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Effect of Municipal Sewage Treatment Plant Effluent on Bioaccumulation of Polychlorinated Biphenyls and Polybrominated Diphenyl Ethers in the Recipient Water

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Water, sediment, and aquatic species including plankton, fish, and turtles were collected from a small lake in Beijing, which receives effluent discharged from a large sewage treatment plant (STP). The samples were examined to investigate polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) releases from a STP and their distributions in the lake. The accumulations of $\sum_{12}$-PBDEs and BDE-209 in the sediment were 62.3 and 1150 ng/cm^2, respectively, while that of ∑PCBs was 99.3 ng/cm^2. BDE-209 was detected in more than 50% of the aquatic species. A strong linear correlation ($R^2 = 0.92$) was found between $\sum_{12}$PBDEs and ∑PCBs levels in aquatic species but not in sediments. The different PBDE congener profiles in sediments and biota samples suggest metabolic debromination in the sampled fish. Bioaccumulations of PBDEs and PCBs were found in aquatic species. The logarithm bioaccumulation factor (BAF) decreases with the number of bromines in PBDEs molecules, while the log BAF versus the number of chlorines in PCBs appears to be parabolic. Biomagnifications of these compounds were not obvious in the food web by analysis of the relationship between $\sum_{12}$PBDEs or ∑PCBs levels and the trophic level of aquatic biota species.

Introduction

Municipal sewage treatment plants (STPs) efficiently remove large proportions of contaminants, such as nutrients and biodegradable organics (*1*). However, for persistent organic pollutants (POPs), such as polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs), STPs can be a potential point source of discharge to the ambient environment via their effluents and sewage sludge disposal (*2*−*6*). North et al. (*7*) tracked the releases of PBDEs in a wastewater treatment plant and found that 4% of the PBDEs that entered the plant was discharged to the estuary through the effluent. Another study estimated that up to 9% of the influent mass of the total of PBDEs was discharged with the final effluent (*8*). The effluents and reclaimed wastewater could be a significant source of PBDE and PCB contamination to recipient water (*9*) and the atmosphere (*10*).

As a class of emerging chemicals, PBDEs have been found throughout the environment (*11*). A previous work that tracked the PBDEs releases in wastewater treatment plant effluent found that PBDE concentrations in discharged effluent ranged from 4 to 20000 pg/L (*7*). An investigation concerning PBDEs in the sewage sludge of wastewater treatment plants in China showed that the concentrations ranged from 6.2 to 57 ng/g (dry weight, excluding BDE-209) (*12*). In heavily polluted areas such as electronic waste recycling sites, the concentrations of PBDEs in river bank sediments were found to reach 16088 ng/g (dry weight) (*13*).

Although PCBs have been banned for many years, their widespread occurrence is still of grave concern. When these pollutants enter aquatic systems, they tend to accumulate in aquatic organisms due to their hydrophobicity and resistance to biodegradation (*14*). In addition, they can biomagnify along the food chains and pose a threat to the health of the ecosystem and human body. The mechanisms and the influencing factors of the bioaccumulation and biomagnification of PBDEs and PCBs have been investigated in a number of studies (*15*−*17*).

In China, about 400 STPs have so far been built. Currently, there are eight STPs in Beijing, and the construction of additional seven new ones is in progress. The wastewater treatment capacity and reclamation rate in Beijing should increase by up to 90 and 50%, respectively, in 2008 before the Olympics Games begins.

Most research on the health effects of STP products in China place an emphasis on toxic metals, while the risks associated with persistent organic pollutants have only been investigated in a limited number of studies (*12*, *18*). In the present work, we carried out a detailed examination of PBDEs and PCBs pollution in Gaobeidian Lake located in the eastern part of Beijing. The STP effluent, lake water, sediment cores, plankton, fish, and turtle samples were collected and analyzed for PBDEs and PCBs in order to evaluate the effect of discharging municipal sewage treatment plant effluent on the bioaccumulation and biomagnifications of PCBs and PBDEs in the recipient water.

Experimental Section

Sampling. Gaobeidian STP processes approximately 1 million tonnes of wastewater per day, of which about 80% is from municipal sources. The STP discharges 30% of its effluent into the lake. The effluent is also used as cooling water for a nearby heat power plant, and its backwater is again discharged into the lake. Therefore, the temperature of this lake water remains higher than those of other local lakes (The water temperature was constantly above 30 °C from May to October and the highest temperature in 2006 was 45 °C).

Figure S1 and S2 in the Supporting Information (SI) show the sampling map and the process flow chart of Gaobeidian STP, respectively, with additional information. Effluent and lake water samples were collected in September and December, 2006. A part of the aquatic food web, sampled in September, 2006 involves primary producers spirogyra and

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March brown (*Limnodrilus hoffmeisteri*), coccid, zooplankton (*Monia rectirostris*, *Monia micrura,* and *Monia macrocopa* as the dominant species), four fish species including common carp (*Cyprinus carpio*), crucian carp (*Carassius auratus*), leather catfish (*Silurus meridionalis*) and java tilapia (*Tilapia nilotica*), and Chinese softshell turtle (*Chinemys reevesii*). In addition, one sediment core was taken from the upstream (core 1) and another from the downstream (core 2) of the STP outfall.

Sample Extraction and Analysis. Chemical analyses followed our previously established method with some modifications (*19*). The samples were analyzed for 13 PBDE congeners (BDEs 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, and 209) and 25 PCB congeners including 12 coplanar congeners (CBs 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189), six indicator congeners (CBs 28, 52, 101, 138, 153, and 180), and other congeners (CBs 3, 15, 19, 202, 205, 208, and 209). The analytical results were obtained based on an isotope dilution method using high-resolution gas chromatography−high-resolution mass spectrometry . Detailed procedures of samples pretreatment, instrumental analysis, and quality assurance and quality control are given in the online SI. The total organic carbon (TOC) of sewage sludge was analyzed by the $KMnO_4$ titration method (*20*).

Results and Discussion

Sediment TOC averaged 12% for core 1 and 10% for core 2 (Table S1, SI). Table S2 (SI) shows the basic information of the fish species. The concentrations and congener profiles of PBDEs and PCBs are illustrated in Figures 1 and 2 for sediment and aquatic species, respectively. Table S3 (SI) summarizes the concentrations of PBDEs and PCBs in water, effluent, plankton, and aquatic species in Gaobeidian Lake.

PBDE and PCB Levels in Lake Water and Effluent. The sum of BDEs 47, 99, 100, and 153 accounted for 80 and 60% of $\sum_{12}$PBDEs (the sum of all measured PBDEs except for BDE-209) in effluent and surface water, respectively. These major congeners of the pentaformulation have been found to bioaccumulate in aquatic species (*21*). The concentration of $\sum_{12}$PBDEs in effluent is far below that found in the STP effluent of Palo Alto, CA (29.02 ng/L) (*7*). That BDE-209 was below the limit of detection in effluent and water was probably due to the hydrophobicity of this bulky congener and small sample volume. Most BDE-209 would likely partition into sludge during wastewater processing (*8*).

PCBs concentrations in the effluents were comparable to PBDEs, but the concentrations in lake water were lower than those of PBDEs. The main congeners of PCBs in the aqueous samples were lowly chlorinated CBs such as CB 15, 28, etc. The indicator PCBs including CBs 28, 52, 101, 138, 153, and 180 account for 74 and 66% of ∑PCBs in effluent and lake water, respectively. The special characteristics of this lake water can disturb the water−air and sediment−water exchange of these contaminants. The higher water temperature of this lake may lead to an increased water−air exchange rate and reduced sorptions on suspended particles and sediment. On the other hand, dissolved organic materials from the effluent of STP may also have an impact on these phase-partitioning processes.

PBDE and PCB Levels in Sediment Cores. Concentrations of $\sum_{12}$PBDEs and BDE-209 in the sediment cores ranged from 3.74 to 10.98 ng/g dry weight (dw) (average 6.33 ng/g) and from under the limit of detection (LOD) to 742.53 ng/g dw (average 237.01 ng/g), respectively. On the basis of organic carbon (OC) levels , $\sum_{12}$PBDEs and BDE-209 ranged from 23.47 to 108.0 ng/g OC and from under LOD to 6366 ng/g OC, respectively. Among the targeted congeners, BDE-209 was found to be the most dominant (Figure 1a), as it was present at much higher concentrations than other congeners. This finding corresponds to the fact that most of the PBDEs produced in China are of the deca-BDE formula. Other main congeners in the sediment cores were tetra- and penta-BDEs, and these two homologues accounted for 33.7 and 29.2% of $\sum_{12}$PBDEs by average (Figure 1b).

∑PCBs concentrations in the sediment cores averaged 10.00 ng/g dw (Figure 1c) and ranged from 3.11 to 18.05 ng/g dw or 29.78 to 158.7 ng/g OC. Toxic equivalents (TEQ) values calculated using WHO toxicity equivalent factors ranged from 0.46 to 3.85 pg/g with an average level of 1.65 pg/g. The concentrations of indicator congeners accounted for 67% of ∑PCBs, and there were significant linear relationships between indicator congeners and ∑PCBs ($R^2 = 0.912$, $P < 0.05$) and between coplanar congeners and ∑PCBs ($R^2 = 0.743$, $P < 0.05$). No linear relationships were found between $\sum_{12}$-PBDEs, BDE-209, or ∑PCBs and OC, indicating that PBDE and PCB loadings to the sediments were independent of the sediment organic content. Regression analysis also suggested that there was no correlation between PBDEs and PCBs concentrations in the sediment of Gaobeidian Lake.

The upper layers (0−12 cm) had higher concentrations of $\sum_{12}$PBDEs and BDE-209 than the deeper layers, reflecting the increasing production and use of commercial PBDEs in recent years in Beijing. In addition, the wastewater processed at Gaobeidian STP has doubled in recent years from its pre-1999 capacity of 50 000 tonnes per day. This may have resulted in increased PBDEs released into the Gaobeidian Lake.

The maximum concentrations of PCBs appeared in the deeper layers, and the highest TEQ of PCBs was found in the depth of 24−32 cm. This profile parallels the decline in the production and use of technical PCBs in China, in agreement with several previous findings regarding the trend of PCB residuals in the aquatic ecosystem in the past 15−20 years (*22*, *23*).

The inventory of pollutant chemicals in the sediments is acquired by calculating the accumulative mass per unit area and is estimated by $\sum C_i \rho_b d_i$, where C_i is the concentration in sediment segment i (ng/g, dw), ρ_b is the dry mass bulk density (g/cm^3), and d_i is the thickness of segment i. The inventories of $\sum_{12}$PBDEs, BDE-209, and ∑PCBs were 62.3, 1150, and 99.3 ng/cm^2, respectively in Gaobeidian Lake. The inventories of $\sum_{12}$PBDEs and BDE-209 were about 10−100 times higher than those at far-from-shore locations in the Great Lakes (*24*−*26*), whereas the PCB inventories were higher than those in Lakes Michigan and Huron (6.21−48.1 ng/cm^2), but lower than those in Lakes Erie and Ontario (169.2−262.9 ng/cm^2). Eganhouse et al. (*27*) found that municipal wastewater effluent exported substantial amount of ∑PCBs to the Palos Verdes Shelf, CA, where the inventories in sediment were as high as 25000−39000 ng/cm^2. The PCB concentrations in cores 1 and 2 in Gaobeidian Lake were at the low end of the worldwide PCB concentration range (0.2−400 ng/g) (*28*).

PBDEs and PCBs Levels in Aquatic Species. Parts a and b of Figure 2 show PBDE congener distributions in aquatic species. Very high concentrations of BDE-209 were found in spirogyra, March brown, coccid, and zooplankton. Especially, the BDE-209 concentration in spirogyra was as high as 1572 ng/g. Other main congeners found in these samples were BDEs 47, 99, 153, and 183. Higher brominated congeners (number of Br >5) account for more than half of the total concentrations ($\sum_{12}$PBDEs + BDE-209).

BDE-209 is only occasionally found in living organisms due to the large molecular size, which decreases its bioavailability, and/or its relatively rapid biotransformation in organisms. In this work, however, BDE-209 was detected in more than 50% of the aquatic species. The relative abundance of BDE-209 in the fish and Chinese softshell turtle is lower than that in spirogyra, March brown, coccid, and zooplankton. Mean concentrations of $\sum_{12}$PBDEs were in the following ranking order: crucian carp > leather catfish > common carp > java tilapia > Chinese softshell turtle. Chinese softshell

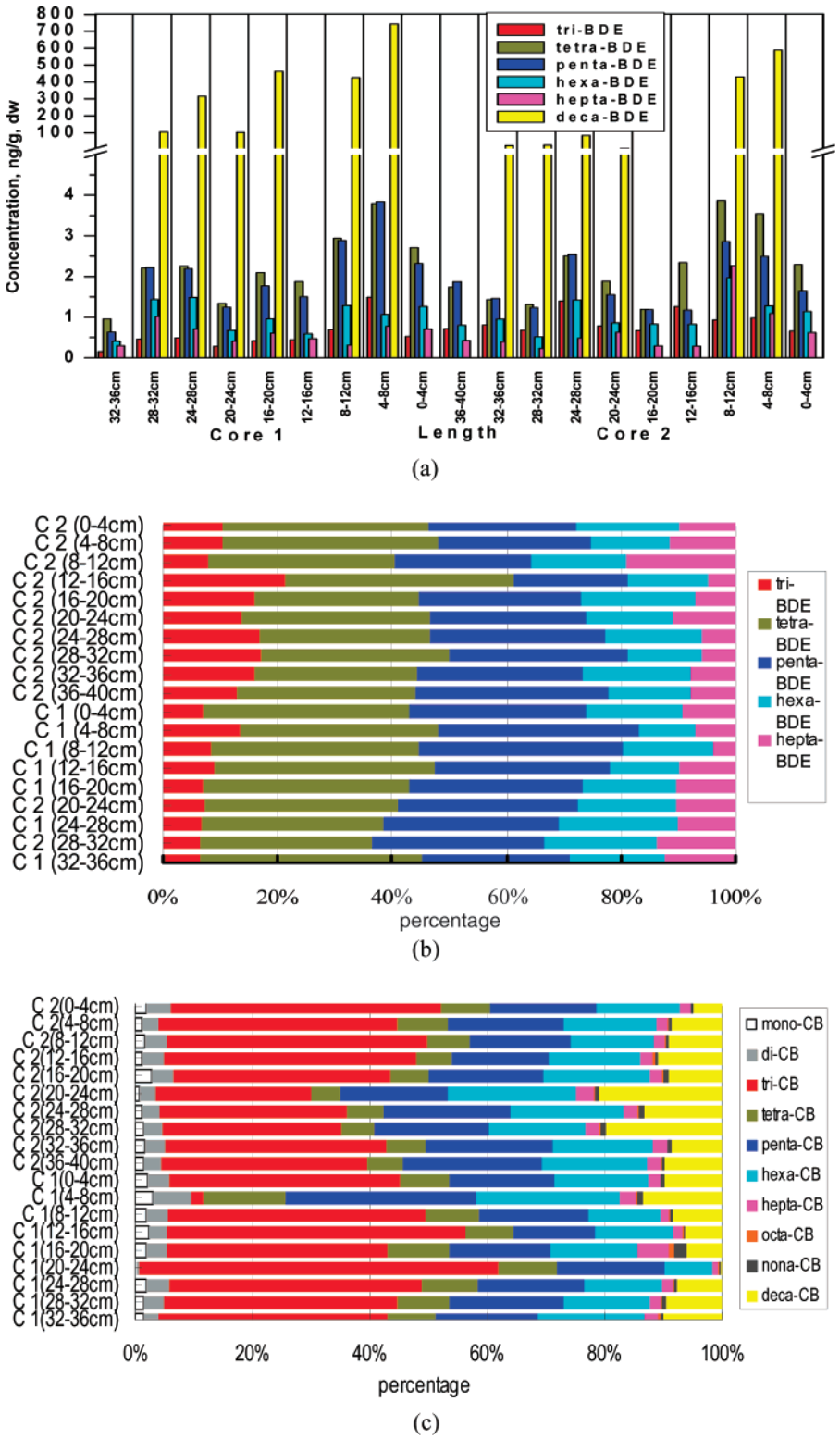

FIGURE 1. Concentrations of $\sum_{12}$PBDEs and BDE-209 in sediments (a); composition profiles of $\sum_{12}$PBDEs (except for BDE-209) in sediments (b); and composition profiles of $\sum$PCBs in sediment core 1 (C1) and core 2 (C2) (c).

turtle have the lowest concentrations of $\sum_{12}$PBDEs although they were greater in age than the other sampled species. The concentrations of lower brominated congeners were higher than those of higher brominated congeners. The linear relationship found between $\sum_{12}$PBDEs and lipid contents ($R^2 = 0.31$, $P < 0.05$) implied that the $\sum_{12}$PBDEs concentrations were dependent on the lipid contents of these aquatic species. However, no linear correlation was found between BDE-209 and lipid contents.

In fish and Chinese softshell turtle, the fraction of tetra-BDEs in $\sum_{12}$PBDEs was up to 61%, whereas that for penta-BDEs was only at 9%. A multichemical food web model suggests that additional an input of BDE-47 in fish may be derived from the debromination of higher brominated congeners. One biotransformation pathway was the debromination of BDE-100 to BDE-47 and the other was the debromination of BDE-153 to BDE-99 and then to BDE-47 (*29*). The higher percentage of tetra-BDEs and lower per-

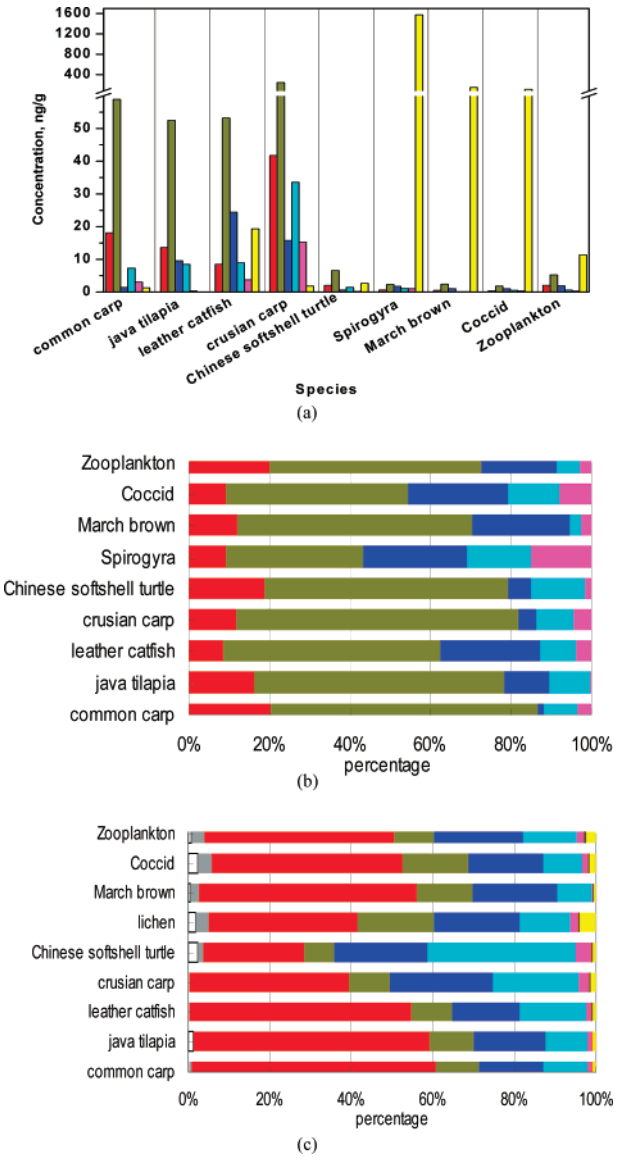

FIGURE 2. Concentrations of PBDEs (a), composition profiles in aquatic species (except for BDE-209) (b), and composition profiles of PCBs (c), found in aquatic species in Gaobeidian Lake. The color codes for homologues are the same as those used in Figure 1.

centage of penta-BDEs in fish compared with those in sediments found in this study seemed to confirm these biotransformation pathways in fish.

In fish species and Chinese softshell turtle, ∑PCBs concentrations were in the same order of magnitude as $\sum_{12}$-PBDEs, with tri-, tetra-, penta-, and hexa-CB homologues as the most abundant congeners. The contribution of indicator congeners in these five aquatic species was 87.9% for common carp, 82.0% for java tilapia, 87.1% for leather catfish, 83.2% for crucian carp, and 71.4% for Chinese softshell turtle, respectively (Figure 2c). A very strong correlation was found between indicator congeners and ∑PCBs in all aquatic species (R^2 is up to 0.998, $P < 0.0001$). There is also a linear relationship between coplanar congeners and ∑PCBs ($R^2 = 0.95$). Trichlorobiphenyls accounted for 38% of ∑PCBs in sediment and 48% in fish and Chinese softshell turtle. The lipid-content dependence of ∑PCBs in fish is, however, relatively weak ($R^2 = 0.38$), although statistically significant ($P < 0.05$). The ∑PCBs concentrations in fish are comparable to the data from other areas in the world despite the fact that the PCB inventories in this lake were at the low end of the world range (*30*). Contrary to the results of sediment analysis, a very strong

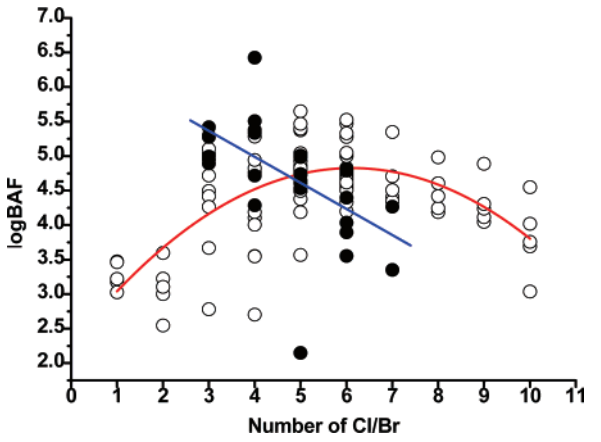

FIGURE 3. Dependence of BAFs on the number of halogen atoms in the PBDE and PCB molecules. Key: (○) PCB; (●) PBDE; (red line) log BAF = 2.28 + 0.83N_{Cl} − 0.07N_{Cl}^2, $R^2 = 0.457$, $P < 0.05$; (blue line) log BAF = 6.50 − 0.38N_{Br}, $R^2 = 0.439$, $P < 0.05$.

linear correlation was found between ∑PCBs and $\sum_{12}$PBDEs ($R^2 = 0.92$, $P < 0.001$) in fish.

Bioaccumulation of PBDEs and PCBs in Food Web. Chemicals are considered to be bioaccumulated if bioaccumulative factors (BAF, which is the ratio of the chemical concentration in biota to the concentration in corresponding lake water) are greater than 5000 in aquatic organisms (*31*). In our study, we found that most of the PBDEs and PCBs congeners analyzed were bioaccumulated in the selected fish species except for CB-3 and CB-15, which are not among the major congeners.

For PBDEs, a significant decrease of BAF with increasing number of bromines was observed ($R^2 = 0.439$, $P < 0.05$) (Figure 3). This is most likely due to the high molecular weights and sizes (which make dietary uptake inefficient in fish) and the debromination and elimination of these compounds in organisms (*21*). The BAFs for lowly brominated diphenyl ethers were found to be higher than those of lowly chlorinated biphenyls. Another study also reported higher bioaccumulation of BDEs 47 and 99 in blue mussels (*Mytilus edulis*) than those of PCBs (except for BDE-153) (*32*).

For PCBs, the correlation between BAF and the number of chlorines was parabolic ($R^2 = 0.439$, $P < 0.05$), as shown in Figure 3. A similar phenomenon has been reported by Danis et al. (*33*). They exposed sea stars to 10 selected PCB congeners. After 30 days, they found that the sea stars concentrated more of hexa-CBs (such as PCBs 138 and 153) than both lower (e.g., PCBs 28 and 52) and higher (PCBs 160 and 180) congeners. The BAFs obtained in this work are shown in Figure S3 (SI) as a function of K_{ow}. Apparently, the relationships between BAF and K_{ow} correspond to those of BAF and the number of chlorines, since there is a significant linear relationship between K_{ow} and the number of chlorines. In Figure 3, the inverse trends when the chlorine number exceeds six may plausibly be explained by the increasing difficulty in the ability of large molecules to migrate across biomembranes and/or their relatively fast metabolic degradation in organisms compared with that of the lower PCB congeners.

Congener specific bioaccumulation of PBDE is much more difficult to assess compared with that of the PCBs. Biotransformation of PBDEs has been proven to occur in fish, which derives from contrasting the half-lives of BDE congeners with a similar K_{ow} value (*21*). The debromination of highly brominated congeners during depuration can also confound their bioaccumulative behavior in fish. Significant debromination has been found in the common carp by independent dietary exposure experiments, converting BDE-99 to BDE-47 and BDE-183 to BDE-154 (*34*). In this study, it was

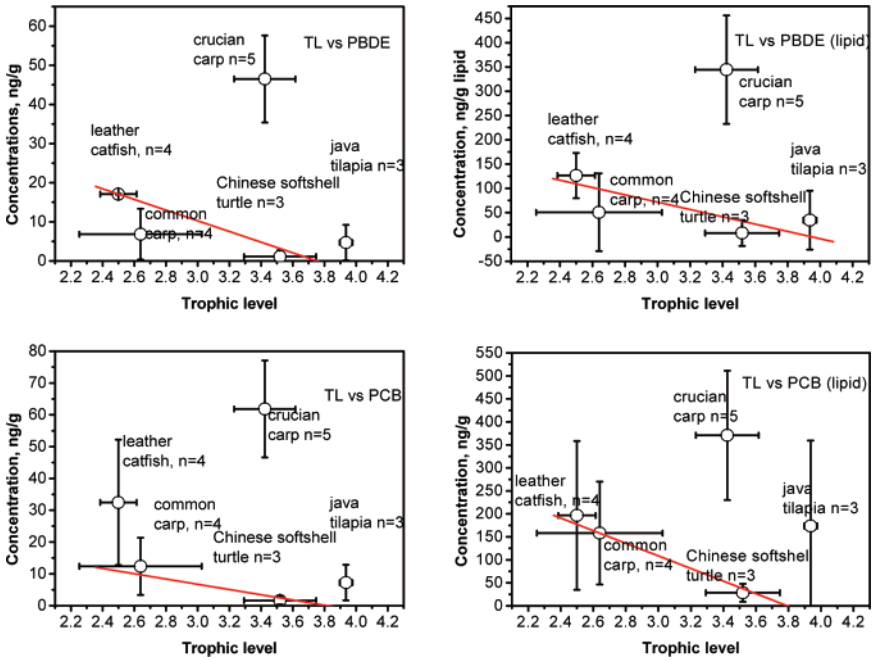

FIGURE 4. Relationships of TL and concentrations of $\sum_{12}$PBDEs and $\sum$PCBs.

found that the fractions of BDE-99 in $\sum_{12}$PBDEs in aquatic species were lower than other BDE congeners, supporting the findings of Stapleton et al. (*34*). Negative correlations were observed between log BAF and log K_{ow} for PBDEs, especially in Java tilapia ($R = -0.701$, $P < 0.05$), in contrast to that found for PCBs. This may be the result of faster increases in the degradation rate of PBDEs than that of PCBs when the number of halogen atoms increases. Elevated water temperature of this lake may also influence the biotransformation and depuration rate of PBDEs and PCBs in aquatic species.

This complex interplay of processes plays a key role in the bioaccumulation of contaminants in organisms, such as gastrointestinal magnification, and its counteraction including growth dilution, respiratory elimination, etc. Consequently, factors such as body length, body weight, age, sex, and environmental temperature can also influence the bioaccumulation of these chemicals in fish. Any attempt to search for only one or two influencing factors is insufficient. The concentrations of PBDEs and PCBs in this work varied among different species. For common carp, $\sum_{12}$PBDEs and $\sum$PCBs were negatively correlated with the body length or weight ($P < 0.05$), while for other species (except for Chinese softshell turtle, for which the correlations between the concentrations and weight or length could not be analyzed), $\sum_{12}$PBDEs and $\sum$PCBs concentrations slightly increased with the increase in weight or length of the fish. It seems that growth dilution plays a key role in common carp since the elimination rate of these compounds in aquatic organisms is inversely related to body size (*35*).

Nevertheless, the influence of other environmental factors cannot be ignored. As the main water source of the lake, the effluent of the STP pours plenty of suspended particles into the lake. Previous studies have convincingly demonstrated that sorbing material can reduce bioavailability and uptake of chemicals by aquatic organisms (*36*) and the extent of sorption increases with increasing K_{ow} (*37*). Therefore, for the highly halogenated congeners, the bioavailability and bioaccumulation should be much more influenced than the lowly halogenated congeners in this suspended particles–water mixture.

Biomagnification of PBDEs and PCBs in Food Web. Several studies indicated that PBDEs and PCBs can biomagnify in aquatic organisms through the food chain, i.e., higher chemical concentrations can be found in an organism with a higher trophic level (TL) than that of its prey (*38*).

To determine if the biomagnification of PBDEs and PCBs occurs in the food webs of the Gaobeidian Lake, TLs of the selected aquatic organisms were measured based on nitrogen isotope ratios, as described by Fisk et al. (*39*): $TL_{consumer} = (\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton})/3.8 + 2$. The results presented some peculiar TL distributions. Java tilapia had the highest TL, while common carp had the widest range δ^{15}N (4.29–7.94) despite their similar length and age (all about 3 years old). The investigation presumed that δ^{15}N can be influenced by municipal waste sources with individuals differing by up to 0.3% in δ^{15}N (*40*). As such, the abnormal TL of these aquatic species may also be influenced by the effluent sources from the STP in addition to their food habits, metabolism, and other factors.

Biomagnification of PBDEs and PCBs via the food web is often considered to occur in aquatic ecosystems. In this study, however, no obvious biomagnifications of PBDEs and PCBs in the selected food web could be found by analyzing the relationship between $\sum_{12}$PBDEs or $\sum$PCBs and TL (Figure 4). Similar negative trends of $\sum_{12}$PBDEs and $\sum$PCBs vs TL were found. Significantly higher $\sum_{12}$PBDEs and $\sum$PCBs concentrations ($P < 0.001$) existed in crucian carp than in other species despite their trophic position. Crucian carp, the smallest in size but the oldest in age, accumulated more $\sum_{12}$PBDEs and $\sum$PCBs than other fish species.

The biomagnification factor (BMF) of halogenated organic compounds has been studied in fish as well as other biota (*38*). Various factors including biotransformation, bioformation, depuration rate, and even extrinsic conditions such as water temperature were discovered to confound efforts to measure the BMF. Especially for PBDEs, it is difficult to measure the BMF due to the rapid elimination of higher brominated BDEs via debromination and depuration, compared with chlorinated contaminants such as PCBs (*37*). For example, the concentrations of BDE-209 as well as $\sum_{12}$PBDEs in crucian carp and leather catfish are higher than those in other species despite their TL position. On the other hand, the fluctuation of the water temperature can also present challenges to assess the food-web dynamics of these chemicals (*26*). The similarity in chemical structures between PBDEs

and PCBs hints at the possibility that they might share similar environmental effects. However, more factors seem to confound the investigation of the bioaccumulation and biomagnification of PBDEs than that of the PCBs. It is likely that the biotransformation of the PBDEs in fish is more prevalent than that of the PCBs, because the Br−C bond is weaker than the Cl−C bond. Also, structural configuration evaluation indicated that PCB families can accommodate their geometry by freely rotating the C−C bond to bind an aryl hydrocarbon receptor. While for PBDE congeners, the large atomic volume of Br distorts the binding site, which can significantly decrease the toxic effects and should influence the bioavailability in aquatic organisms. The present work supports the tenet that STP effluents are potential point sources of PBDEs and PCBs to the recipient waters. Furthermore, the results of this work showed that many factors including metabolic dehalogenation confound the investigation on the bioaccumulation and biomagnification of PBDEs and PCBs.

Acknowledgments

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Supporting Information Available

Detailed procedures of samples pretreatment, instrumental analysis, and quality assurance and quality control; sampling map and process flow chart of Gaobeidian STP in PDF format. This material is available free of charge via the Internet at http://pubs.acs.org.

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Supporting Information

Effect of Municipal Sewage Treatment Plant Effluent on Bioaccumulation of Polychlorinated biphenyls and Polybrominated Diphenyl Ethers in the Recipient Water

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The basic process flow of Gaobeidian sewage treatment plant

Gaobeidian Lake (Figure S1), which was dug during the Ming dynasty, was canalized as a main shipside in Beijing in the past. The water feeds into Beiyun River, which flows into Bohai bay, one of the largest estuaries in China. Beijing Gaobeidian STP, built in the 1990s and is the largest in Asia, directly discharges part of its effluent into Gaobeidian Lake.

The wastewater treatment processing chart is shown in Figure S2. Briefly, the wastewater is kept for 2.5 hours in a primary settling tank, and then passes through an aeration tank in 9 hours. After staying in the secondary settling tank for 4 hours, the effluent is poured into the recipient water.

Most of sewage sludge from secondary clarifier is poured into a sludge gravity thickener to concentrate for about 29 hours, except for a part of sludge that is re-poured into the aeration tank. The second stage of sludge treatment is anaerobic digestion in a primary sludge digester. This stage takes about 21 days and the sludge is then processed in a secondary sludge digester for about 7 days. The digested sludge is further concentrated and dried.

The STP has two outfalls. 30% of the effluent enters Gaobeidian Lake and the remaining is discharged into a river via the other outfall located downstream of the lake.

Materials

All solvents were pesticide residue grade and were purchased from Fisher (Hampton, NH). Silica gel was obtained from Merck (Whitehouse Station, NJ). PBDE standard mixture EO5103 was purchased from Cambridge Isotope Laboratories (Andover, MA). ^{13}C-labeled surrogate standard solutions of PCBs and PBDEs, ^{13}C-labeled injection standard solutions of PCBs (1668a-IS) were obtained from Wellington Laboratories (Guelph, Canada).

Sample collection

1 L lake water or effluent was collected by using Teflon tube. Teflon tube and containers were precleaned thoroughly with acetone prior to sampling. Then the samples were immediately carried to laboratory and stored in a 4°C refrigerator prior to analysis. Two sediment cores were taken by a special sediment core sampler. Core 1 which was collected from the upstream of the STP outfall was about 36 cm in height and the other (Core 2) was about 40 cm in height. The cores were sectioned at 4-cm intervals using a stainless steel blade. Sectioned sediment samples were packed by aluminum foil in sealed plastic bags. Planktons were collected by net with a mesh size of 5 mm and was then concentrated using centrifuge. Aquatic species were stored on ice after being collected and transported back to the laboratory. Weight, length, age and sex were measured and identified. All the planktons, sediments, and aquatic species were stored at -20°C until analysis. In all, 4 water and 4 effluent samples were collected during two sampling events. Sediment Core 1 was sectioned into 9 segments and the other was sectioned into 10 segments. One batch of spirogyra and March brown, coccid, zooplankton were collected during the sampling period. The quantity of the five collected aquatic species was: common carp (4), crucian carp (5), leather catfish (4) and java tilapia (3), and Chinese softshell turtle (3).

Sample extraction and analysis

Sediment samples and biosamples were freeze-dried and homogenized by sieving through a stainless steel 100-mesh sieve. Prior to the extraction, each sample was spiked with ^{13}C-labeled PBDEs and ^{13}C-labeled PCBs surrogate standards solution. Effluent and lake water samples (1 L) were liquid/liquid extracted in dichloromethane (DCM). The extracts were concentrated by rotary evaporation, dried with anhydrous sodium sulfate, and cleaned up on acid silica gel column. The extract were further concentrated and the final volume were reduced under purified nitrogen to about 20 μL for analysis.

For sediment samples and biosamples, different mass (1 g for sediment samples, dry weight, (dw), 2 g for biosamples, dw) was mixed with sodium sulfate and extracted on a accelerated solvent extractor (ASE) in a mixture of 1:1 (v:v) DCM: hexane. Extracts were cleaned up on automated gel permeation chromatography (GPC), multi-layer silica column, which was packed from bottom to top with 1 g activated silica, 4 g basic silica (1.2%, w/w), 1 g activated silica, 8 g acid silica (30%, w/w), 2 g activated silica, and 4 g anhydrous sodium sulfate. The final extracts were also concentrated by rotary evaporation and further reduced with gentle nitrogen flow to final volumes of 20 μL in a minivial. Prior to instrument analysis, ^{13}C-labeled internal standards 1668A-IS (5 μL) was added to the autosampler vial and was vortexed to mix completely.

Stable isotopes were analyzed using a flash EA 1112 elemental analyzer interfaced with a Thermo-Finnagan Delta^{plus} isotope ratio mass spectrometer.

Instrumental analysis

The quantification of PBDEs and PCBs was performed on an Agilent 6890 high-resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS) (Waters Micromass, Manchester, UK) using an electron impact (EI) ion source. A 1 μL split/splitless volume was injected with a CTC PAL autosampler

into a DB-5 (PBDEs: 30 m × 250 μm i.d. × 0.1 μm film thickness; PCBs: 60 m × 250 μm i.d. × 0.25 μm film thickness) capillary column. MS was operated in selective ion monitor (SIM) mode with resolution >10,000. The instrument was mass calibrated against perfluorokerosine (PFK) ions as a reference for mass lock. Helium was the carrier gas at a constant flow rate of 1.2 mL/min. For PBDEs analysis, the initial oven temperature was 100 °C, which was held for 1 min. It was increased to 230 °C at 15 °C/min, then increased to 270 °C at 5 °C/min, and finally ramped to 330 °C at 10 °C/min and held for 8 min. For PCBs analysis, the initial oven temperature was 120 °C, which was held for 1 min. It was increased to 150°C at 30°C/min, and then increased to 300°C at 2.5 °C/min and held for 1 minute.

Quantification was carried out with the internal calibration standard method. Peaks were quantified only if the signal-to-noise ratio was > 3. All PBDE congeners were monitored at the molecular ion clusters $[M]^+$ and $[M+2]^+$. For PBDEs, the method limit of detection (LOD) of tri- through hepta-BDE congeners and BDE-209 ranged from 1 to 100 pg/g, and 1 ng/g, respectively. For PCBs, the LOD ranged from 0.2 to 5 pg/g. Stable isotopes were analyzed using a Flash EA 1112 elemental analyzer interfaced with a Thermo-Finnagan Delta^{plus} isotope ratio mass spectrometer.

Quality assurance and quality control

$\sum_{12}$PBDEs refers to the sum of all targeted PBDEs excluding BDE-209. ∑PCBs represents the sum of all targeted PCBs. Strict quality controls were implemented to ensure the correct identification and accurate quantification of the analysis. All glassware was thoroughly rinsed before and after use with dichloromethane. Each batch of 12 samples included one method blank (30 g pre-cleaned sodium sulfate) to ensure the method was responding properly. The mean concentration of $\sum_{12}$PBDEs of method blank was about 0.18 ng with the dominant congeners BDEs 47 and 99. For ∑PCBs, the method blank was 0.38 ng and the dominant congener is CB-28. The recoveries of ^{13}C-labeled surrogate congeners BDEs 47, 99, and 153 were in the range of 40- 150%. The mean recoveries for ^{13}C-labeled surrogate PCBs were 65-120%.

Some physicochemical water quality data in the environment were as follows: dissolved oxygen: 3.1± 0.6 mg/L; suspended solids: 16.0 ± 3.4 mg/L, total phosphorus: 2.3 ± 0.7 mg/L, total nitrogen: 27.8 ± 4.4 mg/L, chemical oxygen demand: 46.3 ± 6.3 mg/L.

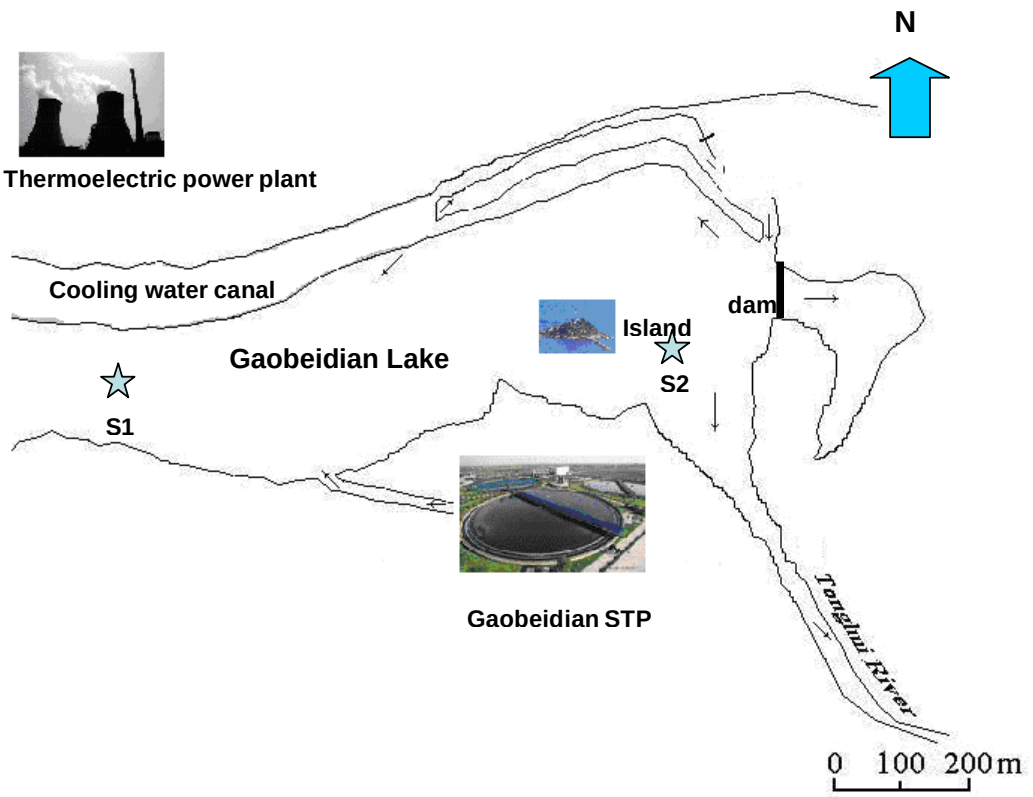


Figure S1 Sampling map. S1 represents the site of Core 1 and S2 is the site of Core 2.

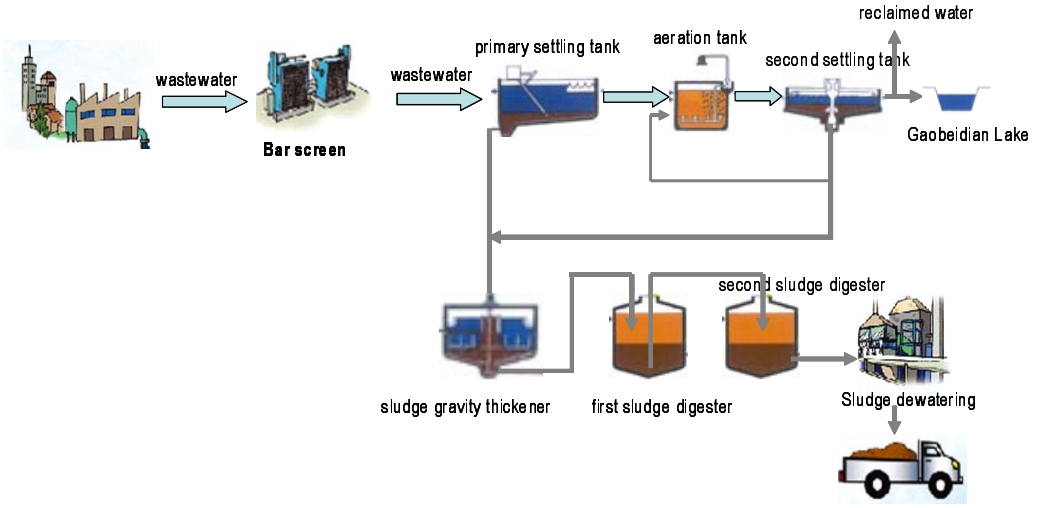
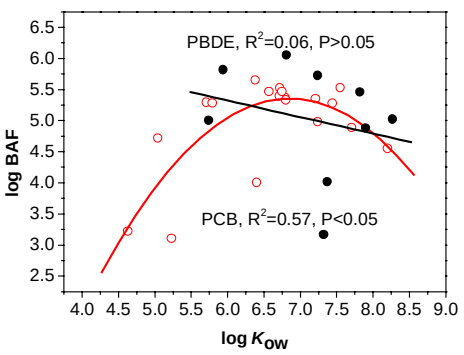
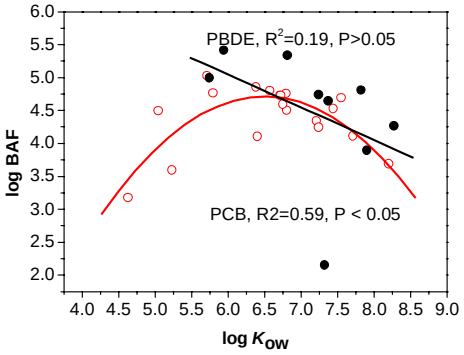


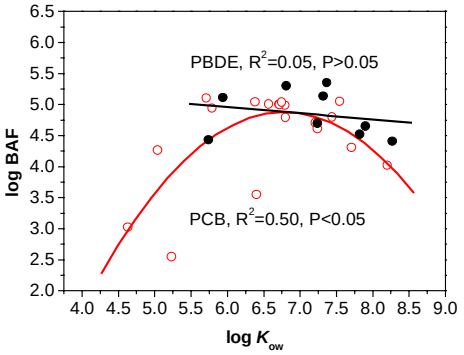
Figure S2 Process flow chart of Gabeidian STP.



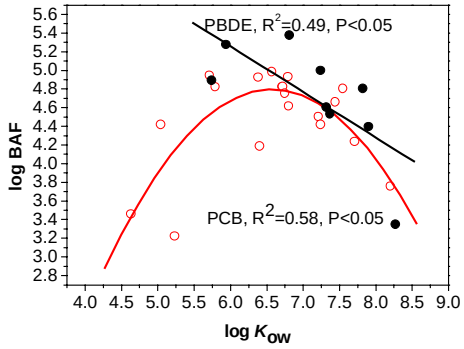
(a) Crucian carp



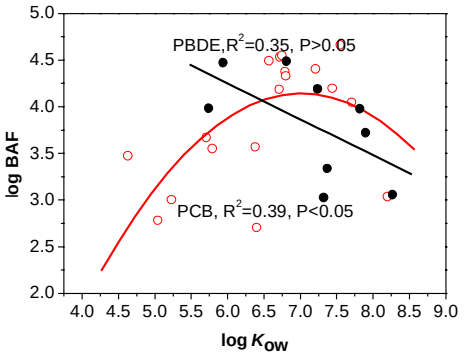
(b) Common carp



(c) Leather catfish



(d) Java tilapia



(e) Chinese softshell turtle

Figure S3. Relationships between log BAF and log K_{ow} of PBDE/PCB congeners. Red cycles represent PCBs and black dots are PBDEs. Value of log K_{ow} of PBDEs are from reference 1 and those of PCBs from reference 2.

Table S1 The total organic carbon of sediment.

Sediment core 1		Sediment core 2	
depth	TOC, %	depth	TOC, %
0-4cm	12.76	0-4cm	7.21
4-8cm	13.26	4-8cm	9.26
8-12cm	10.28	8-12cm	11.02
12-16cm	12.10	12-16cm	8.22
16-20cm	8.90	16-20cm	8.03
20-24cm	15.89	20-24cm	11.38
24-28cm	13.02	24-28cm	12.05
28-32cm	13.07	28-32cm	10.54
32-36cm	10.44	32-36cm	8.25
		36-40cm	9.39

Table S2 The basic information of the samples.

	body length (cm)	body weight (g)	sex	Age (year)	Lipid content
common carp1	40	1500	male	3	0.135
common carp2	36	2750	female	3	0.080
common carp3	43	1600	female	3	0.111
common carp4	30	1200	female	3	0.037
java tilapia1	22	6500	female	1	0.051
java tilapia2	17	500	female	2	0.138
java tilapia3	25	1050	female	1	0.017
leather catfish1	72	2150	female	1	0.156
leather catfish2	57	1600	male	2	0.183
leather catfish3	55	1000	female	2	0.137
leather catfish4	61	1700	female	3	0.256
crucian carp1	18	200	female	4	0.481
crucian carp2	18	99	female	4	0.123
crucian carp3	14	160	male	1	0.168
crucian carp4	17	150	male	4	0.148
crucian carp5	17	149	male	4	0.120
Chinese softshell turtle1		400	male		0.060
Chinese softshell turtle2		463	male		0.028
Chinese softshell turtle3		336	female		0.107

Table S3. Concentrations of PBDEs and PCBs in effluent, lake water, and aquatic species.

Samples	Σ_{12}PBDEs		BDE209		ΣPCBs		
	ng g^{-1} dw	ng g^{-1} OC	ng g^{-1} dw	ng g^{-1} OC	ng g^{-1} dw	ng g^{-1} OC	TEQ, pg/g [a]
effluent	0.84 ng/L	--	<LOD	--	0.93 ng/L	--	--
lake water	0.88 ng/L	--	<LOD	--	0.41 ng/L	--	--
lichen	6.72	--	1572	--	3.11	--	--
March brown	10.05	--	11.37	--	2.08	--	--
Coccid	3.92	--	114.0	--	1.82	--	--
Zooplankton	4.1	--	151.9	--	2.86	--	--
common carp	6.84 (0.25-15.36)[b]	90.86 (3.87-194.5)	5.00 [c]	135.1	12.74 (3.36-23.48)	163.5 (30.25-293.6)	0.71 (0.15-1.37)
java tilapia	4.67 (1.59-9.88)	89.18 (34.70-160.0)	ND		7.62 (2.41-13.51)	184.5 (47.16-408.3)	0.58 (0.16-1.19)
leather catfish	17.08 (9.42-25.83)	100.9 (68.73-101.6)	19.32 (<LOD-60.50)	89.69 (<LOD-236.3)	32.86 (13.02-59.53)	199.7 (71.13-436.6)	2.61 (0.96-4.56)
crusian carp	46.49 (36.68-64.04)	355.0 (104.4-726.9)	1.79 (<LOD-4.80)	12.27 (<LOD-28.59)	62.22 (47.54-86.17)	373.3 (140.9-512.9)	5.40 (3.79-8.33)
Chinese softshell turtle	1.10 (0.14-2.98)	13.60 (3.23-26.33)	2.63 (<LOD-6.74)	83.86 (<LOD-240.82)	1.99 (0.95-3.27)	36.36 (15.82-62.72)	0.24 (0.10-0.47)

[a]: WHO-TEQ in pg/g.

[b]: The values in brackets are the minimum and maximum concentrations.

[c]: BDE-209 was detected in only one sample of common carp, other samples were < LOD.

Table S4 n-octanol-water partition coefficient (logK_{ow}) and n-octanol-air partition coefficient (logK_{oa}) of PBDEs and PCBs

Congener	logK_{ow}	logK_{oa}	Congener	logK_{ow}	logK_{oa}
BDE-17	5.74	*9.20*	CB-3	4.63	7.01
BDE-28	5.94	*9.70*[a]	CB-15	5.23	7.88
BDE-47	6.81	10.34	CB-19	5.04	*6.72*
BDE-66		10.49	CB-28	5.71	*8.03*
BDE-71		*10.59*	CB-52	5.79	*8.33*
BDE-85	7.37	*11.28*	CB-77	*6.40*	9.96
BDE-99	7.32	11.28	CB-101	*6.38*	9.31
BDE-100	7.24	*11.40*	CB-105	6.79	10.27
BDE-138		*10.06*	CB-114	6.71	*9.62*
BDE-153	7.90	12.15	CB-118	6.57	10.08
BDE-154	7.82	*12.18*	CB-123	6.72	*9.83*
BDE-183	8.27	*12.89*	CB-138	6.75	10.09
BDE-209		*15.73*	CB-153	7.55	10.04
			CB-156	7.44	*10.36*
			CB-167	6.80	*10.59*
			CB-180	7.21	10.75
			CB-202	*7.24*	*10.77*
			CB-208	*7.71*	*11.71*
			CB-209	8.20	*12.29*

[a]: Italic represent that the data was obtain by predicted models in the reference 3 and 4.

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