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Managing health risks of perfluoroalkyl acids in aquatic food from a river-estuary-sea environment affected by fluorochemical industry

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ABSTRACT

Substantial perfluoroalkyl acids (PFAAs) production still occurs in China, and the consumption of aquatic products is a critical exposure pathway of PFAAs in humans. In this study, specimens of 16 freshwater and 40 marine species were collected in the river-estuary-sea environment affected by a mega fluorochemical industry park in China in 2015, and the edible tissues of these organisms were analyzed for PFAA levels. Perfluorooctanoic acid (PFOA) was the dominating contaminant with an overall contribution of more than 90%, and concentrations as high as 2161 ng/g wet weight (measured in the freshwater winkle). All species with the greatest PFOA levels were benthic. The trophic magnification factor (TMF) of PFOA was 1.10 for freshwater species and 1.28 for marine species, indicating that PFOA was slightly magnifying. Analysis of carbon source indicated that freshwater species were more benthic feeding, while marine species were more pelagic feeding. Aquatic food consumption screening values of PFOA were modified according to estimated daily intake (EDI) values, which generated recommendations for limited meal categories and the do-not-eat category. Thus, this study provides recommendations for mitigating the health risks of PFAA-contaminated aquatic food, ranging from food selection to consumption frequency and proper food processing.

1. Introduction

Among per- and polyfluoroalkyl substances (PFASs), perfluoroalkyl acids (PFAAs) have a wide array of industrial applications. They consist of a fully fluorinated carbon chain (C_nF_{2n+1}-) and a charged functional group, which gives them enhanced properties, such as strong acidity and high surface activity at very low concentrations, compared to non- or partially fluorinated alkyl compounds (Wang et al., 2017). Earlier research indicated that the eight-carbon (C8) perfluoroalkyl-derived surfactants have the best activity, making perfluorooctanoic acid (PFOA)- and perfluorooctane sulfonic acid (PFOS)-related compounds among the most applied PFAAs. However, more recently, their applications have been restricted due to concerns over their persistent, bioaccumulative, and toxic properties and their presence in wildlife (Betts, 2007). The C4 compound perfluorobutane sulfonic acid (PFBS) was developed by a major producer to replace PFOS (3M, 2002). This development assisted efforts to control PFOS production and emission at a global scale (Stockholm Convention, 2009). But for PFOA, the

Abbreviations: PFASs, per- and polyfluoroalkyl substances; PFAA, perfluoroalkyl acid; PFCA, perfluoroalkyl carboxylic acid; PFSA, perfluoroalkyl sulfonic acid; PFOS, perfluorooctane sulfonic acid; PFOA, perfluorooctanoic acid; PFBA, perfluorobutanoic acid; FIP, fluorochemical industry park; ww, wet weight; TL, trophic level; TMF, trophic magnification factor; BW (bw), body weight; AA-EQS, annual average environmental quality standards; TDI, tolerable daily intake; EDI, estimated daily intake; MDHHS, Michigan Department of Health and Human Services; FCSV, fish consumption screening value; ACSV, aquatic food consumption screening values

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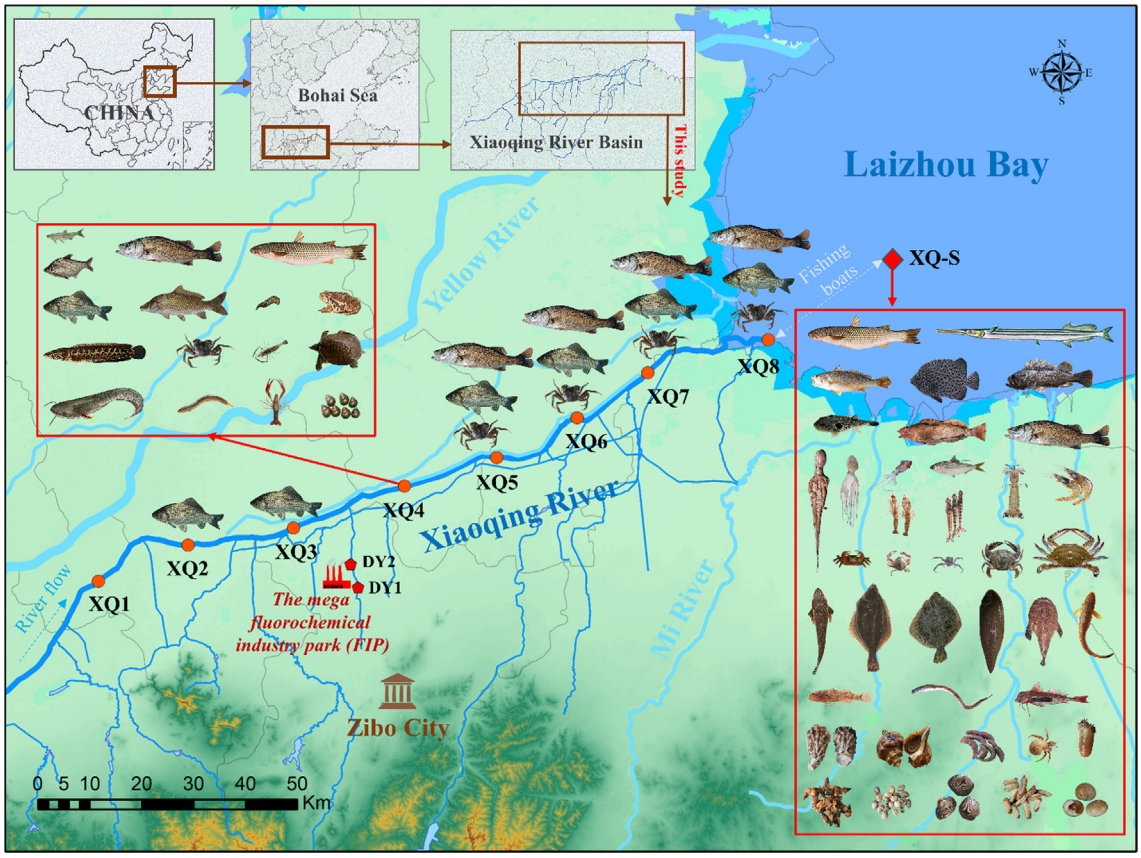

Fig. 1. Sampling sites (marked in red) in the Xiaoqing River catchment and Laizhou Bay. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

restrictions brought more of a shift in production location rather than elimination. For example, the 2010/2015 PFOA Stewardship Program claimed that the goals were met, yet only eight leading companies joined the program (USEPA, 2015); In the EU, it is estimated that the annual import of textiles could contain 1000–10,000 tons of PFOA-related substances (ECHA, 2018). Thus, the production and use of PFOA is expected to continue until effective alternatives are available to the majority of the fluorochemical industries. The occurrence of PFOA remains a major concern that requires global control, and thus, PFOA, its salts, and PFOA-related compounds were listed in Annex A of the Stockholm Convention with specific exemptions (decision SC-9/12) as of May 2019 (Stockholm Convention, 2019).

The widespread presence of PFAAs in the aquatic environment is linked to their water solubility, especially as salt or free acid (USEPA, 2017), and the use of aqueous forms in industrial applications, such as the ammonium salt of PFOA (APFO) in the production of fluoropolymers by emulsion polymerization (European Commission, 2010). Thus, aquatic organisms can accumulate PFAAs, leading to human exposure through consumption of aquatic species. Past estimates of the biomagnification factor (BMF) and trophic magnification factor (TMF) of PFAAs were inconsistent, with values ranging over several orders of magnitude from $\ll 1$ to $\gg 1$ (Franklin, 2015). Apparently, PFOS has a higher bioaccumulation potential than PFOA in aquatic biota, and PFOS displays relatively clear biomagnification along the food chain, especially for top predators like water birds and marine mammals, while there are no clear trends for PFOA among species from different trophic levels (TLs) (Houde et al., 2011; Ahrens and Bundschuh, 2014), which might be due to the faster elimination of PFOA than PFOS. Hence, because TLs or positions in a food chain cannot be used alone as guidance for categorizing aquatic food according to PFAA contamination levels, more factors should be taken into consideration following exposure under natural conditions.

Animal studies indicate that PFAA uptake can lead to many adverse outcomes, including metabolic disorders, endocrine perturbations, and immune and developmental toxicity (Krafft and Riess, 2015). Human health studies also suggest that PFAAs are associated with certain problems like adverse immune outcomes and dyslipidemia, as well as cancer for residents living in manufacturing locations (Sunderland et al., 2019). These concerns have led to stricter regulations of PFAAs for drinking water safety (USEPA, 2016). For food safety, several countries have already published tolerable daily intake (TDI) as health-based guidance values for PFAAs (EFSA, 2008; Danish EPA, 2015; FSANZ, 2017; USEPA, 2017; ATSDR, 2018). These values are critical not only for assessing the human health risk of PFAAs, but also for providing more practical suggestions to reduce the risk. In this case, the calculated TDI values may be improved by considering individual differences in dietary habits.

Despite environmental and health concerns, PFOA production is still ongoing in China. According to the China Fluorosilicon Organic Materials Industry Association, until 2015, the annual production capacity of PFOA was approximately 200 tons. A majority of the PFOA was used as processing aids to produce fluoropolymers with an annual capacity of up to 130,000 tons, with only minor amounts being exported to foreign companies. We are aware that high amounts of PFAAs are emitted from a mega fluorochemical industry park (FIP) located in the Xiaoqing River catchment of North China (Wang et al., 2014; Wang et al., 2016). Continued PFAA emissions have led to local contamination of the surface and groundwater (Liu et al., 2016), indoor and outdoor dust (Su et al., 2016), and plants and animals produced locally for human consumption (Liu et al., 2017; Su et al., 2017). The PFOA that is discharged as waste from the FIP is transported via the river to the adjacent sea (Wang et al., 2016), where an intensive fishery exists.

The objectives of this study included investigating the impact of high PFAA emission on the aquatic organisms in the river-estuary-sea system of the Xiaoqing River catchment of North China. We also aimed to analyze factors that influence the bioaccumulation of PFAAs among different aquatic species, and explore strategies to mitigate human health risks via advice on the selection and consumption of aquatic foods.

2. Materials and methods

2.1. Research design

This study examined the water transportation pathway of PFAAs, starting from the emission source (the FIP). Freshwater and marine aquatic organism samples were collected along this transport route (Fig. 1). In the minor tributary receiving waste from the FIP, limited aquatic organisms were observed. Sites DY1 and DY2 located upstream and downstream of the FIP, respectively, were selected for monitoring PFAA emission. Sites XQ1–XQ8 were distributed from upstream of the Xiaoqing River to its estuary, and two fish species, along with one crab species, were collected to evaluate the transport of PFAAs through the river-estuary-sea environment. The greatest effort in collecting the widest range of freshwater species was focused on site XQ4 located downstream of the confluence point where the tributary meets the Xiaoqing River. Site XQ-S was located in the Laizhou Bay within an intensive fishery area where the widest possible range of marine organisms was collected. Further description of these sampling sites can be found in Wang et al. (2016).

2.2. Sample collection and preparation

The sampling campaign was conducted in October 2015, after the start of the oceanic fishing season (closed from May to September). Fixed fishing nets combined with cast fishing nets were used to catch freshwater organisms, whereas local fishing boats were hired to catch marine organisms (from shore to site XQ-S). We collected common local species at their normal (adult) sizes for analysis. Non-local species that can be bought from local markets were not included. The aquatic organisms were maintained in clean water for a short period (minutes to hours, benthic species took longer time) after being caught to reduce the effect of in-situ water. All aquatic taxa were identified to species level. A few individuals of each species (depending on size and availability) were homogenized to generate the sample material before extraction. To assess the human health risk, only the edible parts were used for PFAA measurements, depending on the dietary habit (muscle, whole body without shell, or whole body, etc.) (Table S1). All aquatic samples were freeze-dried and ground for quantitative extraction. Water samples (XQ1–XQ8, XQ-S, DY1–DY2) were collected in pre-rinsed 1 L polypropylene (PP) bottles at < 20 cm depth, excluding the surface microlayer. Water parameters, including temperature, pH, dissolved oxygen, and salinity, were measured in situ using an HQd Portable and Benchtop Meter Configurator (HACH Company, USA).

2.3. PFAA measurement and QA/QC

A total of twelve linear PFAAs, including nine perfluoroalkyl carboxylic acids (PFCAs) with carbon numbers from C4 to C12 and three perfluoroalkyl sulfonic acids (PFSAs), were quantified in this study (Table S2). The extraction procedures were mostly consistent with previous studies. Briefly, for water samples, 400 mL were extracted using Oasis WAX 6 cc Vac cartridges (Waters Corp., Milford, MA) (Wang et al., 2016). For aquatic organisms, approximately 0.5 g of dry samples were extracted by ion-pair extraction, followed by cleanup with Supelclean™ ENVI-Carb™ cartridges (Sigma-Aldrich Co., St. Louis, MO) and Oasis WAX 6 cc Vac cartridges (Loi et al., 2011). Detailed information on the standards, reagents, and extraction is provided in the supplementary material. Concentrated 1 mL extracts were loaded on an Agilent 1290 Infinity HPLC System coupled to an Agilent 6460 Triple Quadrupole LC/MS System (Agilent Technologies, Palo Alto, CA) (Table S3). Quantification of PFAAs in all samples was conducted using 10-point internal quantification curves of PFAA standards with concentrations ranging from 0.01 to 1000 ng/mL and 10 ng/mL internal standards. The regression coefficients (R^2) were > 0.99 in all batches. LOD and LOQ were 0.01–0.10 ng/g and 0.04–0.50 ng/g dry weight (dw) for aquatic organisms, and 0.01–0.06 ng/L and 0.06–0.22 ng/L for water samples, respectively. Matrix spike recoveries (mean value ± standard deviation, n = 4) ranged from 94 ± 4% to 114 ± 6% for aquatic samples and from 78 ± 3% to 125 ± 6% for water samples (Table S2). Extracts with PFAA concentrations above 1000 ng/mL were adjusted in weight or volume and extracted again to ensure that the sample concentrations were within the calibration ranges. Field, transport, procedure and solvent blanks were prepared with every sample batch to monitor potential interference,

2.4. Natural stable isotope analysis

The nitrogen and carbon stable isotope ratios of the test organism tissues were analyzed using a DELTA V Advantage Isotope Ratio Mass Spectrometer coupled with a Flash 2000 HT Elemental Analyzer (Thermo Fisher Scientific, Inc., USA). The natural nitrogen and carbon stable isotope contents are denoted as δ^{15}N and δ^{13}C (‰), expressing the per mil deviation from international standards, the atmospheric ^{15}N and the ^{13}C content of Vienna Pee Dee Belemnite (VPDB), respectively (Table S8).

Results of the δ^{15}N were used to determine the trophic level (TL) of individual species using the following equation (Eq. (1)):

$$TL_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton})/3.8 \quad (1)$$

The hypothesis is that the TL of zooplankton is 2 and the enrichment factor constant is 3.8 (Loi et al., 2011). We analyzed the community composition and richness of the phytoplankton and zooplankton in the Xiaoqing River. Forty-one phytoplankton and twenty-seven zooplankton species were identified. But the richness was not sufficient for the stable isotope analysis. Instead, we used the mean value of 6.14 as a constant for $\delta^{15}N_{zooplankton}$ derived from Wan et al. (2005) that was conducted near this study area.

Then trophic magnification factor (TMF) of PFAAs was calculated using the following equations (Eqs. (2) & (3)) (Tomy et al., 2004):

$$\ln(PFAA\ \ concentration) = a + (b \times TL) \quad (2)$$

$$\mathrm{TMF} = e^{b} \quad (3)$$

where the unit of PFAA concentration was ng/g, wet weight (ww). Only PFAA concentrations in muscle of the aquatic organisms were used. The value b in Eq. (3) was the slop of Eq. (2). A TMF of > 1 implies that a chemical is biomagnifying.

Results of the δ^{13}C were normalized using the carbon-to-nitrogen ratio (C:N) in the following equation (Eq. (4)):

$$\delta^{13}C_{normalized} = \delta^{13}C_{untreated} - 3.32 + 0.99 \times C{:}N \quad (4)$$

In the study by Post et al. (2007), C:N was proved to be sufficient to normalize the untreated δ^{13}C for lipid content, which is specifically applicable for aquatic organisms.

The normalized δ^{13}C was further used to determine the carbon source of the aquatic species in the following equation (Eq. (5)):

$$\text{Carbon source} = 1 - \frac{\delta^{13}C_{zooplankton} - \delta^{13}C_{consumer} + \Delta\delta^{13}C(TP_{consumer} - TP_{zooplankton})}{\delta^{13}C_{zooplankton} - \delta^{13}C_{benthic}} \quad (5)$$

The hypothesis is that the zooplankton represent the pelagic source, $\Delta\delta^{13}$C is the trophic enrichment factor for consumers analyzed using

muscle tissues, which is set as a constant of 1.3‰ (McKinney et al., 2012). The benthic source in this study is presented using the benthic species with the lowest $\delta^{13}C_{benthic}$ values, which were *Eriocheir sinensis* (−27.1) for the freshwater species and *Zoarces slongatus* (−25.0) for the marine species. Carbon source values closer to 0 indicate more benthic feeding and values closer to 1 indicate more pelagic feeding.

2.5. Screening values of PFOA

Aquatic food consumption screening values (ACSV) of PFOA (ng/g; ww) were calculated using the following equation (Eq. (6)):

$$ACSV_{PFOA} = FCSV_{PFOS} \times R_{PFOA/PFOS} \quad (6)$$

The $FCSV_{PFOS}$ (ng/g, ww) is the fish consumption screening value (FCSV) of PFOS determined by the Michigan Department of Health and Human Services (MDHHS) (State of Michigan, 2016), which includes a series of concentration ranges of PFOS that provide guidelines for fish consumption frequency. The $R_{PFOA/PFOS}$ is the PFOA-to-PFOS ratio from published health guidelines that is a measure for co-occurrences of PFOA and PFOS in the same subject. Because the MDHHS guidelines do not provide the FCSV for PFOA, we hypothesized that the $R_{PFOA/PFOS}$ could be used as a measure for the difference between PFOA and PFOS, and thus, the $ACSV_{PFOA}$ could provide guidance for the consumption of aquatic food with PFOA contamination.

2.6. Estimated daily intake of PFOA

The estimated daily intake (EDI) of PFOA (ng/kg/day) based on the high end of the ranges for $ACSV_{PFOA}$ was further calculated with the following equation (Eq. (7)):

$$EDI_{PFOA} = ACSV_{PFOA} \times F \times R_{IR/BW} \quad (7)$$

where F is the consumption frequency (converted to a daily basis) of the aquatic food that is derived from the meal category of the $FCSV_{PFOS}$. $R_{IR/BW}$ (g/kg) is the ratio of the ingestion rate (IR) per meal (g) to the body weight (BW; kg) of consumers. There are differences in IR and BW among different populations. However, the IR and BW values provided by the MDHHS are divided into age groups and in one-to-one correspondence, which generates a constant ratio (0.10 oz/kg converted to 2.84 g/kg).

2.7. Statistical and spatial analysis.

The PFAA concentrations in aquatic organisms were converted from the dry weight basis to wet weight basis using the moisture content values that were measured during lyophilization (Table S1). Statistical analysis was performed with Microsoft Excel 2016 (Microsoft Corp., Redmond, WA), SPSS Statistics V20.0 (SPSS Inc. Quarry Bay, HK), and OriginPro 2018 (OriginLab Corp., Northampton, MA). Tests of normality on the concentrations of PFAAs were conducted to ensure that data met the assumptions used for further analysis. Spatial distributions of sampling were illustrated using the Arcmap module in ArcGIS V10.0 software (ESRI, Redland, CA) with the actual recording of the coordinates combined with other detailed geographical information (river, tributary, topography of land and sea, etc.).

3. Results and discussion

3.1. PFAAs in aquatic organisms

PFAAs were detected in all freshwater and marine aquatic organisms (Table 1 and Table S4). PFOA was the only compound with a detection ratio of 100%. All long-chain PFAAs had detection ratios over 90%, whereas the short-chain PFAAs had relatively low ratios, except for perfluorobutanoic acid (PFBA) (93%). PFOA concentrations (including mean, median, and maximum values) were several orders of magnitude higher than those of other PFAAs, indicating that PFOA was the dominating PFAA in the aquatic organisms analyzed in this study.

Table 1
Summary of PFAA concentrations (ng/g, ww) in aquatic organisms (n = 43, freshwater species; and n = 42, marine species).

Analytes	Carbon number	Concentration, ng/g, ww				Detection ratio, %
		Min	Median	Mean	Max	
PFCAs						
PFBA	4	nd	0.55	1.32	11.1	93
PFPeA	5	nd	0.11	0.21	1.20	25
PFHxA	6	nd	0.20	0.77	9.46	29
PFHpA	7	nd	0.09	0.54	6.35	55
PFOA	8	0.11	5.58	64.6	2161	100
PFNA	9	nd	0.18	0.42	5.02	91
PFDA	10	nd	0.71	1.51	12.3	91
PFUnDA	11	nd	0.69	0.86	4.64	99
PFDoDA	12	nd	0.36	0.66	3.20	91
PFSAs						
PFBS	4	nd	0.03	0.09	0.51	16
PFHxS	6	nd	0.06	0.06	0.29	20
PFOS	8	nd	1.16	2.16	9.73	98
ΣPFAAs		0.24	14.6	71. 8	2196	100

Note: nd, indicates below LOQ.

Previous studies on PFAAs in aquatic organisms affected directly by fluorochemical emission provided limited information for a comparison with our data. PFOS has received the most attention in the monitoring of contaminants in aquatic animals (Houde et al., 2011; Ahrens and Bundschuh, 2014; Taylor and Johnson, 2016; Pan et al., 2018; Fair et al., 2019). In this study, the PFOS concentrations (max: 3.55 ng/L) in the Xiaoqing River water were relatively low (Table S4), but in aquatic organisms, the maximum PFOS concentration was 9.73 ng/g (ww). This disparity is due to the high bioaccumulation potential of PFOS (Ahrens and Bundschuh, 2014). The tissues of aquatic animals had low concentrations and detection ratios of short-chain PFAAs, which were likely related to their low bioconcentration factors (BCFs) (Martin et al., 2003) and possibly to depuration during the short period when the freshly caught aquatic animals were maintained in clean water (to reduce the effect of in-situ water) (Cerveny et al., 2018; Zhong et al., 2019). In contrast, freshwater plant species affected by fluorochemical industry emissions accumulated substantially higher amounts of short-chain PFAAs than the aquatic animals analyzed in this study (Wang et al., 2019). According to an earlier study on terrestrial animals collected in the emission impact zone of the same FIP described our report (Su et al., 2017), the PFBA-to-PFOA ratio in home cultured eggs was 0.18, which was much higher than the respective ratio in the aquatic animals with the highest concentrations of ΣPFAAs in this study (4.70e−3). These results may indicate that the consumption of aquatic animals is a more important source of long-chain PFAAs in humans than that of terrestrial animals. Thus, the following sections are mostly focused on PFOA.

3.1.1. PFOA in freshwater species

Concentrations of PFOA in the crucian carp (*Carassius auratus*) were only 0.33 and 0.38 ng/g (ww) at the upstream sites XQ2 and XQ3, respectively, but 99.0 ng/g (ww) at XQ4 downstream of the FIP. Among sixteen freshwater species monitored at site XQ4, the highest PFOA concentration was found in a mollusk, the winkle (*Cipangopaludina chinensis*) (2161 ng/g, ww), followed by a fish, the loach (*Misgurnus anguillicaudatus*) (340 ng/g, ww), and a crustacean, i.e., a crayfish species (*Procambarus clarkii*) (241 ng/g, ww), which belong to three different phyla. The nine fish species showed relatively high PFOA concentrations, ranging from 7.04 to 340 ng/g (ww). The two amphibians, a toad (*Bufo raddei*) and a turtle (*Trionyx sinensis*), had the lowest PFOA concentrations of 0.42 ng/g and 0.41 ng/g (ww), respectively)

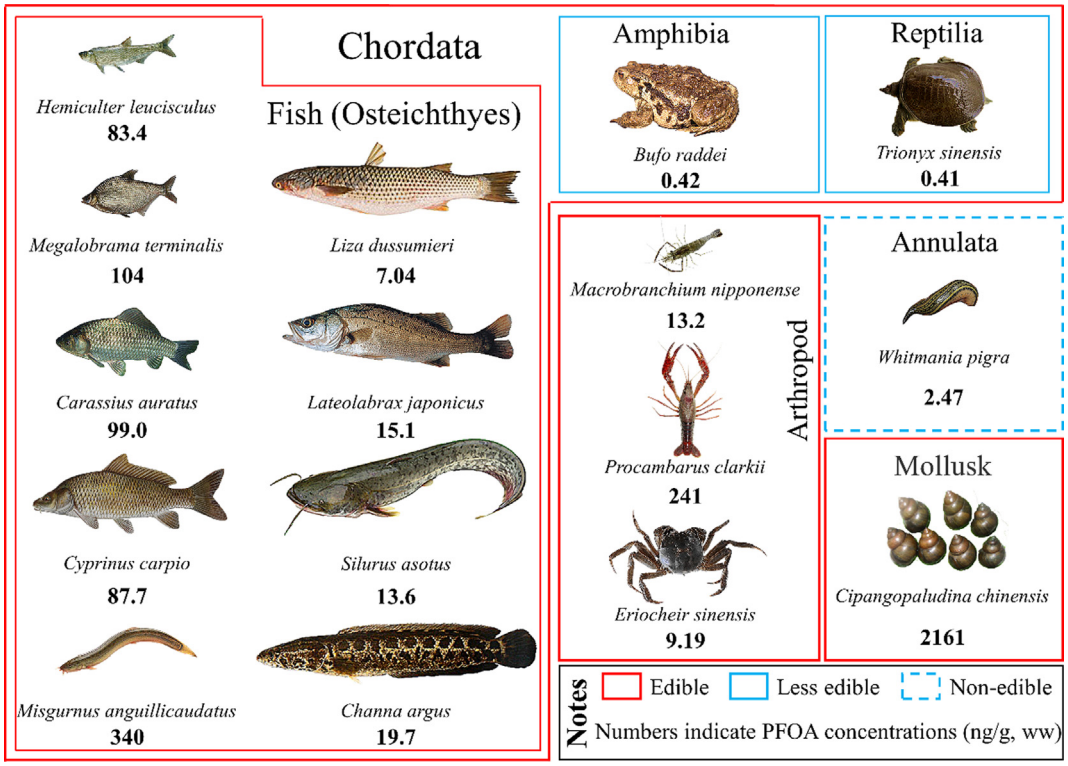

Fig. 2. PFOA concentrations (ng/g, ww) in the freshwater species at site XQ4 (the value for the river crab (*Eriocheir sinensis*) was obtained from muscle tissue of males).

(Fig. 2).

3.1.2. PFOA in marine species

Because the seawater in the Laizhou Bay dilutes the river water PFOA levels through the freshwater-saltwater interface, the forty marine aquatic species at the XQ-S site had lower PFOA concentrations than the freshwater species. The highest PFOA concentration was found in a mollusk, the fur clam (*Scapharca subcrenata*) (642 ng/g, ww), followed by a fish, the fang goby (*Odontamblyopus rubicundus*) (349 ng/g, ww), and a macroinvertebrate sea worm (*Urechis unicinctus*) (167 ng/g, ww). Similar to the findings with the freshwater species, the distribution of PFOA concentrations among the species was not associated with their phylum or type. Fourteen out of eighteen fish species had PFOA concentrations of less than 1.00 ng/g (ww) (Fig. 3).

3.2. Factors affecting the bioaccumulation of PFOA in aquatic organisms

3.2.1. PFOA levels in the aquatic environment

The emission from the FIP was the main source of PFOA in the aquatic environment of the Xiaoqing River (from sites XQ4-XQ8) (Table S5). Compared with our previous studies, the water PFOA levels remained very high in recent years, at mean values from 3100 ng/L in September 2011 (Wang et al., 2014), to 17,361 ng/L in June 2013 (Wang et al., 2016) and 40,368 ng/L in October 2015, measured at sites XQ6 to XQ8, respectively. This trend was corroborated by other studies conducted in the same area in 2014 (Heydebreck et al., 2015; Shi et al., 2015a). Moreover, a study by Chen et al. (2016) on PFASs in the Bohai Sea also suggested broad dissemination of PFOA in the entire Laizhou Bay area, indicating a continued influence of PFOA on local aquatic organisms.

3.2.2. The river-estuary-sea environment

The river-estuary-sea system involves frequent mixing of freshwater and saline water (Table S6). Three common species, including the crucian carp, river crab, and sea bass, were selected to evaluate the influence of the aquatic environment. In the section of the Xiaoqing River with heavy PFOA pollution (sites XQ4–XQ8), the crucian carp had the highest mean PFOA concentration (mean: 90.4 ng/g, ww, range: 33.0–156 ng/g, ww), followed by the river crab (muscle tissue of males, mean: 38.9 ng/g, ww, range: 3.98–150 ng/g, ww) and sea bass (mean: 10.2 ng/g, ww, range: 4.19–15.1 ng/g, ww). Fluctuations of PFOA concentrations were observed throughout the river-estuary-sea environment (Fig. 4a). However, despite the river PFOA concentration decreasing almost 10-fold from site XQ7 (78.0 μg/L) to site XQ8 (8.56 μg/L) (Table S5), PFOA concentrations in the three species collected from estuary (site XQ8) were substantially higher than those from upstream sites, especially in the river crab.

We further analyzed the pairwise correlations between PFOA concentrations in the three species and PFOA concentrations in water, along with four water parameters (temperature, pH, salinity, and dissolved oxygen) (Table S6). There were no strong correlations, except that there was a significant negative correlation ($p < 0.05$) between PFOA concentrations in sea bass and water pH (Table S7). Typically, the water pH is positively correlated with salinity and cation concentrations (the reaction of cations with dissolved CO_2 produces OH^- through hydrolysis and hence increases water pH), but the increasing cation content (e.g., Ca^{2+} and Na^+) in water could decrease the BCF of PFOA in aquatic organisms (Xia et al., 2015).

3.2.3. Sex and tissue differences in PFOA levels of crabs

There are sex-specific differences in the accumulation of PFASs within the same species (Cerveny et al., 2018), but these differences may not be comparable with those between species (Babut et al., 2017). In our study, sex was not a critical factor in most aquatic species used for human consumption, except in crabs. Specifically, we measured PFOA concentrations in the edible parts of crabs, i.e., in muscle and fat samples of male crabs and in muscle and roe samples of female crabs (Table S4, Fig S2). In river crabs, PFOA accumulation was generally higher in males than in females. Among male river crabs, PFOA concentrations were slightly higher in muscle samples (mean: 38.9 ng/g,

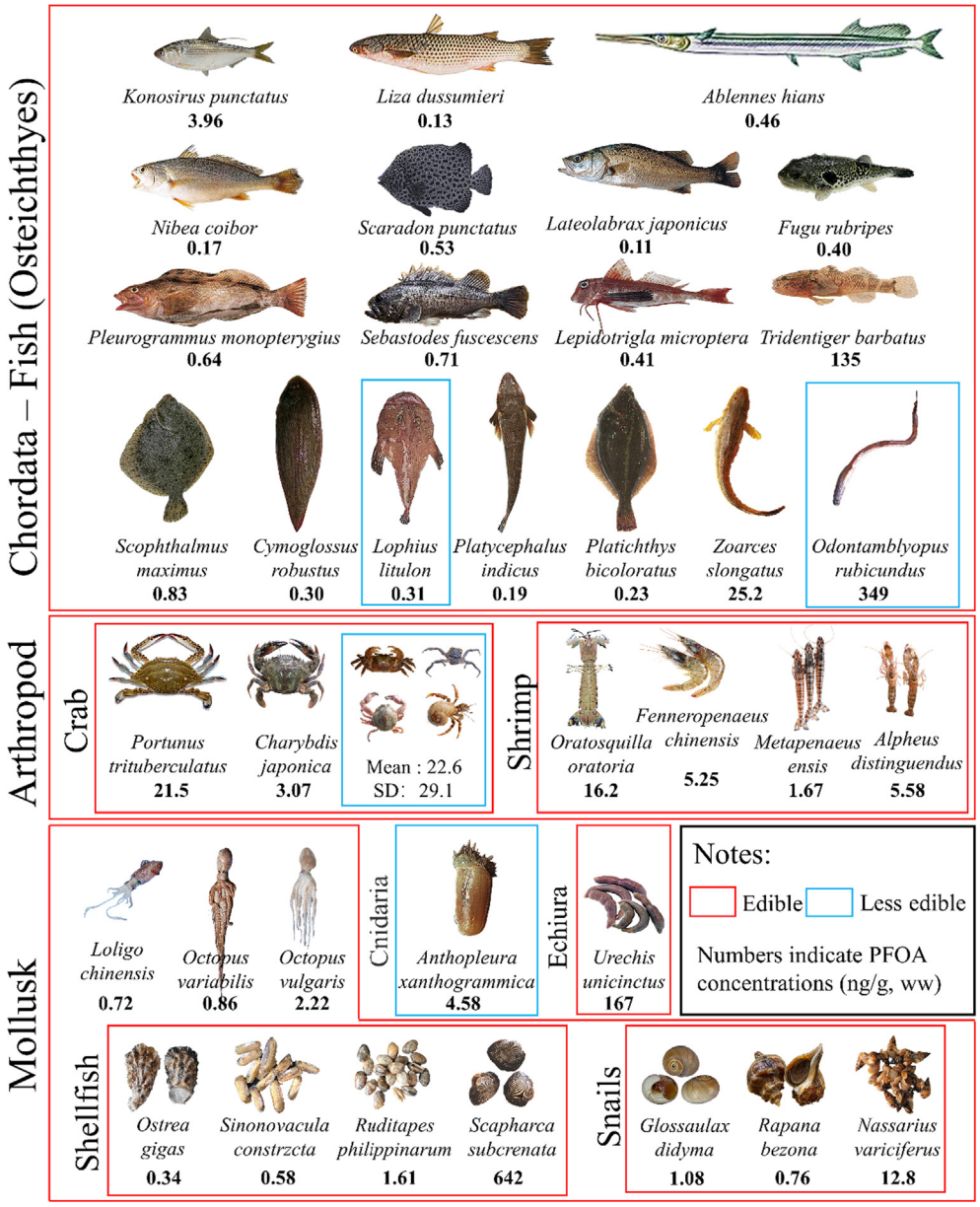

Fig. 3. PFOA concentrations (ng/g, ww) in marine species collected between the shore and site XQ-S (values for the two sea crabs (*Portunus trituberculatus* and *Charybdis japonica*) were obtained from muscle tissue of males).

ww) than in fat samples (mean: 31.5 ng/g, ww), even in crabs from site XQ8 with much higher PFOA concentrations. The same trend was also observed in male sea crabs. But the trend was the opposite in female river crabs, in which PFOA concentrations were slightly higher in roe (mean: 8.14 ng/g, ww) than in muscle samples (mean: 6.07 ng/g, ww). A portion of the PFOA burden is likely disseminated into the eggs, which requires further detailed studies.

3.2.4. Influence of TL

The TLs ranged from 2.21 to 5.57 among freshwater species (3.85 ± 0.8) and from 2.35 to 4.83 among marine species (3.45 ± 0.6) (Table S8). The most contaminated species had relatively low TLs; in freshwater species, the TL was 4.02 for winkle, 2.29 for loach, and 3.62 for crayfish; and in marine species, the TL was 2.38 for fur clam, 3.35 for fang goby, and 2.60 for a sea worm. The TMF of PFOA was 1.10 (p = 0.60) for freshwater species and 1.28 (p = 0.29) for marine species. This indicated that PFOA was biomagnifying among the tested aquatic species, but the trend was not significant, which was a limitation of using field-derived biomagnification factors and trophic magnification factors as indicators of the bioaccumulation potential (Franklin, 2015). Among the three monitored species of the river-estuary-sea environment, the sea bass had the highest TL, followed by the crucian carp and river crab. This trend was similar among the contaminated section of the Xiaoqing River (from site XQ4 to XQ6), whereas their TLs did not differ greatly at the estuary site (XQ8) (Fig. 4b). The freshwater winkle had the highest PFOA level in this study. According to its biology, the winkle grazes on algae and lives on the sediment, perhaps even inside the sediment. PFOA has a high sorption coefficient from water to particles and sediment (Ahrens et al., 2010), and high PFOA levels have been recorded in Xiaoqing River sediments (Shi et al., 2015a; Wang et al., 2016). Furthermore, Robinson et al. (1984) found that the lipids of aquatic organisms can merge with the sediment, generating sediment with an increased nutritive value that is ingested by some aquatic species. This phenomenon could explain the observation that the TL of the winkle (4.02) was comparable to that of some predator fishes.

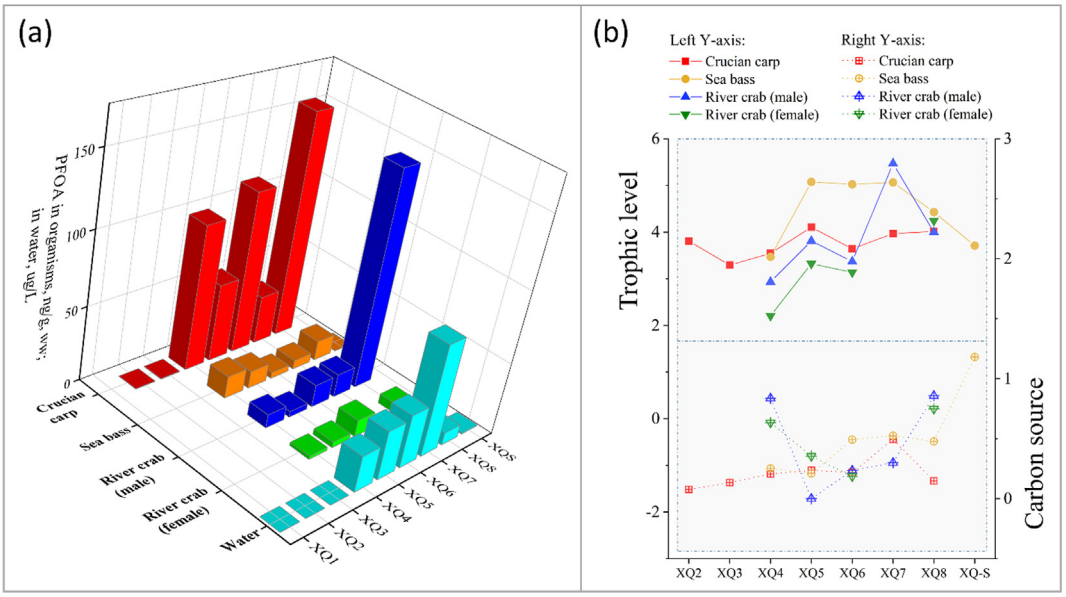

Fig. 4. (a) PFOA concentrations in the three aquatic species (ng/g, ww) of the river-estuary-sea environment and in water (ng/L); (b) The change of trophic level along with carbon source of the three species.

3.2.5. Influence of carbon source

The carbon source of the aquatic organisms is related to their food and habitat (Borgå et al., 2012). In this study, the carbon source was 0.30 ± 0.23 for freshwater species, and 1.11 ± 0.37 for marine species (Table S8). This indicated that freshwater species were more benthic feeding, while marine species were more pelagic feeding. Comparing the TL with carbon source, it seemed that benthic feeding would lead to higher TLs than pelagic feeding. The three species of the river-estuary-sea environment displayed different trends on their TLs and carbon sources (Fig. 4b). The crucian carp showed relatively consistent trends of both TL and carbon source. However, the TL and carbon source of the river crab showed opposite trends, and there were limited sex-specific difference. This might be related to the habitat of the river crab, which lives on the riverbank, ingesting carbon sources that differ from those consumed by species living in the water. The change of carbon sources in the sea bass showed a jump from sea to river, with carbon source altered from pelagic to benthic, and TL increased. The migration of this fish is associated with its physiological adaptations, such as the developmental status, fluid osmotic pressure, etc. How the changing carbon sources or adaptations can affect the bioaccumulation of PFOA in the sea bass, or how PFOA can influence the sea bass migration may require further studies.

3.3. Ecological risk evaluation of PFOA

The exposure of PFOA in both environmental medium and organisms would pose potential ecological risk to the aquatic environment. In the study by Valsecchi et al. (2017), annual average environmental quality standards (AA-EQS) for PFOA were derived. In this study, the PFOA concentrations in both freshwater (XQ4 to XQ7) and estuary (XQ8) almost all exceeded the AA-EQS for the protection of pelagic community in freshwater (30 μg/L) and sea water (3 μg/L), respectively. As mentioned above, the PFOA levels remained very high in recent years. This might be a reason why the richness of phytoplankton and zooplankton in the Xiaoqing River was poor.

The PFOA concentrations in water from sites XQ4 to XQ8 and XQ-S all largely exceeded the AA-EQS for the protection of predators (0.1 μg/L in freshwater and 0.02 μg/L in sea water, respectively). And 72% of the PFOA concentrations in aquatic organisms exceeded the AA-EQS for the protection of predators (0.9 ng/g, ww). Thus, more studies are needed to investigate the influence of PFOA contamination to the local predator species, especially avian in the estuary and wetland environment. And in such monitoring campaign, both whole fish and fillet are necessary to obtain sufficient information on ecological risk evaluation (Mazzoni et al., 2019).

3.4. Managing health risks of PFOA exposure via consumption of aquatic food

3.4.1. Assessment of consumption screening values of PFOA

Previous studies have demonstrated that aquatic food consumption is a major pathway for human exposure to PFAAs. However, the human health risk of PFOA via consumption of contaminated aquatic food depends not only on PFOA concentrations in the food but also on the dietary habits (Fair et al., 2019). In the Michigan Fish Consumption Advisory Program for PFOS by the MDHHS, the FCSV ranges were established based on the dietary habit, which includes 1 to 16 meals per month and 6 meals per year as limited meal categories, and a do-not-eat meal category (State of Michigan, 2016). The TDI values for PFOA and PFOS, as well as the PFOA-to-PFOS ratio from published health guidelines changed greatly by different countries/regions in recent years (Table 2). Especially, the European Food Safety Authority (EFSA) updated the TDI values in 2018 that were much less than those set in 2008 (EFSA, 2008; 2018), but the new EFSA's TDI is not yet put in force in EU because it is not accepted by all the EU Member States. This indicated that with more epidemiological evidences, the toxicity of PFOA and PFOS were more serious, and the toxicity of PFOA was higher than that of PFOS.

To explore risk assessment and management options of dietary frequencies for PFOA in this study, four scenarios were established, with the ACSV calculated using Eq. (6) and EDI calculated using Eq. (7). Then the percentage (P, %) of PFOA concentrations in aquatic organisms measured in this study that fall into the corresponding ACSV ranges were also calculated (Table 3).

Scenario 1: The $R_{PFOA/PFOS}$ is equal to 8 based on the TDI values for PFOA and PFOS set by the FSANZ (2017). Results were presented as $ACSV_1$, EDI_1 and P_1.

Scenario 2: The $R_{PFOA/PFOS}$ is equal to 1 based on the oral non-cancer reference doses (RfDs) for PFOA and PFOS set by the United States Environmental Protection Agency (USEPA) (USEPA, 2017). Results were presented as $ACSV_2$, EDI_2 and P_2.

Scenario 3: The $R_{PFOA/PFOS}$ is equal to 0.44 based on the TDI values

Table 2
Calculation of the PFOA/PFOS ration based on the health-based guideline values.

Regulatory Agency	Year	PFOA	PFOS	PFOA/PFOS	Reference
Danish Environmental Protection Agency (Danish EPA)	2015	TDI = 100 ng/kg·bw/day	TDI = 30 ng/kg·bw/day	3.3	Danish EPA (2015)
Agency for Toxic Substances and Disease Registry (ASTDR)	2015	MRL[a] = 30 ng/kg·bw/day	MRL = 20 ng/kg·bw/day	1.5	ATSDR (2018)
United States Environmental Protection Agency (USEPA)	2016	RfD[b] = 20 ng/kg·bw/day	RfD = 20 ng/kg·bw/day	1	USEPA (2017)
Food Standards Australia and New Zealand (FSANZ)	2017	TDI = 160 ng/kg·bw/day	TDI = 20 ng/kg·bw/day	8	FSANZ (2017)
European Food Safety Authority (EFSA)	2008	TDI = 1500 ng/kg·bw/day	TDI = 150 ng/kg·bw/day	10	EFSA (2008)
European Food Safety Authority (EFSA)	2018	TDI = 0.8 ng/kg·bw/day	TDI = 1.8 ng/kg·bw/day	0.44	EFSA (2018)

[a] MRL: provisional minimal risk level.
[b] RfD: oral non-cancer reference dose.

for PFOA and PFOS set by the EFSA (EFSA, 2018). Results were presented as $ACSV_3$, EDI_3 and P_3.

Scenario 4: The EDI was set as a constant (0.8 ng/kg·bw/day). Results were presented as $ACSV_4$ and P_4.

The calculated EDI values were well within the corresponding TDI values in each scenario for scenario 1 to scenario 2, but exceeded the corresponding TDI values in scenario 3. And scenario 4 was designed to adjust scenario 3. From scenario 1 to scenario 4, the human health risk increased via the consumption of the aquatic food with top PFOA concentrations (Fig. 5). More aquatic organisms fell into stricter meal categories. Especially the do-not eat category, from zero in scenario 1, 4.71% in scenario 2, 10.6% in scenario 3, to 24.7% scenario 4. These results brought critical challenges for human health risk assessment and management of PFOA via consumption of aquatic food with high PFOA residue. Stricter guidelines are better for the protection of human health, but might be difficult for sufficient management by local governments. The governments need to choose proper guidelines to protect the health of local residents, and work with the manufacturers to reduce the emission of PFOA. For aquatic food consumers, taking proper options can also mitigate the health risk.

3.4.2. Health risk evaluation of PFAAs

The aquatic organisms marked as 'edible' in Fig. 2 and Fig. 3 are routinely consumed, and the species with top PFOA levels require special attention focused on the dietary habit to mitigate the health risk. The analysis of factors affecting the bioaccumulation of PFOA in aquatic organisms suggested that certain benthic species accumulate higher amounts of PFOA than other species. But if these species appear on the list of food preferences, the ACSV of PFOA can provide detailed suggestions on meal frequency.

It should be noted that the ACSV results are more suitable to describe the limits for a single food source. Thus, even if these values might be within the TDI guidelines, adding other food sources with substantial PFOA exposure might lead to PFOA overconsumption, leading to increased health risks. Considering that other PFAAs examined in this study had much lower levels than PFOA, the ACSV of PFOA could be considered a summary of the 12 PFAAs for health suggestions. However, other PFASs, such as hexafluoropropylene oxide dimer acid (HFPO-DA) and chlorinated polyfluorinated ether sulfonate (F53B), were also detected at substantial levels in the same study area (Heydebreck et al., 2015; Shi et al., 2015b). Although the information on the health effects of these substances is still limited, there are rising concerns and including them in future risk assessments would improve the ASCV results. Furthermore, the trend to lower the values in health guidelines indicates an increasing need for protecting human health from PFAA pollution.

3.4.3. Treatment of aquatic food with PFAA pollution

There are additional measures for mitigating the health risk of PFAAs, including guidelines for the proper treatment of contaminated aquatic food, which we summarized from previous studies. Maintaining contaminated aquatic organisms alive in clean water for a short period before cooking would decrease the PFOA pollution to some extent; especially, it would clean the contaminated in-situ water as it did in this study. An extended cultivation period would benefit from the detoxification processes. In a study on the fate of PFASs in the food web by Cerveny et al. (2018), the authors observed a decreasing of the PFOA concentration in the liver of brown trout after living for six months in water with similar PFOA levels. Moreover, treatment of edible tissues of aquatic products with uncontaminated water could lead to the

Table 3
ASCV (ng/g, ww) and EDI (ng/kg·bw/d) adjusted for PFOA.

Meal Category	*F*	Scenario 1			Scenario 2			Scenario 3			Scenario 4	
meals/month	meals/day	$ACSV_1$ ng/g	EDI_1 ng/kg·bw/d	P_1 %	$ACSV_2$ ng/g	EDI_2 ng/kg·bw/d	P_2 %	$ACSV_3$ ng/g	EDI_3 ng/kg·bw/d	P_3 %	$ASCV_4$ ng/g	P_4 %
16	0.533	72	109	82.4	9	14	56.5	4	6	43.5	0.53	18.8
12	0.400	104	118	3.53	13	15	7.06	6	7	7.06	0.70	2.35
8	0.267	152	115	5.88	19	14	11.8	8	6	4.71	1.06	7.06
4	0.133	304	115	3.53	38	14	4.71	17	6	20.0	2.11	4.71
2	0.067	600	114	2.35	75	14	2.35	33	6	4.71	4.23	11.8
1	0.033	1200	114	1.18	150	14	8.24	67	6	2.35	8.45	10.6
6 meals/year	0.016	2400	112	1.18	300	14	4.71	133	6	7.06	17.1	20.0
Do Not Eat	0	> 2400		0	> 300		4.71	> 133		10.6	> 17.1	24.7

Note: The meal categories match the ranges provided by the MDHHS (State of Michigan, 2016); *F* is the consumption frequency converted from the meal category; $ACSV_1$, $ACSV_2$, $ACSV_3$ and $ACSV_4$ represent the upper limit of the concentration ranges; *P* is the percentage of PFOA concentrations in aquatic organisms measured in this study that fall into the corresponding ACSV ranges.

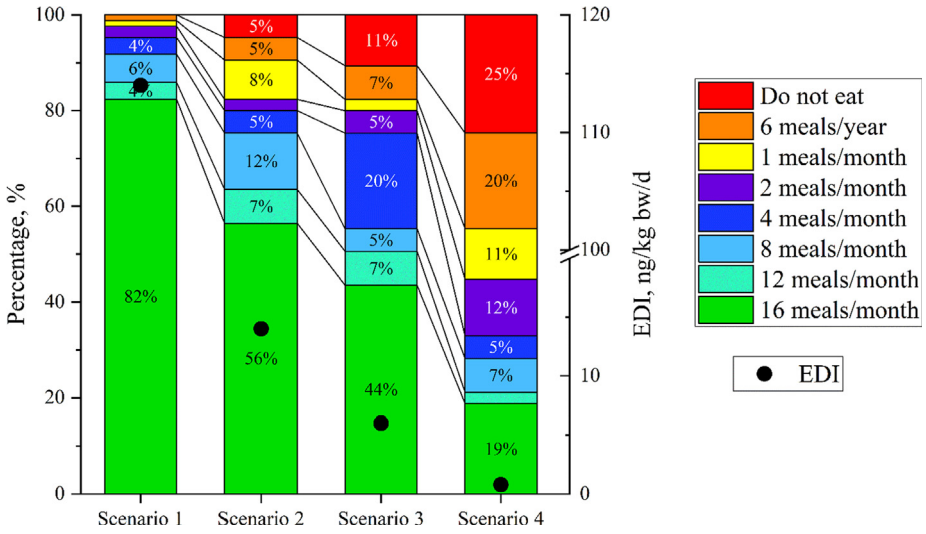

Fig. 5. Percentage (%) of PFOA concentrations (ng/g, ww) in the aquatic organisms measured in this study that fall into the corresponding ACSV ranges from the four scenarios.

depuration of PFOA, using treatment periods of hours or days depending on the species (Taylor et al., 2017).

Selecting the body parts of the contaminated aquatic animals for consumption is also important. In a study by Fair et al. (2019), PFASs concentrations were two to three times higher in the whole fish than in the fillets, indicating that cooking the whole fish (or entire body parts) would result in more PFOA consumption than cooking the fillet (muscle). However, cooking itself cannot consistently reduce contamination by PFASs. Although the concentrations might change, it is likely due to the treatment, while the amounts of PFASs usually remain unchanged during cooking (Bhavsar et al., 2014; Vassiliadou et al., 2015; Taylor et al., 2019).

4. Conclusions

In this study, 16 freshwater species and 40 marine species affected by industrial fluorochemical waste were investigated for exposure to 12 PFAAs. PFOA was dominant with an overall contribution of > 90%. Freshwater species had generally higher PFOA concentrations than marine species related to higher PFOA concentrations in the river water. The winkle (*Cipangopaludina chinensis*) displayed the highest PFOA concentration (2161 ng/g, ww) among freshwater species, whereas the fur clam (*Scapharca subcrenata*) had the highest PFOA concentration (642 ng/g, ww) among marine species. The species with top PFOA levels were all benthic. The TMF of PFOA was 1.10 for freshwater species and 1.28 for marine species, indicating that PFOA was slightly biomagnifying among the tested aquatic species. The carbon source was 0.30 ± 0.23 for freshwater species, and 1.11 ± 0.37 for marine species, indicating that the freshwater species were more benthic feeding, while the marine species were more pelagic feeding. Comparing the TL with carbon source, it seemed that benthic feeding would lead to higher TLs than pelagic feeding. PFOA concentrations in water showed high ecological risk to pelagic community, while PFOA concentrations in both water and aquatic organism showed very high ecological risk to air-breathing predators. We established the PFOA ACSVs and compared the derived EDI values with TDI guidelines for providing practical suggestions on the consumption frequency of PFOA-contaminated aquatic food based on the 1-to-16 meals-per-month and 6 meals-per-year categories, as well as the do-not-eat category. We also assessed studies on detoxification and depuration of aquatic organisms. Thus, this study provides a comprehensive evaluation of how to mitigate the health risk of PFOA in aquatic food, from the food selection and consumption frequency to proper treatment before cooking. However, by analyzing so many species simultaneously, details on individual species were limited in this study, and further interdisciplinary research is needed. Specifically, the biology of benthic species should be considered for assessing the large differences in PFOA exposure levels among these species. Moreover, the effect of the river-sea habitat exchange on PFOA bioaccumulation needs to be further examined in the migrating species, and ecological risk of PFOA to pelagic community and predators should be highly concerned.

CRediT authorship contribution statement

Pei Wang: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Visualization. **Yonglong Lu:** Resources, Supervision, Project administration, Funding acquisition, Writing - review & editing. **Hongqiao Su:** Investigation. **Chao Su:** Investigation, Formal analysis. **Andrew C. Johnson:** Writing - review & editing. **Longfei Yu:** Formal analysis, Writing - review & editing. **Alan Jenkins:** Writing - review & editing.

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Declaration of Competing Interest

The authors declare that there are no conflicts of interest.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2020.105621.

References

3M, 2002. Environmental, Health, Safety, and Regulatory (EHSR). Profile of Perfluorobutane Sulfonate (PFBS). http://multimedia.m.com/mws/media/1720O/ehsr-profile-of-perfluorobutane-sulfonate-pfbs.pdf. (December 22, 2018).
Ahrens, L., Bundschuh, M., 2014. Fate and effects of poly- and perfluoroalkyl substances

in the aquatic environment: a review. Environ. Toxicol. Chem. 33 (9), 1921–1929.
Ahrens, L., Taniyasu, S., Yeung, L.W.Y., Yamashita, N., Lam, P.K.S., Ebinghaus, R., 2010. Distribution of polyfluoroalkyl compounds in water, suspended particulate matter and sediment from Tokyo Bay, Japan. Chemosphere 79 (3), 266–272.
ATSDR, 2018. Toxicological Profile for Perfluoroalkyls, Draft for Public Comment. https://www.atsdr.cdc.gov/toxprofiles/tp200.pdf. (December 22, 2018).
Babut, M., Labadie, P., Simonnet-Laprade, C., Munoz, G., Roger, M.-C., Ferrari, B.J.D., Budzinski, H., Sivade, E., 2017. Per- and poly-fluoroalkyl compounds in freshwater fish from the Rhône River: influence of fish size, diet, prey contamination and biotransformation. Sci. Total Environ. 605–606, 38–47.
Betts, K.S., 2007. Perfluoroalkyl acids: what is the evidence telling us? Environ Health Perspect. 115 (5), A250–A256.
Bhavsar, S.P., Zhang, X., Guo, R., Braekevelt, E., Petro, S., Gandhi, N., Reiner, E.J., Lee, H., Bronson, R., Tittlemier, S.A., 2014. Cooking fish is not effective in reducing exposure to perfluoroalkyl and polyfluoroalkyl substances. Environ. Int. 66, 107–114.
Borgå, K., Kidd, K.A., Muir, D.C., Berglund, O., Conder, J.M., Gobas, F.A., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: Considerations of ecology, ecosystems, and study design. Integr. Environ. Assess. Manage. 8 (1), 64–84.
Cerveny, D., Grabic, R., Fedorova, G., Grabicova, K., Turek, J., Zlabek, V., Randak, T., 2018. Fate of perfluoroalkyl substances within a small stream food web affected by sewage effluent. Water Res. 134, 226–233.
Chen, H., Sun, R., Zhang, C., Han, J., Wang, X., Han, G., He, X., 2016. Occurrence, spatial and temporal distributions of perfluoroalkyl substances in wastewater, seawater and sediment from Bohai Sea. China. Environmental Pollution. 219, 389–398.
Danish EPA, 2015. Perfluoroalkylated substances: PFOA, PFOS and PFOSA. Evaluation of health hazards and proposal of a health-based quality criterion for drinking water, soil and ground water. https://www2.mst.dk/Udgiv/publications/2015/04/978-87-93283-01-5.pdf (January 12, 2020).
ECHA, 2018. Final background document: PFOA salts and PFOA-related substances Final background document: PFOA salts and PFOA-related substances. https://echa.europa.eu/documents/10162/13641/rest_pfoa_final_bd_en.pdf/61e81035-e0c5-44f5-94c5-2f53554255a8. (May 5, 2019).
EFSA, 2008. Perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA) and their salts Scientific Opinion of the Panel on Contaminants in the Food chain. https://www.efsa.europa.eu/en/efsajournal/pub/653. (January 10, 2019).
EFSA, 2018. Risk to human health related to the presence of perfluorooctane sulfonic acid and perfluorooctanoic acid in food. EFSA J. 16 (12), e05194.
European Commission, 2010. Analysis of the risks arising from the industrial use of Perfuorooctanoic acid (PFOA) and Ammonium Perfluorooctanoate (APFO) and from their use in consumer articles. Evaluation of the risk reduction measures for potential restrictions on the manufacture, placing on the market and use of PFOA and APFO. http://ec.europa.eu/enterprise/sectors/chemicals/files/docs_studies/final_report_pfoa_pfos_en.pdf. (March 19, 2015).
Fair, P.A., Wolf, B., White, N.D., Arnott, S.A., Kannan, K., Karthikraj, R., Vena, J.E., 2019. Perfluoroalkyl substances (PFASs) in edible fish species from Charleston Harbor and tributaries, South Carolina, United States: Exposure and risk assessment. Environ. Res. 171, 266–277.
Franklin, J., 2015. How reliable are field-derived biomagnification factors and trophic magnification factors as indicators of bioaccumulation potential? Conclusions from a case study on per- and polyfluoroalkyl substances. Integr. Environ. Assess. Manage. 12 (1), 6–20.
FSANZ, 2017. Hazard assessment report – Perfluorooctane sulfonate (PFOS), Perfluorooctanoic acid (PFOA), Perfluorohexane sulfonate (PFHxS). https://www1.health.gov.au/internet/main/publishing.nsf/Content/2200FE086D480353CA2580C900817CDC/$File/6.sd1-Hazard-assessment-report.pdf. (January 10, 2019).
Heydebreck, F., Tang, J., Xie, Z., Ebinghaus, R., 2015. Alternative and Legacy Perfluoroalkyl Substances: Differences between European and Chinese River/Estuary Systems. Environ. Sci. Technol. 49 (14), 8386–8395.
Houde, M., De Silva, A.O., Muir, D.C.G., Letcher, R.J., 2011. Monitoring of perfluorinated compounds in aquatic biota: an updated review. Environ. Sci. Technol. 45 (19), 7962–7973.
Krafft, M.P., Riess, J.G., 2015. Per- and polyfluorinated substances (PFASs): Environmental challenges. Curr. Opin. Colloid Interface Sci. 20 (3), 192–212.
Liu, Z., Lu, Y., Shi, Y., Wang, P., Jones, K., Sweetman, A.J., Johnson, A.C., Zhang, M., Zhou, Y., Lu, X., Su, C., Sarvajayakesavaluc, S., Khan, K., 2017. Crop bioaccumulation and human exposure of perfluoroalkyl acids through multi-media transport from a mega fluorochemical industrial park, China. Environ. Int. 106, 37–47.
Liu, Z., Lu, Y., Wang, T., Wang, P., Li, Q., Johnson, A.C., Sarvajayakesavalu, S., Sweetman, A.J., 2016. Risk assessment and source identification of perfluoroalkyl acids in surface and ground water: Spatial distribution around a mega-fluorochemical industrial park, China. Environ. Int. 91, 69–77.
Loi, E.I.H., Yeung, L.W.Y., Taniyasu, S., Lam, P.K.S., Kannan, K., Yamashita, N., 2011. Trophic magnification of poly- and perfluorinated compounds in a subtropical food web. Environ. Sci. Technol. 45 (13), 5506–5513.
Martin, J.W., Mabury, S.A., Solomon, K.R., Muir, D.C.G., 2003. Bioconcentration and tissue distribution of perfluorinated acids in rainbow trout (Oncorhynchus mykiss). Environ. Toxicol. Chem. 22 (1), 196–204.
Mazzoni, M., Buffo, A., Cappelli, F., Pascariello, S., Polesello, S., Valsecchi, S., Volta, P., Bettinetti, R., 2019. Perfluoroalkyl acids in fish of Italian deep lakes: Environmental and human risk assessment. Sci. Total Environ. 653, 351–358.
McKinney, M.A., McMeans, B.C., Tomy, G.T., Rosenberg, B., Ferguson, S.H., Morris, A., Muir, D.C.G., Fisk, A.T., 2012. Trophic Transfer of Contaminants in a Changing Arctic Marine Food Web: Cumberland Sound, Nunavut Canada. Environ. Sci. Technol. 46 (18), 9914–9922.
Pan, C.-G., Yu, K.-F., Wang, Y.-H., Zhang, R.-J., Huang, X.-Y., Wei, C.-S., Wang, W.-Q., Zeng, W.-B., Qin, Z.-J., 2018. Species-specific profiles and risk assessment of perfluoroalkyl substances in coral reef fishes from the South China Sea. Chemosphere 191, 450–457.
Post, D.M., Layman, C.A., Arrington, D.A., Takimoto, G., Quattrochi, J., Montaña, C.G., 2007. Getting to the Fat of the Matter: Models, Methods and Assumptions for Dealing with Lipids in Stable Isotope Analyses. Oecologia 152 (1), 179–189.
Robinson, N., Cranwell, P.A., Finlay, B.J., Eglinton, G., 1984. Lipids of aquatic organisms as potential contributors to lacustrine sediments. Org Geochem. 6, 143–152.
Shi, Y., Vestergren, R., Xu, L., Song, X., Niu, X., Zhang, C., Cai, Y., 2015a. Characterizing direct emissions of perfluoroalkyl substances from ongoing fluoropolymer production sources: a spatial trend study of Xiaoqing River, China. Environ. Pollut. 206, 104–112.
Shi, Y., Vestergren, R., Zhou, Z., Song, X., Xu, L., Liang, Y., Cai, Y., 2015b. Tissue Distribution and Whole Body Burden of the Chlorinated Polyfluoroalkyl Ether Sulfonic Acid F-53B in Crucian Carp (Carassius carassius): Evidence for a Highly Bioaccumulative Contaminant of Emerging Concern. Environ. Sci. Technol. 49 (24), 14156–14165.
State of Michigan, 2016. Michigan Fish Consumption Advisory Program: Guidance document. Michigan Department of Health and Human Services. https://www.michigan.gov/documents/mdch/MFCAP_Guidance_Document_500546_7.pdf. (May 5, 2019).
Stockholm Convention, 2009. At COP-4 in 2009, the Conference of the Parties decided to list perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOSF) in Annex B to the Stockholm Convention (decision SC-4/17). http://chm.pops.int/Implementation/IndustrialPOPs/PFOS/Overview/tabid/5221/Default.aspx. (May 5, 2019).
Stockholm Convention, 2019. Chemicals proposed for listing under the Convention. http://chm.pops.int/TheConvention/ThePOPs/ChemicalsProposedforListing/tabid/2510/Default.aspx. (January 10, 2019).
Su, H., Lu, Y., Wang, P., Shi, Y., Li, Q., Zhou, Y., Johnson, A.C., 2016. Perfluoroalkyl acids (PFAAs) in indoor and outdoor dusts around a mega fluorochemical industrial park in China: Implications for human exposure. Environ. Int. 94, 667–673.
Su, H., Shi, Y., Lu, Y., Wang, P., Zhang, M., Sweetman, A., Jones, K., Johnson, A., 2017. Home produced eggs: An important pathway of human exposure to perfluorobutanoic acid (PFBA) and perfluorooctanoic acid (PFOA) around a fluorochemical industrial park in China. Environ. Int. 101, 1–6.
Sunderland, E.M., Hu, X.C., Dassuncao, C., Tokranov, A.K., Wagner, C.C., Allen, J.G., 2019. A review of the pathways of human exposure to poly- and perfluoroalkyl substances (PFASs) and present understanding of health effects. J. Eposure Sci. Environ. Epidemiol. 29 (2), 131–147.
Taylor, M.D., Bowles, K.C., Johnson, D.D., Moltschaniwskyj, N.A., 2017. Depuration of perfluoroalkyl substances from the edible tissues of wild-caught invertebrate species. Sci. Total Environ. 581–582, 258–267.
Taylor, M.D., Johnson, D.D., 2016. Preliminary investigation of perfluoroalkyl substances in exploited fishes of two contaminated estuaries. Mar. Pollut. Bull. 111 (1–2), 509–513.
Taylor, M.D., Nilsson, S., Bräunig, J., Bowles, K.C., Cole, V., Moltschaniwskyj, N.A., Mueller, J.F., 2019. Do conventional cooking methods alter concentrations of per- and polyfluoroalkyl substances (PFASs) in seafood? Food Chem. Toxicol. 127, 280–287.
Tomy, G.T., Budakowski, W., Halldorson, T., Helm, P.A., Stern, G.A., Friesen, K., Pepper, K., Tittlemier, S.A., Fisk, A.T., 2004. Fluorinated Organic Compounds in an Eastern Arctic Marine Food Web. Environ. Sci. Technol. 38 (24), 6475–6481.
USEPA, 2015. EPA's Non-CBI Summary Tables for 2015 Company Progress Reports (Final Progress Reports). https://www.epa.gov/assessing-and-managing-chemicals-under-tsca/epas-non-cbi-summary-tables-2015-company-progress. (May 5, 2019).
USEPA, 2016. PFOA & PFOS Drinking Water Health Advisories. https://www.epa.gov/ground-water-and-drinking-water/supporting-documents-drinking-water-health-advisories-pfoa-and-pfos. (January 10, 2019).
USEPA, 2017. Technical Fact Sheet – Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoic Acid (PFOA). https://www.epa.gov/sites/production/files/2017-12/documents/ffrrofactsheet_contaminants_pfos_pfoa_11-20-17_508_0.pdf. (January 10, 2019).
Valsecchi, S., Conti, D., Crebelli, R., Polesello, S., Rusconi, M., Mazzoni, M., Preziosi, E., Carere, M., Lucentini, L., Ferretti, E., Balzamo, S., Simeone, M.G., Aste, F., 2017. Deriving environmental quality standards for perfluorooctanoic acid (PFOA) and related short chain perfluorinated alkyl acids. J. Hazard. Mater. 323, 84–98.
Vassiliadou, I., Costopoulou, D., Kalogeropoulos, N., Karavoltsos, S., Sakellari, A., Zafeiraki, E., Dassenakis, M., Leondiadis, L., 2015. Levels of perfluorinated compounds in raw and cooked Mediterranean finfish and shellfish. Chemosphere 127, 117–126.
Wan, Y., Hu, J., An, L., An, W., Yang, M., Tao, S., 2005. Analysis of nutrient levels of main species of Bohai Bay food network using stable nitrogen and carbon isotope (In Chinese). Chin. Sci. Bull. 50 (7), 708–712.
Wang, P., Lu, Y., Wang, T., Fu, Y., Zhu, Z., Liu, S., Xie, S., Xiao, Y., Giesy, J.P., 2014. Occurrence and transport of 17 perfluoroalkyl acids in 12 coastal rivers in south Bohai coastal region of China with concentrated fluoropolymer facilities. Environ. Pollut. 190, 115–122.
Wang, P., Lu, Y., Wang, T., Meng, J., Li, Q., Zhu, Z., Sun, Y., Wang, R., Giesy, J.P., 2016. Shifts in production of perfluoroalkyl acids affect emissions and concentrations in the environment of the Xiaoqing River Basin, China. J. Hazard. Mater. 307, 55–63.
Wang, P., Zhang, M., Lu, Y., Meng, J., Li, Q., Lu, X., 2019. Removal of perfluoalkyl acids (PFAAs) through fluorochemical industrial and domestic wastewater treatment plants and bioaccumulation in aquatic plants in river and artificial wetland. Environ. Int. 129, 76–85.
Wang, Z., DeWitt, J.C., Higgins, C.P., Cousins, I.T., 2017. A Never-Ending Story of Per- and Polyfluoroalkyl Substances (PFASs)? Environ. Sci. Technol. 51 (5), 2508–2518.
Xia, X., Rabearisoa, A.H., Dai, Z., Jiang, X., Zhao, P., Wang, H., 2015. Inhibition effect of Na+ and Ca2+ on the bioaccumulation of perfluoroalkyl substances by Daphnia magna in the presence of protein. Environ. Toxicol. Chem. 34 (2), 429–436.
Zhong, W., Zhang, L., Cui, Y., Chen, M., Zhu, L., 2019. Probing mechanisms for bioaccumulation of perfluoroalkyl acids in carp (Cyprinus carpio): Impacts of protein binding affinities and elimination pathways. Sci. Total Environ. 647, 992–999.