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Health Risk Assessment of Organochlorine Contaminants in Fish from a Major Lake (Baiyangdian Lake) in North China

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Abstract Organochlorine contaminants (OCs) in fish were determined to evaluate the potential risk to humans consuming fish originating in Baiyangdian Lake, North China. Relatively low levels of PCBs, HCHs and DDTs were observed, with mean concentrations ranging from 0.28 to 3.28 ng/g, wet weight. Among various fish species tested, the highest burden of OCs was recorded in northern snakehead (7.39 ng/g, wet weight) and the lowest was in grass carp (2.04 ng/g, wet weight). The hazard ratios (HRs), based on noncancer risk were all less than 1.0, while the HRs based on cancer risk exceeded 1.0 only for PCBs based on the 90th percentile concentration.

Keywords Fish · Health risk assessment · Organochlorine contaminants · China

Organochlorine contaminants (OCs), such as organochlorine pesticides (OCPs), including hexachlorocyclohexanes (HCHs), the DDT complex (DDTs), and polychlorinated biphenyls (PCBs) represent a significant health problem, due to their bioaccumulation through the food chain and their highly toxic effects on wildlife and humans (Tanabe et al. 1994). In China, massive amounts of HCHs, DDTs and PCBs were used, especially in the 1970s and early 1980s. Although the open use of HCHs, DDTs and PCBs has been prohibited in China since 1980s, considerable residues still remain in all parts of the environment. Being highly lipophilic and persistent, they tend to accumulate in the fatty tissue of fish and shellfish. Thus, fish are commonly used to monitor the quality of water ecosystems (Yim et al. 2005). Freshwater fish are heavily consumed in the Chinese population. Potential human risk exists universally because of the consumption of fish contaminated by persistent pollutants. It has been found that greater than 90% of human OCs exposure originates from food (Fürst et al. 1990). Because fish generally contain higher levels of OCs than any other food category, a diet containing large amounts of fish may lead to more consumption of OCs even if fish products represent only a small amount (about 10%) in the overall diet (Harrison 1998). Some studies on persistent pollutant contamination in fish tissues associated with human risk assessment have been carried out in China and other countries (Dougherty et al. 2000; Jiang et al. 2005; Yim et al. 2005; Yang et al. 2006). However, in China, most of these studies focused on the fish and shellfish in estuarine and riverine environments (Jiang et al. 2005; Yang et al. 2006; Meng et al. 2007). Little information is available on fish from inland freshwater lakes. In China, freshwater fisheries or inland aquaculture accounted for about two-thirds of total aquaculture production (Wang and Yi 1995), so consumption of contaminated freshwater fish is regarded as the main pathway of human exposure to contaminants.

Baiyangdian Lake, located about 150 km southwest of Beijing, is the largest freshwater lake in North China. Currently, there are more than 243,000 people living in 39 villages scattered around the lake. As an important base of fish farming in North China, Baiyangdian Lake provides aquatic products for nearly seven million people. Although a few studies have investigated the contamination status of OCPs and PAHs in Baiyangdian Lake (Hu et al. 2010; Liu et al. 2010), as far as the authors know, this is the first

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comprehensive study on health risk assessment for consumption of fish from Baiyangdian Lake, China. This study aimed to determine the levels of OCPs and PCBs in fish from Baiyangdian Lake, assess potential health risks associated with fish consumption, and provide preliminary advisories on consumption of fish from Baiyangdian Lake.

Materials and Methods

A total of 121 fish, belonging to 5 species, were collected from fifteen sampling sites throughout Baiyangdian Lake (Fig. 1) in July, 2008. Information on fish length, weight and lipid content are given in Table 1. After transport to laboratory, they were stored at −20°C until chemical analysis.

OCPs, including α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p′*-DDE, *p,p′*-DDD, *o,p′*-DDT, *p,p′*-DDT, and six indicator PCBs (IUPAC Nos. PCB-28, 52, 101, 138, 153, 180) were analyzed. Standards were obtained from AccuStandard Inc (AccuStandard, USA) and all solvents were HPLC grade and purchased from Dikma Technologies (CA, USA). Sample treatment and analysis followed Yang et al. (2006) with minor modifications. Briefly, 10 g of homogenized sample dehydrated with anhydrous Na_2SO_4 was extracted

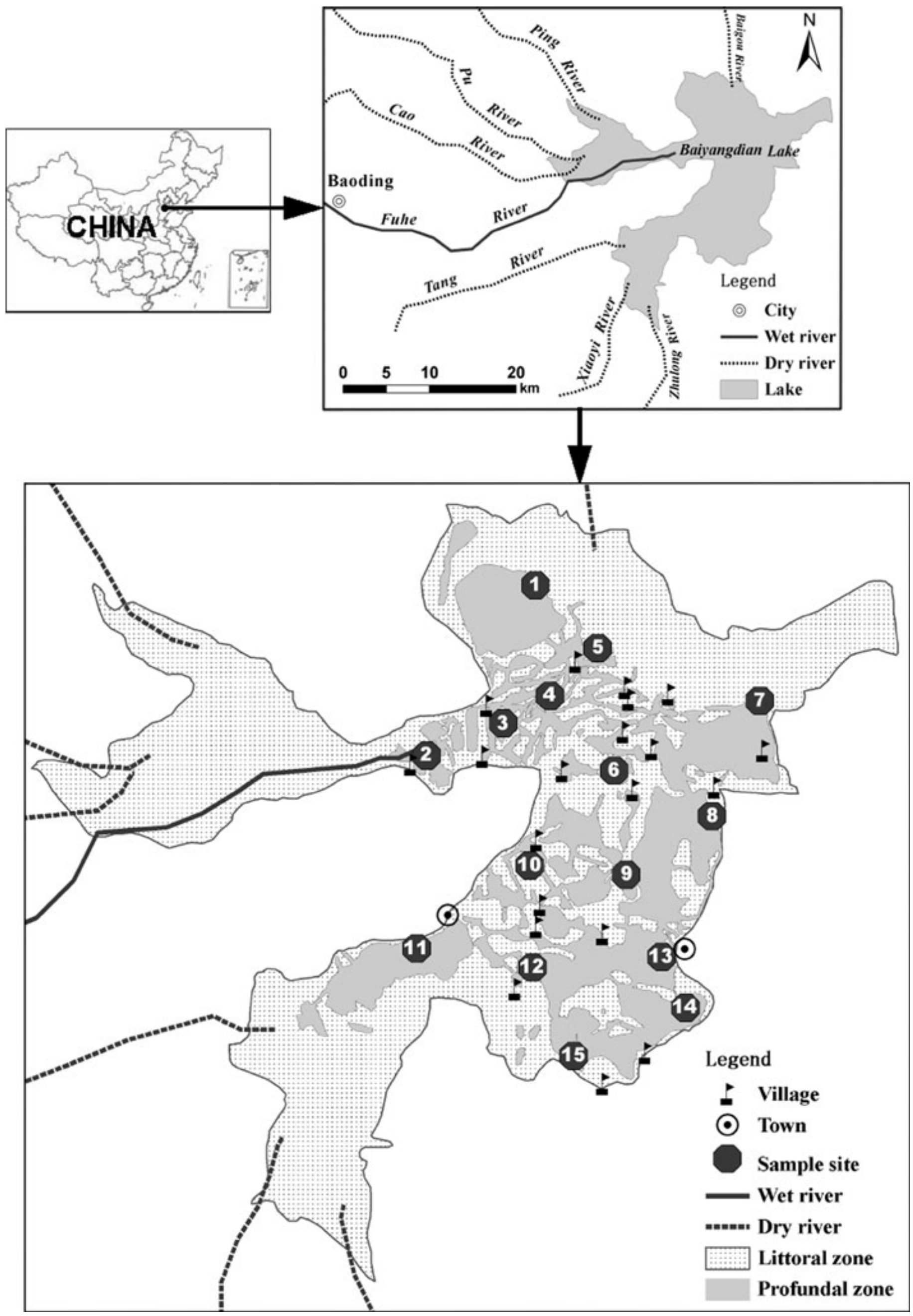


Fig. 1 Map showing sampling locations and surrounding areas in Baiyangdian Lake, China

Table 1 Characteristics of fish species from the Baiyangdian Lake

Common name	Scientific name	No. of specimens	Length (cm)	Weight (g)	Lipid content (%)
Common carp	*Cyprinus carpio*	23	24.8–31.2	630.2–1102.1	5.6–8.8
Crucian carp	*Carassius auratus*	25	19.2–20.1	223.1–267.5	6.3–7.7
Grass carp	*Ctenopharyngodon idella*	27	45.3–50.6	986.8–1927.0	3.5–5.2
Bighead carp	*Aristichthys nobilis*	25	26.8–32.6	498.2–728.5	4.5–5.5
Northern snakehead	*Channa argus*	21	25.1–33.9	241.3–610.2	6.9–8.1

with 100 mL hexane/acetone (3:1, *v*:*v*) in Soxtec 2050 (Foss) for 2 h. From this whole extract, 10% of it was used to gravimetrically determine lipid content. The remaining volume of extraction solvent was treated with concentrated sulphuric acid for lipid purification, and further cleanup was accomplished on a column filled with 8 g acidified silica gel and desiccated sodium sulphate. The column was eluted with 15 mL hexane and 10 mL dichloromethane, respectively. The eluate was concentrated to 100 μL under a gentle nitrogen stream. OCs were quantified with a Varian CP-3800 gas chromatograph with a Varian 4,000 mass spectrometer. Separations were carried out using a VF-5MS column (30 m × 0.25 mm i.d. 0.25 μm film thickness, Varian, USA).

For every six samples, a method blank, a matrix spike, and a sample duplicate were processed together with fish samples by performing the entire pretreatment procedure. Recoveries of spiked OCPs and PCBs into samples which passed through the analytical procedure ranged from 72.8% to 87.1% for OCPs, and 73.8% to 92.3% for PCBs. The relative standard deviation of the repeatability was below 25%. The limit of detection (LOD) for OCPs ranged between 0.02 ng/g and 0.05 ng/g; and PCBs between 0.01 and 0.04 ng/g.

To assess the public health risk of exposure to OCs through fish consumption, hazard ratios (HRs) were calculated by dividing the estimated daily intake (EDI) by the benchmark concentration using equation (1) (Dougherty et al. 2000; Jiang et al. 2005). The benchmark concentration for carcinogenic effect was derived using the USEPA cancer slope factor (CSF), and the benchmark concentration for noncarcinogenic effect is the USEPA reference dose (RfD). Risk associated with carcinogenic and noncarcinogenic health effects due to exposure were considered separately. A hazard ratio that is greater than one indicates that there is potential risk to human health.

$$Hazard\ ratio\ (HR) = {}^{EDI}/_{benchmark\ concentration} \quad (1)$$

For each contaminant, the estimated daily intake (*EDI*, in ng/kg day) via consumption of fish was calculated using the following equation:

$$EDI = \frac{C \times IR}{BW} \quad (2)$$

where C is the measured concentration of chemical contaminant in all species of fish (ng/g wet weight), IR is the average daily consumption rate of fish (g/day) and BW is the body weight, assumed to be 60 kg for an adult. The average fish consumption rate that was used was 12.3 g/day for an adult (Meng et al. 2007).

Two HRs were estimated to assess the potential health risk to humans. One was based on the 50th percentile measured concentrations in fish tissue and the other was based on the 90th percentile measured concentrations.

The risk-based consumption advisory for multiple carcinogens in fish was derived using the methodology developed by USEPA Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories (USEPA 2000). The USEPA approach is designed to manage health risks by providing risk-based consumption advice regarding contaminated fish (for example, one should limit consumption of a particular species to a specified number of meals per month or week). The equations are listed as follows:

$$CR_{\lim} = \frac{ARL \times BW}{\sum_{m=1}^{x} C_m \times CSF_m} \quad (3)$$

$$CR_{mm} = \frac{CR_{\lim} \times T_{ap}}{MS} \quad (4)$$

where CR_{lim} is the maximum allowable fish consumption rate (kg/day), CR_{mm} is the maximum allowable fish consumption rate (meal/month), ARL is the maximum acceptable lifetime risk level (unit-less, 1×10^{-5}), BW is the consumer body weight (kg), X is the number of contaminants, C_m is the measured concentration of contaminant m in a given species of fish (mg/kg), CSF_m is the cancer slope factor for contaminant m, usually the upper 95 percent confidence limit on the linear term in the multistage model used by the USEPA ([mg/kg day]$^{-1}$), MS is the meal size (0.227 kg fish/meal), and T_{ap} is the time average period (30.44 day/month). The CR_{lim} for multiple contaminants assumes that effects are additive. Equation (4) was used to convert daily consumption limits, the number of allowable kilograms per day, to the number of allowable meals per month.

Results and Discussions

Concentrations of HCHs, the DDT complex (DDTs) and PCBs in different fish species collected from the Baiyangdian Lake are summarized in Table 2. The one-sample Kolmogorov–Smirnov test indicated that the concentrations of HCHs, DDTs and PCBs in various fish species were log-normally distributed ($p > 0.05$). The log-normal distribution of micro-organic contaminants in environmental media was also reported in other literatures (Meng et al. 2007; Wang et al. 2009). Therefore, geometric means and geometric standard deviations were calculated to denote the values of HCHs, DDTs and PCBs concentrations in fish samples. DDTs (sum of *p*,*p*′-DDE, *p*,*p*′-DDD, *o*,*p*′-DDT and *p*,*p*′-DDT) represented the predominant group with concentrations ranging from 1.23 to 2.55 ng/g, wet weight. PCBs (sum of PCB-28, 52, 101, 138, 153 and 180) represented the second most common contaminant group with concentrations ranging from 0.53 to 3.28 ng/g, wet weight. HCHs (sum of α-, β-, γ-, and δ-HCH) were relatively low and ranged from 0.28 to 1.56 ng/g, wet weight. Similar results were also reported in fish samples (DDTs > PCBs > HCHs) by Jiang et al. (2005). The concentrations of persistent pollutants in fish samples varied with the species. Northern snakehead had the maximum levels of OCs (7.39 ng/g) followed by common carp (4.0 ng/g), bighead carp (3.56 ng/g), crucian carp (3.16 ng/g), and grass carp (2.04 ng/g). These results were similar to those found in former studies (Yang et al. 2006; Meng et al. 2007), which indicated that bioaccumulation of OCs in fish were highly species dependent, probably due to different ecological characteristic for different aquatic species, such as feeding habits and trophic position (Yang et al. 2006).

In 2005, China reestablished the maximum residual levels of 0.5, 0.1, and 2.0 mg/g for DDTs, HCHs and PCBs, respectively, in fishery products (SEPA (State Environment Protection Agency of China) 2005). For the residues determined in the present study, the maximum concentrations (Table 2) of DDTs, HCHs and PCBs were all much below the respective maximum residual levels. Also, no fish samples contained higher contaminant levels than the action levels and tolerances for DDTs (5,000 ng/g) and PCBs (2,000 ng/g) developed by the US Food and Drug Administration (USFDA 2005).

The compositions of HCHs, DDTs and PCBs in fish from Baiyangdian Lake are shown in Fig. 2a–c, respectively. It was found that α-, β-, γ- and δ-HCH were present in all of the samples collected in the present study, indicating that all the isomers of HCHs are prevalent in Baiyangdian Lake. Technical HCHs usually contains α-, β-, γ- and δ-HCH isomers of 55%–80%, 5%–14%, 8%–15%, and 2%–16%, respectively (Willett et al. 1998). In our samples, the average concentrations of α-, β-, γ-and δ-HCH were found to contribute 36%–51% (mean 41%), 13%–25% (mean 17%), 16%–28% (mean 21%) and 17%–24% (mean 19%), respectively. The ratios of α-HCH/γ-HCH

Table 2 Concentrations of OCPs and PCBs in fish (ng/g, wet weight) from Baiyangdian Lake

Compounds	Common carp	Crucian carp	Grass carp	Bighead carp	Northern snakehead
α-HCH	0.18 ± 0.12	0.22 ± 0.08	0.10 ± 0.09	0.17 ± 0.11	0.79 ± 0.22
β-HCH	0.09 ± 0.02	0.10 ± 0.04	0.07 ± 0.01	0.06 ± 0.02	0.20 ± 0.11
γ-HCH	0.11 ± 0.07	0.15 ± 0.06	0.05 ± 0.01	0.06 ± 0.04	0.44 ± 0.23
δ-HCH	0.12 ± 0.02	0.11 ± 0.05	0.06 ± 0.01	0.09 ± 0.10	0.12 ± 0.05
HCHs	0.50 ± 0.17	0.57 ± 0.10	0.28 ± 0.06	0.38 ± 0.13	1.56 ± 1.06
p,*p*′-DDD	0.94 ± 0.23	0.66 ± 0.12	0.62 ± 0.09	1.08 ± 0.57	0.78 ± 0.35
p,*p*′-DDE	0.24 ± 0.10	0.37 ± 0.07	0.21 ± 0.11	0.11 ± 0.07	0.59 ± 0.23
o,*p*′-DDT	0.52 ± 0.21	0.53 ± 0.09	0.40 ± 0.10	0.37 ± 0.07	0.62 ± 0.14
p,*p*′-DDT	0.10 ± 0.06	nd	nd	0.09 ± 0.02	0.56 ± 0.13
DDTs	1.79 ± 0.46	1.52 ± 0.28	1.23 ± 0.17	1.66 ± 0.43	2.55 ± 0.59
PCB28	0.02 ± 0.03	nd	nd	0.04 ± 0.01	0.06 ± 0.03
PCB52	0.07 ± 0.03	0.06 ± 0.02	0.05 ± 0.01	0.10 ± 0.03	0.16 ± 0.07
PCB101	0.23 ± 0.08	0.17 ± 0.04	0.06 ± 0.02	0.22 ± 0.07	0.46 ± 0.13
PCB138	0.57 ± 0.21	0.34 ± 0.10	0.16 ± 0.05	0.38 ± 0.12	0.79 ± 0.34
PCB153	0.66 ± 0.10	0.43 ± 0.09	0.19 ± 0.06	0.59 ± 0.12	1.39 ± 0.43
PCB180	0.16 ± 0.07	0.07 ± 0.02	0.07 ± 0.03	0.19 ± 0.08	0.42 ± 0.21
PCBs	1.71 ± 0.27	1.07 ± 0.19	0.53 ± 0.18	1.52 ± 0.21	3.28 ± 0.67

nd not detected

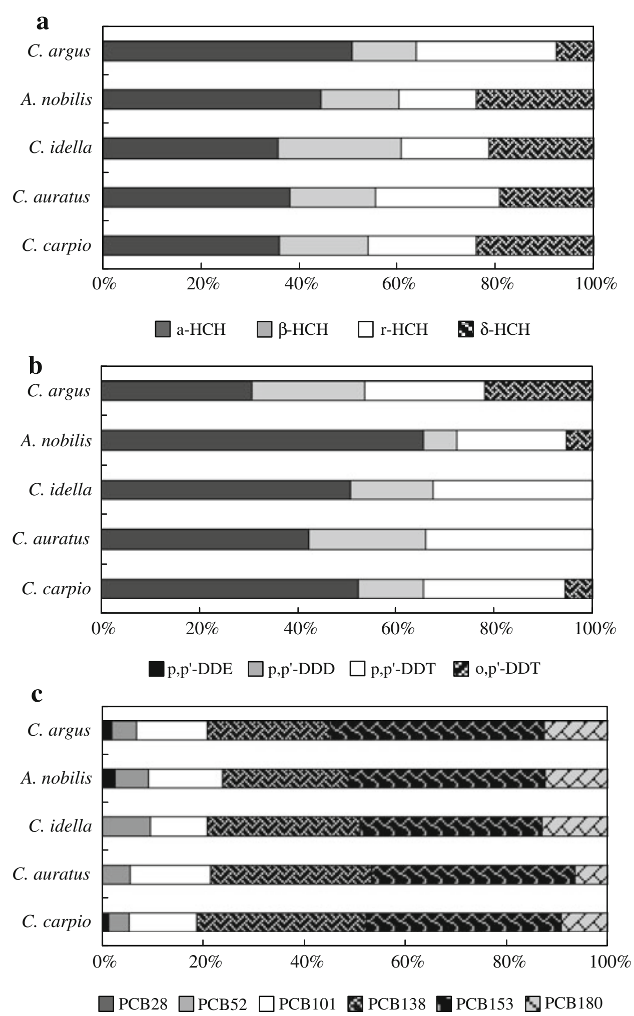


Fig. 2 Composition of HCHs (**a**), DDTs (**b**) and PCBs (**c**) in fish from Baiyangdian Lake

ranged between 1.50 and 2.85, and were significantly lower than the range of 4–10 in technical HCHs, which indicated that the early usage pattern of HCHs was a mixture of technical HCHs and lindane (99% γ-HCH). In addition, the prevalence of all HCH isomers in Baiyangdian Lake indicated the preferential usage of technical HCHs over lindane in the area (Yang et al. 2006).

The composition of DDTs is shown in Fig. 2b. *p,p′*-DDE was at highest level followed by *p,p′*-DDT, *p,p′*-DDD and *o,p′*-DDT. Similarly, Jiang et al. (2005) reported *p,p′*-DDE as main contaminant metabolite of DDT. DDT is degraded into DDD under anaerobic conditions and into DDE under aerobic conditions (Hitch and Day 1992). In general, a ratio of (DDE + DDD)/$\sum$DDTs of more than 0.5 indicates long-term biotransformation of DDT to DDE and DDD, while a ratio of less than 0.5 may indicate recent input of DDT (Hitch and Day 1992). The ratios of (DDE + DDD)/$\sum$DDTs was high (mean 0.70) in this study, indicating aged DDT pollution.

As for PCBs, PCB153 and PCB 138 were dominant contaminants, accounted for about 64% to 72% of total PCBs detected in the present study (Fig. 2c). This distribution of PCB congeners could be explained by the fact that the accumulative properties of PCB congeners increase with the number of chlorine atoms substituted for hydrogen atoms in biphenyl rings, and the resulting increase in their lipophilicity (Bernes 1998). Furthermore, it can also be explained by the fact that higher chlorinated PCBs are so large that they have greater difficulty passing through the body's cell membranes, resulting in greater potential for accumulation in fish samples (Bernes 1998).

To estimate the cancer and noncancer health risk to humans from eating these fish, the HRs was calculated for each contaminant by equations (1) and (2). The oral RfD values, CSF values and cancer benchmark concentrations for OCs are summarized in Table 3. Relevant oral RfD and CSF values were obtained primarily from the Integrated Risk Information System (IRIS) (USEPA 2009).

An evaluation of the noncancer and cancer risks to human health associated with the consumption of fish

Table 3 The benchmark concentrations and estimated daily intakes of OCs by humans

Contaminant	Oral RfD (mg/kg day)	CSF $(mg/kg\ day)^{-1}$	Cancer benchmark concentration (ng/kg day)[c]	50th MC[d]	90th MC[e]	EDI[f] (ng/kg day)	
						50th MC	90th MC
HCHs	0.0003	1.3[a]	0.77	0.52	1.58	0.10	0.32
DDTs	0.0005	0.34	2.94	1.67	2.56	0.34	0.52
PCBs	0.00002	2.0[b]	0.50	1.50	3.31	0.31	0.68

[a] CSF for γ-HCH obtained from USEPA Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories (USEPA 2000)

[b] upper-bound CSF for PCBs

[c] Benchmark concentration for carcinogenic effects equals 10^{-6} divided by the cancer slope factor and represents the exposure concentration at which lifetime cancer risk is one in one million

[d] The 50th percentile measured concentration (MC) of contaminants in fish (ng/g wet weight)

[e] The 90th percentile measured concentration (MC) of contaminants in fish (ng/g wet weight)

[f] The EDI calculated based on the 50th and 90th percentile measured concentrations of contaminants

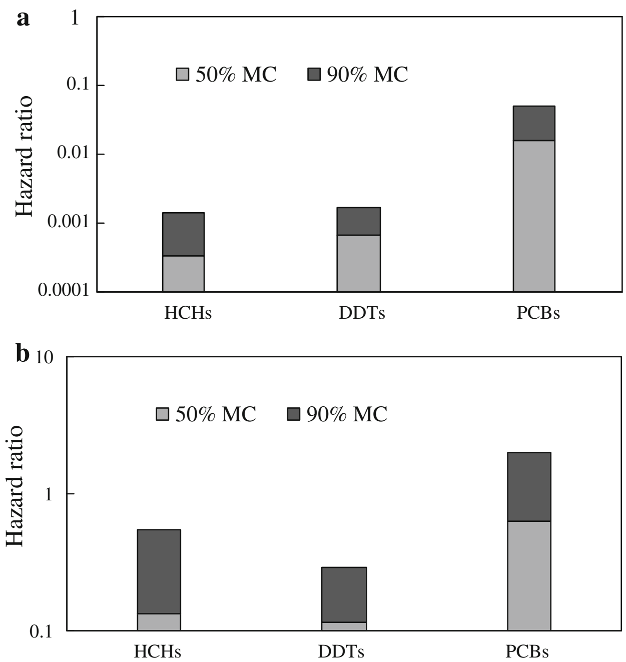
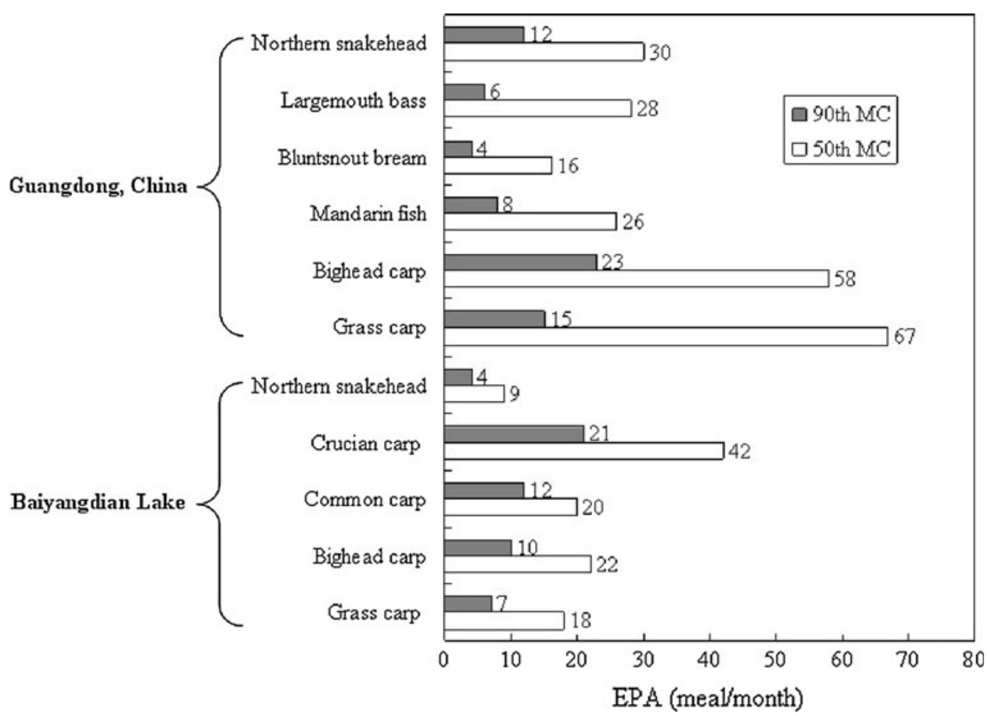


Fig. 3 Hazard ratios of noncancer risk (**a**) and cancer risk (**b**) based on 50th and 90th percentile measured concentrations (MC)

containing OCPs and PCBs was undertaken and the results are shown in Fig. 3a, b. The HRs of noncancer risk based on 50th and 90th percentile concentrations were all less than one (Fig. 3a), due to the relatively low concentrations of OCs in the fish tissue (Table 2). Similarly, a health risk assessment associated with fish consumption in the coastal city of Zhoushan, China, also indicated that the noncancer hazard ratios for DDTs and PCBs were all less than unity (Jiang et al. 2005). An estimation of the lifetime cancer risk based on the 50th and 90th percentile concentrations of HCHs and DDTs were all less than one (Fig. 3b). In contrast, the HR for cancer risk based on the 90th percentile concentrations of PCBs was greater than one (Fig. 3b), suggesting that daily exposure to PCBs due to fish consumption would yield a lifetime cancer risk of greater than one in one million.

Generally, individuals who ingest chemically contaminated fish may be exposed to a number of different chemicals (e.g., HCHs, DDTs, PCBs, heavy metals, and endocrine-disrupting chemicals) simultaneously. The US Environmental Protection Agency (USEPA 2000) developed a risk-based approach to the exposure of multiple chemicals 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 the risk-based fish consumption rates, assuming an average meal size of 0.227 kg, and an adult body weight of 70 kg, consistent with USEPA (2000) risk assessment methods for contaminants in fish. The 50th and 90th percentile measured concentrations of HCHs, DDTs and PCBs and their corresponding cancer slope factors for carcinogenic effects were used to calculate species-specific consumption limits expressed as number of fish meals per month by equations (3) and (4). Fig. 4 shows the risk-based meal consumption limits for the five species of fish from Baiyangdain Lake and the comparison with that from

Fig. 4 Consumption advisories (in meals per month) based on USEPA cumulative risk assessment methods for HCHs, DDTs, and PCBs for five species of fish from Baiyangdian Lake and the comparison with that from Guangdong, China. The numbers on the white bars are calculated with the 50th percentile concentrations, while the numbers on the gray bars are calculated with the 90th percentile concentrations



Guangdong, China. From Fig. 4, it can been seen that the combined 90th percentile measured concentrations of HCHs, DDTs and PCBs triggered EPA consumption advice of no more than 12 meals per month for common carp, grass carp, and bighead carp but crucian carp, for which the advice is no more than 21 meals per month. The 50th percentile measured concentrations of HCHs, DDTs and PCBs triggered less restrictive advice of between 18 and 42 meals per month for common carp, grass carp, bighead carp and crucian carp. For northern snakehead, the most contaminated species, the combined concentrations of HCHs, DDTs and PCBs triggered the most restrictive advice of less than 9 meals (median) and 4 meals (90th percentile concentration) per month, as shown in Fig. 4. In comparison, consumption of northern snakehead, bighead carp and grass carp from Baiyangdian Lake was more likely to raise a health concern than consumption of the same species of fish from Guangdong, China (Meng et al. 2007) (Fig. 4).

Our results indicated that the concentration of HCHs, DDTs and PCBs in fish from Baiyangdian Lake were moderate, with the mean and maximum values much lower than the corresponding maximum admissible residual levels established by the Chinese government, and the action levels and tolerances developed by the USFDA. The HRs of noncancer risk for HCHs, DDTs and PCBs were all less than one based on their 50th and 90th percentile concentrations. However, the HRs of cancer risk for PCBs was greater than one based on its 90th percentile concentration, which indicated that some carcinogenic effects caused by PCBs compounds may occur when consumption of fish from Baiyangdain Lake. Further risk-based consumption advisory study indicated that consumption of northern snakehead from Baiyangdian Lake may result in exposure to a variety of persistent bioaccumulative contaminants with the potential for an elevation in attendant health risks. This study accounted for only a limited number of contaminant. Additional surveys on other toxic substances such as heavy metals, polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and perfluorinated compounds are needed. Attention should also be paid to interactions and combined effects among contaminants.

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