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RESEARCH ARTICLE

Concentrations, bioaccumulation, and human health risk assessment of organochlorine pesticides and heavy metals in edible fish from Wuhan, China

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Abstract The objective of this study was to determine concentration and bioaccumulation of organochlorine pesticides and heavy metals in edible fish from Wuhan, China, in order to assess health risk to the human via fish consumption. Two edible fish species (*Aristichthys nobilis* and *Hypophthalmichthys molitrix*) were collected and analyzed for 11 organochlorine pesticides (OCPs) and eight heavy metals (HMs). Concentrations of ∑HCHs, ∑DDTs, and ∑OCPs in fish samples were in the range of 0.37–111.20, not detected (nd)–123.61, and 2.04–189.04 ng g^{-1} (wet weight), respectively. Bioaccumulation factors (BAFs) of OCPs in bighead carp (*A. nobilis*) were higher than those in silver carp (*H. molitrix*). Concentrations of ∑HMs in bighead carp and silver carp were 352.48 and 345.20 mg kg^{-1} (dw), respectively. Daily exposure of OCPs and HMs for consumers was estimated by comparing estimated daily intake (EDI) with different criteria. The results revealed that the EDIs in our study were all lower than those criteria. Target hazard quotient (THQ) and risk ratio (*R*) were used to evaluate non-carcinogenic and carcinogenic risks, respectively. As regard to non-carcinogenic effects of the contaminants, hazard quotients (THQ) of OCPs and HMs were both lower than 1.0, implying negligible non-carcinogenic risk via fish consumption in study area. Nevertheless, in view of carcinogenic effects of the contaminants, the total value of risk ratio (*R*) of OCPs was lower than the threshold of tolerable risk while the total value of risk ratio (*R*) of HMs was higher than the threshold of tolerable risk due to the high carcinogenic risk ratios of As and Cr, indicating high carcinogenic risks via fish consumption. The results demonstrated that HMs in edible fish from Wuhan, China, especially As and Cr required more attention than OCPs.

Keywords OCPs · Heavy metals · Fish · Health risk · The East Lake · Wuhan

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Introduction

Organochlorine pesticides (OCPs) and heavy metals (HMs) are of global concern in recent decades because of their persistence in environment, bioaccumulation in organisms, and toxicity to human and wildlife. Bioaccumulation of OCPs in biota can lead to some adverse effects on reproduction, nervous systems, and immunity (King et al. 2003; Longcore and Stendell 1977; Misumi et al. 2005). HMs include potentially toxic elements (arsenic, cadmium, lead and mercury, etc.), probably essential elements (nickel, vanadium, cobalt), and essential elements (copper, zinc, iron, manganese) for fish. The potentially toxic elements can be very hazardous at low concentration such as mercury which caused a serious epidemic called Minamata disease in Japan 1950s (Uluozlu et al. 2007), and the essential elements can cause nephritis, anuria, and extensive lesions in kidney if they were excessively took in (Çelik and Oehlenschläger 2007; Luckey and Venugopal 1978; Rahman and Islam 2009).

Among all kinds of aquatic organisms, fish is a suitable indicator for environmental pollution monitoring (Fausch et al. 1984; Roset et al. 2007; Van Ael et al. 2014). The uptake of the pollutants in fish is directly from water via gill and skin and indirectly from diet. Data of pollutants accumulated in fish are indicative not only for the environment but also for the transfer of pollutants through the trophic web (Gobas et al. 1999). The edible fish is an important source of protein and omega-3 polyunsaturated fatty acids which can well guard against cardiovascular disease for human (Hu et al. 2003). The consumption of contaminated edible fish can cause accumulation of pollutants in human body (Li et al. 2008).

Wuhan is the largest city in central China with another name of "city of hundreds of lakes," where the residents are habituated to eating fish and fish consumption is huge every year. In recent years, with the rapid economic growth of Wuhan, many lakes have been subject to serious pollution (Liu et al. 2014; Tang et al. 2007; Yang et al. 2014; Yang et al. 2009; Yun et al. 2014). In the present study, we choose the East Lake as sampling site which not only used to be the largest urban lake in China but also an important producing area of fish in Wuhan. It is a typical shallow lake in middle and lower drainage basin of the Yangtze River and has an average depth of 2.2 m, a mean temperature of 16.7 °C and a maximum temperature of 40 °C all through the year in the lake area. Due to more and more intensive anthropogenic activities in the surroundings for the past few years, the East Lake suffered considerable OCP and HM pollution and the water quality was graded at IV according to 2013 Wuhan Environmental Aspect Bulletin (WEPB 2014a). Reports analyzing concentrations of OCPs in surface water of the East Lake and revealing carcinogenic risk for human have aroused great concern of the government (Yang et al. 2014). Yun et al. and Liu et al. also revealed that OCPs and HMs in sediments of the East Lake might cause potential ecological risk to aquatic organisms (Liu et al. 2014; Yun et al. 2014). What is more, the East Lake was close to a heavy industrial area where several enterprises such as iron and steel company, foundries plants, and machinery factory included into list of the key enterprises of heavy metal emission published by Wuhan Environmental Protection Bureau were located (WEPB 2014b). There are also hundreds of thousands of residents, several hospitals, and farmlands around the lake which might be the source of OCP and HM pollution in the lake. Although OCP and HM pollution in this area was so serious, almost no information of OCPs and HMs in fish was reported either in East Lake or in other lakes of Wuhan.

Therefore, the objectives of this study are to clarify the concentrations and accumulation levels of OCPs and HMs in edible fish in Wuhan, assess the daily exposure and health risk of human via fish consumption in this area, and provide references for pollution research in other urban lakes of the world. This study gives a comprehensive overview of OCP and HMs' status in edible fish of Wuhan and provides a basis for government to perform environmental contamination management and take effective measures to reduce potential health risks.

Materials and methods

Study area and sample collection

Details of sampling sites can be found in Fig. 1. Twenty fish, 42 water, and 42 sediment samples were collected from the East Lake in June 2013. The fish samples included 10 bighead carp (*Aristichthys nobilis*) and 10 silver carp (*Hypophthalmichthys molitrix*) whose dietary habits were all filter feeder, belonging to cyprinid which were also important edible fishes in Wuhan even in China (Xie et al. 2004) (Table 1). The water and fish samples were analyzed for 11 OCPs including α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p*'-DDE, *p,p*'-DDD, *p,p*'-DDT, heptachlor, heptachlor epoxide, aldrin, and methoxychlor. The content of eight heavy metal elements including Cr, Ni, Cu, Zn, As, Cd, Hg, and Pb in water, sediment, and fish samples was also determined simultaneously in this study. Yun et al. had already tested concentrations of OCPs in sediments in June 2013, so the experiment was not repeated in this study (Yun et al. 2014). All of the samples were stored in an ice-chest and transported immediately to laboratory. Throughout the sampling process, the positions of sampling sites were fixed by a global positioning system (GPS).

In the laboratory, fish were dissected and the bladder, intestine, heart, brain, liver, and muscle tissues of the dorsal of fish were taken for analysis. Fish samples were freeze-dried, pulverized, and then stored at −20 °C before extraction. The particulate in the water samples was removed by filtering with 0.45-μm hydrophilic filters. Sediment samples were lyophilized, pulverized, and sifted through 60-mesh sieve. All the sample preparation was finished in 7 days.

Sample extraction and cleanup

Organochlorine pesticides

The concentrations of 11 OCPs were determined in water and fish including the muscle and liver.

OCPs in water samples were tested as described in Muir and Sverko (2006). A recovery surrogate 2,4,5,6-tetrachloro-*m*-xylene (TCmX) was added into 1 L of filtered water sample. The sample was performed liquid-liquid extraction by dichloromethane (35 mL). Water in the organic phase was removed by Na_2SO_4 column, and then, solvent of the extract was replaced into *n*-hexane. After that, a column packed with neutral silica (4.0 g, 3 % deactivated), neutral alumina (2.0 g, 6 % deactivated), and anhydrous sodium sulfate (1 cm) from the bottom to the top was used to remove impurities in the

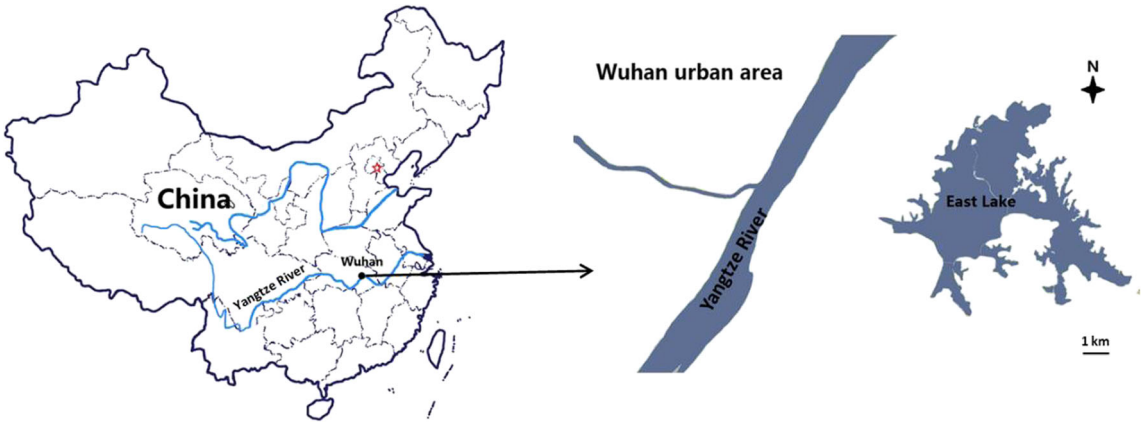

Fig. 1 Sketch map of the sampling site

extract. Finally, the purified extract was blown to dryness by purity nitrogen and the residue was dissolved with 20 μL *n*-hexane.

OCPs in the fish livers and muscles were extracted according to matrix solid-phase dispersion method with some appropriate alternations (Villaverde-de-Sáa et al. 2013). The fish sample (0.5 g) was blended thoroughly with a known amount of 2,4,5,6-tetrachloro-*m*-xylene (TCmX) as a surrogate and 0.5 g octadecylsilane-bonded silica (C18) (SiliCycle Inc., Québec, Canada) as dispersion sorbent in a glass mortar using a glass pestle for 5 min to obtain a homogeneous mixture. The mixture was transferred into a polyethylene syringe barrel column (10 mL) which contained a membrane filter (0.22 μm). From bottom to top, the column was packed with neutral silica (0.5 g, 100–200 mesh, 180 °C 4 h activated), acidic silica (2.5, 30 % fume sulfuric acid, *w*/*w*), florisil (1.75 g, 60–100 mesh, 150 °C 10 h activated), acidic alumina (1.0 g, 150 °C 8 h and 5 % deactivated), and sample mixture. The other membrane filter was placed on the top. The packed column was eluted by 15 mL of dichloromethane flowing through at a rate of 2 mL min^{-1}. Then, the eluent was dried with a gentle stream of high purity nitrogen followed by the redissolution of dry residue with 200 μL of *n*-hexane and addition of internal standard pentachloronitrobenzene (PCNB). The analytes were analyzed qualitatively with gas chromatography-mass spectrometer (GC-MS) and quantified with gas chromatography-electron capture detector (GC-ECD).

Table 1 Biometric data and mean concentrations of ∑OCPs and ∑HMs of fish

	Bighead carp		Silver carp	
Scientific name	*Aristichthys nobilis*		*Hypophthalmichthys molitrix*	
n[a]	10		10	
Fish length (cm)	50.10±4.15		54.29±7.80	
Fish weight (kg)	3.11±0.32		3.50±0.81	
Dietary habits	Filter feeder (zooplankton, phytoplankton, and detritus)		Filter feeder (zooplankton, phytoplankton, and detritus)	
Trophic level[b]	2.51		2.59	
	Muscle	Liver	Muscle	Liver
Water content (%)	74.28±12.22	79.57±1.12	74.51±10.53	76.14±2.48
Lipid (%)	4.71±2.35	12.68±4.32	14.73±8.72	24.77±16.03
∑OCPs[c]	15.23	10.38	24.56	15.85
∑HMs[d]	352.48		345.20	

Data was shown as mean±standard deviation

[a] Number of samples

[b] The data was cited from Yu et al. (2012)

[c] The concentrations were listed in wet weight (ng g^{-1})

∑OCPs=α-HCH+β-HCH+γ-HCH+δ-HCH+*p*,*p*'-DDE+*p*,*p*'-DDD+*p*,*p*'-DDT+heptachlor+aldrin+heptachlor epoxide+methoxychlor

[d] The concentrations listed in dry weight were a sum of the concentration of six tissues of fish (mg kg^{-1})

∑HMs=As+Cd+Cr+Cu+Hg+Ni+Pb+Zn

Heavy metals

The concentrations of eight heavy metals were determined in water, sediment, and fish including the muscle, liver, heart, brain, intestine, and bladder.

About 10 mL water sample was filtered under vacuum with 0.45-μm hydrophilic filters to get rid of particulates. Lyophilized sediment sample (0.1–0.2 g) and fish tissue samples (0.1–0.2 g) including the bladder, intestine, heart, brain, liver, and muscle were weighed directly into acid-washed Teflon digestion vessels. Eight milliliters of pure concentrated HNO_3 and 2 mL of hydrogen peroxide (30 %) were added before microwave digestion. Each batch included a reagent blank and certified reference materials included NRCDOLT-3 (Dogfish liver) and GBW08573 (Yellow-fin tuna) (both from National Standard Material Center, Beijing, People's Republic of China). Samples were digested for 30–40 min at 180 °C. After cooling for at least 1 h, the vessels were placed in a solvent evaporation system at a temperature of 140 °C for 1.5 h to remove most of the extractant. Following cooling for at least 1 h, the digested samples were transferred to 50-mL volumetric flasks, and the volume was made up to 50 mL with deionized water.

Lipid

The lipid content of the fish muscle was determined gravimetrically as described in a previous study with some modifications (Smedes 1999). Twenty milliliters water, cyclohexane, and 16 mL isopropyl alcohol were added to 2 g freeze-dried muscle powder, followed by ultrasonic extraction. The organic phase was collected after the mixture reached the statically separated equilibrium. The extraction was repeated by the addition of 3 mL isopropyl alcohol and 18 mL cyclohexane, and then, organic phase was combined and dried under a gentle nitrogen stream. The residue was weighed. The extraction steps of lipid content in the fish liver were similar to those in the fish muscle, but 0.5 g sample and half dosage of the solvent were used.

Experimental protocol and quality assurance and quality control (QA/QC)

For OCPs, the samples were analyzed on an Agilent 7890A gas chromatography equipped with an electron capture detector (GC-ECD) (Agilent Technologies, Santa Clara, CA, USA) and HP-5 capillary column (30 m×0.25 mm i.d.×0.25-μm film thickness, Agilent). Nitrogen was the carrier gas (1 mL min^{-1}). Temperatures of the injector and detector were kept at 250 and 300 °C, respectively. Starting from 80 °C and holding for 1 min, the oven temperature was risen to 150 °C at the rate of 20 °C min^{-1} and finally to 300 °C (5 min hold) at the rate of 5 °C min^{-1}. The splitless injection volume was 1 μL.

During analysis, the instruments were calibrated with calibration standards every day. A solvent blank and matrix blank were tested every 10 samples throughout whole procedure. Each sample was analyzed in duplicate. High, medium, and low levels of recoveries were analyzed during the experiment. The average recoveries of the surrogate standard TCmX were 80±5 % in water samples and 73±8 % in fish samples. The recoveries of OCPs ranged from 70 to 105 % in water samples and from 62 to 87 % in fish samples. The method detection limits (MDLs) were confirmed by the concentrations of analytes whose signal-to-noise (S/N) ratio was three and ranged from 0.15 to 0.30 ng L^{-1} in water samples and from 0.07 to 0.33 ng g^{-1} in fish samples. Those samples with concentrations detected less than MDLs were treated as not detected (nd).

For HMs, samples were analyzed for Cr, Ni, Cu, Zn, As, Cd, Hg, and Pb using an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) (Thermo Electron Co. Ltd, USA). Results were quantified using an empirical calibration curve generated from the responses obtained from multiple dilutions of a mixed calibration standard prepared from individual element standards (GBW07401, obtained from Chinese Academy of Measurement Science). Analytical quality control was performed with 2 % ultrapure nitric acid blank and a procedural blank. The detection values of all elements were in good agreement with the certified values, with recoveries ranging from 83 to 105 %.

Data analysis

Spearman correlation test was performed by SPSS 16.0 for windows (IBM Corporation, USA). Biota-water accumulation factor (BAF) is calculated as shown in the following equation:

$$\mathrm{BAF} = \frac{C_l}{c}$$

where C_l is the pollutant concentration in fish (ng g^{-1}) normalized by lipid content and c is pollutant concentration in water (ng L^{-1}).

Risk assessment

In order to evaluate potential risk to human health of tested fishes, the estimate daily intake (EDI), the target hazard quotients (THQs), and carcinogenic risk ratio (R) were used in the risk assessment. THQ<1 denotes that the daily exposure seems to cause no deleterious effects to human health and the acceptable level of R was in the range of 1×10^{-6}–1×10^{-4} (Chien et al. 2002; Yu et al. 2014). The determination of THQ and R was referred to method described by Chien

et al. and Yu et al., respectively (Chien et al. 2002; Yu et al. 2014). The calculation formulas of EDI, THQ, and R were listed as follows:

$$\mathrm{EDI} = \frac{C \times W_{\mathrm{F}}}{W_{\mathrm{AB}}}$$

where

C=pollutant concentration in food (μg g^{-1}), W_F=daily average consumption of fish in this area (according to the population size and annual consumption of aquatic products in Wuhan, assuming 75 g day^{-1} $person^{-1}$), and W_{AB}=average body weight (70 kg for adults).

$$\mathrm{THQ} = \frac{E_{\mathrm{F}} \times E_{\mathrm{D}} \times F_{\mathrm{IR}} \times C}{R_{\mathrm{FD}} \times W_{\mathrm{AB}} \times T_{\mathrm{A}}} \times 10^{-3}$$

$$R = \frac{E_{\mathrm{F}} \times E_{\mathrm{D}} \times F_{\mathrm{IR}} \times \mathrm{SF} \times C}{W_{\mathrm{AB}} \times T_{\mathrm{A}}} \times 10^{-3}$$

where

E_F=exposure frequency (350 days $year^{-1}$); E_D=exposure duration (70 years); F_{IR}=food ingestion rate (g $person^{-1}$ day^{-1}), R_{FD}=oral reference dose (mg kg^{-1} day^{-1} (BW)), T_A=average exposure time (365 days $year^{-1}$×lifetime, assuming 70 years), and SF=oral cancer slope factor (mg kg^{-1} $day^{-1})^{-1}$. Concentrations used in risk calculations in the present study are all on a wet weight basis. Risk-based concentration table was used in the study to search for R_{FD} and SF values (US EPA 2009).

Sometimes, the exposure is not only to one signal toxicant. When the toxicants are two or more, some additive and/or interactive effects may come up and the risk addition hypothesis is applicable to this situation. The hazard index (HI) is summation of the non-carcinogenic risks, and R_T is the total carcinogenic risks. The calculation formulas of HI and R_T cited from Yu et al. (2014) were listed as follows:

$$\mathrm{HI} = \sum_{i=1}^{m} \mathrm{THQ}_i \text{ and } R_T = \sum_{i=1}^{m} R_i$$

In addition, it has been assumed that the cooking process does not alter the available contaminant concentration into human bodies and the ingested pollutants are absorbed completely by the consumer.

Results and discussion

Concentrations of OCPs and HMs in water and sediment samples

All of the 11 OCPs were detected in water samples. The mean concentrations of ΣOCPs, ΣDDTs, and ΣHCHs were 16.75, 2.00, and 8.51 ng L^{-1}, respectively (Table 2). Yang et al. also sampled water in the same positions in winter 2012 and tested OCPs (Yang et al. 2014). Research showed that water of the East Lake suffered more serious pollution compared with other lakes in the world. In comparison with their results (the total concentration of 12.38 ng L^{-1} of nine OCPs including α-HCH, β-HCH, γ-HCH, δ-HCH, *p*,*p*'-DDE, *p*,*p*'-DDD, *p*,*p*'-DDT, heptachlor, and heptachlor epoxide), the concentration of 13.79 ng L^{-1} of the nine OCPs in our research performed in summer 2013 was marginally higher. No obviously seasonal change trends of concentrations of individual OCP compound were found in this study. Pollution was mainly from the anthropogenic activities such as fishing in winter, recreational activities in summer, and lake tunnel construction recently.

Only four heavy metals were detectable in water samples with the mean concentration of 0.0076 mg L^{-1} for As, 0.00012 mg L^{-1} for Hg, 0.0012 mg L^{-1} for Ni, and 0.0078 mg L^{-1} for Zn, respectively (Table 3). The concentrations of heavy metals in Bozova of Atatürk Dam Lake (Turkey) which were 0.0154 mg L^{-1} for Ni, 0.025 mg L^{-1} for Cu, and 0.064 mg L^{-1} for Pb were higher than those in the present study (Karadede and Ünlü 2000); the heavy metals in Biétri Bay in Ebrié Lagoon, Ivory Coast, at rainy season were found at the levels of 0.2, 0.1, 3.53, 9.43, and 17.04 mg L^{-1} for Hg, Cd, Pb, Cu, and Zn, respectively, which were much more higher than those in the East Lake (Coulibaly et al. 2012); the mean concentrations of Cu, Cd, Cr, Ni, and Zn in the Taihu Lake (China) were 0.002876, 0.000047, 0.00129, 0.002443, and 0.008778 mg L^{-1}, respectively, which were comparable with those in the present study (Tao et al. 2012). It was obvious that water in the East Lake suffered a slight contamination of heavy metals.

In sediment samples, the mean concentrations of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were 17.72, 0.43, 49.02, 49.89, 0.31, 41.6, 19.59, and 401.76 mg kg^{-1}, respectively (Table 3). Compared with the results of Liu et al. in winter 2012, the concentrations of As, Cd, Cr, and Cu in this study were lower while the concentrations of Hg, Ni, Pb, and Zn were higher (Liu et al. 2014).

Concentrations and tissue distribution of OCPs and HMs in fish samples

As is shown in Table 2, 10 organochlorine pesticides were detected in fish samples except *p*,*p*'-DDT. ΣOCPs concentrations (wet weight, ww) ranged from 2.04 to 139.17 ng g^{-1} in the muscle samples and from 5.22 to 189.04 ng g^{-1} in the liver samples. The mean concentration of ΣDDTs detected in the present study was 3.43 ng g^{-1} in the bighead carp muscle which was lower than those reported before (Li et al. 2008; Wang et al. 2012) and 9.38 ng g^{-1} in the silver carp muscle which was higher than those in the Taihu Lake, Poyang Lake, and Lake Ziway in Ethiopia (Wang et al. 2012; Yohannes et al. 2014; Zhao et al. 2014) (Table S1). With respect to HCHs, the

Table 2 The concentrations of OCPs in fish (ng g^{-1} ww), surface water (ng L^{-1}), and sediment (ng g^{-1}) from the East Lake, China

OCPs	Log K_{ow}[a]	Surface water	Sediment[b]	Bighead carp		Silver carp	
				Muscle	Liver	Muscle	Liver
α-HCH	3.94	1.25±0.44 (0.78–2.88)	3.2 (1.3–8.8)	1.40±0.93 (0.37–8.94)	0.60±0.13 (0.44–3.59)	1.81±0.87 (0.44–9.57)	0.94±0.32 (0.55–6.73)
β-HCH	3.92	3.33±2.25 (0.24–9.81)	4.4 (nd–28)	0.69±1.41 (nd–18.11)	1.63±2.07 (nd–30.87)	1.07±0.97 (nd–11.78)	2.97±7.85 (nd–87.07)
γ-HCH	3.83	0.96±0.25 (0.73–2.09)	4.6 (1.8–14.7)	1.82±1.92 (nd–14.18)	0.73±0.18 (0.27–4.60)	2.59±1.75 (nd–14.57)	0.86±0.25 (0.51–5.66)
δ-HCH	3.19	2.96±1.33 (1.13–7.73)	9.8 (2.6–38)	1.47±1.23 (nd–11.02)	1.12±1.10 (0.72–20.83)	1.70±1.13 (nd–9.91)	1.18±0.71 (0.81–11.74)
∑HCHs	–	8.51±3.55 (2.68–19.32)	22 (5.7–61)	5.38±3.78 (0.37–34.46)	4.08±2.43 (2.27–41.99)	7.17±4.45 (0.44–45.81)	5.96±9.08 (1.89–111.20)
Heptachlor	5.94	3.25±1.79 (1.11–11.59)	6.7 (1.9–27)	1.88±1.62 (0.14–13.41)	0.58±0.20 (0.33–3.75)	2.55±1.53 (0.28–14.54)	0.84±0.19 (0.40–3.91)
Aldrin	6.24	2.94±1.26 (0.55–6.13)	19 (3.1–123)	1.39±0.98 (nd–8.48)	0.62±0.17 (0.37–3.89)	1.80±0.75 (0.42–9.96)	0.83±0.08 (0.71–3.85)
Heptachlor epoxide	5.42	0.03±0.11 (nd–0.30)	2 (nd–7.6)	1.00±1.06 (nd–7.79)	0.45±0.45 (nd–5.02)	1.42±0.97 (nd–7.79)	0.74±0.52 (nd–5.08)
p,p'-DDE	6.93	0.64±0.09 (0.45–0.85)	5.3 (1.6–18)	1.77±1.80 (nd–23.63)	3.67±3.70 (0.15–49.32)	6.75±10.88 (nd–121.62)	5.70±6.15 (nd–62.50)
p,p'-DDD	6.33	0.76±0.73 (0.17–3.25)	3.2 (0.53–14)	1.66±1.75 (nd–13.20)	0.54±0.24 (0.10–4.96)	2.63±1.67 (nd–16.82)	0.97±0.57 (0.12–6.98)
p,p'-DDT	6.39	0.60±1.51 (nd–6.86)	nd	nd	nd	nd	nd
∑DDTs	–	2.00±1.35 (0.81–8.28)	8.5 (2.6–32)	3.43±2.54 (nd–23.63)	4.21±3.85 (0.25–52.26)	9.38±10.25 (nd–123.61)	6.67±6.49 (0.12–68.62)
Methoxychlor	6.47	0.02±0.03 (nd–0.53)	5.66 (0.74–22)	2.15±0.61 (1.00–11.47)	0.44±0.30 (0.17–5.66)	2.24±0.70 (1.72–14.64)	0.81±0.34 (0.50–5.53)
∑OCPs	–	16.75±5.27 (7.34–30.77)	63.86 (14–273)	15.23±9.44 (2.04–95.24)	10.38±5.51 (5.22–98.44)	24.56±10.35 (3.83–139.17)	15.85±13.54 (6.69–189.04)

Data was shown as mean±standard deviation; maximum and minimum values were in parenthesis

[a] The octanol-water partition coefficients (log K_{ow}) of OCPs were suggested by Hale et al. (2010 and Shen and Wania 2005)

[b] The data were cited from Yun et al. (2014)

Table 3 The concentrations of HMs in fish (mg kg^{-1} dw), surface water (mg L^{-1}), and sediment (mg kg^{-1}) from the East Lake, China

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Bighead carp								
Muscle	0.3±0.065 (0.20–0.40)	0.0029±0.002 (0.0068-nd)	1.55±1.14 (0.73–4.57)	0.84±0.35 (0.29–1.4)	0.013±0.011 (nd–0.036)	0.15±0.062 (0.087–0.26)	0.056±0.15 (nd–0.16)	1.66±1.85 (1.4–1.87)
Liver	0.46±0.23 (0.32–1.10)	0.42±0.20 (0.21–0.91)	2.96±3.67 (0.89–11.18)	22.23±25.11 (5.49–90.52)	0.078±0.093 (nd–0.32)	0.15±0.15 (nd–0.52)	0.33±0.10 (0.21–0.54)	106.76±29.36 (56.83–151.33)
Bladder	0.17±0.099 (nd–0.35)	0.0081±0.0060 (nd–0.019)	8.82±2.00 (5.8–12.45)	2.69±3.12 (0.11–10.9)	0.23±0.40 (0.013–1.31)	0.36±0.29 (0.12–1.04)	0.0096±0.015 (nd–0.037)	7.56±17.01 (0.49–55.74)
Intestine	0.87±0.52 (nd–1.83)	0.076±0.056 (0.025–0.20)	10.32±1.76 (8.14–13.38)	4.39±1.35 (2.51–7.27)	nd	0.61±0.71 (0.11–2.41)	0.57±1.06 (nd–3.52)	45.88±30.51 (10.56–125.38)
Heart	0.33±0.16 (0.2–0.62)	0.0039±0.0034 (nd–0.011)	8.97±0.44 (8.29–9.63)	7.48±1.63 (4.08–9.84)	0.014±0.020 (nd–0.055)	0.41±0.47 (0.07–1.42)	0.2±0.16 (0.046–0.48)	72.32±94.59 (29.65–323.89)
Brain	0.8±0.53 (0.047–1.31)	0.00083±0.0026 (nd–0.0064)	5.49±1.55 (2.95–7.20)	2.05±1.27 (0.87–3.86)	nd	0.25±0.34 (0.08–1.00)	0.18±0.12 (0.027–0.34)	33.46±41.39 (3.05–112.35)
Silver carp								
Muscle	0.47±0.18 (0.25–0.80)	0.0024±0.0021 (nd–0.0049)	1.16±0.84 (0.48–3.00)	1.07±1.13 (0.22–3.52)	0.011±0.027 (nd–0.073)	0.28±0.33 (0.0294–0.99)	0.14±0.16 (nd–0.35)	3.9±5.63 (1.06–16.54)
Liver	0.65±0.38 (0.34–1.24)	0.19±0.11 (0.059–0.37)	3.46±2.95 (1.03–8.95)	57.33±26.89 (22.55–108.39)	0.025±0.014 (nd–0.042)	0.07±0.052 (nd–0.15)	0.39±0.24 (0.056–0.67)	121.66±34.06 (56.43–158.16)
Bladder	0.23±0.18 (0.12–0.54)	0.004±0.0019 (0.0019–0.0069)	9.38±1.79 (6.71–11.23)	1.51±0.87 (0.33–2.73)	0.055±0.019 (0.028–0.075)	0.5±0.74 (0.09–1.80)	0.036±0.050 (nd–0.098)	2.59±1.94 (0.99–5.92)
Intestine	0.67±0.20 (0.47–0.98)	0.035±0.012 (0.017–0.046)	8.11±2.65 (4.40–10.19)	4.54±0.63 (3.82–5.04)	nd	0.3±0.12 (0.21–0.48)	0.22±0.20 (0.016–0.55)	52.59±18.88 (36.93–83.40)
Heart	0.22±0.032 (0.18–0.26)	0.0022±0.0021 (nd–0.0055)	9.04±0.86 (8.29–10.51)	8.85±3.61 (2.92–11.63)	0.0021±0.0047 (nd–0.011)	0.53±0.37 (0.19–1.15)	0.2±0.12 (0.10–0.41)	37.9±8.00 (24.60–44.32)
Brain	0.9±0.14 (0.74–1.05)	0.00046±0.0024 (nd–0.0043)	5.69±0.82 (4.56–6.75)	2.59±2.24 (0.90–6.51)	nd	0.13±0.048 (0.078–0.18)	0.33±0.25 (0.16–0.77)	7.24±3.51 (3.31–11.49)
Environmental samples								
Sediment	17.72±3.74 (11.36–22.55)	0.43±0.11 (0.60–0.31)	49.02±14.66 (27.35–66)	49.89±21.75 (28.65–97.84)	0.31±0.19 (0.15–0.73)	41.6±18.30 (21.69–79.82)	19.59±14.75 (10.43–45.81)	401.76±221.77 (212.41–877.56)
Water	0.0076±0.00062 (0.0069–0.0089)	nd	nd	nd	0.00012±0.000076 (nd–0.00028)	0.0012±0.00045 (0.00094–0.0022)	nd	0.0078±0.0039 (nd–0.012)

Data was shown as mean±standard deviation; maximum and minimum values were in parenthesis

general concentration ranges were from 0.37 to 45.81 ng g^{-1} in the muscle samples and from 1.89 to 111.20 ng g^{-1} in the liver samples. Compared with those from other studies, the mean concentrations (5.38 ng g^{-1}) in the bighead carp muscle and (7.17 ng g^{-1}) in the silver carp muscle were higher than those from the Taihu Lake and Lake Ziway, but lower than those found in the Poyang Lake and *Hemiculture leuciscultures* from the Gaobeidian Lake (Table S1). Compared with fish collected from other water bodies, fish in the East Lake were moderately contaminated by OCPs.

A preferential accumulation in the muscle compared with the liver was observed for OCPs except β-HCH in bighead carp as well as *p*'*p*-DDE and β-HCH in silver carp (Table 2). Sun et al. reported 16 OCPs in different organs of silver carp, and the results revealed that the concentrations of OCPs in the muscle and liver were similar. HCHs in the liver were slightly more than those in the muscle, and DDTs in the muscle were marginally more abundant than those in the liver (Sun et al. 2005). A report from Miranda et al. was different: OCPs in the liver of *Hoplias malabaricus* were more than those in the muscle (Miranda et al. 2008). On the contrary, Svobodovà et al. observed more DDTs in the muscle of wels in contrast with the liver (Svobodova et al. 1995). Biotransformation of compounds was always conducted in the liver, and the main function of muscle was mobility (van der Oost et al. 2003). Different accumulation processes might occur in the two tissues which resulted in the difference of chemical accumulation.

As is shown in Table 3, eight elements were detectable in both bighead carp and silver carp except Hg in the intestine and brain of the two fishes. Average concentrations (dry weight, dw) of ΣHMs were 352.48 and 345.20 ng g^{-1} in bighead carp and silver carp, respectively (Table 1). Cu, Zn, and Cr were the predominant elements among eight elements. The general order of HMs in the muscle was Cd<Hg<Pb<Ni<As<Cu<Cr<Zn, in the liver was Hg<Ni<Pb≈Cd<As<Cr<Cu<Zn, in the bladder was Cd<Pb<As≈Hg<Ni<Cu<Zn<Cr, in the intestine was Hg<Cd<Pb<Ni<As<Cu<Cr<Zn, in the heart was Hg<Cd<Pb<As<Ni<Cu<Cr<Zn, and in the brain was Hg<Cd<Pb≈Ni<As<Cu<Cr<Zn. The results of the present study were compared with previous studies of HMs in freshwater fishes of different places and listed in Table S2. For instance, As was found at an average level of 0.39 mg kg^{-1} in the muscle in the present study which was comparable to that in the Danube River, Serbia (0.35 mg kg^{-1}), almost three times of that in the Taihu Lake, Jiangsu, China (0.15 mg kg^{-1}), but much lower than that in the Yangtze River, Nanjing, China (3.45 mg kg^{-1}) (Fu et al. 2013; Hao et al. 2013). Hg levels found in the present study were one order of magnitude lower than those in most of other studies listed in Table S2. The contamination of HMs in fish in this area was at a low level.

Fish tissues, i.e., the muscle, liver, brain, heart, bladder, and intestine, were investigated for the tissue distribution of HMs. As could be seen from Fig. 2, Cu, Zn, and Cr were more than other elements in both two fishes. Numerous studies also reported Cu and Zn were the most abundant elements in fish (Ahmad et al. 2014; Djedjibegovic et al. 2012; Guerin et al. 2011; Turkmen et al. 2013). They were all essential elements for fish, particularly Cu participating in synthesis of several enzymes and hemoglobin (Sivaperumal et al. 2007). Accumulation of Zn trended to occur in more fatty tissue of fish, which could be verified by the concentrations of more fatty silver carp which were higher than those of bighead carp (Rahman et al. 2012). Except Hg, the other seven elements were more easily concentrated in the liver, intestine, and heart which were more metabolically active organs than the muscle and bladder. Many researchers considered that the liver and intestine had relative high potential for accumulation of HMs (Demirak et al. 2006; Karadede et al. 2004; Uysal et al. 2009; Yang et al. 2007; Yılmaz et al. 2007). This point was generally in accordance with our studies. The highest content of Hg was found in the bladder of both fishes. Bladder is an organ related to adjustment of specific gravity of fish, and both of the two fishes we studied are physostomous which means adjustment of specific gravity of fish is conducted by gas exchange with atmosphere. Some study has already reported that one important approach for Hg into the East Lake was by borne particulate in atmosphere, which indicated that the exposure of fish bladder to atmosphere containing Hg particulate might be the reason why bladder accumulated more Hg than other tissues (Su et al. 2004).

Potential sources of OCPs and HMs in water, sediment, and fish samples

Organochlorine pesticides

p,*p*'-DDE and *p*,*p*'-DDD were detectable in all of the samples, and *p*,*p*'-DDT was detected only in 12 of the 42 water samples. It is well known that DDT/(DDE+DDD)>1 indicates new pollution sources which might be existed while DDT/(DDE+DDD)<1 suggests historical use of DDT. Namely, the historical use of DDT was the main source of DDTs in water. However, the ratio of DDT/(DDE+DDD) exceeding 1 in 11 of the 42 water samples meant new input of DDT, which was consistent with the conclusions from Yang et al. (2014). Although the use of DDT has been prohibited since 1983 in China, some reports showed that the usage of DDT for malaria control was still present in our country (Qiu et al. 2005). *p*,*p*'-DDE was the most abundant in fish samples, where the similar results have also been documented in other studies (Eqani et al. 2013; Hu et al. 2010; Sharma et al. 2009). However, in the present study, the non-detectable levels of *p*,*p*'-DDT in both studied fish species might be due to low exposure level

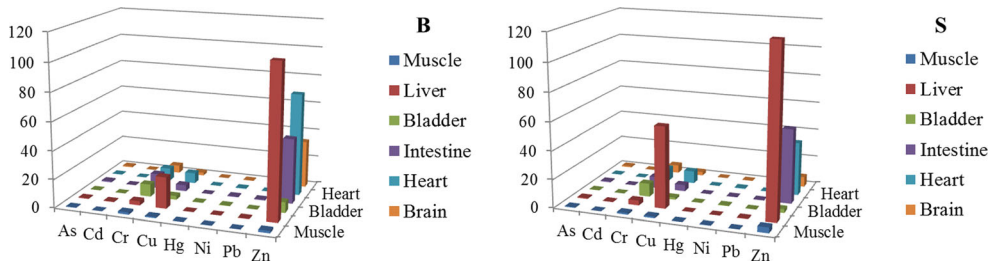

Fig. 2 Heavy metals in tissues of bighead carp (*B*) and silver carp (*S*) (mg kg^{-1}, dw)

since bighead and silver carps are both at low trophic position and DDT was not detected in most of the water and sediment samples.

Gamma-HCH accounted for the most percentage of HCHs in muscle samples. However, in the liver samples, β-HCH accounted for the most percentage of HCHs. Technical HCH usually contains α-HCH (55–90 %), β-HCH (5–14 %), γ-HCH (8–15 %), and δ-HCH (2–16 %), while lindane contains more than 98 % of γ-HCH (Ge et al. 2013). Relative abundances of HCH congeners in fish samples are shown in Fig. 3. HCHs in surface water consisted of 15 % α-HCH, 39 % β-HCH, 11 % γ-HCH, and 35 % δ-HCH. The ratios of α-/γ-HCH varied from 0.74 to 1.87 indicating historical use of technical HCHs and lindane in this area, which was consistent with those from previous studies (Yang et al. 2014; Yun et al. 2014). The ratios of α/γ-HCH in all fish samples ranged from 0.58 to 1.89 indicating the same source of HCHs as that in water and sediments, which also confirmed that these two fishes were good bio-indicators for HCH pollution monitoring in the East Lake.

Regarding rest of the OCPs analyzed, heptachlor, heptachlor epoxide, aldrin, and methoxychlor contributed about one third of the total OCPs in fish. The ranges of concentration of heptachlor were 1.11–11.59, 1.9–27, and 0.14–14.54 ng L^{-1} in water, sediments, and fish samples, respectively, while concentrations of heptachlor epoxide (degradation product of heptachlor) were in the range of nd–0.30, nd–7.6, and nd–7.79 ng g^{-1} in water, sediments, and fish samples, respectively (Yun et al. 2014). Recent usage of heptachlor in the East Lake was revealed by the ratio of heptachlor to its degradation product. Aldrin accounted for about 7 % of total OCPs in fish in present study has never been produced and used on a large scale in China. The most probable source of aldrin in the East Lake might be the global migration (MEPC 2011).

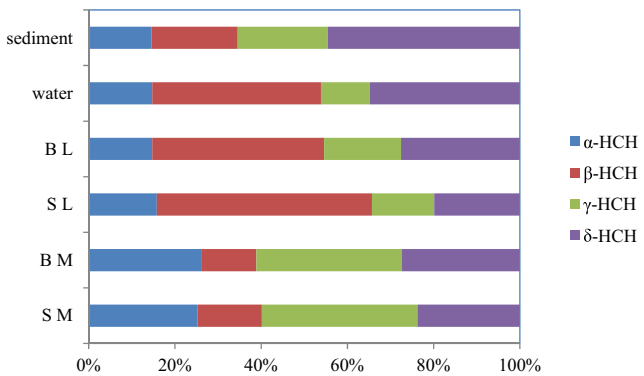

Fig. 3 Relative abundances of HCH congeners in water, sediment, and fish samples (*B* bighead carp, *S* silver carp, *M* muscle samples, *L* liver samples)

Heavy metals

It has been reported that no point source pollution of HMs was found in the proximity of the East Lake (Liu et al. 2014). The most probable origins of HMs in the East Lake were natural and anthropogenic activities such as weathering; erosion of bed rocks; ore deposits; and wastewater from local residents, hospitals, and industries. Table 4 reports correlations between the eight elements in both fish and sediment samples listing Spearman's rho correlation coefficients. Moderate correlations were found in three of the eight elements (Cd-Ni, Cd-Pb, Pb-Ni, $p<0.01$) in fish. However, four of the eight elements (Cu-Cr, Cu-Ni, Cu-As, Cr-Ni, $p<0.01$) were correlated significantly in sediment, which was different from that in fish. Therefore, the conclusion that two elements which present a correlated relationship had the same origin like many studies reported should not be made arbitrarily (Joksimovic et al. 2011; Rahman et al. 2012; Yang et al. 2007). There might be totally different patterns in sediment. Only four elements were detectable in water, so the data were not analyzed for Spearman's correlation.

Bioaccumulation factors of OCPs in fish samples

The bioaccumulation factor (BAF) of fish is the ratio of concentration of a chemical inside the fish to the concentration of surrounding water. Due to the lipophilic characteristic of OCPs, the concentrations in organism needed to be normalized by lipid content. In all investigated samples, BAFs of methoxychlor, heptachlor epoxide, and *p,p*'-DDE were higher than those of the other seven chemicals, especially those BAFs of methoxychlor and heptachlor epoxide which were remarkably high. BAFs of the rest of the seven chemicals were fairly low or none (Fig. 4). Higher BAFs of the three chemicals might be ascribed to the relative high log K_{ow} of them (Table 2). By comparing BAFs of the two fishes, a transparent trend could be obtained: BAFs in bighead carp were higher than those in silver carp. It is known that bioaccumulation of pollutants in fish was potentially

Table 4 Spearman's rho correlation among HMs in fish and sediments from the East Lake

Fish muscle and liver	Cr	Ni	Cu	Zn	As	Cd	Hg	Pb
Cr	1.000	0.018	0.191	−0.105	0.023	0.567**	0.035	0.421
Ni		1.000	0.027	−0.051	0.314	0.699**	−0.065	0.639**
Cu			1.000	−0.095	0.493*	0.077	0.212	0.281
Zn				1.000	−0.185	−0.072	0.259	0.034
As					1.000	0.184	−0.163	0.197
Cd						1.000	−0.185	0.752**
Hg							1.000	−0.005
Pb								1.000
Sediment	Cr	Ni	Cu	Zn	As	Cd	Hg	Pb
Cr	1	0.976**	0.952**	0.214	0.810*	0.5	0.643	−0.143
Ni		1	0.976**	0.19	0.786*	0.381	0.667	−0.238
Cu			1	0.238	0.857**	0.452	0.738*	−0.214
Zn				1	0.214	0	0.333	0.476
As					1	0.643	0.69	0.19
Cd						1	0.143	0.095
Hg							1	0.048
Pb								1

*$p<0.05$ (two-tailed); **$p<0.01$ (two-tailed)

controlled by many physiological and environmental elements such as trophic levels, age, lipid content of fish, environmental concentration of pollutants, and so on (Nichols et al. 1998; Russell et al. 1999). Two fishes (bighead carp and silver carp) had similar trophic level and size, so the possible effects induced by the two environmental elements could be eliminated (Table 1). In general, lipid content always correlated with concentration of hydrophobic chemicals positively. However, in our study, the lipid content of silver carp was almost two times of that in bighead carp while the BAFs of silver carp were lower than those of bighead carp. There were also some studies pointing out no relationships between lipid content and concentration of hydrophobic chemicals (Crimmins et al. 2002; Jackson et al. 2001). BAF is always assessed based on the hypothesis that is the sampled organisms are at steady state with surrounding water; however, the steady state is nearly impossible in natural environment. In addition, silver carp and bighead carp were all pelagic fishes which meant they could be more easily affected by dynamic water. Therefore, lipid content was not the main factor resulting in differences of BAFs of the two fishes and the difficulty for obtaining the actual concentration of pollutants in ambient water enlarged the error of BAFs.

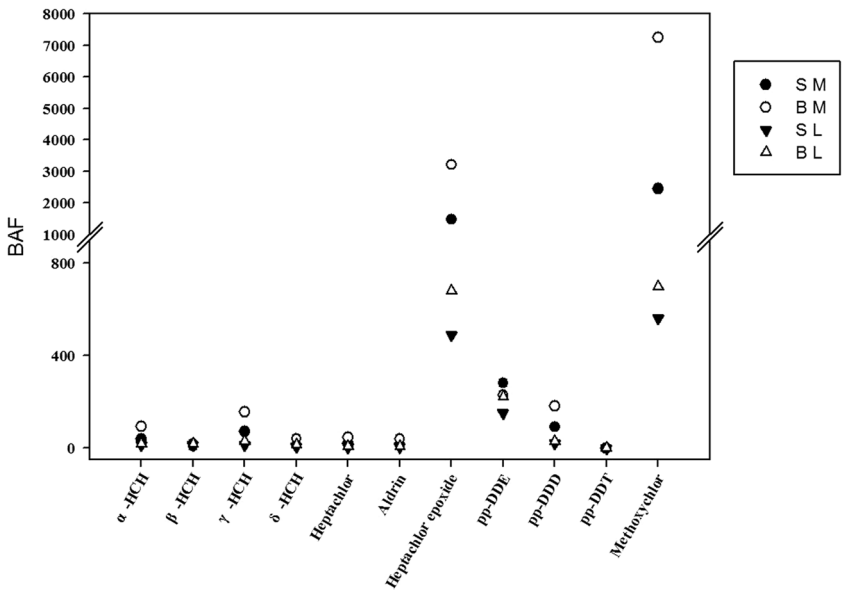

Fig. 4 BAFs of OCPs in fish samples (*B* bighead carp, *S* silver carp, *M* muscle samples, *L* liver samples)

Table 5 Comparison of the estimated daily intake (EDI) of DDTs, HCHs, and heavy metals from fish species studied with different reference values

	EDI (ug/kg bw/day)		Reference value (ug/kg bw/day)		References
	Bighead carp	Silver carp	Value	Criteria	
As	0.56	0.91	15	PTWI	JECFA (2010)
Cd	0.0053	0.0042	7	PTWI	CE (2011)
Cr	0.41	0.31	0.8	ESADDI	NRC (1989)
Cu	0.22	0.29	500	PMTDI	CE (2011)
Hg	0.023	0.020	4	PTWI	JECFA (2010)
Ni	0.039	0.075	5	PMTDI	CE (2011)
Pb	0.11	0.26	25	PTWI	CE (2011)
Zn	0.44	1.05	1000	PMTDI	CE (2011)
∑DDTs	0.0037	0.010	20	ADI	Faroon and Harris (2002)
∑HCHs	0.0058	0.0077	0.3	ADI	EFSA (2005)

PTWI provisional tolerable weekly intake, *ESADDI* estimated safe and adequate daily dietary intake, *PMTDI* provisional maximum tolerable daily intake, *ADI* acceptable daily intake

Risk assessment of OCPs and HMs in the fish collected in the East Lake

One of major ways for contaminants into human body is fish consumption. EDIs were used to carry out the assessment of potential human exposure to OCPs and HMs by calculating with the mean concentration of target contaminants in fish muscle. Table 5 shows the comparison of our results with other prevailing criteria issued by different regulatory agencies. All of the values from the present study were several orders of magnitude lower than the listed standards except for Cr. For bighead carp, the EDI of Cr was 0.41, and for silver carp, was 0.31, which were almost half of 0.8 given by reference. The results implied that potential health risks related to

Table 6 Non-carcinogenic (THQ) and carcinogenic risks (*R*) of studied contaminants

Chemicals	R_{FD} (mg/kg/day)	SF (mg/kg/day)$^{-1}$	THQ		*R*	
			Silver carp	Bighead carp	Silver carp	Bighead carp
α-HCH	0.008	6.3	2.32E−04	1.80E−04	1.17E−05	9.09E−06
β-HCH	-	1.8			1.98E−06	1.27E−06
γ-HCH	0.0003	1.1	8.86E−03	6.23E−03	2.92E−06	2.05E−06
δ-HCH	-	-	-	-	-	-
Heptachlor	0.0005	4.5	5.25E−03	3.86E−03	1.18E−05	8.68E−06
Aldrin	0.00003	17	6.16E−02	4.75E−02	3.14E−05	2.42E−05
Heptachlor epoxide	0.000013	9.1	1.12E−01	7.92E−02	1.33E−05	9.37E−06
p,*p*'-DDE	-	0.34	-	-	2.36E−06	6.20E−07
p,*p*'-DDD	-	0.24	-	-	6.49E−07	4.09E−07
Methoxychlor	0.005	-	4.61E−04	4.42E−04	-	-
HI and R_T	-	-	1.89E−01	1.37E−01	7.61E−05	5.57E-−05
As	0.0003	1.5	4.04E−01	2.56E−01	1.82E−04	1.15E−04
Cd	0.001	-	6.08E−04	7.36E−04	-	-
Cr	0.003	0.5	9.97E−02	1.33E−01	1.50E−04	1.99E−04
Cu	0.04	-	6.89E−03	5.37E−03	-	-
Hg	0.0001	-	2.83E−02	3.23E−02	-	-
Ni	0.02	-	3.61E−03	1.91E−03	-	-
Pb	0.004	-	8.75E−03	3.62E−03	-	-
Zn	0.3	-	3.34E−03	1.42E−03	-	-
HI and R_T	-	-	5.55E−01	4.34E−01	3.31E−04	3.14E−04

"-" means the data is not available at this time according to the risk-based concentration table (US EPA 2009)

daily intake of OCPs and HMs through consumption of bighead carp and silver carp were insignificant.

Table 6 lists the results of health risk assessment of the two fishes. The THQs were all less than 1.0. The THQs of As, Cr, Hg, aldrin, and heptachlor epoxide were one or two orders of magnitude higher than those of the other contaminants in which the THQ of As was the highest. The HI of OCPs which meant the non-carcinogenic risks were 0.19 for silver carp and 0.14 for bighead carp, lower than 1.0. All of the HIs of HMs were also lower than 1.0, which were 0.56 for silver carp and 0.43 for bighead carp, respectively. The HI of OCPs and HMs indicated lower health risk to human via fish consumption in study area. From another aspect, the R and R_T of OCPs associated with fish consumption whose values were several orders of magnitude lower than 1×10^{-4} indicated that the cancer risk of exposure to these contaminants by fish consumption was negligible. Nevertheless, the Rs of As and Cr were all higher than 1×10^{-4} in both two fishes, which suggested that the cancer risk of As and Cr caused by fish consumption cannot be ignored.

Conclusions

Two commonly consumed fishes from Wuhan, China, were collected and analyzed for 11 organochlorine pesticides (OCPs) and eight heavy metals (HMs). The concentrations of OCPs and HMs in fish were all lower or slightly higher than those in other water bodies in the world. Bighead carp trended to accumulate more OCPs than silver carp, and a preferential accumulation in the muscle compared with the liver was observed for most OCPs. Zn as an essential element was most abundant in two fishes. The liver and intestine of the two fishes were proved to be target organs of HMs. The EDIs in our study were all lower than criteria issued by different regulatory agencies. The HI of OCPs and HMs were both lower than 1.0, which indicated lower non-carcinogenic risk to human via fish consumption in study area. Carcinogenic risks of exposure to OCPs by fish consumption were negligible, but carcinogenic risks of exposure to As and Cr were high. In addition, we assessed the consumption rate of fish at the upper band in the present study which meant the actual risks might be lower.

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