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Tissue distribution, bioaccumulation, and carcinogenic risk of polycyclic aromatic hydrocarbons in aquatic organisms from Lake Chaohu, China

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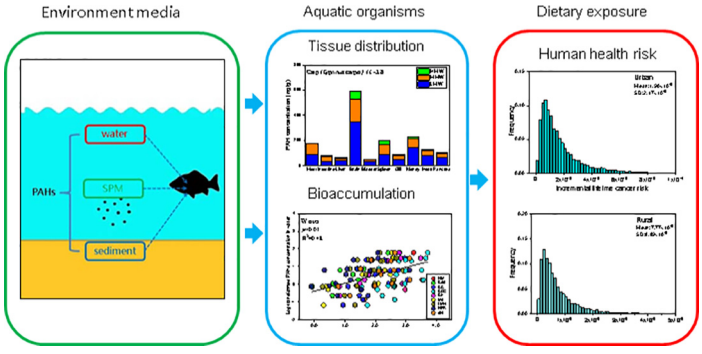
HIGHLIGHTS

- Different tissue distribution patterns of PAHs in three fish species were observed.
- PAHs levels in aquatic organisms were influenced by environment media.
- Bioaccumulation factors and influencing factors were revealed.
- Trophic dilution effect was detected for PAHs in the freshwater food web.
- Potential cancer risk was found in rural and urban consumers of aquatic products.

GRAPHICAL ABSTRACT



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ABSTRACT

Freshwater products consumed in the diet are among the major sources of exposure of humans to polycyclic aromatic hydrocarbons (PAHs). In this study, eight freshwater organisms and environmental samples were collected from Chaohu Lake, the fifth-largest lake in China. The levels of PAHs in the collected organisms were measured using GC–MS. Tissue distribution characteristics in three fish species were studied. Relationship between residual levels and environment concentrations were analyzed and bioaccumulation effect and influencing factors were identified. Finally, the potential carcinogenic risk of aquatic product intake was estimated. The concentrations of ΣPAHs in aquatic organisms varied from 18.4 to 398 ng/g, with a mean value of 157 ± 125 ng/g. For carp, the highest ΣPAHs level was detected in the brain with concentration of 591 ng/g. For topmouth culter, and bighead fish, the organs with the greatest ΣPAHs concentration were gills (440 ng/g) and muscles (200 ng/g), respectively. Significant correlations were found between the PAH content in environment media including water, SPM, sediment and PAH content in aquatic animals. The calculation of food web magnification factors and risk assessment indicates that although the PAH concentration diluted with the increase of the trophic level, PAHs exposure through the aquatic products intake still poses potential carcinogenic risk. The incremental lifetime cancer risk values were 7.68×10^{-6} and 4.75×10^{-6} in urban and rural populations, respectively.

1. Introduction

Pollution of lakes with polycyclic aromatic hydrocarbons (PAHs) has become one of the major environmental problems in China. Freshwater

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fishery is a big aquaculture sector in China and the number of consumers of freshwater fish is high. Freshwater fish production in China increased from 27.5 million tons in 2013 to 31.2 million tons in 2017 (Sun Lin and Shumin, 2018). The production of aquatic products per capita in 2017 was 46.3 kg, showing an increase of 1.6% compared to that reported in 2016. Consumption of aquatic products in large amounts may pose a threat to human health (Khairy et al., 2014) and has been a major route of exposure of humans to PAHs (Olson et al., 2016). Moreover, PAHs at a low level in environmental media may concentrate in the tissues of organism through the process of bioaccumulation and reach a level that are high enough to pose health risks to the consumers (Rotkin-Ellman et al., 2012; Yu et al., 2019).

PAHs are typical persistent organic pollutants with potentially toxic, mutagenic, and carcinogenic properties (Fernandes et al., 1997; Larsen and Baker, 2003; Qin et al., 2013; Shrivastava et al., 2017). They have been proven to be among the potential causes of skin, lung, bladder, and gastrointestinal cancers (Boffetta et al., 1997; Kim et al., 2013; Kuang et al., 2013; Miller et al., 2013). PAHs accumulate in aquatic organisms by direct uptake from water through gills or skin, or through the ingestion of suspended particles and contaminated food (van der Oost et al., 2003). Depending on their lipophilicity and resistance to metabolism, some PAHs can biomagnify in organisms at higher trophic levels (Broman et al., 1992; Fisk et al., 1998). The biomagnification effect considerably increases the risk of exposure to PAHs through ingestion of freshwater products (Burkhard and Lukasewycz, 2000; Pacini et al., 2013; Phillips, 1999). It has been reported that fish meat and fish oil are sources of a variety of organic pollutants, including PAHs (Conti et al., 2011; Ohiozebau et al., 2017). Additionally, epidemiological evidence also confirms the relationship between consumption of aquatic products and internal exposure of humans to PAHs. A significant linear relationship was reported between the concentration of PAHs in the blood plasma of 111 healthy residents and consumption of seafood diet (Qin et al., 2011). Thus, aquatic organisms present in natural water are not only the main “sink” of PAHs in the environment, but are also the “source” of exposure of humans to PAHs.

The profiles of PAHs in the aquatic environment, as well as their trophic transfer and effects on aquatic biota, have been reported previously (Ma et al., 2014; Peng et al., 2014; Zhang et al., 2010). Despite the progress in these directions, studies on three issues remain scarce. First, the relationship between residual levels in organisms and environmental media is seldom reported. Second, the characteristics of bioaccumulation of PAHs in different tissues remain unclear. Third, the factors influencing bioaccumulation are seldom studied. Hence, further studies on PAHs should be performed to obtain a comprehensive understanding of the environmental behavior of PAHs in freshwater organisms and their potential carcinogenic risk to consumers. In this study, the contents of 16 priority control PAHs in aquatic organisms and environmental media were measured, trophic levels were determined, and exposure factors through fish ingestion by the population were estimated. The aims of this study were to: (1) investigate the residual levels of 16 priority PAHs in freshwater organisms and their tissue distribution characteristics in three fish species from Lake Chaohu, (2) elucidate the relationship between the concentrations of these PAHs in organisms and environment media, (3) explore the bioaccumulation and biomagnification factors, and (4) estimate the potential carcinogenic risk through the intake of freshwater organisms.

2. Materials and methods

2.1. Research area and sample collection

Lake Chaohu, the fifth-largest freshwater lake in China, is located near the Yangtze River delta in one of the most developed regions of China. With rapid urbanization of the surrounding area, Lake Chaohu is being increasingly polluted by PAHs, which is expected not only to damage the lake ecosystem but would also compromise the safe use of lake water for drinking. Moreover, accumulation of PAHs in fish tissues also poses a risk to human health upon dietary intake of these products.

Aquatic organisms, including spotted steed (*Hemibarbus maculatus, HM*), carp (*Cyprinus carpio, CC*), snail (*Cipangopaludina chinensis Gray, CCG*), topmouth culter (*Culter eryropterus, CE*), bluntnose black bream (*Megalobrama amblycephala, MA*), Chinese white prawn (*Leander modestus Heller, LMH*), whitebait (*Hemisalanx prognathus Regan, HPR*), and bighead carp (*Aristichthys nobilis, AN*), were collected in January 2010 (Fig. S1). Samples of sediment ($n = 15$) were collected in August 2009. Water and SPM samples were collected once a month at three sampling sites, from May 2010 to April 2011 to minimize the influence of seasonal variation and extreme values. The sampling sites were provided in Fig. S2.

Samples of muscle of aquatic organisms were collected and weighed. For the three large freshwater fish species carp, topmouth culter, and bighead fish, a total of 10 organs including meat, intestine, liver, brain, bladder, spleen, gill, kidney, heart and pancreas were collected. Muscle on both sides of dorsal fin and chest was combined. To minimize the effects of variation among individuals, samples from three to five individuals of the same species were pooled. Samples of tissues were freeze-dried (FDU-830, Tokyo Rikakikai Co., Japan) and weighed to determine their dry mass. The samples were ground to a granular powder with a ball mill (MM400, Retsch GmbH, Germany) and sealed in amber glass bottles until the analysis.

Twenty liters of water was collected from each sampling site. After shaking and mixing, a one-liter aliquot of each was filtered through a 0.45-μm glass fiber filter (combusted at 450 °C for 4 h) using a filtration device consisting of a peristaltic pump (80EL005, Millipore Co., USA) and a filter plate with a diameter of 142 mm. Surrogate standards of 2-fluoro-1,1′-biphenyl and *p*-terphenyl-d14 (J&K Chemical, USA, 2.0 mg/mL) were added to the water samples before extraction to indicate the recovery. The SPM samples in the glass fiber filter were freeze-dried and weighed on an analytical balance. Samples of surface sediment were collected using a grab sampler, freeze-dried, ground, and sieved through a 70-mesh sieve before extraction.

2.2. Sample preparation, instrumental and quantitative analysis

Two-grams of powdered samples from aquatic organisms were placed in an extraction tube, and a recovery indicator and an internal standard were added. After microwave extraction, extracts were pressure filtered and concentrated to approximately 1 mL using a rotary evaporator; the volume was made to 1 mL after then 10 mL ethyl acetate was added. Samples were filtered through a 0.45-μm membrane, and then transferred to gel permeation chromatography (GPC) vials. After adding 3 mL ethyl acetate, the samples were purified by GPC (GPC800+, Lab Tech Ltd., China) using a Bio Beads SX-3 column (300 mm × 20 mm, Bio-Rad Laboratories, Inc. America). The injection volume was 2 mL. A solution of 1:1 ethyl acetate/hexane was used to elute fractions at a flow of 5 mL/min. The fractions eluting from 2 min to 10 min contained macromolecules, including lipids, and were collected in a weighed eggplant-shaped flask. After drying the eluate in a rotary evaporator for 24 h to a constant mass, the flask was weighed again. The quantity of lipid was the difference in the mass of the flask with and without the dried eluate. The fractions eluting from 10 min to 22 min contained the target compounds. The eluate was concentrated to approximately 1 mL by rotary evaporation and then reconstituted to 1 mL, after which 10 mL hexane was added. Subsequently, the concentrates were loaded onto silica gel solid phase extraction (SPE) cartridges (6 mL, 500 mg, Supelco Co, USA). These cartridges were conditioned with 10–15 mL hexane before use. After loading, elution was performed by passing hexane through the cartridge (two times, 5 mL per elution) followed by a mixture of dichloromethane (DCM) and hexane (V:V = 1:1, four times, 5 mL per elution). Extracts were concentrated to 1 mL, transferred to vials, and sealed for analysis.

Filtered water samples were extracted using an SPE system (Supelco). C18 cartridges (500 mg, 6 mL, Supelco) were prewashed with DCM and conditioned with methanol and deionized water. A 1-L sample of water was passed through the SPE system. The cartridges were eluted with 10 mL DCM. The volume of eluate was reduced using a vacuum rotary evaporator (R-201, Shanghai Shen Sheng Technology Co., Ltd., Shanghai, China) placed in a water bath and then adjusted to 1 mL with hexane. Internal standards were added prior to the analysis by gas chromatography (GC). The SPM and sediments were extracted with 25 mL of hexane/acetone mixture (1:1) using a microwave-accelerated reaction system (CEM Corporation, Matthews, NC, USA). The microwave power was set at 1200 W, and temperature was ramped to 100 °C over 10 min and then held at 100 °C for another 10 min. Extracts of both the SPM sediments were concentrated to 1 mL by rotary evaporation at a temperature less than 38 °C and were then transferred to a silica/alumina chromatography column for cleanup. The eluted solution was collected, concentrated, converted to hexane solution, and then spiked with internal standards.

All the samples were identified and quantified using a gas chromatograph with a mass selective detector (MSD; Agilent 6890GC/5973MSD). A capillary column with dimensions 30 m × 0.25 mm i.d. and a 0.25-μm film thickness (HP-5MS, Agilent Technology) was used. The column temperature was programmed to increase from 60 °C to 280 °C at 5 °C/min and then held at this temperature for 20 min. The MSD was operated in the electron impact mode at 70 eV, and the ion source temperature was 230 °C. Mass spectra were recorded using the selected ion monitoring (SIM) mode. The concentrations of 16 PAHs were detected; these included low-molecular-weight (LMW, including Nap, Ace, Acy, Flo, Phe, Ant, and Fla), moderate-molecular-weight (MMW, including Pyr, BaA, Chr, BbF, and BkF), and high-molecular-weight PAHs (HMW, including BaP, DahA, IcdP, and BghiP).

Quantification was performed using the internal standards, Nap-d8, Ace-d10, Ant-d10, Chr-d12, and Perylene-d12 (J&K Chemical, Beijing, China). All the solvents used were HPLC-grade (J&K Chemical). All glassware was cleaned using an ultrasonic cleaner (KQ-500B, Kunshan, China) and heated at 400 °C for 6 h. Field blanks were collected at each sample site. The laboratory blanks and sample blanks were analyzed along with the samples. The recovery of the methods was determined by spiking the standard mixture of 16 PAHs (the standard mixture of 16 PAHs from J&K Chemical Ltd., USA) into the samples and performing the entire analytical methods. Method recoveries and detection limits (MDLs) are shown in Table S1 in the supporting information.

For PAHs detected in blank samples, the MDLs were set to be 4.54 times (degree of freedom 2, level of significance at $\alpha = 0.05$) the standard deviation of concentration in the blank samples, and concentrations of compounds were corrected for the concentrations in the blank. For compounds that were not detected in blank samples, the instrumental minimum detectable amounts were established as a signal-to-noise ratio of 3.

2.3. Parameter measurement and statistical analyses

The content of dissolved organic carbon (DOC) in samples of water, particulate organic carbon (POC) content in SPM samples, and total organic carbon (TOC) content of sediments were determined using a total organic carbon analyzer (TOC-5000A; Shimadzu Corp., Japan). Lipids in the samples of fish were determined gravimetrically in hexane extracts.

Trophic levels (TL) of aquatic organisms were determined using stable nitrogen isotopes (Fisk et al., 2001). The samples of algae from Chaohu Lake were used to estimate the $\delta^{15}N$ baseline and were assumed to represent the trophic position 1.0. Stable isotope ratios ($^{15}N/^{14}N$) were determined by mass spectrometry (Finnigan MAT 253, Thermo Fisher Scientific Inc., USA). The abundance of the stable Nitrogen isotope ($\delta^{15}N$) was expressed as parts per thousand (‰) deviation from the standard, according to the following equation:

$$\delta^{15}N = \left[\left({}^{15}N/{}^{14}N_{sample}/{}^{15}N/{}^{14}N_{standard}\right) - 1\right] \times 10^3 \quad (1)$$

The $^{15}N/^{14}N_{standard}$ values were based on N_2 gas. Based on the measured nitrogen isotope ratios in organisms, TLs were calculated using the following formula (Winemiller et al., 2007):

$$TL = \left[\left(\delta^{15}N_{consumer} - \delta^{15}N_{reference}\right)/3.3\right] + 1 \quad (2)$$

where $\delta^{15}N_{reference}$ is the mean of algae samples and 3.3 is an estimated $\delta^{15}N$ value, which is the enrichment between consumers and their food.

2.4. Bioaccumulation

Bioaccumulation is a fundamental process in environmental toxicology, which controls the internal dose of chemicals (Arnot and Gobas, 2004; Mackay and Fraser, 2000). Bioaccumulation factors (BAFs) are also important indicators in determination of the environmental quality guidelines, thereby categorizing substances that are potential hazards and quantifying hazards or risks posed by chemicals to ecosystems and human health (Froehner et al., 2011). The three widely used indicators, BAFs, biota-suspended solids accumulation factor (BSSAF), and biota sediment accumulation factor (BSAF), are commonly used as indicators of the tendencies of compounds to accumulate in aquatic organisms from water, suspended solids, and sediments (Burkhard, 2003; USEPA, 2003). These three factors were calculated using Eqs. (3)–(5) mentioned below:

$$BAF = C_b/C_w \quad (3)$$

$$BSSAF = C_b/C_{SPM} \quad (4)$$

$$BSAF = C_b/C_s \quad (5)$$

where C_b is the lipid-normalized PAH concentration in tissue (ng/g lipid weight, lw), C_W is the concentration of chemical that is freely dissolved in water (ng/L), and C_{SPM} and C_S are the concentrations of chemical in suspended particles and sediment (ng/g).

The food web magnification factors (FWMFs) are the incremental increase in the concentrations of lipid-normalized concentrations of residues in biota as a function of trophic level (Fisk et al., 2001). Therefore, they are better indicators for description of the trophic transfer of chemicals in food chains (Giesy et al., 2014; Mackay et al., 2014). FWMF is determined by the use of linear regression equation (Eq. (6)).

$$\ln C_L = a + (b \times TL) \quad (6)$$

where C_L is the lipid-normalized concentration of aquatic biota and TL is the trophic level. The slope b is used to calculate FWMF (Eq. (7)).

$$FWMF = e^b \quad (7)$$

Chemicals with FWMFs greater than 1 are considered to biomagnify (Nfon et al., 2008).

2.5. Characterization of health risk assessment

In accordance with the Exposure Factors Handbook (USEPA, 1997), the carcinogenic risk of PAH intake from edible parts of the fish was estimated based on the lifetime average daily dose (LADD). The LADD was calculated using the following equation:

$$LADD = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (8)$$

where C is the BaP equivalent concentration (BaP_{eq}) concentration in the diet (mg kg^{-1}), IR is the ingestion rate of food (kg/day), EF is the exposure frequency (day/year), BW is the body weight, and AT is the average time (70 years × 365 days/year for carcinogens).

The BaP_{eq} and the toxicity equivalency factors (TEFs) are often used to express the carcinogenic risk of PAH mixtures (Nisbet and Lagoy, 1992). To evaluate the total exposure of the dietary PAHs, the BaP_{eq} based on the BaP toxicity was incorporated using the following equation:

$$BaP_{eq} = \sum C_i \times TEF_i \quad (9)$$

where, C_i is the concentration of the PAH species in food, and TEF_i is the toxic equivalence factor of the congener of PAHs, i. Table S1 lists the PAHs and TEFs used in the calculation associated with the evidence of cancer in PAH-exposed individuals.

The incremental lifetime cancer risk (ILCR) of the studied population attributable to the dietary ingestion of PAHs was estimated using following equation:

$$ILCR = LADD_{BaP} \times CSF_{BaP} \quad (10)$$

where $LADD_{BaP}$ is the BaP equivalent daily dietary exposure dose by body weight (mg kg^{-1} day^{-1}). CSF_{BaP} is the oral cancer slope factor for BaP (7.3 per mg kg^{-1} day^{-1}).

3. Results and discussion

3.1. Concentrations of PAHs in environmental media and aquatic organisms

The concentrations of ΣPAHs in aquatic organisms varied from 18.4 ng/g to 398 ng/g wet mass (wm), with a mean value of 157 ± 125 ng/g wm (Table 1). Among all the aquatic organisms, the ΣPAHs was highest in snails (398 ± 266 ng/g), followed by that in spotted steeds (254 ± 34 ng/g) and bighead carps (176 ± 45 ng/g). The ΣPAHs was lowest in whitebait, with an average concentration of 18.4 ± 3.5 ng/g. The concentration of Phe (34.9 ± 23.3 ng/g) was the highest, followed by that of Chr (33.0 ± 24.8 ng/g) and Nap (28.4 ± 33.5 ng/g). The concentration of BaP (0.88 ± 0.83 ng/g) was the lowest. ΣPAHs in the muscle of aquatic organisms was greater than that reported for Lake Baiyangdian (4.76–144 ng/g) (Xu et al., 2011), and also greater than that in fish from Catalonia, Spain (14.5 ng/g) (Falco et al., 2003); however, it was less than those reported in Taiyuan (160 ng/g) (Xia et al., 2010).

3.2. Distribution of PAHs in fish tissue

The concentrations and composition of PAHs in three fish species are illustrated in Fig. 1. The organ averaged ΣPAHs were 171 ± 158, 188 ± 134, and 73.1 ± 48.8 ng/g in carp, bighead fish, and topmouth culter, respectively. For carp, the highest ΣPAHs (591 ng/g) was detected in the brain, followed by that in the kidneys (230 ng/g). For topmouth culter, the highest level was found in the gills (440 ng/g). However, for bighead fish, the highest level was found in the muscle (200 ng/g). Relatively higher levels were found in the brain, kidney, and spleen of carp and topmouth culter. The ΣPAHs in organs were dominated by LMW PAHs. The percentage of LMW PAHs ranged from 42.0%–83.7%, 39.8%–91.0%, and 48.5%–91.8% in carp, topmouth culter, and bighead fish, respectively. High ratios of MMW and HMW PAHs were found in the gills. As the major structures for breathing and filter feeding, gills are available to contact suspend particulate materials and sediment particles, which contain more HMW and MMW PAHs. For edible part of the fish, the ΣPAHs were comparable for carp and bighead fish. Topmouth culter had relatively lower PAH level and HMW ratio. Tissue distribution characteristics of cultured carp and bighead carp from Beijing marked have been reported (Wu et al., 2012). The ΣPAHs in carp brain, liver and muscle are much higher than the data reported in Beijing with values of 6.65 ng/g, 6.74 ng/g and 6.34 ng/g, respectively. Significant gaps can also be found between bighead tissues from Chaohu and Beijing market fish. The results indicated that PAHs pollution in

Table 1
PAHs concentrations in water, SPM, sediment and eight freshwater organisms (ng/g wet Mass) collected from Lake Chaohu.

Compounds	Water[25] (ng/l)	SPM[25] (ng/g)	Sediment[25] (ng/g dry mass)	HM	CC	CCG	CE	MA	LMH	HPR	AN
Nap	68.8 ± 24.0	0.52 ± 0.68	43 ± 187	32.5 ± 1.2	21.3 ± 8.8	107 ± 58	12.1 ± 1.0	28.6 ± 4.0	4.70 ± 1.51	4.55 ± 0.58	16.1 ± 4.1
Acy	1.15 ± 4.52	0.05 ± 0.51	3.49 ± 7.33	0.78 ± 0.06	0.89 ± 0.16	2.82 ± 0.56	0.71 ± 0.14	2.59 ± 0.38	0.51 ± 0.04	0.45 ± 0.01	1.31 ± 0.11
Ace	7.76 ± 5.54	0.17 ± 2.32	9.4 ± 27.3	3.42 ± 0.79	1.69 ± 1.22	15.4 ± 13.2	1.54 ± 0.38	4.90 ± 0.54	0.87 ± 0.13	0.72 ± 0.02	2.83 ± 0.31
Flo	20.3 ± 8.4	0.64 ± 2.57	50.6 ± 78.2	10.5 ± 1.6	7.56 ± 2.49	27.4 ± 15.7	5.67 ± 2.17	14.4 ± 1.6	5.32 ± 1.01	2.63 ± 0.19	12.6 ± 2.9
Phe	42.7 ± 19.2	4.6 ± 16.7	201 ± 342	55.4 ± 10.1	31.6 ± 14.4	74.2 ± 59.2	14.1 ± 6.7	31.4 ± 18.6	18.5 ± 10.7	4.68 ± 1.57	49.3 ± 13.6
Ant	2.81 ± 2.03	0.51 ± 1.34	21.9 ± 41.5	5.55 ± 1.15	3.34 ± 1.18	7.84 ± 6.67	1.30 ± 0.83	3.19 ± 1.52	1.86 ± 1.09	0.41 ± 0.21	5.70 ± 1.52
Fla	9.1 ± 10.9	1.90 ± 5.12	162 ± 404	14.4 ± 2.3	10.8 ± 6.6	16.1 ± 12.2	4.09 ± 3.29	6.07 ± 4.10	2.71 ± 1.18	2.47 ± 0.87	4.73 ± 1.43
Pyr	9.1 ± 12.0	1.12 ± 2.25	116 ± 366	23.1 ± 5.8	19.7 ± 12.3	55.4 ± 38.2	0.03 ± 0.00	17.3 ± 7.2	2.37 ± 2.11	0.03 ± 0.04	15.9 ± 4.4
Baa	0.67 ± 2.83	0.10 ± 0.17	32 ± 109	13.4 ± 2.4	9.37 ± 7.36	15.5 ± 13.0	0.64 ± 0.24	10.6 ± 13.5	3.74 ± 2.99	0.19 ± 0.09	13.0 ± 3.3
Chr	0.81 ± 1.45	0.22 ± 0.36	36 ± 114	61.8 ± 9.4	39.3 ± 40.1	60.3 ± 53.0	0.91 ± 0.24	36.0 ± 44.3	15.1 ± 12.5	0.58 ± 0.05	50.3 ± 14.1
Bbf	0.25 ± 0.23	0.12 ± 0.21	78 ± 145	7.75 ± 0.22	3.03 ± 2.30	4.40 ± 3.62	0.50 ± 0.17	2.05 ± 2.21	0.94 ± 0.56	0.12 ± 0.05	3.73 ± 1.21
Bkf	0.14 ± 0.12	0.14 ± 0.15	23.8 ± 40.3	7.37 ± 0.31	0.44 ± 0.33	0.55 ± 0.47	0.14 ± 0.08	0.21 ± 0.07	0.08 ± 0.03	0.04 ± 0.02	0.24 ± 0.13
Bap	0.09 ± 0.11	0.06 ± 0.14	31.3 ± 69.2	1.06 ± 0.29	0.99 ± 0.80	2.74 ± 2.23	0.20 ± 0.08	0.82 ± 0.74	0.37 ± 0.21	0.21 ± 0.30	0.61 ± 0.65
IcdP	0.23 ± 0.08	0.04 ± 0.04	26.2 ± 26.9	0.67	0.50 ± 0.80	6.40 ± 6.68					
DahA	0.21 ± 0.16	0.01 ± 0.01	1.17 ± 2.40		0.30	0.84 ± 0.45		0.21	0.63		
BghiP	0.19 ± 0.24	0.08 ± 0.09	32.5 ± 57.9	17.2 ± 1.4		1.50 ± 1.56		0.18	1.14 ± 0.09	1.29 ± 0.09	
ΣPAH	171 ± 71	11.8 ± 27.7	908 ± 1878	254 ± 34	150 ± 85	398 ± 266	41.8 ± 13.5	158 ± 89	58.4 ± 30.4	18.4 ± 3.5	176 ± 45
Lipid content (%)				0.47 ± 0.07	0.72 ± 0.05	1.15 ± 0.99	0.60 ± 0.07	2.37 ± 0.61	0.78 ± 0.16	0.40 ± 0.02	0.95 ± 0.37
TL				2.90 ± 0.02	2.41 ± 0.01	2.36 ± 0.02	3.61 ± 0.17	2.48 ± 0.02	2.49 ± 0.08	3.64 ± 0.02	2.73 ± 0.02
N	12	12	14	18	7	10	9	5	30	40	10

HM = Spotted steed (*Hemibarbus maculatus*); CC=Carp (*Cyprinus carpio*); CCG = Snail (*Cipangopaludina chinensis Gray*); CE = Topmouth culter (*Culter erythropterus*); MA = Bluntnose black bream (*Megalobrama amblycephala*); LMH=Chinese white prawn (*Leander modestus Heller*); HPR = whitebait (*Hemisalanx prognathus Regan*); AN=Bighead carp (*Aristichthys nobilis*); ΣPAH is the total concentration of 16 priority controlled PAHs; TL is trophic level; Unit of SPM content has been converted to ng/g.

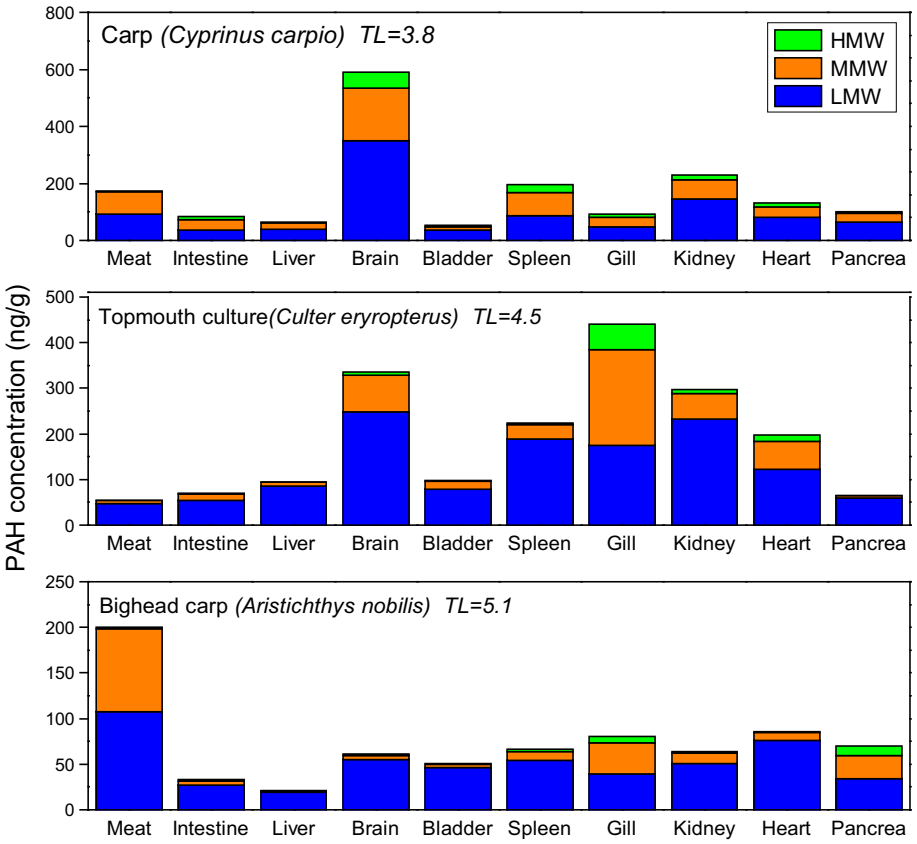

Fig. 1. Tissue distribution of polycyclic aromatic hydrocarbons in three large freshwater fish species.

natural water system is the main reason accounting for the high levels in the Chaohu fish tissues.

The tissue distribution is affected by multiple reasons including exposure, tissue lipid content, nutrition and metabolism of PAHs in fish (Jafarabadi et al., 2019; Rose et al., 2012; Zhao et al., 2014). Significant correlations have been found between lipid content in the tissue and many organic pollutants (Liu et al., 2018; Wu et al., 2012). Higher lipid content than the other tissues can be a reason accounting for the high PAH levels in brains of carp and topmouth culter. It can also be found that PAH levels in tissues of bighead carp are much lower than the other two species. Taking into account the different life habits such as the height of water and feeding habits of the three fish species, different exposure condition can be the main reason.

3.3. Relationship between the residual levels and environmental concentration

Freshwater media, including water, SPM, and sediment are the major sources for the exposure to PAHs. For fishes, PAHs can not only enter their body in the dissolved state through the gills, but also through gill filtering in the form of suspended matters; for benthos, such as snails and shrimps, PAHs can be ingested with sediment particles. Therefore, environmental factors may have an important effect on the enrichment of PAHs in aquatic animals. To eliminate the influence of fat in the study, the fat standardized muscle content of aquatic animals was used for analysis.

The Pearson's correlation analysis of lipid standardized content of PAHs with water, SPM, and sediment in aquatic animals is shown in Fig. 2. The correlation analysis of lipid-standardized content of PAHs with water, SPM, and sediment PAHs in eight aquatic animals in Chaohu Lake is shown in Figs. S3–S5. Based on the results of correlation analysis, correlations ($p < .01$) were found between the three environmental media and the content of PAHs in aquatic animals, which indicated that environmental media had a significant effect on the content of PAHs in the animals. Significant positive correlation was found between water concentration and residual levels in seven organisms, and between SPM concentration and residual levels in eight organisms. It seems that sediment has a relative low influence on animals because correlation could only be found in four aquatic organisms.

3.4. Bioaccumulation and influencing factors

Bioaccumulation is defined as a process that causes an increased chemical concentration in an aquatic organism compared to that in water due to the uptake through all the routes of exposure, including dietary absorption, transport across respiratory surfaces, and dermal absorption (Antunes et al., 2007). Three different BAFs were calculated to evaluate the influence of the different sources of contaminants by comparing the levels in the organism with those in the sources.

The BAF for eight aquatic animals ranged from 3.62 to 720. The averaged BAF of LMW, MMW, and HMW PAHs were 16.3, 74.5, and 300, respectively. It was also found that the HMW PAHs were more easily enriched in aquatic animals. The results were quite different for BSAF (range from 0.38 to 15.42). LMW had the highest average BSAF of 7.88, followed by HMW (5.79) and MMW (3.93). Compared to the bioaccumulation from water and sediment, the BSSAF values were relative lower in aquatic animals (0.004–0.18). The average BSSAF for LMW, MMW, and HMW PAHs were 0.04, 0.06, and 0.10, respectively.

Physical and chemical properties of pollutants are important factors affecting the bioaccumulation of PAHs from environmental media. We determined the relationship between the logarithms of these factors and logKow values. Significant positive correlation was found in seven

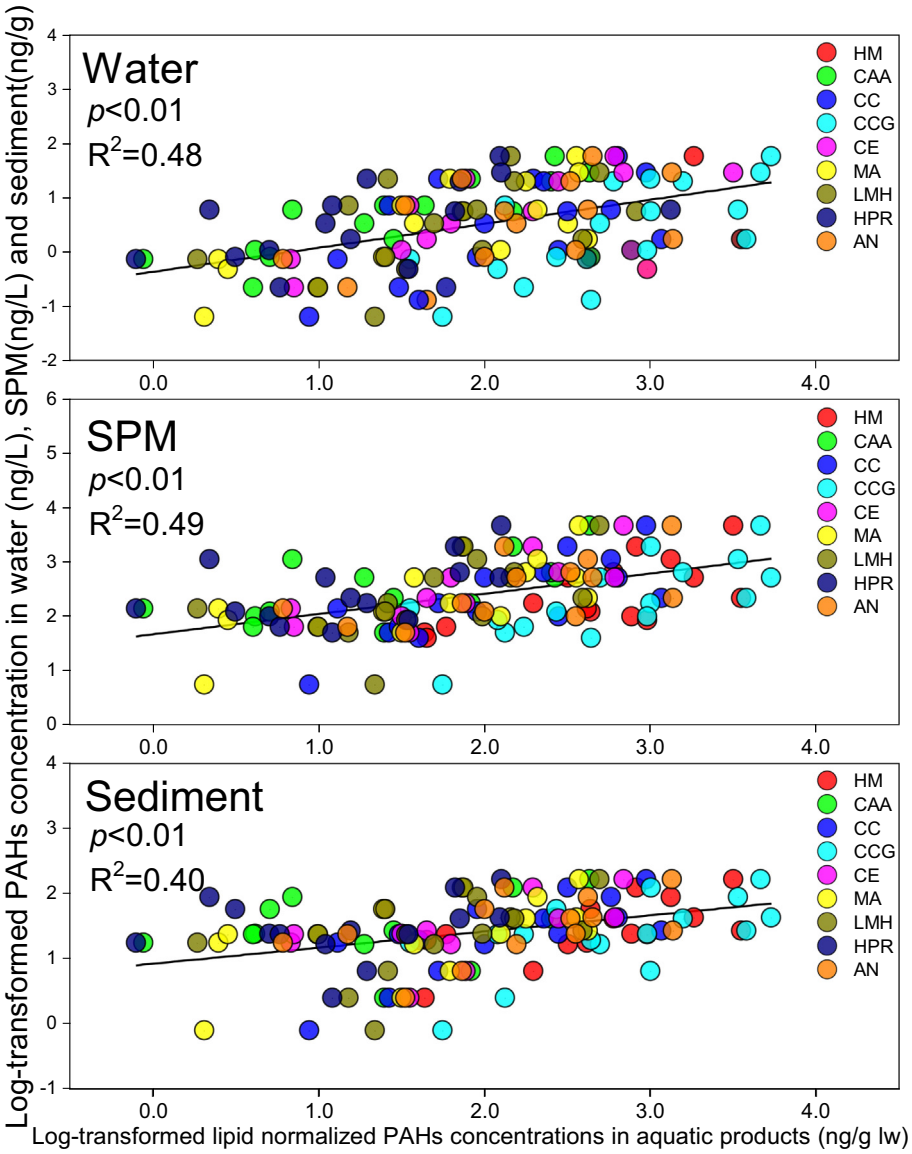

Fig. 2. Relationship between lipid-normalized concentration of polycyclic aromatic hydrocarbons (PAHs) in the tissues of aquatic organisms and PAH content in the environmental media.

species between logBAF and logKow Table 2. Significant correlations ($p < .01$) were found in spotted steed, carp, and white bait. It appears that the PAHs with high Kow can be easily adsorbed on the gill surface and can enter the fish when the PAHs in water flows through the gill.

In contrast, significant negative correlations were found between logBSSAF for four aquatic organisms and their logKow values. The results indicate the competition between SPM and organisms. The higher

Table 2
Correlation between BAF, BSSAF and BSAF and Kow.

	LogBAF-logKow		logBSSAF-logKow		logBSAF-logKow	
	p	R	*p*	R	*p*	R
HM	$p < .01$	0.90				
CC	$p < .01$	0.79				
CCG	$p < .05$	0.63	$p < .01$	−0.66		
CE	$p < .05$	0.64	$p < .01$	−0.94	$p < .01$	−0.73
MA			$p < .01$	−0.67		
LMH	$p < .01$	0.72				
HPR	$p < .05$	0.53	$p < .05$	−0.61		
AN	$p < .05$	0.68				

Table 3
Results of correlation analysis and regression between the PAHs concentrations and the trophic levels, and the FWMF values.

	R	N	Slope	Intercept	R^2	FWMF
Nap	−0.320	8	−0.33	3.72	0.11	0.08
Acy	−0.365	8	−0.64	3.79	0.15	0.01
Ace	−0.341	8	−0.35	3.5	0.13	0.06
Flo	−0.402	8	−0.54	4.15	0.18	0.01
Phe	−0.465	8	−0.49	4.27	0.23	0.02
Ant	−0.502	8	−0.5	3.8	0.26	0.02
Fla	−0.247	8	−0.29	3.49	0.07	0.10
Pyr	−0.768*	7	−0.34	3.52	0.6	0.07
BaA	−0.754*	8	−0.52	3.95	0.58	0.02
Chr	−0.799*	8	−0.49	4.13	0.64	0.02
BbF	−0.557	8	−0.43	3.56	0.32	0.03
BkF	−0.226	8	−0.14	2.96	0.06	0.33
BaP	−0.548	8	−0.57	3.56	0.31	0.01
IcdP	−0.466	3	−0.27	3.09	0.28	0.12
DahA	−0.490	4	−0.059	2.5	0.37	0.63
BghiP	0.152	5	0.08	2.63	0.02	1.87
ΣPAH	−0.526	8	−0.052	4.67	0.29	0.67

the Kow value is, the more difficult it is to bioaccumulate from SPM. Furthermore, no evident correlation was found between the BSAF and logKow values for most species. The result was consistent with those reported in previous studies (Moermond et al., 2007; Thorsen et al., 2004). In addition to the characteristics of the pollutants, the feeding method and metabolism may also have an influence on the bioaccumulation, which requires further research.

Food web magnification factors were calculated to investigate the concentrations of residues among different trophic levels in food webs (Table 3). The FWMF values for the 16 PAHs ranged from 0.008 to 0.749, which indicated that the concentrations of PAHs decreased as a function of the trophic level, which is referred to as trophic level biodilution. These results are consistent with those of previous studies on marine food webs (Wan et al., 2007). The results of the Pearson's correlation analysis showed that the relationships between the concentration and trophic levels were not significant for most PAHs, except for Pyr, BaA, and Chr. In contrast, significant negative correlations between trophic levels and log-transformed BAFs were observed for seven individual PAHs (Fig. 3) Phe, Ant, BbF, and BaP ($p < .05$) and Pyr, BaA, and Chr ($p < .01$). Among the regression models, Pyr ($R^2 = 0.78$), BaA ($R^2 = 0.71$), and Chr ($R^2 = 0.75$) had the best relationships, followed by Phe, Ant, BbF, and BaP, with R^2 of 0.46, 0.49, 0.45, and 0.53, respectively. The relationships for PAHs of lesser molecular mass, such as Nap, Ace, and Acy, were not obvious. A possible explanation could be the difference in metabolism and efficiencies of assimilation among PAHs. Bioaccumulation is the net result of competing rates of chemical uptake and elimination from an organism (Arnot et al., 2009). Metabolism and assimilation are two important factors that determine the efficiencies of the trophic transfer of PAHs in ecosystems (Burkhard and Lukasewycz, 2000; Wan et al., 2007). Greater efficiencies of assimilation for PAHs of lesser molecular mass suggest that they are degraded in the biota.

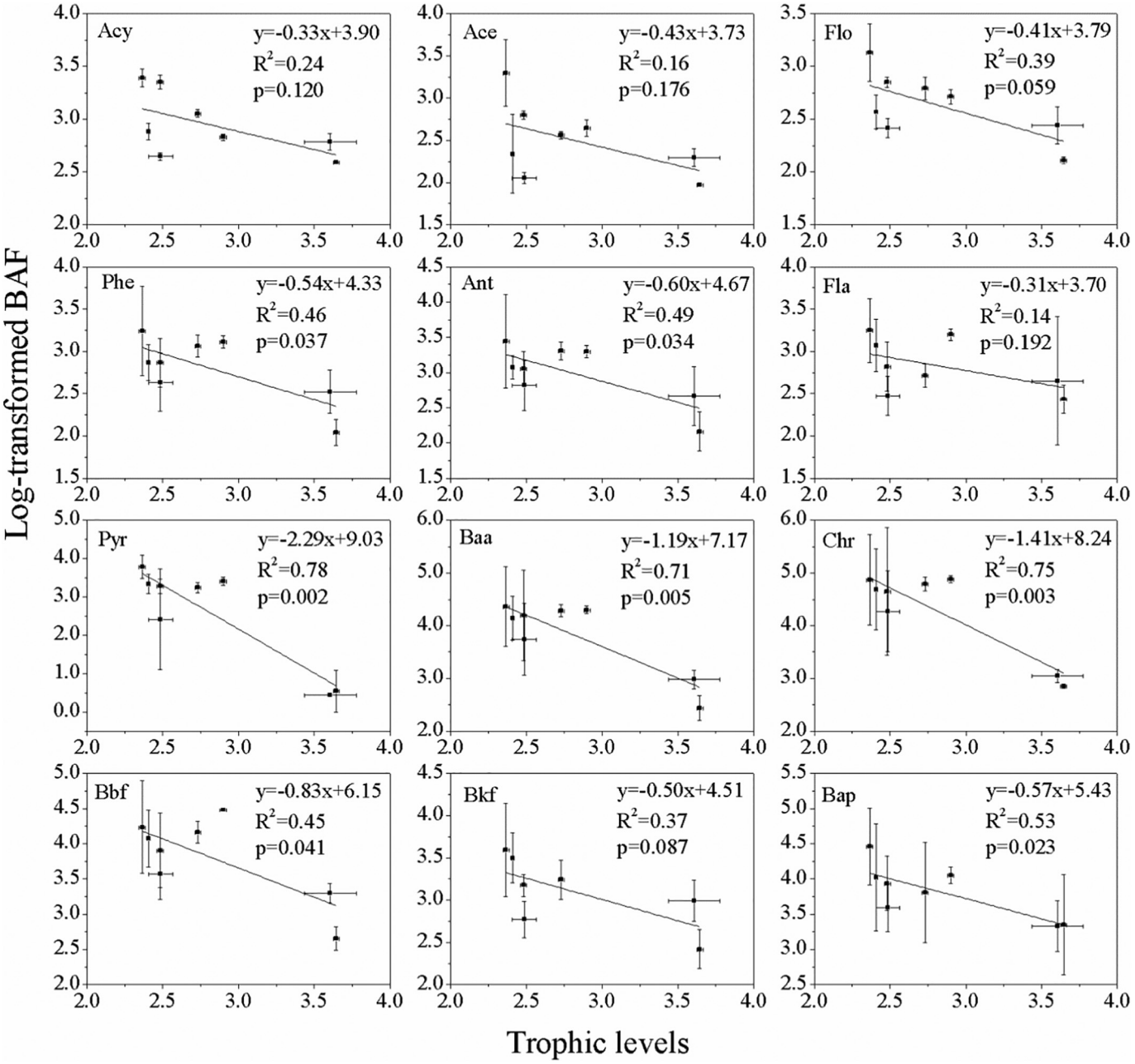

Fig. 3. Relationships between bioaccumulation factors for polycyclic aromatic hydrocarbons and trophic levels for the aquatic organism food web in Lake Chaohu.

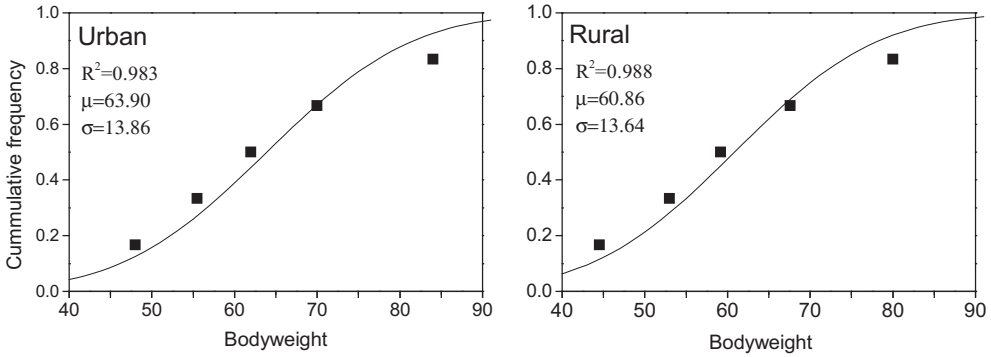

Fig. 4. Fitting of bodyweight of urban and rural population in Anhui Province. μ and σ are mean and standard variation of normal distribution, respectively.

3.5. Assessment of the health risk

Consumers of aquatic products from Chaohu Lake include not only the population from cities, such as Hefei, the capital of Anhui Province, but also the population from the surrounding villages. Significant differences in the exposure behavior parameters can be observed between rural and urban population subgroups because of the income levels and consumption habits. According to a survey conducted between 2010 and 2013, the averaged daily intake of freshwater products per capita were 19.0 g/day and 11.1 g/day for the urban and rural populations, respectively (Zhao and He, 2018). Despite the exposure behavior, differences exist in physiological parameters between urban and rural people. Here, we considered a normal distribution for BW and log-normal distributions for the concentration of BaP_{eq} (Shapiro-Wilk test, $p > .05$) because the normal and ilog-normal distribution models are the most widely applied in studies on the exposure parameters (Chen and Liao, 2006; Chiang et al., 2009; Liao et al., 2011). We obtained quartiles and medians using the Exposure Factors Handbook of Chinese Population. The weight distribution of the population was fitted using the Gaussian function. The fit curves and parameters are shown in Fig. 4.

The ILCR of the studied population attributed to BaP_{eq} of the ingested aquatic products was estimated using a 10,000-times Monte Carlo simulation. Parameters for the Monte Carlo simulation are shown in Table S3.

The ILCR distribution of population groups was derived using the Monte Carlo simulation (Fig. 5). The mean values of the ILCR for urban and rural residents were 7.68×10^{-6} and 4.75×10^{-6}, respectively, which showed a noticeable higher carcinogenic risk for urban residents than that for rural residents.

It has been reported that for most non-occupationally exposed individuals, diet is the main route of exposure (Falco et al., 2005; Martorell et al., 2012). The risk values determined in Chaohu are higher than those reported in Korea (2.85×10^{-6}). The results were lower than those reported in another research on food, including aquatic products, in Shanxi (3.87×10^{-5} to 4.04×10^{-5}). According to the criteria suggested by the US EPA, a one-in-a-million chance of an additional human cancer over a 70-year lifetime is an acceptable level of risk and a one-in-ten thousand or greater chance is considered to be a serious risk. Based on this, almost the entire population within the research area exceeded the acceptable limits. These results indicate that the intake of PAHs through the ingestion of aquatic products is related to a certain carcinogenic risk. Moreover, our research is based on raw uncooked aquatic products. Comparative research between raw and cooked food has confirmed that processed food contains greater amounts of PAHs than those reported in raw food, especially in the case of meat (Alomirah et al., 2011; Zhang et al., 2014). The process of cooking may considerably increase the exposure to PAHs and enhance the risk of cancer in consumers.

4. Conclusions

In this study, the residual levels of PAHs in eight major freshwater organisms and environmental media, including water, SPM, and sediment from Chaohu Lake were measured. Tissue distribution and

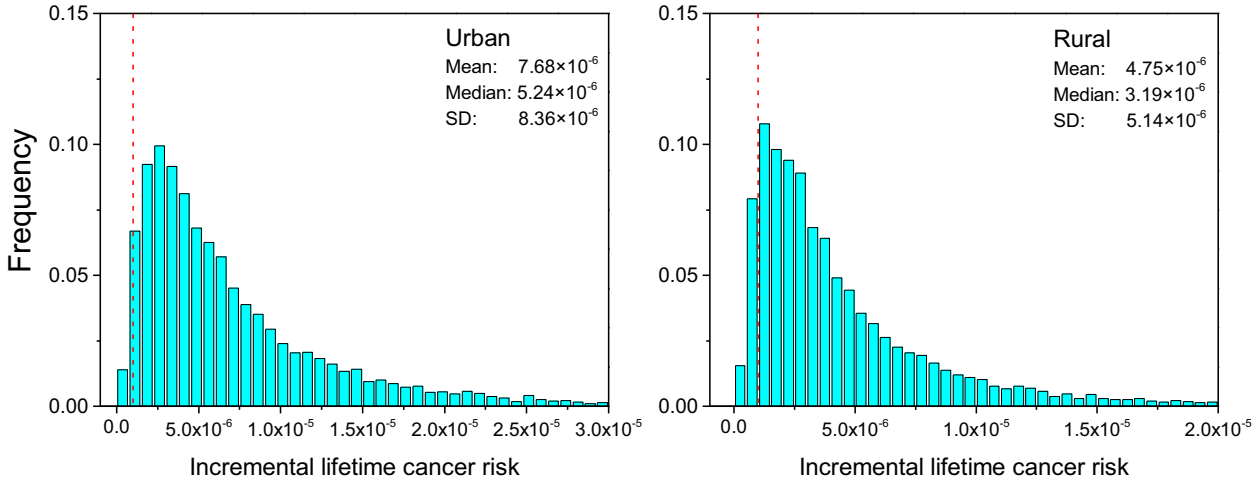

Fig. 5. Distributions of incremental lifetime cancer risk for urban and rural populations derived using the Monte Carlo simulation. Red dotted lines represent the US EPA acceptable levels. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

bioaccumulation characteristics were studied. Different tissue distribution modes were observed among the different fish species. We observed correlations ($p < .01$) between the three environmental media and the content of PAHs in aquatic animals. Based on the FWMF values and risk assessment, it was found that although diluted because of the food web, the intake of PAHs through the ingestion of aquatic products, at levels higher than the US EPA acceptable levels, poses a potential risk of cancer. Further control strategies need to be formulated to reduce the cancer risk.

CRediT authorship contribution statement

Ning Qin: Conceptualization, Methodology, Experiment, Formal analysis, Data curation, Writing - original draft, Visualization. **Wei He**: Investigation, Experiment. **Wenxiu Liu**: Investigation, Experiment. **Xiangzhen Kong**: Investigation. **Fuliu Xu**: Supervision, Project administration, Funding acquisition. **John P. Giesy**: Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.141577.

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Supporting Information for

Tissue distribution, bioaccumulation, and carcinogenic risk of polycyclic aromatic hydrocarbons in aquatic organisms from Lake Chaohu, China

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Supplementary caption

Table S1 Recoveries, method detection limits (MDLs) and toxicity equivalency factors (TEFs) in aquatic organisms, water, SPM samples from Lake Chaohu

Table S2 BAF, BSSAF and BSAF values of priory PAHs in aquatic organisms from Lake Chaohu

Table S3 Slope and p value of regression analysis between logarithm lipid-normalized concentrations and trophic levels, and FWMF values

Table S4. Exposure parameters for Monte Carlo simulation.

Fig. S1 Sampling zones and the sampling sites in Lake Chaohu; fourteen sediment sites include west lake area (S1), east lake area (S2-S4), water source areas (S5-S10) and river sites (S11-S14); three water & SPM sites were located on Mushan Island (W1), in Zhongmiao Town (W2) and Yuxi Environmental Monitoring Station (W3).

Fig. S2 Aquatic organisms in Lake Chaohu

Fig. S3 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in water.

Fig. S4 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in SPM

Fig. S5 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in sediment.

Table S1 Recoveries, method detection limits (MDLs) and toxicity equivalency factors (TEFs) in aquatic organisms, water, SPM samples from Lake Chaohu

	Aquatic Organisms		SPM/sediment		Water		TEF[1]
	Recoveries (%)	MDL ng/g wm	Recoveries (%)	MDL ng/g dm	Recoveries (%)	MDL ng/l	
Nap	114.9	0.65	45.5	0.28	80.9	0.62	0.001
Acy	101.0	0.07	79.7	0.11	87.4	0.57	0.001
Ace	117.4	0.16	73.8	0.09	80.6	1.24	0.001
Flo	104.9	0.91	76.7	0.10	103.4	0.48	0.001
Phe	106.5	1.08	95.1	0.10	107.8	0.46	0.001
Ant	100.9	0.40	85.1	0.11	93.1	0.54	0.01
Fla	112.8	0.07	95.3	0.11	88.7	0.56	0.001
Pyr	122.1	1.45	96.0	0.15	88.8	0.56	0.001
BaA	102.1	0.08	93.9	0.14	62.7	1.59	0.1
Chr	118.6	0.30	95.0	0.17	62.2	1.61	0.01
BbF	105.4	0.06	95.3	0.14	42.7	2.34	0.1
BkF	102.2	0.10	95.1	0.17	44.3	2.26	0.1
BaP	87.2	0.65	88.8	0.16	60.3	1.66	1
IcdP	88.5	0.11	89.0	0.21	31.2	3.20	0.1
DahA	93.2	0.11	85.5	0.22	23.6	4.24	1
BghiP	109.9	0.11	91.2	0.17	23.7	4.22	0.01

Table S2 BAF, BSSAF and BSAF values of priory PAHs in aquatic organisms from Lake Chaohu

	BAF			BSSAF			BSAF		
	Mean	GM	SD	Mean	GM	SD	Mean	GM	SD
Nap	18.81	9.04	29.16	0.18	0.08	0.27	25.45	12.23	39.46
Acy	5.53	4.33	5.10	0.07	0.05	0.06	15.88	12.44	14.64
Ace	7.90	3.62	14.03	0.08	0.04	0.15	28.03	12.84	49.77
Flo	20.41	13.88	23.15	0.05	0.04	0.06	9.95	6.77	11.29
Phe	46.37	27.23	52.23	0.02	0.01	0.03	8.24	4.84	9.28
Ant	41.23	21.70	48.80	0.02	0.01	0.03	8.53	4.49	10.10
Fla	56.33	34.43	62.84	0.01	0.01	0.02	2.57	1.57	2.87
Pyr	125.98	25.78	192.12	0.07	0.04	0.09	8.62	1.76	13.14
BaA	269.17	86.98	321.95	0.25	0.08	0.29	12.14	3.92	14.52
Chr	696.87	201.02	864.33	0.45	0.13	0.56	44.66	12.88	55.40
BbF	135.38	49.65	184.75	0.07	0.03	0.10	1.89	0.69	2.58
BkF	73.64	9.13	186.74	0.03	0.00	0.08	3.05	0.38	7.73
BaP	159.29	73.88	249.16	0.04	0.02	0.06	1.50	0.70	2.34
IcdP	1359.36	719.92	1781.34	0.33	0.18	0.43	9.14	4.84	11.98
DahA	-	-	-	-	-	-	28.06	15.42	30.32
BghiP	476.21	108.03	847.84	0.26	0.10	0.41	9.78	2.22	17.42

Table S3 Slope and p value of regression analysis between logarithm lipid-normalized concentrations and trophic levels, and FWMF values

	b	R^2	p value	FWMF
Nap	-0.563	0.071	-0.267	0.570
Acy	-0.289	0.067	-0.258	0.749
Ace	-0.611	0.085	-0.292	0.543
Flo	-0.503	0.114	-0.338	0.605
Phe	-0.813	0.180	-0.424	0.444
Ant	-0.950	0.219	-0.467	0.387
Fla	-0.293	0.024	-0.157	0.746
Pyr	-4.799	0.725	-0.851	0.008
BaA	-2.288	0.583	-0.764	0.101
Chr	-2.792	0.630	-0.794	0.061
BbF	-1.458	0.294	-0.541	0.233
BkF	-0.564	0.028	-0.165	0.569
BaP	-0.999	0.227	-0.475	0.368

Table S4. Exposure parameters for Monte Carlo simulation.

Parameters	Urban	Rural	References
BaP_{eq} (ng/g)	LN(1.14, 0.86)	LN(1.14, 0.86)	Measured
BW (kg)	N(63.9, 13.9)	N(60.9, 13.6)	[2]
IR (g/day)	19.0	11.1	[3]

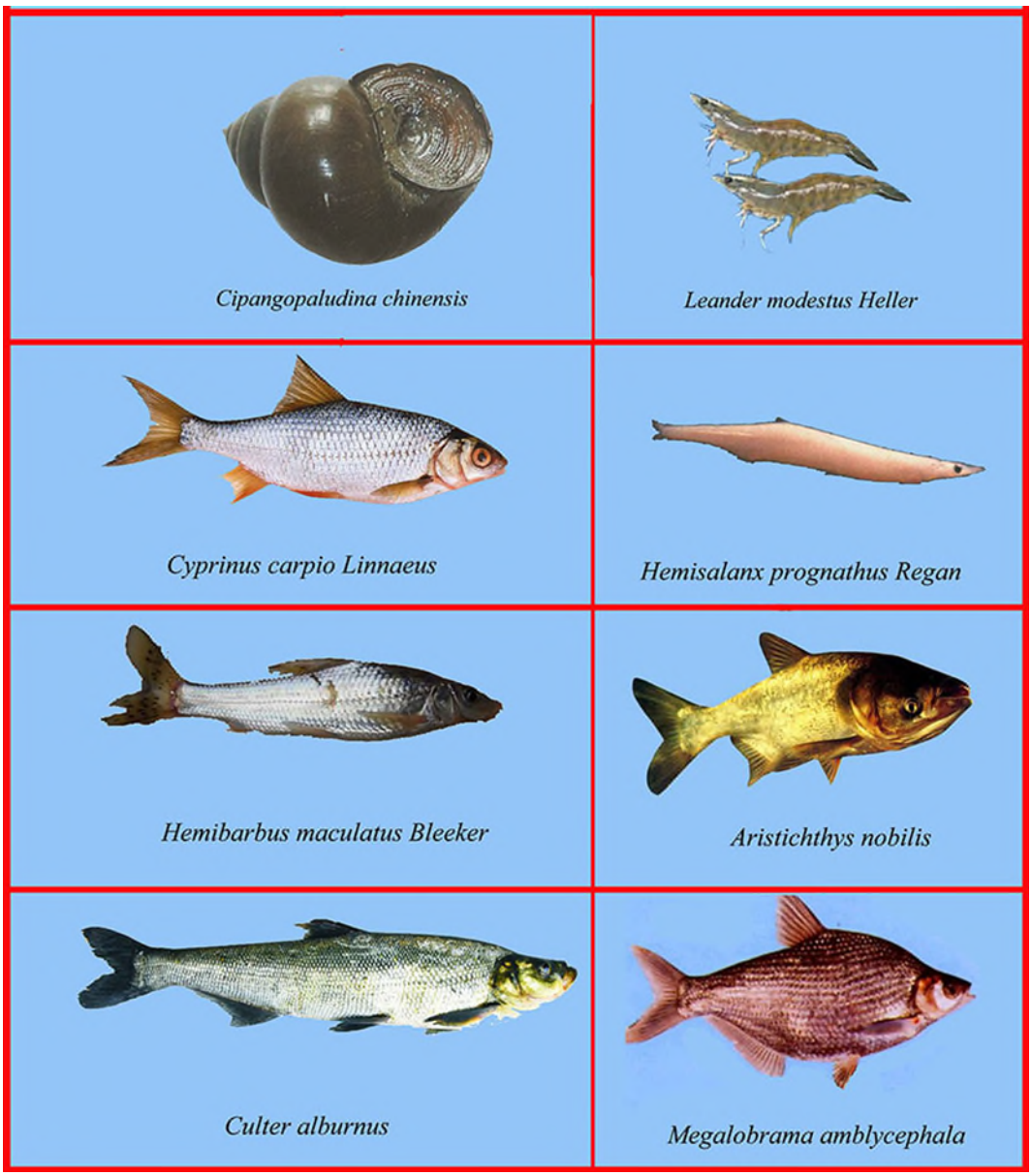

Fig. S1 Aquatic organisms in Lake Chaohu

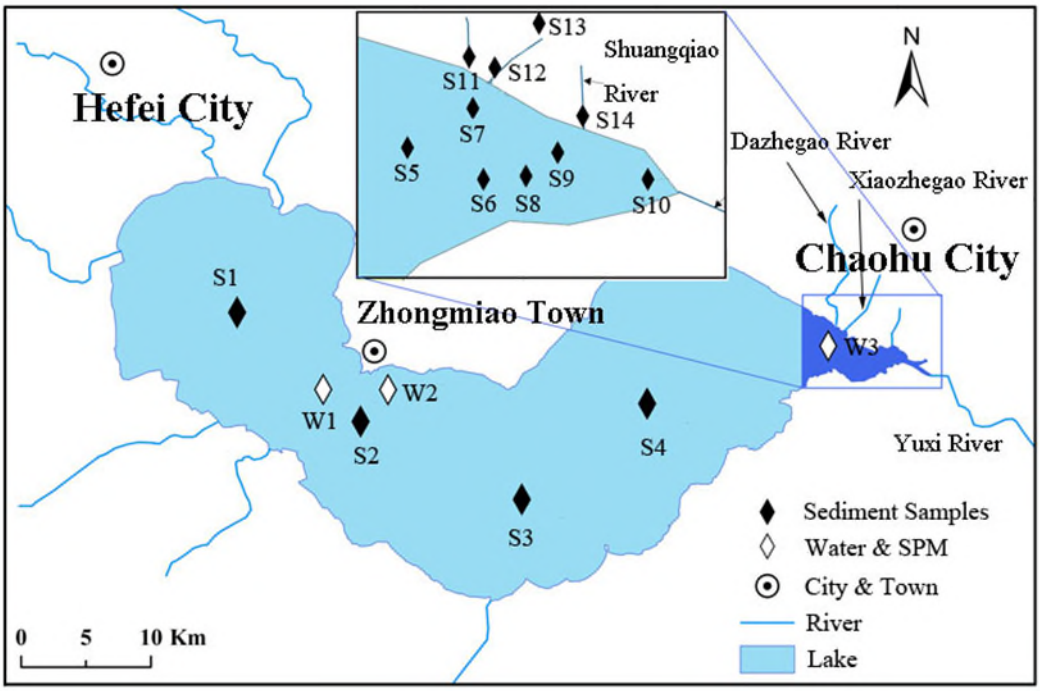

Fig. S2 Sampling zones and the sampling sites in Lake Chaohu; fourteen sediment sites include west lake area (S1), east lake area (S2-S4), water source areas (S5-S10) and river sites (S11-S14); three water & SPM sites were located on Mushan Island (W1), in Zhongmiao Town (W2) and Yuxi Environmental Monitoring Station (W3).

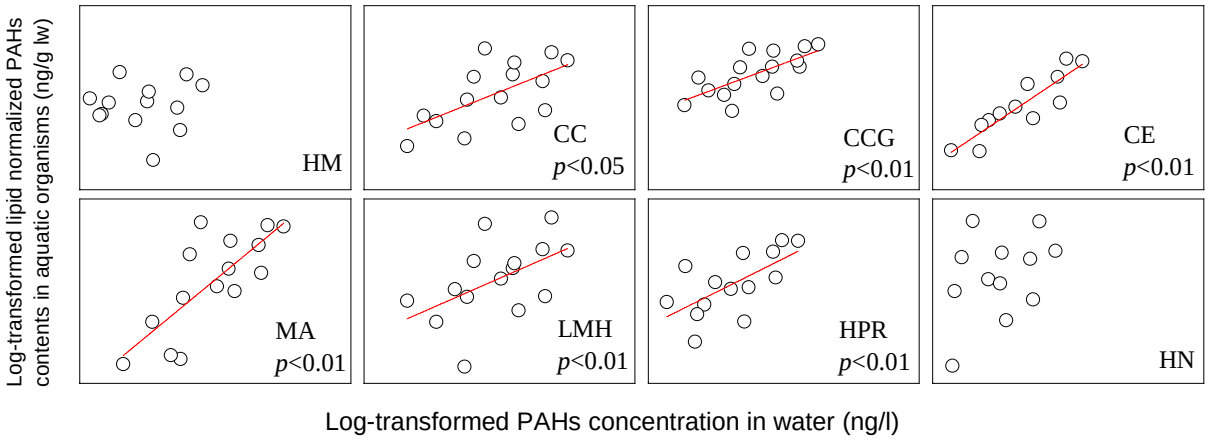

Fig.S3 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in water.

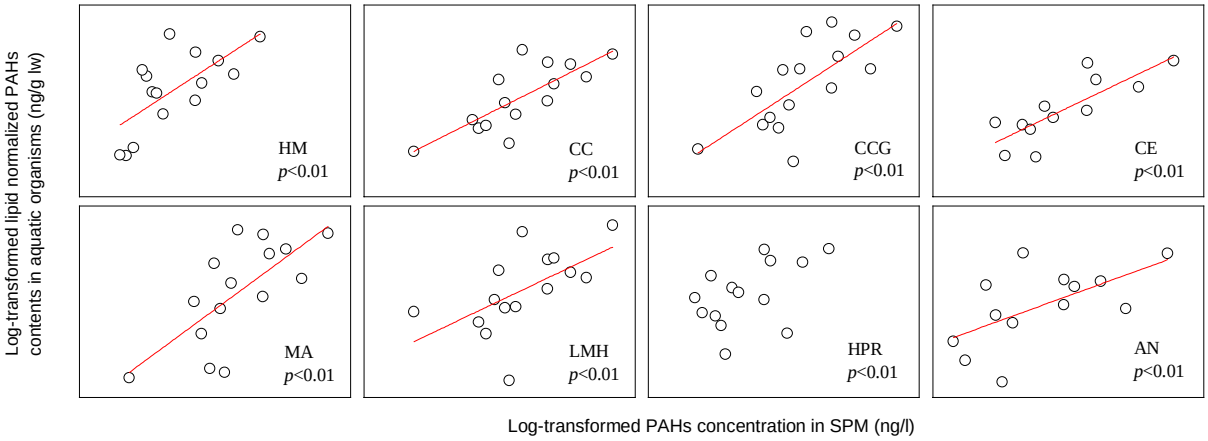

Fig.S4 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in SPM

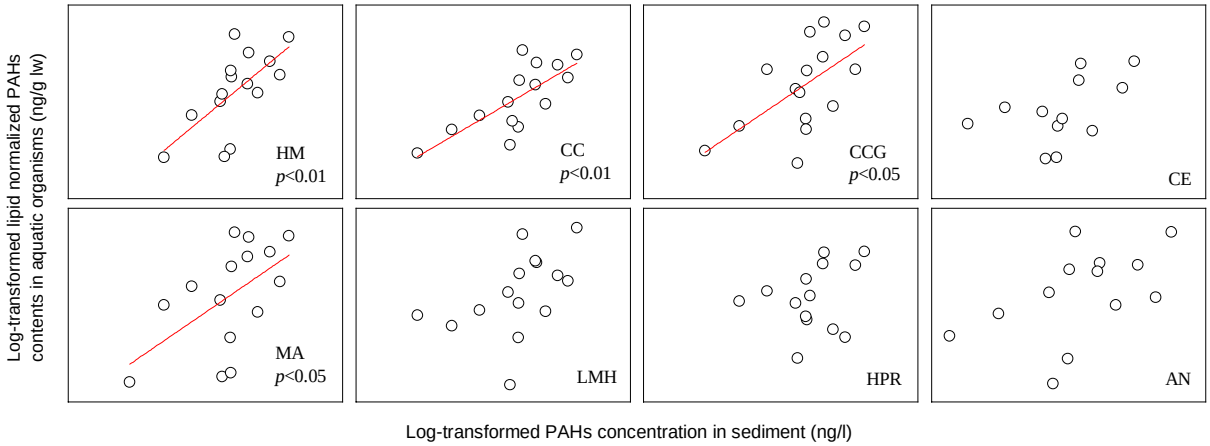

Fig.S5 Relationship of lipid-normalized PAH concentration in aquatic organism organs and PAHs content in sediment.

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