Persistent Halogenated Hydrocarbons in Consumer Fish of China: Regional and Global Implications for Human Exposure

XIANG-ZHOU MENG,[†,‡] EDDY Y. ZENG,[*,†] LI-PING YU,[†,‡] BI-XIAN MAI,[†] XIAO-JUN LUO,[†] AND YONG RAN[†]

State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China, and Graduate School, Chinese Academy of Sciences, Beijing 100049, China

Concentrations of persistent halogenated hydrocarbons (PHHs) were determined in 13 consumer fish species (a total of 390 individuals) collected from a major fish-farming region of China. The potential health risks of PHHs associated with consumption of fish from China was also systematically assessed regionally and globally. In all fish samples, DDTs, HCHs, PCBs, and PBDEs were the predominant PHH residues, with the median levels (ranges) being 6.0 (0.14−698.9), 0.50 (0.13−24.06), 0.10 (<0.02−7.65), and 0.15 (<0.0012−3.85) ng/g wet weight, respectively. The upper-bound (90th percentile) values of estimated daily intakes of DDT, HCHs, PCBs, and PBDEs via fish consumption were 45.5, 1.35, 0.46, and 0.30 ng/kg bw/d (urban), and 15.9, 0.47, 0.16, and 0.10 ng/kg bw/d (rural). Globally, the upper-bound outflows via fishery exportation of DDT, HCHs, PCBs, and PBDEs were 185, 5.51, 1.86, and 1.22 kg, respectively, in 2005. Japan was the largest recipient of PHHs, followed by Korea and the United States. Fish consumption assessments indicated that consumption of freshwater farmed and wild marine fish generally does not subject consumers to significant health risk as far as PHHs are concerned, while limited consumption of seawater farmed fish is advised.

Introduction

Global fish production has grown impressively, with the average per capita supply doubling from 8 kg in 1950 to 16.2 kg in 2002 (live weight equivalent) (*1*). Overall, fish provided more than 2.6 billion people with at least 20% of the average per capita animal protein consumption (*1*). Many studies indicated that fish were good sources of proteins, n-3 polyunsaturated fatty acids, which were beneficial for human health. Balanced against this, however, are potential health risks associated with consumption of contaminated fish (*2*, *3*).

Currently, China (mainland) has the largest fishery production in the world with an output of 44.3 million metric tons in 2002, accounting for 33.3% of the total global production. China has overtaken Thailand to become the world's largest exporter of fishery products since 2002 (*1*). The main exporting destinations include Japan, Korea, the United States, Germany, Canada, Hong Kong, and Russia, etc. Despite the increasingly important impact of China's fishery products on the global population, the state of fish contamination by persistent halogenated hydrocarbons (PHHs) and associated human health risk have not been systematically assessed in China.

This study aimed to determine the levels of PHHs, including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs), in typical consumer fish of China, assess potential health risks associated with fish consumption, estimate the outflows of PHHs from China to its global trade partners, and provide preliminary advisories on consumption of fish from China. Sampling was conducted in Guangdong Province of South China (Figure S1; "S" designates figure and tables in the Supporting Information), where the fishery production reached 6.65 million metric tons in 2004, representing 13.6% of the total output in China, and ranked No. 2 among all the provinces in the country (*4*). As a result, fish collected from this region are sufficiently representative of the overall fish output and exportation from China.

Materials and Methods

Sample Collection. A total of 390 fish individuals were randomly collected between November 2004 and January 2005, from local markets in 11 coastal cities of Guangdong Province, one of the economically fastest growing and typical fishery-producing regions in China (Figure S1). The sampled species include seven freshwater farmed fish (tilapia (*Tilapia*), grass carp (*Ctenopharyngodon idellus*), bighead carp (*Aristichthys nobilis*), bluntsnout bream (*Megalobrama amblycephala*), largemouth bass (*Micropterus salmoides*), mandarin fish (*Siniperca chuatsi*), and northern snakehead (*Ophicephalus argus*)), three seawater farmed fish (red drum (*Sciaenops ocellatus*), snubnose pompano (*Trachinotus blochii*), and crimson snapper (*Lutjanus erythopterus*)), and three wild marine fish (hairtail (*Trichiurus lepturu*), golden thread (*Nemipterus virgatus*), and common mullet (*Mugil cephalus*)). To ensure sufficient statistical power for data analysis, approximately 30 individuals were collected for each species. Such a number of samples ensure that the error associated with a comparison of analyte distribution in different species is approximately 10% with a 90% confidence interval (*5*), which is expected to be within the analytical error. After transport to laboratory, they were stored at −20 °C until chemical analysis.

Sample Preparation and Extraction. Materials used in sample preparation and instrumental analysis are described in detail in the Supporting Information. About 20 g (wet weight) of skin-off fish muscle was taken, homogenized, freeze-dried, and ground into fine powder. After spiking with the surrogate standards of 2,4,5,6-tetrachloro-*m*-xylene (TMX), PCB 67, PCB 191, PCB 209, and ^{13}C−PCB 141, each was Soxhlet extracted with a 1:1 (v:v) acetone/hexane mixture for 48 h. Lipid content was determined gravimetrically using 20% of the extract. The remaining extract was applied to the head of a gel permeation chromatography column, packed with 40 g of SX-3 "Bio-Beads" (Bio-Rad Laboratories, Hercules, CA) in a 50 cm × 2.5 cm i.d. glass column with a PTFE stopcock. The column was eluted with 1:1 dichloromethane/hexane (v:v) for lipid removal. The fraction from 90 to 280

* Corresponding author phone: 86-20-85291421; fax: 86-20-85290706; e-mail: eddyzeng@gig.ac.cn.
† Guangzhou Institute of Geochemistry.
‡ Graduate School, Chinese Academy of Sciences.

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mL was collected and concentrated to 4 mL by a TurboVap II evaporator (Zymark, Hopkinton, MA). For analysis of OCPs, 2 mL of extract was purified on a 10 mm i.d. silica/alumina column packed, from the bottom to top, with neutral alumina (6 cm, 3% deactivated) and neutral silica gel (12 cm, 3% deactivated). The first fraction was eluted with 5 mL hexane and discarded. The second fraction was eluted with 80 mL of dichloromethane/hexane (3:7 in volume) and collected. For PCBs and PBDEs, the remaining 2 mL extract was cleaned on a 10 mm i.d. silica/alumina column packed, from the bottom to top, with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), 25% sodium hydroxide silica (5 cm), neutral silica gel (2 cm, 3% deactivated), and 50% sulfuric acid silica (6 cm). The PCBs and PBDEs mixtures were eluted with 70 mL of dichloromethane/hexane (1:1 in volume). Both extracts were further concentrated to 100 μL under a gentle stream of N_2. Finally, the internal standards (PCB 82 for OCPs, PCB 24 and PCB 189 for PCBs, and ^{13}C−PCB 208 for PBDEs) were added before instrumental analysis.

Instrumental Analysis. The detailed analytical and quantification procedures are presented in the Supporting Information. Briefly, OCPs and PCBs were quantified with a Hewlett-Packard (HP) 5890 gas chromatograph (GC) coupled to a 5972 mass spectrometer (MS) and a Varian 3800 GC interfaced with a Saturn 2000 MS, respectively, in the selective ion monitoring (SIM) mode. Injection of 1-uL samples was conducted manually on the HP GC/MS and automated with a Varian CP-8410 autosampler, both in the splitless mode. For PCBs, a total of 70 chromatographic peaks containing 112 individual or coeluting congeners (designated as domains) were quantified by a secondary Aroclor mixture of 1242/1248/1254/1260 (1:1:1:1). The concentration of PCBs was defined as the sum of the concentrations of individual congeners and domains. For PBDEs, the detailed procedure was described elsewhere (*6*). With a signal/noise ratio better than 3, the wet-weight-based limit of detection (LOD; all normalized to wet weight except where indicated) for most OCPs (except for endrin and endosulfan for which the LOD was 25 pg/g) ranged from 10 to 20 pg/g. The LODs varied from 1.2 to 2.5 pg/g for all tri- to hepta- BDEs congeners (the LOD for BDE 209 was 100 pg/g). The LODs for individual PCB congeners varied from 20 to 40 pg/g.

Quality Assurance/Quality Control. For each batch of 20 fish samples, a procedural blank, a spiked blank, a matrix spiking sample, a matrix spiking duplicate, and a standard reference material (NIST SRM1946) were processed. The spiked samples contained 21 OCPs (α-HCH, β-HCH, γ-HCH, δ-HCH, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin, endrin ketone, endrin aldehydes, *o,p′*-DDT, *o,p′*-DDE, *o,p′*-DDD, *p,p′*-DDT, *p,p′*-DDE, *p,p′*-DDD, endosulfan I, ensosulfan II, endosulfan sulfate, and methoxychlor) and 11 PBDE congeners (BDE 28, 47, 66, 85, 99, 100, 138, 153, 154, 183, and 209). Recoveries of 21 OCPs and 11 PBDEs in these spiked samples were 86.3 ± 18.4% and 87.2 ± 19.0%, respectively. The surrogate recoveries of TMX, PCB 67, PCB 191, ^{13}C−PCB 141, and PCB 209 were 55.3 ± 11.1%, 83.2 ± 12.5%, 89.8 ± 11.0%, 81.8 ± 17.3%, and 84.8 ± 18.4%, respectively. Only low concentrations of OCPs, PCBs, and BDE-47 were detected in procedural blanks. Hence, blank values were not subtracted from the sample measurements. Reported concentrations were not surrogate recovery corrected.

Data Analysis. For samples with contaminant concentrations below detection limits, zero was used for the calculations. All PHH data were examined using a nonparametric test (Kolmogorov−Smironov Z) before ANOVA analyses (SPSS v 13.0). The detailed procedures for estimated daily intakes (EDIs) and risk-based fish consumption advisories are presented in the Supporting Information.

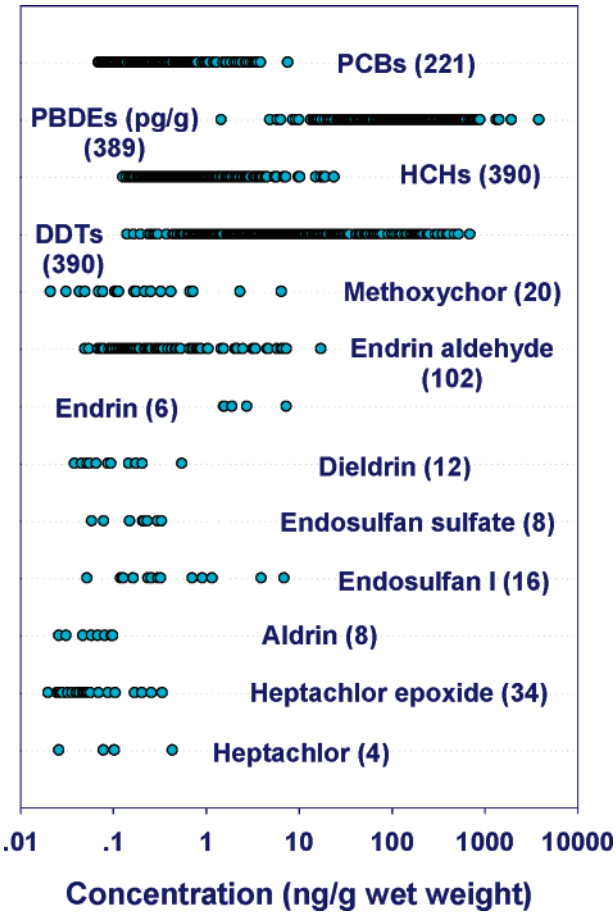

FIGURE 1. Concentrations (ng/g wet weight; pg/g for PBDEs) of 13 PHHs determined in fish samples of China. The numbers in parentheses are the numbers of samples containing detectable analytes.

Results and Discussion

Levels of PHH Contamination. Preliminary analysis indicated that DDTs (sum of *o,p′*-DDT, *o,p′*-DDE, *o,p′*-DDD, *p,p′*-DDT, *p,p′*-DDE, and *p,p′*-DDD), HCHs (sum of α-HCH, β-HCH, γ-HCH, and δ-HCH), PCBs, and PBDEs (sum of BDE 28, 47, 66, 85, 99, 100, 138, 153, 154, and 183) were the predominant and ubiquitous PHH residues in the fish samples (Figure 1). The occurrence frequencies for other OCP compounds, such as heptachlor, heptachlor epoxide, aldrin, dieldrin, endosulfan (I), endosulfan sulfate, endrin, endrin aldehyde, and methoxychlor, were 1%, 9%, 2%, 3%, 4%, 2%, 2%, 26%, and 5%, respectively. No endosulfan (II) or endrin ketone were detected. Therefore, only the data of DDTs, HCHs, PCBs, and PBDEs were reported herein. The 50th and 90th percentile concentrations were used as the mean and upper-bound values for comparison and assessments.

The concentration ranges of DDTs, HCHs, PCBs, and PBDEs were 0.14−698.9, 0.13−24.06, <0.02−7.65, and <0.0012−3.85 ng/g (all normalized to wet weight except where indicated), respectively, when all fish species were considered. Detailed concentration data of PHHs are given in Table S1. It is worthwhile to note that the concentrations of all analytes varied widely with individual fish samples. A large variability of analyte concentrations within the same species was also evident (Figure 2). A recent study by Domingo et al. (*7*) obtained generally narrower concentration ranges in marine species with three composite samples (20 individuals per composite) for each species, but large variabilities remained in a few species. Clearly, the occurrence and

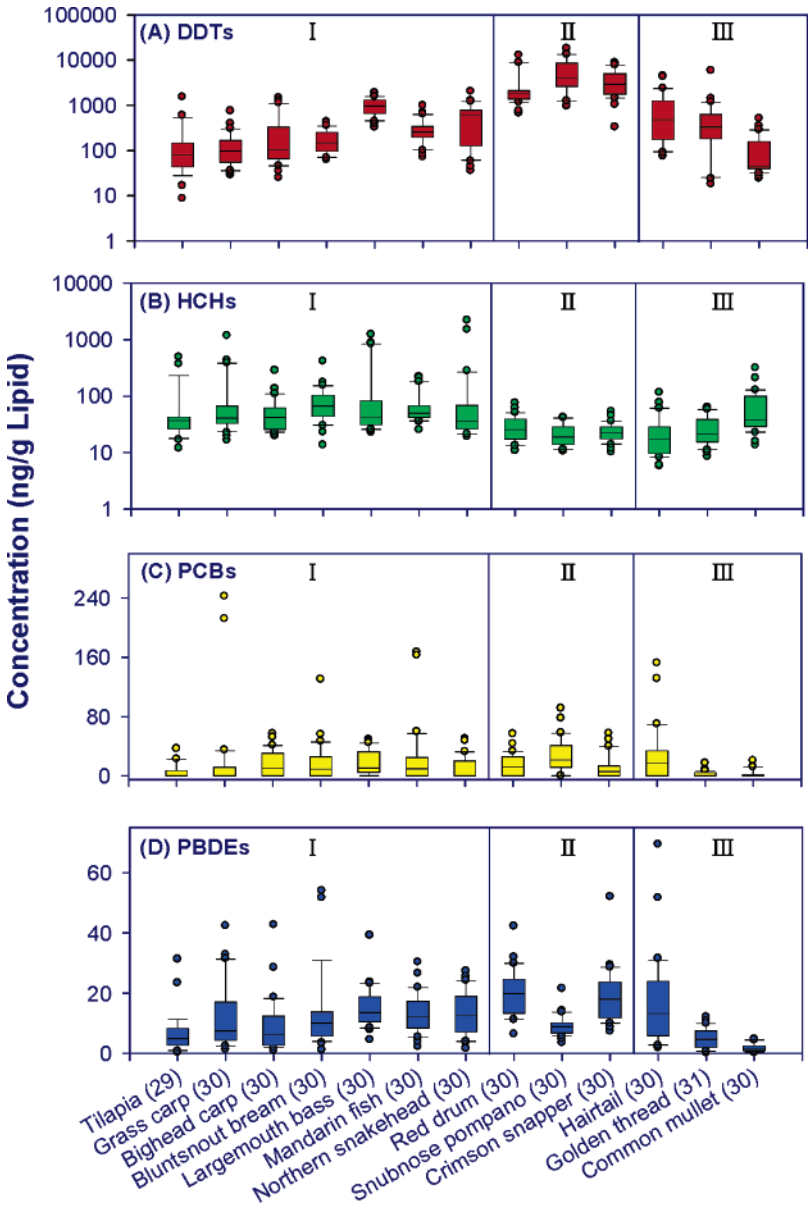

FIGURE 2. Concentrations (ng/g lipid weight) of DDTs, HCHs, PCBs, and PBDEs in (I) seven species of freshwater farmed fish, (II) three species of seawater farmed fish, and (III) three species of wild marine fish. The numbers of individuals for different species are given in parentheses. The horizontal lines represent 10th, 50th, and 90th percentiles, and the boxes represent 25th and 75th percentiles. Outliers are shown as individual points.

distribution of PHHs were highly variable even in fish collected from a relatively small region. In any sampling effort of a regional or global scale with limited sample numbers, large error can therefore be expected especially if samples representing the highest or lowest end of the concentration range were unintentionally collected. Occurrence of individual PHH groups and comparison with those in edible fish from other countries or regions are detailed below.

DDTs. Overall, the concentrations of DDTs were 1−2 orders of magnitude higher than those of HCHs, PCBs, and PBDEs (Table S1). In addition, all 390 samples contained detectable DDTs, indicating that these pollutants were ubiquitous in the biosphere of China. This resulted from the large production and widespread usage of DDTs in China from the 1950s to 1983 (*8*). The median concentration (6.0 ng/g wet weight or 352 ng/g lipid weight) of DDTs in all fish species was lower than those reported by Kannan et al. (Table S2). Mean concentrations as high as 28, 26, and 22 ng/g were reported in fish from Indonesia, Vietnam, and Australia, respectively (*9*). Darnerud et al. (*10*) reported a mean concentration of 7.02 ng/g in fish from Swedish markets, which was comparable to our median concentration. Jacobs et al. (*11*) reported a maximal concentration of 249.5 ng/g (lipid weight) in fish from European markets, which was lower than our median concentration (352 ng/g lipid weight). On the other hand, the maximum concentrations of DDTs in individual samples were higher than those from other regions of China and other countries in Asia, Oceania, America, Europe, and Africa. This may suggest the presence of current point sources of DDTs in the study areas, which may include intermittent production of DDT mixtures to satisfy export demand and the manufacture and use of dicofol technical products known to contain a large amount of DDT residues (*12*).

HCHs. The general concentration range of HCHs (0.13−24.06 ng/g) was wider than those reported in Dalian (0.15−4.25 ng/g), Tianjin (0.35−0.54 ng/g), Shanghai (0.044−2.64) (*13*), and Zhoushan (<0.1 ng/g) (*14*) areas of China (Table S3). Compared with those obtained from other countries, our median value (0.50 ng/g) was similar to those from

Sweden (0.96 ng/g) (*10*), Thailand (0.82 ng/g), Australia (0.34 ng/g), Indonesia (0.73 ng/g), and Korea (0.94 ng/g), but was much lower than those found in India (28 ng/g) and Vietnam (1.8 ng/g) (*9*) (Table S3). Also, the median concentration of HCHs (34.82 ng/g lipid weight) from the present study was lower than those in biota from the Danube Delta, Romania (mean concentration of 181 ng/g lipid weight) (*15*) and in fish from Turkey (*16*), but was higher than those in salmon from European markets (*11*).

PCBs. PCBs were detectable in 57% (221 individuals out of 390) of the samples analyzed, with a concentration range (<0.02−7.65 ng/g) comparable to those reported in Dalian (1.11−8.04 ng/g), Tianjin (1.26−5.60 ng/g) and Shanghai (0.83−11.4 ng/g) (*13*) and wider than those obtained in Zhoushan (0.24−1.4 ng/g) (*14*) (Table S4). Our median concentration of PCBs (0.10 ng/g wet weight or 5.67 ng/g lipid weight) was much lower than those reported in fish from Salton Sea, CA (30.21 ng/g) (*17*), Marmara, Turkey (253.08 ng/g lipid wt) (*16*), Sweden (9.58 ng/g) (*10*), Australia (55 ng/g), and Korea (23 ng/g) (*9*) (Table S4). These results supported an early suggestion (*18*) that production and/or usage of PCBs was relatively modest in China.

PBDEs. The range of PBDEs (<1.2−3850 pg/g or 118−69410 pg/g lipid wt) was within the range in fish from Canada (1.3−5500 pg/g) (*19*), but wider than the range obtained in Japan (79−547 pg/g) (*20*) and the Danube Delta, Romania (<0.1−14300 pg/g lipid wt) (*15*) (Table S5). The overall PBDE levels, with a median concentration of 153 pg/g or 9706 pg/g lipid wt, in consumer fish of China were clearly lower than those reported in market-purchased fish from the United States (median concentration 1735 pg/g) (*21*), Spain (mean concentration 564 pg/g) (*7*), and Sweden (mean concentration 634 pg/g) (*10*), and in salmon from Southern Chile (1460 pg/g) (*22*) (Table S5). However, the maximum concentration of PBDEs observed in the present study was higher than those from other countries (except for Canada), likely due to the presence of hot spots in the study area.

Occurrence of PHHs in Different Fish Species. Aquaculture has become a dominant practice in China's fish production since the early 1980s, as reliance on wild fish stocks cannot meet the growing demand. In 2004, the output from freshwater and seawater farming accounted for more than 65% of the total fish production, compared to 26% in 1978 (*4*). Figure 2 shows the concentration levels of PHHs in various fish species, which were log-normally distributed as determined by Kolmogorov−Smironov Z nonparametric test ($P > 0.01$). ANOVA analyses were conducted to compare the contaminant levels in freshwater farmed, seawater farmed, and wild marine fish. Freshwater and seawater farmed fish were taken as a single type when farmed and wild fish were compared. The PHHs were significantly more concentrated in farmed fish as a group than in wild fish. This was consistent with the results obtained in previous studies (*23*). Additionally, levels of DDTs and PBDEs in seawater farmed fish were significantly higher than those in freshwater farmed fish. However, HCHs concentrations were significantly lower in seawater farmed fish than in freshwater farmed fish. Furthermore, seawater farmed fish contained significantly higher concentrations of DDTs, PCBs, and PBDEs but lower concentrations of HCHs than wild marine fish. Freshwater farmed fish also contained significantly higher concentrations of HCHs and PBDEs, but lower DDTs concentrations than wild marine fish. These results indicated that seawater farmed fish accumulated more DDTs, PBDEs, and PCBs than freshwater farmed fish and wild marine fish. This probably reflected the impact of contaminant discharge from the Pearl River Delta of southern China on the coastal environment where most seawater farmed fish are raised (*24*, *25*).

The different pattern with HCHs compared to those with other PHHs may be contributed to the manner of usage of HCHs in China. Technical HCHs were banned in 1983 by the Chinese government, and in 1991 technical HCHs were replaced by a purified active isomer γ-HCH, known as lindane (*26*). As a result, HCHs levels have declined dramatically in foodstuffs (*27*) and biota (*28*) over the past two decades because of the low persistency, high biodegradability, and moderate volatility with HCHs compared to DDTs (*27*). Typically, seawater farmed fish are cultured in intertidal zones, small bays, and shallow seas, while freshwater farmed fish are raised in ponds, lakes, reservoirs, and river channels close to agricultural areas. Freshwater farmed fish may be more easily influenced than seawater farmed fish and marine wild fish by HCHs residues residing in agricultural areas.

Human Exposure. Potential health risks from exposure to PHHs were assessed using two criteria. The first criterion was the maximum residue level (MRL), the concentration of a pesticide allowable in agriculture products, or maximum level (ML) for other chemicals. In 2005, China reestablished MRLs of 0.5 and 0.1 mg/kg for DDTs and HCHs, respectively, and a ML of 2.0 mg/kg for PCBs in fishery products (*29*, *30*). Currently no MRL is available for PBDEs. In the fish samples analyzed in the present study, the mean and upper-bound concentrations (Table S1) of DDTs, HCHs, and PCBs were all well below the respective MRLs or MLs. Also, no fish samples contained higher contaminant levels than the action levels and tolerances for DDTs (5.0 mg/kg) and PCBs (2.0 mg/kg) developed by the U.S. Food and Drug Administration (U.S. FDA) (*31*). However, two fish samples contain higher DDTs levels than the MRL, and 13.8% of the samples contain DDTs at levels higher than the maximum admissible concentration (0.05 mg/kg wet weight) established by the Europe Union for human consumption (*32*).

The second criterion was the acceptable daily intakes (ADIs) and provisional tolerable daily intakes (PTDIs) recommended by the Food and Agriculture Organization and World Health Organization (FAO/WHO) to assess human exposure to target contaminants over a lifetime without appreciable health risk, expressed as ng/kg body weight/day (ng/kg bw/d) assuming a typical body weight of 60 kg (*33*). The per capita fish consumption values were 34.19 g for urban and 12.30 g for rural residents (assuming 365 days a year), respectively, reported by the Chinese government/organizations (*4*). Using the PHHs data from the present study, we calculated EDIs of PHHs via fish consumption. As shown in Table 1, the mean values of EDIs (EDI_{50}) of DDTs for urban and rural residents of China were 3.66 and 1.27, while the upper-bound values (EDI_{90}) were 45.5 and 15.9 ng/kg bw/d, respectively. All these values were far below the PTDIs for DDTs (10 000 ng/kg bw/d) (*33*). EDI_{50} values of HCHs were 0.30 and 0.11 ng/kg bw/d, respectively, for urban and rural residents, and EDI_{90} values were 1.35 and 0.47 ng/kg bw/d, also well below the ADIs for γ-HCH (5000 ng/kg bw/d) (*33*). No ADIs were available for PCBs and PBDEs. The EDI_{50} values were 0.06 and 0.02 ng/kg bw/d and the EDI_{90} values were 0.46 and 0.16 ng/kg bw/d for PCBs, whereas the EDI_{50} values for PBDEs were 0.09 and 0.03 ng/kg bw/d and the EDI_{90} values were 0.30 and 0.10 ng/kg bw/d. Geographically speaking, residents in costal regions, such as Shanghai, Hainan, Zhejiang, Guangdong, and Fujian, were subject to greater exposure to PHHs than those living in the inland regions (Figure S1). Furthermore, the maximum EDI_{90} among all regions of China was 52.9 ng/kg bw/d for DDTs, whereas it was 1.63 ng/kg bw/d for HCHs. These were all much lower than the PTDIs (for DDTs) or ADIs (for HCHs) (Table S6).

Outflows of PHHs from China. In 2005, China (mainland) exported a total of 2.48 million metric tons of fishery products (including fish and seafood products) (*34*). Importation occurred into Asia, Europe, North America, South America, Africa, and Oceania. Japan, Korea, and the United States were the largest recipients, accounting for 25.9, 20.8, and

TABLE 1. Estimated Dietary Intakes (EDIs) (ng/kg Body Weight/Day) of PHHs by Residents in Different Regions of China and Other Countries or Regions (Associated with Chinese Fish)[a]

	DDTs		HCHs		PCBs		PBDEs	
country or region	EDI_{50}	EDI_{90}	EDI_{50}	EDI_{90}	EDI_{50}	EDI_{90}	EDI_{50}	EDI_{90}
China (urban)	3.66	45.5	0.30	1.35	0.06	0.46	0.09	0.30
China (rural)	1.27	15.9	0.11	0.47	0.02	0.16	0.03	0.10
Hong Kong	5.66	70.4	0.47	2.09	0.09	0.71	0.14	0.46
Macau	7.06	87.9	0.59	2.61	0.12	0.88	0.18	0.58
Taiwan	0.82	10.2	0.07	0.30	0.01	0.10	0.02	0.07
Japan	1.38	17.1	0.11	0.51	0.02	0.17	0.03	0.11
Korea	2.92	36.4	0.24	1.08	0.05	0.37	0.07	0.24
United States	0.37	4.58	0.03	0.14	0.006	0.05	0.009	0.03
Canada	0.36	4.44	0.03	0.13	0.006	0.04	0.009	0.03
Germany	0.35	4.39	0.03	0.13	0.006	0.04	0.009	0.03
United Kingdom	0.15	1.92	0.01	0.06	0.003	0.02	0.004	0.01
Poland	4.95	61.6	0.41	1.83	0.08	0.62	0.12	0.40
Sweden	3.02	37.6	0.25	1.12	0.05	0.38	0.08	0.25
Other Asian countries	0.02	0.29	0.002	0.009	0.0004	0.003	0.0006	0.002
Other European countries	0.11	1.42	0.01	0.04	0.002	0.01	0.003	0.009
Other North American countries	0.09	1.10	0.007	0.03	0.001	0.01	0.002	0.007
South America	0.006	0.08	0.0005	0.002	0.0001	0.0008	0.0002	0.0005
Africa	0.009	0.12	0.0008	0.003	0.0002	0.001	0.0002	0.0008
Oceania	0.21	2.65	0.02	0.08	0.004	0.03	0.005	0.02

[a] The EDI_{50} and EDI_{90} were calculated using the 50th and 90th percentile concentrations of PHHs.

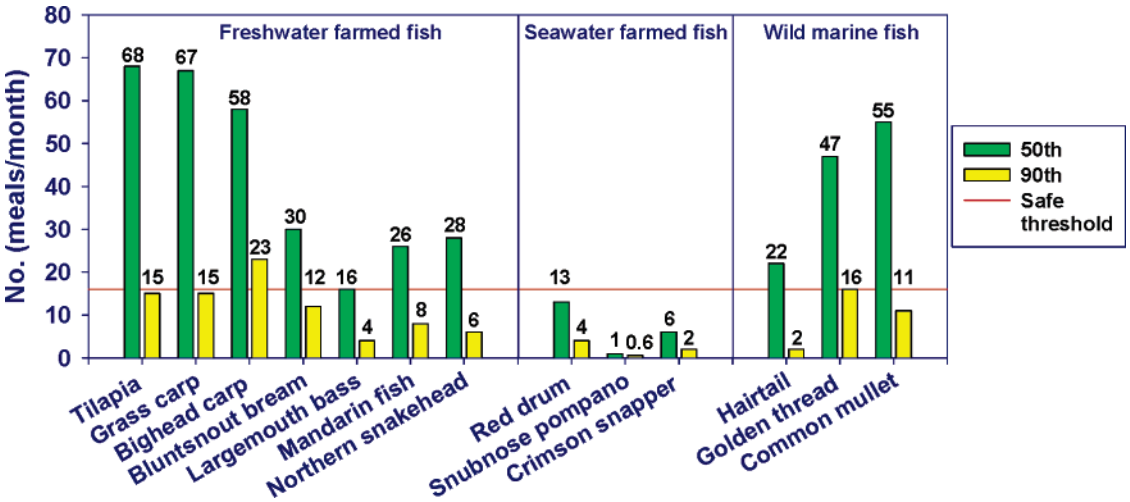

FIGURE 3. Risk-based consumption advisories using the concentration data for DDTs, HCHs, and PCBs for seven species of farmed freshwater fish, three species of farmed seawater fish, and three species of wild marine fish. The solid red line is a safe fish consumption threshold, suggesting that the consumption of this species does not require restriction based on the PHHs evaluated (16 meals/month). The numbers on the green bars are calculated with the 50th percentile concentrations, while the numbers on the yellow bars are calculated with the 90th percentile concentrations.

16.1%, respectively, of the total. Using the mean concentration values, the mean outflows of DDTs, HCHs, PCBs, and PBDEs from China to the globe were about 14.9, 1.24, 0.25, and 0.37 kg (Table S7). The corresponding upper-bound outflows were 185, 5.51, 1.86, and 1.22 kg, respectively, in 2005. Japan accepted the largest upper-bound outflows of DDTs, HCHs, PCBs, and PBDEs at 48.0, 1.43, 0.48, and 0.32 kg, respectively, followed by Korea at 38.6, 1.15, 0.39, and 0.25 kg. The United States ranked third at 29.7, 0.88, 0.30, and 0.20 kg. Based on these estimates and the populations of the importing countries or regions (*35*), exposure to PHHs via consumption of fish from China can be estimated. As shown in Table 1, the residents of Macau were subject to the highest exposure to PHHs in association with consumption of fish from China, followed by those of Hong Kong, Poland, Sweden, Korea, and Japan. By contrast, the residents of Africa and South America were subject to the least exposure to PHHs.

Advisories on Fish Consumption. Generally, most humans are simultaneously exposed to a suite of contaminants (e.g., DDTs, HCHs, PCBs, heavy metals, and endocrine-disrupting chemicals). The U.S. Environmental Protection Agency developed a risk-based approach (*31*) to multiple chemical exposure for assessing carcinogenic and noncarcinogenic effects caused by DDTs, HCHs, and PCBs. Here we used this approach with an acceptable risk level of 1 in 100,000 to derive consumption advisories for consumer fish of China. The mean and upper-bound concentrations of DDTs, HCHs, and PCBs in this study and the corresponding cancer slope factor for carcinogenic effects were used to calculate species-specific consumption limits expressed as number of fish meals per month. The cancer slope factor developed for γ-HCH was used for HCHs. PBDEs were not included in the calculation due to the lack of toxicity data. The risk-based meal consumption limits for the fish species and PHHs under consideration (Figure 3) suggested that

consumption of seawater farmed fish was more likely to raise health concern than that of wild marine fish or freshwater farmed fish. If the mean concentrations of PHHs were used, no restrictive limits should be imposed for freshwater farmed fish and wild marine fish (i.e., more than 16 meals per month allowed). Consumption of bighead carp (freshwater farmed) and golden thread (wild marine) would not result in health concern even if the upper-bound concentrations of PHHs were used in the calculation. On the other hand, less than 13 meals (mean) and 4 meals (upper-bound) per month were allowed for seawater farmed fish. The worst case occurred with snubnose pompano, a cage-cultured seawater fish, where no more than one meal per month should be allowed (Figure 3).

Our results suggested that the concentrations of PHHs in consumer fish of China were moderate, with the mean and upper-bound values much lower than the corresponding MRLs or MLs established by the Chinese government, and the action levels and tolerances developed by the U.S. FDA. Regionally and globally, the human daily intakes of DDTs and HCHs were far below the PTDIs and ADIs recommended by the FAO/WHO. Intakes of PCBs and PBDEs were also low. PHHs were more concentrated in farmed fish than wild fish, particularly marine species. Accordingly more stringent consumption advisories were desirable for seawater farmed fish than for wild and freshwater farmed fish. In addition, further extensive investigations of other toxic substances, such as heavy metals and endocrine-disrupting chemicals, are required in Chinese consumer fish.

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Supporting Information Available

Detailed description of methods, tables of data, additional figures. This material is available free of charge via the Internet at http://pubs.acs.org.

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