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# Bioaccumulation and changes of trace metals over the last two decades in marine organisms from Guangdong coastal regions, South China

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## ARTICLE INFO

### Article history:

Received 4 December 2019

Revised 5 May 2020

Accepted 6 May 2020

Available online 14 June 2020

### Keywords:

Trace metal

Bioaccumulation

Marine organism

South China

## ABSTRACT

Trace metal (Cr, Ni, Cu, Zn, Cd and Pb) exposures, distribution and bioaccumulation were investigated in marine organisms from Guangdong coastal regions, South China. The results showed that all of the selected metals were observed in marine organisms with a predomination of Cu and Zn. The metal exposure levels exhibited obvious variations between species with the decreasing order of crab > shellfish > shrimp > fish. The higher metals enrichment seen in shellfish and crab species primarily attributed to their living habits and the higher sediment background values of trace metals. Endpoint bioaccumulation factor (BAF<sub>fd</sub>) was used to characterize the bioaccumulation potentials of marine organisms to trace metals, of which Cu and Zn were the most accumulated elements. The exposure of trace metals in the cultured organisms was far lower than those in wild marine organisms, which is probably due to the effect of growth dilution. Comparisons with previous studies demonstrated that the concentration profiles of most trace metals declined over the last one to two decades, except Cu, that increased indistinctively.

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## Introduction

Trace metal pollution has become a global concern, especially in developing countries due to their high toxicity to both humans and livestock (Shafi et al., 2015; Wei et al., 2008). These metals are introduced

into aquatic environments through a variety of natural and anthropogenic sources, including industrial and domestic discharges, mining, smelting, and e-waste recycling (Pan and Wang, 2012). Trace metals can be bioaccumulated in the body of marine organisms after they enter marine environments. In addition, these metals can be transferred to human beings through the food chain, thereby causing the potential health risks (Li et al., 2017; Wong, 2017). The exposure levels of trace metals in marine organism species varied from both their physiological and metabolizable processes and their living environ-

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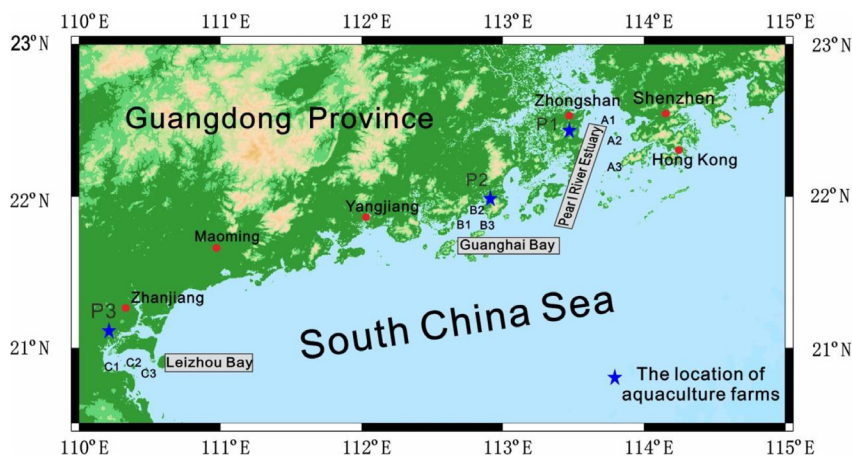


Fig. 1 – Sample locations (A1-A3, B1-B3, C1-C3, P1-P3) in Guangdong coastal zone.

ments. Thereafter, some marine organisms can act as an indicator to monitor the long-term metal accumulation from coastal environments (Gu et al., 2017). For example, trace metals in fish tissues are usually used as a biomonitor to estimate the exposure levels in waters (Zhou et al., 2008).

Guangdong Province has the longest coastline of  $3.37 \times 10^6$  m in China (Zhang et al., 2015). However, Guangdong coastal environments have faced significant challenges in terms of ecological health during in the past few decades due to its rapid urbanization and high industrialization—especially in the Pearl River Delta (PRD) region, which had been called the World's Factory (Chen et al., 2012; Ip et al., 2005). A greatly large amount of various pollutants, including trace metals from both industrial and civil sources, have been discharged into the Guangdong coastal environments through rivers and land runoff. These finally entered the open South China Sea (SCS) (Wang et al., 2018; Zhang et al., 2015).

Previous studies have documented that trace metals were ubiquitous in seawater sediments and marine organisms in most Chinese coastal regions, and as well as Guangdong coasts (Hao et al., 2019; Qiu, 2015a; Zhang et al., 2016b). In addition, trace metal contaminations have high potential ecological risks to the marine organisms and posed a potential threat to human health through food chains (Qiu, 2015a; Wang et al., 2018). Although a number of studies have been performed in the past one to two decades to investigate trace metal contaminations in marine organisms of the coasts of Guangdong Province (Gu et al., 2018; Zhang et al., 2015), the rapid industrial and urban developments of Guangdong Province as well as in the PRD region could have caused changes to the metal exposures in marine organisms over the last one to two decades. Therefore, the objectives of this study are to investigate and assess the current situation regarding the trace metals pollution and spatial distribution in the four categories of marine organisms (fish, shrimp shellfish and crab) from Guangdong coastal environments, and further to study the temporal variations during the two decades for seeing the differences in concentrations between wild and aquaculture marine organisms.

## 1. Materials and methodology

### 1.1. Samples collection

A total of 135 samples of wild marine organisms were collected via trawl net at three sampling regions from November 2016 to April 2017. The three sampling regions included the Pearl River Estuary (PRE), Guanghai Bay (GH) and Leizhou Bay (LZ); these are shown in Fig. 1. Synchronously, a total of 110 samples of aquaculture organism were purchased from several aquaculture farms (P1, P2 and P3); these were distributed in the coastal areas of the PRE, GH and LZ, respectively. The samples generally include four common groups: fish, shrimp, shellfish and crab. Nine species of fish, eight species of shrimp, two species of

shellfish and four species of crab were sampled in this study. The details of the species of the samples are summarized in Table 1.

### 1.2. Samples treatment and analysis

All of the samples were individually stored in polyethylene bags. The entire edible muscles of the organisms were taken out and stored frozen at  $-20^\circ\text{C}$  prior to freeze-drying. The freeze-dried samples were then characterized by the contents of trace and major elements. Solids were digested by nitric and perchloric acids using methods described previously (Xu et al., 2012); only a brief description is given here. Briefly, ground samples were treated with high purity nitric acid and perchloric acid in an aluminum heating block and heated to complete dryness with a designed temperature variation procedure. The residue was dissolved in 5% high purity nitric acid. The metal concentrations were measured by inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500, Agilent Technologies, CA, USA).

### 1.3. QA/QC

For quality assurance, reagent blanks, sample replicates and standard reference materials (NIST SRM 1566a and DORM-2) were included during the analytical measurement to assess the accuracy and precision of the analysis. The quality control tests show that the blanks were below 1% of the mean concentration for all metals. The QA/QC results showed no signs of contamination. The recovery rates for in the standard reference material NIST 1566a ranged from 85% to 108%, and the precision (RSD) was generally lesser than 5%.

## 2. Results and discussion

### 2.1. Exposures and distribution of trace metals in wild marine organisms

The trace metal concentrations in the marine organism are shown in Table 2. Generally, all metals were detected in the marine organisms, dominated by Cu and Zn. The Cr, Ni, Cu, Zn, Cd and Pb concentrations ( $\mu\text{g/g}$ , dry weight, dw) were in 0.49–3.04, 0.09–4.66, 1.13–168.00, 10.50–169.00, 0.01–5.76, and no data (n.d.)–1.90 respectively. The mean concentrations decreased as  $\text{Zn} > \text{Cu} > \text{Ni} > \text{Cr} \approx \text{Cd} > \text{Pb}$  in the marine organisms from the three sampling areas. Cu and Zn are the non-essential elements, and their contents in fish and shrimp were lower than the limits (50 and 150 mg/kg, respectively) of the national seafood safety standard (NSPRC, 2005). However, it should be noted that their exposure in crabs from the three sampling regions were remarkably higher than the limits. Therefore, more concern is needed on the toxicity effects produced by these two elements. Pb mainly originates from automobile emissions and industrial discharges in the Pearl River Delta region (Ip et al., 2005). Vehicles emit a large amount of lead-containing exhaust gas into the atmosphere, which then settles

**Table 1 – The sizes and weight of wild organisms and aquaculture organisms samples.**

	Scientific name	Species	Number	Sample locations	Body length (cm)	Weight (g)
Wild organisms	<i>Leiognathus brevirostris</i>	Fish	9	A1, A2	6–11	26–90
	<i>Collichthys lucidus</i>	Fish	5	A2, A3	9–16	36–68
	<i>Argyrosomus argentatus</i>	Fish	10	B1, B3, C2	7–13	61–105
	<i>Ophichthus apicalis</i>	Fish	3	B1, B2	36–51	75–113
	<i>Cynoglossus sinicus</i>	Fish	3	B1, B2	14–17	106–124
	<i>Siganus canaliculatus</i>	Fish	7	C1, C2	7–13	24–56
	<i>Therapon theraps Cuvier et Valenciennes</i>	Fish	3	C2, C3	10–14	61–109
	<i>Metapenaeus ensis</i>	Shrimp	8	A2, A3,	5–10	10–15
	<i>Penaeus japonicus</i>	Shrimp	13	B1, B2, B3	6–11	14–18
	<i>Penaeus penicillatus</i>	Shrimp	10	B1, B2	7–15	15–25
	<i>Oratosquilla oratoria</i>	Shrimp	18	B1, B2, B3, C1,C2	5–12	15–31
	<i>Parapenaeopsis hardwickii</i>	Shrimp	11	C1, C2, C3	3–8	6–13
	<i>Paphia gallus</i>		14	C1, C2, C3	2.5–4 <sup>a</sup>	5–12
	Aquaculture organisms	<i>Ostrea rivularis Gould</i>	Shellfish	4	C2,C3	6–9 <sup>a</sup>
<i>Scylla serrata</i>		Crab	8	A2, A3,	4–8	33–105
<i>Matuta lunaris</i>		Crab	4	B1, B3	3–5	12–42
<i>Portunus sanguinolentus</i>		Crab	6	C1, C2, C3	4–7	23–65
<i>Plectorhinchus cinctus</i>		Fish	20	P1, P3	14–22	280–390
<i>Sciaenops ocellatus</i>		Fish	10	P2	26–45	360–510
<i>Penaeus orientalis</i>		Shrimp	20	P1, P3	11–15	38–55
<i>Penaeus vannamei</i>		Shrimp	20	P1	9–12	30–46
<i>Macrobrachium rosenbergii</i>		Shrimp	10	P2	10–15	40–61
<i>Scylla serrata</i>		Crab	30	P1, P2	8–10	98–156

<sup>a</sup> The length of shell; <sup>b</sup>The length of carapace.

**Table 2 – Heavy metals concentrations in wild marine organisms.**

Sampling region	Species	Average content (µg/g)						Scientific name	
		Cr	Ni	Cu	Zn	Cd	Pb		
Pearl River Estuary	Fish	0.57±0.07	0.16±0.05	8.64±1.12	23.10±2.72	0.05±0.01	n.d. <sup>a</sup>	<i>Leiognathus brevirostris</i> (n=9)	
	Shrimp	0.87±0.10	0.27±0.06	1.44±0.27	30.30±6.95	0.05±0.01	0.82±0.27	<i>Collichthys lucidus</i> (n=5)	
Guanghai Bay	Crab	0.81±0.13	0.51±0.12	23.70±3.38	61.37±6.45	0.03±0.01	0.14±0.03	<i>Metapenaeus ensis</i> (n=8)	
	Fish	1.05±0.20	1.74±0.34	119.33±27.00	111.67±27.63	3.08±0.28	0.39±0.07	<i>Scylla serrata</i> (n=8)	
	Shrimp	Fish	0.84±0.09	0.82±0.05	1.86±0.37	31.33±6.64	0.05±0.01	0.08±0.01	<i>Argyrosomus argentatus</i> (n=6)
		Fish	1.68±0.14	1.46±0.05	20.90±2.13	51.20±14.95	0.041±0.01	0.33±0.03	<i>Ophichthus apicalis</i> (n=3)
		Fish	0.75±0.18	0.19±0.05	2.25±0.65	12.97±3.00	0.059±0.01	1.80±0.13	<i>Cynoglossus sinicus</i> (n=3)
	Shrimp	0.81±0.06	0.34±0.03	36.93±8.61	59.45±15.97	0.064±0.01	n.d.	<i>Penaeus japonicus</i> (n=13)	
	Shrimp	0.76±0.07	0.23±0.05	31.55±7.00	52.98±12.52	0.44±0.09	n.d.	<i>Penaeus penicillatus</i> (n=10)	
Shrimp	0.89±0.06	1.72±0.47	101.25±27.91	104.25±26.85	4.59±0.96	0.16±0.02	<i>Oratosquilla oratoria</i> (n=12)		
Leizhou Bay	Crab	2.64±0.54	3.43±0.83	149.00±23.30	108.33±26.01	2.92±0.32	0.52±0.12	<i>Matuta lunaris</i> (n=4)	
	Fish	1.22±0.18	0.71±0.54	1.61±0.11	21.30±5.08	0.01±0.00	0.18±0.07	<i>Siganus canaliculatus</i> (n=7)	
	Shrimp	Fish	0.88±0.35	0.48±0.30	1.72±0.53	28.40±8.77	0.078±0.04	n.d.	<i>Argyrosomus argentatus</i> (n=4)
		Fish	0.83±0.08	0.66±0.50	1.47±0.17	32.87±6.56	0.028±0.02	0.79±0.13	<i>Therapon theraps Cuvier et Valenciennes</i> (n=3)
	Shrimp	0.72±0.03	0.65±0.05	74.60±5.45	99.53±23.08	2.31±0.44	n.d.	<i>Oratosquilla oratoria</i> (n=6)	
	Shrimp	0.81±0.04	0.61±0.04	10.53±3.45	62.78±12.88	0.31±0.05	0.08±0.01	<i>Parapenaeopsis hardwickii</i> (n=11)	
	Shellfish	2.22±0.15	4.39±0.26	6.31±1.42	81.90±12.83	1.11±0.12	0.99±0.10	<i>Paphia gallus</i> (n=14)	
Crab	Crab	1.28±0.17	2.01±0.16	67.90±9.67	149.33±21.73	3.08±0.23	0.70±0.15	<i>Ostrea rivularis Gould</i> (n=3)	
	Crab	0.60±0.07	1.72±0.07	52.37±15.48	101.17±22.75	1.18±0.17	n.d.	<i>Portunus sanguinolentus</i> (n=6)	

<sup>a</sup> Not detected; <sup>b</sup>Number of organisms for detection.

onto water surfaces and is eventually washed into the water column and sewage. Thus, the exposure levels of Pb in water are higher than those in sediment (Zhang et al., 2016b). In this study, Pb was detected with remarkably lower concentrations and detection frequencies, suggesting its low environmental exposures in Guangdong coastal areas, which probably contributed to the use of unleaded gasoline since 2003

(Lee et al., 2007). Cr and Cd derived from wastewater discharge and acid mine drainage (Wei et al., 2009), had relatively low concentrations. However, their potential ecological risks could not be neglected due to their high toxicities.

The accumulation of trace metal in organisms usually depends on their living habits (Sfakianakis et al., 2015). Here, the concentrations of

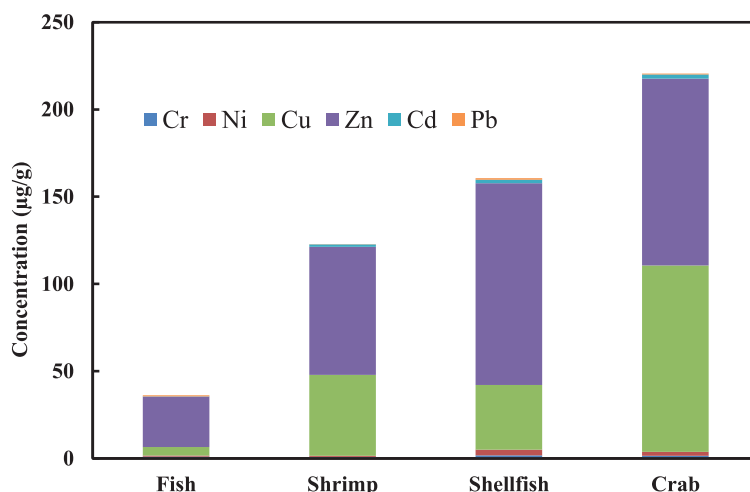


Fig. 2 – Metal concentrations in different marine species.

Table 3 – Heavy metals concentrations in aquaculture organisms.

Sampling region	Species	Number	Average content (µg/g)						Remarks
			Cr	Ni	Cu	Zn	Cd	Pb	
Pearl River Estuary	Fish	10	0.71	0.27	1.22	20.16	0.01	<0.05	<i>Plectorhinchus cinctus</i>
	Shrimp	20	0.7	0.17	25.86	49.46	0.01	<0.05	<i>Penaeus orientalis</i>
Guanghai Bay	Crab	20	0.65	0.22	13.64	32.42	0.01	<0.05	<i>Penaeus vannamei</i>
	Fish	15	1.05	0.53	43.00	70.44	0.30	<0.05	<i>Scylla serrata</i>
	Shrimp	10	0.64	0.14	1.71	17.16	0.03	<0.05	<i>Sciaenops ocellatus</i>
Leizhou Bay	Crab	20	0.75	0.22	23.66	53.36	0.04	<0.05	<i>Macrobrachium rosenbergii</i>
	Fish	15	0.98	0.58	42.06	62.02	0.26	<0.05	<i>Scylla serrata</i>
Leizhou Bay	Fish	10	0.56	0.15	2.06	19.24	0.01	<0.05	<i>Plectorhinchus cinctus</i>
	Shrimp	20	0.73	0.24	20.26	50.42	0.01	<0.05	<i>Penaeus orientalis</i>

these metals showed variations between species with decreasing order of crab > shellfish > shrimp > fish (Fig. 2). The considerably high Cu observed in crab (up to 168 µg/g) indicated that Cu was prone to bioaccumulation in crab species (Liu et al., 2019). Marine sediments in coastal environments usually act as a trace metals reservoir and could transfer to seawater under dynamic conditions (Xu et al., 2015). In addition, suspended sediments are generally enriched by trace metal due to the high abundance of fine particles. Hence, it is reasonable that crab and shellfish that live in sand or sediment and use suspended matters as their primary food source are likely to accumulate trace metals (Gu et al., 2018; Jia et al., 2018). In this study, trace metal concentrations varied indistinctively in the same species from different sampling areas, except for the species *Ophichthus apicalis*. The Cu detected in *Ophichthus apicalis* which belongs to benthic species and was one to two orders magnitude higher than in the other fish species.

Generally, the total metals in the marine organisms did not show evidently spatial variation. This indicated that the overall contamination situation in marine organisms from different areas of Guangdong coasts was similar. Besides, some individual metals displayed spatial variation, suggesting their specific sources. In comparison with PRE and LZ Bay, the Cu in GH Bay is evidently higher in shrimp and slightly higher in fish and crab. The trace metals in the Guangdong coastal environments predominantly came from the sewage and industrial wastewater discharge and atmospheric deposition. Hence, the local industry structure could affect the metals exposures in the local environment. The Cu industries from the GH industrial park could be responsible for the high Cu. LZ Bay has large-scale crude oil dock (Zhanjiang Harbor). It is also close to Zhanjiang City, an important industrial city in Guangdong Province. Zn and Pb from LZ Bay were higher than the other two bays—particularly in shrimp and shellfish species.

## 2.2. Comparisons of trace metals from aquatic organisms in worldwide coastal areas

Appendix A Table S1 summarizes the environmental exposures of trace metals in marine organisms from global marine and estuarine regions. The results indicated that the trace metal contents along the Guangdong coast were much higher than those from other listed regions, this is likely due to China's rapid development and large-scale urbanization in the past few decades. Obvious higher levels of trace metals were found in bivalves in dry weight from the Gulf of Oman, Arabia (De Mora et al., 2004). Besides, human factors such as diverse degrees of industrialization and activities, natural conditions, including salinity, temperature, and tidal currents could impact the bioaccumulation and distribution of trace metals in marine organisms (Krishnakumar et al., 2016).

## 2.3. Relationship between trace metal contents and biomass

Table 3 summarizes the concentrations of metals in sampled aquaculture organisms. In comparison to wild marine organisms, the average concentrations of trace metals were evidently lower in artificially reared organisms. Trace metal contents followed the order of Zn > Cu > Cr > Ni > Cd ≈ Pb in all organisms. Metals in different species, except for Zn and Cu, were ranked as crab > shrimp ≈ fish. The concentrations of trace metals in marine organisms are extremely relevant to their bioaccumulation potential and their natural habitat (Pini et al., 2015; Zhang et al., 2016b). The bioaccumulation potential of organisms has a distinct effect on absorbing chemical substances from their habitat environments. The endpoint bioaccumulation factor (BAF<sub>fd</sub>) is appropriately used to characterize the bioaccumulation potential. It

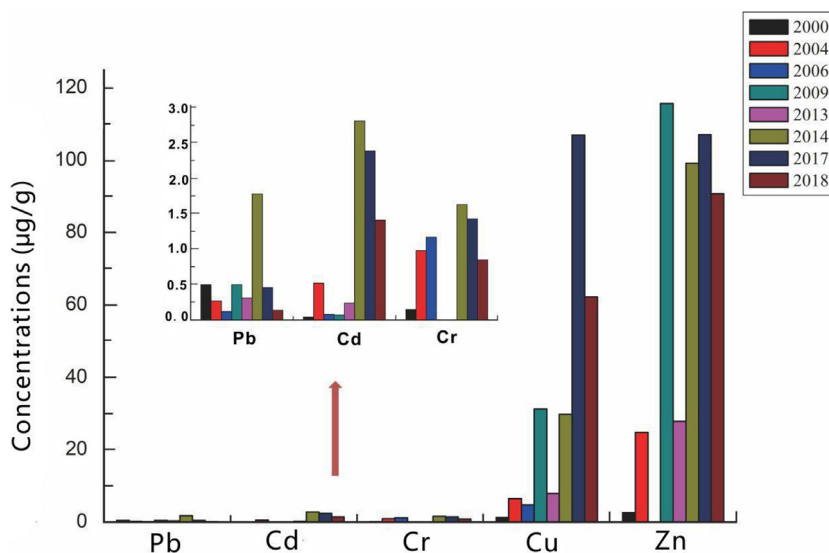


Fig. 3 – Metal contents in crustacean from the recent two decades.

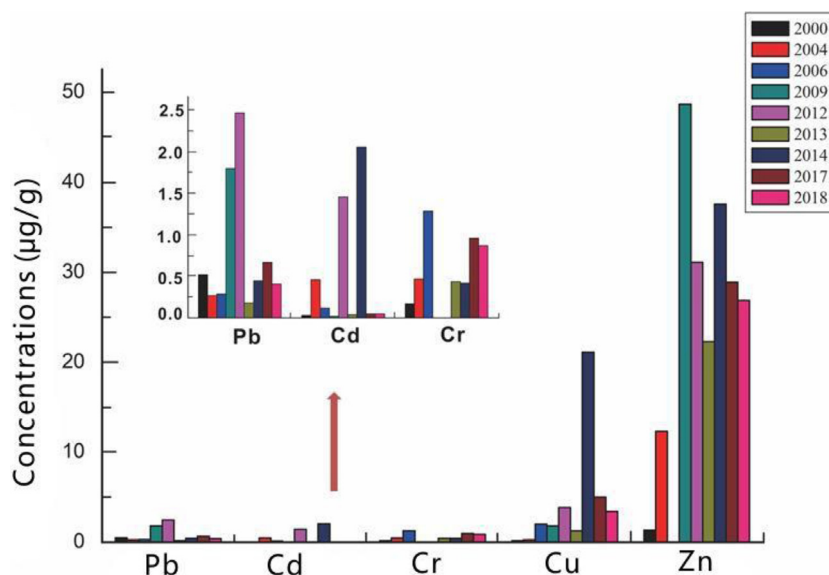


Fig. 4 – Metal contents in fish from the recent two decades.

is generally calculated as follows:

$$BAF_{fd} = C_B / C_{WD}$$

$C_B$  (g/kg) is the chemical concentration in the organism, and  $C_{WD}$  (g/L) is the freely dissolved chemical concentration in the water (Arnot and Gobas, 2006). A previous study investigated the average values of Cu, Zn, Cr and Cd in seawater around the study area. These values were 2.23, 10.44, 1.95 and 1.38  $\mu\text{g/L}$ , respectively (Zhang et al., 2016a). Appendix A Table S2 lists  $BAF_{fd}$  in different biotas.  $BAF_{fd}$  values followed the order of  $\text{Cu} \approx \text{Zn} > \text{Cr} > \text{Cd}$  in fish,  $\text{Cu} > \text{Zn} > \text{Cd} > \text{Cr}$  in shrimp, and  $\text{Cu} \approx \text{Zn} > \text{Cd} > \text{Cr}$  in crab. These organisms had similar levels of Cu and Zn, which were several orders of magnitude higher than those of Cr and Cd.

Appendix A Table S3 shows the relationships between fish lengths and metal contents. Strong positive correlations between Cu and Zn were found which probably resulted from their similar sources. Conversely, Cd was negatively correlated with Zn. Fewer metals were found in the artificially reared organisms, which is probably due to the effect of growth dilution: these values are remarkably higher than

the wild marine organisms in body length and weight (Lefebvre et al., 2004).

#### 2.4. Changes of the contents of trace metals recent ten years

Figs. 3 and 4 present the trace metal contents in crustacean and fish species in or near our study areas from last 10–20 years (Hao et al., 2019; Leung et al., 2014; Qin et al., 2010; Wei et al., 2002; Zeng et al., 2012, 2014; Zhang et al., 2015, 2016a). In general, the trends of most trace metal contents in marine organisms of Guangdong coastal regions declined during this period, especially in the most recent 5–10 years. Owing to the rapid development in the Guangdong coastal economic belt, the total quantity of sewage and industrial wastewater discharges has continued to increase (Hao et al., 2019). Hence, this decline in metals probably benefited from the effective pollution control measures in Guangdong Province and the PRD region including the increased removal efficiencies for metals from wastewater and less untreated wastewater discharge.



It is interesting to note that Pb increased before 2012 and then sharply decreased. The high value of Pb was mainly attributed to vehicle exhaust. The environmental exposure of Pb remarkably reduced since 2012 due to the use of unleaded gasoline (He et al., 2009). It is worthy to note that Cu contents increased in the past two decades, which is consistent with the Cu finding in the sediment from the Pearl River Estuary (Chen et al., 2012). Generally, the inevitable discharge from industrial production and copper mining is the key source. Data from the Guangdong Statistical Bureau revealed that the output of electrolytic Cu kept increasing over the last one to two decades. However, a slight decrease in Cu was observed in the most recent two years.

### 3. Conclusions

The individual metal showed various distribution in different aquatic species. The mean concentrations were Zn > Cu > Ni > Cr ≈ Cd > Pb in the marine organisms. Metal pollution in the Guangdong coasts was relatively high than those in other coastal regions. The aquaculture organisms have lower metal contents than wild marine organisms—this is largely attributed to the growing dilution effects. Except for Cu, an overall decreasing trend was found over the past one to two decades, which probably benefited from the effective pollution control measures in Guangdong Province.

### 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.

### Acknowledgments

This work was supported by the Science and Technology Program of Guangzhou, China (No. 201707010219), the National Natural Science Foundation of China (No. 41877295) and the Project of China Geological Survey (No. DD20190627). We thank LetPub (www.letpub.com) for its linguistic assistance during the preparation of this manuscript.

### Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.05.007.

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