



Heavy metal distribution and bioaccumulation ability in marine organisms from coastal regions of Hainan and Zhoushan, China

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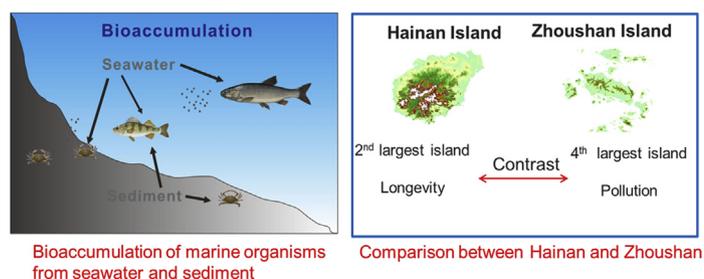
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HIGHLIGHTS

- Across all samples, Zn and Hg had the highest and lowest concentrations, respectively.
- Heavy metal concentrations (except for Zn) were higher in Zhoushan than in Hainan.
- Heavy metal concentrations were higher in crab than in fish.
- BAFs of organisms for heavy metals were higher in seawater than in sediment.
- BAFs of organisms were higher for Cu and lower for Pb.

GRAPHICAL ABSTRACT



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ABSTRACT

We analyzed the distribution and bioaccumulation of six heavy metals (Cu, Pb, Zn, Cr, Cd and Hg) in marine organisms, water, and sediments from China's Hainan and Zhoushan coastal regions. In all marine organism, seawater, and sediment samples, Zn and Hg had the highest and lowest concentrations, respectively. Heavy metals in marine organisms varied by region and species, with concentrations being higher in Zhoushan than in Hainan (except for Zn) and in crab than in fish. Marine organisms' ability to digest and eliminate heavy metals (bioaccumulation ability), based on bioaccumulation factors, was significantly higher for heavy metals in seawater than in sediment. Higher sediment background values may explain the higher heavy metal concentrations in crab. Generally, marine organisms' bioaccumulation ability was higher for Cu and Zn and lower for Pb. Findings specific to the coastal environments may be generalizable to other coastal areas in China or worldwide.

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1. Introduction

Marine organisms are a popular food source for coastal residents (Bosch et al., 2016; Burger et al., 2014; Kwoczek et al., 2006; Rahnuzzaman et al., 2016) and offer numerous health benefits

because they are high in protein and low in saturated fats (Golden et al., 2016; Gu et al., 2018; Kennedy et al., 2009). However, these benefits may be compromised by dangerous levels of contaminants in marine organisms (Burger et al., 2014; Gochfeld and Burger, 2005; Meador et al., 1998), which may contain substantially higher contaminant levels than other foods (Lobet et al., 2003).

Heavy metals are considered a serious global environmental threat (Burger et al., 2002; Wang et al., 2018). Toxic heavy metals such as Pb, Cd, and Hg damage both marine species diversity and ecosystems (Mertz, 1993; Zhang et al., 2012), resulting in potential health risks (Kalantzi et al., 2013; Zhang et al., 2015) and deleterious effects on humans (Fraser et al., 2013; Jarup and Akesson, 2009). However, heavy metal intake is beneficial in small doses, e.g., Zn and Cu are essential for human health, particularly for maintaining normal physiological function (Gu et al., 2017, 2018; Jiang et al., 2015; Kwoczek et al., 2006; Liu et al., 2015). Marine organisms can absorb heavy metals from the surrounding water and sediment as well as through their food (Bosch et al., 2016; Ginsberg and Toal, 2009; Hao et al., 2013; Jiang et al., 2015; Velusamy et al., 2014) and can therefore serve as indicators in the long-term monitoring of metal accumulation (Burger et al., 2002; Gu et al., 2017; Zhong et al., 2015). To some extent, high heavy metal concentrations in marine organisms reflect the surrounding environmental pollution levels. Marine organisms thus represent a direct link between heavy metal contaminants in the environment and the health of coastal residents (Burger and Gochfeld, 2009; Fraser et al., 2013; Rajeshkumar et al., 2018). As such, an understanding of the distribution and bioaccumulation of heavy metals in marine organisms in coastal regions is crucial for scientists and government regulatory agencies.

China is the largest fish producer and exporter in the world (Food and Agriculture Organization, 2012). Hainan and Zhoushan, located in the South and East China Seas, are two of the largest islands in China and export large amounts of seafood (Huang et al., 2007; Li et al., 2017; Liu et al., 2015; Wan et al., 2017; Ye, 2000). However, the different environmental conditions on each island may result in different heavy metal concentrations in marine organisms and associated health conditions of coastal residents. Hainan has a lower level of industrial activity, a higher level of environmental protection, and an older population (Hao et al., 2015). A comparison of these two coastal regions would aid understanding of the distribution and bioaccumulation of heavy metals in marine organisms.

Previous studies have considered heavy metal contamination in marine organisms from the East China Sea (Huang et al., 2007; Wan et al., 2017), and others have focused on heavy metal contamination in marine organisms, seawater, and sediments from southern China's coastal regions (Qiu et al., 2011, 2015; Zhang and Wang, 2012). However, few studies have focused on heavy metal contamination in marine food chains from the environment (water and sediments) to the organisms. Studies that consider heavy metal enrichment levels and accumulation patterns in marine organisms are also lacking. Some studies have shown that heavy metal concentrations and bioaccumulation varied significantly between different marine organisms (Canli and Atli, 2003; Copat et al., 2012; Raknuzzaman et al., 2016; Zhang et al., 2012, 2015). Variations in heavy metal concentration are possibly related to the different living environments, seasons, feeding patterns, and trophic level of marine organisms (Rajeshkumar et al., 2018; Liu et al., 2018).

The response of different marine species and regional environments on heavy metal contamination warrant further investigation. Therefore, the objectives of this study were to (1) measure the distribution of various heavy metals (Cu, Pb, Zn, Cr, Cd, and Hg) in two marine organisms (fish and crab) and in associated seawater/sediments from two marine environments (Hainan and Zhoushan

coastal regions) (2) analyze any categorical (e.g., crab or fish, seawater or sediment); or regional differences in heavy metal distribution; and (3) assess marine organisms' ability to digest and eliminate heavy metals (bioaccumulation ability) based on these differences.

Results from this study will contribute to the understanding of heavy metal distribution and bioaccumulation in marine organisms. Specifically, the results will identify potential factors that affect heavy metal contamination in marine organisms and support the development of targeted response measures for protecting marine ecosystems and public health. Findings specific to marine organisms can support the development of targeted seafood consumption guidelines for humans. Findings specific to coastal environments may indicate the transferability of these findings to other coastal areas in China or globally.

2. Methods

2.1. Study area and sampling

The study area comprised various sampling sites along the coastal regions of two islands: Hainan and Zhoushan (Fig. 1 and Appendix 1). Located in the northern South China Sea, Hainan is the second largest island in China (Li et al., 2017), with an area of $35.4 \times 10^3 \text{ km}^2$ (Hu et al., 2013). Its climate is characterized as seasonal and oceanic tropical, with a mean annual temperature and precipitation of 23.8°C and 1685 mm, respectively. Located in the East China Sea, Zhoushan is the fourth largest island in China, with an area of 476.17 km^2 . Zhoushan has a mean annual temperature and precipitation of 16.2°C and 1273 mm, respectively (Wan et al., 2017; Ye, 2000).

Sample collection methods and analyses were based on the Marine Industry Standards of the People's Republic of China (HY/T 132-2010) for organisms and sediments and the Specification for Marine Monitoring of the State Standard of the People's Republic of China (GB17378-2007) for seawater. Marine organisms were collected at 15 sites along Hainan's coast (near Lingshui) and 14 sites along Zhoushan's coast. Various fish and crab species were collected from the bottom of the sea using a trawling net during the spring and fall of 2016 and 2017. Overall, the numbers of marine organisms sampled were 29 (17 fish and 12 crab) and 27 (17 fish and 10 crabs) in Hainan and 31 (20 fish and 11 crab) and 25 (14 fish and 11 crab) in Zhoushan in spring and fall, respectively. Immediately after sampling, the marine organisms were placed in polyethylene plastic bags and preserved with ice at -20°C for later analysis in the laboratory.

Seawater and sediment samples were collected concurrently with the marine organism samples at each site in the fall. Seawater samples were collected and immediately filtered through a $0.45 \mu\text{m}$ membrane and acidified ($\text{pH} < 2.0$) using HNO_3 , as part of a preliminary analysis to detect Cu, Pb, Zn, and Cd, and HSO_4^- , to detect Hg and Cr. Where the depth was less than 10 m, only surface water was collected, where it was 10–25 m, both surface and bottom water were collected, and where it was 25–50 m, the 10 m water was also collected as well as the surface water. Sediment samples were collected at a depth of 0–5 cm and were stored at -20°C in a freezer for later analysis in the laboratory.

2.2. Laboratory analysis

Before the laboratory analysis, the fish and crab samples were identified to species level according to *Fauna Sinica, Fish Fauna of the East China Sea* and *Fish Fauna of the South China Sea*. The samples were thawed and washed in the laboratory, and the most commonly consumed tissues of each sample (dorsal fin muscles for

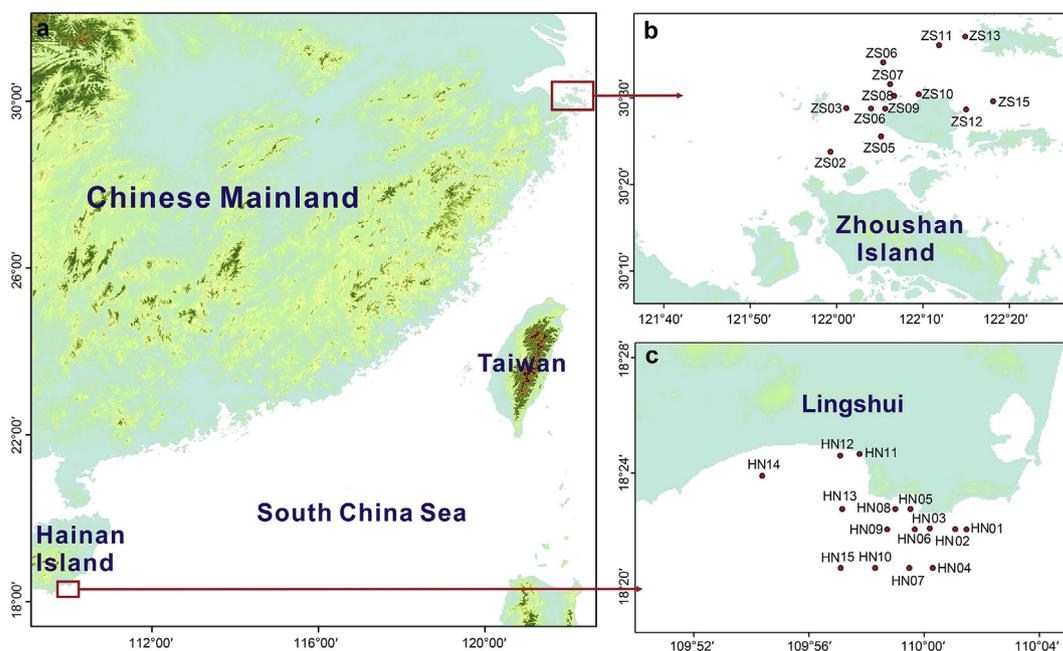


Fig. 1. Study area and sampling sites in Hainan and Zhoushan, China.

fish and abdominal muscles for crab) were dissected by plastic knife and tweezers. Tissues with a wet weight of 1–2 g were weighed and freeze-dried for 24 h until the weight difference were less than 0.5% of the total. Samples of approximately 200 mg were weighed and analyzed using microwave-assisted heating with an $\text{HNO}_3\text{--H}_2\text{O}_2$ (with volume ratio of 4:2) mixture and heated with an electric plate to 120–140 °C to evaporate water and acid.

Similarly, sediment samples were analyzed with an $\text{HNO}_3\text{--HCl}$ (with volume ratio of 4:2) mixture after being dried and ground. Seawater samples were digested after the preliminary analysis to detect Hg and chelated and extracted to detect other metals. Cu, Pb, Cd, and Cr concentrations were determined using flameless atomic absorption spectrometry (XGY-1011A, China). The Zn concentration was determined using flame atomic absorption spectrometry (XGY-1011A, China), and the Hg concentration was determined using atomic fluorescence spectrometry (PinAAcle900T, USA). Operating conditions were clarified and listed in Appendix 2.

2.3. Quality assurance and control

Standard solutions were prepared from stock standard solutions of the metals (Multi-Element Standard, Merck, Germany). The calibration curve demonstrated good linearity for each concentration, with correlation coefficients (r) of greater than 0.99. Accuracy and precision of the analytical procedure were tested with a reference material DORM-2 (dogfish muscle, National Research Council, Canada) for organisms, China Stream Sediment Reference Materials (GBW07314) for sediments, and China Seawater Reference Materials (GBW(E) 080040 and 080042) for seawater. The Cu, Pb, Zn, Cr, Cd, and Hg recovery rates for a standard organism sample were 80.58%, 92.02%, 100.15%, 98.44%, 90.19%, and 117.65%, respectively. Blank and duplicate samples were added every 10 samples. The relative standard deviations of the duplicate samples were less than 10%.

2.4. Statistical analyses

All statistical analyses were performed using SPSS 16.0 for

Windows (IBM, Chicago, IL, USA). Comparative t -tests with a significance level of $p < 0.05$ were performed to detect significant categorical (e.g., crab or frog, seawater or sediment) or regional differences. Cluster analyses were used to detect accumulation patterns in marine organisms and any relevance among heavy metal contaminants. Microsoft Excel 2010 was used to graphically display and visually contrast any observed categorical or regional differences.

2.5. Bioaccumulation factor calculation

The bioaccumulation factor (BAF) has been used to quantify the bioaccumulation of environmental pollutants in previous studies. It illustrates the importance of sediments and water in terms of their influence on aquatic organisms (Sankar et al., 2018; Zhang et al., 2015). In this study, a BAF was used to quantify marine organisms' ability to digest and eliminate heavy metals (bioaccumulation ability); a high BAF suggests a low bioaccumulation ability and vice versa. BAFs for each marine organism sample and studied heavy metal were calculated with Eq. (1) to assess the concentration of heavy metals in the organism (C_{organism}) relative to that in the associated seawater and sediment environment (C_{sediment} or C_{seawater}).

$$\text{BAF} = C_{\text{organism}} / C_{\text{sediment}} \text{ OR } C_{\text{seawater}} \quad (1)$$

3. Results and discussion

3.1. Heavy metal distribution

3.1.1. Seawater and sediments

Heavy metal concentrations measured in seawater and sediments from the Hainan and Zhoushan coastal regions provided the environmental background values for this study. The average depths of seawater were 10.8 m and 14.0 m in Hainan and Zhoushan, respectively. Appendix 3 shows the distribution of heavy

metals in the seawater and sediments from each region. Heavy metal concentrations in seawater followed the following trends in the respective regions: $Zn > Cu > Pb > Cr > Cd > Hg$ and $Zn > Pb > Cu > Cr > Cd > Hg$, and sediment concentrations followed similar trends: $Zn > Cr > Pb > Cu > Cd > Hg$ and $Zn > Cr > Cu > Pb > Cd > Hg$, respectively. In both regions and across both mediums, Zn and Hg had the highest and lowest concentration, respectively. Heavy metal concentrations were generally higher in Zhoushan than in Hainan, except for seawater Zn concentration and overall Cd concentrations, which were higher in Hainan. These results suggest a more favorable environmental background value in Hainan.

The higher Zn concentration in the seawater from Hainan may be attributable to external effects. Compared with stable sediments, flowing seawater is more likely to be affected by environmental and human activities. In particular, rivers are important sources of heavy metal contaminants in seawater. Possible pollutant sources were determined according to the Marine Environment Quality Bulletin and field visit (Appendix 1). According to the 2017 Hainan Marine Environment Quality Bulletin, the total amount of heavy metals flowing into the sea via the Nandu River was 75.5 t, of which Zn accounted for 67.25 t. In addition, Hainan supports an extensive and historic houseboat community (*Danjia*). The bottom coating of these boats, which protect them against corrosion, contain Zn that is released into the sea (Zhou et al., 2007).

3.1.2. Marine organisms

Heavy metal concentrations in marine organisms from the Hainan and Zhoushan coastal regions are summarized in Appendix 4. Among the heavy metals in marine organisms, Zn and Hg concentrations were consistently highest and lowest, respectively, across all regions and categories. High concentrations of both Zn and Cu in marine organisms have also been reported by other studies (Bazzi, 2014; Karadede and Unlu, 2000; Zhang et al., 2015). Nonetheless, heavy metal concentrations in each marine organism sample met the safety standards and guidelines of the Ministry of Public Health of China (2012) and the safety standards of the Joint Food and Agriculture Organization/World Health Organization Expert Committee on Food Additives (Food and Agriculture Organization, 1983).

3.2. Comparative heavy metal distribution in marine organisms

To better understand heavy metal distribution in marine organisms, regional and categorical differences were analyzed. To control for potentially confounding effects, we limited this analysis to marine organism samples collected in the fall season. Overall, more species were collected in the fall, and the same species (*Portunus*) was collected from both Hainan and Zhoushan, enabling a direct comparison between these two regions.

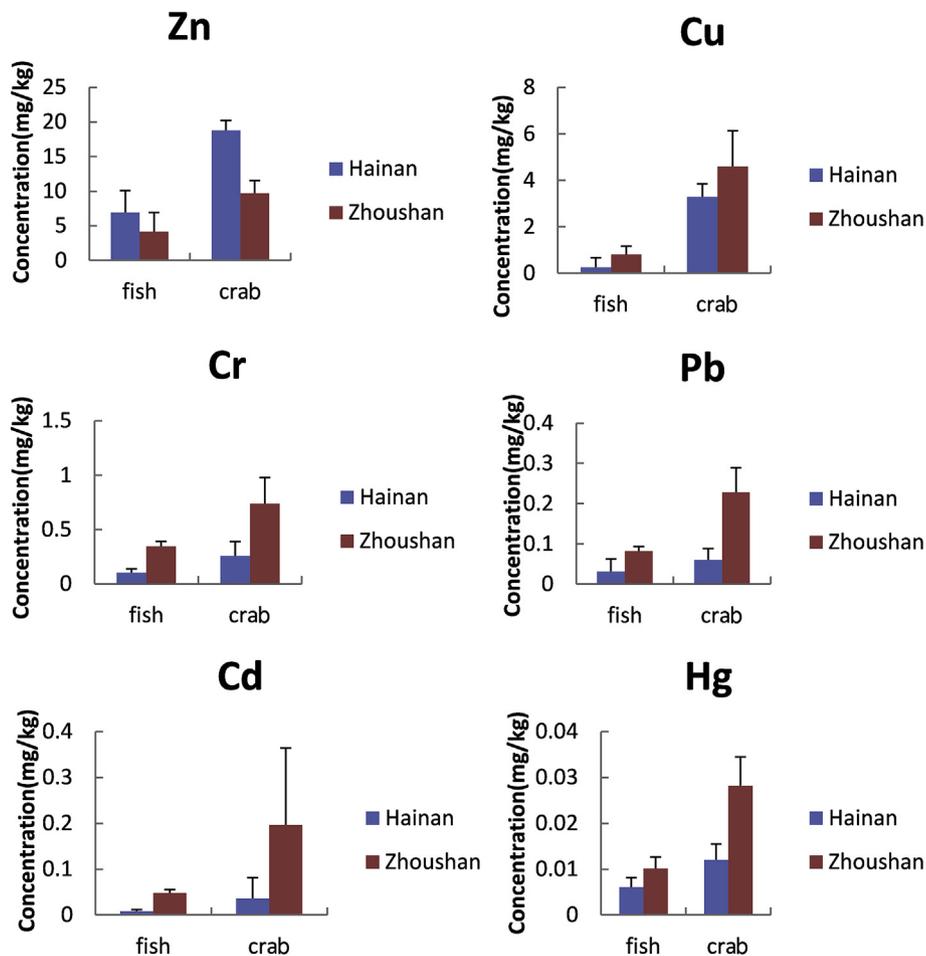


Fig. 2. Comparison of heavy metal concentrations in marine organisms from the Hainan and Zhoushan coastal regions.

3.2.1. Regional differences

Heavy metal concentrations in marine organisms from Hainan and Zhoushan were compared (Fig. 2) and were generally higher in Zhoushan for both crab and fish. Similar to seawater heavy metal concentrations, Zn concentrations were higher in marine organisms from Hainan. All regional differences were statistically significant ($p < 0.05$).

Metal bioaccumulation in marine organisms can seriously affect human health (Zhong et al., 2015) in terms of both quality of life and longevity. However, previous studies have reported that Zn is essential for fish health at low daily doses (Plum et al., 2010). Compared to other metal ions with similar chemical properties, Zn is relatively harmless. In humans, a Zn deficiency may cause aging, disease, or deregulated homeostasis. The moderate intake of Zn from marine organisms may therefore promote health (Plum et al., 2010).

In general, the higher concentration of Zn combined with lower concentrations of all other heavy metals enhanced the quality of marine organisms from Hainan. Based on the same reasoning, the environmental background values in seawater and sediments was more favorable in Hainan. These findings suggest that the surrounding marine environment affects heavy metal distribution in marine organisms. The high quality of marine organisms from Hainan may contribute to better health for its residents.

Cluster analyses were used to detect correlations between heavy metal concentrations in marine organisms. Metals grouped in a cluster likely shared similar sources or analogous transformation and migration processes. Fig. 3 shows the results of this cluster

analysis depicted as hierarchical dendrograms. The heavy metals considered in this study generally grouped in two clusters: Cluster 1 included Zn, and Cluster 2 contained all other heavy metals. These results were generally consistent across both species and region, although Cu was somewhat distinguished from other Cluster 2 elements in the crab samples from Zhoushan. Based on these results, Zn was likely derived from a unique source before being absorbed by the marine organisms. Maritime transport may be a source of heavy metal contamination, as boats made of alloys generally contain both Zn and Cu (Zhou et al., 2007), which may be released into the environment (Zhang et al., 2015).

3.2.2. Categorical differences

Fig. 4(a) and (b) compare the heavy metal concentrations in fish and crab from Hainan and Zhoushan, respectively. In both regions, crab contained higher concentrations of heavy metals than fish species did ($p < 0.01$). Previous studies have also found that crabs (crustaceans) had higher levels of heavy metals (Raknuzzaman et al., 2016; Zhang et al., 2015). Heavy metal concentrations in marine organisms can be affected by their bioaccumulation abilities and their environment (Burger et al., 2014; Canli and Atli, 2003). Crabs and other crustaceans should therefore be considered discrepant aquatic species and, acting as a typical benthic organism, may be good indicators of surface sediment contamination levels (Ololade et al., 2011).

Although the fish samples in this study were collected from the sea bottom, fish typically swim in the middle/upper seawater layers and are thus less affected by sediments. Therefore, from a human

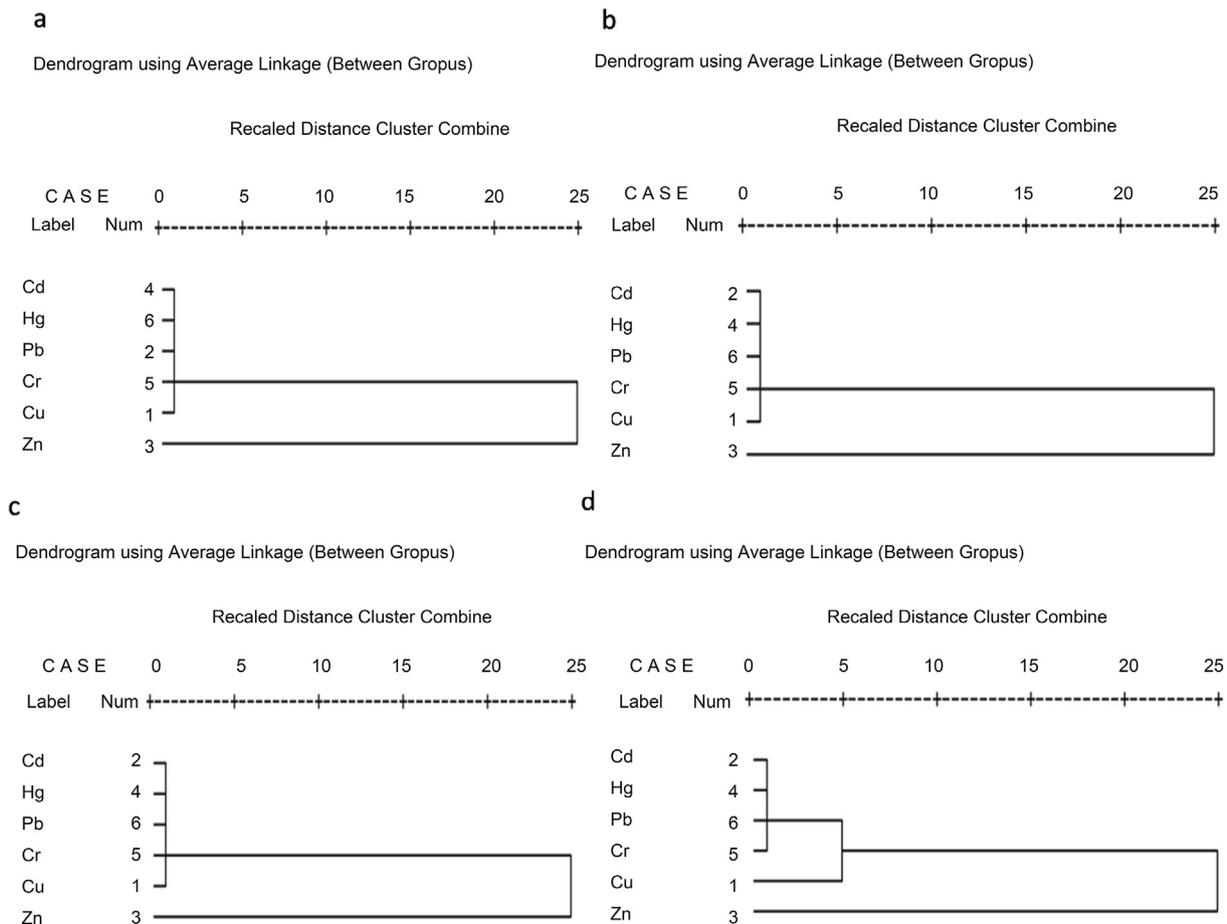


Fig. 3. Hierarchical dendrograms showing the clustering of heavy metals in (a) fish from Hainan, (b) fish from Zhoushan, (c) crab from Hainan, and (d) crab from Zhoushan.

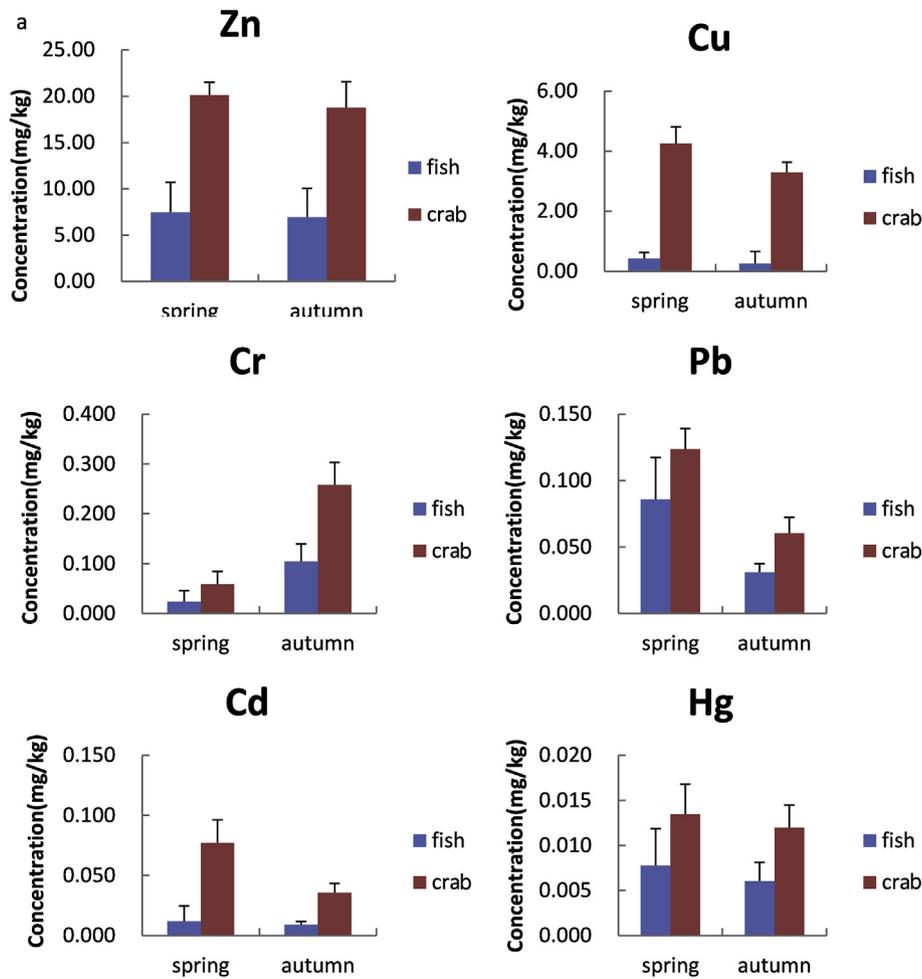


Fig. 4. Categorical differences in heavy metal concentrations from (a) Hainan, (b) Zhoushan, and (c) all samples.

health perspective, it may be safer to eat fish rather than crab, as fish had lower concentrations of heavy metals.

The average values of heavy metal concentrations in fish and crab may obscure important individual characteristics of different species. Fig. 4(c) shows the heavy metal concentrations in 17 different species of fish and crab from the Hainan and Zhoushan coastal regions. Samples of *Portunus* (15 and 16 in Fig. 4(c)) were collected from both regions. *Leiognathus elongatus* (2) from Hainan and *Collichthys lucidus* (11) from Zhoushan had the highest heavy metal concentrations among the fish species. Heavy metal concentrations were generally higher in Zhoushan than in Hainan (except for Zn concentrations, which were higher in Hainan), and in crab than in fish.

Cluster analyses were used to detect correlations among marine organisms with respect to heavy metal concentrations (Fig. 5). Marine organisms were generally grouped into three statistically significant clusters based on their heavy metal concentrations. Cluster 1 included two crab species (*Charybdis anisodon* (14) and *Portunus pelagicus* (15)) and one fish species (*Leiognathus elongatus* (2)) from Hainan. Cluster 2 included two crab species (*Portunus trituberculatus* (16) and *Charybdis japonica* (17)) from Zhoushan and one fish species (*Trichiurus haumela* (3)) from Hainan. Cluster 3 contained all other fish species.

With *Leiognathus elongatus* (2) and *Trichiurus haumela* (3) as

exceptions, Cluster 3 and Clusters 1 and 2, respectively, differentiated fish and crab based on their heavy metal bioaccumulation patterns. Heavy metal concentrations in most fish species did not significantly differ by region, and most fish species were therefore grouped in Cluster 3 regardless of location. Conversely, heavy metal concentrations in most crab species differed according to region and were subsequently split into Clusters 1 and 2.

These findings provided additional evidence that heavy metal distribution and bioaccumulation differ between fish and crab. These differences were exacerbated by seawater and sediment conditions. Crabs, as benthic organisms, may directly indicate regional contamination conditions. Comparatively, the more dynamic environment and feeding habits of fish present more complex species–environment interactions, resulting in inconspicuous regional differences.

3.3. Heavy metal bioaccumulation

BAFs were used to quantify marine organisms' bioaccumulation ability (Table 1). For fish and crab from Hainan, the BAF_{sediment} values followed a similar trend: $Hg > Zn > Cd > Cu > Cr > Pb$ and $Hg > Zn > Cu > Cd > Cr > Pb$, respectively, as did those in Zhoushan: $Hg > Cd > Zn > Cu > Cr > Pb$ and $Cd > Hg > Cu > Zn > Cr > Pb$, respectively. Comparatively, the BAF_{seawater} values differed

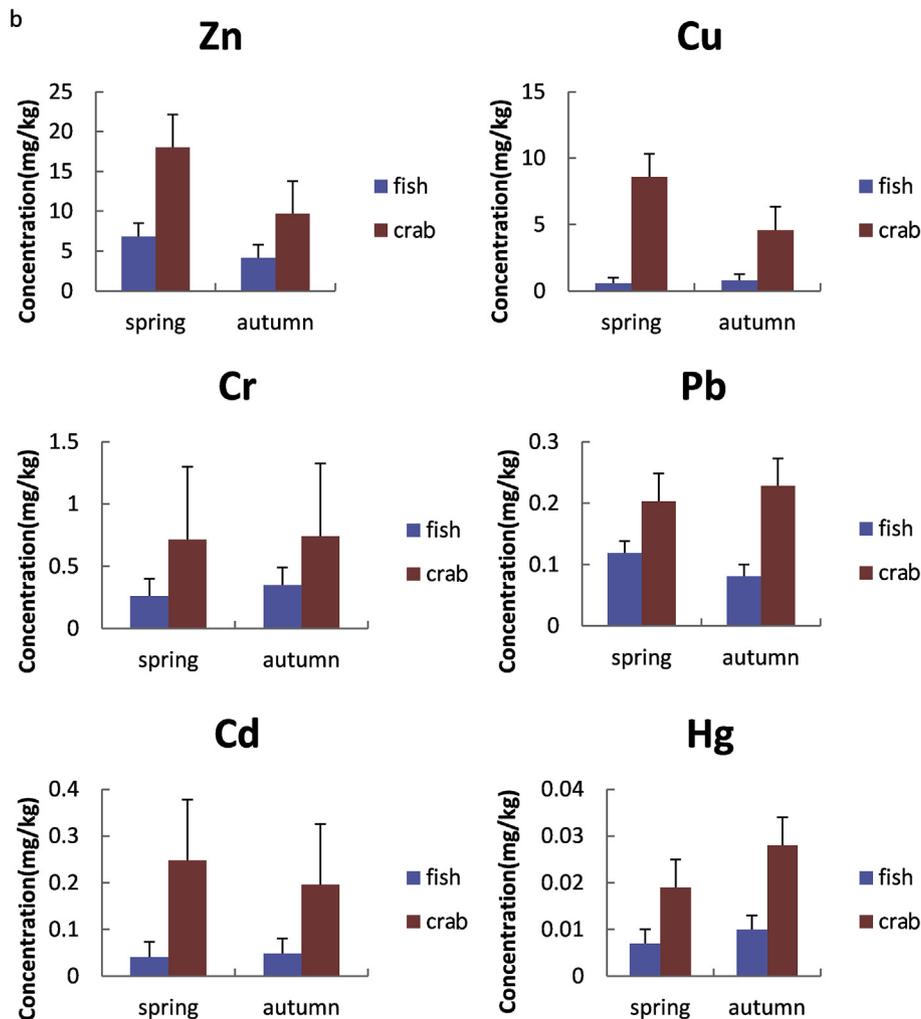


Fig. 4. (continued).

significantly between the organisms: For fish and crab from Hainan, these values had the following trends: Zn > Cu > Hg > Cr > Cd > Pb and Cu > Zn > Hg > Cr > Cd > Pb, respectively; those from Zhoushan had the following trends: Cu > Cr > Zn > Hg > Cd > Pb and Cu > Cr > Zn > Hg > Cd > Pb, respectively.

The marine organisms exhibited different heavy metal bioaccumulation abilities. Marine organisms from Zhoushan had higher BAF values than those in Hainan did for most heavy metals considered in this study, suggesting a lower ability for digesting and eliminating contaminants. Within a certain threshold, marine organisms from environments with higher seawater and sediment heavy metal concentrations generally had higher BAF values. However, Zn was an exception. Marine organisms from Hainan had higher BAF values for Zn, suggesting a lower ability for digesting and eliminating this element. Increased Zn in marine organisms may result in higher Zn intake by humans, potentially improving their health. This finding may explain the noted differences in health and longevity of the Hainan and Zhoushan populations. Within the same environment, the BAF values were higher for crab than for fish. Higher concentrations of heavy metals from both seawater and sediment were able to accumulate in crab.

The BAF_{sediment} values were generally <1, indicating that the bioaccumulation of heavy metals from sediment was not significant (Rumisha et al., 2012). Exceptions included *Portunus trituberculatus*

and *Charybdis japonica* samples that accumulated Cd and Hg, respectively. All other marine organisms in this study, particularly the fish species, exhibited low bioaccumulation of heavy metals from sediment. Regarding heavy metals from seawater, marine organisms may accumulate an element if the BAF_{seawater} values are >1, but this is generally not considered to be significant unless the BAF_{seawater} values are >100 (Yu et al., 2012). The BAF_{seawater} values for Cu, Zn, Cd, and Hg were >100 for all marine organisms in this study.

These findings indicate that heavy metals are more likely to be absorbed by marine organisms from seawater. Seawater quality is thus important and should not be ignored. However, higher heavy metal concentrations in crab, which presumably has more exposure to sediments, suggest that the environmental background value contributed more to heavy metal distribution than bioaccumulation ability did. These results provide another explanation of Zn concentrations in marine organisms and seawater being higher in Hainan than in Zhoushan but lower in sediments overall.

To determine the generalizability of this study's results to other environments, the BAF values were compared with those from other areas (Table 2). We focused on BAF_{seawater} values because of the absorption properties of seawater. Although the BAF values varied according to category and region, similar trends in the heavy metal distribution and bioaccumulation of marine organisms were

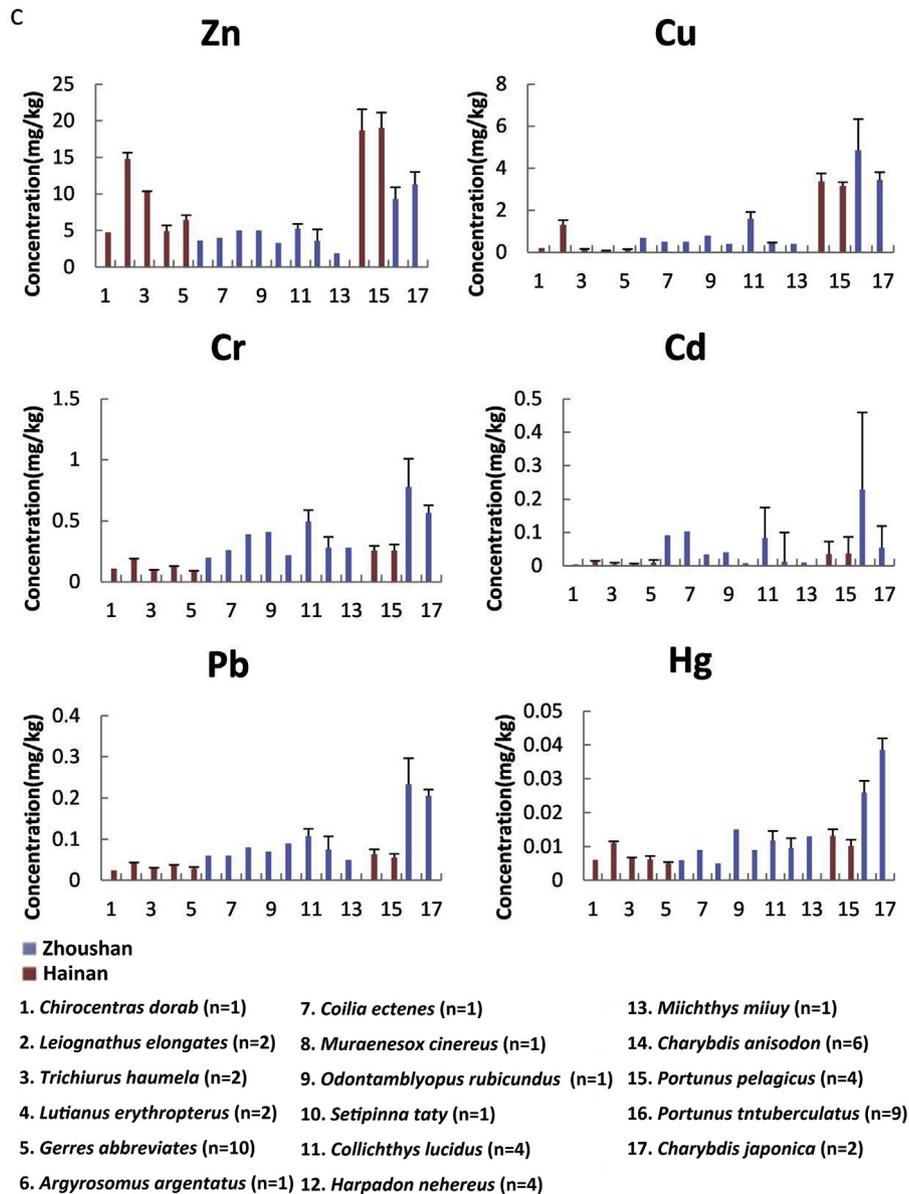


Fig. 4. (continued).

observed within China more broadly. For example, marine organisms exhibited a relatively low bioaccumulation ability for Pb from both seawater and sediment and a high bioaccumulation ability for Cu and Zn from seawater. This may be a common feature of heavy metal accumulation in China, and environments or species unique to China may explain this phenomenon. Further research is needed within and outside China in this regard. Previous studies have reported that industrial activities and fuel combustion contribute considerable quantities of Pb to the terrestrial environment (Harlavan et al., 2010), which may subsequently be deposited in the marine environment (Bosch et al., 2016; Reuer and Weiss, 2002). However, the low Pb bioaccumulation ability of marine organisms limits the health risk to humans.

4. Conclusions

We analyzed the distribution and bioaccumulation of six heavy

metals—Cd, Cr, Cu, Pb, Hg, and Zn—in marine organisms from China's Hainan and Zhoushan coastal regions. Fish and crab samples were collected from 15 sites in Hainan and 14 sites in Zhoushan. Seawater and sediment samples were also collected to elucidate the environmental background values. In all marine organism samples, Zn and Hg had the highest and lowest concentrations, respectively. Similar trends were observed in the sediment and seawater samples.

Heavy metal distribution and bioaccumulation in marine organisms varied according to region and species. Both distribution and bioaccumulation results indicated higher-quality marine organisms and environments in Hainan, as both exhibited higher Zn concentrations and lower concentrations of all other heavy metals. Heavy metal concentrations across both coastal regions were higher in crab than in fish. Marine organisms' bioaccumulation ability was also significantly higher for heavy metals in seawater than in sediments. Higher sediment background values may

Dendrogram using Average Linkage (Between Groups)

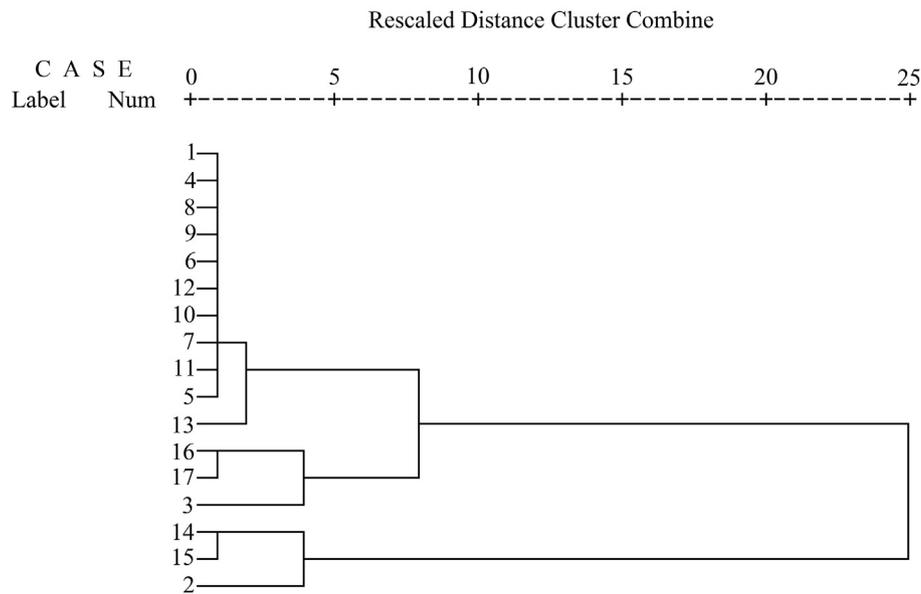


Fig. 5. Hierarchical dendrogram showing the clustering of marine organisms based on heavy metal concentrations.

Table 1

Bioaccumulation factors for marine organisms from the Hainan and Zhoushan coastal regions.

No.	Region	Category	Species	N	Organism-sediment						Organism-seawater					
					Cu	Pb	Zn	Cd	Cr	Hg	Cu	Pb	Zn	Cd	Cr	Hg
1	Hainan	Fish	<i>Chirocentras dorab</i>	1	0.02	0.001	0.08	0.01	0.00	0.34	272	42	369	35	166	233
2			<i>Leiognathus elongatus</i>	2	0.12	0.002	0.24	0.04	0.00	0.61	1821	76	1152	105	285	425
3			<i>Trichiurus haumela</i>	2	0.01	0.002	0.17	0.02	0.00	0.37	178	53	801	43	141	254
4			<i>Lutianus erythropterus</i>	2	0.01	0.002	0.08	0.02	0.00	0.35	111	66	381	53	194	240
5			<i>Gerres abbreviatus</i>	10	0.01	0.001	0.11	0.03	0.00	0.27	151	49	500	82	129	189
Average					0.02	0.002	0.11	0.03	0.003	0.33	347	54	541	75	159	231
6	Zhoushan	Fish	<i>Argyrosomus argentatus</i>	1	0.03	0.002	0.03	0.48	0.00	0.20	467	88	338	242	308	194
7			<i>Coilia ectenes</i>	1	0.02	0.002	0.04	0.54	0.00	0.30	333	88	376	271	400	290
8			<i>Muraenesox cinereus</i>	1	0.02	0.003	0.05	0.18	0.01	0.17	333	118	469	89	600	161
9			<i>Odontamblyopus rubicundus</i>	1	0.06	0.003	0.08	0.13	0.01	0.83	1111	123	389	342	621	577
10			<i>Setipinna taty</i>	1	0.03	0.003	0.05	0.03	0.01	0.50	556	158	257	67	333	346
11			<i>Collichthys lucidus</i>	4	0.13	0.004	0.09	0.26	0.01	0.65	2222	189	409	692	750	452
12			<i>Harpadon nehereus</i>	4	0.03	0.003	0.06	0.04	0.01	0.53	590	132	278	104	424	365
13			<i>Miichthys miiuy</i>	1	0.03	0.002	0.03	0.03	0.01	0.72	556	88	148	83	424	500
Average					0.03	0.003	0.04	0.25	0.005	0.33	540	119	390	126	534	323
14	Hainan	Crab	<i>Charybdis anisodon</i>	6	0.27	0.003	0.31	0.11	0.01	0.73	4687	111	1452	292	392	506
15			<i>Portunus pelagicus</i>	4	0.25	0.003	0.31	0.12	0.01	0.57	4384	98	1481	307	390	392
Average					0.26	0.003	0.31	0.11	0.007	0.67	4569	105	1463	300	391	462
16	Zhoushan	Crab	<i>Portunus trituberculatus</i>	9	0.18	0.009	0.09	1.20	0.01	0.86	3230	343	875	600	1198	835
17			<i>Charybdis japonica</i>	2	0.13	0.008	0.11	0.29	0.01	1.28	2300	301	1061	143	869	1242
Average					0.17	0.009	0.09	1.03	0.011	0.93	3060	335	909	516	1138	903

explain the higher heavy metal concentrations in crab. Environmental background values contributed more to heavy metal concentrations in marine organisms than their bioaccumulation ability did. In general, marine organisms' bioaccumulation ability was higher for Cu and Zn and lower for Pb. Marine organisms from China, within and outside the study area, exhibited a relatively low bioaccumulation ability for Pb from both seawater and sediment and a high bioaccumulation ability for Cu from seawater.

Results from this study contribute to the understanding of heavy

metal distribution and bioaccumulation in marine organisms. Specifically, the results can support the development of targeted response measures to protect marine ecosystems and public health. Findings specific to marine organisms can support the development of targeted seafood consumption guidelines for humans. Findings specific to the coastal environments may be generalizable to other coastal areas in China or worldwide. Future research should consider human health risk assessment and the influencing mechanism of heavy metals in marine organisms.

Table 2
Bioaccumulation factors for marine organisms from outside the study area.

	Source	Cu	Pb	Zn	Cd	Cr	Hg	Order
BAFs-sediment								
Fish in Hainan	This study	0.02	0.002	0.11	0.03	0.003	0.33	Hg > Zn > Cd > Cu > Cr > Pb
Crab in Hainan	This study	0.26	0.003	0.31	0.11	0.01	0.67	Hg > Zn > Cu > Cd > Cr > Pb
Fish in Zhoushan	This study	0.03	0.003	0.04	0.25	0.01	0.33	Hg > Cd > Zn > Cu > Cr > Pb
Crab in Zhoushan	This study	0.17	0.01	0.09	1.03	0.01	0.93	Cd > Hg > Cu > Zn > Cr > Pb
<i>Scylla serrata</i> in Xiangshan	Zhao et al. (2018)	0.29	0.003	0.34	0.41	0.001	0.70	Hg > Cd > Zn > Cu > Pb > Cr
Fish in West Guangdong	Zhang et al. (2015)	1.45	0.01	0.41	3.79	0.01	/	Cd > Cu > Zn > Cr > Pb
Fish in Jiangsu	Yuan et al. (2005)	0.67	0.07	0.97	0.27	0.01	0.00	Zn > Cu > Cd > Pb > Cr > Hg
Indicus in Southeast Coast of India	Sankar et al. (2018)	2.86	0.47	4.38	0.76	/	0.51	Zn > Cu > Cd > Hg > Pb
oyster in oman sea	Bazzi (2014)	3.12	0.36	4.73	0.36	0.85	/	Zn > Cu > Cr > Pb > Cd
Fish in Chennai	Thomas et al. (2014)	/	0.48	/	0.30	0.62	0.20	Cr > Pb > Cd > Hg
BAFs-seawater								
Fish in Hainan	This study	347	54	541	75	159	231	Zn > Cu > Hg > Cr > Cd > Pb
Crab in Hainan	This study	4569	105	1463	300	391	462	Cu > Zn > Hg > Cr > Cd > Pb
Fish in Zhoushan	This study	540	119	390	126	534	323	Cu > Cr > Zn > Hg > Cd > Pb
Crab in Zhoushan	This study	3060	335	909	516	1138	903	Cu > Cr > Zn > Hg > Cd > Pb
<i>Scylla serrata</i> in Xiangshan	Zhao et al. (2018)	4609	59	2287	270	100	1423	Cu > Zn > Hg > Cd > Cr > Pb
Fish in West Guangdong	Zhang et al. (2015)	14,381	110	3236	8000	259	/	Cu > Cd > Zn > Cr > Pb
Fish in Jiangsu	Yuan et al. (2005)	189	3	34	237	0.8	8	Cd > Cu > Zn > Hg > Pb > Cr
Ennore Creek, Southeast Coast of India								
Indicus in Southeast Coast of India	Kumara et al. (2013)	308	694	487	399	164	382	Pb > Zn > Cd > Hg > Cu > Cr
oyster in oman sea	Sankar et al. (2018)	17,417	12,345	19,633	1467	/	633	Zn > Cu > Pb > Cd > Cr > Hg
Fish in Chennai	Bazzi (2014)	40,199	4154	13,636	1938	1470	/	Cu > Zn > Pb > Cd > Cr
	Thomas et al. (2014)	/	47,733	/	15,481	11,317	8667	Pb > Cd > Cr > Hg

Conflicts of interest

The authors declare no conflicts of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2019.03.132>.

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