Ecotoxicology and Environmental Safety 185 (2019) 109666

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Comparisons of tissue distributions and health risks of perfluoroalkyl acids (PFAAs) in two fish species with different trophic levels from Lake Chaohu, China

Jing-Yi Wu[a,1], Wen-Xiu Liu[a,b,1], Wei He[a,c], Fu-Liu Xu[a,*]

[a] MOE Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing, 100871, China
[b] Center for Enviornmental Health Risk Assessment and Research, Chinese Research Academy of Environment Sciences, Beijing, 100012, China
[c] MOE Key Laboratory of Groundwater Circulation and Environmental Evolution, China University of Geosciences (Beijing), Beijing, 100083, China

ARTICLE INFO

Keywords:
Perfluoroalkyl acids (PFAAs)
Fish tissues
Trophic level
Residues
Health risk
Lake Chaohu

ABSTRACT

Perfluoroalkyl acids (PFAAs) are a type of persistent organic pollutants that are widely distributed in multiple environmental media and organisms and have a teratogenic effect on and toxicity to animals and humans. The residual levels of seventeen PFAAs in the tissues of two regular consumption fish species, *Culter erythropterus* and *Aristichthys nobilis* in Lake Chaohu were measured by a high-performance liquid chromatograph – mass spectrometer (HPLC-MS). The distributions of PFAAs and the effect of the lipid contents were analyzed, and the health risks of typical PFAAs were evaluated. The results showed that perfluorohexanoic acid (PFHxA) was the predominant contaminant (80.50 ± 58.31 ng/g and 19.17 ± 12.57 ng/g wet weight, ww), followed by perfluorooctanesulfonic acid (PFOS) (55.02 ± 34.82 and 14.79 ± 6.24 ng/g, ww) in both fish. The level of total PFAAs was the highest in the liver tissues of *Culter erythropterus* (359.87 ng/g, ww) and the lowest in the kidney tissues in *A. nobilis* (10.06 ng/g, ww). Due to the higher trophic level of *C. erythropteru*, the total PFAA concentrations were significantly higher in all tissues than those in *A. nobilis*. Liver muscle ratio of *C. erythropteru* was the highest, indicating the most accumulation in the liver. The concentrations of PFAAs in fish tissues were influenced by the lipid content, resulting in a difference between the lipid-normalized concentrations and the wet weight concentrations of the PFAAs. The non-carcinogenic risks of PFOS were higher than those of PFOA through the ingestion of *C.* erythropterus *and A. nobilis*. Both the carcinogenic and non-carcinogenic risks of *C. erythropterus* were greater than those of *A. nobilis,* and fish tissue intake could cause an increasing of risks up to 60%, indicating that long-term and large amount ingestion of carnivorous fish and related tissues with higher trophic level, such as *C. erythropterus* should be avoided.

1. Introduction

Perfluoroalkyl acids (PFAAs), including perfluorosulfonic acids (PFSAs) and perfluoroalkyl carboxylic acids (PFCAs) are typical persistent organic pollutants (POPs) that are widely distributed in water, sediments, atmosphere, and organisms. They are difficult to degrade and can survive long-distance transportation. They are widely used in floor polish, cleaning products, paper, textile treatment and fire-fighting foam as surfactants because of their stability to high temperatures, ultraviolet radiation, strong acids and bases, and oxidants (Bultman and Pike, 1981; Asahi, 1982; Cella et al., 1976; Zoellner-Braue, 1995; Schultz and Quessy, 1992; Enders, 1961). The representative substance PFOS and its salts were listed in Annex B to the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2009), moreover, PFOA and PFHxS and their salts were also proposed to be added into the Annex (UNEP, 2015, 2017). It has been proved that the building of wastewater treatment plants (WWTPs) is the most effective approach to control and prevent the PFAAs pollution in water, since WWTPs could serve as a barrier to purify wastewater and prevent PFAAs from directly entering the water environment. Worldwide researchers have made intensive efforts to figure out the occurrence, environmental behavior and mass load of perfluoroalkyl substances (PFASs) in WWTPs from different countries or regions, including the United States (US) (Hu et al., 2016), European Union (Eriksson et al., 2017), Australia (Gallen et al., 2018, 2016), South Korea (Kwon et al., 2017), China (Chen et al., 2018) and so on.

* Corresponding author.
E-mail address: xufl@urban.pku.edu.cn (F.-L. Xu).
[1] Co-first author.

https://doi.org/10.1016/j.ecoenv.2019.109666
Received 15 May 2019; Received in revised form 27 August 2019; Accepted 7 September 2019

Since the occurrence of PFOS in animal tissues, including those of fish, birds, and marine mammals, was first reported (Giesy and Kannan, 2001), PFAAs have been globally detected in various media, including freshwater (Zhou et al., 2012, 2013; Sun et al., 2011), drinking water (Xiao et al., 2013), sediment (Benskin et al., 2011), air (Dreyer et al., 2010; Webster and Ellis, 2012; Liu et al., 2018a,b), etc. The biomagnification of PFAAs in the aquatic food web has been shown in several studies. (Kannan et al., 2002a, 2002b; Taniyasu et al., 2003; Houde et al., 2006; Kumar et al., 2009). The hepatotoxicity, developmental toxicity, immunotoxicity, and carcinogenicity of PFAAs, as well as their ability to interfere with hormone levels have been reported. (Seacat et al., 2002; Takacs and Abbott, 2007; Wolf et al., 2007; DeWitt et al., 2008; White et al., 2011; Du et al., 2013; Keiter et al., 2016). PFAAs could be accumulated through the trophic level, and pose health risks. Therefore, it is crucial to study the tissue distribution of PFAAs in fish, especially in edible tissues, for assessing the health risk of PFAAs to human body through eating fish. Meanwhile, studying their distribution characteristics in fish tissues is of great significance for further understanding the toxic effects, target organs and toxic mechanisms of PFAAs in fish. In China, the concentrations of PFAAs in fishes have been reported in several studies (Gulkowska et al., 2006; Yang et al., 2012; Fang et al., 2014; Liu et al., 2018a,b); however, there is still less studies on the tissue distributions of PFAAs in fishes. More studies are necessary because it is important for studying the environmental behavior of PFAAs and their influence on health risk. This paper presents a case study on the tissue distributions of PFAAs in two fish species with different trophic levels from Lake Chaohu, China, and our aim is to investigate their influencing factor and the carcinogenic and non-carcinogenic risks of PFAAs.

Lake Chaohu, located in the middle of Anhui Province, is the fifth largest freshwater lake in China. Large amount of domestic and industrial sewage has been poured into Lake Chaohu by the surrounding cities, especially Hefei City, in recent decades. Pollution in Lake Chaohu and the surrounding environment is attributed to an increase in population and the dramatic development in industry and agriculture during this period. Water quality deterioration pose a direct threat to the lives of residents, ecosystems, biota, flora, and industries along the lake. Studying the distribution and health risks of PFAAs in the organisms of Lake Chaohu is significant for understanding the present pollution levels, the environmental behaviors, and the ecological effects of PFAAs in human and biota; and for providing theoretical support for pollution prevention and environmental management.

Two main edible fish species, *Culter erythropterus* and *Aristichthys nobilis,* found in Lake Chaohu were investigated in this study. The residual levels of PFAAs in the fish tissues were measured to elucidate the distribution and the influencing factors of PFAAs and to evaluate the carcinogenic and non-carcinogenic risks of typical PFAAs.

2. Materials and methods

2.1. Sample preparation and extraction

Two main local edible fish in Lake Chaohu, the topmouth culter (*Culter erythropterus*) and the bighead carp (*Aristichthys nobilis*), were collected in August 2011. The fish tissues were sampled separately, including muscle, gill, heart, liver, pancreas, swim bladder, intestine, spleen, and, kidney. Tissue samples were then transported to the lab for frozen storage. Basic physiological characteristics of the *Culter erythropterus* and *Aristichthys nobilis* are listed in Table S1. However, the GPS data was unavailable due to obtaining the fish at on a fishing boat by the lakeside.

To reduce individual fish differences, tissues from three to five fish of the same species were combined into one mixed sample. After the wet weights were obtained, samples were freeze-dried for three to four days to completely remove any moisture, weighed to obtain the dry weight, and grounded into powder. Afterwards, samples were bottled for further processing. PFAAs extraction from the tissue samples followed a published method (Giesy and Kannan, 2001) with several modifications.

The 0.5-g tissue samples were weighed into a 15-ml polypropylene centrifuge tube. A five nanogram of isotope internal standard, including [1,2,3–13C3]PFHxS, [1,2,3,4–13C4]PFOS, [1,2,3,4–13C4]PFBA, [1,2,3,4–13C4]PFOA, [1,2–13C2]PFDoA (Wellington Labs, Ontario, Canada), 1 ml of 0.5 ml/L TBAS (Tetrabutylammonium sulfate), and 2 ml of 0.25-mol/L sodium carbonate solution (pH = 10) were added to the tube. After 5 ml of MTBE (methyl tert-butyl ether) was added, the tube was vortexed for 15 min and centrifuged, and the supernatant was then transferred. The procedure was repeated twice, and the supernatants were mixed, dried under high purity nitrogen, and then redissolved by methanol. The ENVI-CARB cartridges (3 ml, 250 mg, Sigma-Aldrich Co., USA) and WAX cartridges (6 ml, 150 mg, Waters Corp., USA) were used sequentially to purify the samples. The ENVI-CARB cartridges were conditioned by 1 ml of methanol for three times and the WAX cartridges were conditioned by 4 ml of 0.1% NH_4OH in methanol, then 4 ml of methanol, followed by 4 ml of ultrapure water before use. The samples were loaded onto the ENVI-CARB, and the cartridges were washed with 1 ml of methanol for three times. Then, the eluents were loaded on the WAX cartridges. After drying for 1 min, the WAX cartridges were washed with 4 ml of ammonium acetate (pH 4.0, 25 Mm) and then dried with a vacuum pump. The SPE cartridges were preserved for further elution and purification. Then, 4 ml of methanol and 4 ml of 0.1% NH_4OH in methanol were added to the cartridges for elution. The eluents were concentrated to 1 ml under high purity nitrogen and were transferred to brown glass vials with a needle polypropylene filter (13 mm, 0.2 μm, Pall Corp., USA).

2.2. PFAAs measurement and quality control

The PFAA contents were measured using a published method (Wang et al., 2014). Agilent 1290–6460 HPLC-MS (Agilent Technologies, Palo Alto, CA) was applied with a Zorbax Eclipse Plus C18 column (2.1 × 100 mm, 3.5 μm, Agilent Technologies, Palo Alto, CA). The column temperature was 40 °C. The 5-μl samples were injected and flowed at the rate of 0.3 ml/min. The 2-mM ammonium acetate solution was applied as the mobile phase A and acetonitrile as mobile phase B. Gradient elution was initiated with 80% of mobile phase A and 20% of mobile phase B for 13 min, followed by 10% of mobile phase A and 90% of mobile phase B for 1 min.

The mass spectrometry conditions were as follows: negative ESI modes, atomization temperature at 350 °C, auxiliary gas (N_2) at a flow rate of 9 L/min and a capillary voltage of 3500 V.

Polypropylene bottles were used for sample collection. Fluorine materials were strictly avoided during extraction and analysis. A field blank, a process blank and a reagent blank were measured to ensure data quality. The procedure recovery rate and detection limit were tested before sample analysis (Table S2.).

2.3. Lipid content analysis

Two grams of freeze-dried samples were weighed into an extraction tube. After microwave extraction, the extracts were pressure filtered and concentrated to 1 ml by rotary evaporation. Then, 10 ml of ethyl acetate was added to the tube and again concentrated to 1 ml by rotary evaporation. A 0.45-μm filter was applied for purification, and the filtrates were transferred to the vials. Three milliliters of ethyl acetate was added to the vial, and the gel chromatograph (GPC800 +, Labtech, China) was used for sample purification.

Instrument conditions were as follows: Bio Beads SX-3 column (300 mm × 20 mm, Bio-Rad Laboratories, Inc. USA), the injection volume of 2 ml, a ratio of 1:1 ethyl acetate/cyclohexane at the flow rate of 5 ml/min.

Lipid contents were collected from 2 to 10 min and transferred to a

constantly weighed eggplant-shaped flask. The flasks were kept in a desiccator for 24 h after being dried by rotary evaporation. The weight increase was the weight of lipid contents.

2.4. Health risk evaluation

Health risks are typically divided into carcinogenic and non-carcinogenic risks. In this study, both carcinogenic and non-carcinogenic risks of PFOA and only non-carcinogenic risks of PFOS are discussed. Health risks of other PFAAs are not mentioned due to a lack of exposure parameters.

Potential carcinogenic risks are calculated using the method recommended by the United States Environmental Protection Agency (USEPA, 1989; USEPA, 2006). Carcinogenic risks of low-dose pollutants (Risk < 0.01) are calculated as follows:

$$Risk = CDI \times SF$$

CDI refers to chronic daily intake (mg/(kg·d)). SF refers to the pollutant slope factor (1/(mg/(kg·d))). The SF of PFOA is 0.07(1/(mg/(kg·d)))(USEPA, 2016a). The Lifetime Average Daily Dose model (USEPA, 1992) is applied for the CDI calculation. CDIs of PFAAs through fish consumption are calculated as follows:

$$CDI = C\times CR/BW$$

C refers to pollutant concentrations in the fish (mg/L). BW refers to the average human body weight (kg). CR refers to the consuming rate of fish (kg/d).

Non-carcinogenic risks are calculated as follows:

$$HQ = CDI/RfD$$

HQ refers to hazard quotient. RfD refers to reference dose, which is 0.00002 mg/(kg·d) for both PFOA and PFOS (USEPA, 2016a; USEPA, 2016b).

2.5. Other analysis and data processing

To calculated the trophic level, a published method waa used (Winemiller et al., 2007; Liu et al., 2015; Kong et al., 2016). All biota samples were analyzed for stable isotope ratios (15N/14N) using mass spectrometry (Finnigan MAT253, Thermo Fisher Scientific, Inc., USA).

The concentration data (ng/g) used in this study was calculated as the total weight (dry) of the PFAAs divided by the wet weight (ww), and lipid normalized weight (lw) of the fish samples. Microsoft Excel 2016 and SPSS 20.0 were used for data analysis.

3. Results and discussion

3.1. Distribution of PFAAs in the tissues of two fish

Seventeen PFAAs were detected in the fish tissues collected from Lake Chaohu. For most PFAAs, the detection rates were 100% in samples, except PFODA (73%) and PFHxDA (93%), which possess the longest carbon chain PFAAs. The mean concentrations of PFAAs in the fish tissues are shown in Table 1. PFHxA was the predominant contaminant in the tissues of *Culter erythropterus* and *Aristichthys nobilis* (80.50 ± 58.31 and 19.17 ± 12.57 ng/g, ww), respectively, followed by PFOS (55.02 ± 34.82 and 14.79 ± 6.24 ng/g, ww), and PFODA (0.01 ± 0.01 and 0.00 ± 0.01 ng/g, ww) had the lowest level. The total PFAAs concentrations were 194.22 ± 114.38 and 51.85 ± 25.05 ng/g, respectively.

The composition of PFAAs in the tissues of *A. nobilis* and *C. erythropterus* are presented in Fig. 1. The composition of PFAAs in the two fish species is similar, and both PFOS and PFHxA were the predominant contaminants. In *A. nobilis*, PFOS accounted for the highest proportion, ranging from 24.2% (intestine) to 41.9% (heart), and the percentage of PFHxA ranged from 14.6% (heart) to 48.3% (liver). The proportions of PFDA and PFUdA were roughly equivalent, ranging from 6.4% to 16.0% and 6.7%–12.8%, respectively. The percentage of PFODA was the lowest, ranging from 0.00% to 0.03%, and these three contaminants were all greatest in gill. In *C. erythropterus*, the proportion of PFHxA was the highest, ranging from 3.9% (meat) to 48.8% (pancreas), followed by the proportion of PFOS with the range from 19.7% (intestine) to 45.7% (meat). The percentages of PFHxDA and PFODA were the lowest, ranging from 0.00% to 0.05% and 0.00%–0.08%, respectively. A study showed that sulfonic acid groups form more hydrogen bonds with amino acids at the protein binding site than carboxylic acids, which may lead to the accumulation of PFOS more easily in tissues (Zhang et al., 2013). In addition, the carbon number of PFAAs was also an important influencing factor on distribution of PFAAs in biota, and the accumulation of long carbon chain PFAAs (more than 10) could decrease with the carbon number (Wen et al., 2019), which could explain the composition of PFHxDA and PFODA accounted for lower proportion.

Table 1
Distributions of different PFAAs in two fish tissues (ng/g, ww).

	Culter erythropterus	*Aristichthys nobilis*	Sample quantity	Detection rate
PFBA	4.49 ± 5.44	2.51 ± 4.15	41	100%
PFPeA	1.98 ± 2.85	1.02 ± 1.38	41	100%
PFHxA	80.50 ± 58.31	19.17 ± 12.57	41	100%
PFHpA	0.18 ± 0.20	0.05 ± 0.05	41	100%
PFOA	0.67 ± 0.23	0.25 ± 0.13	41	100%
PFNA	4.54 ± 2.57	1.35 ± 0.65	41	100%
PFDA	16.66 ± 9.18	4.78 ± 2.21	41	100%
PFUdA	18.82 ± 11.15	4.53 ± 1.98	41	100%
PFDoA	2.51 ± 1.41	0.67 ± 0.30	41	100%
PFTrDA	5.84 ± 3.48	1.27 ± 0.55	41	100%
PFTeDA	0.75 ± 0.45	0.22 ± 0.09	41	100%
PFHxDA	0.03 ± 0.03	0.02 ± 0.02	41	93%
PFODA	0.01 ± 0.01	0 ± 0.01	41	73%
PFBS	0.43 ± 0.41	0.18 ± 0.20	41	100%
PFHxS	1.69 ± 1.62	1.00 ± 0.60	41	100%
PFOS	55.02 ± 34.82	14.79 ± 6.24	41	100%
PFDS	0.10 ± 0.09	0.04 ± 0.03	41	100%
Total	194.22 ± 114.38	51.85 ± 25.05		

The distributions of total PFAAs in different fish tissues are shown in Fig. 2. The concentrations of total PFAAs in the tissues were quite different in fish at two trophic level. Among the tissues in *A. nobilis*, PFAA level was the highest in the intestine (78.93 ng/g, ww), followed by in the swim bladder (71.52 ng/g, ww), and was the lowest in the kidney tissue (10.06 ng/g, ww). Among the tissues collected from *C. erythropterus*, the PFAA levels were highest in the liver (359.87 ng/g, ww), followed by the gill and kidney (283.79 ng/g and 283.58 ng/g, ww, respectively), and in the muscle was the lowest (30.98 ng/g, ww). The relatively higher levels of PFAAs in liver were observed in both fish. This might be related to the uptake route and the enhancement of fish (Labadie and Chevreuil, 2011; Qiang et al., 2016). In addition, liver is an important organ for fat metabolism and protein synthesis, which might influence the association of PFAAs with lipid and protein. The highest level in intestine was observed in *A. nobilis*; it was consistent with other studies (Tomy et al., 2004; Xu et al., 2014; Hong et al., 2015), and suggested PFAAs could be accumulated through diet from water and sediment intake. This indicated the pollution condition of fish habitat could influence the concentrations in fish directly. Meanwhile, the PFAA level in muscle was lower. Studies showed that protein could influence the accumulation of PFAAs, and the protein content was usually at higher level in liver while lower in muscle (Vanden Heuvel et al., 1991, Labadie and Chevreuil, 2011; Wen et al., 2016, 2019). This could explain the high PFAAs concentrations in liver than muscle. Overall, as a carnivorous fish species, *C. erythropteru* could accumulate more PFAAs than the omnivorous fish *A. nobilis* due to the higher trophic level (Liu et al., 2015), and PFAAs could be transferred though

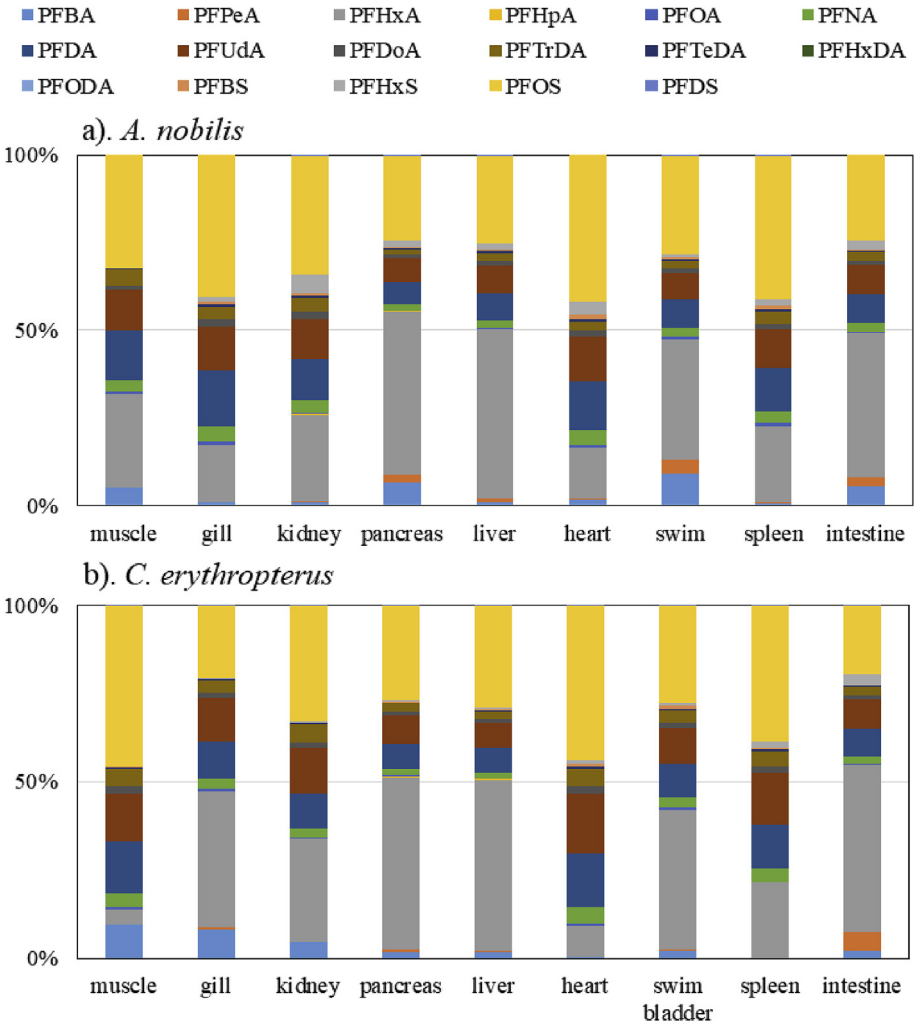

Fig. 1. The mean percent composition of seventeen PFAAs in different tissues of (a) *A. nobilis*, (b) *C. erythropterus* from Lake Chaohu.

predation and feeding in the food web (Müller et al., 2011; Xu et al., 2014; Liu et al., 2018a,b).

As an important organ, the bioaccumulation of contaminants in liver was concerned by liver-muscle-ratio (LMR) (Shi et al., 2012; Yang et al., 2012). LMR was calculated by dividing the individual PFAA concentration in liver by concentration in muscle. In *A. nobilis*, the LMRs for main contaminant PFOS, PFHxA, PFDA, and PFUdA were 3.41, 7.95, 2.47 and 2.91, while 7.40, 143.57, 5.26 and 6.18 in *C. erythropterus*. They were significant higher in *C. erythropterus* than those in *A. nobilis* ($p < 0.05$), which indicated the more accumulative in the fish at higher trophic level. This was consistent with previous studies (Murakami et al., 2011; Jörundsdóttir et al., 2014), and corresponding with the result that the liver of carnivorous fish was more easily to accumulate PFAAs than that of omnivorous fish.

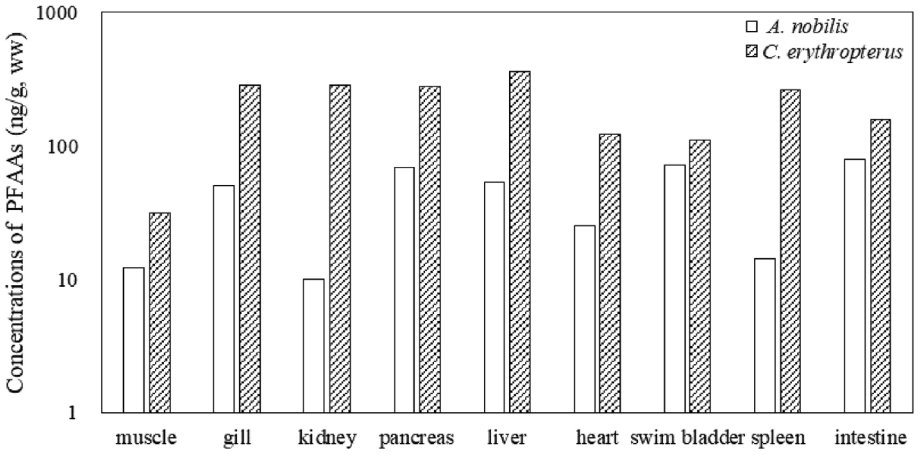

Fig. 2. Distributions of total PFAAs in tissues of *Aristichthys nobilis* and *Culter erythropterus* in Lake Chaohu.

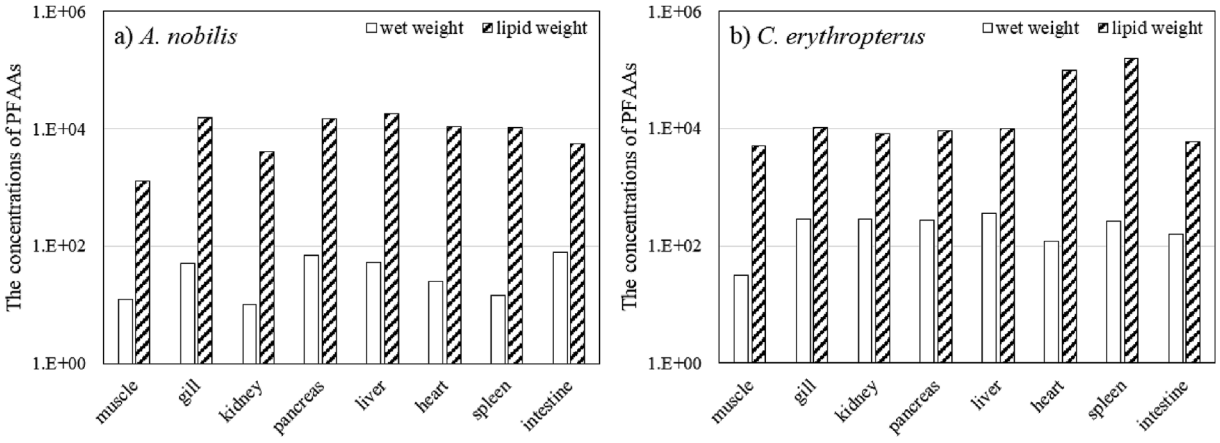

Fig. 3. Comparison of the wet weight concentrations and lipid-normalized concentrations of total PFAAs in tissues of *Aristichthys nobilis* and *Culter erythropterus* from Lake Chaohu.

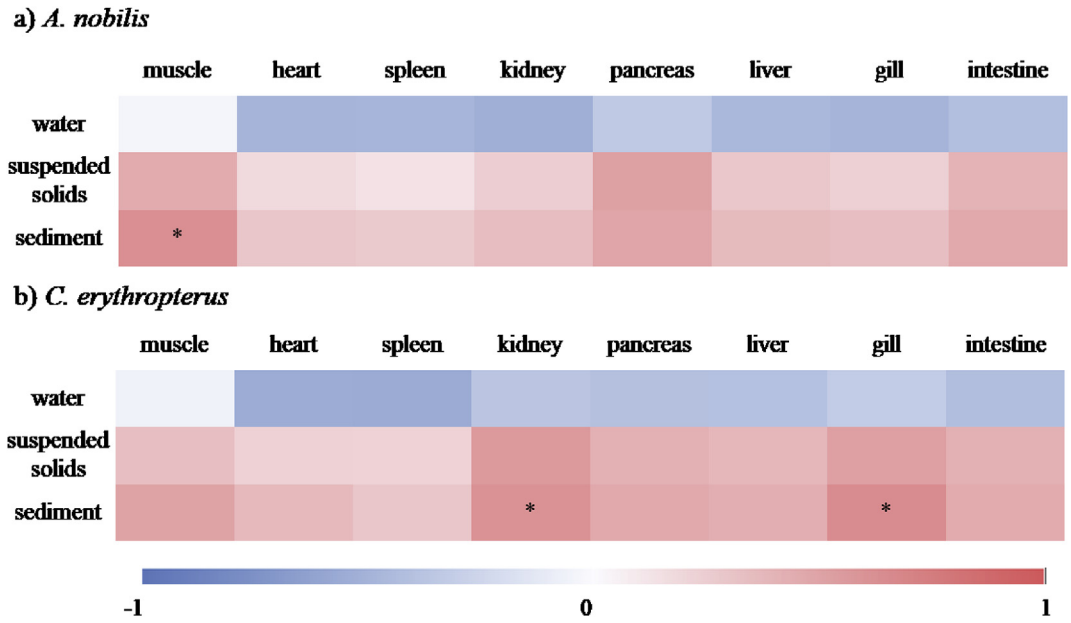

Fig. 4. The results of Pearson's Test between the concentrations in environmental media and fish tissues from Lake Chaohu (* significant at 0.05 level).

Table 2
Chronic intake risk of PFOA and PFOS in *Culter erythropterus* and *Aristichthys nobilis*.

	Muscles intake only			Considering tissue intake		
	PFOA		PFOS	PFOA		PFOS
	Carcinogenic	Non-carcinogenic	Non-carcinogenic	Carcinogenic	Non-carcinogenic	Non-carcinogenic
Culter erythropterus	9.27E-09	6.62E-03	3.94E-01	1.25E-08	8.92E-03	4.71E-01
Aristichthys nobilis	3.43E-09	2.45E-03	1.09E-01	5.27E-09	3.76E-03	1.70E-01

3.2. The influencing factors of PFAA distributions in fish tissues

3.2.1. Influence of lipid content

PFAAs are hydrophobic, oleophobic but lipophilic because of their chemical structure (Jing et al., 2009). Lipid content may influence the distributions of PFAAs in the tissues of an organism. The lipid content of tissues from the *C. erythropterus* and the *A. nobilis* from Lake Chaohu are shown in Table S1. The lipid content in the intestine of *C. erythropterus* was the highest (17.21%), while lowest in the heart (0.64%). The lipid-normalized concentrations of PFAAs in all tissues from two fish species are shown in Fig. 3. The mean lipid-normalized concentration of PFAAs in all tissues from *A. nobilis* and *C. erythropterus* were $1.00 \times 10^4 \pm 0.60 \times 10^4$ ng/g and $3.81 \times 10^4 \pm 5.79 \times 10^4$ ng/g (lipid-normalized, lw), respectively. The distribution of lipid-normalized concentrations of PFAA was different from that of wet weight concentrations (Fig. 3). The distribution of lipid-normalized PFAA concentrations in tissues from *A. nobilis* was similar to that of wet weight concentrations, due to the similar lipid contents in each tissue. However, the lipid contents varied in tissues from *C. erythropterus*.

The Pearson's test was used to analyze the relationship between the lipid contents of each tissue and the concentrations of each PFAAs in that fish tissue (Table S3). As a result, we found significant positive

correlations between the PFHxA and PFPeA levels found in the tissues of the *C. erythropterus* and the PFHxS level in the tissues of the *A. nobilis* and their lipid contents ($p < 0.05$). This finding indicates that lipid content might improve the accumulation of low-carbon chain PFAAs, compared to high-carbon chain PFAAs, in aquatic organisms, and research on the influence of serum lipid on PFOA and PFNA also reported that lipid could decrease the concentrations of contaminant (Nelson et al., 2010; Fu et al., 2014), which could supported our findings about the effect of lipid. However, the mechanism by which lipid reduced the accumulation of PFAAs is still not clear. In addition, the protein, which could bond with PFAAs, is another important factor and further study is necessary.

3.2.2. Influence of octanol-water partition coefficient (Kow)

The Spearman test was employed to analyze the correlations between lipid-normalized concentrations and logKow of PFAA (Table S4). However, no significant relationship was observed. It was reported that the accumulation of PFAAs in organisms could be influenced by the protein contents (Wen et al., 2016), which thus suggested that the influence of Kow on the PFAA concentrations in lipids was slightly lower than traditional persistent organic pollutants, such as organochlorine pesticides.

3.2.3. Influence of environmental media

Aquatic animals in lakes can ingest dissolved PFAAs in water through gill, meanwhile PFAAs in suspended matter and sediments may also enter aquatic animals through filter feeding and preying of benthic organisms. Therefore, the environment may have an important influence on the enrichment of PFAAs by aquatic animals. Pearson's test was performed between the concentrations of PFAAs in fish tissues and those in water, suspended solids, and sediments. In order to eliminate the interference of fat, the lipid-normalized concentrations of PFAAs in aquatic animals were used. The results are shown in Fig. 4. The concentrations of PFAAs in water and suspended solids were not significantly correlated with those in each organ, but the concentration of PFAAs in the sediment showed a significant positive correlation with those in the muscle of *A. nobilis*, as well as the kidney and gill of *C. erythropterus*. It suggested that PFAAs in water did not directly affect the enrichment of PFAAs in aquatic animals, and sediments had a significant effect on the enrichment of PFAAs in biota. This may be related to the species-specific path of intake of PFAAs.

3.3. Health risk assessment of PFOA and PFOS

The model recommended by USEPA was used to estimate the exposures of PFOA and PFOS on the residents near Lake Chaohu. In the present study, only the adult health risks of PFOA and PFOS are discussed due to a lack of toxicity data and exposure parameters for children. The exposure parameters of Anhui province were cited from the Exposure Factors Handbook of Chinese Population (Chinese Ministry of Environmental Protection, 2013). Exposure of the *C. erythropterus* and the *A. nobilis* are separately discussed (Table S5), indicating that the consumption of *C. erythropterus* led to a higher exposure to both PFOA and PFOS than that of *A. nobilis*. This implies that the health risk of eating *C. erythropterus* were higher than that of eating *A. nobilis*, due to the toxicity of PFOA and PFOS.

The health risk was discussed in terms of carcinogenic risks and non-carcinogenic risks. To consider the influence of consuming other fish tissues in addition to the muscle, edible tissues, such as the swim bladder, were taken into consideration based on their ratio of wet weight. CDIs and risks of both pollutants are shown in Table S5 and Table 2. The carcinogenic risks of PFOA, through the consumption of the muscle tissue of the fishes, were calculated (Table S5). Compared to the maximum acceptable risk recommended by USEPA (10^{-6}), the risks caused by PFOA in Lake Chaohu were far below this threshold. Consequently, at present exposure level, there is less carcinogenic risk of PFOA for adult residents of the Lake Chaohu area through the ingestion of *C. erythropterus* and *A. nobilis*.

Non-carcinogenic health risks of PFOA and PFOS to the residents of the Lake Chaohu area were assessed according to the reference doses provided by USEPA. HQ values are shown in Table 2. The non-carcinogenic risks of PFOS were higher than those of PFOA through the ingestion of *C. erythropterus and A. nobilis*. At the current exposure level, the two pollutants will pose lower non-carcinogenic risk to residents of the Lake Chaohu area through the consumption of *C. erythropterus* and *A. nobilis* considering that both HQs are less than 1. However, as both the carcinogenic and non-carcinogenic risks of *C. erythropterus* are greater than those of *A. nobilis,* people should avoid long-term consumption of large amounts of large carnivorous fish such as *C. erythropterus.*

After the risk values were calibrated by considering the tissue intake, the carcinogenic risks of PFOA were still far below the maximum acceptable risk of 10^{-6} recommended by the USEPA. Regarding the non-carcinogenic risks of PFOA and PFOS, the HQ values increased but were still less than 1. Considering that *C. erythropterus* accounts for approximately 4% of the edible fish caught from Lake Chaohu (Liu et al., 2014), PFOA and PFOS could still threat the health if people increase the consumption of *C. erythropterus.* Although the health risks of PFOA and PFOS did not exceed the reference values, it is worth noting that fish tissue intake could cause an increasing risks of up to 60%, and chronic ingestion of fish tissues, such as the swim bladder, should be avoided.

4. Conclusion

The residual levels and distributions of PFAAs in fish tissues from Lake Chaohu are shown in this study. A total of seventeen PFAAs were detected, ranging from 9.55 to 423.26 ng/g, ww, with a mean concentration of 124.53 ± 106.86 ng/g, ww. The concentrations of PFAAs found in different tissues of the *Culter erythropterus* and *A. nobilis* fluctuated. The PFAA levels in the liver of *C. erythropterus* were the highest and the lowest in muscle of *A. nobilis*: PFAAs were tended to accumulate in the liver of carnivorous fish *C. erythropterus* than omnivorous fish *A. nobilis*. Because the lipid contents of heart and spleen from the *Culter erythropterus* were lower than those of other tissues, their lipid-normalized concentrations of PFAAs were higher than their wet weight concentrations. PFOA and PFOS were posing carcinogenic and non-carcinogenic risks to humans, and the consumption of carnivorous fish and related tissues with higher trophic level, such as *Culter erythropterus*, leads to greater health risks. Although the risks did not exceed the reference values, it is worth noting that fish tissue intake could cause an increasing risks of up to 60%, and chronic ingestion of fish tissues, such as the swim bladder, should be avoided.

Acknowledgments

Funding for this study was provided by the National Natural Science Foundation of China (NSFC) (41977312, 41271462, 41030529). This work was also supported by grants from the Undergraduate Student Research Training Program of the Ministry of Education.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecoenv.2019.109666.

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