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Human health risk of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in edible fish from Huairou Reservoir and Gaobeidian Lake in Beijing, China

Xuemei Li [a,c], Yiping Gan [b], Xiangping Yang [b], Jun Zhou [b], Jiayin Dai [a], Muqi Xu [a,*]

[a] *Key Laboratory of Animal Ecology and Conservation Biology, Institute of Zoology, Chinese Academy of Sciences, Beijing 100080, China*
[b] *Beijing Municipal Drainage Corporation, Beijing 100022, China*
[c] *Graduate School of the Chinese Academy of Sciences, Beijing 100039, China*

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Abstract

Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were measured by a gas chromatography–mass spectometry (GC–MS) in some edible fish from Huairou Reservoir and Gaobeidian Lake in Beijing, China. The concentrations of OCPs and PCBs were higher in fish (except *Misgurnus anguillicaudatus*) from Gaobeidian Lake than in those from Huairou Reservoir. The average concentrations of HCHs, DDTs and PCBs in fish ranged from 0.58 to 8.48, 7.54 to 88.3, below limit of detection (nd) to 22.7 ng/g wet weight, respectively. β-HCH, *p,p*′-DDE and PCB153 were the most abundant compounds among HCHs, DDTs and PCBs, respectively. Risk assessments of OCPs and PCBs for humans were estimated according to three different guidelines. The results indicated that fish intake would not pose a health risk to humans with a consumption of 7.4 ± 8.6 g/person day according to the acceptable daily intake (ADI) and minimal risk level (MRL) in the two environments. However, the hazardous ratio of the 95th percentile for PCBs in fish from Gaobeidian Lake exceeded 1, which suggested that daily exposure to PCBs had a lifetime cancer risk of greater than 1 in 1,000,000.

Keywords: Composition; POPs; HCHs; DDTs; Potential risk

1. Introduction

The global environment is contaminated by persistent organic pollutants (POPs). The contamination of POPs is a significant health problem because POPs can be accumulated and magnified through the food web or food chain, and then can cause several adverse effects to human health and wildlife survival. Many examples of accidental contamination of POPs have occurred (Binelli & Provini, 2004; Chen, Luo, Wong, & Chen, 1982; Kannan, Tanabe, Ramesh, Subramanian, & Tatsukawa, 1992; Rappe et al., 1987) and the risk assessment of POPs in food for human health is important and necessary. Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) are two principal pollutants. In China, massive amounts of OCPs were used, especially in the 1970s and early 1980s. Although the usage for agriculture of DDTs and HCHs has been banned since 1980s, DDTs are still being used in low amounts to control certain insects in tropical and subtropical countries, including China (UNEP Regional Report, 2002).

Humans take up POPs through skin absorption, respiration and ingestion of contaminated food. Skin absorption and respiration are not the main route. Some researches have confirmed that more than 90% of contaminants come from food (Fürst, Fürst, & Groebel, 1990). Among all foods, fish is one of the main sources of contaminants although fish products account only for about 10% of diet (Alcock, Behnisch, Jones, & Hagenmaier, 1998; Harrison et al., 1998) or less. POPs in fish from some areas were detected to assess the risk for human health (Binelli &

* Corresponding author. Tel.: +86 10 6480 7169; fax: +86 10 6480 7099.
E-mail address: xumq@ioz.ac.an (X. Li).

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Provini, 2004; Jiang et al., 2005; Yang, Matsuda, Kawano, & Wakimoto, 2006). With the banning of massive usage and production of the compounds, the residual levels in foodstuffs have decreased significantly. However, there is not sufficient information on OCPs and PCBs in edible fish from some freshwaters in China. Most freshwater fish consumed by humans come from some local lakes or reservoirs which are more polluted by POPs because of the slow water cycling.

Huairou Reservoir, located in the northeast of Beijing, China, is one of the main water resources for industry, agriculture and domestic use in Beijing. There are some agricultural activities around Huairou Reservoir. Compared with Huairou Reservoir, Gaobeidian Lake is the receptor of treated industrial wastewater (about 70%) and population-used wastewater (about 30%) by the Gaobeidian Wastewater Treatment Plant (WWTP), the biggest WWTP in Beijing, China, with a treatment capacity of one million tons of wastewater daily. The ecosystem of Gaobeidian Lake is much impacted by its in-flowing water from Gaobeidian WWTP. So, Huairou Reservoir and Gaobeidian Lake are two typical bodies of freshwater affected by different contamination sources. Moreover, a lot of fish from these two waters are consumed by the local or non-local population. Therefore, it is very important to clarify the status of POPs in fish from the two waters, and especially in those that are popular with the local population. The present study will provide more information on the residues of OCPs and PCBs in fish from Huairou Reservoir and Gaobeidian Lake. The risk assessment of POPs for human health is also considered in detail.

2. Materials and methods

2.1. Sample collection

Huairou Reservoir and Gaobeidian Lake were selected for study (Fig. 1). The volume of Huairou Reservoir is approximately 9670×10^4 m^3 of water in most seasons. Gaobeidian Lake has a surface area of about 0.15 km^2. Its water source is mainly the water effluent of Gaobeidian WWTP. Water in Gaobeidian Lake is used as a coolant by the nearby Beijing Guohua Thermal Power Plant and then recycled to the aquatic environment at a higher temperature than it was originally. Water temperature in the lake is between 12 and 41 °C corresponding to seasonal changes, about 5–10 °C higher than the ambient temperature. The water in Gaobeidian Lake discharges into Tonghui River and finally into the largest estuaries, Bohai Bay in China. Fish samples, including *Carassius auratus*, *Misgurnus anguilicauatus* and *Pseudobagrus fulvidraco* were collected from Huairou Reservoir, and *Carassius auratus*, *Misgurnus anguilicauatus* and *Hemiculture leucisculture*s were collected from Gaobeidian Lake in May, 2006. The geographical position and related information of fish samples are listed in Table 1. The edible portions of fish were homogenized and frozen at −20 °C.

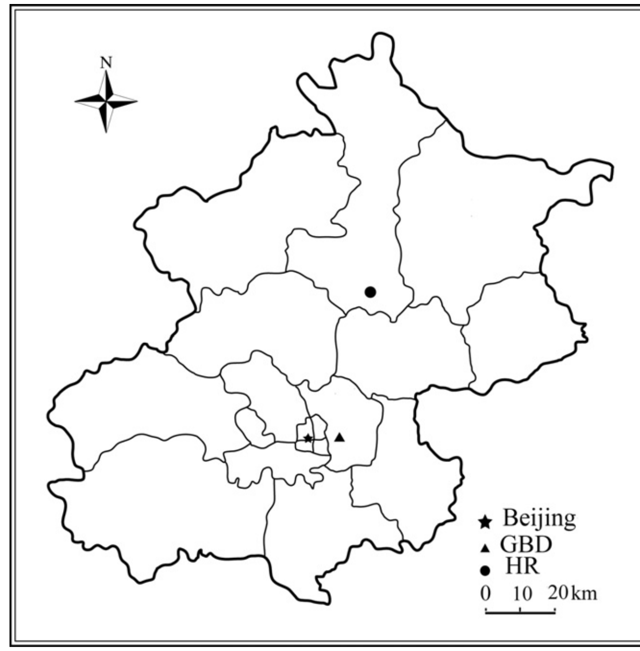

Fig. 1. Map of sampling locations.

2.2. Experimental protocol and quality assurance

Fish were extracted and cleaned up according to the method in the reference (Yang, Yao, Xu, Jiang, & Xin, 2007). The samples were analyzed for OCPs (α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p*′-DDD, *p,p*′-DDE, *p,p*′-DDT, and *o,p*′-DDT) and the six indicator PCBs (PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180). The results were obtained by Agilent 6890 with a ^{63}Ni electron capture detector (micro-ECD) with 30 m length × 0.25 mm

Table 1
Description of biological data of the fish samples

Sample site	Species	No.	Weight (g)	Length (cm)	Mean moisture content (%)	Mean lipid content (%)
Gaobeidian Lake	*C. auratus*	8	25.5–68.6	6.5–7.9	75.1–84.6	3.87
	M. anguillicaudatus	5	4.5–8.3	7.8–0.1	77.5–86.1	2.83
	*H. leucisculture*s	8	2.6–8.2	5.0–8.5	73.8–79.6	5.88
Huairou Reservoir	*C. auratus*	5	28.4–65.6	9.7–13.3	72.7–85.9	3.05
	M. anguillicaudatus	6	11.5–16.6	10.5–13.7	76.6–87.2	2.97
	P. fulvidraco	4	21.8–53.2	6.8–10.3	74.9–82.8	0.84

i.d × 0.25 μm film thickness used for separation of OCPs and PCBs. The main peaks in the selected samples were confirmed by a Finnigan GC–MS (PolarisQ, USA) using a 50 m DB-5 MS column with He carrier gas. Blanks and recovery were checked between every 8 and 12 samples. High, medium, and low levels of blank recoveries were analyzed during the experiment. The recoveries from samples of fish spiked with OCP and PCB standard solutions were also analyzed. One point standard solution was run every eight to 10 samples during the GC analysis to detect any deviation in the response of the system. Recoveries of OCPs and PCBs ranged from 73.8% to 98.1%. MDLs ranged from 0.08 ng/g for α-HCH to 0.34 ng/g for *p,p′*-DDT. The blank, recoveries and deviations were within the acceptable ranges.

3. Results and discussion

3.1. Concentrations of OCPs and PCBs

The concentrations of OCPs and PCBs in fish samples in the present study are summarized in Table 2. Relatively high concentrations of DDTs (from 7.54 ± 4.90 for *C. auratus* from Huairou Reservoir to 88.3 ± 68.0 ng/g wet wt for *H. leuciscultures* from Gaobeidian Lake) were found in fish from the two sampling locations, which showed that a large amount of DDT was used in past decades in China (Yang et al., 2006). There are no significant differences in concentrations of DDT between the two waters ($p > 0.05$). The highest concentrations of γ-HCH (2.04 ± 0.60 ng/g wet wt) and δ-HCH (0.99 ± 0.54 ng/g wet wt) were detected in *M. anguillicaudatus* from Huairou Reservoir, while the highest concentrations of the other compounds were found in *H. leuciscultures* from Gaobeidian Lake.

The concentrations of PCBs ranged from 6.43 ± 0.82 to 22.7 ± 15.4 ng/g wet wt in fish from Gaobeidian Lake; however, the concentrations of PCBs were below the limit of detection (LOD) in the fish from Huairou Reservoir. Low concentrations of PCBs were found previously in China and the results were similar to those in fish and shellfish (Nakata et al., 2002; Yang et al., 2006), which suggested lower background levels of PCBs in the Chinese environment than those in developed countries such as Japan and America (Yang et al., 2006). The fact is that only about 8000 tons of PCBs were produced in China (Jiang, Li, Chen, & Jin, 1997). The above results indicated that the bioaccumulation of OCPs and PCBs were species-specific for their different ecological characteristics, such as feeding habits, habitat (Yang et al., 2006) and compounds' physical and chemical properties.

The contaminants were in the following ranking order: DDTs > PCBs > HCHs and DDTs > HCHs > PCBs in fish from Gaobeidian Lake and Huairou Reservoir, respectively. The distributions of HCHs and PCBs were different in fish from the two environments and the concentrations of OCPs were higher in *M. anguillicaudatus* from Huairou Reservoir than in those from Gaobeidian Lake. These differences indicated that there were different sources in the two environments. Gaobeidian Lake was affected mainly by Gaobeidian WWTP, which treats about 70% of industrial wastewater and may bring a large amount of industrial contaminants such as PCBs. However, Huairou Reservoir is mainly affected by the ambient farmland and long-range atmospheric transport.

3.2. Compositions of OCPs and PCBs

The compositions of HCHs, DDTs and PCBs in fish from Gaobeidian Lake and Huairou Reservoir are shown in Fig. 2a–c, respectively. The compositions of PCBs in Fig. 2c are only shown for fish from Gaobeidian Lake

Table 2
The concentrations of OCPs and PCBs in fish from Gaobeidian Lake and Huairou Reservoir (ng/g)

Compounds	LOD	Gaobeidian Lake			Huairou Reservoir		
		C. auratus	*M. anguillicaudatus*	*H. leuciscultures*	*C. auratus*	*M. anguillicaudatus*	*P. fulvidraco*
α-HCH	0.08	1.05 ± 0.27	0.48 ± 0.14	1.83 ± 1.28	0.15 ± 0.11	1.71 ± 0.52	0.63 ± 0.14
β-HCH	0.19	4.36 ± 0.40	1.52 ± 0.90	7.40 ± 5.57	nd	1.67 ± 0.65	0.62 ± 0.41
γ-HCH	0.09	1.00 ± 0.43	0.61 ± 0.26	1.91 ± 1.01	0.19 ± 0.13	2.04 ± 0.60	0.68 ± 0.05
δ-HCH	0.13	0.61 ± 0.16	0.39 ± 0.08	0.96 ± 0.55	0.21 ± 0.02	0.99 ± 0.54	0.28 ± 0.04
HCHs		7.01 ± 2.89	2.99 ± 1.00	8.48 ± 5.61	0.58 ± 0.29	6.41 ± 2.14	2.21 ± 0.61
p,p′-DDD	0.14	3.67 ± 1.04	1.28 ± 0.78	8.76 ± 6.26	0.83 ± 0.49	5.83 ± 2.05	5.42 ± 4.36
p,p′-DDE	0.10	15.9 ± 5.15	11.6 ± 4.87	72.4 ± 47.1	6.42 ± 4.50	37.8 ± 12.6	28.5 ± 18.6
p,p′-DDT	0.34	2.39 ± 2.66	1.20 ± 0.94	3.24 ± 1.33	0.28 ± 0.17	0.54 ± 0.07	0.55 ± 0.20
o,p′-DDT	0.23	3.50 ± 1.47	1.30 ± 0.68	15.21 ± 8.23	nd	nd	nd
DDTs		25.4 ± 8.94	15.4 ± 6.26	88.3 ± 68.0	7.54 ± 4.90	44.2 ± 14.5	34.5 ± 22.9
PCB28	0.19	1.58 ± 0.64	0.49 ± 0.52	4.96 ± 3.46	nd	nd	nd
PCB52	0.18	0.74 ± 0.45	0.25 ± 0.23	4.60 ± 2.66	nd	nd	nd
PCB101	0.15	0.27 ± 0.22	0.31±0.35	4.26±5.54	nd	nd	nd
PCB153	0.14	4.47 ± 0.77	4.59 ± 0.44	8.71 ± 8.05	nd	nd	nd
PCB138	0.16	0.48 ± 0.18	0.48 ± 0.21	1.96 ± 1.34	nd	nd	nd
PCB180	0.11	0.42 ± 0.58	0.30 ± 0.37	4.57 ± 3.79	nd	nd	nd
PCBs		7.90 ± 1.98	6.43 ± 0.82	22.7 ± 15.4	nd	nd	nd

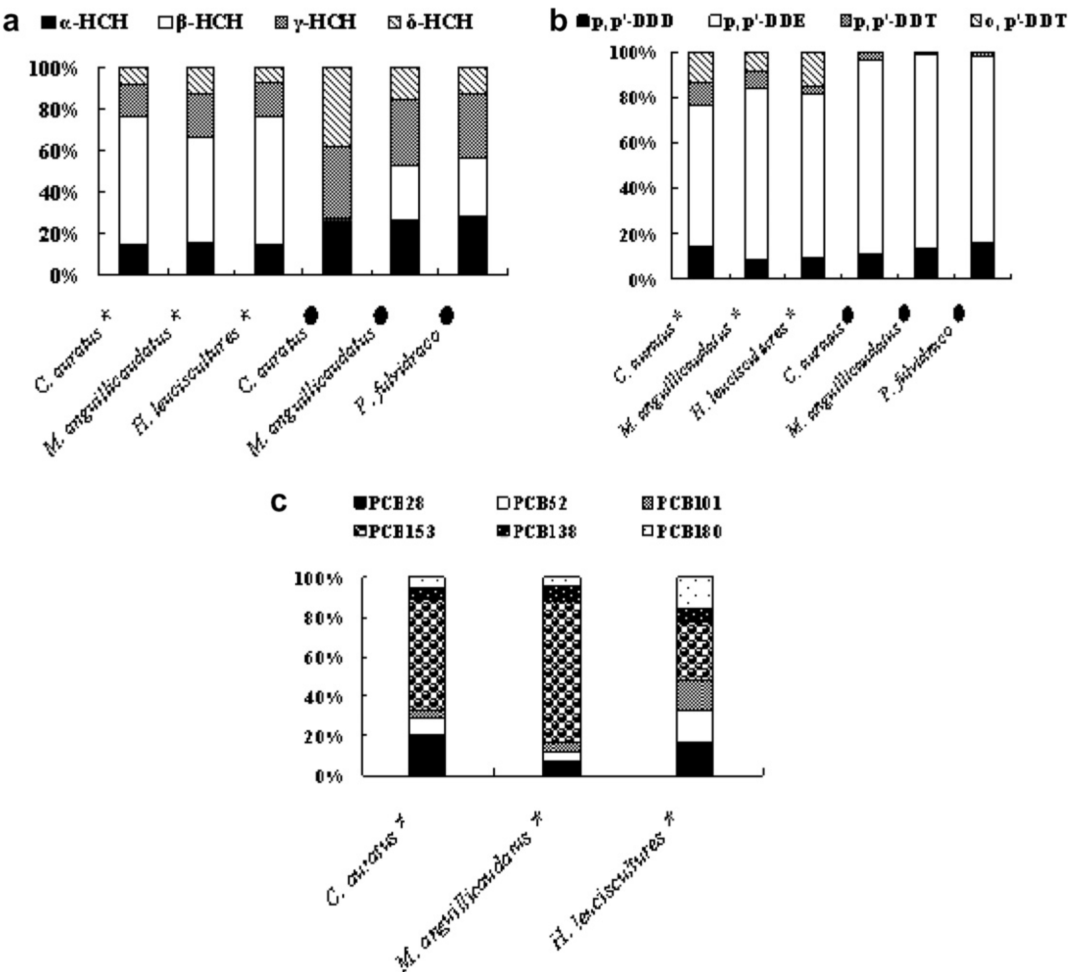

Fig. 2. The compositions of HCHs, DDTs and PCBs in fish from Gaobeidian Lake and Huairou Reservoir.

because the concentrations of PCBs were below the limit of detection in fish from Huairou Reservoir.

The compositions of HCH isomers were similar in fish from Gaobeidian Lake, while the compositions of HCH isomers were different in *C. auratus* and the other two fish species from Huairou Reservoir. The bioaccumulation of HCH isomers seems to be related to species and other physicochemical factors, such as feeding habit, water temperature, or structure of contaminants. β-HCH was dominant and the ratios of β-HCH/HCHs exceeded 0.50 in fish samples from Gaobeidian Lake. The results were similar to those in fish and shellfish (Yang et al., 2006), which suggested that Gaobeidian Lake was more influenced by technical mixtures of HCHs (α-HCH: 60–70%, β-HCH: 5–6%, γ-HCH: 12–14% and δ-HCH: 6%) than by lindane (γ-HCH more than 99%) in China. The reason for higher levels β-HCH might be that α-HCH can be isomerized into β-HCH, which is more persistent with respect to microbial degradation and also has a lower volatility (Andersen et al., 2001; Howard, 1992; Kouras, Zouboulis, Samara, & Kouimtzis, 1998; Rajendran, Imagawa, Tao, & Ramesh, 2005).

p,p′-DDE was the dominant compound in the present study. The highest concentration of *p,p′*-DDE (72.4 ± 47.1 ng/g wet wt) was found in *H. leucisculture* from Gaobeidian Lake. The ratio of *p,p′*-DDE/DDTs ranged from 0.53 to 0.93 in fish in the present study. DDT usually contains 75% of *p,p′*-DDT, 15% of *o,p′*-DDT, 5% of *p,p′*-DDE, and less than 5% of other species (Kim et al., 2002). *p,p′*-DDE was dominant because *p,p′*-DDE is the metabolite of *p,p′*-DDT and generally is detected as a main DDT isomer in fish. The ratio of *p,p′*-DDT /DDTs was very low in fish from Huairou Reservoir, which indicated that no new polluted source of DDT was discharged into Huairou Reservoir in recent years.

As for PCBs, PCB153 was dominant in fish from Gaobeidian Lake in the present study. The distribution of PCB congeners could be explained by the fact that higher chlorinated congeners accumulated more than did lower chlorinated congeners in most samples (Yang et al., 2006). It is easier for lower chlorinated PCBs to pass through the body's cell membranes than for higher chlorinated PCBs, which means that higher chlorinated PCBs have much stronger abilities of accumulation in fish samples (Bernes, 1998). A higher percentage of PCB153 (0.71) was found in *M. anguillicaudatus* than in *C. auratus* (0.56) and *H. leucisculture* (0.29). The reason for the difference could be that *M. anguillicaudatus* lives at the bottom of the lake and has more chance to be in contact with the sediment containing more highly chlorinated PCBs (Rajendran et al., 2005).

3.3. Risk assessment to humans

3.3.1. General

There are some guidelines supplied by different international organizations to evaluate the risk in fish for human health. Three guidelines were used to assess the risk of OCPs and PCBs in fish in the present study. Firstly, the risk assessment could be carried out through comparison of the determined levels of POPs in the present samples with the levels set by laws and guidelines. Generally, the acceptable daily intake (ADI) formulated by WHO was used to assess the risk, which can protect the world population and did not take into consideration other factors such as different eating habits and different consumption rates (Binelli & Provini, 2004). Secondly, minimal risk level (MRL) was formulated by US FDA and can be used to evaluate the chronic effects. Thirdly, potential risk can be evaluated by taking both chronic effects and carcinogenic effects into consideration. This method was proposed by the US EPA. In order to investigate the effect of fish in the diet, 153 local people were surveyed. A consumption of 7.4 ± 8.6 g/person day was used to determine ADI, MRL and potential risk in the present study.

3.3.2. ADI

For PCBs, the EU Directive 1999/788 establishes a maximum value of 200 ng/g lipids for the sum of seven main PCB congeners (PCB28, 52, 101, 118, 138, 153, 180) in some foodstuffs, including meat, eggs, poultry and related products. The standard did not include fish and fish products and PCB118 was not detected in fish samples in our study. Nevertheless, all the results for fish did not exceed the threshold. For DDTs and HCHs, the residual concentrations are proposed as 2000 and 5000 ng/g wet wt, respectively in China (Chen et al., 2002). The detected levels of PCBs and OCPs were below these guidelines (Table 2) in the present study. Related to the rate of consumption fish, ADIs were estimated by: average daily exposure = fish consumption × contaminants concentrations. The daily intake of OCPs and PCBs by humans through fish consumption are shown in Table 3. The estimated daily intake of DDTs and γ-HCH by humans were far below the ADI recommended by FAO/WHO, indicating that this intake would not pose a health risk when considering the ADI in the present study. A similar approach had also been adopted in previous studies (Yang et al., 2006).

3.3.3. MRL

The intake of contaminants and exposure time should be considered when evaluating the potential risk to human health of POPs (Binelli & Provini, 2004). The no observed adverse effect level/uncertainty factor (NOAEL/UF) approach is used by ATSDR to derive MRLs for hazardous substances. MRLs are obtained for acute (1–14 days), intermediate (14–364 days) and chronic (365 days and longer) exposure duration and for the oral and inhalation routes of exposure. In general, MRLs are based on the most sensitive substance-induced end-point, which is considered to be of relevance to humans. Serious health effects (such as irreparable damage to the liver or kidney, or birth defects) are not used as a basis for establishing MRLs. Based on the duration of exposure, different consumption values should be used (shown in Table 4). As compared with Table 4, the MRL of PCBs in fish from Gaobeidian Lake was 1.62 ng/kg day and did not exceeded 20 ng/kg day when using the average daily consumption of 7.4 g/person day to estimate the MRL of compounds (Table 3).

Table 3
Estimated daily intakes of OCPs and PCBs in fish by humans (ng) (average rates of consumption fish and weight were 7.4 g/ person day and 60 kg, respectively) in Beijing

Compounds	Average concentration (ng/g wet wt)		EDI (ng/kg day)		ADI (ng/kg day)
α-HCH	1.00[a]	0.90[b]	0.12[a]	0.11[b]	
β-HCH	3.98[a]	0.84[b]	0.49[a]	0.10[b]	
γ-HCH	1.05[a]	1.06[b]	0.13[a]	0.13[b]	8000
p,p′-DDT	2.21[a]	0.46[b]	0.27[a]	0.06[b]	20,000
HCHs	6.62[a]	3.35[b]	0.82[a]	0.41[b]	
DDTs	46.99[a]	29.38[b]	5.80[a]	3.62[b]	
PCBs	13.19[a]	0.22[b]	1.62[a]	nd[b]	

[a,b]Mean the values obtained in fish from Gaobeidian Lake and Huairou Reservoir, respectively.
nd represents target analytes with concentrations lower than limit of detection (LOD) and was treated as zero when calculating the mean values.

3.3.4. Potential risk

In order to estimate the potential risk to human health, percent probability associated with a particular point of departure was estimated from a plot of percent cumulative probability as a function of compound concentrations in fish (Olsen et al., 2003). Hazard ratios (HRs) were calculated and the detailed methods are described in other references (Jiang et al., 2005). In brief, HRs were estimated by dividing the average exposure by the benchmark

Table 4
Minimal risk level (MRL) for PCBs and OCPs by the Agency for Toxic Substances and Disease Registry (ATSDR) (1996) according to the duration of oral exposure

Compounds	Duration	MRL (ng/kg day)	Factor of uncertainty	Endpoint
α-HCH	Chr.	800	100	Hepatic
β-HCH	Acute	50,000	100	Neurol.
	Int.	600	300	Hepatic
γ-HCH	Acute	300	300	Develpo.
	Int.	10	1000	Immuno.
p,p′-DDT	Acute	500	1000	Develpo.
	Int.	500	100	Hepatic
PCBs	Int.	30	300	Neurol.
	Chr.	20	300	Immuno.

Table 5
National average exposure and benchmark concentrations for contaminants in fish

Compounds	Oral RfD	Cancer slope factor (mg/kg day)	Cancer benchmark concentrations (ng/kg day)	50th percentile measured concentrations and HRs (ng/g wet wt)		95th percentile measured concentrations and HRs (ng /g wet wt)	
DDTs	0.50	0.34	23.85	30.6[a]	0.16[a]	141.8[a]	0.73[a]
				19.4[b]	0.10[b]	117[b]	0.61[b]
PCBs	0.02	2.0	4.05	10.1[a]	0.31[a]	33.0[a]	1.01[a]

[a,b]Mean the values obtained in fish from Gaobeidian Lake and Huairou Reservoir, respectively.

concentrations (the benchmark concentration is derived by setting the cancer risk to 1,000,000 for lifetime exposure). HRs exceeding 1 indicated that there was potential risk to human health. Two HRs were estimated to assess the potential risk, in which one was on the 50th percentile exposure and the other was on the 95th percentile exposure (Jiang et al., 2005). Among OCPs, the 95th percentile concentration for DDTs was large: 142 and 117 ng/g wet wt in fish from Gaobeidian Lake and Huairou Reservoir, respectively. All of the concentrations of OCPs and PCBs were less than their oral RfD. Similar results were found in the Chinese coastal population (Jiang et al., 2005). For cancer assessment, the oral reference dose (RfD) and cancer benchmark concentrations for contaminants are shown in Table 5.

The 50th and 95th percentile of PCBs in fish from Huairou Reservoir were not estimated because the concentrations of PCBs in fish were below LOD. The HRs of the 50th and 95th percentile for OCPs did not exceed 1 although the concentrations were higher. The reason may be that the rate of consumption of fish was very low compared with the average rate of fish consumption (30.5 g/person day) in 1997 (Du, Zai, & Yang, 2004) and 105 ± 182 g/person day in China (Jiang et al., 2005). The results for cancer risks indicated that there was no potential risk for human health from OCPs. However, the 95th percentile of PCBs in fish from Gaobeidian Lake exceeded unity, which suggested that daily exposure to PCBs had a lifetime cancer risk of greater than 1 in 1,000,000.

4. Conclusions

Among OCPs and PCBs, DDT was the dominant contaminant in the present study. The accumulation and composition of OCPs and PCBs were different in the two environments. The accumulation and distribution patterns of OCPs and PCBs were consistent with the previous studies observed in fish from the other areas where PCBs were not the dominant compounds in China.

The risk assessments of OCPs and PCBs were estimated for human health through different limits or guidelines. The results indicated that consumption of fish from the two environments will not be risky to humans with a consumption of 7.4 ± 8.6 g/person day according to the acceptable daily intake (ADI) and minimal risk level (MRL) in the two environments, while some chronic and carcinogenic effects caused by PCBs may occur if consuming fish from Gaobeidian Lake. Based on the 50th and 95th percentile measured concentrations of DDTs, the thresholds of fish consumption rates were 13.1 and 8.42 g/person day, respectively in fish from Gaobeidian Lake, and 16.51 and 9.26 g/person day, respectively in fish from Huairou Reservoir. For PCBs, the thresholds of fish consumption rate were 9.44 and 7.19 g/person day, respectively, when taking the 50th and 95th percentile measured concentrations. Among the 153 people surveyed, there were about 22% of people consuming freshwater fish at more than 7.19 g/person day, which suggested that those with a high rate of fish consumption should be cautious.

The present survey is limited and other surveys on other possible contaminants such as polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs), polybrominated diphenyl ethers (PBDEs) and perfluorinated compounds (PFCs) are needed. Attention should also be paid to interactions and combined effects among contaminants.

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