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Distribution of polycyclic aromatic hydrocarbon (PAH) residues in several tissues of edible fishes from the largest freshwater lake in China, Poyang Lake, and associated human health risk assessment

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ABSTRACT

The residual levels, tissue distribution and human health risk assessment of polycyclic aromatic hydrocarbons (PAHs) in edible fishes, bighead carp (*Aristichthys nobilis*) and silver carp (*Hypophthalmichthys molitrix*), from the largest freshwater lake in China, Poyang Lake, were studied. PAH concentrations ranged from 105 to 513 ng g^{-1} ww and from 53.9 to 401 ng g^{-1} ww in different tissues of bighead carp and silver carp, respectively. Low molecular weight (LMW) PAHs were the predominant compounds, suggesting the gill-water transfer might be the major exposure route for PAHs in the studied fish species. Tissue distribution indicated that the hepatobiliary system accumulated higher concentrations of PAHs than the extrahepatic tissues with bile being the most predominant tissue for both species. Composition analysis demonstrated that PAHs were from the combined petrogenic and pyrogenic origin, and the gasoline combustion might be the main source. A preliminary evaluation of human health risk using benzo[*a*]pyrene (BaP) potency equivalent concentration (PEC) as well as the incremental lifetime cancer risk (ILCR) indicated that PAHs in fish would induce potential carcinogenic effects.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group complex mixture with more than 10,000 individual compounds formed by two or more fused aromatic rings of carbon and hydrogen atoms (Logan, 2007) originating from natural and anthropogenic sources, such as incomplete combustions, industrial incinerations, transport or uncontrolled spills (Nácher-Mestre et al., 2010). PAH input has increased extensively in the 20th century (Vives et al., 2004), and thus the U.S. Environmental Protection Agency (USEPA) has established 16 PAHs as the priority control pollutants, 7 of which are potentially carcinogenic to humans according to the International Agency for Research on Cancer (Kannan et al., 2005; Qin et al., 2013). PAHs entering the aquatic ecosystem can be first accumulated in fine grained sediments and suspended particles due to their hydrophobic nature, later remobilized in the water column, then become bioavailable to native organisms (Wetzel and Van Vleet, 2004), and finally accumulated in biota of higher trophic levels (Arias et al., 2009). In addition, PAHs may pose toxicity to fish and birds, by interfering with cellular membrane function and the associated enzyme systems (Malik et al., 2008). It was suggested PAHs in many aquatic environments are an important risk factor for various health aspects of fish (Payne et al., 2003), such as adverse histopathologic and immunological responses in tilapia (Liang et al., 2007). Furthermore, PAHs can be metabolized in aquatic fauna to active and potent carcinogenic forms. In fact, metabolites of PAHs found in benthic fish are strongly associated with hepatic lesions and liver neoplasm (Varanasi et al., 1989; Malik et al., 2008; Johnson-Restrepo et al., 2008).

The dietary intake of contaminated food, and especially aquatic products, is considered to be one of the major sources of the total human exposure to toxic chemicals (Smith and Gangolli, 2002). Dougherty et al. (2000) revealed that freshwater fish consumption accounted for 75 percent of exposure such as dichlorodiphenyltrichloroethane (DDT) (whereas other food items contribute not more than 5 percent) across the United States. For Asian countries, it has been noted that mean daily intake of persistent organic pollutants (POPs, such as dioxin-like compounds) was the highest in Japanese people from eating fish and shellfish (Tsutsumi et al., 2001), although fish products accounted only for about 10 percent of diet or less (Li et al., 2008). Furthermore, global freshwater fish production has grown impressively, with the average per capita supply increasing from 1.5 kg in 1961 to 6.1 kg in 2009 (Food and Agriculture Organization of the United Nations (FAOSTAT), 2013).

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As for China, it has the largest fishery production in the world with an output of 44.3 million metric tons in 2002, accounting for 33.3 percent of the total global production (Meng et al., 1821–1827), while the supply quantity of freshwater fish also increased impressively from 1.1 kg capita^{-1} yr^{-1} in 1961 compared to 13.3 kg capita^{-1} yr^{-1} in 2009 (Food and Agriculture Organization of the United Nations (FAOSTAT), 2013). In addition, it is also expected that PAH emissions have been increasing greatly due to the increasing energy demand associated with the rapid population growth and economic development, and to the low efficiency of energy utilization (Xu et al., 2006; Zhang et al., 2007). It was estimated that the total PAH emission in China was 25,300 t in 2003 (Xu et al., 2006). Despite the increasingly important impact of China's fishery products on the global population and widespread PAHs, the state of fish contamination by PAHs and associated human health risk through dietary exposure have not been assessed adequately so far.

Most previous studies on the occurrence of PAHs in different fish species were reported during last decades (Pointet and Milliet, 2000; White and Triplett, 2002; Barron et al., 2004; Liang et al., 2007; Perugini et al., 2007), which mainly focused on PAH residual levels in fish muscles. Although some studies also pointed out the tissue-distribution of specific PAHs (such as phenanathrene (Phe), pyrene (Pyr) and benzo[*a*]pyrene (BaP)) and their metabolites through the laboratory experiments (Lazartigues et al., 2010, 2011; Carrasco Navarro et al., 2012), the patterns and distributions of PAHs in different fish tissues (such as fish brain, kidney, liver and gill, etc.), which could provide more clues about the bioaccumulation and metabolism of PAHs in fishes, are still not well documented (Deb et al., 2000). The residues of PAHs in edible fishes could also provide more information on the risk levels of PAHs to human health though fish consumption (Xu et al., 2011).

In China, some investigations have been carried out on the pollution status of PAHs in freshwater and marine fishes, such as Lake Small Bai-Yang-Dian (Xu et al., 2011), Hong Kong (Cheung et al., 2007), Pearl River Delta (Kong et al., 2005), Xiamen coastal waters (Klumpp et al., 2002a, 2002b) and Bohai Bay (Wan et al., 2007). Poyang Lake, the largest freshwater lake of China, is located in the north of Jiangxi province and lies on the southern bank of the Yangtze River. Most previous studies around this basin mainly focused on the heavy metal pollution since Jiangxi province is famous for its nonferrous metal resources (Luo et al., 2008; Yuan et al., 2011), little information is available on POP pollution in such an important aquatic ecosystem of China. However, during the past decades, with the rapid economic development and population growth in the watershed and neighbor regions, Poyang Lake has suffered from the considerable increasing load of PAHs. Therefore, it is urgent to elucidate the pollution status and potential human health risk of PAHs in edible fishes from this lake considering its importance on providing abundant fishery products for both Jiangxi province and the whole region of mid-lower reaches of the Yangtze River.

In this study, the residual levels of PAHs in edible fishes collected from Poyang Lake were investigated to illustrate the tissue-specific bioaccumulation of PAHs. Compositions of individual compounds as well as the particular characteristic indices were also analyzed to elucidate the pollution sources of PAHs around Poyang Lake. In addition, human health risk assessment of PAHs by dietary intake was also performed to evaluate their potential carcinogenic effects on human beings through fish consumption from Poyang Lake.

2. Materials and methods

2.1. Area description and sampling

Poyang Lake (28°04'–29°46'N, 115°49'–116°46'E), the largest freshwater lake in China, plays an important role in maintaining the unique biota of the Yangtze River floodplain ecosystem and supports diverse species of fish (136 species, 25 families) (Wang et al., 2011), with taxa representing a wide range of ecological attributes and life history strategies. Two popular fish species, bighead carp (*Aristichthys nobilis*) and silver carp (*Hypophthalmichthys molitrix*), which are both filter feeders and

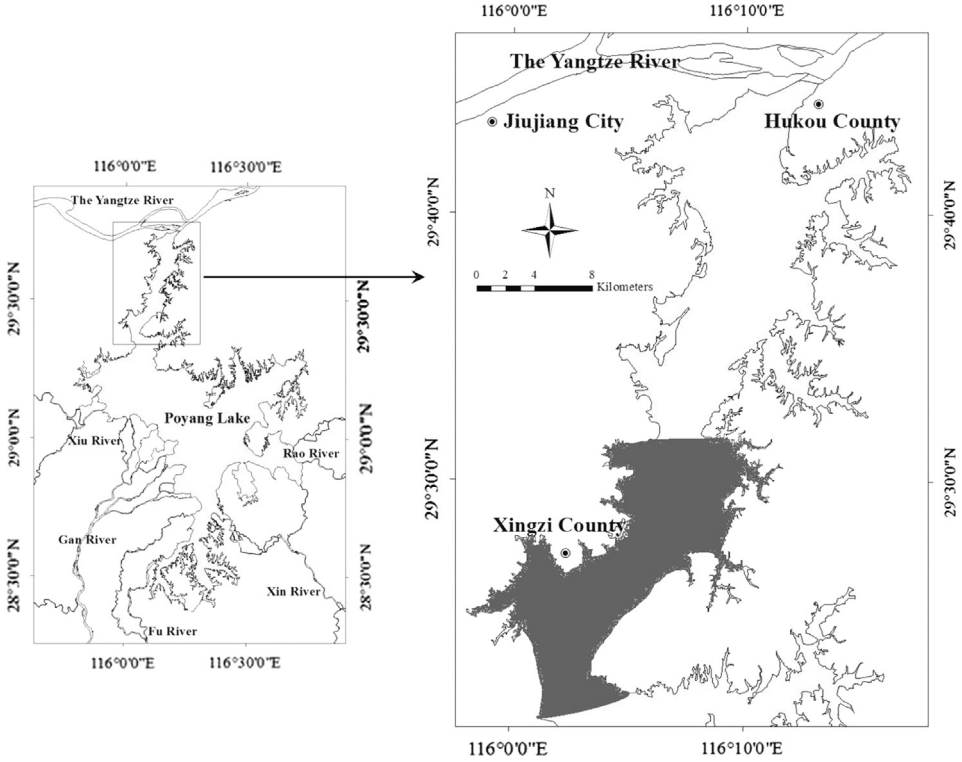

Fig. 1. Study area and sampling locations (the black area) of fish samples from Poyang Lake.

belong to the cyprinid fish were selected as the studied samples due to their importance for human consumption (Cremer and Smitherman, 1980). Fish samples were collected from the northern water region located near Xingzi County (Fig. 1) from August 15th to 18th, 2011 with the help of local fishermen. Seven individuals were picked out from each fish species with similar fresh weight (36.0 ± 1.6 cm of bighead carp and 36.0 ± 0.7 cm of silver carp) and similar body length (958.4 ± 101.5 g for bighead carp and 796.3 ± 40.1 g for silver carp) for the following analysis. The fish age of bighead carp and silver carp calculated according to the length-growth model ranged from 1.62 years to 1.78 years and from 1.62 years to 1.71 years, respectively, and ranged from 1.96 years to 2.13 years and from 1.41 years to 1.50 years based on the weight-growth model, respectively, which both demonstrated the selected specimens were the young fish. All fish samples were stored in incubator with constant temperature of 4 °C, and then were immediately transported to the laboratory for the following treatment.

2.2. Chemicals and reagents

Sixteen priority PAHs recommended by EPA 610 method was determined, which was purchased from Supelco (USA) containing naphthalene (NaP), acenaphthylene (Any), acenaphthene (Ana), fluorene (Flu), Phe, anthracene (Ant), fluoranthene (Flt), Pyr, benz[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]flouranthene (BkF), BaP, dibenz[*a*,*h*]anthracene (DahA), benzo[ghi]perylene (BghiP) and indeno[1,2,3-cd]pyrene (IP). The stock standard solutions were dissolved in methanol/dichloromethane (1:1, *v*/*v*), and then the standard solutions were kept at 4 °C before use. The working solutions were prepared with suitable dilutions daily before use. All solvents used for dilution and following sample extraction were pesticide grade and stored in glass containers in dark before use. Deionized water used for HPLC analysis was taken from a Milli-Q water system (Millipore Milli-Q system, USA).

2.3. Sample pretreatment

The fish were washed by the deionized water and then were freshly dissected carefully to obtain muscle (taken from the dorsal surface of the fish), gill, liver, heart, kidney, bile, swim bladder, squama, skin (without muscle layer and squama), respectively. Each tissue sample was washed, dried in filter paper and weighed, packed in aluminum foil and then kept at −20 °C until analysis. The samples were freeze-dried and the same tissue from individuals of each fish species were mixed together, and then were ground in an agate mortar to obtain fine powders (about 100 mesh). The samples were extracted using accelerated solvent extraction (ASE) system and purified by gel permeation chromatography (GPC) and silica gel–alumina (2:1) column in due order. The detailed operations and conditions of sample extraction and cleanup as well as the lipid determination were shown as the supplemental information, which could also be found in Zhao et al. (2009, 2013).

2.4. Instrument analysis

Determination of PAHs was completed using high performance liquid chromatography (HPLC) (Agilent 1200 HPLC, USA) equipped with a diode-array detector (DAD) coupled with a series-wound fluorescene detector (FLD). The separation column was a Supelcosil LC-PAHs (ODS, 3.0 mm i.d. × 250 mm length, particle size: 5 μm, Supelco, USA). The wavelength of DAD was 254 nm, while the excitation and emission wavelengths were changed according to a time program for FLD (Ex/Em: 260/380 nm at 0 min, 280/330 nm at 2 min, 260/380 nm at 9.2 min, 280/450 nm at 14.0 min, 260/380 nm at 16.5 min, 290/410 nm at 28 min, 290/500 nm at 35.5 min). The mobile phase for gradient elution was a mixture of purified water and acetonitrile with the gradient program of 60 percent acetonitrile at start, 15 min hold, 15 min linear gradient to 100 percent and 10 min hold. The stop time of pump was set at 48 min for each sample. The injection volume was 20 μL and the flow rate was 0.75 mL min^{-1}. During the whole process, column temperature was maintained at 30 °C.

2.5. Quality assurance and quality control

Peak identification of PAHs was based on both the UV spectra and retention time of standard components analyzed under the same instrument conditions, while the quantification was completed by the external standard method with 6 different contents being applied to build the standard curves between concentration and peak area for each compound (r=0.995–0.999). PAH concentrations were expressed on the wet weight basis (ng g^{-1} wet weight, ng g^{-1} ww). During the whole test, method detection limits (MDLs) of individual PAHs were defined as the concentration of target analytes giving a peak with a signal-to-noise (S/N) of 3, which ranged from 0.07 to 3.52 ng g^{-1} ww for individual compounds. Results of laboratory blanks indicated no interferent contaminants were detected during the whole tests. The quartz (baked at 550 °C for 6 h) were used as the substitute matrix, and then were spiked with the known concentration of standards (up to 5 ng g^{-1} dw) containing 16 priority PAH compounds to study on the recoveries of the whole method. The spiked recoveries ranged from 90 to 116 percent for individual PAHs. Obtained results were not corrected for recovery efficiency at present. Relative standard deviations (RSDs) of the whole method conducted with 5 different standard solutions were less than 12 percent. In addition, all readings were recorded in duplicate and the variations of chemical concentrations of replicated samples were all less than 10 percent. Additionally, standard solution was added every fifteen samples to recalibrate the retention time of target compounds.

2.6. Statistical analysis

Statistical treatment involved in the correlation analysis of compositions and concentrations between fish tissues or chemicals were performed with SPSS software (SPSS 16.0 for Windows, SPSS Inc. USA). Spearman's rank correlation was used to test the strength of associations between parameters, and the statistical significance was considered for $p < 0.05$ (two tailed tests).

3. Results and discussion

3.1. PAH residues in fish

The concentrations of total PAHs and individual congeners observed in different tissues of bighead carp and silver carp are listed in Table 1. PAHs were detected in all tissue samples with the

Table 1
PAH concentrations (ng g^{-1} ww) in tissues of bighead carp and silver carp from Poyang Lake.

Compounds	Bighead carp									Silver carp								
	Bile	Scale	Heart	Muscle	Gill	Skin	Bladder	Liver	Kidney	Bile	Scale	Heart	Muscle	Gill	Skin	Bladder	Liver	Kidney
Nap	81.0	1.44	1.43	17.4	8.65	13.5	12.4	5.79	31.4	8.42	5.90	4.19	0.24	16.5	10.4	25.2	4.95	6.11
Any	22.9	15.7	68.0	20.6	18.4	29.5	25.1	58.4	75.6	34.0	20.1	47.0	6.74	16.8	6.61	8.61	33.1	28.5
Ana	nd	nd	nd	nd	nd	0.06	nd	0.08	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Flu	36.9	11.0	18.9	9.34	5.65	8.42	10.5	9.03	11.4	36.8	17.6	34.6	5.03	9.21	7.31	13.9	7.97	4.14
Phe	95.7	28.8	81.2	50.7	34.9	48.0	59.2	55.0	65.3	42.1	16.4	20.6	6.60	9.78	4.17	11.9	8.73	5.84
Ant	44.0	13.5	25.4	10.1	6.85	8.89	11.9	9.87	12.7	69.4	28.2	44.1	8.49	11.3	9.32	16.6	11.0	5.79
Flt	nd	nd	nd	nd	nd	nd	nd	nd	0.58	nd	nd	nd	nd	nd	nd	nd	nd	nd
Pyr	66.4	21.7	39.0	17.0	10.6	16.1	19.1	14.6	21.2	113.2	12.4	56.3	12.7	15.7	11.8	22.7	14.9	8.60
BaA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Chr	28.7	9.42	20.6	3.46	2.04	2.99	3.69	1.33	7.30	25.8	13.5	27.2	6.00	6.90	6.46	13.1	8.03	3.20
BbF	89.4	24.2	33.4	18.8	11.7	1.98	20.5	10.9	27.1	44.7	20.0	26.2	6.31	45.2	32.0	64.5	37.7	1.79
BkF	32.2	9.42	16.5	6.81	4.30	1.40	7.65	1.66	9.22	5.48	1.06	2.58	0.52	0.69	0.58	0.86	0.56	0.22
BaP	6.85	1.91	3.17	1.43	0.79	0.53	1.63	0.23	1.49	2.99	0.97	1.15	0.33	0.36	0.31	2.53	0.46	0.31
DahA	4.84	1.15	2.91	0.75	0.75	0.57	0.80	0.52	1.50	3.98	0.95	3.68	0.70	1.29	0.52	0.77	0.61	0.07
BghiA	3.78	0.90	1.37	0.07	0.08	0.08	0.11	0.13	0.12	14.1	0.22	1.14	0.23	0.30	0.14	0.36	0.19	0.21
IP	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
∑PAHs	513	139	312	157	105	132	173	168	265	401	137	269	53.9	134	89.6	181	128	64.8

nd: The concentrations of chemicals less than the MDLs were defined as not detected (nd).

concentrations ranging from 105 to 513 ng g^{-1} ww for bighead carp and from 53.9 to 401 ng g^{-1} ww for silver carp, respectively. Significant positive correlations of PAH concentrations were observed between tissues of both bighead carp ($r=0.615$–0.994, $p<0.05$, $n=16$) and silver carp ($r=0.503$–0.992, $p<0.05$, $n=16$) except for the weaker positive correlation between kidney with other tissues of silver carp ($r=0.137$–0.669, $p=0.005$–0.613, $n=16$), documenting the potency of evaluating environmental PAH pollution in any studied tissues of fish. Except for the higher concentrations detected in gill and bladder of silver carp than bighead carp, total PAHs observed in other tissues of bighead carp were all higher than silver carp. Additionally, sum of PAH residues in all tissues of different fish species was found at the concentrations of 1964 and 1458 ng g^{-1} ww for bighead carp and silver carp, respectively. Nap, Any, Phe and BkF mainly accounted for such difference with obviously higher concentrations of 173, 334, 519 and 89.2 ng g^{-1} ww detected in bighead carp comparing with silver carp (81.9, 202, 126 and 12.6 ng g^{-1} ww for Nap, Any, Phe and BkF, respectively). However, the concentrations of Flu, Ant, Pyr, Chr, BbF, and BghiA observed in bighead carp (ranging from 6.65 to 238 ng g^{-1} ww) were a little lower than silver carp (ranging from 16.9 to 278 ng g^{-1} ww).

The results of the present study indicated that fish of Poyang Lake were contaminated mainly with low-molecular weight (LMW) compounds (total concentrations of two- and three-ringed PAHs ranged from 70.4 to 281 ng g^{-1} ww for bighead carp and from 27.1 to 191 ng g^{-1} ww for silver carp), while the high-molecular weight (HMW) PAHs (four-, five- and six-ringed PAHs) were detected at lower concentrations of 23.6–232 ng g^{-1} ww for bighead carp and 14.4–210 ng g^{-1} ww for silver carp, respectively, which was consistent with the studies carried out in Gomti River of India (Malik et al., 2008) and Hiroshima Bay (Deb et al., 2000). Among PAHs, Phe was detected at the highest concentration in most tissues of bighead carp included bile, scale, heart, muscle, gill, skin and bladder with the concentrations ranging from 28.8 to 95. The 7 ng g^{-1} ww, while Any was the most predominant compound in kidney (75. 6 ng g^{-1} ww) and liver (58.4 ng g^{-1} ww), respectively. While for other congeners, BbF (1.98–89.4 ng g^{-1} ww), Pyr (10.6–66.4 ng g^{-1} ww), Nap (1.43–81.0 ng g^{-1} ww) and Ant (6.85–44.0 ng g^{-1} ww) were also the main contributors to total PAHs in bighead carp. Vives et al. (2004) also pointed out the Phe was the most abundant contributor to PAHs in fish samples from remote and high mountain lakes in Europe and Greenland. In addition, the predominance of Phe is consistent with the PAH compositions found in fish liver from other freshwater (Pointet and Milliet, 2000) and marine systems (Baumard et al., 1998). BaP and IP were not detected in any bighead carp samples, while Ana was only detected in skin and liver and Flt was only observed in kidney. As for silver carp, Pyr showed the highest concentrations in bile (113 ng g^{-1} ww), heart (56.3 ng g^{-1} ww) and muscle (12.7 ng g^{-1} ww), while BbF was the most dominant congener in gill, skin, bladder and liver ranging from 32.0 to 64.5 ng g^{-1} ww. Ant and Any were found at the highest concentrations in scale (28.2 ng g^{-1} ww) and kidney (28.6 ng g^{-1} ww), respectively. Therefore, Any, Phe, BbF, Nap and Ant were also the main contributors to PAHs in silver carp, showing similar compositions of individual PAHs as bighead carp. Ana, Flt, BaA and IP also showed the least detection frequency without being detected in any tissues of silver carp. Relative higher concentrations of such PAH congeners as Nap, Phe and Ant with lower octanol-water partition coefficients (Log Kow less than 5.0) in both fishes were due to their higher gill-water transfer efficiencies as suggested by Cheung et al. (2007), while the lower residues of compounds with larger molecular weight and higher Kow (Log Kow larger than 5.0) might be mainly attributed to the enhanced biotransformation as well as the decreased gut assimilation in fish (Liang et al., 2007). This phenomenon was also in good agreement with the findings of DouAbul et al. (1997) who found that Nap was the most abundant PAH compound in fish from Red Sea Coast of Yemen.

3.2. Tissue distribution of PAHs

The PAHs present in the water column are available to fish via different uptake routes, such as dietary intake and/or through direct gill-water transfer. As a general rule in fish, water is the dominant source of exposure for organic compounds with low Kow, while sediment particles (as food) can contribute substantially to bioaccumulation for those with high Kow (Beyer et al., 2010). Therefore, the higher concentrations of LMW PAHs in both fishes implied the gill-water transfer might be mainly responsible for the detected PAHs in the selected filter-feeding fish.

For both fish species, the highest concentration of PAHs was found in bile, while the lowest residues of PAHs were found in gill of bighead carp and muscle of silver carp, respectively. Higher residues were also found in heart for both species, with the concentrations of 312 ng g^{-1} ww for bighead carp and 269 ng g^{-1} ww for silver carp, respectively. In addition, PAH residues in kidney and liver were also higher than in muscle for both fish (Table 1). The relative lower residues of PAHs found in gill for both fish indicated the tissue distribution first to gills and second to the remaining tissues might not be very efficient due to the enhanced biotransformation capacity of the liver (Carrasco Navarro et al., 2012).

Since PAHs are lipophilic, they would prefer to be accumulated in tissues with higher lipid contents after external membrane absorption or dietary intake. However, the extremely weak positive correlations were found out between PAHs and lipid contents (g g^{-1} ww) in tissues (bighead carp: $r=0.009$, $p=0.982>0.05$, $n=9$; silver carp: $r=0.462$, $p=0.211>0.05$, $n=9$), documenting lipid content was not the key factor determining the tissue-specific accumulation of PAHs in fishes. On the other hand, PAHs can be oxidized by enzyme systems (such as cytochrome P450 enzymes) to epoxides and hydroxylated derivatives during phase I biotransformation, and these products are further converted into highly water-soluble conjugates (e.g., glucuronides or sulfates) during phase II biotransformation to facilitate excretion. Generally, liver is the major site of fish for phase I and phase II biotransformation of PAHs, and the intermediates are subsequently stored in the bile in the gall bladder for elimination (Beyer et al., 2010). As a result, PAHs may act as stowaways in the enterohepatic circulation and will thereby stay longer in the liver-bile biotransformation-excretion system, which thus induced higher residues in the hepatobiliary system as demonstrated in our study. For both fish, muscle was not the target organ for the accumulation of PAHs, which was consistent with the exposure experiments performed with BaP showing that the concentrations in liver were proximately 100 times higher than that in muscle (Varanasi and Stein, 1991; Vives et al., 2004; Ballesteros et al., 2011). Furthermore, the distribution of PAHs in muscle and viscera was also in agreement with that previously reported, with the hepatobiliary system (such as bile and liver) accumulated higher PAHs and their metabolites than in extrahepatic tissues (such as muscle) (Liang et al., 2007; Balk et al., 1984), but different routes of exposure (the exchange between water and gill as well as the dietary intake) resulted in the similar distribution pattern (Varanasi et al., 1989). Generally, the residues in gill as well as other external membrane tissues mainly reflected the concentrations of contaminants in the waters where the fish live; whereas, the concentrations in bile, liver and kidney represented the final storage of hydrophobic chemicals (Karadede et al., 2004). It can therefore be expected that PAHs in fish are more easily detected in the viscera than in the muscle, and then the PAH levels in muscle as well as in the whole body can be well predicted and calculated from those in the viscera

tissues as suggested by previous studies (Oliveira Ribeiro et al., 2005; Johnson-Restrepo et al., 2008; Beyer et al., 2010). Furthermore, the higher residues of parent PAHs observed in bile for both fishes also indicated recent and continuous PAH input around Poyang Lake.

3.3. Compositions and sources of PAHs

In terms of the number of fused rings present in the chemical structure of PAHs, it was observed that three-ringed hydrocarbons were the dominant contributors to the PAH burden in tissues of bighead carp (38.9–79.0 percent) and silver carp (28.1–68.4 percent), followed by four- and five-ringed chemicals (Fig. 2). Nap detected in bighead carp contributed to 0.5–15.8 percent of total PAHs, while six-ringed PAHs were the lowest contributors accounting for 0.04–0.7 percent of PAHs. As for silver carp, Nap accounted for 0.4–13.9 percent in tissues and six-ringed chemicals only accounted for 0.15–3.5 percent of total PAHs, thus documenting a similar PAH profiles in both fish species. It was clear that in fish tissues LMW PAHs were prevalent than HMW PAHs, which was in agreement with the reported dominating accountancy (about 90 percent) of two- and three-ringed PAHs in fish muscles conducted by Liang et al. (2007).

Although fish can biotransform PAHs to hydrophilic intermediates using an active oxidative enzymatic system, some studies also pointed out the occurrence of parent PAHs in fish organs had been related to recent episodes of pollution exposure in surrounding environment (Collier and Varanasi, 1991; Pointet and Milliet, 2000; da Silva et al., 2007). PAHs emitted from different sources would exhibit different molecular compositions. Generally, the petrogenic sources are more abundant in LMW PAHs and the pyrogenic sources contain greater percentage of HMW PAHs. Molecular indices based on the ratios of selected PAHs have been widely used to differentiate PAHs from pyrogenic and petrogenic origins (Aichner et al., 2007; Chen et al., 2005; Peng et al., 2011). The usefulness of these indices relies on the fact that during low temperature processes such as catagenesis of organic matter leading to the formation of petroleum, the PAH distribution is governed by thermodynamic properties. For high temperature processes, such as pyrolysis of organic matter, their distribution is governed by kinetic characteristics (Tang et al., 2005). Based on the PAH isomer pair ratio measurements compiled by Yunker et al. (2002), the ratios of LMW/HMW, Ant/(Ant+Phe), BaP/BghiP and CombPAH/∑PAHs were used to preliminary distinguish between combustion and petroleum sources of PAHs around Poyang Lake (Table 2). Although the ratios of LMW/HMW in bile, gill, skin and bladder were less than 1 for silver carp, most of the values, especially for bighead carp (ranged from 1.03 to 4.71), accompanied by the percentage analysis aforementioned indicated the predominance of LMW PAHs originating from low temperature pyrolytic processes such as biomass burning and/or petrogenic sources (Ping et al., 2007). This phenomenon could also be concluded from the ratios of combPAHs/∑PAHs (sum of combustion originated PAHs to total PAHs) observed for bighead carp (0.18–0.49) and silver carp (0.22–0.58). In addition, the ratios of Ant/(Ant+Phe) detected in both bighead carp and silver

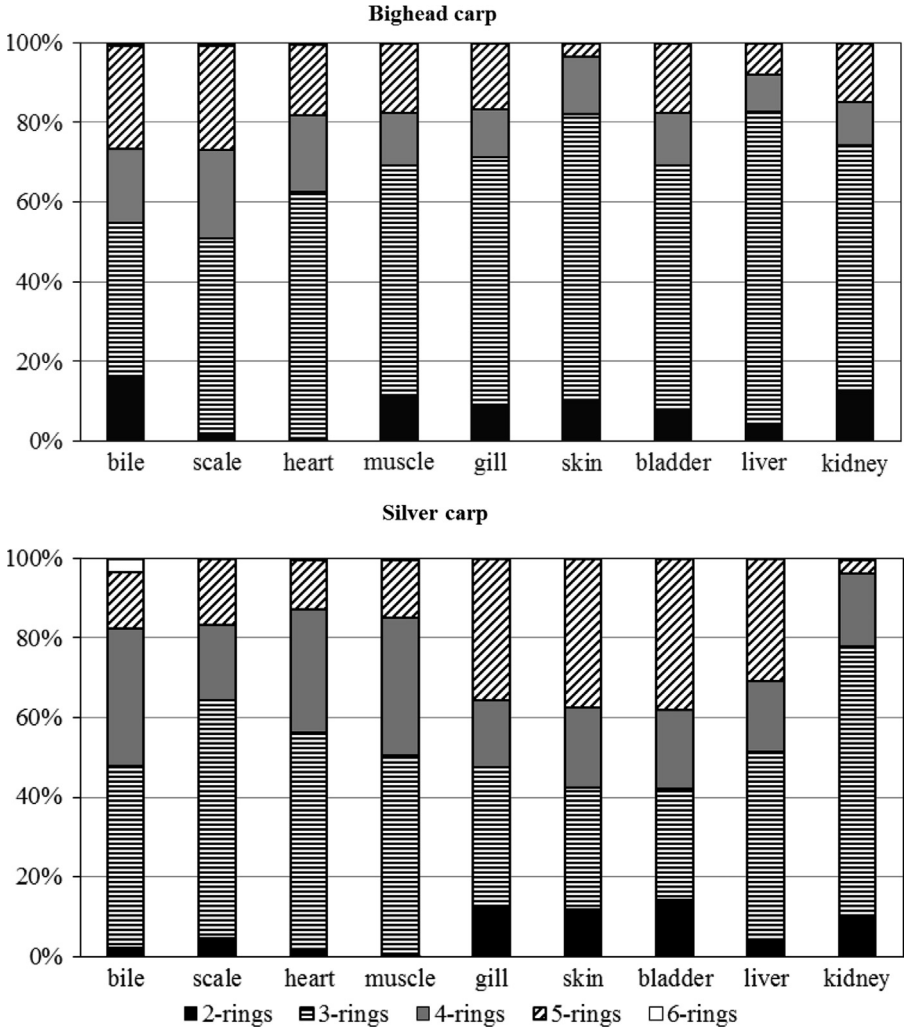

Fig. 2. PAH profiles with regard to the aromatic-ring number of PAHs in different tissues of bighead carp and silver carp from Poyang Lake.

Table 2
Molecular diagnostic ratios of PAHs in different tissues of bighead carp and silver carp.

Tissues	Bighead carp			
	LMW/HMW[a]	Ant/(Ant+Phe)[b]	BaP/BghiP[c]	CombPAHs/∑PAHs[d]
Bile	1.21	0.32	1.81	0.45
Scale	1.03	0.32	2.12	0.49
Heart	1.67	0.24	2.31	0.38
Muscle	2.24	0.17	20.38	0.31
Gill	2.46	0.16	10.32	0.29
Skin	4.59	0.16	6.39	0.18
Bladder	2.23	0.17	14.48	0.31
Liver	4.71	0.15	1.79	0.18
Kidney	2.87	0.16	12.15	0.26
	Silver carp			
Bile	0.91	0.62	0.21	0.52
Scale	1.80	0.63	4.42	0.36
Heart	1.27	0.68	1.01	0.44
Muscle	1.01	0.56	1.42	0.50
Gill	0.90	0.54	1.19	0.53
Skin	0.73	0.69	2.25	0.58
Bladder	0.73	0.58	6.93	0.58
Liver	1.05	0.56	2.46	0.49
Kidney	3.50	0.50	1.46	0.22

[a] The ratios of total LMW chemicals (two- and three-ringed PAHs) to HMW chemicals (four-, five- and six-ringed PAHs). The petrogenic sources are more abundant in LMW PAHs and the pyrogenic sources contain greater percentage of HMW PAHs.

[b] The ratio <0.1 is taken as an indication of petroleum while a ratio >0.1 indicates a dominance of combustion.

[c] The ratio BaP/BghiP could be used to elucidate the traffic (>0.6) and non-traffic sources (<0.6).

[d] The ratios of PAHs from combustion origin (Flt, Pyr, BaA, Chr, BbF, BkF, BeP (not included here), BaP, BghiP, IP) to total 16 PAHs.

carp were all less than 1.0 with the values at the range of 0.15–0.32 in bighead carp and 0.50–0.69 in silver carp, respectively, documenting the dominance of combustion origin of PAHs. The Phe/Ant ratios were also calculated to fractionate the detailed sources of PAHs (2.13–5.57 for bighead carp and 0.45–1.01 for silver carp, respectively), which demonstrated the gasoline combustion might be responsible for PAH residues in fish from Poyang Lake (Yunker et al., 2002). While for the BaP/BghiP ratios, they were found in the range of 1.79–20.38 in bighead carp and 0.21–6.93 in silver carp, respectively, indicating the potential origin of PAHs from traffic sources as suggested by Yunker et al. (2002). Therefore, PAHs observed in fishes from Poyang Lake originated from both the petrogenic and pyrogenic sources, and gasoline combustion coupled with increasing traffic sources might be the dominant process needed further detailed identification in the future study.

3.4. Human health risk assessment

Some PAHs and especially their metabolic products are of great concern due to their documented carcinogenicity, however, reports concerning dietary health risk assessment for PAHs are rather limited (Xia et al., 2010). BaP is the only PAH for which toxicological data are sufficient for derivation of a carcinogenic potency factor among all known potentially carcinogenic PAHs (Agarwal et al., 2009). To understand the magnitude of dietary exposure of PAHs by fish consumption, the toxic equivalency factors (TEFs) were used to quantify the carcinogenicity of other PAHs relative to BaP and to estimate BaP-equivalent concentration (BaPeq). The calculated TEF is 0.001 for NaP, Any, Ana, Flu, Phe, Flt and Pyr, 0.01 for Ant, Chr and BghiP, 0.1 for BaA, BbF, BkF and IP, 1 for BaP and DahA (Nisbet and LaGoy, 1992; Liao and Chiang, 2006; Peng et al., 2011). The BaPeq was defined as the potency equivalent concentration (PEC) of total PAHs, which was calculated according to the following Eq. (1) suggested by Xu et al. (2011) for comparison with the screening values of PEC of total PAHs (0.67 ng g^{-1} ww) suggested by USEPA (2000) for human fish consumption. In addition, the incremental life time cancer risk (ILCR) of the dietary exposure to PAHs was also calculated based on Eqs. (2) and (3) (Xia et al., 2010). A public screening criteria for carcinogens which is set at a carcinogenic risk level of 1.0E−06 was used for assessment. One in a million chance of additional human cancer over a 70 year lifetime (ILCR=1.0E−06) is the level of risk considered acceptable or inconsequential, while the additional lifetime cancer risk of one in ten thousand or greater (ILCR=1.0E−04) is considered serious and there is high priority for paying attention to such health problems.

$$\mathrm{PEC} = \sum_{i=1}^{n} \mathrm{TEF}_i \times C_i \quad (1)$$

$$\mathrm{ILCR} = \frac{\mathrm{ED} \times \mathrm{EF} \times \mathrm{EDI} \times \mathrm{SF} \times \mathrm{CF}}{\mathrm{AT}} \quad (2)$$

$$\mathrm{EDI} = \frac{\mathrm{CR} \times C}{\mathrm{BW}} \quad (3)$$

where PEC is the potency equivalent concentration of total PAHs; TEF is the toxic equivalent factors of individual PAH compounds; C is the concentration of each PAH observed in target samples. ILCR is the incremental lifetime cancer risk of the dietary exposure (dimensionless); ED is the exposure duration (43 for adults, year); EF is the exposure frequency (365 day yr^{-1}); SF is the oral cancer slope factor of BaP (geometric mean of 7.3 mg kg^{-1} day^{-1}); CF is the conversion factor (10E−06 mg ng^{-1}); AT is the average lifespan for carcinogens (25,500 days). EDI is the estimated daily dietary PAH exposure for human (ng kg^{-1} body weight (bw) day^{-1}); CR is the consumption rate of fish (30.5 g $person^{-1}$ day^{-1}); BW is the average body weight of people (70 kg for adults).

For PAHs, total concentrations of potentially carcinogenic PAHs, including BaA, BkF, BbF, BaP, DahA, IP and Chr (relatively weak carcinogen) (Bhargava et al., 2004; Nadal et al., 2004), varied from 7.48 to 162 ng g^{-1} ww for bighead carp and from 5.59 to 82.9 ng g^{-1} ww for silver carp, respectively. Both the highest residues of such 7 carcinogenic PAHs were observed in bile, which accounted for 5.7–33.1 percent of total PAHs for bighead carp and 8.6–45.2 percent for silver carp, respectively. Additionally, the PEC of total PAHs in different tissues ranged from 1.68 to 24.9 ng g^{-1} ww for bighead carp and from 0.73 to 13.3 ng g^{-1} ww for silver carp (Fig. 3), respectively, which were all higher than the recommended PEC of total PAHs (0.67 ng g^{-1} ww) for human consumption. As for the main edible parts of fish, PEC in muscle was 5.00 ng g^{-1} ww for bighead carp and 1.89 ng g^{-1} ww for silver carp, demonstrating potential toxic effects on human beings induced by PAHs in both fish species by fish consumption around Poyang Lake.

The cumulative probability distributions of calculated ILCR for different tissues of bighead carp and silver carp from Poyang Lake are presented in Fig. 3. ILCR values in different tissues of bighead carp and silver carp were in the range of 3.3E−06–4.9E−05 and 1.4E−06–2.6E−05, respectively, being higher than the acceptable risk level (1.0E−06) and lower than the priority risk level (1.0E−04). As for the major edible parts, ILCR values in muscle were 9.8E−06 for bighead carp and 3.7E−06 for silver carp, respectively. In addition, higher ILCR was also found in liver of silver carp with the value of 1.0E−05 comparing to other major edible parts (muscle and skin) of fish, thus documenting high potential carcinogenic risk for fish consumption at present around Poyang Lake. Furthermore, biotransformation of PAHs in fish and other aquatic organisms generally accompanied by side effects resulting from the formation of carcinogenic intermediates (Johnson-Restrepo et al., 2008; Lazartigues et al., 2011). In fact, those

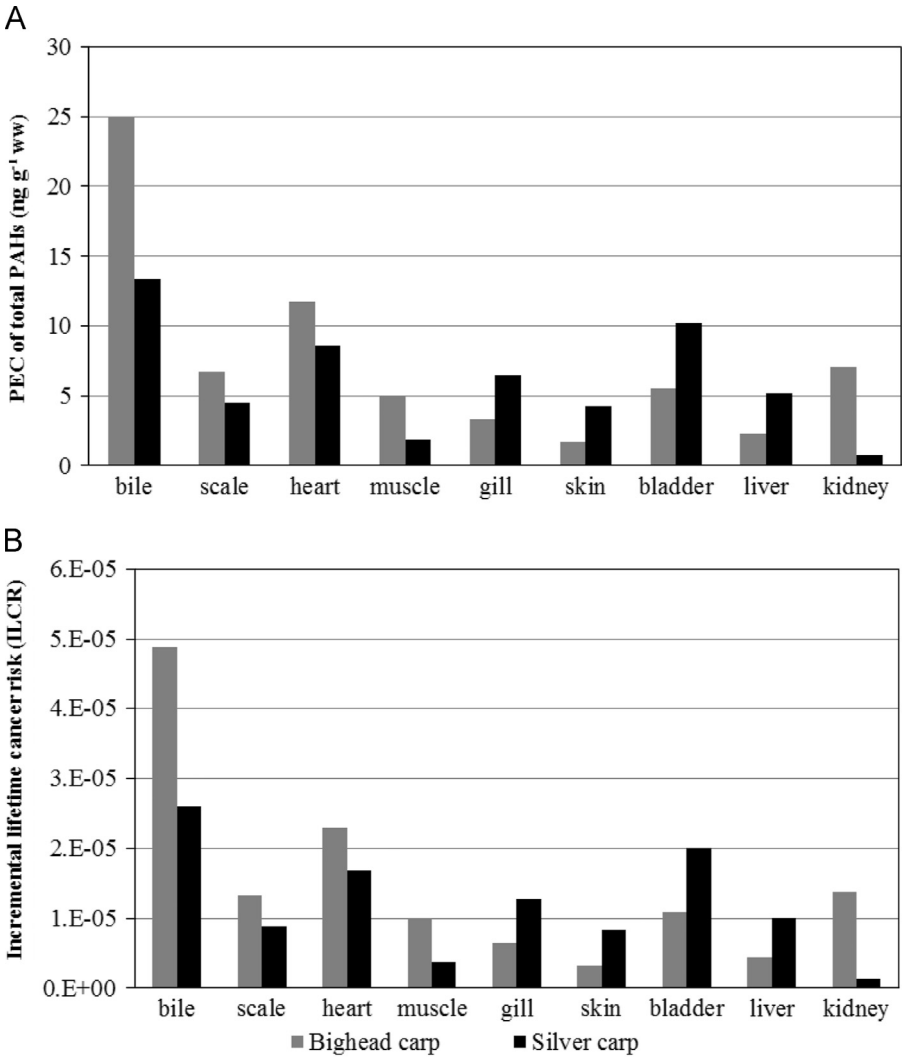

Fig. 3. The potency equivalent concentration (PEC) (A) and incremental lifetime cancer risk (ILCR) (B) of total PAHs in tissues of bighead carp and silver carp from Poyang Lake.

PAH metabolites formed by photochemical/chemical reactions are known to be more toxic than their parent PAHs base on the tumorigenicity studies in a new born mouse bioassay (Jung et al., 2010). Therefore, the potential carcinogenic risk of PAH exposure might be underestimated without considering the concentrations of their metabolites at present. On the other hand, many PAHs and their metabolites are aryl hydrocarbon receptor (AhR) ligands and may activate estrogen receptors (ER) (Santodonato, 1997), which has also been demonstrated by Kummer et al. (2008) that BaP, BaA, Flt and BkF could induce significant estrogenic effects in vivo, and thus might affect their toxic impact and carcinogenicity. Therefore, 100 percent detection frequency of both Flt and BkF in tissues of bighead carp and silver carp would more or less increase the carcinogenic risk of PAHs in fishes from Poyang Lake.

4. Conclusions

PAHs were detected in all tissues of freshwater fish, bighead carp and silver carp, collected from Poyang Lake. The predominance of LMW PAH congeners indicated the gill-water transfer might be the key exposure route for studied fish species. Tissue distribution demonstrated hepatobiliary system accumulated higher concentrations of PAHs than the extrahepatic tissues. PAHs around Poyang Lake were from the combined pollution sources of petrogenic and pyrogenic processes. Gasoline combustion coupled with increasing traffic sources might be the dominant origin of PAHs, which needed further detailed studies conducted on different environmental compartments around this area. Human health risk assessment documented PAHs might induce potential carcinogenic effects on human beings through fish consumption at present. More related studies should be carried out around Poyang Lake in the future considering the increasing toxic effects of PAH metabolites.

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