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Comparison of the health risks associated with exposure to toxic metals and metalloids following consumption of freshwater catches in China

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RESEARCH ARTICLE

Abstract

Crayfish, carp, and crab threaten the consumer's health due to the enrichment of the toxic elements. The concentration of the toxic elements (As, Cr, Cd, and Pb) in these aquatics was less investigated simultaneously *in situ*. In this study, the toxic elements in these aquatics from the same site of Hanchuan, Hubei Province, China were measured by ICP-MS to assess the enrichment of the four toxic elements in them and the health risk to the consumer. The results showed that the concentrations of these elements in muscle were lower than those in hepatopancreas/liver, except for Cr in crayfish. Furthermore, the concentrations of Cd in the hepatopancreas of crayfish and crab exceeded the standards of China and the European Union. Accordingly, the hepatopancreas of crayfish and crab are not recommended for consumption. The estimated daily intake of these elements in the muscle of crayfish, carp, and crab did not exceed the provisional tolerable daily intakes, the corresponding target hazard quotient (THQ), and the cancer risk (CR) values for As were lower than 1 and 1×10^{-6}, respectively. However, the accumulated consumption of the muscle of the three freshwater catches could result in a significant increase in the total THQ value. The total ingestion of the muscle of these catches should be limited. Collectively, these findings may renew interest in food safety and the health risk to humans owing to the consumption of freshwater catches.

Keywords: Bighead carp (*Hypophthalmichthys nobilis*); Chinese mitten crab (*Eriocheir sinensis*); Red swamp crayfish (*Procambarus clarkii*); Health risk assessment; Toxic elements

Introduction

The global consumption of aquatic animals, such as fish, crayfish, shrimp, and crab, has remained high as these animals contain beneficial proteins, polyunsaturated fatty acids, and minerals (Jiang *et al*., 2014). The production of freshwater catches has increased on a yearly basis with increasing consumption. In China, up to 2,089,600 tons of crayfish (*Procambarus clarkii)* were produced in 2019 owing to the increased area of rice-crayfish co-culture (National Fisheries Technology Extension Center and China Society of Fisheries, 2020). Bighead carp (*Hypophthalmichthys nobilis*) is one of the most important commercial aquaculture fish in China owing to its high production, fast growth, and low price (Hui *et al*., 2012). In 2019, the production of bighead carp reached 3,101,637 metric tons in China. Crab, including Chinese mitten crab (*Eriocheir sinensis*), is another important aquatic product. In fact, the demand for this product has sharply increased due to its delicious taste and high nutritional value (Jiang *et al.*, 2014). The production of Chinese mitten crab continues to increase, with 778,682 metric tons produced in 2019 in China (Ministry of Agriculture and Rural Affairs of the People's Republic of China *et al.*, 2020). Although aquatic organisms have many benefits to human health, their consumption could result in health risk to humans as they contain toxic elements, such as heavy metals and metalloid (Varol *et al.*, 2017).

Nowadays, the accumulation of toxic elements in crayfish, fish, and crab has attracted increasing attention. In fact, few researchers sought to examine and assess the bioaccumulation of toxic elements, such as arsenic (As), chromium (Cr), cadmium (Cd), and lead (Pb), in these aquatic animals (Fernández-Trujillo *et al.*, 2021; Ramon *et al.*, 2021; Solgi and Mirmohammadvali, 2021; Yang *et al.*, 2021). In general, the bioconcentration of toxic elements in aquatic animals is associated with the environment and food chain (Köker *et al.*, 2021). Previously, fish was reported to absorb toxic elements by ingesting particulate matter suspended in the water (Varol *et al.*, 2017). However, crayfish and crabs are benthic organisms that mainly feed on benthic food in sediments, and can accumulate toxic elements from the sediments. Ultimately, the toxic elements from water and/or the food chain can be absorbed and accumulated in the tissues of gills, hepatopancreas, and the edible part of the muscle of these aquatic animals (Nagarajan *et al.*, 2019).

Due to the characteristics of bioaccumulation and biomagnification of the toxic elements, exposure to the toxic elements could serve as a health risk to humans (Samuel *et al.*, 2021). Hexavalent chromium (Cr^{6+}) was reported to display heavy toxicity, and chronic exposure to this compound can cause skin cancer (Iwegbue *et al.*, 2015). As a carcinogen, Cr^{6+} can attack the respiratory system, kidneys, and other vital organs (Varol *et al.*, 2017). Cd is a cell poison that can cause different types of damage, including morphological damage and biochemical changes. Exposure to Cd can damage various tissues, such as the liver, kidney, and lung, of humans (Huo *et al.*, 2017; Iwegbue *et al.*, 2015). Pb is a neurotoxic compound that can cause mental retardation in children and exert adverse effects on the liver, kidney, and reproductive system (Egbueri, 2020; Tahmasbian *et al.*, 2014). As Pb is also a highly toxic carcinogen, chronic exposure to this compound can also cause functional damage in most organ systems in humans (Ozturk *et al.*, 2022).

The accumulation of toxic elements in aquatic animals has garnered increasing global attention. However, several studies only compared the concentrations of heavy metal in sediment, water, and individual species, such as crayfish (Bellante *et al.*, 2015; Goretti *et al.*, 2016), fish (Rather *et al.*, 2019), or crab (Çoğun *et al.*, 2017). Rajeshkumar *et al.* investigated seasonal pollution of heavy metals in water, sediment, and tissues of fish and oyster from the Meiliang Bay of Tai Lake in China (Rajeshkumar *et al.*, 2018). However, these researchers did not assess the health risk to humans. Furthermore, many studies have assessed the health risk induced by toxic element contamination of single species, such as crayfish (Peng *et al.*, 2022; Sarkar *et al.*, 2016; Xiong *et al.*, 2020), fish (Arisekar *et al.*, 2020; Jiang *et al.*, 2018; Zaqoot *et al.*, 2017), or crab (Barath Kumar *et al.*, 2019). In fact, several of these studies were performed with fish and shellfish (Kumar *et al.*, 2021) or fish and crabs (Liu *et al.*, 2020) obtained from different regions in different countries. The consumption of freshwater catches, including fish, crayfish, and crab, by humans is diverse and mixed in most situations. To date, the health risk associated with the accumulation of toxic elements in fish, crayfish, and crabs has only been assessed in few comparative studies. For instance, it was reported that the metal concentrations showed great variability among fish species, and crabs had higher toxic elements concentrations than shrimp and lobster (Baki *et al.*, 2018). Besides, crabs were more likely to accumulate heavy metals than fish because of different bioaccumulation patterns (Raknuzzaman *et al.*, 2016).

In this study, the crayfish, carp, and crab and their aquaculture environment (culture water and surface sediments) were collected from Hanchuan of Hubei Province, China. The main objectives of this study were to: (1) perform an *in situ* analysis of the bioaccumulation differences of four toxic elements (Cd, Cr, As, and Pb) in the tissues of these three species from the same environment, (2) conduct a health risk assessment and comparison of the toxic elements caused by the consumption of crayfish, carp, and crab using the estimated daily intake (EDI) risk model.

Materials and Methods

Sample collection

The three freshwater catches samples were collected in Hanchuan, Hubei Province, China (N30°61272, E113°82944) (Figure S1), which belongs to the lower reaches of Hanjiang River and the hinterland of Jianghan Plain, with a subtropical monsoon climate. Rice, fish, crayfish, and crab are the major products of the local farming. Samples, including red swamp crayfish (*P. clarkii*), Chinese mitten crab (*E. sinensis*), and bighead carp (*H. nobilis*), were randomly obtained from three cultivated ponds in September, 2020. The body weight of the collected crayfish, carps, and crabs ranged from 14 g to 58 g, 120 g to 988 g, and 51 g to 119 g, respectively. A total of 135 specimens of crayfish, carp, and crab (45 for each species) were collected to assess the accumulation of the toxic elements. Three samples of culture water and sediment were collected from each pond in plastic bottles and bags.

Sample treatment and analysis

Crayfish, carp, and crab specimens were dissected to obtain the (abdominal) muscle and hepatopancreas/liver tissues. The tissue samples were dehydrated twice with anhydrous ethanol to remove the oil and fat, enabling easy grinding into powder once dried. The tissues and sediment samples stored in petri dishes and dried at 65°C were ground into powder and screened (100 mesh). Thereafter, 500 mL culture water and 0.2 g powdered tissue and sediment samples were mixed with 5 mL 65% HNO_3, respectively. The mixed tissue and sediment samples were digested in a microwave digestion system (CEM, Mars6, USA, North Carolina) from 120°C to 180°C for 45 min, cooled to 25°C, and diluted with deionized water (Goretti *et al.*, 2016; Yang *et al.*, 2018).

The concentrations of Pb, Cd, Cr, and As in the samples were determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700 series, USA). The quality assurance/quality control (QA/QC) protocol included blanks, with recalibration performed after every 20 samples using standard solutions. The certified reference material (CRM) values of the four toxic elements (As, Cd, Cr, and Pb) were 87.2 ± 4.7 μg kg^{-1}, 71.4 ± 6.7 μg kg^{-1}, 534.8 ± 54.6 μg kg^{-1}, and 1526.6 ± 107.7 μg kg^{-1}, respectively. The limit of detection (LOD) values for As, Cd, Cr, and Pb were 0.02 ± 0.004 μg kg^{-1}, 0.01 ± 0.001 μg kg^{-1}, 0.03 ± 0.002 μg kg^{-1}, and 0.01 ± 0.002 μg kg^{-1}, respectively.

Health risk assessment

As the four toxic elements are nonbiodegradable, their accumulation can be significant in crayfish, carp, and crab. As a result, the consumption of these aquatic organisms may have adverse effects in humans due to the strong probability of translocation into the human digestive systems through ingestion and subsequent transport to other organ systems (Kumari and Maiti, 2019; Nagarajan *et al.*, 2019).

Calculation of the EDI

In general, the abdominal muscle of crayfish and crab is edible; however, some individuals also favour the hepatopancreas of crayfish and crab. Therefore, the (abdominal) muscle and hepatopancreas were selected in the present study to assess the health risk. To estimate the average daily toxic element load in the body system of a consumer with a specific weight, the abdominal muscle and hepatopancreas of crayfish and crab and the muscle of carp were used to calculate accumulation risk value of the four toxic elements. EDI was calculated using the following equation (Griboff *et al.*, 2017; Hossain *et al.*, 2022):

$$EDI = Ci \times \frac{IR}{BW} \tag{1}$$

where

Ci is the metal concentration in tissue samples (mg kg^{-1}, dry weight);
IR is the ingestion rate (g day^{-1});
BW is the body weight (kg).

The element concentration in the tissue by wet weight was converted using a conversion factor of 4.8 (Maurya *et al.*, 2019; Peng *et al.*, 2022; Xiong *et al.*, 2020). IR refers to the daily average adult per capita consumption of crayfish, carp, or crab based on the item—China, mainland (the crayfish and crab from Hubei, China were mainly consumed by Chinese people living on the mainland); food supply quantity for "Crustacean" (10.54 g $capita^{-1}$ day^{-1}, wet weight) (FAOSTAT, 2013). Furthermore, the carp from Hubei, China was mainly consumed by Chinese people; food supply quantity for "Freshwater Fish" (43.47 g $capita^{-1}$ day^{-1}, wet weight) (FAOSTAT, 2013). The mean ratios of the abdominal muscle and hepatopancreas to crayfish weight (abdominal muscle/crayfish weight = 13.5%, hepatopancreas/crayfish weight = 6.32%) were determined using 150 crayfish whose body weight ranged from 25 g to 50 g. The mean ratios of the muscle to carp weight were 57.73% (Wang *et al.*, 2013). The mean ratio of abdominal muscle and hepatopancreas to crab weight was 22.8% (Zhang, 2005) and 7.20% (calculated from experimental samples in this study), respectively. The BW was set as 70 kg for adults (Iqbal and Shah, 2014; Zhou *et al.*, 2022) and 20 kg for children (Griboff *et al.*, 2017).

Target hazard quotient (THQ) assessment

The THQ, which can be used to assess the risk level of noncarcinogenicity due to pollutant exposure, can be calculated using the following equation (Xiong *et al.*, 2020):

$$THQ = \frac{ED \times EF \times EDI}{RfD \times AT} \times 10^{-3} \tag{2}$$

where

ED is the ingestion exposure duration (70 years);
EF is the ingestion exposure frequency (365 days $year^{-1}$);
EDI is the estimated daily intake;
AT is the average exposure time (equivalent to EF × ED) (Naji *et al.*, 2016; Zhou *et al.*, 2022),
RfD is the oral reference dose (μg kg^{-1} day^{-1}; As, 0.3; Cr, 1500; Cd, 1; Pb, 4) (Peng *et al.*, 2022; Varol *et al.*, 2017). The limit of acceptable exposure is less than 1. Accordingly, when the THQ exceeds 1, there is a risk of hazardous impact (Kumar *et al.*, 2021).

Cancer risk (CR) assessment

Daily exposure to some potential carcinogens will increase the possibility of cancer development in humans.

The CR owing to life-long exposure to toxic elements can be calculated using the cancer slope factor (CSF) and the following equation:

$$CR = \frac{EF \times ED \times IR \times CSF}{BW \times AT} \times 10^{-3} \quad (3)$$

where

EF, ED, IR, BW, and AT represent the variables in equations (1) and (2).

The CSF of inorganic As is 1.5 mg kg^{-1} d^{-1} (USEPA, 2016; Zhou *et al.*, 2022). Inorganic As accounts for approximately 3% of the total As (Copat *et al.*, 2013; Varol *et al.*, 2017). According to the USEPA, a CR value between 1×10^{-6} and 1×10^{-4} is acceptable for humans. However, the CR value should not exceed 1×10^{-4} (Hossain *et al.*, 2022).

Statistical analysis

The data were statistically analysed using SPSS software (v.20.0, SAS Institute, Inc. USA). The statistical means and standard deviations were calculated. To assess the correlation of toxic element concentrations in samples, Pearson's correlation was adopted. Differences were considered statistically significant at $P < 0.05$. Duncan's test was used for multiple comparisons to compare the bioaccumulation differences of four toxic elements in the tissues of these three species with two significance levels of $P < 0.05$ and $P < 0.01$.

Results

Comparison of the toxic elements in three freshwater catches

The concentrations of four toxic elements (As, Cd, Cr, and Pb) in different tissues of the three freshwater catches (crayfish, carp, and crab) are presented in Table 1. The concentrations of As, Cd, and Pb in the liver/hepatopancreas of the three freshwater catches were arranged in the order: crayfish > crab > carp. The same trend was also found for the concentration of As in the (abdominal) muscle (Table 1). The concentrations of Cr and Pb in the (abdominal) muscle appeared in the order carp > crayfish > crab. Furthermore, the concentrations of Cr in the liver/hepatopancreas and Cd in the (abdominal) muscle were arranged in the order: carp > crab > crayfish and crab > crayfish > carp, respectively (Table 1). On the other hand, the concentrations of the four toxic elements in the edible part of the abdominal muscle of crayfish and crab were arranged in the order: As > Cr > Pb > Cd. The concentrations of As and Cd were higher than those of Cr and Pb in the hepatopancreas of crayfish and crab. The concentration trend of Cr > As > Pb > Cd was observed in the liver and muscle of carp (Table 1). Furthermore, the concentrations of the four toxic elements in the liver/hepatopancreas were higher than those in the (abdominal) muscle, except for Cr in crayfish (Table 1). The concentrations of Cr, Pb, and Cd in the (abdominal) muscle and hepatopancreas/liver were lower than the national standards (GB2762-2017) of China and the EC standards (NO. 629/2008; NO. 1881/2006), except for Cd in the hepatopancreas of crayfish and crab (crayfish, 6.14 mg kg^{-1}; crab, 2.51 mg kg^{-1}; GB/EC, 2.40 mg kg^{-1}) (Table 1).

Relationship between the three freshwater catches and their surrounding environment in terms of the toxic elements

The concentrations of the four toxic elements appeared in the order: As > Pb > Cr > Cd in culture water, and Cr > Pb > As > Cd in sediment (Table S1). The concentrations of the four toxic elements in the culture environment (culture water and sediment) did not exceed the

Table 1. Concentrations of the four toxic elements in different tissues of three freshwater catches (mg kg^{-1}, dry weight).

Tissues	Crayfish		Carp		Carb		GB	EC
Elements	**Am**	**He**	**Muscle**	**Liver**	**Am**	**He**		
Cr	0.53 ± 0.08^{ab}	0.39 ± 0.23^{B}	0.75 ± 0.04^{a}	1.00 ± 0.48^{A}	0.41 ± 0.15^{b}	0.42 ± 0.25^{B}	9.6	–
CV	0.15	0.51	0.05	0.48	0.37	0.6		
As	1.64 ± 0.17^{a}	12.93 ± 2.64^{A}	0.38 ± 0.03^{b}	0.67 ± 0.22^{C}	1.49 ± 0.08^{a}	3.48 ± 0.92^{B}	–	–
CV	0.10	0.2	0.08	0.33	0.05	0.26		
Cd	$10.15 \pm 5.15^{*b}$	6.14 ± 1.03^{A}	$4.49 \pm 0.14^{*c}$	0.12 ± 0.02^{C}	$14.98 \pm 1.20^{*a}$	2.51 ± 0.30^{B}	2.4	2.4
CV	0.51	0.17	0.03	0.17	0.08	0.12		
Pb	$78.75 \pm 16.98^{*ab}$	0.48 ± 0.13^{A}	0.1 ± 0.02^{a}	0.3 ± 0.09^{B}	$52.77 \pm 4.89^{*b}$	0.35 ± 0.05^{B}	2.4	2.4
CV	0.22	0.27	0.2	0.3	0.09	0.14		

Data are presented as mean ± SD; Carp, bighead carp (*H. nobilis*); Crab, Chinese mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas, Am, abdominal muscle; CV, Coefficient of Variation; "*", represent μg kg^{-1}; A, B, and C, ranked by Duncan test at $P < 0.05$ in the hepatopancreas/liver; a, b, and c, ranked by Duncan's test at $P < 0.05$ in the abdominal muscle; GB2762-2017; (EC) NO.629/2008, (EC) NO.1881/200.

national criterion (GB11607-89 & GB15618-2018) of China (Table S1).

The correlation coefficients for the four toxic elements between the tissues of the three freshwater catches and sediments, and culture water are shown in Table 2 and Table S2, respectively. The concentrations of Cr in the abdominal muscle and Cd in the hepatopancreas of crayfish had a significant negative correlation with the concentrations found in sediments (r_{Cr} = –0.63, $P < 0.05$; r_{Cd} = –0.79, $P < 0.01$) (Table 2). In contrast, the concentration of Pb in the liver of carp had a significant positive correlation with the concentration found in sediments (r_{Pb} = 0.68, $P < 0.05$) (Table 2), and the concentration in the liver of carp had a significant negative correlation with that in culture water (r_{Pb} = –0.75, $P < 0.05$) (Table S2). In addition, the concentrations of As in the muscle and Cd in the liver of carp were significantly negatively correlated with those in sediments (r_{As} = –0.69, $P < 0.05$; r_{Cd} = –0.63, $P < 0.05$) (Table 2). Furthermore, the concentration of Cr in the abdominal muscle of crab was significantly positively correlated with that in sediments (r_{Cr} = 0.95, $P < 0.01$), and significantly negatively correlated with that in culture water (r_{Cr} = –0.70, $P < 0.05$). In contrast, the concentration of Cr in the hepatopancreas of crab was significantly positively correlated with that in culture water (r_{Cr} = 0.97, $P < 0.01$), and significantly negatively correlated with that in sediments (r_{Cr} = –0.70, $P < 0.05$) (Table 2, Table S2).

The correlation coefficients of the four toxic elements in the same tissues of the three freshwater catches are shown in Table 3. A significant positive correlation was found between the concentration of As and Cr in the hepatopancreas of crayfish and crab (r = 0.66, $P < 0.05$; r = 0.93, $P < 0.01$) (Table 3). In contrast, a significant negative correlation was found between the concentration of Cd and Pb in the liver/hepatopancreas of carp and crab (r = –0.92, $P < 0.01$; r = –0.70, $P < 0.05$). Notably, the same relationship was found in the muscle of carp (r = –0.88, $P < 0.01$) (Table 3). Significant positive correlation and negative correlation were derived for the concentration of Cr and Pb in the abdominal muscle of crayfish (r = 0.66, $P < 0.05$) and crab (r = –0.64, $P < 0.05$), respectively (Table 3). A markedly significant negative correlation was found between the concentration of As and Pb in the (abdominal) muscle of crayfish and carp (r = –0.89, $P < 0.01$; r = –0.98, $P < 0.01$), while a markedly significant positive correlation (r = 0.87, $P < 0.01$) was found between the concentration of As and Pb in the abdominal muscle of crab (Table 3). Besides, a significant positive correlation was found for the concentration of As and Cd in the muscle of carp (r = 0.69, $P < 0.05$) (Table 3).

Human health risk assessment

The EDI values for the four toxic elements in the three freshwater catches are shown in Table 4. The EDI values of the four toxic elements for children (20 kg) and adults (70 kg) were calculated based on the intake by one Chinese person (Mainland) (FAOSTAT, 2013) and

Table 2. Correlation coefficients of the four toxic element concentrations between tissues and sediments.

Tissues	Crayfish		Carp		Carb	
Elements	**Am**	**He**	**Muscle**	**Liver**	**Am**	**He**
Cr	–0.63*	0.49	0.39	–0.27	0.95**	–0.70*
As	–0.02	0.25	–0.69*	–0.39	–0.18	–0.04
Cd	–0.26	–0.79**	0.03	–0.63*	0.13	0.58
Pb	0.53	–0.39	–0.24	0.68*	–0.31	–0.16

Carp, bighead carp (*H. nobilis*); Crab, Chinese mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas, Am, abdominal muscle; "*" indicates $P < 0.05$; "**" indicates $P < 0.01$.

Table 3. Correlation coefficients of the four toxic element concentrations in the same tissue.

Am/M / He/L	Cr	As	Cd	Pb
Cr		$0.12^{a}/0.38^{b}/0.17^{c}$	$0.24^{a}/0.55^{b}/–0.53^{c}$	$0.66^{a*}/0.45^{b}/–0.64^{c*}$
As	$0.66^{a*}/–0.45^{b}/0.93^{c**}$		$–0.57^{a}/0.69^{b*}/0.11^{c}$	$–0.89^{a**}/–0.98^{b**}/–0.87^{c**}$
Cd	$–0.38^{a}/–0.08^{b}/–0.03^{c}$	$–0.50^{a}/–0.22^{b}/0.09^{c}$		$0.42^{a}/–0.88^{b**}/0.26^{c}$
Pb	$–0.56^{a}/0.20^{b}/–0.48^{c}$	$–0.001^{a}/–0.03^{b}/–0.32^{c}$	$–0.18^{a}/–0.92^{b**}/–0.7^{c*}$	

He, hepatopancreas, Am, abdominal muscle, M, muscle, L, liver; "*" indicates $P < 0.05$, "**" indicates $P < 0.01$; "a" indicates red swamp crayfish (*P.* clarkii); "b" indicates bighead carp (*H. nobilis*); "c" indicates Chinese mitten crab (*E. sinensis*).

the concentrations of the toxic elements in the hepatopancreas and abdominal muscle of crayfish and crab, with the exception of carp, whose only edible tissue is muscle. The EDI values for the four toxic elements in the tissues of crayfish, carp, and crab were less than 1.97×10^{-4} mg kg^{-1} day^{-1}, which is markedly less than the provisional tolerable daily intakes (PTDIs) (JECFA, 2013) for children (20 kg) and adults (70 kg) (Table 4). The THQ values for the four toxic elements in the hepatopancreas/liver and (abdominal) muscle of the three freshwater catches are presented in Table 5. The maximum THQ values for As owing to the consumption of each of the three freshwater catches by children were determined ($0.21 \leq THQ \leq 0.38$). The THQ values for the other three toxic elements were markedly less than 1. As exposure to the four toxic elements via consumption of the freshwater catches simultaneously occurs, the total THQ (tTHQ) value for the four toxic elements was calculated to assess the additive health risk from the various toxic elements (Lu *et al.*, 2021; Madden, 2003). The tTHQ values for the consumption of the (abdominal) muscle of carp and crayfish were the highest and lowest for children (0.34 and 0.08) and adults (0.10 and 0.02), respectively (Table 5). The tTHQ value for the consumption of the hepatopancreas of crayfish by children was markedly higher (0.34) than that of the consumption of the abdominal muscle (0.08) of crayfish. In contrast, the tTHQ value for the consumption of the hepatopancreas of crab was slightly lower than that for the consumption of the abdominal muscle of crab (Table 5). Because the CSF was only available for As, its CR value for the tissues in crayfish, carp, and crab was calculated and presented in Table 6. For children, the CR values of As in the (abdominal) muscle of crayfish, crab, and carp were 1.10×10^{-9}, 1.68×10^{-9}, and 4.48×10^{-9}, respectively. A similar trend was found for the CR values (crayfish < crab < carp) for adults. The CR value of As was higher for the consumption of crayfish hepatopancreas than the abdominal muscle. However, the opposite result was obtained for the consumption of the crab tissues. Altogether, the CR values in this study were markedly less than 1×10^{-6} (Table 6).

Discussion

Different levels of bioaccumulation of the toxic elements in freshwater catches

Metals and metalloids are ubiquitously present in the environment and can be enriched in organisms mainly through the food chain (Gedik *et al.*, 2017; Goretti *et al.*, 2016; Mohanasrinivasan *et al.*, 2014; Xiong *et al.*, 2020). Compared with fish, crayfish and crabs are typical benthic organisms. Accordingly, different bioaccumulation

Table 4. Estimated daily intake (EDI) of the four toxic elements (mg kg^{-1} day^{-1}).

Body weight	Tissue	Crayfish				Carb				Carb			
		Cr	As	Pb	Cd	Cr	As	Pb	Cd	Cr	As	Pb	Cd
Children (20 kg)	Am/M	7.89×10^{-6}	2.44×10^{-5}	1.17×10^{-6}	1.50×10^{-7}	1.96×10^{-4}	9.95×10^{-5}	2.69×10^{-5}	1.17×10^{-6}	1.03×10^{-5}	3.74×10^{-5}	1.32×10^{-6}	3.70×10^{-7}
	He	2.68×10^{-6}	8.97×10^{-5}	3.33×10^{-6}	4.26×10^{-5}	–	–	–	–	3.36×10^{-6}	2.75×10^{-5}	2.77×10^{-6}	1.98×10^{-5}
Adults (70 kg	Am/M	2.26×10^{-6}	6.96×10^{-6}	3.30×10^{-7}	4.30×10^{-8}	5.60×10^{-5}	2.84×10^{-5}	7.68×10^{-6}	3.40×10^{-7}	2.93×10^{-6}	1.07×10^{-5}	3.80×10^{-7}	1.10×10^{-7}
	He	7.70×10^{-7}	2.56×10^{-5}	9.50×10^{-7}	1.22×10^{-5}	–	–	–	–	9.60×10^{-7}	7.85×10^{-6}	7.90×10^{-7}	5.66×10^{-6}
PTDI		–	2.14×10^{-3}	3.57×10^{-3}	0.83×10^{-3}	–	2.14×10^{-3}	3.57×10^{-3}	0.83×10^{-3}	–	2.14×10^{-3}	3.57×10^{-3}	0.83×10^{-3}

Carp, bighead carp (*H. nobilis*); Crab, Chinese mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas, Am, abdominal muscle, M, muscle; PTDI, provisional tolerable daily intake.

Table 5. Target hazard quotient (THQ) of the four toxic elements.

Species	Body weight	Tissue	Cr	As	Pb	Cd	tTHQ
Crayfish	Children (20 kg)	Am	5.26×10^{-6}	8.13×10^{-2}	2.93×10^{-4}	1.50×10^{-4}	0.08
		He	1.79×10^{-6}	0.3	8.33×10^{-4}	4.26×10^{-2}	0.34
	Adults (70 kg)	Am	1.51×10^{-6}	2.32×10^{-2}	8.25×10^{-5}	4.30×10^{-5}	0.02
		He	5.13×10^{-7}	8.53×10^{-2}	2.38×10^{-5}	1.22×10^{-2}	0.10
Carp	Children (20 kg)	M	1.31×10^{-4}	0.33	6.73×10^{-3}	1.17×10^{-3}	0.34
	Adults (70 kg)		3.73×10^{-5}	9.47×10^{-2}	1.92×10^{-3}	3.40×10^{-4}	0.10
Crab	Children (20 kg)	Am	6.87×10^{-6}	0.125	3.30×10^{-4}	3.70×10^{-4}	0.13
		He	2.24×10^{-6}	9.17×10^{-2}	6.93×10^{-4}	1.98×10^{-2}	0.11
	Adults (70 kg)	Am	1.95×10^{-6}	3.57×10^{-2}	9.50×10^{-5}	1.10×10^{-4}	0.04
		He	6.40×10^{-7}	2.62×10^{-2}	1.98×10^{-4}	5.66×10^{-3}	0.03

Carp, bighead carp (*H. nobilis*); Crab, Chinese mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas, Am, abdominal muscle, M, muscle; The tTHQ (total THQ value) was treated as the arithmetic sum of the individual metal THQ values.

patterns of metals and metalloid may occur in these three freshwater catches. However, simultaneous investigations of the accumulation of toxic elements in three freshwater catches from the same environment, including crayfish, carp, and crab, which are frequently consumed by humans, have rarely been performed. In this study, the accumulation of four toxic elements was investigated in these freshwater catches and their common environment. The concentrations of Cd (10.15 μg kg^{-1}) and Pb (78.75 μg kg^{-1}) in the abdominal muscle of crayfish were lower than those in crayfish (Cd, 60 μg kg^{-1}, Pb, 2.24 mg kg^{-1}) from Louisiana, America (Gedik *et al.*, 2017), and crayfish (Cd, 2.4 mg kg^{-1}, Pb, 2.2 mg kg^{-1}) from Italy (Goretti *et al.*, 2016). The concentration of Cd (14.98 μg kg^{-1}) in the abdominal muscle of crab was lower than the previously reported concentration (0.10 mg kg^{-1}) for crab from Jiangsu province, China (Fan *et al.*, 2021). The concentrations of Cd (4.49 μg kg^{-1}) and Pb (0.1 mg kg^{-1}) in the muscle of carp were lower than those of carp (Cd, 50 μg kg^{-1}, Pb, 0.71 mg kg^{-1}) from Shandong, China (Li *et al.*, 2015). The concentration of As (0.38 mg kg^{-1}) in the muscle of carp was close to the previously reported concentration (0.36 mg kg^{-1}) (Li *et al.*, 2015). Besides, the crayfish and crab evaluated in this study had a similar accumulation pattern of the toxic elements (As > Cd > Pb) in their hepatopancreas. These three toxic elements appeared as follows in the hepatopancreas/liver of the freshwater catches: crayfish > crab > carp. These results further indicate that the three toxic elements had similar enrichment in crayfish and crab, which differed from the enrichment in carp. This finding corresponds to reports that revealed that different aquatic animals have different enrichment levels of metal and metalloid (crustaceans > fish) (Lu *et al.*, 2021). Furthermore, the concentrations of the four toxic elements were higher in the hepatopancreas/liver than the (abdominal) muscle of the three freshwater catches, except for Cr in crayfish (Table 1), which aligns with previous studies that found that the hepatopancreas/liver play a major role in metabolism and tend to accumulate higher levels of metals and metalloids than the (abdominal) muscle (Goretti *et al.*, 2016). Of note, the concentrations of Cr and Pb in the (abdominal) muscle of the three freshwater catches showed the same trend (crab < crayfish < carp), opposing that found for the concentration trend of As and Cd (crayfish/crab > carp) in the (abdominal) muscle of the three freshwater catches (Table 1). Together, these results suggest that the hepatopancreas of crayfish and crab generally bioaccumulates more toxic elements than that of carp. However, different bioaccumulation levels of the toxic elements were found in the (abdominal) muscle of the three freshwater catches. Such a finding may be attributed to the distinct genetic

Table 6. Cancer risk (CR) estimate for As.

Body weight	Tissue	As		
		Crayfish	Carp	Crab
Children (20 kg)	Am/M	1.10×10^{-9}	4.48×10^{-9}	1.68×10^{-9}
	He	4.04×10^{-9}	–	1.24×10^{-9}
Adults (70 kg)	Am/M	3.13×10^{-10}	1.28×10^{-9}	4.81×10^{-10}
	He	1.15×10^{-9}	–	3.53×10^{-10}

Carp, bighead carp (*H. nobilis*); Crab, Chinese mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas; Am, abdominal muscle, M, muscle.

regulation of absorption, transport, and metabolism of the toxic elements among the freshwater catches.

Differences in the levels of bioaccumulation of the toxic elements may be species-dependent

Fish can absorb toxic elements by ingesting particulate matter suspended in water (Varol *et al.*, 2017). However, crayfish and crab are benthic organisms and thus mainly feed on benthic food in sediments, which can accumulate toxic elements from sediments. In this study, the concentration of four toxic elements in the hepatopancreas/liver and (abdominal) muscle and in sediment and cultural water was investigated. Based on the results, the concentration of the four toxic elements in the sediments had a significant correlation with that in tissues of the three freshwater catches (Table 2). For example, the concentration of As and Pb in the sediment had significant negative ($r = -0.69$, $P < 0.05$) and positive ($r = 0.68$, $P < 0.05$) correlations with the concentration in the muscle and liver of carp, respectively. The concentration of Cr in the sediment had a significant negative ($r = -0.63$, $P < 0.05$) or positive ($r = 0.95$, $P < 0.01$) correlation with that in the abdominal muscle of crayfish and crab, respectively. However, only the concentrations of Cr and Pb in cultured water correlated with those found in the tissue of carp and crab, respectively (Table S2). In particular, no significant correlation was found between the tissues of crayfish and cultured water for any of the toxic elements. Compared to cultured water, sediments had a higher correlation with the bioaccumulation of toxic elements in the freshwater catches. These results align with those of previous reports (Nagarajan *et al.*, 2019; Varol *et al.*, 2017). Furthermore, the concentrations of Cr in the hepatopancreas of crab, As in the muscle of carp, and Cd in the hepatopancreas of crayfish were, respectively, and significantly negatively correlated with those in the sediment; however, the concentrations of Cr in the abdominal muscle of crab and Pb in the liver of carp were significantly positively correlated with the concentrations in sediment, respectively (Table 2). These results indicate that the accumulation of toxic elements in different freshwater catches might not be completely dependent on their living environment, but may be also species dependent.

The consumption of crayfish is associated with a lower risk to human health than carp and crab

Aquatic products are important food sources for human beings. These products are not only rich in nutrients but also impact people's choice of eating. Nonetheless, the consumption of aquatic animals has markedly increased human exposure to toxic elements (Varol *et al.*, 2017). To date, the health risk associated with the individual consumption of crayfish, carp, or crab was assessed in different studies from several countries (Jiang *et al.*, 2018; Kumar *et al.*, 2021; Lu *et al.*, 2021). However, only few comparative studies have been conducted to assess the health risk associated with the consumption of fish, crayfish, and crabs that accumulate toxic elements in their tissues. Consumers often consume various freshwater catches, such as crayfish, carp, and crab at the same time. The health risk caused by the consumption of multiple food types should thus be assessed. In the present study, the health risk caused by the consumption of crayfish, carp, and crab were assessed together. In addition, the health risks for adults and children were separately considered as children are more sensitive to pollutants than adults (Lu *et al.*, 2021). The EDI values for the four toxic elements in the (abdominal) muscle of crayfish, carp, and crab were less than the PTDIs (JECFA, 2013) for children (20 kg) and adults (70 kg) (Table 4). Such findings suggest that a very low risk is associated with the consumption of the three freshwater organisms. Furthermore, the THQ and CR associated with the consumption of the (abdominal) muscle of the freshwater catches were markedly below the acceptable thresholds ($THQ < 1$ and $CR < 10^{-6}$). However, consuming the three freshwater organisms at the same time could increase the THQ and CR values. In particular, the sum of the tTHQ values for the consumption of the hepatopancreas of crayfish and crab was 0.45 for children (20 kg) and 0.13 for adults (70 kg); these values are remarkably higher than those (0.21 for children, 0.06 for adults) obtained for the abdominal muscle. In addition to the high concentration of the toxic elements in the hepatopancreas of crayfish and crab, the Cd concentration in the hepatopancreas of crayfish and crab exceeded the national standards (GB2762-2017) of China and the EC standards (NO. 629/2008; NO. 1881/2006) (Table 1). Therefore, the hepatopancreas of crayfish and crab should not be consumed by humans. The THQ values associated with the consumption of the (abdominal) muscle of the three freshwater catches were arranged in the order of crayfish (0.08 for children, 0.02 for adults) < crab (0.13 for children, 0.04 for adults) < carp (0.34 for children, 0.10 for adults). As tTHQ increases with the mixed consumption of the (abdominal) muscle of the three freshwater catches, a multiple-low-dose approach should be adopted for the (abdominal) muscle, especially that of the carp when consumed by children.

Conclusion

The concentrations of toxic elements (As, Cd, and Pb) in the (abdominal) muscle were found to be markedly lower than those in the hepatopancreas/liver of the three freshwater catches (crayfish, carp, crab). Therefore, it is suggested that the hepatopancreas of crayfish and crab should not be consumed due to their high concentration of toxic elements. The EDI of these elements in the (abdominal) muscle of crayfish, carp, and crab did not exceed the PTDIs, and the THQ and CR associated with the consumption of the (abdominal) muscle were markedly below the acceptable thresholds (THQ < 1 and CR < 10^{-6}). Of note, consuming the (abdominal) muscle of one of the freshwater animals does not pose a health risk to human beings. However, the tTHQ value could significantly increase if the (abdominal) muscles of the three freshwater catches are simultaneously consumed. Altogether, a multiple-low-dose approach is suggested for the consumption of the (abdominal) muscle of the three freshwater catches by humans.

Limitations of study and perspectives for future research

The three toxic elements (As, Cd, and Pb) had a higher enrichment in the hepatopancreas/liver of the three freshwater catches compared to that of the (abdominal) muscle. On the contrary, Cr had a higher enrichment in the abdominal muscle than that in the hepatopancreas of crayfish. However, the molecular mechanism of the different enrichment of the toxic elements among the different freshwater catches could not be explored here, which needs to be further dissected in a later study. Besides, due to experimental limitations, only three freshwater catches were investigated to carry out toxic elements detection and health risk assessment in this study. The additional aquatics from the different regions of the world should be further investigated in future studies.

References

Arisekar, U., Shakila, R.J., Shalini, R. and Jeyasekaran, G., 2020. Human health risk assessment of heavy metals in aquatic sediments and freshwater fish caught from Thamirabarani River, the Western Ghats of South Tamil Nadu. Marine Pollution Bulletin 159: 111496. https://doi.org/10.1016/j.marpolbul.2020.111496

Baki, M.A., Hossain, M.M., Akter, J., Quraishi, S.B., Haque Shojib, M.F., Atique Ullah, A.K.M. and Khan, M.F., 2018. Concentration of heavy metals in seafood (fishes, shrimp, lobster and crabs) and human health assessment in Saint Martin Island, Bangladesh. Ecotoxicology and Environmental Safety 159: 153–163. https://doi.org/10.1016/j.ecoenv.2018.04.035

Barath Kumar, S., Padhi, R.K. and Satpathy, K.K., 2019. Trace metal distribution in crab organs and human health risk assessment on consumption of crabs collected from coastal water of South East coast of India. Marine Pollution Bulletin 141: 273–282. https://doi.org/10.1016/j.marpolbul.2019.02.022

Bellante, A., Maccarone, V., Buscaino, G., Buffa, G., Filiciotto, F., Traina, A., Del Core, M., Mazzola, S. and Sprovieri, M., 2015. Trace element concentrations in red swamp crayfish (*Procambarus clarkii*) and surface sediments in Lake Preola and Gorghi Tondi natural reserve, SW Sicily. Environmental Monitoring and Assessment 187: 404. https://doi.org/10.1007/s10661-015-4613-4

Çoğun, H.Y., Firat, Ö., Aytekin, T., Firidin, G., Firat, Ö., Varkal, H., Temiz, Ö. and Kargin, F., 2017. Heavy Metals in the Blue Crab (*Callinectes sapidus*) in Mersin Bay, Turkey. Bulletin of Environmental Contamination and Toxicology 98: 824–829. https://doi.org/10.1007/s00128-017-2086-6

Copat, C., Arena, G., Fiore, M., Ledda, C., Fallico, R., Sciacca, S. and Ferrante, M., 2013. Heavy metals concentrations in fish and shellfish from eastern Mediterranean Sea: consumption advisories. Food and Chemical Toxicology 53: 33–37. https://doi.org/10.1016/j.fct.2012.11.038

Egbueri, J.C., 2020. Heavy metals pollution source identification and probabilistic health risk assessment of shallow groundwater in Onitsha, Nigeria. Analytical Letters 53: 1620–1638. https://doi.org/10.1080/00032719.2020.1712606

Fan, Z., Zou, J., Wang, Q., Qiu, L., Hu, G., Song, C. and Chen, J., 2021. Quantitative benefit and risk assessment of cadmium and nutrientlevels in Chinese mitten crab (*Eriocheir sinensis*). Environmental Science and Pollution Research 28: 7322–7331. https://doi.org/10.1007/s11356-020-10977-9

FAOSTAT, 2013. China. Food Balances. Available at: http://www.fao.org/faostat/en/#data/CL

Fernández-Trujillo, S., López-Perea, J.J., Jiménez-Moreno, M., Martín-Doimeadios, R.C. and Mateo, R., 2021. Metals and metalloids in freshwater fish from the floodplain of Tablas de Daimiel National Park, Spain. Ecotoxicology and Environmental Safety 208: 111602. https://doi: 10.1016/j.ecoenv.2020.111602

Gedik, K., Kongchun, M., DeLaune, R. and Sonnier, J., 2017. Distribution of arsenic and other metals in crayfish tissues (*Procambarus clarkii*) under different production practices. Science of the Total Environment 574: 322–331. https://doi.org/10.1016/j.scitotenv.2016.09.060

Goretti, E., Pallottini, M., Ricciarini, M.I., Selvaggi, R. and Cappelletti, D., 2016. Heavy metals bioaccumulation in selected tissues of red swamp crayfish: an easy tool for monitoring environmental contamination levels. Science of the Total Environment 559: 339–346. https://doi.org/10.1016/j.scitotenv.2016.03.169

Griboff, J., Wunderlin, D.A. and Monferran, M.V., 2017. Metals, As and Se determination by inductively coupled plasma-mass spectrometry (ICP-MS) in edible fish collected from three eutrophic reservoirs. Their consumption represents a risk for human health? Microchemical Journal 130: 236–244. https://doi.org/10.1016/j.microc.2016.09.013

Hossain, M.B., Tanjin, F., Rahman, M.S., Yu, J., Akhter, S., Noman, M.A. and Sun, J., 2022. Metals bioaccumulation in 15 commonly consumed fishes from the lower Meghna River and Adjacent Areas of Bangladesh and associated human health hazards. Toxics 10: 139. https://doi.org/10.3390/toxics10030139

Hui, H., Luo, Y., Zhu, S. and Shen, H., 2012. Establishment of quality predictive models for bighead carp (*Aristichthys nobilis*) fillets during storage at different temperatures. International Journal of Food Science and Technology 47: 488–497. https://doi.org/10.1111/j.1365-2621.2011.02868.x

Huo, J.F., Dong, A.G., Yan, J.J., Wang, L., Ma, C.G. and Lee, S.C., 2017. Cadmium toxicokinetics in the freshwater turtle, *Chinemys reevesii*. Chemosphere 182: 392–398. https://doi.org/10.1016/j.chemosphere.2017.05.029

Iqbal, J. and Shah, M.H., 2014. Study of seasonal variations and health risk assessment of heavy metals in *Cyprinus carpio* from Rawal Lake, Pakistan. Environmental Monitoring and Assessment 186: 2025–2037. https://doi.org/10.1007/s10661-013-3515-6

Iwegbue, C.M.A., Bassey, F.I., Tesi, G.O., Overah, L.C., Onyeloni, S.O. and Martincigh, B.S., 2015. Concentrations and health risk assessment of metals in chewing gums, peppermints and sweets in Nigeria. Journal of Food Measurement and Characterization 9: 160–174. https://doi.org/10.1007/s11694-014-9221-4

JECFA, 2013. Evaluation of certain food additives and contaminants: seventy-seventh report of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), JECFA/77/TRS. Food & Agriculture Organisation, Rome.

Jiang, K.J., Zhang, F.Y., Pi, Y., Jiang, L.L., Yu, Z.L., Zhang, D., Sun, M.M., Gao, L.J., Qiao, Z.G. and Ma, L.B., 2014. Amino acid, fatty acid, and metal compositions in edible parts of three cultured economic crabs: Scylla paramamosain, *Portunus trituberculatus*, and *Eriocheir sinensi*s. Journal of Aquatic Food Product Technology 23: 73–86. https://doi.org/10.1080/10498850.2012.695761

Jiang, Z., Xu, N., Liu, B., Zhou, L., Wang, J., Wang, C., Dai, B. and Xiong, W., 2018. Metal concentrations and risk assessment in water, sediment and economic fish species with various habitat preferences and trophic guilds from Lake Caizi, Southeast China. Ecotoxicology and Environmental Safety 157: 1–8. https://doi.org/10.1016/j.ecoenv.2018.03.078

Köker, L., Aydın, F., Gaygusuz, Ö., Akçaalan, R., Çamur, D., İlter, H., Ayoğlu, F.N., Altın, A., Topbaş, M. and Albay, M., 2021. Heavy metal concentrations in trachurus mediterraneus and merlangius merlangus captured from Marmara sea, turkey and associated health risks. Environmental Management 67: 522–531. https://doi.org/10.1007/s00267-020-01352-y

Kumar, P., Sivaperumal, P., Manigandan, V., Rajaram, R. and Hussain, M., 2021. Assessment of potential human health risk due to heavy metal contamination in edible finfish and shellfish collected around Ennore coast, India. Environmental Science and Pollution Research 28: 8151–8167. https://doi.org/10.1007/s11356-020-10764-6

Kumari, P. and Maiti, S.K., 2019. Health risk assessment of lead, mercury, and other metal(loid)s: a potential threat to the population consuming fish inhabiting, a lentic ecosystem in Steel City (Jamshedpur), India. Human and Ecological Risk Assessment 25: 2174–2192. https://doi.org/10.1080/10807039.2018.1495055

Li, P., Zhang, J., Xie, H., Liu, C., Liang, S., Ren, Y. and Wang, W., 2015. Heavy Metal Bioaccumulation and Health Hazard Assessment for Three Fish Species from Nansi Lake, China. Bulletin of Environmental Contamination and Toxicology 94: 431–436. https://doi.org/10.1007/s00128-015-1475-y

Liu, Q., Liao, Y., Xu, X., Shi, X., Zeng, J., Chen, Q. and Shou, L., 2020. Heavy metal concentrations in tissues of marine fish and crab collected from the middle coast of Zhejiang Province, China. Environmental Monitoring and Assessment 192: 285. https://doi.org/10.1007/s10661-020-8234-1

Lu, J., Lin, Y., Wu, J. and Zhang, C., 2021. Continental-scale spatial distribution, sources, and health risks of heavy metals in seafood: challenge for the water-food-energy nexus sustainability in coastal regions? Environmental Science and Pollution Research 28: 63815–63828. https://doi.org/10.1007/s11356-020-11904-8

Madden, E.F., 2003. The role of combined metal interactions in metal carcinogenesis: a review. Reviews on Environmental Health 18: 91–110. https://doi.org/10.1515/reveh.2003.18.2.91

Maurya, P.K., Malik, D.S., Yadav, K.K., Kumar, A., Kumar, S. and Kamyab, H., 2019. Bioaccumulation and potential sources of heavy metal contamination in fish species in River Ganga basin: possible human health risks evaluation. Toxicology Reports 6: 472–481. https://doi.org/10.1016/j.toxrep.2019.05.012

Ministry of Agriculture and Rural Affairs of the People's Republic of China (MARA), National Fisheries Technology Extension Center (NFTEC) and China Society of Fisheries (CSF), 2020. China fisheries statistical yearbook. China Agriculture Press. MARA, NFTEC and CSF, Beijing, China.

Mohanasrinivasan, V., Mishra, M., Paliwal, J.S., Singh, S.K., Selvarajan, E., Suganthi, V. and Devi, C.S., 2014. Studies on heavy metal removal efficiency and antibacterial activity of chitosan prepared from shrimp shell waste. 3 Biotech 4: 167–175. https://doi.org/10.1007/s13205-013-0140-6

National Fisheries Technology Extension Center (NFTEC) and China Society of Fisheries (CSF), 2020. Report on the development of crayfish industry in China. Chinese Fisheries Economics, Beijing, China.

Nagarajan, R., Anandkumar, A., Hussain, S.M., Jonathan, M.P., Ramkumar, M., Eswaramoorthi, S., Saptoro, A. and Chua, H.B., 2019. Geochemical characterization of Beach sediments of Miri, NW Borneo, SE Asia: implications on provenance, weathering intensity, and assessment of coastal environmental Status. Ch. 12. In "Coastal Zone Management". M. Ramkumar, R.A. James, D. Menier and K. Kumaraswamy (Ed.), 279–330. https://doi.org/10.1016/B978-0-12-814350-6.00012-4

Naji, A., Khan, F.R. and Hashemi, S.H., 2016. Potential human health risk assessment of trace metals via the consumption of marine fish in Persian gulf. Marine Pollution Bulletin 109: 667–671. https://doi.org/10.1016/j.marpolbul.2016.05.002

Ozturk, M., Metin, M., Altay, V., Bhat, R., Ejaz, M., Gul, A., Unal, B., Hasanuzzaman, M., Nibir, L., Nahar, K., Bukhari, A., Dervash, M. and Kawano, T., 2022. Arsenic and human health: genotoxicity, epigenomic effects, and cancer signaling. Biological Trace Element Research 200: 988–1001. https://doi.org/10.1007/s12011-021-02719-w

Peng, F., Li, J., Gong, Z., Yue, B., Wang, X., Manyande, A. and Du, H., 2022. Investigation of bioaccumulation and human health risk assessment of heavy metals in crayfish (*Procambarus clarkii*) farming with a rice-crayfish-based coculture breeding modes. Foods 11: 261. https://doi.org/10.3390/foods11030261

Peng, Q., Nunes, L.M., Greenfield, B.K., Dang, F. and Zhong, H., 2016. Are Chinese consumers at risk due to exposure to metals in crayfish? A bioaccessibility-adjusted probabilistic risk assessment. Environment International 88: 261–268. https://doi.org/10.1016/j.envint.2015.12.035

Rajeshkumar, S., Liu, Y., Zhang, X., Ravikumar, B., Bai, G. and Li, X., 2018. Studies on seasonal pollution of heavy metals in water, sediment, fish and oyster from the Meiliang Bay of Taihu Lake in china. Chemosphere 191: 626–638. https://doi.org/10.1016/j.chemosphere.2017.10.078

Raknuzzaman, M., Ahmed, M.K., Islam, M.S., Habibullah-Al-Mamun, M., Tokumura, M., Sekine, M. and Masunaga, S., 2016. Trace metal contamination in commercial fish and crustaceans collected from coastal area of Bangladesh and health risk assessment. Environmental Science and Pollution Research International 23: 17298–17310. https://doi.org/10.1007/s11356-016-6918-4

Ramon, D., Morick, D., Croot, P., Ran, B., Scheinin, A., Dan, T., Nadav, D. and Malka, B., 2021. A survey of arsenic, mercury, cadmium, and lead residues in seafood (fish, crustaceans, and cephalopods) from the south-eastern Mediterranean Sea. Journal of Food Science 86: 1153–1161. https://doi.org/10.1111/1750-3841.15627

Rather, M.Y., Tilwani, Y.M. and Dey, A., 2019. Assessment of heavy metal contamination in two edible fish species *Carassius carassius* and *Triplophysa kashmirensis* of Dal Lake, Srinagar, Kashmir, India. Environmental Monitoring and Assessment 191: 242. https://doi.org/10.1007/s10661-019-7382-7

Samuel, M.S., Datta, S., Khandge, R.S. and Selvarajan, E., 2021. A state of the art review on characterization of heavy metal binding metallothioneins proteins and their widespread applications. Science of the Total Environment 775: 145829. https://doi.org/10.1016/j.scitotenv.2021.145829

Sarkar, T., Alam, M.M., Parvin, N., Fardous, Z., Chowdhury, A.Z., Hossain, S., Haque, M.E. and Biswas, N., 2016. Assessment of heavy metals contamination and human health risk in shrimp collected from different farms and rivers at Khulna-Satkhira region, Bangladesh. Toxicology Reports 3: 346–350. https://doi.org/10.1016/j.toxrep.2016.03.003

Solgi, E. and Mirmohammadvali, S., 2021. Comparison of the heavy metals, copper, iron, magnesium, nickel, and zinc between muscle and gills of four benthic fish species from Shif Island (Iran). Bulletin of Environmental Contamination and Toxicology 106: 658–664. https://doi.org/10.1007/s00128-021-03155-1

Tahmasbian, I., Nasrazadani, A., Shoja, H. and Sinegani, A.A.S., 2014. The effects of human activities and different land-use on trace element pollution in urban topsoil of Isfahan (Iran). Environmental Earth Sciences 71: 1551–1560. https://doi.org/10.1007/s12665-013-2561-2

United States Environmental Protection Agency (USEPA), 2016, Washington. Integrated Risk Information System (IRIS). Available at: https://www.epa.gov/iris.

Varol, M., Kaya, G.K. and Alp, A., 2017. Heavy metal and arsenic concentrations in rainbow trout (*Oncorhynchus mykiss*) farmed in a dam reservoir on the Firat (Euphrates) River: risk-based consumption advisories. Science of the Total Environment 46: 1288–1296. https://doi.org/10.1016/j.scitotenv.2017.05.052

Wang, J.N., Tang, L., Liu, K.Q., Hui, L.I., Yao, J.J. and Miao, A.N., 2013. Analysis of nutritions in the muscle between cultured and wild variegated carp. Hebei Fisheries 2: 8–16. https://doi.org/10.3969/j.issn.1004-6755.2013.02.003

Xiong, B., Xu, T., Li, R.P., Johnson, D., Ren, D., Liu, H.G., Xi, Y. and Huang, Y.P., 2020. Heavy metal accumulation and health risk assessment of crayfish collected from cultivated and uncultivated ponds in the Middle Reach of Yangtze River. Science of the Total Environment 739: 139963. https://doi.org/10.1016/j.scitotenv.2020.139963

Yang, C.P., Liu, Y., Shan, B.B., Xu, J., Yu, W., Sun, D.R. and Zhang, Z.W., 2021. Heavy metal concentrations and associated health risks in edible tissues of marine nekton from the outer Pearl River Estuary, South China Sea. Environmental Science and Pollution Research 28: 2108–2118. https://doi: 10.1007/s11356-020-10605-6

Yang, M., Lu, K., Zhao, F.J., Xie, W., Ramakrishna, P., Wang, G., Du, Q., Liang, L., Sun, C., Zhao, H., Zhang, Z., Liu, Z., Tian, J., Huang, X.Y., Wang, W., Dong, H., Hu, J., Ming, L., Xing, Y., Wang, G., Xiao, J., Salt, D.E. and Lian, X., 2018. Genome-wide association studies reveal the genetic basis of ionomic variation in rice. The Plant Cell 30: 2720–2740. https://doi.org/10.1105/tpc.18.00375

Zaqoot, H.A., Aish, A.M. and Wafi, H.N., 2017. Baseline concentration of heavy metals in fish collected from Gaza fishing harbor in the Mediterranean Sea along Gaza coast, Palestine. Turkish Journal of Fisheries and Aquatic Sciences 17: 101–109. https://doi.org/10.4194/1303-2712-v17112

Zhang, T.Q., 2005. Nutrient parameter comparison and analysis on Chinese-Mitten Handed Crab from different proliferation and culturing water. Nanjing Agricultural University, Nanjing, China.

Zhou, H., Ge, T., Li, H., Fang, T., Li, H., Shi, Y., Zhang, R. and Dong, X., 2022. A multi-medium analysis of human health risk of toxic elements in rice-crayfish system: a case study from middle reach of Yangtze River, China. Foods 11: 1160. https://doi.org/10.3390/foods11081160

Supplementary

Figure S1. Location map of specimen collection. "📍" indicates the sampling site.

Table S1. Concentrations of the four toxic element in culture environment.

Elements	Cr	As	Cd	Pb
Culture water (μg L^{-1})	1.33 ± 0.29	13.19 ± 9.32	0.02 ± 0.002	1.71 ± 0.51
GB	100	50	5	50
Sediment (mg kg^{-1})	59.99 ± 4.20	8.77 ± 1.11	0.30 ± 0.05	24.34 ± 2.81
GB	250	30	0.4	100

Data are presented as mean ± SD; The reference standard of culture water samples is GB11607-89; The reference standard of sediment samples is GB15618-2018.

Table S2. Correlation coefficients of the four toxic element concentrations between tissues and culture water.

Tissues	Crayfish		Carp		Crab	
Elements	Am	He	Muscle	Liver	Am	He
Cr	0.27	–0.56	–0.08	0.09	–0.70*	0.97**
As	0.46	–0.04	–0.45	–0.50	0.26	–0.27
Cd	–0.32	0.10	–0.54	0.30	0.5	0.22
Pb	–0.33	0.33	–0.11	–0.75*	0.48	0.24

Carp, bighead carp (*H. nobilis*); Crab, Chinese Mitten crab (*E. sinensis*); Crayfish, red swamp crayfish (*P. clarkii*); He, hepatopancreas, Am, abdominal muscle; "*" indicates $P < 0.05$; "**" indicates $P < 0.01$.