



# Human biomonitoring of heavy metals exposure in different age- and gender-groups based on fish consumption patterns in typical coastal cities of China

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## ABSTRACT

The study aimed to investigate heavy metals (HMs) concentrations in human hair based on fish consumption patterns in Qingdao, Xiamen, and Zhoushan. The (HMs) concentrations were determined using acid digestion and an inductively coupled plasma mass spectrometer (ICP-MS, NexION 300X, PerkinElmer). The associated health risks were investigated using risk assessment models described by the United States Environmental Protection Agency (USEPA). The order of fish HMs concentration in Qingdao was Cd<Pb<As>Cr<Cu<Zn, while in Zhoushan and Xiamen the order was Cd<Pb<Cr<Cu<As<Zn. The highest concentrations of HMs in fish were found in Zhoushan and lowest in Xiamen. Zn showed the highest values in fish, while Cd and Pb were the lowest. The Cr concentration in *Scorpaenopsis niphonius* collected in Qingdao exceeded the safety standards. The HMs in both male and female hair followed a descending order of Zn>Cu>Pb>Cr>As>Cd in all three study areas. The hair Zn concentration in 28 % of the studied population exceeded the safety standards. Overall, the hair HMs concentration was found to be high in middle-aged groups (19–45 and 45–59), and the hair HMs concentrations were high, especially in the case of females. A significant correlation was noticed between hair As (0.119;  $p < 0.05$ ), Cr (0.231;  $p < 0.05$ ), and Cu (0.117;  $p < 0.05$ ), and fish consumption frequency. High Odd ratios (>2) were noticed for As, Cu and Zn in high fish-eating frequency. A significant non-carcinogenic risk was noticed in human Cr exposure (1.10E+00) in Xiamen, and the hazard index values indicated non-carcinogenic risk in Xiamen and Zhoushan. The carcinogenic risk for human As exposure (2.50E-05–7.09E-03) indicated a significant cancer risk.

## 1. Introduction

Elements with densities higher than 5 g/m<sup>3</sup> are termed HMs, and it includes elements such as arsenic (As), lead (Pb), cadmium (Cd), nickel (Ni), copper (Cu), mercury (Hg), and tin (Sn) (Arumugam et al., 2020; Mohammadi et al., 2022). These pollutants can be found naturally in the aquatic environment (earth crust) or from anthropogenic activities such as mining, smelting, pesticides, coal combustion, industrial processing, and waste incineration through runoff and atmospheric deposition or disposal of industrial effluents (Soltani et al., 2019). These elements are of great concern due to their non-biodegradable nature, high toxicity,

persistence and bioaccumulation (Angaru et al., 2022). Some of these HMs, such as Co, Cr, Cu, Zn, etc., are essential elements that are vital constituents of some enzymes and play an essential role in different oxidation-reduction processes; however, high levels of these elements can cause cellular and tissue damage (Faraji et al., 2023; WHO, 1996). In contrast, other HMs such as Pb, As, Cd and Hg have no biological function and are highly toxic even at a very low concentration. Pb can cause renal failure, coma, mental retardation, liver damage, and even death (ATSDR, 2007a). As can lead to a high risk of skin cancer and lesions such as dermatomycosis (Kamala-kannan et al., 2008). A high dose of Cd can cause kidney injury, growth reduction, and hypocalcemia

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(ATSDR, 2012a).

In China, the annual production of aquatic products has been on the rise for the last ten consecutive years. In 2020, the total aquaculture production in China was 52.24 Mt, of which 27.61 Mt (52.8 %) was attributed to fish (China Fisheries Society, 2021). It is an essential source of carbohydrates, vitamins, micronutrients, and proteins (Varol and Sünbül, 2020). Owing to the worldwide advancement of industries and urbanization, aquatic environments are contaminated with various toxicants and heavy metals (HMs). These HMs undergo various biogeochemical cycles upon entering the aquatic environment and can be bioaccumulated and biomagnified through the food web. Consumption of aquatic products such as fish is considered one of the critical routes of HMs exposure in human beings (Jia et al., 2017). These pollutants enter the food web and bioaccumulate from low trophic to higher trophic levels (Jia et al., 2018). The bioaccumulation of HMs in different fish depends on their concentration and bioavailability in the aquatic environment. Furthermore, adsorption and precipitation processes, complexation kinetics, chemical speciation, lipid solubility, and particulate/water partition coefficients are all influenced by physical and chemical variables. Additionally, biological factors including species, trophic relationships, and biochemical and physiological adaptability are crucial (Bonsignore et al., 2018). Other factors such as age, sex, swimming pattern, living environment, feed habits, and reproductive cycle also play crucial roles in the bioaccumulation of HMs (Hao et al., 2019). In fact, understanding the distribution of this class of contaminants in marine organisms is an important matter of investigation and frequently a crucial challenge for thorough investigation of environmental impact on the marine ecosystem due to variable chemical affinities of metals to fish tissues, different uptake, deposition, and excretion rates (Bonsignore et al., 2018).

Previous studies conducted in China revealed that the contribution of Cd derived from aquatic products in residents' diets was estimated to be 21.5 % in the northeast and 24.8 % in southern coastal cities, while in the case of Pb, it was 58.7 % in Fujian Province. The contribution of As in Zhejiang and Fujian was as high as 78.5 % and 92 % (Li, 2012). The HMs can enter fish bodies through several pathways, such as ingestion, surface contact with sediments, adsorption through the skin, and ion exchange through gills. Therefore these aquatic organisms act as a secondary source of exposure to humans (Dang et al., 2016). Human exposure to these HMs based on their concentration in different environmental media and food products is hard to assess, therefore bio-monitoring using human bio-materials can be of key importance to evaluate the nutritional status and occupational or environmental exposure (Oyoo-Okoth et al., 2010).

Hair is one of the most important biomaterial used in biomonitoring studies as it can reflect the physiological status and impact of environmental factors on humans (Ali et al., 2019a, 2019b). Furthermore, it has the advantage over other biomaterials due to easy sampling, transport and storing HMs information for longer (Zhou et al., 2016). Human hair is mainly composed of fibrous proteins, especially in the form of keratin which makes it more robust and stable. The protein that makes up keratin is helical. In hair, keratin fibers come in two varieties: type I, which contains acidic amino acid residues, and type II, which contains basic amino acid residues. Coiled-coil dimers are created when one strand of type I fiber and one strand of type II fiber spiral together. These dimers then form tetramers by coiling together in an antiparallel fashion (Yang et al., 2014). Previously many studies such as Liang et al. (2017), Wang et al. (2017), Oyoo-Okoth et al. (2010) and Batool et al. (2015) have found a significant association between hair HMs and dietary intake which indicates the reliability of hair as a bio-indicator of HMs entering the human body. Fish is one of the most commonly used aquatic products and it is essential to thoroughly investigate the HMs pollution levels. Previously studies have focused mostly on HMs content, but only a few followed it with biomonitoring to find out the source and accumulation patterns in the exposed population.

The key objectives of the study are to; (1) investigate the

accumulation and distribution of HMs (As, Cr, Cu, Cd, Pb, and Zn) in fish collected from the three study areas; (2) investigate the concentrations of HMs in hair based on age gender groups and fish consumption frequency; (3) evaluate the carcinogenic and non-carcinogenic health risk associated with HMs exposure due to consumption of selected aquatic products.

## 2. Materials and methods

### 2.1. Sampling areas

In this study, a total of 214 fish samples were collected from three coastal cities, Qingdao, Xiamen, and Zhoushan. Details of the study area and sampling sites are presented in Fig. 1. Qingdao is located in the Shandong Province, with a total coastline of about 817 km, 49 bays, and a total sea area of 12,240 km<sup>2</sup>. Qingdao has excellent shipping conditions and plays a vital role in developing China's marine economy and industry. It has rich sea resources, and in 2017, Shandong province produced approximately 7.95 Mt of seafood, which was the highest in the country (China Fisheries Society, 2021). Xiamen is located in Fujian province, with a coastline of about 234 km and a sea area of about 390 km<sup>2</sup>. There are about 157 common fish species found in Xiamen. In 2017, the seafood output in Fujian province was about 6.7 Mt, the second-highest in the country (Li et al., 2013). Zhoushan is located in Zhejiang Province, with a coastline of more than 2444 km and a sea area of 20,800 km<sup>2</sup>. The Zhoushan Fishing Ground is one of the largest fishing grounds in China and is known as the "Fish Capital". Zhoushan's annual output accounts for about one-tenth of the national marine production. In 2017, Zhejiang Province produced approximately 4.9 Mt of seafood, ranking first for seafood production (Hao et al., 2019).

### 2.2. Collection of samples

In this study, 214 fish samples were collected in Qingdao, Xiamen, and Zhoushan from April 2016 to January 2018. Samples were collected randomly based on residents' dietary structure and consumption level. The fish samples were collected using a dissection knife, tissue scissors, tweezers made of stainless steel, and ethanol was used for cleaning before and after use. After collection, fish samples were washed and ice packed in a clean polyethylene bag and brought to laboratory on same day. The edible part of fish (muscles tissue) was cut into small pieces and oven dried at 70–80 °C to attain a constant weight. The dried fish samples were grinded and packed in airtight zip lock bags and stored in refrigerator until further chemical analysis (Kwok et al., 2014; Rajeshkumar and Li, 2018).

In April 2016, 106 fish samples were collected systematically from two large aquatic markets (Donghe Vegetable Market and Nanzhen Vegetable Market) in Zhoushan. The collected samples include 15 kinds of marine fish (small yellow croaker, large yellow croaker, dragon head fish, hairtail, mullet, sea eel, sea bass, silver pomfret, anchovy, brown scorpionfish, yellow catfish, gray mackerel, blue mackerel, Spot mackerel, striped sole, yellowfin mackerel (Table S1).

Samples from Qingdao City were collected in September 2017. A total of 61 fish samples were collected systematically from the three aquatic product markets in Qingdao. The three aquatic product markets were the Chengyang vegetable market, aquatic products wholesale market, Qingdao Shazikou seafood market, and Qingdao aquatic products wholesale market. The collected samples include seven kinds of fish (barracuda, mackerel, plaice, small yellow croaker, turbot, sea bass, and tongue sole) (Table S2).

A total of 47 fish samples from three large aquatic product markets in Xiamen were collected systematically in September 2017. The three aquatic product markets were the central fishing port of Xiamen and Taiwan, Dongdu aquatic products wholesale market, and Xiamen no. 8 aquatic products market. The collected aquatic samples included seven kinds of marine fish (mackerel, large yellow croaker, grouper, sea bass,

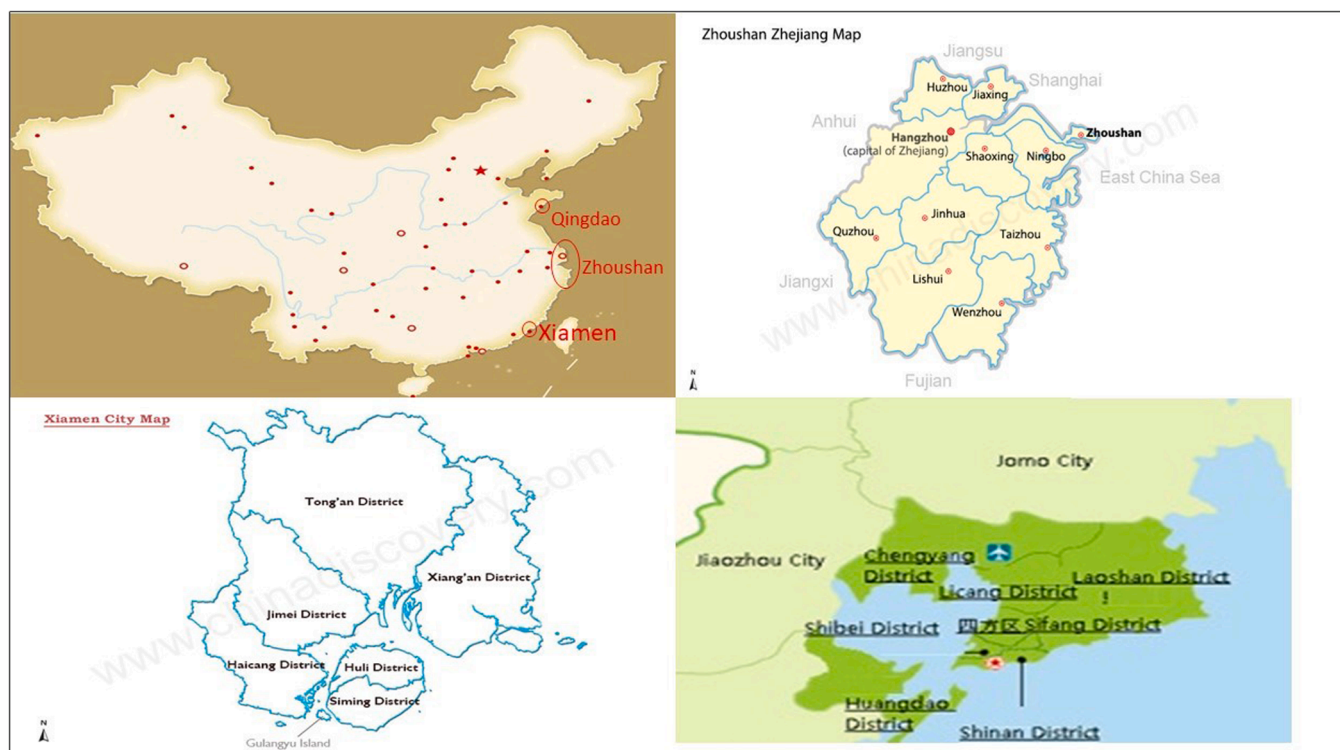


Fig. 1. Study areas.

horse-face puffer, yellowfin bream, and blue trevally). The detailed information on fish samples is shown in Table S3.

### 2.3. Hair sample collection

Ethical approval was obtained from the Institute of Geochemistry Chinese Academy of Sciences and all the participants must sign a consent form. Based on particular inclusion and exclusion criteria. A total of 391 hair samples were collected using USEPA-7473 method. Within 3 cm of the occipital or front hairline, 1–2 g of hair was cut using clean stainless-steel scissors and placed in Ziplock bags for further analysis. The samples were soaked and in 1 % detergent and rinsed using deionized distal water (DDW) two to three times and oven dried at 60 °C before digestion. A questionnaire survey was conducted simultaneously with informed consent. The main content of the questionnaire included height, weight, gender, age, occupation, smoking, drinking, makeup, whether rice is the staple food, and fish consumption. The inclusion criteria were: residents  $\geq 6$  months, age  $> 2$  years old; exclusion criteria: persons with dyed hair and perm within one year, and persons who may be exposed to HMs by occupation. The fish-eating frequency of residents was divided into three groups: high, medium, and low: high group, fish-eating frequency  $\geq 20$  times/month; medium group, fish-eating frequency 5–19 times/month; low group, fish-eating frequency  $\leq 4$  times/month. The demographic characteristics of the surveyed population are presented in Table S4.

### 2.4. Analytical methods

The heavy metal (As, Cd, Cr, Cu, Pb and Zn) concentrations (wet weight) were determined using acid digestion and an inductively coupled plasma mass spectrometer (ICP-MS, NexION 300X, PerkinElmer). The samples were prepared for HMs analysis using the method presented by Qin et al. (2021). Approximately 2 g of sample was weighed and transferred to a Teflon digestion flask along with HNO<sub>3</sub> (purity 65 %; manufacturer HUSHI Ltd) H<sub>2</sub>O<sub>2</sub> (purity 30 %; manufacturer HUSHI Ltd) (5:3). The samples were heated using a hotplate in 4

stages: 1st 120 °C for 30 min, 2nd 140 °C for 20 min, 3rd 160 °C for 60 min, and 4th 140 °C for 100 min. At the end of the digestion process, the samples were left to cool down to room temperature. The solution was then rinsed with three portions of deionized distilled water (DDW), transferred to a 10 mL volumetric flask, and diluted to the scale (Yang et al., 2021). In order to eliminate interference from the solution and reagents, method blank samples were carried out with each set of digestion.

### 2.5. Health risk assessment

#### 2.5.1. Estimated daily intake

In order to evaluate the health risk from HMs due to the consumption of aquatic food, the estimated daily intake (EDI) was calculated by employing the following equation:

$$EDI = \frac{C \times FIR}{BW} \quad (1)$$

where C is the concentration of HMs in the aquatic food samples, FIR is the daily intake of fish (g/day), which was 166 g/person/day for Xiamen and 105 g/person/day for Qingdao and Zhoushan were used in this study (Wang et al., 2021), B.W. represents the average body weight of an adult (58.1 kg) (Qin et al., 2021).

#### 2.5.2. Non-carcinogenic risk

In order to find out the risk due to the consumption of fish, the target hazard quotient (THQ) was calculated using Eq. 2.

$$THQ = \frac{C \times ED \times EF \times FIR}{BW \times RfD \times AT} \times 10^{-3} \quad (2)$$

Where ED is the lifetime exposure duration (70 years), EF is the exposure frequency (360 days/year), RfD is the oral reference dose for different HMs ( $\mu\text{g kg}^{-1} \text{day}^{-1}$ ): current study: (As (0.0003), Cu (0.04), Cr (0.003), Cd (0.001), Pb (0.0015) and Zn (0.03) and AT is representing the exposure duration of non-carcinogens (25,550 days) (USEPA, 2011).

To find out the non-cariogenic of multiple HMs due to fish food consumption, total hazard index (HI) was calculated by adding the THQ values of the HMs using Eq. (3) (Cui et al., 2015).

$$HI = \sum_{i=1}^n THQ \tag{3}$$

THQ assesses human health risks due to individual HMs, while HI is a combined hazard due to all the HMs. If the value of THQ or HI is < 1, there was no significant non-carcinogenic risk. However, if the value is > 1, there is a non-carcinogenic risk (USEPA, 1989).

### 2.5.3. Carcinogenic risk

The cancer risk (CR) due to fish food consumption was estimated using the following Eq. (4) (USEPA, 1989).

$$CR = \frac{C \times ED \times EF \times FIR \times CSF}{BW \times AT} \times 10^{-3} \tag{4}$$

Where CSF is the carcinogenic slope factor, in the current study, CSF values for Cr (6.3), Pb (0.0085), As (1.5), and Cd (0.5) mg kg<sup>-1</sup> day<sup>-1</sup> based on USEPA Integrated Risk Information System database were

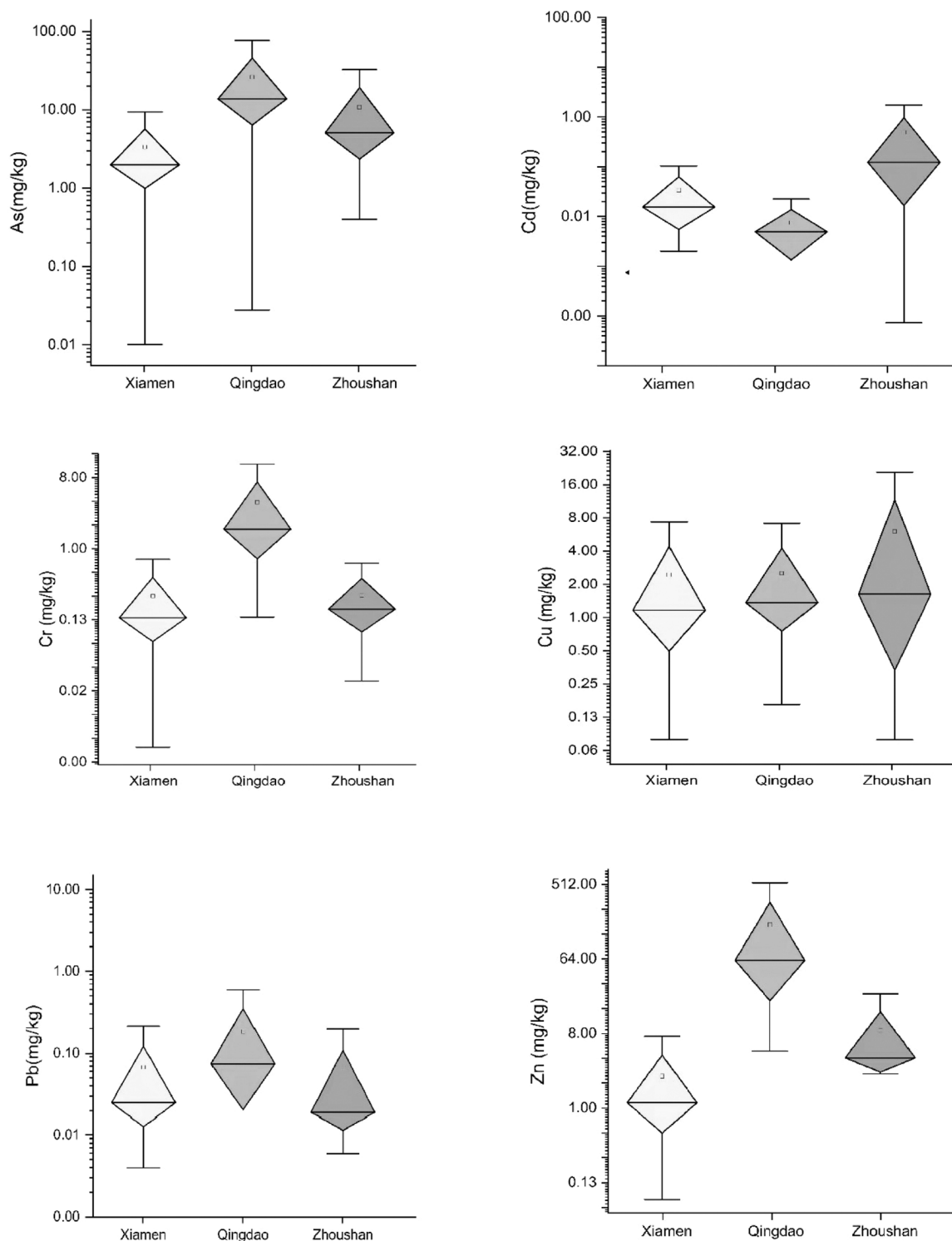


Fig. 2. Heavy metal concentrations in different fishes in three Cities.

used. If the CR value is  $> 1 \times 10^{-4}$  represents a carcinogenic risk, and if  $1 \times 10^{-6} < CR < 1 \times 10^{-4}$ , it shows acceptable levels of cancer risk, while  $CR < 1 \times 10^{-6}$  indicates negligible risk (USEPA, 2015).

## 2.6. Quality control and data precision

Strict quality control was carried out throughout the experiment to ensure the method's accuracy and reliability. Certified reference materials (fish tissue, NRC-Tort3, National Research Council of Canada) were used during each experiment batch to ensure accuracy. The replicates and reference sample's relative standard deviations (RSDs) were less than 10%. The recoveries of certified reference materials ranged within 90–110% of the certified values, which shows that the experimental method was accurate and reliable. The limits of detection for the determination of HMs Cd, As, Pb, Cr, Cu and Zn were 0.0011 µg/kg, 0.0263 µg/kg, 0.017 µg/kg, 0.0497 µg/kg, 0.114 µg/kg and 1.43 µg/kg. The descriptive data was analyzed statistically using SPSS 25.0 statistical software and graphs were plotted using Sigmaplot (Systat Software Inc., San Jose, California, USA). All data was tested for normality using the Shapiro-Wilk method. Mann-Whitney test was used to analyze differences between variables; Spearman rank correlation analysis was used for the correlation test.

## 3. Results and Discussion

### 3.1. Characteristics of heavy metal content in fish

A total of 214 fish samples were collected in this research. Among the cities, the overall concentration of HMs in fish was highest in Zhoushan city, while the lowest concentrations were found in the case of Xiamen. In the case of Qingdao, the arithmetic means of element concentration showed an ascending order of  $Cd < Pb < As < Cr < Cu < Zn$ . The order was found to be the same for Zhoushan and Xiamen ( $Cd < Pb < Cr < Cu < As < Zn$ ). Overall, Zn was found to be the highest among all the fish species, while the lowest concentrations were observed for Cd and Pb. The Cr contents in Mackerel (*Scomberomorus niphonius*) from Qingdao city were relatively high and exceeded the national safety standard. Detailed information on the heavy metal content of fish in different regions is presented in Fig. 2.

According to the living environment, fish are divided into marine fish and freshwater fish. The means of Cd and As concentration in marine fish were  $0.0081 \text{ mg kg}^{-1}$  and  $4.65 \text{ mg kg}^{-1}$ , which were significantly higher ( $p < 0.05$ ) than the mean content of Cd ( $0.0007 \text{ mg kg}^{-1}$ ) and As ( $0.117 \text{ mg kg}^{-1}$ ) in freshwater fish. Marine fish have a longer food chain and can easily accumulate HMs. The means of Cr concentration in mackerel (*Scomberomorus niphonius*) from Qingdao was  $0.743 \pm 0.981 \text{ mg kg}^{-1}$ , and 10% (1/10) of the samples exceeded the limit of  $2.0 \text{ mg kg}^{-1}$  set by China "National Food Safety Standard (GB 2762–2017). Fish normally uptake the HMs from water through ingestion of suspended particles, food web, and ion exchange during breathing, which may explain the lower levels of HMs in fish samples (Han et al., 2021). Heavy metal concentrations were found to be higher in marine fish than in freshwater fish. The results were consistent with those previously presented by Pragnya et al. (2021). The Cd and As contents of marine fish were significantly higher than those of freshwater fish ( $p < 0.05$ ), which may be attributed to their longer food chain, and adsorption of metals from surrounding water and sediments (Han et al., 2021). The comparison between HMs content presented in current study and those reported previously is given in Table S7.

### 3.2. Impact of sizes and feeding habits on fish heavy metal

Body length and weight are important factors that affect the metal content of fish (Bonsignore et al., 2018). A significantly positive correlation ( $p < 0.05$ ) between Cd and Pb concentration and body length of tonguesoles (*Cynoqlossus robustus Gunther*) in Qingdao was observed.

The Cu contents of hairtail (*Trichiurus lepturus*) in the Zhoushan area were positively correlated to body length ( $p < 0.05$ ), while Cr contents showed a positive correlation to the bodyweight of pufferfish (*Thamnaconus hypargyreus*) in the Xiamen area. The weight of the large yellow croaker (*Pseudosciaena crocea*) was positively correlated to Zn content ( $p < 0.05$ ). In the case of other fish samples, no obvious correlation was observed between the heavy metal and body weight or body length.

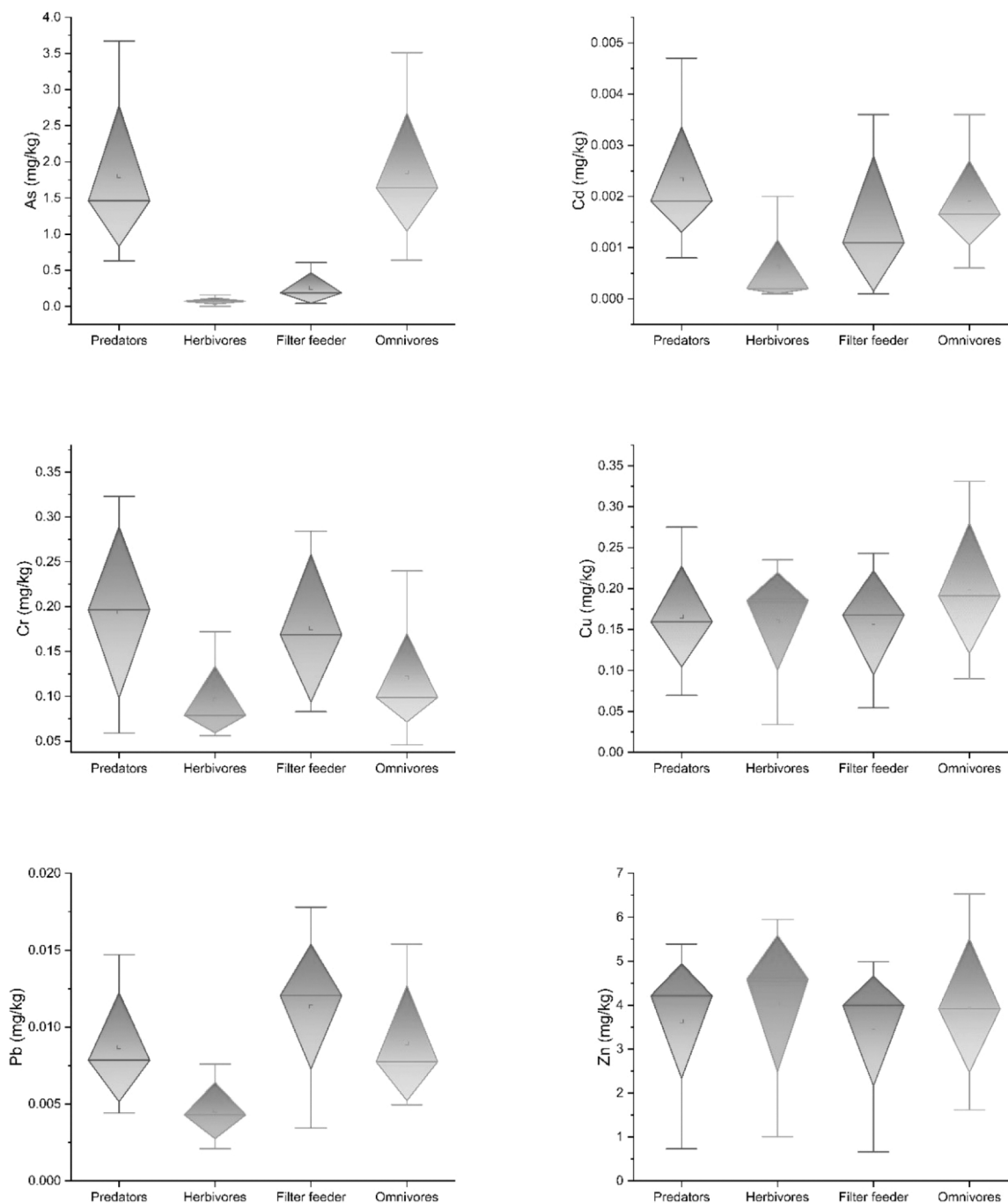
Body length and weight are not only a measure of the fish's body size but also one of the main factors that affect the metal content of a fish body. Only some fishes showed a positive correlation between heavy metal content and body length/weight in this study. The results were consistent with those previously presented by Jia et al. (2017). The HMs concentration in the fish body is controlled by the balance between uptake and elimination, which is influenced by various factors such as habitat, age, feeding behavior, and location. Higher accumulation of HMs was reported in younger fish due to their ingestion rate and metabolic activities compared to older ones. However, continuous accumulation of HMs from the surrounding environment results in elevated metal concentration with increasing fish weight and length (Yi and Zhang, 2012). This also may be due to the fact that the fish samples from various regions were collected and purchased in different aquatic product trading markets. Each fish's growth environment may differ, so the HMs content of some fish in this study showed no significant correlation with their body length and weight. Previous studies also found a negative correlation between HMs content and fish body weight and length. Even if the same fish grows in waters with different pollution levels, the degree of enrichment of HMs in the body will vary depending on the environment (Garai et al., 2021).

The collected fish were divided into different categories based on their feeding habits, such as carnivorous, filter-feeding, omnivorous, and herbivorous. The detailed information is presented in Fig. 3. In this study, the mean concentration of HMs in fishes with different dietary types showed a decreasing order of carnivorous > omnivorous > filter-feeding > herbivorous. Carnivorous fishes are more likely to accumulate HMs due to their higher nutritional level. Overall, the contents of Cd and As in carnivorous and omnivorous fish were significantly higher than those in filter-feeding and herbivorous fish ( $p < 0.05$ ), and the Pb contents of herbivorous fish were significantly lower ( $P < 0.05$ ) than those in carnivorous, omnivorous and filter-feeding fish.

Feeding habits are also an important factor affecting the content of HMs in fish. In this study, the average content of Cd, As, Pb, Cr, Cu, and Zn of the various feeding fishes were in the order of carnivorous > omnivorous (filter-feeding) > herbivorous, which is consistent with the results of previous studies (Jia et al., 2018). Generally, the heavy metal content of fish with high trophic levels is higher than that of fish with low trophic levels. The diet of carnivores and omnivores mainly includes small fish, aquatic insects, crustaceans, algae, and detritus that lead to high accumulation and biomagnification of HMs. Conversely, herbivores are primary consumers, and their diet includes aquatic macrophytes, plants, and algae. Being at lower trophic levels, herbivores' diet includes fewer items than carnivores and omnivores. Therefore, the biomagnification of HMs is lower in the case of herbivores (Hashim et al., 2014).

### 3.3. Distribution of heavy metals in male and female hair

The heavy metals distribution in male and female hair samples from Qingdao, Xiamen, and Zhoushan is presented in Fig. 4. The descending order of HMs in both male and female hair followed an order of  $Zn > Cu > Pb > Cr > As > Cd$ . In both males and females, the highest mean values were noticed in the case of Zn and Cu, while the lowest values were noticed in the case of Cd and As. The order was also found to be similar in the other two study areas, Xiamen and Zhoushan. The concentration of Zn in 28% of the subjects (37/130) from Qingdao exceeded the recommended values in China. The proportion of people with excessive Cr and Zn content was relatively large, indicating that



**Fig. 3.** Distribution of heavy metal concentrations in different feeding fishes. The concentration of As, Cd, Cr followed a descending order of carnivorous>omnivorous>filter-feeding>herbivorous, while Cu was found to be high in omnivorous, Pb in filter feeders and Zn in herbivorous.

residents may have a certain risk of exposure to Cr and Zn. Zinc is an essential and second most abundant element in the human body. It plays a vital role in proper functioning alcohol dehydrogenase, carbonic anhydrase, superoxide dismutase, leucine aminopeptidase, metalloenzymes, etc. However, excess of Zn can cause coronary artery disease, anemia, metal fume fever and artery disease (ATSDR, 2005; Waseem and Arshad, 2016). The Cr concentration above permissible limits can lead to various diseases, such as asthma, nerves, kidney and circulatory malfunctions (ATSDR, 2012b). The current results showed that the Cu content in 36 % (181/508), Zn content in 10 % (49/508) and Cr content (70/390) in the studied population was less than the recommended values. It is worth noting that Cr, Cu, and Zn are also elements necessary for the human body and insufficient intake can harm human health.

Overall, the concentration was mostly found to be high in females. Previously Ali et al. (2019a), (2019b) and Liang et al. (2017) also reported that for most of the elements, the concentration was higher in female hair than male. However, González-Muñoz et al. (2008) and Li et al. (2020) reported higher concentrations in male hair samples. The variation between the current study and previous literature can also be attributed to several factors, such as geographical variation, analytical approach, methods, and environmental factors (Sahoo et al., 2015).

#### 3.4. Age-wise distribution of hair heavy metals

The age-wise distributions of hair HMs in Qingdao, Xiamen and Zhoushan are presented in Fig. 4. The results illustrate that the Zn

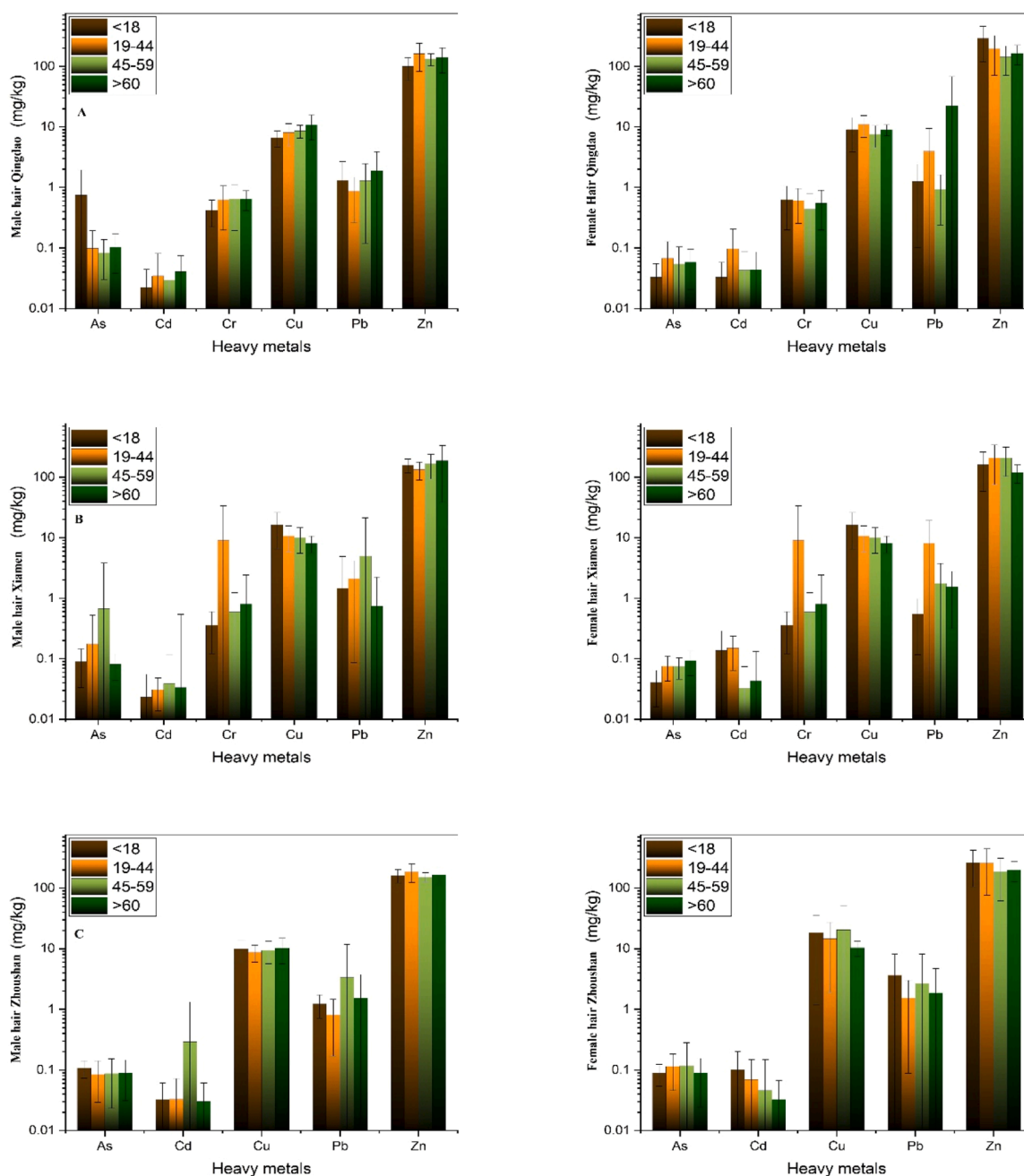


Fig. 4. Heavy metals concentration in male and female hair samples from Qingdao (A) and Xiamen (B) and Zhoushan (C) based on gender and age groups.

concentration in males and As and Cu in females was highest in age 19–44 and > 60, followed by 45–59. The Cd content in males and Pb in females was found to be the highest in the case of age group > 60, followed by 19–44 and 45–59. The content of Cr and Cu in males was normally high in middle and old age groups (19–44 and 45–59). In female subjects Cr and Zn were found to be highest in age groups < 18 and > 60. In Xiamen, the As and Cd concentrations were significantly high in middle-aged groups (19–44 and 45–59) in males, while in females, the concentrations were high in age groups > 60 and 19–44. In the case of both males and females the highest content of Cr was noticed for the age group 19–44. A decreasing trend was noticed in the case of Cu with increasing age. The concentration of Pb was found to be high in age groups 19–44 and 45–59 for both male and female. For male the Zn concentration was highest in age group > 60, while in female middle age groups were the most vulnerable. In Zhoushan, the As concentration for male was found to be high in the age group < 18, while in female the

content was high in middle-age groups. The Cd content from male was high in age group 45–59, while in female the concentration significantly decreased with age. In the case of Pb, the 45–59 and < 18 age groups were the most vulnerable for both males and females. The lowest concentrations were noticed in the young age group (<18).

The normal concentration of As in the human body is 3–4 mg, which increases with increasing age regardless of gender (ATSDR, 2007b, 2006). Ali et al. (2019a), (2019b) reported that the lowest concentration of As was noticed in the age group > 20, and it significantly increased with increasing age, with high values in the age above 40. Wang et al. (2017) reported that the concentration of Zn and Cu increased significantly with age. Furthermore, the study stated that the Cu concentration increased from 10.03 mg kg<sup>-1</sup> in teenagers to 20.25 mg kg<sup>-1</sup> in a population above 59. However, Khalique et al. (2005) stated that the Zn concentration increased with age and decreased in the case of male. The high Zn concentration in the middle-aged group may also be attributed

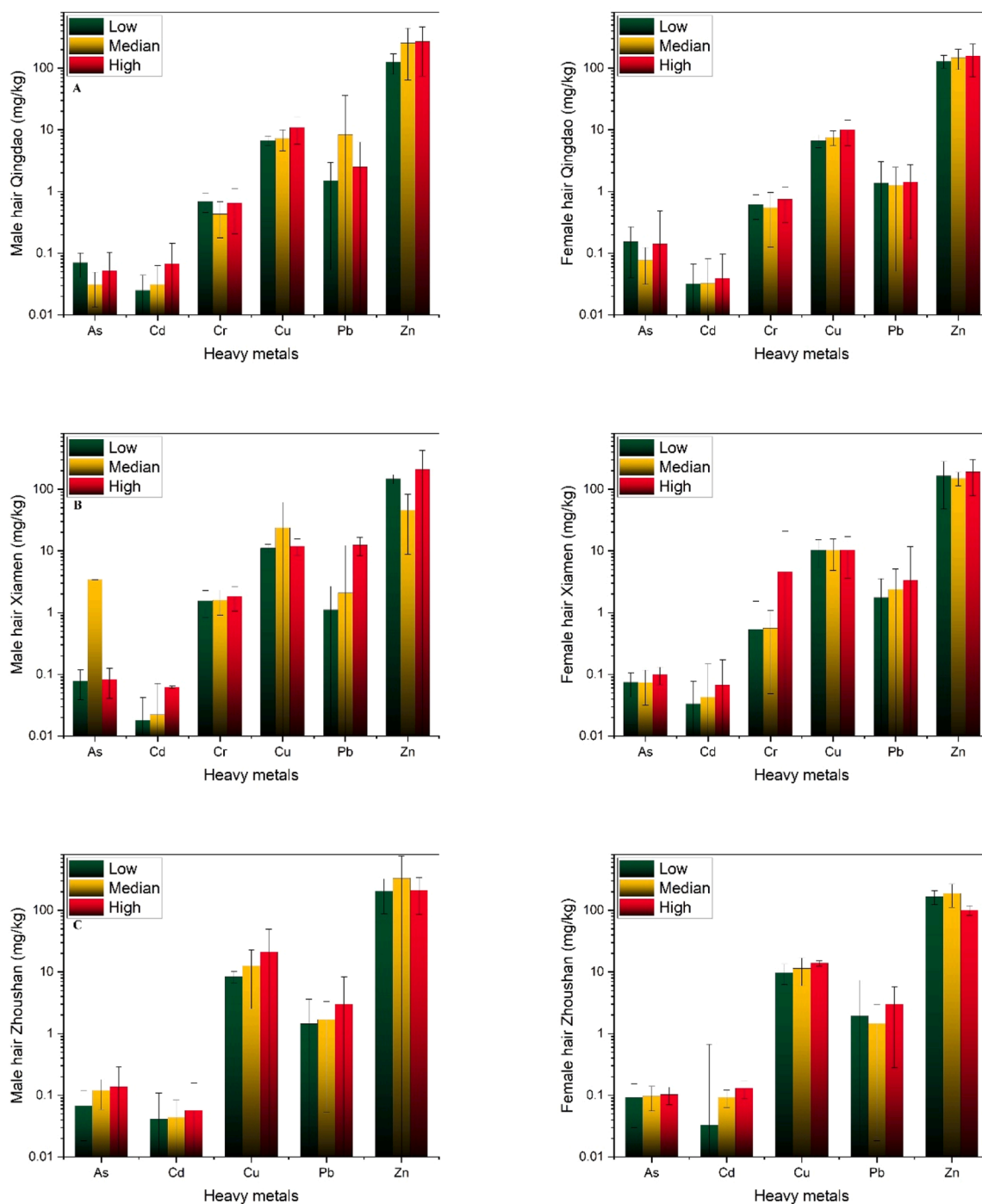
to increased puberty demands, outdoor activities and exposure (Afridi et al., 2006; Zhu et al., 2018). In the case of Cd and Cr, the results were consistent with those presented by Wang et al. (2017). According to their results, the hair Cd content increased from 0.14 mg kg<sup>-1</sup> in teenagers to 0.33 mg kg<sup>-1</sup> in middle and old-age groups. Another study reported that the Cd content increases significantly with age, reaching the maximum value at 25–45 and decreasing slightly in old age (Szynkowska et al., 2009). The Cr concentration was high in young and middle age groups and decreased by 49 % at old age. The variation in HMs in male and female hair may be attributed to cumulative behavior of HMs, dietary habits and lifestyle, consumption of refined carbonate by males,

and hormonal changes in female (Davies et al., 1997).

For most elements, the HMs concentration was within the permissible limits except for some. In Qingdao, Cd concentration in female (<18), Zn in female 19–44 and Pb in female 45–59 exceeded the permissible limit (Cd >0.6 mg kg<sup>-1</sup>, 1120–210 mg kg<sup>-1</sup> and Pb <10 mg kg<sup>-1</sup>). In Zhoushan, Zn in female age group <18 and <60 exceeded the permissible limits.

### 3.5. Hair HMs and fish-eating frequency

Residents' fish-eating frequency was divided into three categories:



**Fig. 5.** Distribution of heavy metals in male and female hair from (A) Qingdao and (B) Xiamen and (C) Zhoushan based on fish consumption frequency. Note: The concentration of Cd, Cu and Zn in Qingdao and Cd, Cr and Pb in Xiamen followed a sequence of low < medium < high for both male and female. In Zhoushan, As, Cd, Cu and Pb in male and As, Cd and Cu in female followed a sequence of low < medium < high.



high, medium, and low: high group, fish-eating frequency  $\geq 20$  times/month; medium group, fish-eating frequency 5–19 times/month; low group, fish-eating frequency  $\leq 4$  times/month. The concentrations of HMs in different age groups based on fish consumption frequency are presented in Fig. 5. In Qingdao, the content of Cd, Cu and Zn in the case of male and As and Cu in female hair showed an increasing trend with an increase in fish-eating frequency (high>low>medium). The trend for Cd, Cr, and Pb in female hair was high>low>medium. In Xiamen, for male the content of Cd, Cr and Cu showed an increasing trend with an increase in fish-eating frequency. For, female all the studied elements except for Zn showed an increasing trend (high>low>medium). In Zhoushan, the As and Cu content in male showed an increasing trend with an increase in fish-eating frequency (High>medium>low), while in the case of female a similar trend was noticed for As, Cd and Cu. In case of Cd, Cr and Pb in male and Pb in female the trend was high>low>medium.

A significant correlation was noticed in case As ( $r = 0.221$ ) and Cr ( $r = 0.231$ ) and fish eating frequency. While in the case of Cu, a slightly high correlation with fish-eating frequency was noticed ( $r = 0.117$ ) (Table 1). Among HMs, a significant correlation was noticed between Cd and Pb, Cd and Cu, Pb and Cu, and Cd and Zn, indicating that the accumulation of these HMs in hair follows the same physiological pathways and exposure sources.

Except As, Cu and Zn, there was no effect of fish-eating frequency on heavy metals detected among the population. The odds ratio greater than 2.0 for As, Cu and Zn shows that with high fish-eating frequency, the probability of Cu and Zn contamination will increase two times. It is indicated from the logistic regression analysis that low and medium frequency of fish-eating has less impact on heavy metal contamination; however, when the population indulges in highly frequent fish-eating habits, the likelihood of metal contamination is expected to rise for most of the metals such as Cd and Cr (apart from As, Cu and Zn) due to positive values of the regression coefficient (beta) (table S5).

The results demonstrated a significant association between fish-eating frequency and age ( $p = 0.05$ , Chi-Squair). However, no association was noticed in the case of gender (Table S6). A significant association was noticed in the case of smoking, indicating that smoking can be a possible exposure source.

### 3.6. Health risk assessment

#### 3.6.1. Estimated daily intake, total hazard quotient and hazard index

The estimated daily intake is presented in Table 2. The relative mean of the EDI values showed an ascending order of Cd < Pb < Cr < Cu < As < Zn. The highest mean EDI values were observed in the case of Zn ( $4.84 \mu\text{g kg}^{-1} \text{day}^{-1}$ ) and As ( $1.38 \mu\text{g kg}^{-1} \text{day}^{-1}$ ). The total hazard quotient (THQs) and hazard index (HI) of HMs in different fish from the three study areas is presented in Table 3. In all three study areas, the THQ values of As, Cu, Pb and Zn were < 1, indicating no significant non-carcinogenic risk due to fish consumption. However, a significant non-carcinogenic risk was found in the case of Cr (1.09), especially through consuming fish in Xiamen. Overall, The THQ values for all the elements due to fish consumption were < 1 except for Cr in Xiamen. The HI values indicated a significant non-carcinogenic risk through fish

consumption in Xiamen (1.55) and Zhoushan (1.164). Overall, the results of THQ and HI indicated a significant non-carcinogenic risk due to fish consumption.

In the current study, no significant non-carcinogenic risk was found to be associated with the studied HMs in all the study areas except for Cr in fish from Xiamen. The results were consistent with previous literature, which showed negligible non-carcinogenic risk (Sharifian et al., 2022). The higher risk of Cr in Xiamen through fish consumption may be attributed to the fact that Xiamen is a coastal area and the consumption of fish by the locals is common due to its ready and easy availability, which may lead to higher exposure to Cr. In the current study, the THQ and HI values, especially in the case of Cr in fish, were high, indicating a higher chance of non-carcinogenic risk in the exposed population. In order to reduce the risk, precautionary measures should be taken, and consumption should be minimized.

#### 3.6.2. Total carcinogenic risk (CR)

The total carcinogenic risk (CR) from HMs due to the consumption of fish is presented in Table 3. The CR values for fish indicate negligible risk. For Pb, the CR values were in the range of  $1.91\text{E}03$  to  $6.08\text{E}03$ , which indicates slight to acceptable cancer risk ( $\text{CR} < 1 \times 10^{-6}$ ). The CR values were found to be in the range of  $1.64\text{E}-04$ – $6.7\text{E}05$  in the case of Cr, which indicates a slight cancer risk through consumption of fish in Xiamen ( $1.64\text{E}-04$ ) and Zhoushan ( $1.16\text{E}-04$ ), while the rest of the values were well within the acceptable limits of cancer risk, which indicates negligible risk. For As the CR values were found to be in the range of  $4.09\text{E}-07$ – $6.08\text{E}-03$ . The CR values for As through fish consumption in Xiamen, Qingdao and Zhoushan were  $6.08\text{E}-03$ ,  $1.89\text{E}-03$  and  $5.54\text{E}-03$ , respectively, indicating the prevalence of carcinogenic risk. The study results indicate that the chance of cancer development due to Cd, Pb and Cr exposure was acceptable to negligible range through fish consumption. However, As posed a significant carcinogenic risk through fish consumption in all study areas.

In this study, a significant carcinogenic risk was found to be associated with As exposure through the consumption of fish. The study's results were consistent with those previously presented by Wang et al. (2020) and Liu et al. (2015). In this study, the carcinogenic risk due to Pb, Cd, and Cr exposure was negligible; however, significant cancer risk was associated with As exposure through fish consumption. The toxicity of As depends on its chemical form, and it is widely reported to cause adverse health impacts in humans. Chronic exposure to As may result in adverse health impacts such as cancer, neurological disorders, and cardiovascular problems. Although the cancerous effects of As are not very clear, it is believed that intracellular biosynthesis of reactive oxygen species that is mediated by As exposure may lead to DNA damage and cause carcinogenic impacts (NRC, 2001). To minimize adverse health impacts, the government and environmental agencies should implement proper awareness, protocols, and regulations.

## 4. Conclusions

The HMs concentration in Qingdao was Cd<Pb<As>Cr<Cu<Zn, while in Zhoushan and Xiamen the order was Cd<Pb<Cr<Cu<As<Zn. The Cr concentrations in Spanish mackerel from Qingdao City exceeded

**Table 1**

Correlation analysis among variables pertaining to heavy metals and fish eating frequency.

Heavy metals	Fish Eating Frequency	As	Cd	Cr	Pb	Cu	Zn
As	0.221**	1					
Cd	-0.039	0.002	1				
Cr	0.231**	-0.014	-0.032	1			
Pb	-0.002	-0.004	0.299**	-0.013	1		
Cu	0.117*	-0.022	0.190**	0.001	0.142**	1	
Zn	-0.102*	-0.033	0.029	-0.019	-0.001	0.296**	1

Note: level of significance for correlation \*  $p < 0.05$ ; \*\*  $p < 0.01$ .

Correlation between heavy metals detected in hair samples with population's fish eating frequency tapped on three point scale (1 high; 2 medium & 3 low).

**Table 2**  
Estimated daily intake (EDI) of heavy metals through consumption of aquatic food.

Study area	Sample	Estimated daily intake (EDI, µg/kg/day)					
		Cd	As	Pb	Cr	Cu	Zn
Xiamen	Fish	5.71E-03	4.23E+00	8.57E-03	3.43E-01	4.57E-01	1.18E+01
Qingdao		3.61E-03	1.32E+00	2.35E-02	1.41E-01	6.69E-01	9.85E+00
Zhoushan		7.23E-03	3.85E+00	2.35E-02	2.42E-01	3.61E-01	8.93E+00

**Table 3**  
Non-carcinogenic and carcinogenic risk of heavy metals through consumption of fish.

Study area	Total hazard quotient (THQ)						HI
	Cd	As	Pb	Cr	Cu	Zn	
HMs							
Xiamen	5.48E-02	1.35E-02	5.48E-03	<b>1.10E+00</b>	1.10E-02	3.76E-01	<b>1.56E+00</b>
Qingdao	3.47E-02	4.21E-03	1.50E-02	4.51E-01	1.60E-02	3.15E-01	8.35E-01
Zhoushan	6.93E-02	1.23E-02	1.50E-02	7.74E-01	8.66E-03	2.85E-01	<b>1.16E+00</b>
Carcinogenic risk (CR)							
HMs							
Xiamen	3.45E-05	<b>6.08E-03</b>	6.99E-08	1.64E-04	//	//	//
Qingdao	2.18E-05	<b>1.89E-03</b>	1.91E-07	6.76E-05	//	//	//
Zhoushan	4.37E-05	<b>5.54E-03</b>	1.91E-07	1.16E-04	//	//	//

the food safety standards. In all three study areas, the HMs in male and female hair followed a descending order of Zn>Cu>Pb>Cr>As>Cd. The high content of Zn in 28 % population needs attention even if Zn is an essential element. The odds ratio for hair As (2.89), Cu (2.23) and Zn (2.041) indicate that with high fish-eating frequency, the probability for these HMs accumulation in hair will increase two times. The study provides evidence that fish consumption frequency can strongly predict HMs accumulation in the human body, and biomonitoring is a helpful tool to assess the HMs internal burden and external pollution. In all the study areas, THQ and HI values showed acceptable to negligible non-carcinogenic risk from all studied HMs exposure, except Cr exposure through consumption of fish in Xiamen. The CR values were under the permissible limits for all the studied elements, except for As in all the study areas indicating chances of developing cancer. Therefore, avoiding excessive consumption of these fish is suggested to minimize the health impacts due to HMs exposure. Furthermore, the government and environmental agencies should regularly carry out pollution monitoring of water bodies and fish products, and awareness plans and necessary preventive measures should be implemented. This study can provide important guidance for the dietary intake of fish and the risk control of HMs exposure in typical aquatic cities in China.

### Ethical approval

The study entitled "Human biomonitoring of heavy metals exposure in different age- and gender-groups based on fish consumption patterns in typical coastal cities of China" conducted by Prof. Ping Li's group has obtained the ethics approval by the Institute of Geochemistry, Chinese Academy of Sciences. All participants in the study were agreed to take part in the survey and to sign a consent form.

### CRediT authorship contribution statement

**Muhammad Ubaid Ali:** Methodology, Writing – original draft; Writing – review & editing. **Chuan Wang:** Data curation, Methodology. **Yuan Li:** Data curation, Methodology. **Xingang Jin:** Data curation, Methodology. **Shaochen Yang:** Data curation, Methodology. **Li Ding:** Data curation, Investigation, Methodology. **Lin Feng:** Data curation, Methodology. **Bo Wang:** Data curation, Methodology. **Ping Li:** Conceptualization, Funding acquisition, Investigation, Methodology, Supervision, Writing – original draft, Writing – review & editing.

### Declaration of Competing Interest

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

### Data availability

Data will be made available on request.

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### Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.ecoenv.2023.115316](https://doi.org/10.1016/j.ecoenv.2023.115316).

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