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Research Article

Pollution Characteristics and Health Risk Assessment of Halogenated Organic Pollutants (HOPs) in Fish from the Laibin Section of Hongshui River, China

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This study used gas chromatograph-mass spectrometer (GC-MS) to determine three kinds of halogenated organic pollutants (HOPs) in the edible muscles of nine fish species in the Laibin section of the Hongshui River of Xijiang River in the Pearl River system. The results showed that the sum of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) in fish muscle was 8.94~43.32 ng/g ww, 1.22~4.09 ng/g ww, and ND~0.13 ng/g ww, respectively. Evaluation indicators include analysis of the composition of homologous compounds, residues in different diets and habitats, and estimated daily intake (EDI) calculations. The analysis of the composition characteristics of homologous compounds showed that endrin was the most dominant in OCPs; hexachlorobenzene was the most dominant in PCBs; and BDE 154 was the most dominant PBDEs. Calculations of the levels of the HOPs in fish of different diets and habitats showed that OCPs and PCBs had the highest residues in carnivorous fish, and PBDEs had the highest residues in herbivorous fish. The residual OCPs, PCBs, and PBDEs in demersal fishes were the most. The estimated daily intake (EDI) results showed that daily fish consumption in the Laibin section of the Hongshui River would not cause HOPs health risks to the local residents. However, there is still a need for continuous monitoring of HOPs to reduce pollutant emissions and prevent pollution conditions from arising.

1. Introduction

Halogenated organic pollutants are a class of persistent organic pollutants containing halogenated elements, which are characterized by long-term residual, bioaccumulation, long-range migration, and high biotoxicity. They can be amplified along the food chain step by step, which can cause great harm to biological and human health [1]. The low cost and widespread use of these chemicals in the industrial, agricultural and health sectors have led to their continued use in many developing countries [2]. Studies have shown that residues of HOPs are still detectable worldwide [3–6]. Even today, there are still some HOPs being produced and used in China, which makes HOPs pollution a constant problem [7]. Therefore, it is necessary to test the Laibin section of the Hongshui River for HOPs levels to see if any pollutants remain.

In recent years, river ecosystems around the world have been polluted to varying degrees due to rapid population growth and economic development. Human activities change the hydromorphological, physicochemical, and biological parameters of surface water bodies, affecting their biodiversity and ecological functioning [8]. To monitor the pollution status of rivers, many researchers consider aquatic organisms as an important object of study. Aquatic organisms are capable of absorbing and enriching bioaccumulative pollutants through the food chain or directly from water and sediment [9]. Fish are almost ubiquitous in aquatic environment and play an important ecological role in aquatic food chain because they are energy carriers from low to high trophic levels. Therefore, understanding uptake, behavior and response of toxicant in fish may have a high ecological significance [10]. At present, using fish as a bioindicator of chemical pollutants has been a common procedure in several countries worldwide like China, Brazil, Argentina, the Atlantic Ocean, and other regions [11–14].

The Hongshui River is the main stream of the Xijiang River system in the Pearl River basin. It has an excellent ecological environment for the growth and reproduction of aquatic organisms, high fishery yield, and wide variety of fish species (Huang and Zeng, 2008). However, in recent years, there are fewer studies related to HOPs in fish from the Hongshui River, and there is a lack of powerful data to explain whether fish from the Hongshui River are contaminated with HOPs. Therefore, in this study, nine common food fish species from the Laibin section of the Hongshui River were selected to determine the content of HOPs in their edible muscle, calculate the ecological contamination risk level, and assess the potential health risk of human consumption of HOPs.

2. Materials and Methods

2.1. Sample Collection and Preparation. In July 2021 and December 2021, nine common edible fish (*Cirrhinus molitorella*, *Ctenopharyngodon idella*, *Cyprinus carpio*, *Squaliobarbus curriculus*, *Ptychidio jordani*, *Carassius auratus*, *Silurus asotus*, *Tachysurus fulvidraco*, and *Channa argus*) were collected from the Laibin River section in the Hongshui River (Figure 1). Five mature and well-formed individuals of each species were collected as experimental samples. The basic characteristics of the fish are shown in Table 1. All samples were sealed in plastic bags and stored at −20°C until chemical analysis.

The samples thawed at room temperature were cleaned with deionized water, scaled, and peeled. The edible muscles of their backs were dissected with a stainless-steel scalpel and weighed wet. Then, put the sample into a tin foil paper bag, freeze-dried for 48 h, and weighed for dry weight. We grounded the freeze-dried samples into fine powder. The operation is carried out on aluminum foil paper to avoid pollution.

2.2. Chemicals. Three types of HOPs were determined as follows: OCPs: α-, β-, γ-, δ-Hexachlorocyclohexanes (ΣHCHs), o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'-DDT, p,p'-DDT (ΣDDTs), p,p'-methoxychlor (MXC), heptachlor, aldrin, dieldrin, endrin, endrin-ketone, endrin-aldehyde, heptachlor epoxide, endosulfan I, endosulfan II, and benzoepin sulfate; PCBs: PCB 3, 18, 19, 77, 81, 114, 118, 123, 126, 128, 138, 153, 169, 170, 180, 187, 208, and 209; PBDEs: BDE 28, 99, 100, 154, and 208 [15].

Figure 1: The study area and sampling sites.

2.3. Extraction and Analysis. The extraction and analysis of HOPs referred to a previous study [16] with minor modifications according to laboratory conditions. Approximately 0.5 g of freeze-dried muscle sample was grounded into fine powder and then mixed with dichloromethane. 100 ng of PCB 24, 82, and 198; 20 ng of PBDE 118 and 128; and 50 ng of $^{13}C_{12}$-BDE 209 were added as an internal standard before extraction. The sample was ultrasonically extracted with 4 ml of dichloromethane three times. The lipids were removed with 5 ml of concentrated sulfuric acid and then centrifuged at 3000 rpm for 5 minutes. The supernatant was transferred to a new tube and concentrated to 1 ml under gentle nitrogen flow. The extracts were purified using a multilayer silica column packed with neutral silica gel (8 cm, 3% deactivated, activated at 180°C), 50% (on a weight basis) sulfuric acid silica (8 cm), anhydrous sodium sulfate, and 1 g anhydrous sodium sulfate (activated at 450°C). We used dichloromethane to elute HOPs. The final eluate was concentrated to near dryness under a gentle stream of pure nitrogen. Known amounts of recoveries of surrogate standards (100 ng of PCB 30, 65, and 204; 20 ng of PBDE 77, 181, and 205) were added to the extract and make up to 100 μl with isooctane prior to instrumental analysis.

PBDEs were analyzed by a 7890 Agilent gas chromatograph (GC) coupled with a 5975 mass spectrometer (MS) operated in an electron capture negative ionization mode. The concentrations of PCBs and OCPs were determined by a 7890 Agilent GC coupled with a 5975 MS in an electron ionization mode.

2.4. Quality Control and Quality Assurance. Set the program blank in the chemical analysis process of the sample to check the sample pollution caused during the experiment. The results showed that no target compound was detected in the blank sample of the program or the content was very low, indicated that there was no sample contamination caused by human or other environmental factors during the experiment. The average recoveries of surrogate standards in all samples were as follows: 73.58–90.06% for PCB 30, 80.72–103.58% for PCB 65, and 84.86–101.14% for PCB 204

Table 1: Basic characteristics of nine fish species in the Laibin section of Hongshui River.

Species	*n*	Feeding habits	Habitats	Body length (cm)	Weight (g)
Cirrhinus molitorella	4	Herbivorous	Lower middle	17.75 ± 1.93	131.73 ± 56.74
Ctenopharyngodon idella	5	Herbivorous	Lower middle	27.49 ± 1.23	390.75 ± 25.62
Cyprinus carpio	4	Omnivorous	Lower middle	19.84 ± 3.62	216.31 ± 27.85
Squaliobarbus curriculus	4	Omnivorous	Midwater	20.82 ± 3.54	191.18 ± 93.98
Ptychidio jordani	5	Omnivorous	Demersal	14.67 ± 3.42	75.73 ± 67.44
Carassius auratus	5	Omnivorous	Demersal	21.51 ± 2.69	255.96 ± 49.58
Silurus asotus	5	Carnivorous	Lower middle	27.92 ± 3.58	326.91 ± 78.16
Tachysurus fulvidraco	5	Carnivorous	Demersal	11.14 ± 2.00	16.24 ± 4.89
Channa argus	4	Carnivorous	Demersal	24.33 ± 2.18	409.59 ± 74.31

and 71.55–110.61% for BDE 77, 70.86–130.17% for BDE 181, and 75.80–97.74% for BDE 205.

In this study, the standard internal method was used to detect the target compound, and the multipoint calibration curve was used for quantification. The correlation coefficients of the standard curves were greater than 0.99. The instrument limit of detection (LOD) and quantitative limit of quantitation (LOQ) was defined as the concentrations corresponding to 3 and 10 times the signal-to-noise ratio, respectively. The LODs and LOQs for the fish samples were 0.0001–0.0514 ng/g ww and 0.0002–0.3931 ng/g ww, respectively.

2.5. Evaluation Method. The assessment of human exposure to HOPs is usually expressed in terms of estimated daily intake (EDI), expressed as follows [17]:

$$\mathrm{EDI} = \frac{\mathrm{PR} \times \mathrm{C}}{\mathrm{BW}}, \tag{1}$$

where PR is the daily per capita intake of fish. In this paper, the per capita fish consumption in southern China is 59.3 g per person per day [18, 19]. C is the wet weight concentration of the target compound in fish muscle tissue (ng/g ww), and BW is the average body weight of local residents 60 kg (National Bureau of Statistics, 2019).

The hazard quotient (HQ) for human consumption of fish can be estimated from the acceptable daily intake (ADI) or reference intake (RfD) of PCBs, DDTs, HCHs, and BDE 99. HQ is usually determined by the ratio of EDI to ADI or RfD. If HQ is greater than 1, that is, EDI > ADI or RfD, there is a health risk, otherwise, there is no health risk [20] (Naqvi et al., 2020).

2.6. Statistical Analysis. The results were analyzed using descriptive statistical parameters such as mean value, standard deviation, and concentration range [21]. We used Origin 2021 to make the composition characteristic map of three kinds of different homologs in fish and the composition characteristic map of homologs in fish of different feeding habits and habitats.

3. Results and Discussion

3.1. Levels of HOPs in Fish Species. The detection rate of OCPs was 93.10–100%, and all selected OCP homologs were detectable in fish samples. Table 2 shows the measured concentrations for all samples. The levels of Σ_{18}OCPs in fish muscle tissue ranged from 8.94 to 43.32 ng/g ww, with a mean of 18.61 ng/g ww. Among them, *C. idella* had the highest average concentration, the concentration range was 13.94–43.32 ng/g ww and the average value was 23.81 ng/g ww. *C. molitorella* have the lowest average concentration, with the concentration range of 8.94–11.38 ng/g ww and the average value of 10.23 ng/g ww. Compared with those freshwater fish concentrations of Σ_{19}OCPs in Lake St Lucia, KwaZuluNatal (KZN), ranged from 74 to 510 ng/g lw [22], Σ_6OCPs in Oguta Lake, Nigeria, mean concentration was 236.9 ng/g (2021), Σ_6OCPs in Lake Tana, Ethiopia, ranged from ND to 5142.15 ng/g [23], concentrations of OCPs in the present study were in relatively low levels. Although the use of most pesticides has been restricted, their long half-life and long-distance mobility make them still detectable worldwide.

As can be seen from Table 3, the detection rate of PCBs ranged from 79.31 to 100% for all 18 PCB monomers selected for this study. Σ_{18}PCBs in fish muscle tissue ranged from 1.22 to 4.09 ng/g ww, with an average value of 1.94 ng/g ww. It was noteworthy that *T. fulvidraco* had the highest PCBs content in the total PCBs, which was 2.80 ng/g ww. The lowest concentration was found in *C. molitorella*, with a concentration range of 1.22–1.61 ng/g ww and an average value of 1.45 ng/g ww. The highest and lowest average contents of the 18 monomers analyzed were PCB 169 and PCB 18, with 0.51 ng/g ww and 0.00 ng/g ww, respectively. The concentration of Σ_{18}PCBs in the present study was moderate compared to the concentrations reported in other studies, in Lake Chapala, Mexico (Σ_{39}PCBs, 1.06–6.07 ng/g) [24] and Cook County IL, Chicago (11.8–505 ng/g ww) [25].

The detection rate of PBDEs ranged from 34.48% to 86.21% and the concentration ranged from ND to 0.13 ng/g ww, with an average value of 0.05 ng/g ww. The average concentrations of omnivorous *C. auratus* and carnivorous *T. fulvidraco* were the highest and lowest, respectively, with average values of 0.09 ng/g ww and 0.01 ng/g ww, respectively. The average content of BDE 154 and BDE 99 was the highest and lowest among the five PBDE monomers, with 0.02 ng/g ww and ND, respectively. Please see Table 4 for details. ΣPBDE content in this study is lower than Σ_6PBDEs in the Yellow River Delta, China (5.3–149 ng/g lw) [26], Σ_6PBDEs in Lake Geneva, France and Switzerland (0.54–2.60 ng/g ww) [27], and Σ_5PBDEs in Newfoundland, Canada (ND-368.27 ng/g) [28].

Table 2: Average concentration and detection rate of OCPs in fish in the Laibin section of the Hongshui River (ng/g ww).

Homologs	Detection rate (%)	*C. auratus*	*C. argus*	*S. curriculus*	*C. carpio*	*C. molitorella*	*T. fulvidraco*	*S.asotus*	*C. idella*	*P. jordani*
α-HCH	100	0.824 ± 0.082	1.212 ± 0.459	1.219 ± 0.396	1.011 ± 0.167	0.884 ± 0.228	0.237 ± 0.000	1.092 ± 0.709	0.950 ± 0.181	1.169 ± 0.122
β-HCH	100	1.504 ± 0.070	2.982 ± 0.547	1.917 ± 0.445	2.050 ± 0.340	1.294 ± 0.393	3.964 ± 0.000	3.475 ± 1.251	1.962 ± 0.347	2.018 ± 0.800
γ-HCH	100	0.600 ± 0.267	1.121 ± 0.468	0.419 ± 0.125	0.303 ± 0.127	0.469 ± 0.166	2.133 ± 0.000	0.848 ± 0.397	0.312 ± 0.194	0.762 ± 0.014
δ-HCH	100	1.552 ± 0.299	2.262 ± 0.462	0.922 ± 0.400	0.770 ± 0.154	0.525 ± 0.117	2.026 ± 0.000	3.483 ± 1.406	0.738 ± 0.480	1.379 ± 0.392
ΣHCHs		1.120 ± 0.465	1.894 ± 0.911	1.120 ± 0.653	1.034 ± 0.675	0.793 ± 0.414	2.090 ± 0.000	2.224 ± 1.622	0.991 ± 0.687	1.332 ± 0.639
o,p'-DDE	100	0.432 ± 0.028	0.203 ± 0.064	0.269 ± 0.088	0.350 ± 0.060	0.303 ± 0.058	0.657 ± 0.000	0.099 ± 0.026	0.202 ± 0.055	0.318 ± 0.161
p,p'-DDE	100	0.434 ± 0.104	0.145 ± 0.038	1.059 ± 0.772	0.171 ± 0.127	0.127 ± 0.037	2.814 ± 0.000	0.459 ± 0.300	0.495 ± 0.242	2.506 ± 2.383
o,p'-DDD	100	0.133 ± 0.071	0.192 ± 0.121	0.197 ± 0.101	0.147 ± 0.064	0.076 ± 0.049	0.175 ± 0.000	0.072 ± 0.020	0.232 ± 0.147	0.197 ± 0.024
p,p'-DDD	100	0.088 ± 0.034	0.088 ± 0.019	0.178 ± 0.034	0.099 ± 0.030	0.078 ± 0.023	0.442 ± 0.000	0.197 ± 0.102	0.168 ± 0.055	0.181 ± 0.105
o,p'-DDT	100	0.087 ± 0.034	0.085 ± 0.020	0.168 ± 0.021	0.075 ± 0.009	0.047 ± 0.028	0.401 ± 0.000	0.184 ± 0.102	0.148 ± 0.036	0.180 ± 0.105
p,p'-DDT	100	0.261 ± 0.068	0.173 ± 0.088	0.236 ± 0.030	0.201 ± 0.069	0.180 ± 0.034	0.388 ± 0.000	0.317 ± 0.096	0.143 ± 0.072	0.179 ± 0.154
ΣDDTs		0.239 ± 0.161	0.148 ± 0.083	0.351 ± 0.452	0.174 ± 0.114	0.135 ± 0.095	0.813 ± 0.000	0.221 ± 0.194	0.231 ± 0.174	0.594 ± 1.301
Heptachlor	100	1.300 ± 0.081	1.199 ± 0.159	1.441 ± 0.327	1.203 ± 0.235	1.249 ± 0.068	1.098 ± 0.000	0.759 ± 0.107	0.986 ± 0.266	1.405 ± 0.166
Aldrin	96.55	0.380 ± 0.031	0.213 ± 0.077	0.487 ± 0.035	0.174 ± 0.048	0.324 ± 0.052	0.398 ± 0.000	0.108 ± 0.068	0.237 ± 0.225	0.138 ± 0.007
Endrin	100	6.872 ± 2.894	8.970 ± 5.075	5.487 ± 2.189	8.485 ± 2.822	2.770 ± 0.551	1.961 ± 0.000	3.284 ± 1.778	13.381 ± 10.982	5.957 ± 4.137
Dieldrin	100	0.089 ± 0.076	0.036 ± 0.016	0.036 ± 0.010	0.033 ± 0.012	0.025 ± 0.006	0.038 ± 0.000	0.014 ± 0.008	0.056 ± 0.024	0.047 ± 0.032
MXC	100	1.200 ± 0.663	1.035 ± 0.251	1.036 ± 0.107	1.067 ± 0.352	0.738 ± 0.349	4.895 ± 0.000	1.175 ± 0.333	0.905 ± 0.232	1.216 ± 0.386
Heptachlor epoxide	93.10	0.002 ± 0.002	0.003 ± 0.001	0.005 ± 0.005	0.007 ± 0.003	0.003 ± 0.001	0.006 ± 0.000	0.005 ± 0.002	0.010 ± 0.009	0.003 ± 0.002
Endosulfan I	100	0.095 ± 0.017	0.083 ± 0.019	0.094 ± 0.050	0.126 ± 0.136	0.043 ± 0.019	0.104 ± 0.000	0.069 ± 0.048	0.088 ± 0.043	0.125 ± 0.088
Endosulfan II	96.55	0.048 ± 0.048	0.036 ± 0.011	0.049 ± 0.025	0.061 ± 0.020	0.042 ± 0.025	0.125 ± 0.000	0.078 ± 0.028	0.161 ± 0.060	0.076 ± 0.004
Benzoepin sulfate	100	0.064 ± 0.007	0.042 ± 0.009	0.049 ± 0.008	0.033 ± 0.016	0.033 ± 0.011	0.025 ± 0.000	0.041 ± 0.006	0.055 ± 0.032	0.095 ± 0.065
Endrin-ketone	100	0.032 ± 0.003	0.027 ± 0.012	0.029 ± 0.013	0.037 ± 0.018	0.013 ± 0.005	0.091 ± 0.000	0.019 ± 0.007	0.041 ± 0.017	0.017 ± 0.009
Endrin-aldehyde	100	1.983 ± 0.864	1.814 ± 0.670	1.957 ± 0.441	1.403 ± 0.803	1.006 ± 0.389	1.454 ± 0.000	1.325 ± 0.512	2.537 ± 2.340	2.659 ± 0.384

Table 3: Average concentration and detection rate of PCBs in fish in Laibin section of the Hongshui River (ng/g ww).

Homologs	Detection rate (%)	*C. auratus*	*C. argus*	*S. curriculus*	*C. carpio*	*C. molitorella*	*T. fulvidraco*	*S.asotus*	*C. idella*	*P. jordani*
PCB 3	100	0.024 ± 0.000	0.022 ± 0.006	0.033 ± 0.005	0.093 ± 0.088	0.037 ± 0.009	0.032 ± 0.000	0.022 ± 0.001	0.025 ± 0.006	0.036 ± 0.007
PCB 18	79.31	0.002 ± 0.001	0.003 ± 0.002	0.001 ± 0.001	0.001 ± 0.002	0.002 ± 0.001	0.002 ± 0.000	0.001 ± 0.001	0.002 ± 0.001	0.001 ± 0.001
PCB 19	89.66	0.002 ± 0.002	0.023 ± 0.026	0.022 ± 0.018	0.008 ± 0.008	0.010 ± 0.009	0.037 ± 0.000	0.022 ± 0.015	0.017 ± 0.004	0.016 ± 0.007
PCB 77	100	0.051 ± 0.008	0.070 ± 0.011	0.117 ± 0.011	0.067 ± 0.009	0.062 ± 0.007	0.003 ± 0.000	0.031 ± 0.010	0.058 ± 0.045	0.107 ± 0.006
PCB 81	100	0.007 ± 0.002	0.028 ± 0.015	0.016 ± 0.009	0.018 ± 0.006	0.025 ± 0.009	0.024 ± 0.000	0.025 ± 0.008	0.023 ± 0.025	0.030 ± 0.001
PCB 114	93.10	0.021 ± 0.013	0.015 ± 0.008	0.043 ± 0.008	0.009 ± 0.006	0.007 ± 0.005	0.043 ± 0.000	0.028 ± 0.018	0.017 ± 0.010	0.003 ± 0.003
PCB 118	96.55	0.051 ± 0.028	0.038 ± 0.009	0.074 ± 0.045	0.032 ± 0.009	0.021 ± 0.017	0.035 ± 0.000	0.034 ± 0.022	0.022 ± 0.014	0.068 ± 0.037
PCB 123	89.66	0.162 ± 0.063	0.011 ± 0.007	0.024 ± 0.010	0.015 ± 0.005	0.007 ± 0.008	0.104 ± 0.000	0.063 ± 0.022	0.058 ± 0.032	0.078 ± 0.053
PCB 126	93.10	0.127 ± 0.108	0.127 ± 0.053	0.059 ± 0.056	0.076 ± 0.028	0.064 ± 0.058	0.094 ± 0.000	0.037 ± 0.014	0.057 ± 0.035	0.084 ± 0.074
PCB 128	100	0.065 ± 0.017	0.044 ± 0.010	0.046 ± 0.010	0.025 ± 0.013	0.033 ± 0.007	0.013 ± 0.000	0.026 ± 0.011	0.029 ± 0.010	0.051 ± 0.025
PCB 138	100	0.140 ± 0.095	0.112 ± 0.035	0.273 ± 0.182	0.018 ± 0.010	0.018 ± 0.007	0.127 ± 0.000	0.144 ± 0.071	0.026 ± 0.024	0.241 ± 0.237
PCB 153	100	0.271 ± 0.183	0.188 ± 0.052	0.487 ± 0.360	0.057 ± 0.009	0.071 ± 0.010	0.213 ± 0.000	0.297 ± 0.152	0.077 ± 0.068	0.420 ± 0.362
PCB 169	100	0.847 ± 0.075	0.412 ± 0.094	0.438 ± 0.148	0.794 ± 0.106	0.398 ± 0.123	1.194 ± 0.000	0.398 ± 0.095	0.392 ± 0.062	0.587 ± 0.352
PCB 170	100	0.420 ± 0.055	0.497 ± 0.075	0.512 ± 0.081	0.472 ± 0.051	0.535 ± 0.116	0.788 ± 0.000	0.308 ± 0.063	0.438 ± 0.099	0.505 ± 0.134
PCB 180	96.55	0.062 ± 0.022	0.053 ± 0.018	0.071 ± 0.043	0.037 ± 0.018	0.020 ± 0.011	ND	0.041 ± 0.015	0.030 ± 0.014	0.038 ± 0.034
PCB 187	100	0.082 ± 0.058	0.075 ± 0.012	0.127 ± 0.113	0.037 ± 0.015	0.018 ± 0.010	0.066 ± 0.000	0.121 ± 0.050	0.045 ± 0.024	0.135 ± 0.106
PCB 208	96.55	0.019 ± 0.000	0.041 ± 0.021	0.045 ± 0.016	0.035 ± 0.013	0.012 ± 0.011	0.023 ± 0.000	0.028 ± 0.007	0.076 ± 0.027	0.032 ± 0.017
PCB 209	96.55	0.066 ± 0.009	0.135 ± 0.025	0.324 ± 0.264	0.108 ± 0.018	0.064 ± 0.037	ND	0.086 ± 0.012	0.153 ± 0.008	0.160 ± 0.036

Note: ND means not detected.

Table 4: Average concentration and detection rate of PBDEs in fish in Laibin section of the Hongshui River (ng/g ww).

Homologs	Detection rate (%)	*C. auratus*	*C. argus*	*S. curriculus*	*C. carpio*	*C. molitorella*	*T. fulvidraco*	*S.asotus*	*C. idella*	*P. jordani*
BDE 28	68.97	0.028 ± 0.026	0.006 ± 0.002	0.012 ± 0.009	ND	0.002 ± 0.001	0.004 ± 0.000	0.002 ± 0.002	0.008 ± 0.012	0.002 ± 0.000
BDE 99	34.48	ND	0.001 ± 0.001	ND	0.001 ± 0.001	0.001 ± 0.001	ND	0.002 ± 0.001	0.001 ± 0.002	ND
BDE 100	86.21	0.002 ± 0.000	0.003 ± 0.001	0.003 ± 0.001	0.001 ± 0.000	0.001 ± 0.001	0.001 ± 0.000	0.001 ± 0.001	0.009 ± 0.007	0.004 ± 0.002
BDE 154	75.86	0.016 ± 0.015	0.031 ± 0.005	0.011 ± 0.016	0.014 ± 0.010	0.013 ± 0.015	ND	0.028 ± 0.017	0.047 ± 0.008	0.051 ± 0.051
BDE 208	51.72	0.046 ± 0.026	0.004 ± 0.007	0.006 ± 0.009	0.006 ± 0.006	0.010 ± 0.011	ND	0.005 ± 0.005	0.005 ± 0.005	0.008 ± 0.008

Note: ND means not detected.

Currently, China attaches great importance to the emission situation of HOPs and has adopted emission reduction, waste incineration shutdown, and treatment process improvement to reduce the emission of HOPs. However, the late start and the imperfect monitoring system have resulted in residues of HOPs still being detected in various locations. Ten kilometers upstream from the sampling site, there is a large area of industrial and agricultural land, which is flat and far from living areas and suitable for industrial and agricultural industries. OCPs are likely to originate mainly from residues of pesticide use on agricultural land. PCBs accumulated in freshwater originate mainly from their industrial application in paints, insulation additives, and dielectric fluids (2012). There are metal, paper, and timber mills upstream of the Hongshui River, and the wastewater discharge may lead to residual PCBs in the Hongshui River. In addition, historical residues may also be a contributing factor. The generally low concentration may be related to the limited use and short consumption history of PCBs in China [29]. PBDEs, an organic bromine compound, are widely used in the electronics, clothing, and furniture industries and help to reduce the flammability of products containing bromine [30]. The increasing number of flame retardants being put on the market to sustain people's productive lives may explain the growing concern over PBDEs pollution in recent years [31].

3.2. Composition Characteristics and Sources of Target Pollutants

3.2.1. OCPs. The composition of OCPs in muscle tissues of different fish species is shown in Figure 2(a). The composition patterns of OCPs in the nine fish species were similar, with HCHs and endrin being the main contributing contaminants, followed by DDTs, heptachlor, endrin-aldehyde, MXC, and aldrin, with other OCPs contributing less significantly. The total contribution of HCHs and endrin ranged from 44.04% to 75.48%. The similar composition pattern shows that OCPs contained in nine fish species have certain similarity or similar origin.

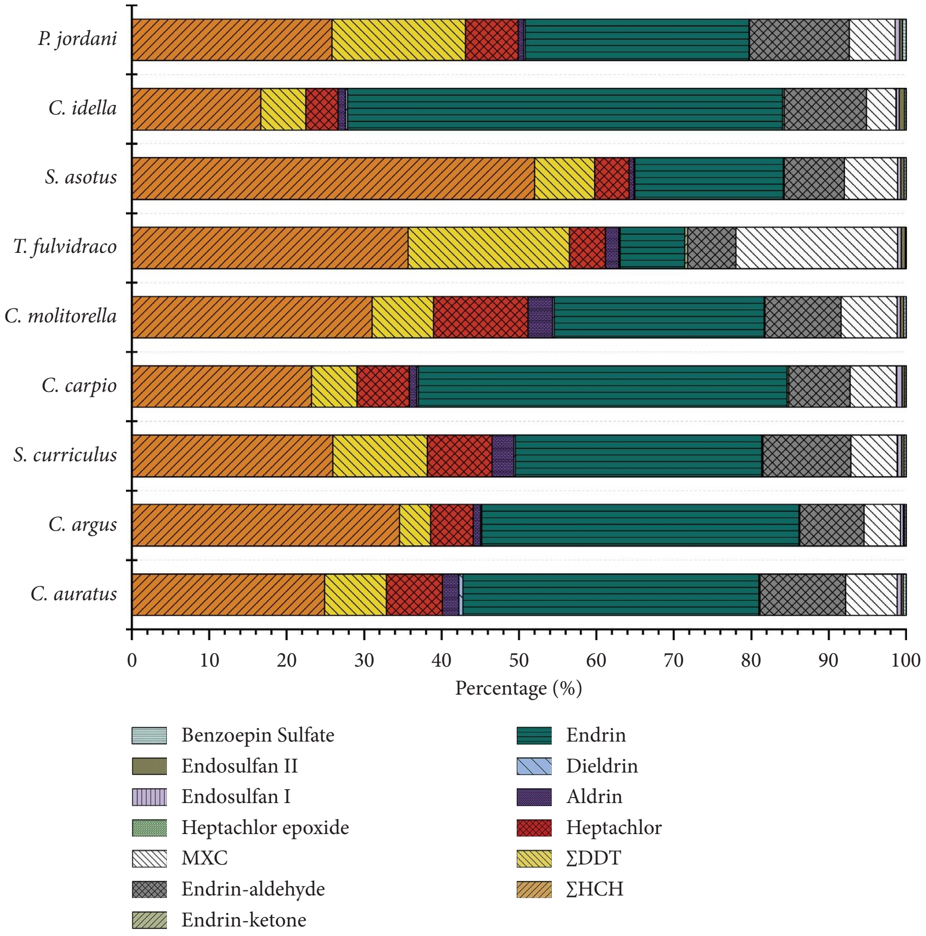
The main components of HCHs are α-HCH, β-HCH, γ-HCH, and δ-HCH. The highest percentage of HCHs in this study was β-HCH (Figure 3). There are the following two main sources of HCHs in the environment: industrial HCH and Lindane, which is still used as a broad-spectrum pesticide in some areas [32]. Biodegradation of β-HCH in environmental media is challenging; in contrast, α-HCH and γ-HCH are highly volatile and can be transported over long distances in the environment. Endrin is a highly toxic pesticide. Although endrin has never been produced on a large scale in China, its residues have been detected throughout the country [33]. Therefore, from the analysis of the compositional characteristics of OCPs, it could still be concluded that the main sources of OCPs in the region are historical residues and emissions as by-products from the manufacture and use of various industrial and agricultural chemicals [34].

3.2.2. PCBs. In this study, 18 kinds of PCB monomers were classified according to the number of chlorine atoms, mainly including tetra-CBs, penta-CBs, hexa-CBs, hepta-CBs, and deca-CBs. From Figure 2(b), it could be seen that hexa-CBs made the most significant contribution, accounting for 64.89% of the total. The second was penta-CBs, accounting for 10.78% of the total. The percentages of deca-CBs, tetra-CBs, and hepta-CBs in the total were similar at 8.23%, 8.15%, and 7.95%, respectively.

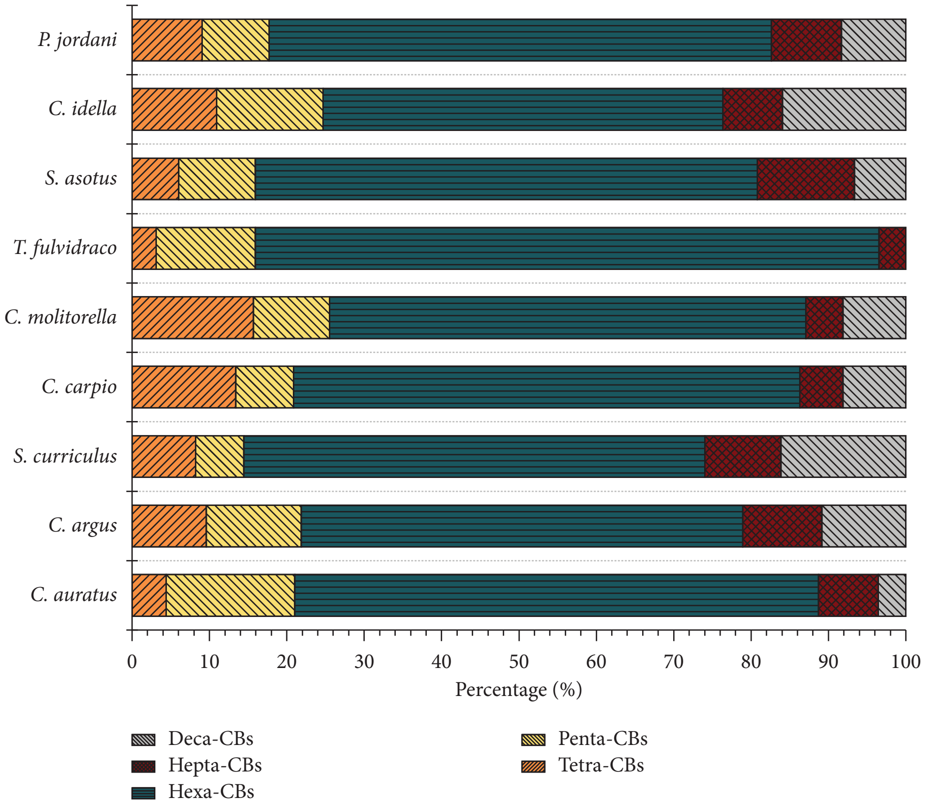
It is generally accepted that higher chlorinated congeners of PCBs accumulate more in organisms than lower chlorinated congeners due to their greater lipophilicity. In contrast, the content of higher chlorinated PCB congeners, such as deca-CBs, are very low, probably because these chlorinated compounds are too large to pass through the cell membranes of the organisms, resulting in low levels of accumulation [35]. This is consistent with the results of studies on PCBs in food fish from the Negro River Basin, Patagonia, Argentina [36], where Ondarzav noted that PCB 138 and PCB 153 are more resistant to metabolism and slower to be eliminated, and that the inherent high lipophilicity, stability, and persistence of hexa-CBs lead to their easier accumulation in fish. PCBs are industrial products and no natural sources, so it is inferred that the PCBs detected in the area are mainly from upstream industrial activities and historical residues.

3.2.3. PBDEs. The distribution of PBDEs in nine species of fish was different (Figure 2(c)). There was almost no BDE 99 in the *P. jordani*, and BDE 154 had the highest contribution rate; there were five kinds of PBDEs in *C. idella*, BDE 154 had the highest contribution rate and BDE 99 had the lowest contribution rate; there were five kinds of PBDEs in *S. asotus*, BDE 154 had the highest contribution rate and BDE 100 had the lowest contribution rate; there were almost no BDE 99, BDE 154 and BDE 208 in *T. fulvidraco*, and the contribution rate of BDE 28 was higher than that of BDE 100; there were five kinds of PBDEs in *C. molitorella*, BDE 154 had the highest contribution rate, followed by BDE 208 and had little difference with BDE 154, BDE 99 and BDE 100 had little difference; there was almost no BDE 28 in *C. carpio*, BDE 154 had the highest contribution rate, and BDE 99 and BDE 100 had a small contribution rate and little difference; there was almost no BDE 99 in *S. curriculus*, and BDE 28 had the highest contribution rate; there were five kinds of PBDEs in *C. argus*, BDE 154 had the highest contribution rate and BDE 99 had the lowest contribution rate; there was almost no BDE 99 in *C. auratus*, and BDE 208 had the highest contribution rate.

It could be seen from Figure 2(c) that the composition characteristics of PBDEs homologs in the nine fish species are different. This is similar to the research conclusion of Zhou et al. in Dianshan Lake, Shanghai, China [37]. This may be due to the different habitats of the fish and the different metabolic and biotransformation mechanisms of each fish [38]. The main sources of PBDEs are local wastewater discharges and wastewater from plastic and textile product factories [39]. Therefore, it was tentatively determined that the residues of PBDEs in the water

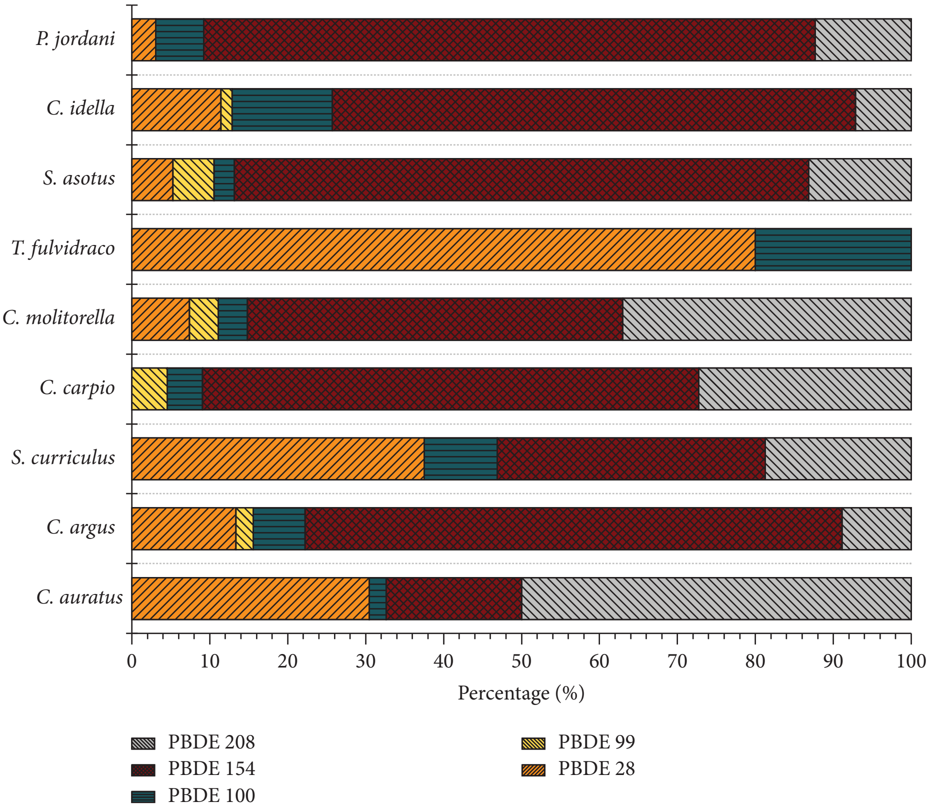


(a)



(b)

FIGURE 2: Continued.



(c)

Figure 2: (a) Characterization of the composition of OCPs homologs in fish from the Laibin section of the Hongshui River. (b) Characterization of the composition of PCBs homologs in fish from the Laibin section of the Hongshui River. (c) Characterization of the composition of PBDEs homologs in fish from the Laibin section of the Hongshui River.

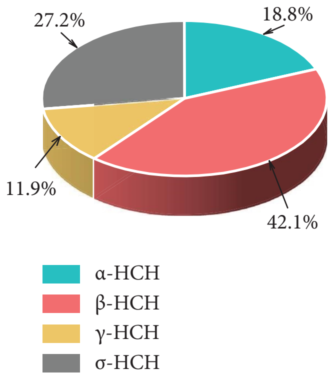


Figure 3: The percentages of major components of the four HCHs in this study.

environment of the area are mainly of industrial origin. However, the distribution characteristics of the various PBDEs homologs are different due to the fish themselves.

3.3. Compositional Characteristics of Pollutants in Fish of Different Feeding Habits and Habitats.

In this study, fish of three different feeding habits and two different habitats were analyzed separately. In order to ensure the rationality of the data, *C. molitorella* and *C. idella* were selected for the analysis of herbivorous fish. The omnivorous fish that were selected for the analysis were *C. carpio* and *S. curriculus*,. The carnivorous fish that were selected for the analysis were *T. fulvidraco* and *C. argus*. Selection was based on good recovery rates and preference for a high number of strips. As only one midwater fish species was collected, midwater fish were not involved in the analysis of the different habitat. The remaining eight species were divided into four lower midwater fish and four demersal fish.

As can be seen in Figures 4(a) and 4(b), the three HOPs (OCPs, PCBs, and PBDEs) have different compositional characteristics in fish of different diets and inhabiting water layers. According to the analysis of three different feeding habits (carnivorous, omnivorous, and herbivorous), the order of OCPs and PCBs content was carnivorous > omnivorous > herbivorous; the order of PBDEs content was omnivorous > herbivorous > carnivorous. The composition of the three contaminants in fish from the two different habitats was characterized by demersal > lower midwater.

In a study of OCPs in fish from Lake Koka, Ethiopia [40], it was noted that OCPs in the water bodies are more efficiently transferred to phytoplankton, which herbivorous fish

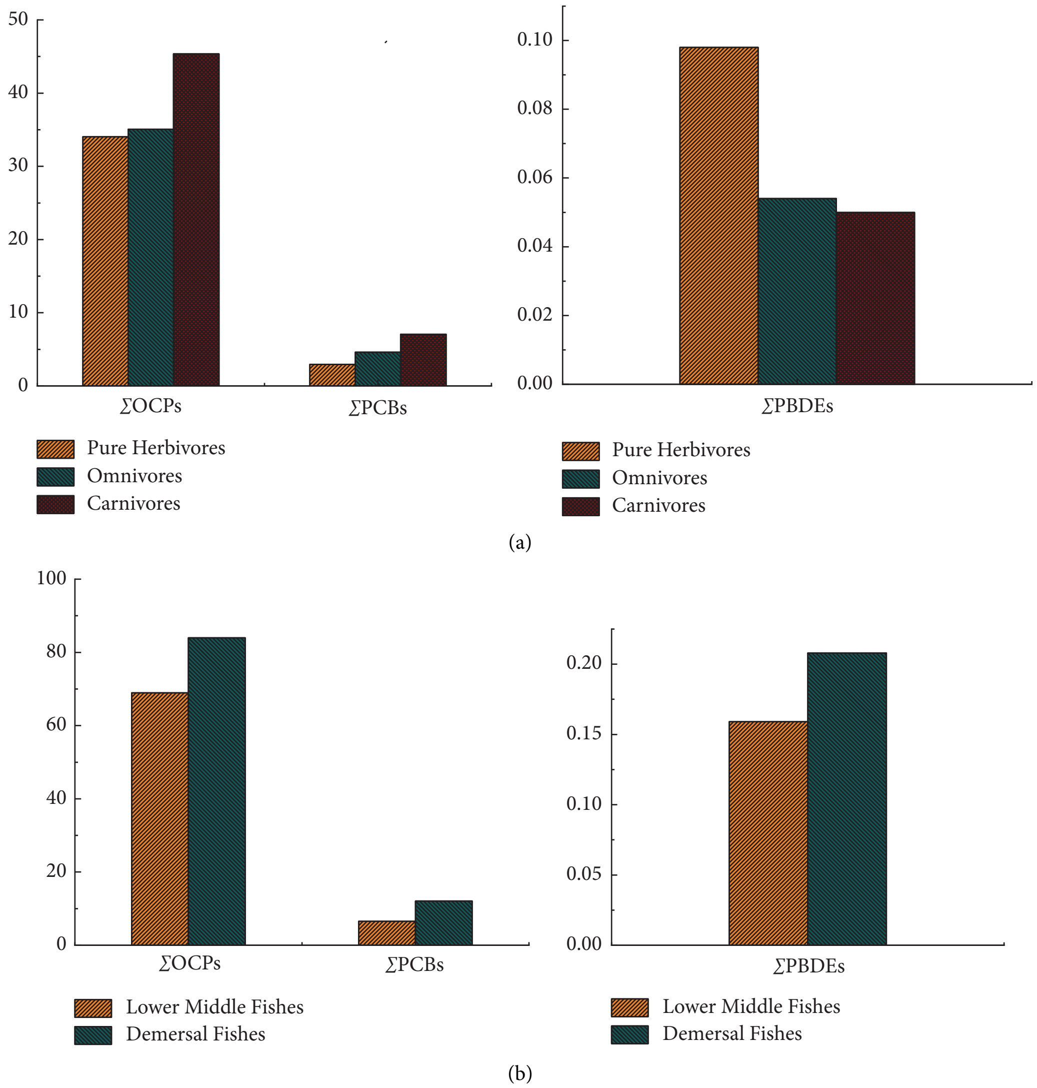


Figure 4: Characteristics of the homologue composition of fishes of different feeding and habitat in the Laibin of the Hongshui River.

can take in. In contrast, carnivorous fish that feed on these fish accumulate more degraded forms of these contaminants, which are more persistent, resulting in higher levels of OCPs accumulated in carnivorous fish. OCPs and PCBs have been found in many studies to have characteristics that are transmitted in the food chain species because of their high octanol-water partition coefficient (K_{ow}) [18, 41, 42]. It makes them more concentrated in carnivorous fish with higher trophic level. This is consistent with the findings in this study. However, in some studies, it is found that there is no biomagnification feature of PBDEs, so the enrichment degree of PBDEs is more dependent on the physicochemical properties and habitat [43, 44].

In aquatic ecosystem, halogenated persistent organic pollutants are usually attached to sediments, making it easier for benthic fish to absorb them into their bodies from their skin, gills, and feeding. As a result, demersal fish typically have higher levels of contaminants than other water-layer fish. However, many factors still influence the concentration of HOPs in fish, such as the age of the fish, habitat environment, and the stability of pollutants. Therefore, more detailed studies are needed to analyze the characteristics of the composition of different pollutants in fish in different environments.

3.4. Risk for Fish Consumers. EDI_{50} and EDI_{90} were calculated based on the 50th and 90th concentrations of pollutants in fish, respectively, representing the average and maximum level of HOPs intake by Chinese residents through fish consumption. The results of the calculations are shown in Table 5. The maximum daily intake of the five compounds (except MXC) calculated in this study was far lower than the allowable daily intake (ADIs) recommended by the International Food and Health Organization (FAO/WHO) and the reference dose (RfD, USEPA 2000a) of PCBs set by the United States Environmental Protection Agency [26, 45–47]. EDI of each compound was lower than the corresponding allowable daily intake (ADIs), that is, HQ > 1. At present, there are no national regulations or standards for

Table 5: Daily intake of HOPs by Chinese residents consuming fish from the Laibin section of the Hongshui River (ng/kg bw/d).

	EDI_{50}	EDI_{90}	RfD (μg/kg bw/d)
ΣPCBs	10.209	14.077	0.02
ΣDDTs	1.199	2.456	0.5
ΣHCHs	4.316	8.644	0.3
BDE 99	0.000	0.003	0.1
ΣPBDEs	0.033	0.091	—
MXC	1.028	1.498	—

MXC. This means that the estimated daily intakes in the nine fish samples in this experiment were much smaller than the specified reference measurements. To sum up, the daily consumption of fish in the Laibin section of Hongshui River may not pose a health risk to local residents for PCBs, DDTs, HCHs, and BDE 99 compounds. However, there are still many persistent halogenated organic pollutants not involved in this study, so the risk of fish consumption in this area still needs to be further studied.

4. Conclusion

In this study, three types of persistent halogenated organic pollutants (OCPs, PCBs, and PBDEs) could be detected. Endrin dominated in OCPs; hexachlorobiphenyl dominated in PCBs; BDE 154 dominated in PBDEs, but its composition characteristics were different in nine species of fish. From the perspective of different feeding habits, OCPs and PCBs had the largest residues in carnivorous fish, and PBDEs had the largest residues in herbivorous fish. From the perspective of different habitats, the residues of OCPs, PCBs, and PBDEs in the demersal fish were the largest. There were many factors that affect the enrichment of HOPs.

Based on the test data and analysis in this paper, the following suggestions can be made: (1) Build a comprehensive environmental monitoring system, increase the number of environmental monitoring points, conduct long-term monitoring of pollutants and take corresponding treatment measures immediately if the standards are found to be exceeded. (2) Upgrade environmental monitoring technology, step up talent building and keep up with changes in technology. (3) Vigorously carry out pollution prevention and control publicity to enhance public awareness of pollutant emissions and control, so that the public can properly understand the hazards of pollutant emissions.

Data Availability

Data will be made available on request.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

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Supplementary Materials

The additional data are included in Supplementary file. (*Supplementary Materials*)

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