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Polybrominated diphenyl ethers (PBDEs) and indicator polychlorinated biphenyls (PCBs) in various marine fish from Zhoushan fishery, China

Xiaohong Shang [a], Guixian Dong [b], Hongxia Zhang [c], Lei Zhang [a], Xinwei Yu [d], Jingguang Li [a], Xiangyong Wang [e], Bing Yue [a], Yunfeng Zhao [a], Yongning Wu [a, *]

[a] Key Laboratory of Food Safety Risk Assessment, Ministry of Health, China National Center for Food Safety Risk Assessment, Beijing 100021, China
[b] Yantai Center for Disease Control and Prevention, Yantai 264003, China
[c] Maternal and Child Care Service Centre of Shiquan County, 725200 Shiquan County, Shaanxi Province, China
[d] Zhejiang Provincial Key Laboratory of Health Risk Factors for Seafood, Zhoushan Center for Disease Control and Prevention, Zhoushan 316021, China
[e] Sanquan Foods Co.,LTD, Zhengzhou 450044, China

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A B S T R A C T

Levels of polybrominated diphenyl ethers (PBDEs) and indicator polychlorinated biphenyls (indicator-PCBs) were measured in thirty-two commonly consumed marine fish species from Zhoushan Fishery, China. Factors effecting the accumulation of the compounds in sea fish were discussed, as well as the associated health risk via fish consumption. Levels and congener profiles of these contaminants varied by fish type. Due to the different accumulation influenced by fat content, feed habits and living zone in the sea area, levels of total nine PBDEs ranged from 0.0085 ng/g wet weight (Pagrosomus major) to 1.6819 ng/g wet weight (Bullet mackerel). The corresponding values for six indicator-PCBs ranged from 0.0124 ng/g wet weight (*Navodon septentrionalis)* to 3.8244 ng/g wet weight (*Bullet mackerel*). Total PBDEs and total indicator-PCBs intakes for Chinese population were estimated according to the contaminant level in each fish species and mean fish consumption in China, using an average body weight as 60 kg for the general population. The estimated intake of total PBDEs and total indicator-PCBs ranged from 0.007 ng/kg bw/day (*Pagrosomus major*) to 1.402 ng/kg bw/day (*B. mackerel*) and 0.010 ng/kg bw/day (*N. septentrionalis*) to 3.187 ng/kg bw/day (*B. mackerel*), respectively. The evaluated health risk associated with PBDEs intakes and indicator-PCBs intakes via consumption of all the fish species are relatively low for Chinese resident.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) are both lipophilic, persistent pollutants found worldwide in environmental and human samples. PBDEs are a group of effective brominated flame retardants and have long been used in the manufacture of electrical equipment, furniture, and mattresses etc. PBDEs have attractive growing concern worldwide regarding their properties of persistence, lipophilicity, bioaccumulation and biomagnify (Domingo, 2012; Frederiksen, Vorkamp, Thomsen, & Knudsen, 2009; Haffner and Schecter, 2014). Particularly, lower brominated congeners have more toxic effects on biota than higher brominated congeners. Exposure to PBDEs has been associated with alteration of thyroid hormones in adults (Huang et al., 2014; Turyk et al., 2008) and adverse neurological outcomes in children (Eskenazi et al., 2013; Herbstman et al., 2010). Therefore, penta-BDEs and octa-BDEs were banned in Japan, Europe and North American marketplace (Ross et al., 2009). PCBs are classic persistent organic pollutants (POPs) and were banned all over the world since 1980s due to significant toxic effects on human health, such as neurotoxicity, reproductive and developmental toxicity etc (EFSA, 2005).

In general, dietary intake, especially the intake of animal origin foods, is the main route for general population exposure to PBDEs and PCBs (Akutsu et al.,2008; Arnich et al., 2009; Baars et al.,2004; Fromme et al., 2009; Roosens et al., 2010; Zhang et al., 2013). Some

* Corresponding author.
E-mail addresses: shangxh@cfsa.net.cn (X. Shang), wuyongning@cfsa.net.cn (Y. Wu).

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studies have indicated that intake of PBDEs and PCBs mainly originates from fish or other aquatic foods because of the high contamination level in these foods, as well as high fish consumption (Chen et al.2013; Fattore, Fanelli, Dellatte, Turrini, & Domenico, 2008; Kiviranta, Ovaskainen, & Vartiainen, 2001; Liem, Furst, & Rappe, 2000; Voorspoels, Covaci, Neels, & Schepens, 2007). In China, aquatic foods also contributed the greatest quantity of PBDEs and PCBs to the dietary intake in southeast coastal regions (Zhang et al., 2013). Considering the increasing trend of fish consumption for Chinese population, it is necessary to regularly monitor these contaminants in fish. The objective of this study was to measure PBDEs and indicator-PCBs in marine fish species collected from the largest fishery in China and evaluate the associated health risk.

2. Materials and methods

2.1. Sampling

A total of 32 marine fish species (6 samples per specie) that most consumed by local residents were collected from Zhoushan fishery, the largest fish farming area in southeast China, from September to October in 2011. Edible parts of various fish samples were prepared by collectors immediately. The processed filets of samples were mixed and homogenized by species, weighed and freeze–dried. Meanwhile, the water content of each sample was recorded.

2.2. Analytical methods

Analysis of PBDEs and indicator-PCBs was described elsewhere, with slight modification (Zhang et al., 2011). Briefly, approximately 0.5 g of freeze–dried sample was spiked with ^{13}C labeled internal standards of PBDEs (^{13}C-BDE-28, -47, -66,-85,-99, -100, -153,-154, and -183) and ^{13}C-labeled internal standards of indicator-PCBs (^{13}C-PCB-28, -52, -101, -138, -153, and -180). The samples were extracted with a mixture of hexane/methylene chloride (1:1, v/v) using pressurized liquid extraction—ASE 300 system (Dionex, Sunnyvale, CA, USA) at 120 °C and 1500 psi. The bulk lipid was removed by stirring with acid-modified silica gel after solvent evaporation. Further cleanup was conducted using a Power Prep instrument (Fluid Management Systems, Waltham, MA, USA), and the eluting fraction containing PBDEs and indicator PCBs was collected and concentrated to approximately 50 μL. Then ^{13}C -labeled injection standards (^{13}C -PCB-70, -111, and -170 and ^{13}C -BDE-77 and -138) were added prior to instrument analysis.

The measurements of nine PBDE congeners and six indicator PCB congeners were performed using high resolution gas chromatography-high resolution mass spectrometry (HRGC-HRMS, MAT95XP, Thermo Finnigan, Germany) with DB-5MS capillary columns, 15 m × 0.25 mm i.d. × 0.1 μm, and 60 m × 0.25 mm i.d. × 0.25 μm, applied for the analysis of PBDEs and indicator-PCBs, respectively. The injector temperature and interface temperature both were 270 °C and the injection volume was 1 μL.

The temperature programme for determination of PBDEs was as follows: started at 120 °C (2 min), increased at 15 °C/min to 230 °C, at 5 °C/min to 270 °C, at 9 °C/min to 325 °C, and held for 2 min. For indicator-PCBs, the temperature programme started at 110 °C (1 min), increased by 15 °C/min to 180 °C, held for 1 min, 3 °C/min to 300 °C, held for 2 min.

The ΣPBDEs value was the sum of nine BDE congeners (BDE-28,47, 66, 85,99, 100, 153,154,183) and the ΣPCBs value was the sum of six indicator PCB congeners (CB-28,52, 101, 138,153,180).

2.3. Quality assurance and quality control

In this study, method blank samples were carried out for every eight samples. Spiking experiments of PBDEs and indicator-PCBs indicated the recoveries were in the range of 57%–115%. The limit of detection (LOD) ranged between 0.01 pg/g wet weight and 0.50 pg/g wet weight for PBDEs and between 0.01 pg/g wet weight and 0.40 pg/g wet weight for indicator-PCBs. The laboratory performance was validated by successfully participating in the interlaboratory comparison of PBDEs and indicator-PCBs in food organized by the Norwegian Institute of Public Health in 2010–2012. Z scores values ranged from −0.41 to 0.70 for sum PBDEs (u/BDE-209) and from −0.063 to 1.5 for sum indicator-PCBs.

2.4. Estimation of dietary intake and risk assessment

PBDEs and indicator-PCBs intakes from various fish species for average population were estimated by multiplying total PBDEs levels and total indicator-PCBs levels in each fish species with the fish consumption data (50 g/d) obtained from the Guide to Chinese Diet (Chinese Nutrition Society, 2007). A standard body weight of 60 kg was used (Song et al., 2011).

In the absence of a tolerable daily intake (TDI) value for PBDEs, the risk assessment of dietary intake of PBDEs was conducted using a margin of exposure approach (MOE approach) recommended by EFSA (EFSA, 2005). The MOE is calculated by comparing estimated dietary exposure to PBDEs with benchmarked dose lower confidance limit 10% ($BMDL_{10}$). Since relevant toxicity data were available only for BDE-47, -99, and -153, $BMDL_{10}$ recommended by EFSA were available only for these three individual PBDEs, which are 172 ng kg^{-1} bw day^{-1}, 4.2 ng kg^{-1} bw day^{-1} and 9.6 ng kg^{-1} bw day^{-1} respectively (EFSA, 2005). Therefore, risk assessment was carried out only for BDE-47, -99, and -153 in the present work (EFSA, 2011). The larger the MOE is, the smaller the potential health concern becomes.

2.5. Data analysis

SPSS 16.0 (SPSS of Windows; SPSS Inc., Chicago, IL) was used for the data analysis. The differences among groups were assessed by the paired samples t-test. A p-value of 0.05 (2-tailed) was chosen as the criterion for statistical significance.

3. Results and discussion

3.1. Residue levels and congener profiles of PBDEs

In the present study, nine PBDE congeners (BDE-28, 47, 66, 85, 99, 100,153, 154 and 183) were measured in 32 edible marine fish species and the levels of PBDEs were summarized in Table 1. The results showed that levels of PBDEs varied widely according to fish species with a factor about 200. The highest level of $\sum_9$PBDEs was found in *Bullet mackerel* (1.6819 ng/g wet weight), and the lowest level was observed in *Pagrosomus major* (0.0085 ng/g wet weight). Overall, the mean and median levels of $\sum_9$PBDEs were 0.2047 ng/g wet weight and 0.1452 ng/g wet weight, respectively.

Several studies have reported PBDEs levels in fish sample from coastal area of China. Liu (Liu et al., 2011) reported the total concentration of seven BDE (BDE-28, 47, 99, 100,153,154 and 183) was 0.115 ng/g w w, which was lower than that in the present work (0.1832 ng/g wet weight). Shen (Shen et al., 2009) reported PBDE levels in fish sample collected from several coastal provinces area in China. Among those provinces, Zhejiang and Fujian are located in the East China Sea coast, which is the same area to the present work. The average sum of nine BDE (BDE-28,47, 66, 85,99, 100, 153,154 and 183) in fish samples from Zhejiang and Fujian was 0.163 ng/g wet weight, which was a little lower than that in the present work (0.2047 ng/g wet weight). In the First Hong Kong

Total Diet Study (Chen et al., 2013), the mean PBDE levels (0.350 ng/g wet weight) in fish and seafood and their products, were also similar to the mean sum of PBDE in the present study.

The percentage contribution of PBDE congeners in various fish species were shown in Fig. 1. BDE-47 was the most abundant congener in nearly two third fish species, which was similar to the study on fatty fish samples from six coastal provinces of China (Shen et al.,2009). In this study, BDE-66 was another predominated congener in about one third fish species. The sum of BDE-47 and BDE-66 contributed nearly 50% to the total nine BDE. BDE-154 averagely contributed 20%, and other BDE averagely contributed less than 10%.

3.2. Residue levels and congener profiles of indicator-PCBs

The levels of six indicator-PCBs (PCB-28,52, 101, 138,153,180) were detected and the results were given in Table 2. The highest level of $\sum_6$ indicator-PCBs was found in *B. mackerel* (3.8244 ng/g wet weight), followed by *Mugil cephalus* (2.0981 ng/g wet weight) and *Astroconger Myriaster* (1.7381 ng/g wet weight). The lowest level was observed in *Navodon septentrionalis* (0.0124 ng/g wet weight) followed by *Lepidotrigla micropterus* (0.0374 ng/g wet weight). Overall, the mean level of $\sum_6$ indicator-PCBs was 0.805 ng/g wet weight, which was close to the results of previous study conducted in China (mean 0.797 ng/g wet weight, Shen et al., 2009).

According to the $\sum_7$PCB (PCB-28,52, 101, 118,138,153 and 180) maximum limit (500 ng/g wet weight) fixed for aquatic animals and their products issued by Chinese legislation, the $\sum_6$ indicator-PCBs in all the fish samples in the present study were far below the maximum limit. The $\sum_6$ indicator-PCBs for fish samples in this work were also below the maximum levels set for non dioxin-like PCBs (sum of PCB-28, 52, 101, 138,153 and 180) in fish and fishery products by EU, which is 75 ng/g wet weight. The percentage contributions of PCBs were shown in Fig. 2. Interspecies differences were also observed in the congener patterns for the various fish species. For most fish species, the predominating congeners were PCB-153 (average 30%), followed by PCB-138 (20%), similar to that in previous study in China (Liu et al., 2011; Shen et al.,2009).

In the present study, indicator-PCBs levels in fish species were much higher than PBDEs levels. This was consistent with the study on PBDEs and indicator-PCBs in marine fish in China (Liu et al., 2011). Significant correlation was observed between levels of $\sum_6$indicator-PCBs and $\sum_9$PBDEs ($r = 0.85$, $n = 35$). This suggested that PBDEs and indicator-PCBs in the studied fish species were probably from similar sources. Meanwhile, the highest levels of $\sum_9$PBDEs and $\sum_6$indicator-PCBs were both found in *B. mackerel*, and the lowest levels of $\sum_9$PBDEs and $\sum_6$indicator-PCBs were both found in *N. septentrionalis*. In our previous work (Wang et al., 2015), the highest and lowest levels of total PCDD/Fs and dl-PCBs were also found in this two fish species. This phenomenon can be explained by the difference in bioaccumulation among fish species.

As mentioned above, the contaminant levels were affected by many factors. Tables 3 and 4 showed the correlations between levels of $\sum_6$PCBs + $\sum_9$PBDEs and feeding habit, fat content, living habit in omnivorous fish and carnivorous fish respectively. The significant correlation between fat content and $\sum_6$PCBs + $\sum_9$PBDEs level was found ($r = 0.78$, $n = 32$, $p < 0.01$). For example, *B. mackerel*, which had the highest level of $\sum_6$PCBs + $\sum_9$PBDEs, also had the highest fat content (16.70%) among all the fish species. However, there was also exception. *Lateolabrax japonicus* had the low fat content (0.53%), while its

Table 1
Sample weight (kg) and levels of PBDEs and their sum ($\sum_9$PBDEs) in various fish species (ng/g wet weight).

Species	Sample weight		BDE28	BDE47	BDE66	BDE85	BDE99	BDE100	BDE153	BDE154	BDE183	$\sum_9$PBDEs
	Mean	Std										
Ablennes anastomella	1.15	0.22	0.0227	0.0949	0.0010	0.0003	0.0130	0.0309	0.0079	0.0378	0.0015	0.2100
Anchoa lucida	0.04	0.01	0.0098	0.0439	0.0060	0.0006	0.0113	0.0110	0.0072	0.0313	0.0041	0.1252
Astroconger Myriaster	1.08	0.15	0.0196	0.0801	0.0065	0.0064	0.0177	0.0331	0.0510	0.1407	0.0156	0.3707
Bombay duck	1.13	0.20	0.0034	0.0216	0.0014	0.0006	0.0039	0.0058	0.0037	0.0144	0.0001	0.0549
Bullet mackerel	1.25	0.15	0.0785	0.5003	0.0955	0.0158	0.1713	0.2400	0.1085	0.4701	0.0019	1.6819
Cephalopholis boenak	1.02	0.23	0.0084	0.0606	0.0012	0.0011	0.0074	0.0103	0.0058	0.0081	0.0018	0.1047
Collichthys lucidus	0.92	0.13	0.0030	0.0149	0.0033	0.0002	0.0061	0.0044	0.0033	0.0094	0.0002	0.0448
Engraulis japonicus	0.26	0.12	0.0027	0.0255	0.0010	0.0033	0.0043	0.0178	0.0040	0.0380	0.0089	0.1055
Goniistius quadricornis	0.93	0.24	0.0103	0.0750	0.0090	0.0011	0.0515	0.0263	0.0164	0.0441	0.0002	0.2339
Ilisha elongata	1.37	0.15	0.0113	0.0646	0.0050	0.0004	0.0155	0.0167	0.0122	0.0452	0.0008	0.1717
Lateolabrax japonicus	1.23	0.19	0.0073	0.0262	0.0470	0.0008	0.0025	0.0080	0.0020	0.0113	0.0028	0.1079
Lepidotrigla micropterus	0.32	0.11	0.0011	0.0091	0.0167	0.0004	0.0005	0.0027	0.0010	0.0038	0.0008	0.0361
Miichthys miiuy	1.03	0.08	0.0045	0.0339	0.0599	0.0010	0.0105	0.0111	0.0060	0.0249	0.0008	0.1526
Mugil cephalus	1.10	0.13	0.0337	0.0650	0.1143	0.0003	0.0076	0.0153	0.0047	0.0397	0.0204	0.3010
Muraenesox bagio	1.13	0.16	0.0131	0.1044	0.0050	0.0019	0.0232	0.0291	0.0129	0.0616	0.0015	0.2527
Navodon septentrionalis	1.33	0.17	0.0013	0.0042	0.0007	0.0002	0.0010	0.0012	0.0009	0.0020	0.0005	0.0120
Nimbochromis livingstonii	0.42	0.15	0.0028	0.0499	0.0885	0.0019	0.0362	0.0155	0.0139	0.0230	0.0018	0.2335
Oplegnathus fasciatus	1.07	0.13	0.0136	0.1231	0.0020	0.0027	0.0052	0.0226	0.0092	0.0508	0.0157	0.2449
Pagrosomus major	1.09	0.20	0.0009	0.0030	0.0005	0.0001	0.0007	0.0009	0.0007	0.0014	0.0003	0.0085
Pampus chinensis	1.14	0.13	0.0052	0.0431	0.0036	0.0019	0.0155	0.0192	0.0093	0.0351	0.0048	0.1377
Pangio kuhlii	0.87	0.21	0.0107	0.0629	0.0035	0.0005	0.0077	0.0235	0.0075	0.0478	0.0025	0.1666
Paralichthys olivaceus	0.96	0.12	0.0016	0.0152	0.0271	0.0003	0.0009	0.0043	0.0015	0.0066	0.0015	0.0590
Percocypris pingi	0.53	0.17	0.0015	0.0080	0.0145	0.0006	0.0021	0.0024	0.0018	0.0021	0.0068	0.0398
Pneumatophorus japonicus	1.13	0.25	0.0336	0.1129	0.0185	0.0064	0.0656	0.0323	0.0260	0.0614	0.0066	0.3633
Pseudosciaena polyactis	0.58	0.10	0.0111	0.0671	0.0067	0.0027	0.0214	0.0231	0.0117	0.0597	0.0011	0.2046
Rachycentron canadum	3.20	0.40	0.0046	0.0324	0.0022	0.0014	0.0059	0.0091	0.0038	0.0115	0.0009	0.0718
Scomberomorus niphoniu	1.08	0.23	0.0106	0.0723	0.0017	0.0006	0.0135	0.0266	0.0038	0.0278	0.0020	0.1589
Sebastiscus marmoratus	1.20	0.16	0.0040	0.0228	0.0408	0.0003	0.0009	0.0053	0.0021	0.0113	0.0006	0.0881
Setipinna taty	0.87	0.21	0.0231	0.1627	0.0164	0.0019	0.0214	0.0556	0.0204	0.1234	0.0033	0.4282
Tenualosa reevesii	0.93	0.24	0.0313	0.1223	0.0088	0.0015	0.0080	0.0139	0.0117	0.0105	0.0101	0.2181
Trichiurus lepturus	0.92	0.25	0.0054	0.0412	0.0020	0.0015	0.0075	0.0114	0.0045	0.0245	0.0006	0.0986
Zebrias zebra	0.56	0.18	0.0048	0.0108	0.0196	0.0002	0.0010	0.0047	0.0038	0.0164	0.0016	0.0629

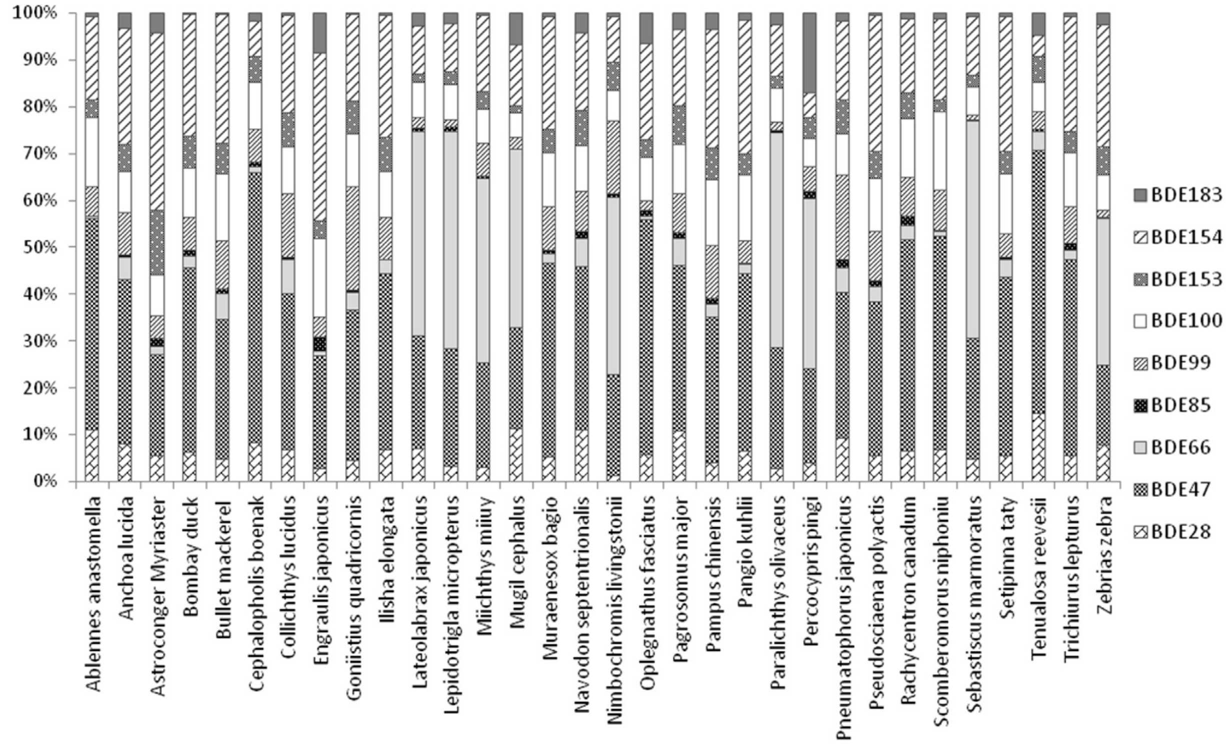

Fig. 1. The profiles of PBDE congeners in various fish species.

Table 2
Levels of PCBs and their sum ($\sum_6$PCB) in various fish species (ng/g wet weight).

Species	PCB28	PCB52	PCB101	PCB138	PCB153	PCB180	$\sum_6$PCB
Ablennes anastomella	0.1526	0.1135	0.1002	0.1395	0.1804	0.0361	0.7223
Anchoa lucida	0.0822	0.1371	0.1035	0.0913	0.1247	0.0386	0.5772
Astroconger myriaster	0.1295	0.1472	0.2078	0.4164	0.6462	0.1909	1.7381
Bombay duck	0.0327	0.0313	0.0466	0.0645	0.0892	0.0209	0.2851
Bullet mackerel	0.4096	0.2892	0.6002	0.8775	1.2156	0.4322	3.8244
Cephalopholis boenak	0.1051	0.1787	0.1854	0.4254	0.6248	0.1930	1.7124
Collichthys lucidus	0.0437	0.0307	0.0292	0.0385	0.0756	0.0192	0.2369
Engraulis japonicus	0.0127	0.0176	0.0450	0.0994	0.1702	0.0644	0.4093
Goniistius quadricornis	0.2384	0.1266	0.0868	0.1300	0.1740	0.0299	0.7856
Ilisha elongata	0.2255	0.1111	0.0988	0.1388	0.2487	0.0925	0.9154
Lateolabrax japonicus	0.0399	0.0528	0.0297	0.0527	0.0701	0.0279	0.2731
Lepidotrigla micropterus	0.0014	0.0012	0.0043	0.0101	0.0144	0.0059	0.0374
Miichthys miiuy	0.0377	0.0492	0.0619	0.0915	0.1248	0.0390	0.4042
Mugil cephalus	0.3980	0.3308	0.3435	0.3206	0.5806	0.1245	2.0981
Muraenesox bagio	0.0601	0.1343	0.1793	0.2923	0.4286	0.1159	1.2105
Navodon septentrionalis	0.0007	0.0015	0.0021	0.0028	0.0038	0.0016	0.0124
Nimbochromis livingstonii	0.0262	0.0360	0.0460	0.0661	0.0920	0.0268	0.2931
Oplegnathus fasciatus	0.0890	0.1319	0.2550	0.3060	0.3981	0.0806	1.2606
Pagrosomus major	0.1964	0.1327	0.1506	0.1916	0.2677	0.0879	1.0269
Pampus chinensis	0.0556	0.0448	0.0648	0.0900	0.1445	0.0550	0.4548
Pangio kuhlii	0.0501	0.0522	0.1102	0.1472	0.2229	0.0648	0.6474
Paralichthys olivaceus	0.0167	0.0135	0.0204	0.0346	0.0474	0.0146	0.1472
Percocypris pingi	0.0122	0.0101	0.0077	0.0445	0.0967	0.0378	0.2090
Pneumatophorus japonicus	0.2299	0.2443	0.2396	0.1869	0.2764	0.0710	1.2481
Pseudosciaena polyactis	0.0841	0.0705	0.0902	0.0984	0.1458	0.0443	0.5334
Rachycentron canadum	0.0415	0.0316	0.0383	0.0665	0.0894	0.0253	0.2927
Scomberomorus niphoniu	0.0543	0.1008	0.1173	0.1503	0.2026	0.0599	0.6852
Sebastiscus marmoratus	0.0308	0.0330	0.0312	0.0393	0.0590	0.0149	0.2083
Setipinna taty	0.0920	0.1729	0.2964	0.3402	0.5245	0.1455	1.5716
Tenualosa reevesii	0.2647	0.2654	0.2202	0.2154	0.3201	0.0852	1.3711
Trichiurus lepturus	0.0482	0.0579	0.0728	0.0777	0.1204	0.0381	0.4152
Zebrias zebra	0.0287	0.0435	0.0153	0.0169	0.0322	0.0076	0.1442

$\sum_6$PCBs + $\sum_9$PBDEs level was not much low. There might be other factors except for fat contents affecting $\sum_6$PCB + $\sum_9$PBDEs levels in fish.

The living zone areas could influence the compounds levels in fish species, too. As shown in Table 3, for omnivorous fish species like *Astroconger Myriaster*, *M. cephalus*, *Oplegnathus fasciatus and*

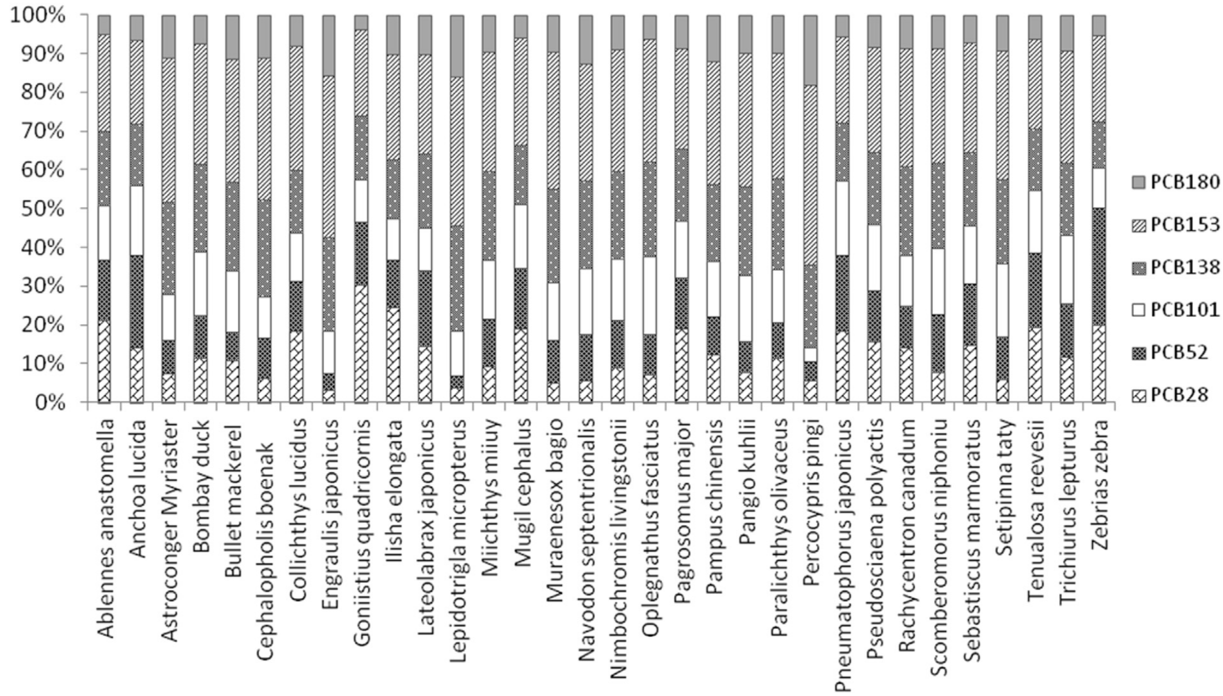

Fig. 2. The profiles of indicator-PCBs congeners in various fish species.

Tenualosa reevesii, which have higher levels of $\sum_6$PCB + $\sum_9$PBDEs usually live in shallow waters, rocks area and water plants, or belong to oceanodromous migration. For carnivorous fish (Table 4), *B. mackerel*, *Setipinna taty*, *Pneumatophorus japonicas*, which have higher levels of $\sum_9$PBDEs, live in rocks area, sediments or belong to oceanodromous migration. However, *L. micropterus*, which live in sediments have the lowest $\sum_6$PCB + $\sum_9$PBDEs level. For fish species live in shallow waters, the $\sum_6$PCB + $\sum_9$PBDEs level also varied significantly. Therefore, contaminants levels in fish might be influenced by complicated factors including fat content, feeding habit, as well as fish living environment and so on.

3.3. *Estimation of intake of PBDEs and indicator-PCBs and risk assessment*

Based on $\sum_9$PBDEs and $\sum_6$indicator-PCBs levels, the estimated PBDEs and indicator-PCBs intakes were calculated for Chinese population (Table 5). The estimated intakes of total PBDEs from the various fish species ranged from 0.007 ng/kg bw/day (*P. major*) to 1.402 ng/kg bw/day (*B. mackerel*) with a mean level of 0.171 ng/kg bw/day.

In the absence of a tolerable daily intake (TDI) value for PBDEs, the associated health risk of PBDEs intake was evaluated by MOE approach, which is recommended by EFSA. In the consideration of EFSA, it was unlikely to be a health concern in the case of PBDEs in principle any MOE larger than 2.5 (EFSA, 2005). Based on the PBDEs intakes in the present study, the calculated MOE ranged from 71,348 to 14,117,647, with average of 1,775,123. Therefore, such large MOE values suggested that the health risk of PBDEs intake via consumption of the studied fish species is very low.

With respect to PCBs, the estimated intakes from the various fish species ranged from 0.010 ng/kg bw/day (*N. septentrionalis*) to 3.187 ng/kg bw/day (*B. mackerel*). The mean level of intakes (0.671 ng/kg bw/day) in the present study was less than those from Dutch (1.25 ng/kg bw/day) (Baars et al.,2004), France (3.62 ng/kg bw/day) (Arnich et al., 2009), and Italian (4.58 ng/kg bw/day) (Fattore et al.,2008). The results indicated that the health risk of PCBs intake via consumption of the studied fish species was relatively low.

4. Conclusion

Nine PBDEs and six indicator-PCBs were analyzed in 32 marine fish species collected in the largest fishery in China. Both the levels and congener profiles of these contaminants showed species difference. There was no notably associated health risk with PBDEs

Table 3
Correlations between levels of $\sum_6$PCB and $\sum_9$ PBDEs and feeding habit, fat content, living habit in omnivorous fish.

Species	$\sum_6$PCBs (ng/g ww)	$\sum_9$PBDEs (ng/g ww)	$\sum_6$PCBs + $\sum_9$PBDEs (ng/g ww)	Fat content (%)	Living habit
Mugil cephalus	2.0981	0.3010	2.3991	8.08	Live in shallow water
Astroconger myriaster	1.7381	0.3707	2.1088	12.56	Live in the rocks area and water plants
Tenualosa reevesii	1.3711	0.2181	1.5892	14.4	Oceanodromous migration
Oplegnathus fasciatus	1.2606	0.2449	1.5055	4.92	Live in shallow water
Ablennes anastomella	0.7223	0.2100	0.9323	5.86	Live in the depth of 1–50 m
Pangio kuhlii	0.6474	0.1666	0.8140	3.24	Live in the rocks area and water plants
Bombay duck	0.2851	0.0549	0.3400	1.02	Live in the middle water
Navodon septentrionalis	0.0124	0.0120	0.0244	0.41	Live in the depth of 50–120 m
Mean			1.2142		

Table 4
Correlations between levels of $\sum_6$PCB and $\sum_9$PBDEs and feeding habit, fat content, living habit in carnivorous fish.

Species	$\sum_6$PCB (ng/g ww)	$\sum_9$PBDEs (ng/g ww)	$\sum_6$PCB + $\sum_9$PBDEs (ng/g ww)	Fat content (%)	Living habit
Bullet mackerel	3.8244	1.6819	5.5063	16.7	Live in the rocks area
Setipinna taty	1.5716	0.4282	1.9998	11.26	Live in the depth of 4–13 m and in the sediments
Cephalopholis boenak	1.7124	0.1047	1.8171	7.46	Live in the depth of 1–64 m
Pneumatophorus japonicus	1.2481	0.3633	1.6114	15.51	Oceanodromous migration
Muraenesox bagio	1.2105	0.2527	1.4632	6.17	Live in the depth of 50–80 m
Ilisha elongata	0.9154	0.1717	1.0871	7.85	Live in nearshore shallow water
Pagrosomus major	1.0269	0.0085	1.0354	8.34	Live in nearshore shallow water and in the rocks area as well as
Goniistius quadricornis	0.7856	0.2339	1.0195	7.57	Live in nearshore shallow water
Scomberomorus niphoniu	0.6852	0.1589	0.8441	3.34	Oceanodromous migration
Pseudosciaena polyactis	0.5334	0.2046	0.7380	3.60	Oceanodromous migration, live in the depth of 20–80 m
Anchoa lucida	0.5772	0.1252	0.7024	8.72	Live in shallow water
Pampus chinensis	0.4548	0.1377	0.5925	6.10	Live in the depth of 1–10 m
Miichthys miiuy	0.4042	0.1526	0.5568	2.05	Live in the depth of 15–70 m
Nimbochromis livingstonii	0.2931	0.2335	0.5266	3.83	Live in shallow water
Engraulis japonicus	0.4093	0.1055	0.5148	0.97	Live in the depth of 50–60 m
Trichiurus lepturus	0.4152	0.0986	0.5138	3.74	Live in the middle water
Lateolabrax japonicus	0.2731	0.1079	0.3810	0.53	Live in nearshore shallow water
Rachycentron canadum	0.2927	0.0718	0.3645	3.74	Oceanodromous migration
Sebastiscus marmoratus	0.2083	0.0881	0.2964	1.84	Live in the rocks area
Collichthys lucidus	0.2369	0.0448	0.2817	1.10	Live in the depth of about 20 m and in the sediment
Percocypris pingi	0.2090	0.0398	0.2488	0.69	Live in the depth of 1–70 m
Zebrias zebra	0.1442	0.0629	0.2071	1.01	Live in the sediments
Paralichthys olivaceus	0.1472	0.0590	0.2062	0.34	Live in shallow water
Lepidotrigla micropterus	0.0374	0.0361	0.0735	0.56	Live in the sediment
Mean			0.9412		

Table 5
The intakes of total PBDEs and total indicator-PCBs and MOEs for total PBDEs.

Species	$\sum_9$PBDEs		$\sum_7$PCBs estimated intake (ng/kg bw/day)
	Estimated intake (ng/kg bw/day)	MOE	
Ablennes anastomella	0.175	571429	0.602
Anchoa lucida	0.104	958466	0.481
Astroconger Myriaster	0.309	323712	1.448
Bombay duck	0.046	2185792	0.238
Bullet mackerel	1.402	71348	3.187
Cephalopholis boenak	0.087	1146132	1.427
Collichthys lucidus	0.037	2678571	0.197
Engraulis japonicus	0.088	1137441	0.341
Goniistius quadricornis	0.195	513040	0.655
Ilisha elongate	0.143	698893	0.763
Lateolabrax japonicus	0.090	1112141	0.228
Lepidotrigla micropterus	0.030	3324100	0.031
Miichthys miiuy	0.127	786370	0.337
Mugil cephalus	0.251	398671	1.748
Muraenesox bagio	0.211	474871	1.009
Navodon septentrionalis	0.010	10000000	0.010
Nimbochromis livingstonii	0.195	513919	0.244
Oplegnathus fasciatus	0.204	489996	1.051
Pagrosomus major	0.007	14117647	0.856
Pampus chinensis	0.115	871460	0.379
Pangio kuhlii	0.139	720288	0.540
Paralichthys olivaceus	0.049	2033898	0.123
Percocypris pingi	0.033	3015075	0.174
Pneumatophorus japonicus	0.303	330306	1.040
Pseudosciaena polyactis	0.171	586510	0.444
Rachycentron canadum	0.060	1671309	0.244
Scomberomorus niphoniu	0.132	755192	0.571
Sebastiscus marmoratus	0.073	1362089	0.174
Setipinna taty	0.357	280243	1.310
Tenualosa reevesii	0.182	550206	1.143
Trichiurus lepturus	0.082	1217039	0.346
Zebrias zebra	0.052	1907790	0.120

and indicator-PCBs via consumption of the studied fish species for Chinese population. In order to protect health, we should select the fish species which accumulate less PBDEs and PCBs. In addition, the monitoring of contaminants in fish should be carried out regularly to assess the exposure level in time.

Conflict of interest

The authors declare that there are no conflicts of interest.

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Xiaohong Shang and Guixian Dong contributed equally to this work.

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