Environ. Sci. Technol. **2010,** *44,* 5781–5786

Tissue Concentrations of Polybrominated Compounds in Chinese Sturgeon (*Acipenser sinensis*): Origin, Hepatic Sequestration, and Maternal Transfer

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Received January 31, 2010. Revised manuscript received June 3, 2010. Accepted June 29, 2010.

Information on concentrations of polybrominated compounds in various tissues of wild fish is limited. Concentrations of polybrominated diphenyl ethers (PBDEs), methoxylated PBDEs (MeO-PBDEs), and hydroxylated PBDEs (OH-PBDEs) were measured in 12 organs and eggs of 17 female Chinese sturgeon (*Acipenser sinensis*). The highest concentrations of PBDEs (42.8 ± 39.4 ng/g ww), and MeO-PBDEs (135 ± 63.6 pg/g ww) occurred in adipose followed by liver (PBDEs: 25.0 ± 27.0 ng/g ww, MeO-PBDEs: 32.3 ± 29.1 pg/g ww) and eggs (PBDEs: 21.2 ± 19.4 ng/g ww, MeO-PBDEs: 120 ± 119 pg/g ww), and the highest concentration of OH-PBDEs was observed in liver (185 ± 174 pg/g ww) and eggs (178 ± 294 pg/g ww). The lack of in vitro transformation of 6-MeO-BDE47 or BDE47 by microsomes prepared from Chinese sturgeon liver suggests that most 6-OH-BDE47 was directly accumulated as a natural product. Lipid-normalization revealed preferential accumulation of PBDEs in liver, and ratios of concentrations between eggs and liver were 0.10 ± 0.11 to 0.22 ± 0.26, which was lower than that for MeO-PBDEs (6-MeO-BDE47: 0.57 ± 0.60, 2′-MeO-BDE68: 0.65 ± 0.85) and 6-OH-BDE47 (0.59 ± 0.51). Concentrations of PBDEs were negatively correlated with age, but no significant relationships between concentrations of OH-PBDEs or MeO-PBDEs and age were observed.

Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of flame retardants used in a wide range of products such as textiles, construction materials, and electronic equipment (*1*). Environmental occurrences (*2–4*), fates (*5*), and toxicities of PBDEs (*6–9*) have been reported. Methoxylated polybrominated diphenyl ethers (MeO-PBDEs) and hydroxylated polybrominated diphenyl ethers (OH-PBDEs) are also toxic (*10*). The presence of OH-PBDEs in wildlife and humans is of concern due to their higher toxic potencies relative to PBDEs and MeO-PBDEs (*11*). Some OH-PBDEs bind to human transthyretin with affinities 3-fold higher than the natural ligand thyroxine (T4) (*12*). Some OH-PBDEs such as 4′-OH-BDE17 can displace 17-β estradiol (E2) from the estrogen receptor (ER)-α with a relative estrogenic potency approximately 30% higher than that of E2 (*13*).

OH-PBDEs have been reported to originate from both anthropogenic and natural sources. They have been reported to be transformed from PBDEs by female Sprague−Dawley rats (*14*) or from MeO-PBDEs by rainbow trout, chicken, and rat microsomes (*15*). OH-PBDEs and MeO-PBDEs have also been reported to be synthesized by marine organisms such as algae and sponges or their associated microorganisms (*16, 17*). There are reports that OH-PBDE can be formed in vitro and in vivo from PBDE, but the literature is ambivalent with some authors reporting transformation while others did not observe transformation. The proportions transformed are small even when exposures were large (*5, 15, 18, 19*). Purity of the exposure mixtures is always an issue in these sorts of studies and the reported formation of OH-BDEs and MeO-BDE from PBDE may be an artifact. When the precursor materials were confirmed to not contain OH-BDE or MeO-BDE no transformation was observed (*15, 18*). OH-PBDEs and MeO-PBDEs have been reported in many aquatic organisms including marine and freshwater fish, birds, and higher trophic level organisms such as marine mammals including beluga whales, ringed seals, and polar bears (*20–22*). However, information on concentrations of OH-PBDEs and MeO-PBDEs is mainly restricted to blood, adipose, and liver of wildlife. There have been few studies of the toxicokinetics of OH-PBDEs, MeO-PBDEs, and PBDEs (*23, 24*), which could help clarify the tissue concentration, maternal transfer, and age-related accumulation of these polybrominated compounds in organisms and help identify possible target organs to guide monitoring and studies of toxicity.

For oviparous organisms, such as some fish, transfer of hydrophobic chemicals from females to eggs along with yolk proteins is a pathway of exposure of eggs (*25*). Organisms usually exhibit higher sensitivity to pollutant exposure during early life stage than adult life stages (*26, 27*), and various effects of polybrominated compounds on embryos have been reported. Exposure of zebrafish embryos to BDE47 resulted in development of morphological, cardiac, and neural deficits (*7*). Bromkal 70-DE, a commercial mixture containing predominantly BDE47, 99, and 100, delayed and altered activity level, fright response, predation rates, and learning ability in subsequent life stages (*8*). Exposure of zebrafish embryos to 6-OH-BDE47 caused a wide range of developmental defects, including pericardial edema, yolk sac deformations, reduced pigmentation, and lowered heart rate as well as delayed development, with an EC_{50} value of 25 nM (*28*).

The Chinese sturgeon (*Acipenser sinensis*) is a predatory fish that can live for 40 years or longer and weigh as much as 500 kg (*29*). Previous studies have shown that Chinese sturgeon can be exposed to pollutants and accumulate especially high concentrations into their eggs (*27, 30*). In this study, concentrations of 9 OH-PBDEs, 12 MeO-PBDEs, and 11 PBDEs were measured in thirteen organs, including 8

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10.1021/es100348g
Published on Web 07/07/2010

liver, 8 muscle, 6 heart, 7 gonad, 5 stomach, 7 intestine, 5 adipose, 6 gill, 2 pancreas, 1 kidney, 1 gallbladder, 1 spleen, and 15 egg samples from 17 Chinese sturgeon. Tissue-dependent accumulations of the three classes of compounds were examined and hepatic sequestration and maternal transfer rates were calculated. Finally, age-related accumulation was compared among PBDEs, OH-PBDEs, and MeO-PBDEs.

Materials and Methods

Sample Collection. Because the Chinese sturgeon has been listed as a grade I protected animal in China since the 1980s, the capture of Chinese sturgeon was authorized strictly for scientific purposes only. Artificial propagation has been implemented to save this endangered species. This program captures adult Chinese sturgeon from the Yangtze River. Chinese sturgeon migrate from the East Sea and spend approximately one year in the river before spawning. After propagation, sturgeon were released back into the Yangtze River, but some did not survive. Eggs were collected before propagation and the other organs and tissues came from the 17 sturgeon that died during artificial propagation in the period between 2003 and 2006. To avoid contamination of stomach and intestine with gut contents, only the inner layer of the stomach and intestine were collected. This was possible because of their large body size and because the muscle layer of the stomach and intestine are very thick. After collection, tissues were frozen immediately at −20 °C until analysis. The ages of fish were determined by counting growth layers in the cleithrum, as described previously (*29*). The total number of sturgeon samples was 72, and the detailed information about the samples is given in Supporting Information Table S1.

In Vitro Microsomal Incubations. Microsomes were isolated from cultured two-year-old Chinese sturgeon, according to the method improved by Benedict et al. (*31*) and included dithiothreitol (DTT) in the homogenization, wash and resuspension buffers to preserve catalytic activity of reductases and deiodinases. Ethoxyresorunfin *O*-deethylase (EROD) activity and protein content of microsomes were determined simultaneously by use of a fluorescence kit (Genmed Scientific Inc., USA). The final reaction volume was 100 μL and contained either 50 μL of the microsomal preparation and 3 μL of exposure chemicals. Individual congeners (BDE47, BDE99, BDE154, BDE183, and 6-MeO-BDE47) were used, and the concentration in the incubation mixture was 150 ng/mL. The protein concentration in the reaction vial was 5.2 mg/mL and the CYP1A1-catalyzed EROD activity was 3.5 pmol/mg/min. Reactions were performed at 37 °C for 20 h with constant agitation. Incubations without chemicals and without microsomes were used as negative controls to assess background contaminants and the possibility of nonenzyme-mediated changes in chemical structure. After the incubation, the samples were extracted immediately for chemical analysis.

Quantification of Target Analytes and Transformation Products in Tissues and Microsomes. The method used to quantify PBDEs, OH-PBDEs, and MeO-PBDEs has been described previously (*15*) and details specific to this study are given in Supporting Information.

Quality Assurance and Quality Control (QA/QC). Details of the QA/QC of the method have been described previously (*15*). Concentrations of all congeners were quantified by the internal standard isotope-dilution method with mean relative response factors determined from standard calibration runs. PBDEs and MeO-PBDEs were quantified in sample extracts relative to ^{13}C-PBDEs, and OH-PBDEs were quantified relative to 6′-OH-BDE17. Recoveries of ^{13}C-PBDEs and 6′-OH-BDE17 were 74.9 ± 38.8% to 130 ± 60.0% and 98.5 ± 33.7%, respectively. All equipment rinses were carried out with acetone and hexane to avoid sample contamination. A laboratory blank was incorporated in the analytical procedures for every batch of 12 samples. The method detection limits (MDLs) were set to be the mean of the concentration plus three times the standard deviation in the blank samples in which BDE28, BDE47, BDE85, BDE154, BDE153, and BDE138 were detected. The MDLs for the other compounds, which were not detected in blank samples, were set to the instrumental minimum detectable amounts. Based on an average sample with a wet mass of 10 g, the detection limits were 0.4 pg/g ww for MeO-PBDEs; 2.0 pg/g ww for 2′-OH-6′-Cl-BDE7; 4.0 pg/g ww for 3-OH-BDE47, 5-OH-BDE47, 2′OH-BDE68, 6-OH-BDE47, 4′-OH-BDE49, 2′-OH-6′-Cl-BDE68, 6-OH-BDE90, and 2-OH-BDE123; 1.6 pg/g ww for BDE28; 26.8 pg/g ww BDE47; 48.8 pg/g ww for BDE85; 22.4 pg/g ww for BDE154; 3.2 pg/g ww for BDE153; 11.2 pg/g ww for BDE138; and 0.4 pg/g ww for BDE66, BDE77, BDE100, BDE99, and BDE183. Purity of stock chemicals was checked by measuring, and concentrations of potential transformation products in stocks were determined by quantification of concentrated standards.

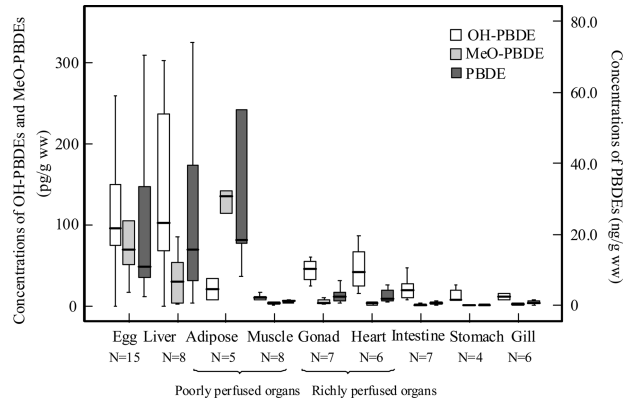

FIGURE 1. Concentrations of ΣOH-PBDEs, ΣMeO-PBDEs, and ΣPBDEs in tissues of Chinese sturgeon. Due to the limited number of samples, kidney, spleen, and gallbladder are not presented. The horizontal line represents the median concentration. The 25th and 75th percentiles define the boxes and the whiskers represent the 10th and 90th percentiles. Of the five stomach samples, concentrations of PBDEs in one sample exceeded three times the standard deviation so that data point was not included in calculating the median.

Statistical Analysis. In this study, for those results lower than the MDL, half of the MDL was assigned to avoid missing values in statistical analyses. The average liver/adipose and egg/liver ratios calculated on a lipid weight basis were calculated for individual PBDE and MeO-PBDE congener and the egg/liver ratios base on wet weight was calculated for the 6-OH-BDE47 in Chinese sturgeon. All data analyses such as linear regression were performed with SPSS 15.0. Statistical significance was defined as $p < 0.05$.

Results and Discussion

Concentrations among Tissues. Higher concentrations of ΣPBDEs were observed in adipose (42.8 ± 44.0 ng/g ww), liver (25.0 ± 27.0 ng/g ww), and eggs (21.2 ± 19.4 ng/g ww) (Figure 1). Detailed information on concentrations of PBDEs, OH-PBDEs, and MeO-PBDEs in tissues of 17 Chinese sturgeon are shown in Supporting Information Tables S2 and S3. Concentrations of ΣPBDEs in eggs of Chinese sturgeon (21.3 ± 19.4 ng/g ww) were higher than those of sturgeons from Azerbaijan (*Acipenser Huso huso*), Bulgaria (*Acipenser guldenstaedti*), and Russia (*Acipenser stellatus*) (0.010−0.27 ng/g ww) (*32*). Concentrations of PBDEs in skipjack tuna from the East China Sea were relative high compared to other areas of the world (*33*). This is consistent with the concentrations

of PBDEs observed in the Chinese sturgeon which is an anadromous fish that spends its first 14 years of life in the East China Sea (*29*).

The highest concentrations of MeO-PBDEs were found in adipose (135 ± 63.6 pg/g ww), eggs (120 ± 119 pg/g ww), and liver (32.3 ± 29.1 pg/g ww). Concentrations of 6-MeO-BDE47 in liver of Chinese sturgeon were lower than those in beluga whales from Eastern Hudson Bay and the Hudson Strait (*34*) and in tuna (*Katsuwonus pelamis*) from the North Pacific Ocean (*15*).

Both PBDEs and MeO-PBDEs were preferentially accumulated in organs with higher lipid contents such as adipose (65.9 ± 13.8%), egg (35.1 ± 8.5%), and liver (12.7 ± 6.6%). When concentrations of PBDEs and MeO-PBDEs were expressed on a lipid weight basis, differences in concentrations among tissues were less (Supporting Information Figure S1). But, concentrations of PBDEs were still higher in liver (157 ± 114 ng/g lw) than in other tissues, which suggests that the lipid of tissues is not the sole factor influencing the tissue concentrations of PBDEs in Chinese sturgeon.

The highest concentrations of OH-PBDEs occurred in liver (185 ± 174 pg/g ww) and eggs (178 ± 294 pg/g ww). Concentrations of 6-OH-BDE47, a major congener of OH-PBDEs, in liver (166 ± 182 pg/g ww) of Chinese sturgeon were higher than those (17.8−44.0 pg/g ww) in liver of tuna (*Katsuwonus pelamis*) collected from the North Pacific Ocean (*15*). The tendency of OH-PBDEs to associate with liver may be due to their biotransformation formation in this tissue and/or competitive binding to the major thyroid hormone transport protein (TTR) (*12*), which is mainly produced and expressed in liver (*35*). Among target tissues, gonad and heart could be classified as richly perfused tissues, and adipose and muscle as poorly perfused tissues (*36*). Concentrations of OH-PBDEs in two richly perfused tissues (gonad: 42.8 ± 39.4 pg/g ww, heart: 46.1 ± 26.5 pg/g ww) were higher than those in poorly perfused tissues (adipose: 35.0 ± 40.1 pg/g ww, muscle: 11.1 ± 14.3 pg/g ww). This observation indicates that blood flow could be an important factor to influence the tissue concentrations of OH-PBDEs. OH-PBDEs have been shown to bind competitively to TTR leading to their accumulation in blood (*12, 37*).

Relatively high concentrations of PBDEs (28.5 ng/g ww, $n = 1$) and OH-PBDEs (138 pg/g ww) were observed in gall bladder. This indicates possible preferential disposition of PBDEs and OH-PBDEs in bile, which could facilitate excretion of these compounds from Chinese sturgeon.

Patterns of Relative Concentrations. All of identifiable tri- to hepta-BDE congeners (BDE28, 47, 66, 77, 99, 100, 85, 153, 154, and 183) were detected, and the patterns of relative concentrations were similar among tissues except for stomach (Figure 2). BDE47 (52.5 ± 13.8%) was the predominant congener, followed by BDE154 (16.7 ± 8.4%), BDE100 (12.2 ± 2.8%), BDE28 (6.1 ± 2.1%), BDE153 (4.1 ± 2.3%), and BDE99 (2.4 ± 2.2%). In the stomach, the profile of PBDEs was dominated by BDE183, accounting for 27.7 ± 27.5%, which is largely different from those in the extra-hepatic tissues. While such a different pattern observed in stomach could be attributed to the molecule size of BDE183, which makes it difficult to pass through the stomach wall or intestines into the bloodstream and further to other tissues (*38*), an exposure experiment with Juvenile lake trout (*Salvelinus namaycush*) showed that the assimilation efficiency of BDE183 (22.8%) was similar to those of BDE153 (33.3%), BDE 154 (47.0%), BDE47(21.8%), BDE99(31.3%), and BDE100 (41.9%) (*39*). Thus, the most likely explanation for the concentration of BDE 183 observed in other organs would be biotransformation. To better understand the potential biotransformation of BDE183 in Chinese sturgeon, in vitro studies of BDE183, BDE154, and BDE99 were conducted in microsomal fractions of Chinese sturgeon liver (Supporting Information Table S4).

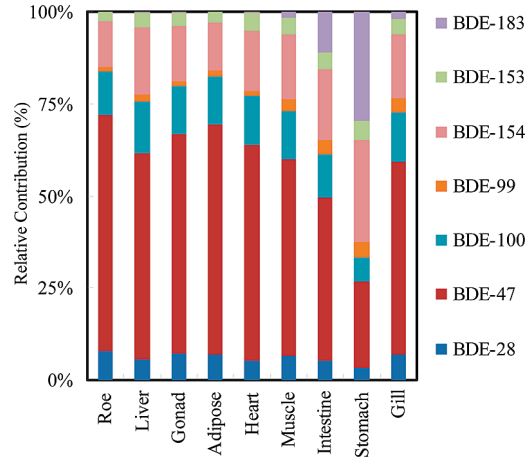

FIGURE 2. Relative contribution (%) of seven major PBDE congeners (% congener contribution on a mass per mass basis) in tissues of Chinese sturgeon.

After a 20-h exposure, 94 ± 1% of BDE183 and 78 ± 1% of BDE99 were biotransformed while concentrations of BDE154 were comparable to the starting concentrations indicating that BDE183 is readily metabolized in Chinese sturgeon. A significant amount of BDE154 was formed by conversion of BDE183 (73 ± 1.2%), while BDE47 was formed from BDE99 (52 ± 2%). This result demonstrated that part of the BDE99 and BDE183 were transformed to unquantified products or becoming unextractable. This result is similar to those observed when common carp (*Cyprinus carpio*) were exposed to BDE99 and BDE183 (*40*). Thus, it is likely that biotransformation was responsible for the relatively large BDE183/154 and BDE99/47 ratios in stomach and intestine relative to other tissues (Supporting Information and Figure 2). Furthermore, application of these two ratios can be used to estimate the extent of biotransformation in other tissues.

Of the 11 MeO-PBDEs congeners monitored, 4-MeO-BDE17, 2′-MeO-BDE68, 6-MeO-BDE47, 5-MeO-BDE47, and 5′-MeO-BDE100 were detected in eggs and liver, but their concentrations were lower than the detection limit in stomach, pancreas, and spleen. 6-MeO-BDE47 was the predominant congener, followed by 2′-MeO-BDE68 (Supporting Information Table S3). A similar pattern of relative concentrations was observed in organisms from the Canadian Arctic marine food web and pike (*Esox lucius*) from the Swedish coast (*34, 41*). Generally, MeO-PBDEs are detected in marine organisms at concentrations sometimes higher than those of PBDEs, and concentrations of OH-PBDEs are lower than those of MeO-PBDEs (*42–44*). But in the tissue samples of Chinese sturgeons, the concentration of MeO-PBDEs was 100−1000 times lower than those of PBDEs and comparable to those of OH-PBDEs. A recent study found that concentrations (31 ng/g lw) of MeO-PBDEs in the anadromous anchovy (*Coilia nasus*) from the Yangtze River estuary were higher than those (9.1 and 14 ng/g lw) farther upstream in the Yangtze River near the city of Nanjing (*45*). The Chinese sturgeon is an anadromous fish in the Yangtze River Basin. Chinese sturgeon were collected from their spawning habitat in the middle-stem of the Yangtze River. The fact that Chinese sturgeon may have been in the river for as much as a year may have resulted in the lower concentrations of OH-PBDE and MeO-PBDEs.

Of the 9 OH-PBDEs monitored, 2′-OH-BDE68, 6-OH-BDE47, 5-OH-BDE47, and 4-OH-BDE49 were detected (Supporting Information Table S3). 6-OH-BDE47 was the predominant compound in all tissues. This result is similar to the pattern observed in blood plasma of bottlenose dolphin from Indian River Lagoon, Florida and fish collected from the Detroit River and Hudson Bay region of northeastern

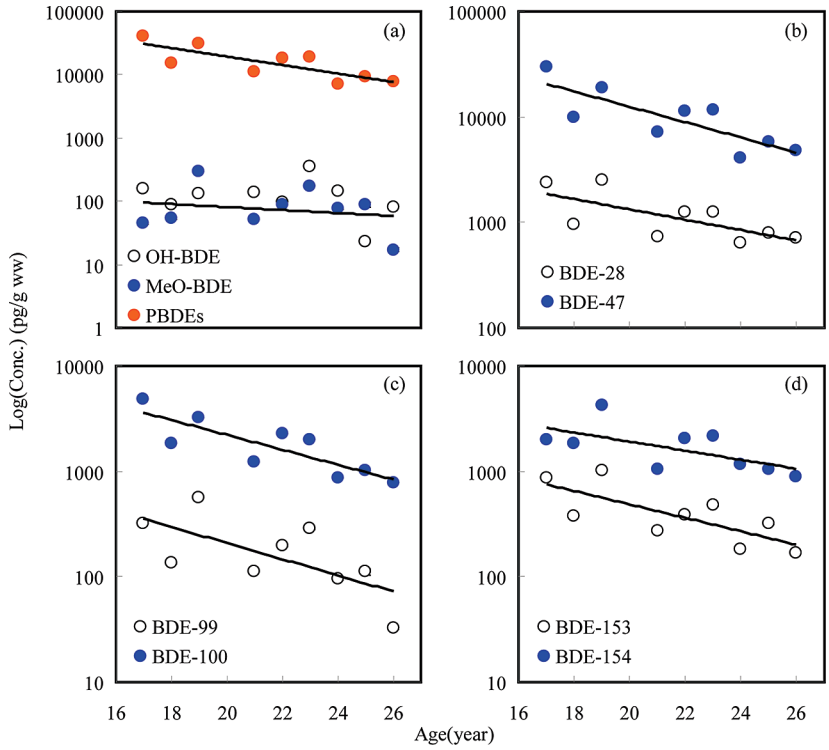

FIGURE 3. Correlations among concentrations (pg/g ww) of PBDEs, OH-PBDEs, and MeO-PBDEs in eggs as a function of age.

Canada (*21, 34, 43*). Relatively high 6-OH-BDE47 concentrations and comparable 2′-OH-BDE68 concentrations were detected in eggs. 5-OH-BDE47 and 4-OH-BDE49 with higher frequencies of detection were observed in liver (Supporting Information Table S3). It was reported that OH-PBDEs with a hydroxyl moiety at the *meta* or *para* position have been hypothesized to be biotransformation products of PBDEs based on the results of in vivo exposures while OH-PBDEs of natural origin all have a hydroxyl group at the *ortho* position (*14, 18*). Since the hydroxyl groups in 5-OH-BDE47 and 4-OH-BDE49 are at the *meta* and *para* positions, respectively, they likely would originate from biotransformation of PBDEs in liver rather than from accumulation of natural products (*15*). 2′-OH-BDE68 and 6-OH-BDE47 which have a hydroxyl group at the *ortho* position would be derived from natural products. There was a significant correlation between concentrations of 6-OH-BDE47 and 6-MeO-BDE47 ($R^2 = 0.534$, $p = 0.003$) (Supporting Information Figure S3) in the eggs of Chinese sturgeon. Such correlation was also observed in albatross (*Thalassarche chlororhynchos, Phoebetria palpebrata, Thalassarche chrysostoma, Thalassarche cauta,* and *Thalassarche melanophrys*), and polar bear (*Ursus maritimus*) (*15*). This result suggested that 6-OH-BDE47 would be from metabolic demethoxylation of MeO-PBDEs as demonstrated by a recent study based on in vitro metabolism using rainbow trout (*Oncorhynchus mykiss*), chicken (*Gallus gallus*), and rat (*Rattus norvegicus*) microsomes (*15*). On the other hand, 6-OH-BDE47 has also been observed in rats exposed to BDE47 as metabolite (*18*). To further understand the origin of 6-OH-BDE47 in Chinese sturgeon, in vitro metabolism of 6-MeO-BDE47 and various PBDE individual congeners by microsomal fractions of Chinese sturgeon was conducted (Table S4 in Supporting Information). Concentrations of 6-OH-BDE47 were lower than the MDL when dosing with PBDEs and 6-MeO-BDE47 at concentration of 150 ng/mL. Considering that 6-OH-BDE47 and 6-MeO-BDE47 have been reported in the marine environment (from formation in algae/sponges) and Chinese sturgeons spend most of their life in the sea, a large proportion of 6-OH-BDE47 in Chinese sturgeons would result from direct bioaccumulation of OH-PBDEs from natural sources.

Liver/Adipose Ratios. Ratios of lipid-normalized concentrations in liver and adipose (liver/adipose) have been used as in some physiologically based pharmacokinetic (PBPK) models (*46*). The liver/adipose ratios based on lipid normalized concentrations were 1.1 ± 1.0 for MeO-PBDEs and 2.6 ± 1.7 for PBDEs, indicating preferential deposition of PBDEs in liver. This result is similar to the hepatic sequestration of PCDD/Fs that has been observed in birds (*47*). Hepatic sequestration of PCDD/Fs is due to binding to proteins such as cytochrome P450 (CYP) enzymes (*48, 49*) via the aromatic hydrocarbon receptor (AhR) mediated pathway. A statistically significant correlation (Supporting Information Figure S4) was also observed between liver/adipose ratios of individual PBDEs and their reported AhR binding affinities ($r^2 = 0.934$, $p < 0.001$), suggesting that AhR might be involved in the hepatic sequestrations. However, PBDEs are relatively weak AhR agonists relative to PCDD/Fs or *nonortho*, 2,3,7,8-substituted, PCB (*50, 51*), the actual mechanism(s) responsible for hepatic sequestrations of PBDEs should be further studied.

Maternal Transfer of PBDEs to Eggs. Hydrophobic chemicals can be transferred from the female to eggs along with yolk proteins formed in the liver of the female parent (*25*). Ratios of lipid-normalized concentrations of PBDEs and MeO-PBDEs egg to those in liver (E/L) were used to assess maternal transfer in Chinese sturgeon. Since lipid is not a major factor affecting concentrations of OH-PBDEs in different tissues, wet weight concentrations were used to calculate egg to liver ratios. The E/L ratios of PBDEs ranged from 0.10 ± 0.11 for BDE153 to 0.22 ± 0.26 for BDE28. Ratios of 6-OH-BDE47 (0.59 ± 0.51), 6-MeO-BDE47 (0.57 ± 0.60), and 2′-MeO-BDE68 (0.65 ± 0.85) were higher. Previous investigations of maternal transfer of organochlorines in Chinese sturgeon (*30*) provide an opportunity to compare the E/L ratios of the polybrominated compounds with other chemicals. The E/L ratios of PBDEs were low than those of *p,p*-DDD (0.27 ± 0.15), *p,p*-DDE (0.30 ± 0.19), and HCB (0.98

± 0.46), but those of 6-OH-BDE47, 6-MeO-BDE47, and 2′-MeO-BDE68 were higher than the E/L ratios of DDTs. It was found that the more PBDEs were hepatic sequestrated in the liver, the lower the ratio of E/L, indicating that hepatic sequestration of PBDEs would affect their maternal transfer in Chinese sturgeon.

Maternal transfer can influence accumulation of pollutants in females, and generally higher transfer ratios will result in lower accumulation by females than males (*52*). While there is no correlation between age and concentrations of OH-PBDEs or MeO-PBDEs, the concentration of ΣPBDEs significantly decreased with age (Figure 3a). As shown in Figure 3b, c, and d the lipid normalized concentrations of six major PBDEs congeners (BDE28, BDE47, BDE99, BDE100, BDE153, and BDE154) also show a negative trend with age of female Chinese sturgeon ($p < 0.05$ except for BDE99 and BDE154, Supporting Information Table S5). This result is different from those for DDE and HCB which were positively correlated with age of Chinese sturgeon (*30*). A similar negative correlation between concentrations of PBDEs with age was observed in tissues of both male and female Harbour seals (*Phocoena phocoena*) (*53*). Since maternal transfer ratios (E/L) of HCB and DDE were higher than those of PBDEs, maternal transfer would not be the primary factor resulting in accumulation of PBDEs in Chinese sturgeon. The higher metabolism of PBDEs such as BDE99 and 183 is a more likely factor. In addition, rapid excretion could be another potential factor since higher concentrations were observed in gall bladder as discussed above.

Acknowledgments

Financial support for this study was obtained from the National Basic Research Program of China (2007CB407304) and the National Natural Science Foundation of China (40632009). We thank Qiwei Wei, Yangtze River Fisheries Research Institute, Chinese Academy of Fisheries Science, Ministry of Agriculture of China, for providing samples of eggs and tissues from Chinese sturgeon. Portions of this research were supported by a Discovery Grant from the National Science and Engineering Research Council of Canada (Project 326415-07) and a grant from Western Economic Diversification Canada (Projects 6578 and 6807). J.P.G. was supported by the Canada Research Chair program and an at large Chair Professorship at the Department of Biology and Chemistry and State Key Laboratory in Marine Pollution, City University of Hong Kong.

Supporting Information Available

Text, figures, and tables addressing (1) chemicals and standards used in the analysis; (2) extraction and cleanup of tissues; (3) instrumental conditions; (4) details of Chinese sturgeon samples; (5) mean concentrations and ranges of OH-PBDEs, MeO-PBDEs, and PBDEs in tissues of Chinese sturgeon; (6) percentages of brominated compounds relative to the dosing concentration after metabolism with Chinese sturgeon microsomes exposed to PBDEs and 6-MeO-BDE47; (7) association of concentrations of PBDE congeners with age of Chinese sturgeon; (8) levels of MeO-PBDEs and ΣPBDEs in various tissues in Chinese sturgeon after lipid normalization; (9) ratios of concentrations of BDE99 to BDE47 (BDE99/47) and BDE183 to BDE154 (BDE183/154) in Chinese sturgeon tissues; (10) relationships between the concentrations of 6-OH-BDE47 and the concentration of 6-MeO-BDE47; (11) ratios of lipid-normalized ratio of concentration in liver to that in adipose of Chinese sturgeon plotted versus Ah receptor binding affinities. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES100348G