ± 0.1	ND	2.4 ± 0.8	0.8 ± 0.4	2.7 ± 1.0	2.1 ± 1.8	3.8 ± 0.5	1.6 ± 0.6	174 ± 53.7
BDE-153	0.5	ND	ND	ND	5.6 ± 0.4	ND	ND	2.3 ± 1.9	ND	ND	2.2 ± 0.2	0.7 ± 0.3	280 ± 184
BDE-138	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	45.8 ± 36.5
BDE-183	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	173 ± 249
sum (PBDEs)	18.5	10.5 ± 8.6	13.1 ± 3.5	21.3 ± 6.1	78.2 ± 11.5	11.3 ± 5.4	33.7 ± 9.1	25.4 ± 9.1	89.7 ± 46.5	23.6 ± 8.7	198 ± 71.4	37.8 ± 1.7	3320 ± 515

[a] ZK, zooplankton; AI, bay scallop (*Argopecten irradians*); RP, short-necked clam (*Ruditapes philippinarum*); RV, veined rapa whelk (*Rapana venosa*); US, burrowing shrimp (*Upogebia* sp.); PT, crab (*Portunus trituberculatus*); LS, mullet (*Liza so-iuy*); OR, wolffish (*Obontamblyopus rubicundus*); PI,bartail flathead (*Platycephalus indlcus*); CS, catfish (*Chaeturichthys sitgmatias*); NA, white flower croaker (*Nibea albiflora*); LJ, weever (*Lateolabras japonicus*); LA, herring gull (*Larus argentatus*). TL, trophic level; ND, not detected. [b] Pooled sample collected from six locations.

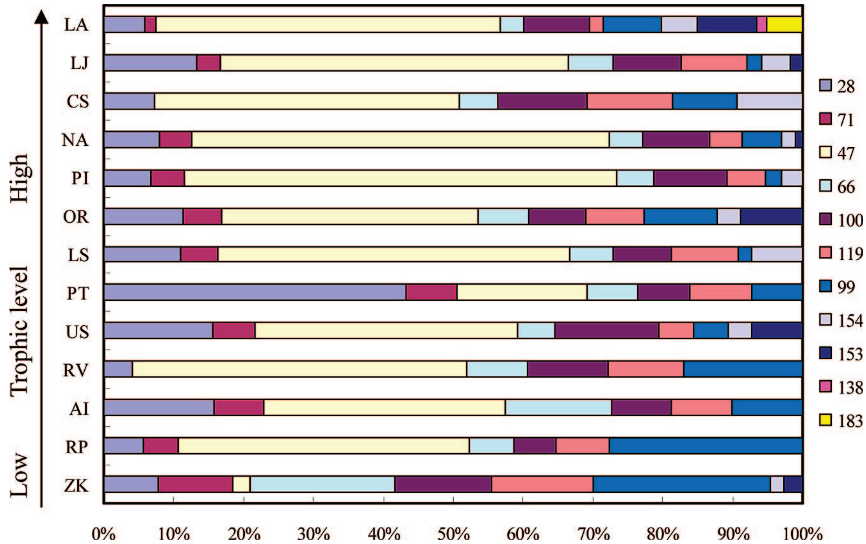

FIGURE 1. PBDE profiles in organisms collected from Bohai Bay, North China. ZK, zooplankton; AI, bay scallop (*Argopecten irradians*); RP, short-necked clam (*Ruditapes philippinarum*); RV, veined rapa whelk (*Rapana venosa*); US, burrowing shrimp (*Upogebia* sp.); PT, crab (*Portunus trituberculatus*); LS, mullet (*Liza so-iuy*); OR, wolffish (*Obontamblyopus rubicundus*); PI, bartail flathead (*Platycephalus indlcus*); CS, catfish (*Chaeturichthys sitgmatias*); NA, white flower croaker (*Nibea albiflora*); LJ, weever (*Lateolabras japonicus*); LA, herring gull (*Larus argentatus*).

lower compared with those in aquatic birds from Belgium and herring gulls from the Great Lakes (*46*). Overall, the PBDE concentrations in organisms from Bohai Bay in North China were low compared with other marine organisms worldwide, although global monitoring of PBDEs using skipjack tuna as a bioindicator reported relatively high concentrations of PBDEs in the East China Sea and South China Sea (*11*).

PBDE Profiles. Figure 1 shows the PBDE profiles in organisms from Bohai Bay. Of all the congeners, BDE-47 was the predominant compound in most samples, with a contribution of 41 ± 16.1% to the total concentrations. Excluding BDE-47, the general order of decreasing contribution to the total PBDE concentrations is BDE-28 > BDE-99 or BDE-100 > BDE-119, which accounts for 12.0 ± 10.1%, 10.2 ± 8.4%, 10.0 ± 2.6%, and 8.3 ± 3.4% of total PBDE concentrations, respectively. Profiles with predominant BDE-47 and high contribution of BDE-99 and BDE-100 have been reported in many marine organisms (*23–26*). It is interesting that relatively high proportions of BDE-28, especially in crab (43%), compared with those of BDE-99 and BDE-100, were only reported in organisms (shrimp, fish, and penguin) from the Ross Sea, Antarctica (*47*), indicating that organisms in Bohai Bay might be exposed to low brominated PBDEs. The most abundant congener in zooplankton was found to be BDE-99, accounting for 27% of the total PBDEs, which differed from the profiles of zooplankton in the Baltic Sea (*29*), svalbard (*25*), and in Lake Winnipeg (predominant congener: BDE-47) (*30*). In herring gulls, the PBDE profiles (BDE-47: 49 ± 4.6%, aquatic birds) were similar with those of aquatic birds reported previously (*45, 46, 48*), and different from those of terrestrial birds with equal proportions of BDE-47, BDE-153, and BDE-99 (*46, 48*), further proving that herring gulls in Bohai Bay would be exposed to lower brominated BDE congeners. As for high brominated congeners, BDE-138 (1.4%) and BDE-183 (5.2%) were only detected in birds, and high detection frequency and proportion of BDE-154 and BDE-153 were found in high trophic levels organisms: BDE-154 was detected in zooplankton (2.1%), burrowing shrimp (3.4%), and all fish and birds (1.9–9.3%), BDE-153 in zooplankton (2.6%), burrowing shrimp (6.2%), three fish and birds (1.1–9.0%), suggesting that these congeners might biomagnify in the marine food web.

Trophodynamics of PBDEs. Regression analysis was conducted between the lipid-normalized concentrations of PBDEs (log-transformed) and the trophic levels (Figure 2), and the statistical results of the regression analysis are listed in Table 2. Lipid-normalized concentrations of total PBDE concentrations increased significantly with increasing trophic level, with a TMF value of 3.53 ($p = 0.046$). As for individual PBDE congeners detected, TMFs ranged from 1.56 (BDE-99) to 7.24 (BDE-47), and significant positive relationships were obtained for BDE-28 (3.57, $p = 0.024$), BDE-47 (7.24, $p = 0.006$), BDE-100 (3.23, $p = 0.049$), and BDE-119 (2.60, $p = 0.042$), with relatively high concentration contributions, as shown in Figure 3and Table 2. The TMFs for PBDEs could be found in Law's study in a Lake Winnipeg food web (*30*). In that study, the TMFs of four individual congeners (BDE-47, BDE-99, BDE-100, and BDE-209) and total PBDEs were reported, ranging from 0.7 (BDE-99) to 3.6 (BDE-209), and significant relationships were only obtained for BDE-47 (1.5, $p = 0.01$) and BDE-209 (3.6, $p = 0.0001$) (*30*). The TMFs of the PBDEs in Bohai Bay were higher than those in Lake Winnipeg, which may be due to the different ecological factors such as the food web length. The TMFs based on the food webs including only zooplankton, invertebrates, and fish in the present study were calculated, and only BDE-47 shows significant value (4.85, $p = 0.012$), which were lower than those estimated based on the whole food web. This result was also demonstrated by several previous studies, in which TMFs of HCB, HCHs, p,p'-DDE, and PCBs estimated based on a food web including only poikilotherms (invertebrates, fish) are usually lower than those estimated based on a food web including both piokilotherms and homeotherms (seabirds and mammals) (*33, 34, 49*).

Previous investigations about the trophodynamics of dioxins, PAHs, and organochlorines in the same food web provide an opportunity to compare the TMFs of PBDEs with those of other chemicals. The TMF of total PBDEs was 3.53, which was comparable with that of total DDTs (TMF: 3.98) (*37*). It should be noted that the TMF of BDE-47 was 7.24, which was much higher than those of p,p'-DDE and DDMU (3.26–3.83) in the food web (*37*), and the possible reason would be due to the fact that BDE-47 was formed by biotransformation of high brominated PBDEs as exemplified by the common carp exposed to BDE-99 (*50*). A similar phenomenon was also reported for p,p'-DDE, which is a major metabolite of DDT in organisms and always shows high TMF values (*33, 34, 37*). It should be noted that the TMF

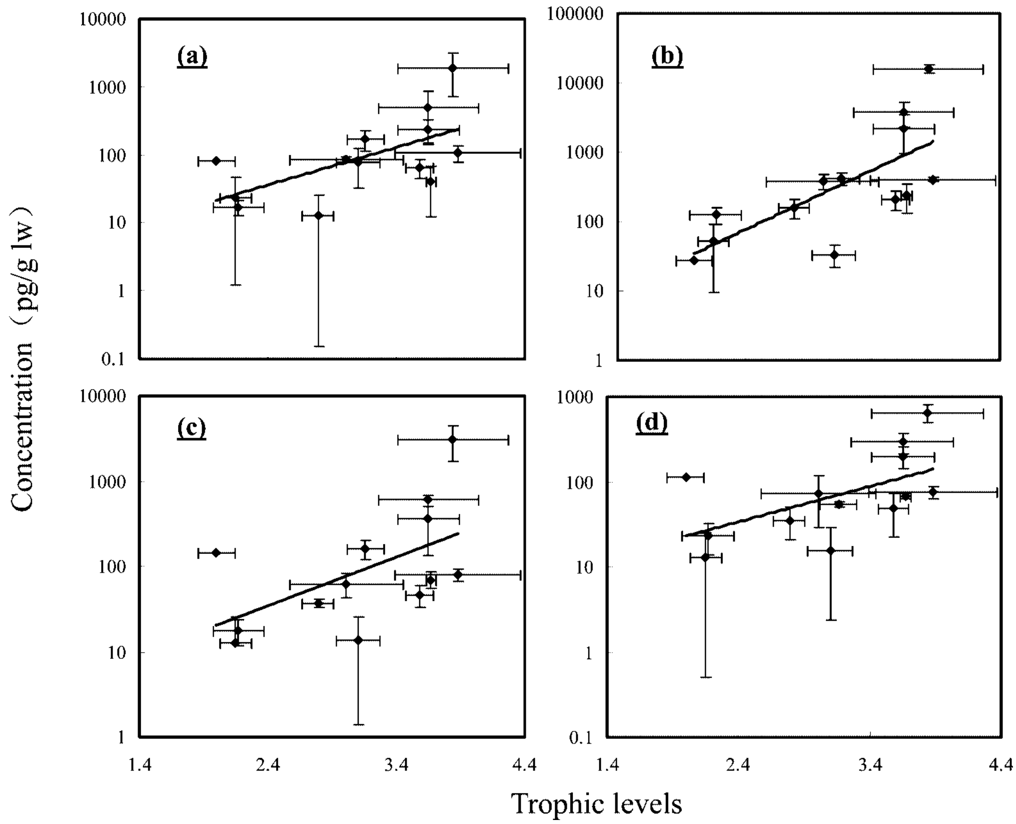

FIGURE 2. Relationship between concentrations of PBDE congeners (ng/g lw) and trophic levels of organisms in Bohai Bay, north China. (a) BDE-28, slope = 0.5531, r^2 = 0.3827, p = 0.024; (b) BDE-47, slope = 0.8598, r^2 = 0.5094, p = 0.006; (c) BDE-100, slope = 0.5695, r^2 = 0.308, p = 0.049; (d) BDE-119, slope=0.4158, r^2 = 0.3243, p = 0.042.

TABLE 2. Slope and p-Value of Slope of Regression Analysis between Logarithm of Concentration and Trophic Levels, and TMFs for PBDEs[a]

compound	slope	r^2	TMF	p
BDE-28	0.5531	0.3827	3.57	**0.024**
BDE-71	0.3295	0.1071	2.14	0.275
BDE-47	0.8598	0.5094	7.24	**0.006**
BDE-66	0.3042	0.1343	2.01	0.218
BDE-100	0.5695	0.308	3.23	**0.049**
BDE-119	0.4158	0.3243	2.60	**0.042**
BDE-99	0.1918	0.038	1.56	0.523
Sum (PBDEs)	0.5479	0.316	3.53	**0.046**

[a] p-values in bold print represent statistically significant increases or decreases of the lipid equivalent concentration (i. e., <0.05).

of BDE-47 in the present study was just in the range of values reported for PCB-47 in marine food webs (2.5–14.5) (*33, 34*). Overall, the TMFs of PBDEs (2.60–7.24, log K_{ow}: 5.94–7.32) (*13*) were slightly lower than those of coplanar PCBs (TMFs: 3.40–12.26) with the same range of log K_{ow} (6.65–7.71) (*36*), higher than those of HCB (TMFs: 2.96), and much higher than those of PAHs (TMFs: 0.11–0.45), nonylphenol and nonylphenol ethoxylates (TMFs: 0.45–1.15) in the food web (*36*), which could be related to their metabolism in organisms: the half-lives of PBDEs (TeBDE: 9.8–115 days; PeBDE: 27–32 days) (*51*) were found to be slightly lower than those of coplanar PCBs (28–107 days), but higher than those of PAHs (2.6–15.6 days) in invertebrates (*52, 53*). This suggested that metabolism would be the main factor influencing the trophodynamics of these chemicals.

Ratio of BDE-99/BDE-100 and BDE-99/BDE-47. Another important topic in PBDE accumulation studies was the variations of concentration ratios between congeners, and

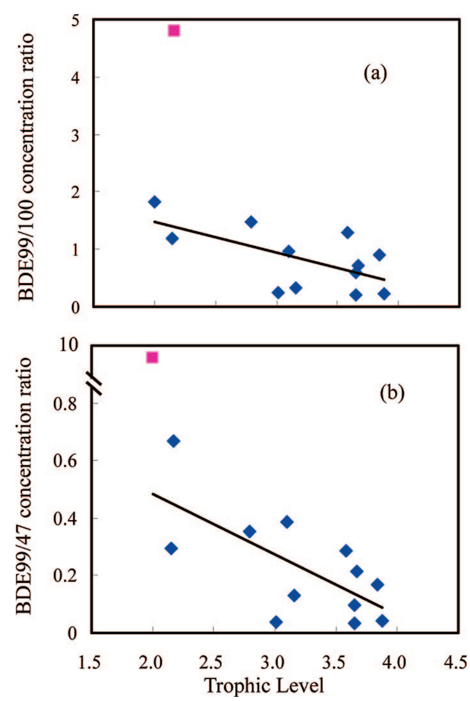

FIGURE 3. Relationship between concentration ratios and trophic levels of organisms in Bohai Bay, north China. (a) BDE-99/BDE-100 concentration ratios, slope = −0.5343, r^2 = 0.3926, p = 0.029; (b) BDE-99/BDE-47 concentration ratios, slope = −0.2094, r^2 = 0.4654, p = 0.015.

there have been several interesting investigations of concentration ratios between BDE-99 and BDE-100 in various matrices. Christensen et al. (2002) reviewed many investigations and found that the ratio between BDE-99 and BDE-100 in abiotic samples (air and sediment) were 80:20, which was similar to those in the industrial product Bromkal 70–5DE (84:16), but averaged 30:70 in biotic samples (fish and marine mammals) (*16*). The possible reason might be the higher bioavailibility of BDE-100 compared to BDE-99, or low biotransformation of BDE-100 (*16*). In 2003, Voorspoels et al. studied the ratios in various organisms in the Belgian North Sea, but found that the ratios varied among the species, ranging from 30:70 to 80:20 (three invertebrates and six fish species) (*23*). This phenomenon was explained as possibly being due to the different metabolism abilities of the organisms, which would increase as organisms climb the evolutionary ladder (*23*). Large differences among species were also found in the present study (zooplankton, five invertebrates, six fish species, and one bird): the concentration ratios between BDE-99 and BDE-100 ranged from 83:17 for short-necked clam to 17:83 for bartail flathead. To explore the main factors influencing the variation of the concentration ratios among organisms, a regression analysis was conducted between concentration ratios between BDE-99 and BDE-100 and trophic levels, as shown in Figure 3(a). It was found that the concentration ratios significantly decreased with increasing trophic levels except for clam, indicating that the trophic level of organisms would be a factor influencing the concentration ratios between BDE-99 and BDE-100. In fact, it has been reported that BDE-99 could be debrominated to BDE-47 by biotransformation in carp exposed to BDE-99 (*50*). So, we also estimated the concentration ratios between BDE-99 and its major metabolite (BDE-47), and then correlated these ratios with the trophic levels, as shown in Figure 3(b). The ratio was very high in zooplankton (9.7), and then decreased significantly with increasing trophic levels ($p = 0.015$), implying that the ability of BDE-99 to be biotransformed to BDE-47 increased with the trophic level. Thus, the trophic-level-dependent biotransformation possibly resulted in the increasing dominance of BDE-100 compared with BDE-99 and, therefore, the relatively high TMF of BDE-47 (7.24, $p = 0.006$) compared with other PBDE compounds.

Now, one of paradoxes about PBDE profile in accumulation studies is that BDE-47 was found to be the predominant congener in most organisms especially in high trophic level organisms (*54–58*), although the abundance of BDE-47 was relatively low in commercial PBDE mixtures, which were mainly consisted of deca-BDE (*59*). Our results suggested that the high concentrations of BDE-47 in high trophic level animals would be contributed by the debromination of high brominated PBDEs (e.g., BDE-99) and its relative high biomagnification potential in food webs.

Acknowledgments

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Supporting Information Available

Text, figures, and tables addressing (1) sample collection; (2) chemicals used in the analysis; (3) GC-MS condition; (4) details about quantitation and quality assurance quality control (QA/QC) for analysis; (5) lipid content analysis; (6) calculations of trophic magnification factor and statistical analysis; (7) sampling figure; (8) Table about the comparison of PBDEs levels in organisms from Bohai Bay with those reported in other locations. This material is available free of charge via the Internet at http://pubs.acs.org.

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