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Distribution Characteristics and Controlling Factors of Heavy Metals in Surface Sediments from the Bay-Island-Estuary System (BIES): A Case Study in Coastal Waters of Fujian Province, China

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Abstract Based on the contents of six heavy metal elements in surface sediments from coastal areas of Fujian Province, the distribution characteristics and controlling factors of six heavy metals in a bay-island-estuary system (BIES) were studied. This paper focuses on the influence of the hydrodynamic environment, and systematically discusses how grain size compositions, chemical environment, tidal current, ocean circulation and human activities influence the distribution and transportation of the heavy metals. The results indicated that the distribution and migration of Cu, Pb, Zn and Cr elements were mainly controlled by natural factors such as regional geological background, grain size compositions, and tidal residual currents. In contrast, As and Hg was mainly affected by human factors such as agriculture and industrial manufacturing. In the BIES, where the chemical environment exerted limited influence, the accumulation and migration of heavy metals are mainly influenced by human activities and enhanced by estuary processes as well as the complex sedimentary dynamic environment caused by many bays and islands.

Key words heavy metals; sediments; source to sink; controlling factors; hydrodynamic environment

1 Introduction

Over the past few decades, heavy metal pollution has become one of the most concerning environmental issues in the world (Wang *et al*., 2018b; Wang *et al*., 2020b). Due to its biotoxicity, non-degradability and bio-enrichment effects, it is regarded as a potential long-term threat to both human health and the ecosystem (Reddy *et al*., 2004; Green and Planchart, 2018; Naifar *et al*., 2018; Rai *et al*., 2019). Heavy metal elements are mainly supplied by natural processes such as rock weathering and coastal erosion (Lv *et al*., 2021). Meanwhile, human activities such as agricultural production, industrial manufacturing, fossil fuel combustion, just to name a few, have largely contributed to the process, since the industrial revolution, in particular, being responsible for the dramatic increase in the amount of heavy metals (Thevenon *et al*., 2011; Chen *et al*., 2014). Heavy metal elements produced by different natural processes and human activities are delivered to the ocean *via* surface runoff and accumulate in marine sediments (Yin *et al*., 2016). Thus, marine sediments can act as the 'sink' of heavy metals. Later, under certain marine environmental conditions (chemical environment, hydrodynamic environment, *etc*.), marine sediments can act as a 'source' of heavy metals by releasing them into water (Pan and Wang, 2012; Chen *et al*., 2016). Therefore, marine sediments play a key role in both the accumulation and the migration of heavy metals.

The study area is located in the coastal waters of the Fujian Province on the inner shelf of the East China Sea (ECS), 150 km to the northeast of the Taiwan Strait (Fig. 1a). It is a typical bay-island-estuary system (BIES) with many islands, bays and estuaries (Fig.1b). For example, the Sansha Bay, the Funing Bay and the Qingchuan Bay are representative ones; while the islands, which affect the hydrodynamic environment, include Fuying Island, Changbiao Islands, *etc*. Similarly, rivers such as the Huotongxi River, the Jiaoxi River and others also flow into the study area (Fig.1b), with the river basins having been developed for industrial and agricultural purposes. The study area is

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subject to the influence of Zhejiang-Fujian Coastal Current (ZFCC), which is a seasonal ocean circulation (Guan, 1983; Liu *et al*., 2010). The Oujiang River, Qiantang River and Yangtze River, to the northeast of the study area, supply large amounts of terrestrial materials (2.32×10^6 t, 6.08×10^6 t and 4.68×10^6 t respectively) into the coastal sea every year (Jin, 1988; Shi *et al*., 2010). It has been suggested that nearly 50% of the sediments derived from the Yangtze River have been transported to the ECS, of which approximately 30% have been transported more southwardly along the coastline by ZFCC (Xiao *et al*., 2006; Liu *et al*., 2007). And few sediments from the small rivers near the south of the Yangtze River, such as Oujiang River, Qiantang River, have been transported more southwardly along the coastal line by ZFCC (Qin *et al*., 1987). As commonly observed in the case of BIES, industrial manufacturing, agricultural production, port shipping and other intensive human activities may cause the heavy metal pollution to the sea, including in the study area (Lv *et al*., 2021).

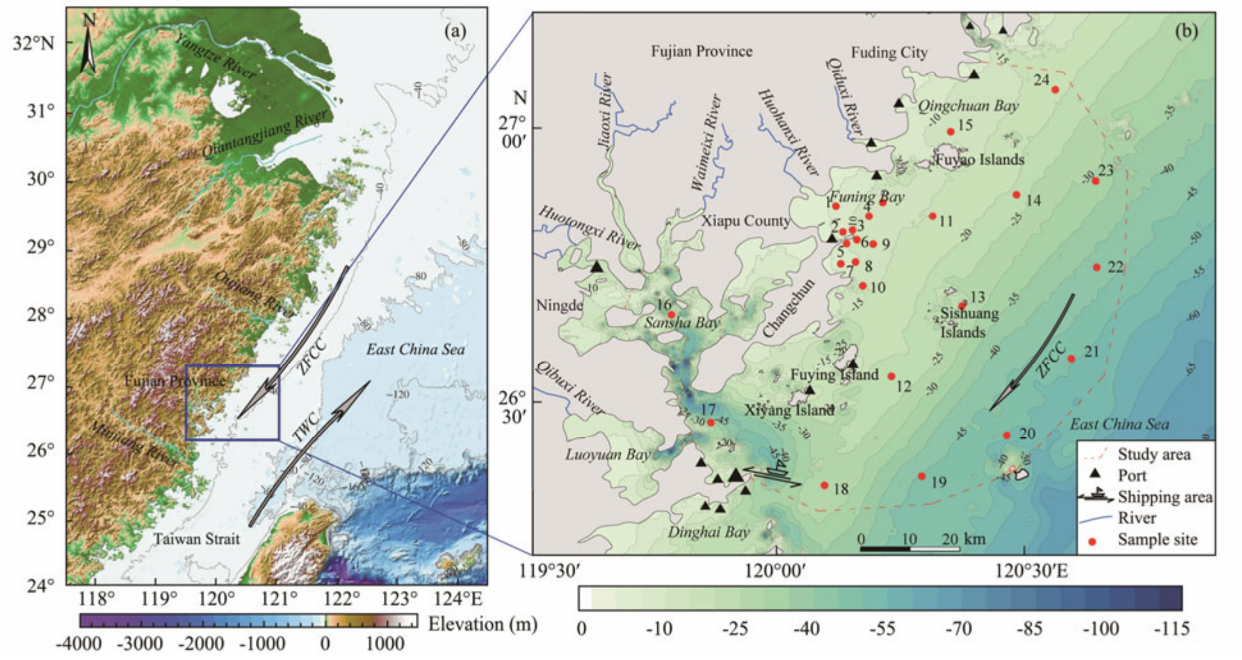

Fig.1 Location of study area (a) and sampling stations (b). Circulation system is after Guo *et al*. (2007). ZFCC, Zhejiang-Fujian Coastal Current; TWC, Taiwan Warm Current.

It should be noted that the study of marine environmental pollution should not be limited to a single discipline, but instead, it should focus on the integration and coordination of different environmental sciences such as geochemistry, geology, marine sedimentary dynamics, ecology as well as other disciplines. Due to varying environmental conditions in different areas of the sea, the dominant factors identified to control the heavy metal pollution by different disciplines are likely to be different. For example, Sindern *et al*. (2016) believed, from a geological perspective, that the weathering of volcanic rocks was the main factor affecting the content of As, Cr and part of Cu in the Jakarta Bay region. On the other hand, from an ecological perspective, Zhou *et al*. (2010) found that mangrove reforestation promoted the enrichment of heavy metal elements in intertidal surface sediments. Estuaries, bays and coastal waters, as major zones of sea-land interaction, are not only important places for frequent exchanges of matter and energy (Liu *et al*., 2020), but they are also the areas where human activities and economic constructions are significant. Hence, they are complex source-to-sink systems for heavy metals, which are under the control of multiple factors. Many scholars assessed the pollution level and discussed the distribution and sources of heavy metals in BIES from the perspective of environmental science and geochemistry (Brady *et al*., 2014; Zhao *et al*., 2018; Ota *et al*., 2021). Some researchers have found that sedimentary dynamic environment has a significant effect on the distribution and transport of heavy metal elements (Wang *et al*., 2013; Webb *et al*., 2020). Therefore, it is necessary to further discuss heavy metal pollution in BIES from the perspective of sedimentary dynamic environment. In this paper, the influence of the sedimentary dynamic environment was emphasized, especially considering the influence of sediment particle size compositions, the chemical environment, the tidal current, the ocean circulation and human activities on the transport of heavy metals in the study area. This work also provides a reference case for understanding the source-to-sink process of heavy metals in global BIES by highly interdisciplinary analysis, while providing important scientific guidance for the prevention and control of heavy metal pollution in coastal waters.

2 Materials and Methods

2.1 Sample Collection

From 25th to 31st in August 2015, 24 sediment samples were grabbed from the coastal waters of the Fujian

Province (119˚30′–121˚00′E, 26˚00′–27˚30′N) by using a grab sampler of the Second Institute of Oceanography (SOA) (Fig.1b). The upper 10 cm surface sediments were collected and sent to the testing center of SOA for analysis.

The data used in the partition of the sedimentary dynamic environment in the study area come from 268 sediment samples collected and analyzed by the 'Project on Coastal Investigation and Research'.

2.2 Methods

Before testing for particle size compositions, an appropriate amount of subsample was taken and put in a tube which was added successively excess amount of 30% H_2O_2 and 3 mol L^{-1} HCl to remove organic matters and carbonate minerals respectively. Then, $[NaPO_3]_6$, at a concentration of 0.05 mol L^{-1}, was added before applying ultrasonic vibrations to disperse the sediment particles. Samples, pretreated in this way, were subsequently analyzed with a laser particle size analyzer (Mastersizer 2000, Malvern Instruments Ltd., UK) in the range of 0.02–2000 μm, with an error of less than 1%.

The contents of the six typical pollution heavy metal elements in the surface sediment samples were determined according to GB17378.5-2007 the Specification for Marine Monitoring Part 5: Sediment Analysis (CSBQTS, 2008). For this purpose, the samples were first naturally air-dried, and then ground in an agate mortar before being passed through an 80-mesh nylon sieve and stored. After HNO_3-$HClO_4$-HCl and HF digestion, the Cu, Pb and Zn contents in the samples were determined by the flame atomic absorption spectroscopy (Thermo M6). Similarly, the amount of Cr was determined by the flameless atomic absorption spectroscopy (Thermo M6), while Hg and As contents were determined by the atomic fluorescence spectrometry (Jitian AFS-920), after digestion with an HCl-HNO_3 solution. The detection limits for Cu, Pb, Zn, Cr, Hg and As were 2.0, 3.0, 6.0, 2.0, 0.002 and 0.06 mg kg^{-1}, respectively, with the procedures re- ported by Wu *et al.* (2014a) and Wu *et al.* (2014b). For the analysis, parallel samples and a standard material, GBW-07314 were used to control and correct the sample detection process. All the standard deviations of the parallel samples were less than 8%, with the recovery rates of heavy metals ranging from 92% to 106%.

The specific analytical methods for determining total organic carbon (TOC) contents, redox potential (Eh), pH and sulfide contents in the samples were carried out according to GB17378.5-2007 the Marine Monitoring Code Part 5: Sediment Ana- lysis (CSBQTS, 2008). The TOC content in sediments was determined by the potassium dichromate oxidation-reduction volumetric method. The Eh value was determined by using a field potentiometer, while the pH measurements involved the use of a field pH meter. Finally, the sulfide content was spectrophotometrically determined by methylene blue, with a detection limit of 0.3 $\times 10^{-6}$ mg kg^{-1}.

2.3 Statistical Analysis

IBM SPSS Statistics 24 was used for analyzing the data to conduct the correlational analysis, principal component analysis and Q-type hierarchical cluster analysis after the data standardization by the Z-score standardization method in order to eliminate the impact of dimensional differences for the different indicators.

2.4 Numerical Modeling

The Flow Model Hydrodynamics Module in Mike 21 has been widely applied in the simulation of tidal current field and material transport in estuaries and shallow seas (Hanapiah *et al.*, 2020; Bai *et al.*, 2021). The numerical model was used to simulate the tidal current field in the study area. The model applied unstructured grids to divide the computational domain and the standard Galerkin finite element method to solve the equations.

The coordinate range of the calculation domain is 37˚00′36.63″–40˚57′05.37″N, 117˚23′54.53″–122˚39′25.35″E. Local encryption processing was carried out on the grid of the research area (Fig.2). There were 240093 triangles and 14603 nodes in the simulation area. The smallest area of the triangles was 24.2 m^2 and the smallest angle was 26.0˚. The time integration and space discretization in Solution Technique were set to low order, and the minimum time step was 0.1 s. The critical CFL number was 0.8. The drying depth, flooding depth, and wetting depth were set to 0.05 m, 0.1 m, and 0.15 m, respectively. Cs in Smagorinsky formulation was 0.28. The format of Maning number was set to varying in domain, and the value of Maning number ranged from 50 to 56 $m^{1/3}s^{-1}$. The open boundary of the model was controlled by the tidal level process and the tidal level boundary was calculated by the model China Tide. The simulation period was from August 1 to September 10, 2015, with the first 20 days as spin-up time, and the extraction time for the average tidal residual current was from August 20 to September 5 (a spring tide and neap tide cycle). The simulated tide results were verified by data collected from 3 stations in August 2015 (Fig.2). The simulated results for the tidal current were verified by current data from 13 stations which were recorded in August 2015. The specific method which was eventually used for establishing the numerical model is referred to Jiang *et al.* (2018).

3 Results

3.1 Grain Size, pH, Eh, Sulfide and TOC

Based on the results of the particle size analysis, the surface sediments in the study area were dominated by clayey silt (Fig.3a). The overall median particle size (D_{50}) ranged from 5.24 to 9.93 μm, with an average of 7.25 μm. Over the study area, relatively fine sediments are found in Sansha Bay, Fuying Island Waters, Changbiao Islands Waters and Qingchuan Bay (Fig.3a), with D_{50} ranging from 5.24 to 7.06 μm, and an average of 6.32 μm. On the other hand, sediments in the southern outer waters of the study area, the mouth of Luoyuan Bay as well as the mouth of Funing Bay were relatively coarse, with D_{50} values ranging from 7.13 to 9.93 μm and an average of 8.03 μm.

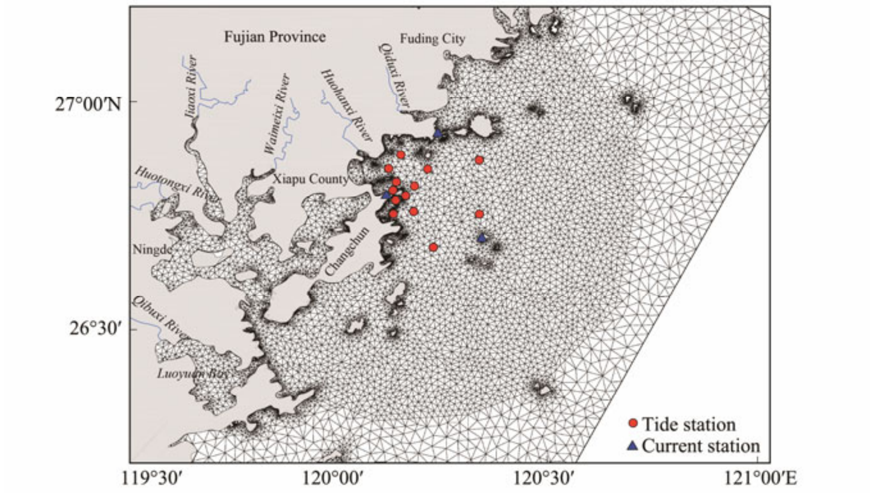

Fig.2 Computational domain of Mike 21 used in this study and verifying stations for current data.

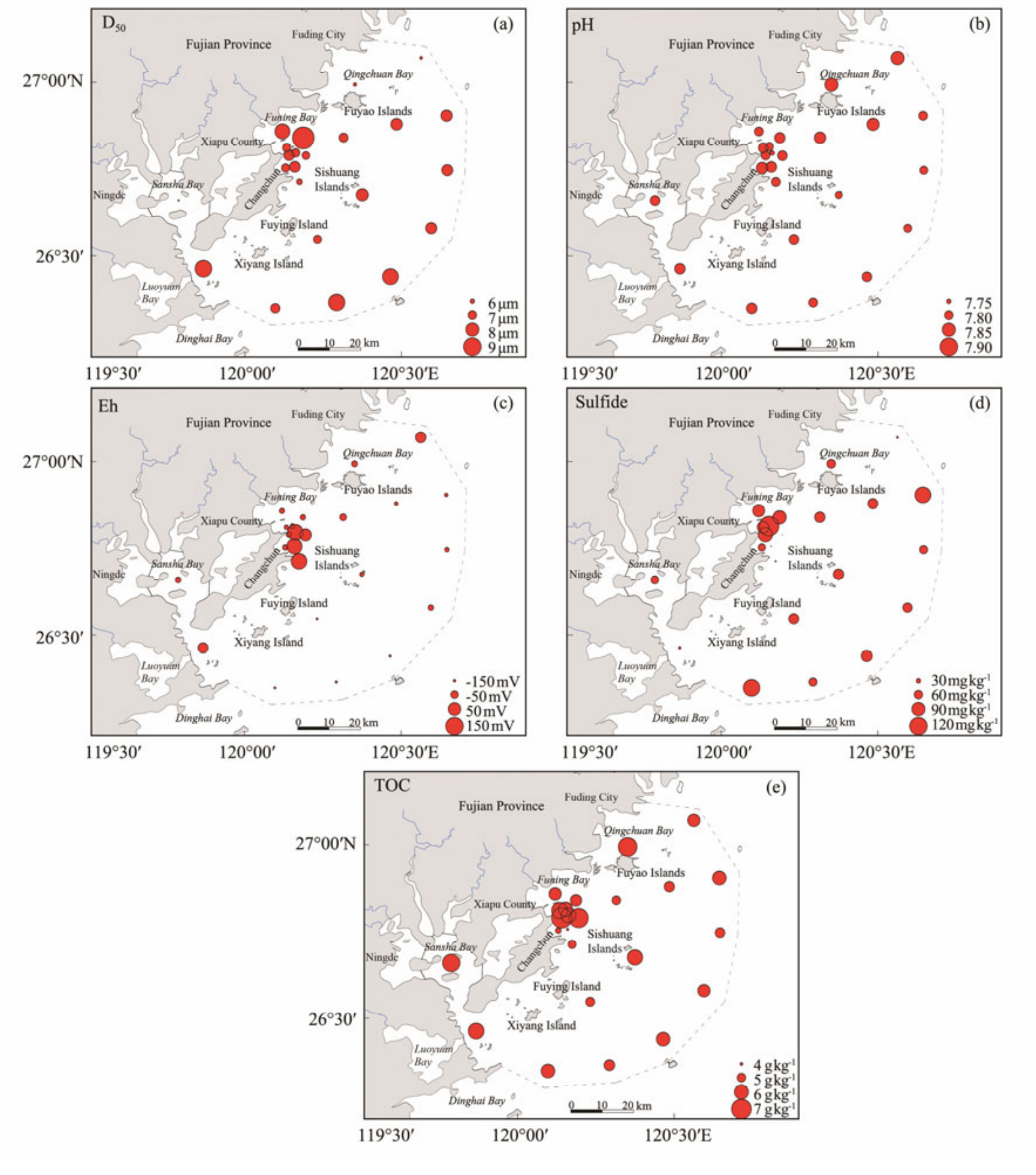

Fig.3 Distribution of D_{50} (a), pH (b), Eh (c), sulfide (d) and TOC (e).

The distribution of pH, Eh, contents of sulfide and TOC in the study area are shown in Figs.3b–3e. The pH values range from 7.75 to 7.85, with an average value of 7.81, thereby showing weak alkalinity. The range of the Eh value was −162 – 110 mV, with an average value of −70.6 mV, indicating that the study area is in a reduction environment as a whole. The concentrations of sulfide were in the range of 2 – 133.2 mg kg^{-1}, with a mean value of 59.88 mg kg^{-1}; By comparison Fig.3d with Fig.3c, it can be seen that the high value zones and low value zones of the sulfide concentrations are consistent with the low value zones and high value zones of Eh values, respectively. The concentration of TOC ranged from 3.9 to 7.1 mg kg^{-1}, with an average of 5.75 mg kg^{-1}.

3.2 Contents and Distribution Features of Heavy Metals

The distributions of heavy metals in the study area were shown in Figs.4a–4f. The concentrations of Cu, Pb, Zn, Cr, Hg and As are 26.7 –36.5 mg kg^{-1}, 31.10 – 41.80 mg kg^{-1}, 65.90–126.40 mg kg^{-1}, 46.30 – 61.60 mg kg^{-1}, 0.036–0.049 mg kg^{-1} and 9.80 –13.50 mg kg^{-1}, respectively, relatively higher than those of the sediments from adjacent waters (Table 1) (Liu *et al*., 2011; Bi *et al*., 2017). Similarly, the concentrations of heavy metals differed greatly from those from other typical BIES around the world (Table 1) (Zhang *et al*., 2007; Yu *et al*., 2016; Li *et al*., 2017; El-Sorogy *et al*., 2018; Liu *et al*., 2018). For example,

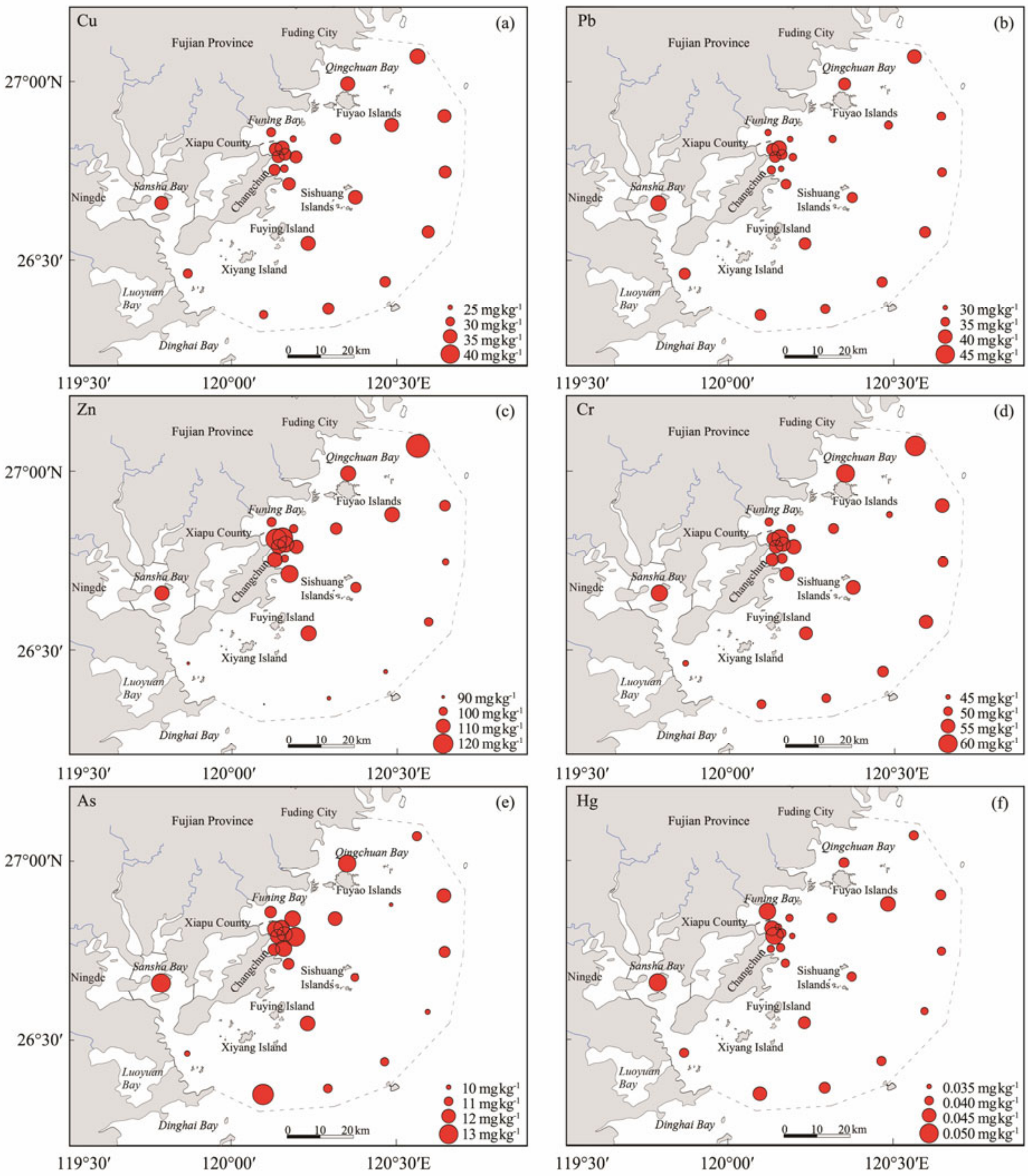

Fig.4 Distribution of Cu (a), Pb (b), Zn (c), Cr (d), As (e) and Hg (f) in the study area.

Table 1 Heavy metal concentrations in the surface sediments from the study area compared with other typical BIES around the world (unit: $\mathrm{mg\,kg^{-1}}$)

Areas	Location		Cu	Pb	Zn	Cr	Hg	As	Reference
Study area and adjacent sea areas	Coastal waters of Fujian Province (*n*=24)	Range	26.7–36.5	31.10–41.80	65.90–126.40	46.30–61.60	0.036–0.049	9.8–13.5	This study
		Mean	32.75	35.93	105.23	53.29	0.04	11.75	
		C.V.	7.6%	8.0%	12.1%	7.4%	9.0%	8.5%	
	Inner continental shelf of the East China Sea (*n*=59)	Range	18.8–49.2	24.4–52.9	80.2–140.5	45.1–142.8	–	7.5–15.8	Liu *et al.* (2011)
		Mean	37.40	34.13	116.03	93.48	–	–	
	Yangtze River Estuary (*n*=860)	Range	6.5–79.9	13.5–48.2	39.9–312.0	42.9–301.0	0.004–0.19	3.5–19.3	Bi *et al.* (2017)
		Mean	21.93	22.72	74.20	76.06	0.053	9.23	
	Minjiang River Estuary (*n*=175)	Range	0.13–44.30	13.00–83.60	10.60–158.80	1.23–105.00	0.00–0.22	2.39–14.48	Bi *et al.* (2017)
		Mean	21.09	35.80	88.54	54.41	0.054	8.83	
Typical BIES	Xiamen Bay, China (*n*=8)	Range	19–97	45–60	65–223	37–134			Zhang *et al.* (2007)
		Mean	44.0	50.0	139.0	75.0			
	Quanzhou Bay, China (*n*=175)	Range	24.8–160.6	37.0–93.2	120.4–324.5	52.8–166.0	0.011–0.350	–	Yu *et al.* (2016)
		Mean	60.81	66.98	186.70	84.72	0.107	–	
	Dammam, Arabian Gulf (*n*=26)	Range	32–790	0.6–9.4	1.2–106	11–111	1.2–2.8	11.8–51.3	El-Sorogy *et al.* (2018)
		Mean	297.29	5.25	48.26	63.79	2.02	30.61	
	Weihai coast waters，China (*n*=10)	Range	5.2–21.9	6.0–54.2	11.6–115.9	4.2–51.3	–	2.9–18.7	Li *et al.* (2017)
		Mean	11.6	20.0	40.0	23.9	–	9.0	
	Daya Bay, China (*n*=14)	Range	9.5–61.3	15.9–30.0	33.5–207.3	36.4–90.3	–	7.8–18.4	Liu *et al.* (2018)
		Mean	24.58	22.64	111.65	65.04	–	12.41	
	Thessaloniki Bay, northern Greece (*n*=32)	Range	21.3–180.1	29.4–195.4	48.6–538.2	65.5–173.5	–	–	Christophoridis *et al.* (2019)
		Mean	77.23	84.19	218	115.40	–	–	

Notes: *n*, number of sampling sites; C.V., coefficients of variation.

the concentrations of heavy metal elements in the study area were much higher than those in Weihai Coast sediments (Li *et al.*, 2017), but much lower than those in Thessaloniki Bay (Christophoridis *et al.*, 2019).

The coefficients of variation of heavy metals in the study area (Table 1), in decreasing order, were as follows: Zn (12.1%) > Hg (9.0%) > As (8.5%) > Pb (8.0%) > Cu (7.6%) > Cr (7.4%). The contents of all the heavy metals did not change much, and their spatial distribution was relatively balanced, with scattered low value and high value sites. For example, obvious high concentrations occur in Sansha Bay, Fuying Island Waters, Changbiao Islands Waters, and Qingchuan Bay to the north of the study area. Nevertheless, the distribution of Cu, Pb, Zn, and Cr may be basically considered to be the same. Furthermore, it was found that the distributions of these four heavy metal elements was clearly correlated with the grain size compositions of the surface sediments, *i.e.*, enriched in the fine sediments. Compared with the distributions of the other elements, those of As and Hg were clearly different. The high concentrations of As occur in the south of Sansha Bay, Fuying Island, Xiyang Island and Qingchuan Bay; whereas the high value areas of Hg are in the south of Sansha Bay, Xiyang Island and the inner waters of Funing Bay.

3.3 Numerical Simulation Results of the Tidal Current

The tidal current field of the whole study area was generated by numerical simulation. During the flood tide (Fig.5a), the tidal current velocity in the offshore area was generally lower than $40\,\mathrm{cm\,s^{-1}}$, and that of open sea was basically $40-80\,\mathrm{cm\,s^{-1}}$, while that of the isthmus between Changchun and Fuying Island and the mouth of Sansha Bay was even higher than $80\,\mathrm{cm\,s^{-1}}$. During the ebb tide (Fig.5b), the velocity around the most islands and in the Sansha Bay was greater than $40\,\mathrm{cm\,s^{-1}}$, while in relatively open sea, it was $0-40\,\mathrm{cm\,s^{-1}}$. The tidal residual current in the study area was extracted from the numerical simulation results covering a spring tide and neap tide cycle (Fig. 5c). It was found that the areas with high values for the residual current were mainly located in the southern and southeastern seas of the study area. In this case, the values varied between 5.0 and $14.0\,\mathrm{cm\,s^{-1}}$, with high outliers found at some isthmus or island heads. On the other hand, the areas with low residual current velocities were located in the middle and northeast of the study area, with the values being between 1.0 and $5.0\,\mathrm{cm\,s^{-1}}$. Overall, the direction of the tidal residual current was from the northeast to the southwest, although residual current vortices were also identified in several sea areas.

3.4 Partition of the Sedimentary Dynamic Environment in the Study Area

The sedimentary dynamic environment largely influences the migration and accumulation of sediments and control the distribution of heavy metal elements in the sediments (Sundaray *et al.*, 2011; Liu *et al.*, 2019a). In order to have an overall understanding of the sedimentary dynamic environment in the study area, the trigonometric map zoning method proposed by Pejrup (1988) is used to divide the sedimentary dynamic environment of the study area.

According to the partition results (Fig.6), only one station is located in zone AIV, 15 stations in zone BIII, 46 stations in zone DIII, and 206 stations in zone DIII. Zone AIV and BIII show relatively high energy sedimentary environments, which are mainly located in the southwest of the study area. Zone DIII implies that the material movement intensity in this area is low, and the sedimentary dynamic environment is relatively weak, which is mainly located in the southwest sea area, Funing Bay and the outer sea area. The study area is mostly divided into zone DIII and secondly in zone DIV, with higher clay percentages than other zones in the study area, which reveals weaker sedimentary dynamic environment. Therefore, the study area as a whole is in a weak sedimentary dynamic environment.

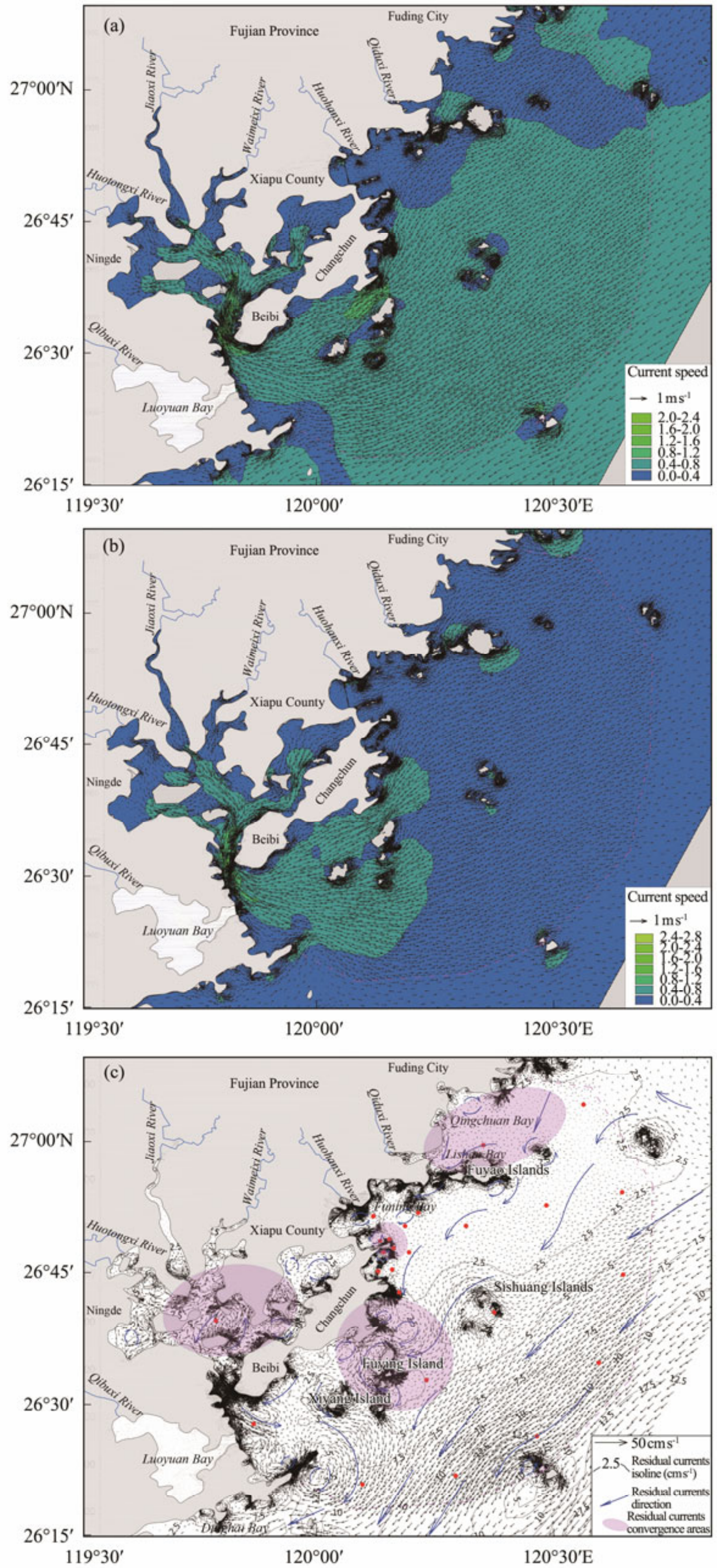

Fig.5 Tidal currents and tidal residual currents in the study area. (a), flood tide; (b), ebb tide; (c), tidal residual currents.

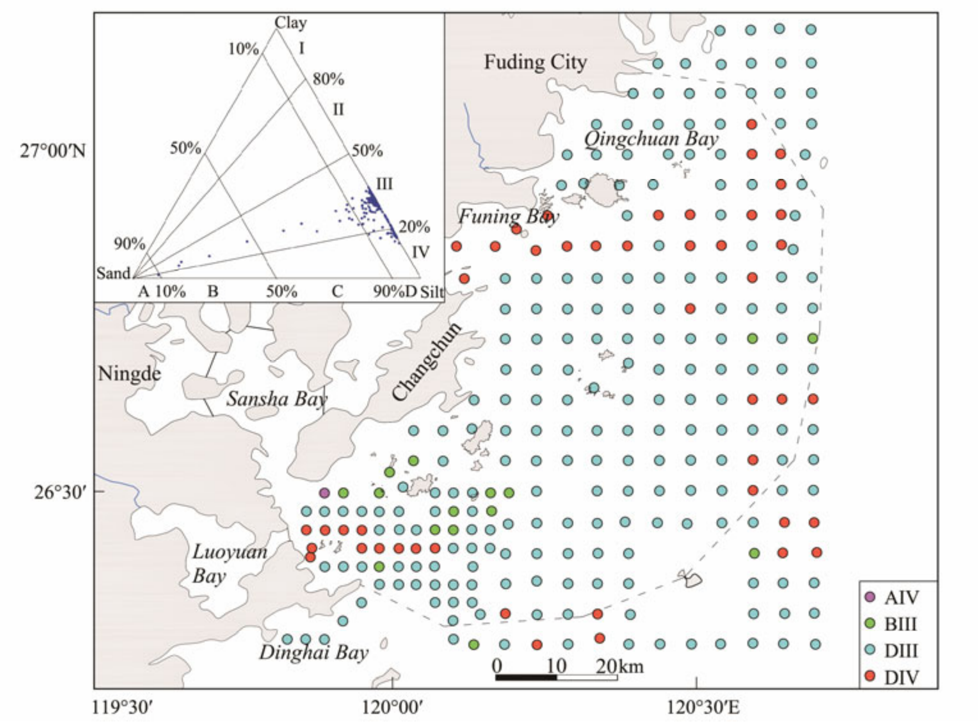

Fig.6 Sedimentary dynamic environment division in the study area.

4 Discussion

4.1 Effect of Grain Size Compositions

Significant correlations between the contents of two heavy metal elements or between the contents and D_{50} values may indicate that they share a common source or geochemical process (Zaharescu *et al*., 2009), and therefore, the correlation analysis was carried out for the study area sediments (Table 2). The results showed that Cu, Pb, Zn and Cr were highly and positively correlated at 0.01 significance level and in this case, a Pearson correlation coefficient value of up to 0.692 between Cu and Cr was obtained. However, Hg and As were not correlated with the other four elements and as such, it was speculated that the Cu, Pb, Zn and Cr could have similar sources or geochemical processes.

Table 2 Pearson correlation coefficients among the values of heavy metal element contents, D_{50}, TOC, pH, Eh and Sulfide contents

	Cu	Pb	Zn	Cr	Hg	As	D_{50}	TOC	pH	Eh	Sulfide
Cu	1										
Pb	0.633**	1									
Zn	0.658**	0.319	1								
Cr	0.692**	0.628**	0.639**	1							
Hg	0.070	0.212	−0.033	−0.133	1						
As	−0.138	0.089	0.001	0.325	0.122	1					
D_{50}	−0.681**	−0.653**	−0.571**	−0.789**	−0.035	−0.393	1				
TOC	0.305	0.489*	0.099	0.349	0.307	0.221	−0.159	1			
pH	−0.037	−0.034	0.044	−0.043	0.074	0.011	−0.116	−0.147	1		
Eh	−0.094	−0.158	0.283	0.196	−0.363	0.095	−0.217	−0.160	−0.117	1	
Sulfide	−0.032	0.136	−0.194	−0.110	0.315	0.132	0.107	0.198	−0.038	−0.806**	1

Notes: *, correlation is significant at the 0.05 level (two-tailed); **, correlation is significant at the 0.01 level (two-tailed).

The D_{50} values had significant negative correlations with Cu, Pb, Zn and Cr at 0.01 level, but were not correlated to either Hg or As. From Figs.3a and 4, it can also be found that the distribution of fine grain sediments was consistent with that of high concentrations of Cu, Pb, Zn and Cr. According to 'the control rules of grain size compositions to elements' (Zhao, 1983), the contents of most minor elements are negatively correlated with the amounts of coarse sediments but positively correlated with those of fine ones, thereby suggesting that, heavy metal elements were more likely to be enriched in fine sediments. Therefore, it can be considered that the grain size composition of surface sediments is an important factor affecting the distribution of Cu, Pb, Zn and Cr in the study area.

4.2 Effect of the Chemical Environment

The chemical parameters, such as pH, Eh, chlorinity, TOC and sulfide contents, of marine sediments in different sea areas exert different effects on the distribution of heavy metals. Many scholars have found that the chemical environment plays an important role in the enrichment of heavy metals of marine sediments (*e.g.*, Clark *et al.*, 1998; Fang *et al.*, 2005). However, other studies have also found that the chemical characteristics of some sea areas were not the major factor affecting the distribution of heavy metals. For example, Yavar Ashayeri and Keshavarzi (2019), who studied the Shadegan Wetland, found that the physical and chemical parameters of the sediments, in terms of electrical conductivity (EC), pH, cation exchange capacity (CEC) and TOC, had no significant correlation with the contents of heavy metals. Similarly, Yang (2006) studied the chemical environment of the Modaomen Estuary and found that pH values and chlorinity were not related to heavy metal elements, indicating that both parameters are not the main factors affecting their distribution.

In order to explore the influence of the chemical environment on the distribution of heavy metal elements, correlation analyses between heavy metal contents, TOC, pH, Eh and sulfide contents were conducted (Table 2). Overall, there was no obvious correlation between the heavy metal elements and other parameters, except for a weak one between Pb and TOC at 0.05 significance level (correlation coefficient 0.489). In addition, the correlation coefficient of −0.806 indicated significant but negative correlation between Eh and the sulfide content due to the direct influence of the redox environment on the concentration of sulfide. Indeed, as the Eh value increases, the sulfide in sediments may be oxidized and metal ions may be released, thus decreasing the sulfide content (Casas and Crecelius, 1994). Based on the results, it was, therefore, concluded that, compared with other factors, the chemical environment of the study area exerted little influence on the distribution of heavy metal elements.

4.3 Influence of the Sedimentary Dynamic Environment

4.3.1 Effects of the tidal current

According to the numerical simulation results, the tidal current velocity in the study area is relatively high. With the research results of Guo *et al.* (2021), surface sediments in most of the study area can be activated under the influence of tidal current, and the flood current and ebb current have significant resuspension and transport effects on sediments. Tidal residual current reflects the net transport direction of water body and is closely related to the long-term transport of materials. Tidal currents may also give rise to the residual eddy which is formed by the non-linear interactions between tidal currents and the variable topography at headlands (Guyondet and Koutitonsky, 2008), thus, suspended materials carried by the residual current often deposit in its vortex (Howarth and Huthnance, 1984). As shown in Fig.5c, four tidal residual currents convergence areas, usually located in bays and waters near islands, were found in Sansha Bay, Fuying Island Waters, Changbiao Island Waters and Qingchuan Bay. When combined with results of the grain size compositions and the distribution of heavy metals (Figs.3a and 4), it was found that these convergence areas correspond not only to regions with relatively fine sediments, but also those with high concentrations of Cu, Pb, Zn and Cr. This is because the grain size distribution of surface sediments is greatly affected by residual currents (Gao *et al.*, 1994; Cheng and Gao, 2000; Liang *et al.*, 2019). Indeed, tidal residual currents directly affect the transport and accumulation of heavy metal elements by moving and depositing the sediments. At the same time, based on 'the control rules of grain size to elements' (Zhao, 1983) as mentioned before, it is already known that fine particulate matter has a significant enrichment effect on heavy metal elements. Thus, tidal residual currents can further affect the enrichment effects by controlling the grain size distribution of surface sediments. In terms of these results aforementioned, it can be concluded that tidal currents play a significant role in the transport of heavy metal elements in the study area, while tidal residual currents (intensified by many bays and islands) play a dominant role in the accumulation of heavy metal elements.

4.3.2 Influence of the ocean circulation

The marine circulation system in the study area and adjacent areas includes the ZFCC and the TWC (Fig.1a), with some sites in the southwest of the ZFCC's influence area (Guan, 1983; Liu *et al.*, 2010; Liu *et al.*, 2011). The ZFCC is a monsoon circulation, mainly distributed in the coastal waters of Zhejiang and Fujian Provinces, and displays obvious seasonal variations. In winter, due to the prevailing northeast winds, the range of influence of the ZFCC reaches its maximum. Thereby, the low temperature/low salt coastal waters of Fujian and Zhejiang transport lots of materials from the northeast to the shallower southwest area (Liu *et al.*, 2009). However, due to the prevailing southwest winds in summer, the ZFCC flows to the northeast, leading to smaller velocity and smaller range of influence. In fact, Wang *et al.* (2018a) believed that, in summer, the ZFCC exerts little influence on the study area. Therefore, the maximum influence of ZFCC on the sediments in the study area is exerted as it flows from the northeast to the southwest. Many scholars have found that the sediments in the coastal waters of Fujian mainly came from the Yangtze River, the Qiantang River and the Oujiang River (Guo *et al.*, 2000; Shi *et al.*, 2010) and the amount of suspended sediments transported annually by the Yangtze River can reach up to 4.175×10^{8} t, 30 times that of the Qiantang River, the Oujiang River and the Minjiang River combined. Due to insufficient geochemical tracers (to identify the source) and physical oceanographic data (to determine transport mechanisms), the level of influence of the different rivers on the study area could not be determined definitely. However, it is clear, from previous studies, that, compared to the three rivers, the Changjiang River was the main source of sediments in the coastal area of Fujian (He, 1991). Under the action of the ZFCC, the sediments are transported from the northeast to the southwest, parallel to the shoreline and may pass through or deposit in the study area as a result of weak hydrodynamics (usually forming fine-grained deposits) (Liu *et al.*, 2011). Wang *et al.* (2020a) also believed that the southwesterly ZFCC has a significant influence on the distribution of heavy metal elements in the Southern Inner Shelf where the study area is located. Therefore, it can be speculated that the ZFCC also exerts a certain influence on the transport and enrichment of heavy metals in sediments from the study area. The TWC, on the other hand, is a mixture of seawater mainly from the Taiwan Strait and the Kuroshio. It basically travels northeast along the 50 m isobath line, and as such, it has little influence on the transport of materials or heavy metal enrichment in the study area (Bao *et al.*, 2005; Liu *et al.*, 2007).

4.4 Effects of Human Activities

The geological accumulation index was used to evaluate heavy metal pollution of surface sediments. This index is given by:

$$I_{\text{geo}} = \log_2 \left[C_i / (k * B_i) \right], \tag{1}$$

where, C_i and B_i are respectively the content of heavy metal element i in the sample and the geochemical background value. k is a constant, usually valued at 1.5. Seven classes of I_{geo} have been defined, starting from the unpolluted level (indicated by $I_{\text{geo}} < 0$) up to the extremely polluted one (Müller, 1981). In this study, B_i represented the background value of the soils from the coastal zone of Fujian Province (Liu, 1995), which are sampled in 1987, when the level of urbanization and industrialization were relatively low. The results showed that the average I_{geo} value for each of the six heavy metals decrease as following order: As (0.29) > Cu (−0.04) > Cr (−0.20) > Zn (−0.26) > Pb (−0.71) > Hg (−1.20). Therefore, heavy metal elements were almost all at the unpolluted level in the surface sediments except for As, for which the high I_{geo} values were mainly concentrated in Sansha Bay, Funing Bay, Qingchuan Bay and the southeast sea area.

Principal component analysis was also carried out for the contents of the six heavy metal elements. The Kaiser-Meyer-Olkin (KMO) measure of sampling was 0.692 (> 0.50) with a significance level of less than 0.05, so the data was considered to be suitable for principal component analysis. The standardized heavy metal contents were taken as variables and the varimax rotation was performed to obtain the results of the principal component analysis (Table 3). The first three factors with eigenvalues greater than 1 were then extracted, and since they could explain 46.85%, 19.85% and 17.60% of the total variance (a cumulative variance contribution of 84.30%), these three factors could reflect most of the information on the heavy metal elements in the surface sediments of the study area.

Table 3 Loads and cumulative variances of heavy metals on the first three PCs

PCs	PC1	PC2	PC3
Cu	0.91	−0.19	0.10
Pb	0.74	0.10	0.38
Zn	0.80	−0.04	−0.16
Cr	0.87	0.37	−0.15
Hg	−0.02	0.05	0.96
As	0.01	0.98	0.07
Variability (%)	46.85	19.85	17.60
Cumulative (%)	46.85	66.70	84.30

It was seen from Table 3 that the load of Cu, Pb, Zn and Cr on PC1 was relatively high (0.74–0.91). Combined with previous findings that the distribution of Cu, Pb, Zn and Cr was mainly affected by natural factors such as grain size compositions and the hydrodynamic environment, it can be deduced that the variations along with PC1 are mostly mediated by natural processes.

In the case of PC2, As had a high load (0.98) while Cr had a relatively low one (0.37). The areas with high As contents were mainly concentrated in Sansha Bay, Changbiao Island, Qingchuan Bay and the southern waters of Xiyang Island, close to the land and estuaries, where the influence of land agricultural production may be significant. Indeed, the river basin in the study area is industrially and agriculturally developed and this encourages the use of chemical fertilizers and pesticides in which heavy metal elements such as As and Cr are important components (Pan and Wang, 2012; Tian *et al.*, 2020). Therefore, agricultural wastewater may be considered to be the main source of As, most probably of Cr, in the sea area. In addition, the aquaculture industries in these waters are densely distributed. Due to the excessive use of As-containing baits in aquaculture areas, the As content of some feeds used by the fishermen of Fujian also exceeds the standard (Tu *et al.*, 2011). Altogether, these different sources cause the river to carry large amounts of pollutants produced by human activities to estuaries (Fan *et al.*, 2021). Hence, PC2 represented the influence of human factors related to the land agricultural production, which is aggravated by the enrichment process in estuaries, as well as marine aquaculture.

Hg had a high load (0.96) on PC3, with Pb which had a low one (0.38). Human pollution is the main factor affecting the Hg content in surface sediments of the coastal waters of Fujian and Zhejiang (Zhang, 2015). In Fig.4f, the areas with high Hg contents were mainly concentrated in the inner waters of the Sansha Bay and the Funing Bay, along with the southern waters of Xiyang Island, where many rivers flow into. These river basins are industrially developed, and it is believed that rivers, polluted by the industrial discharge of Hg, could be the main source of this element in the sea area. In addition, these regions also possess developed aquaculture and shipping industries and this is especially the case for the southwest sea area where waterways and many ports are present (Fig.7). Since the fuel oil used in ships contains both Hg and Pb, fuel gas and oil leakage may not only be the source of Hg in the sea area, but they may also influence the distribution of Pb. Therefore, PC3 represented the influence of human factors mainly in terms of industrial manufacturing on land and shipping.

4.5 Source to Sink System and Factors Controlling Heavy Metal Elements in the Surface Sediments of a Typical Bay-Island-Estuary System (BIES)

The concentrations of Cu, Pb, Zn, Cr, Hg and As were analyzed by Q-type hierarchical cluster analysis. The Ward clustering method was used for cluster analysis, and the average Euclidean distance method was used for interval measurements. Through the hierarchical cluster analysis, the study area was divided into three sea areas namely Zoning I, Zoning II and Zoning III (Fig.7).

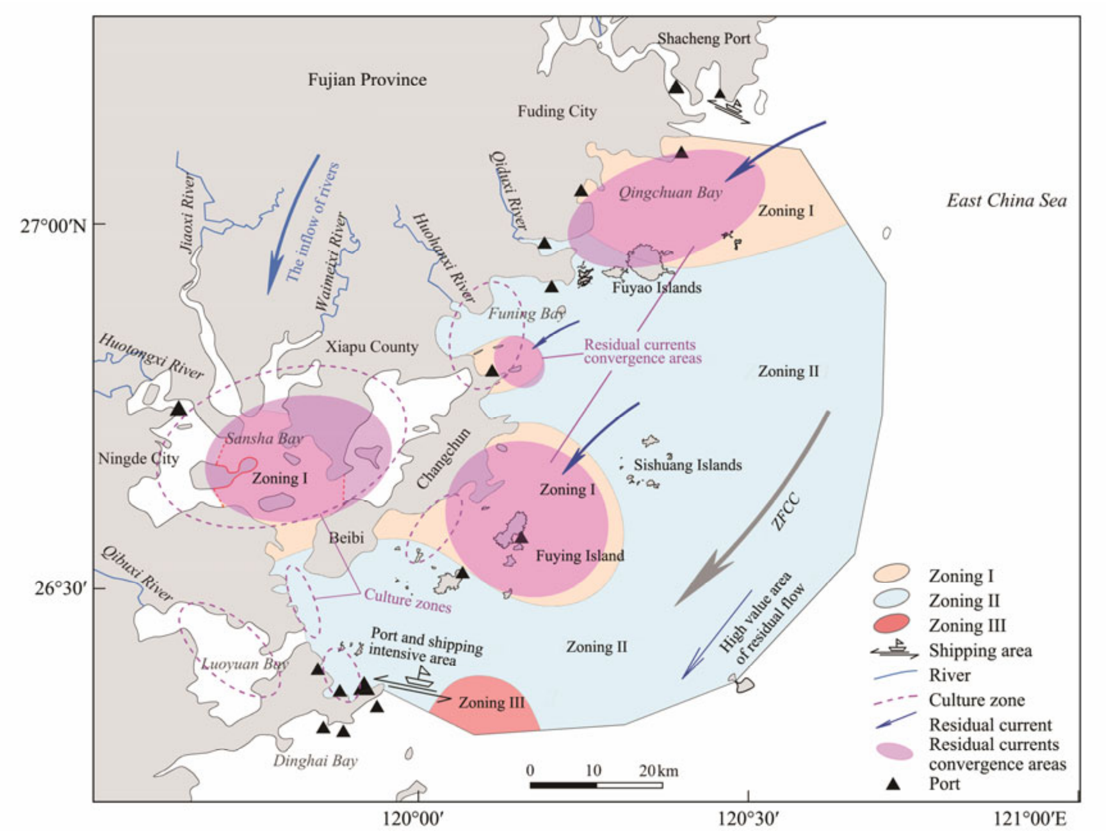

Fig.7 Environmental zoning of heavy metals in surface sediments from Fujian Province coastal areas.

Zoning I was mainly located in islands, bays and estuaries, with highest contents of heavy metal elements. Zoning I was distributed in the zone DIII of the sedimentary dynamic division, which represents a relatively weak depositional dynamic environment. the Zoning I is consistent with the fine sediment area (Fig.3a), as well as the tidal residual currents convergence areas (Fig.5c). Therefore, the transport and enrichment of heavy metal elements in the Zoning I were mainly controlled by natural factors such as the surface sediment type and the sedimentary dynamic environment. Similarly, the discussion of Section 4.4 pointed out that the enrichment of heavy metal elements in this zone was also affected by human factors. Therefore, the pollution of heavy metals in this sea area was affected by both natural and human factors, although the natural factors were more dominant. Zoning II was mainly distributed in the outer sea area in the southern part and the sea area from Funing Bay to the eastern part of the study area. The Zoning II was mainly corresponded to the zone DII and DIV for the partition of the sedimentary dynamic environment, and was in a relatively weak depositional dynamic environment as a whole. The contents of heavy metals in this zone were lower than those in other study areas. Heavy metal sources include human sources and natural ones, with the latter including mainly parent rock weathering and coastal erosion (Zhang *et al*., 2017; Xu *et al*., 2018). Based on earlier analyses, human activities basically had no impact on the concentration of heavy metal elements in this region. Therefore, the heavy metals in the sediments from this sea area were more likely to come from geological processes. In general, the overall sedimentary dynamic environment in the study area was weak, and there was no significant correspondence between the partition of the sedimentary dynamic environment and the distribution of heavy metal elements. However, there was a significant correspondence between the convergence area of tidal residual currents and the high concentration area of heavy metals in the study area. Therefore, we believed that tidal residual currents played a leading role in the dis- tribution of heavy metal elements in the study area under the weak sedimentary dynamic environment.

Finally, Zoning III was distributed in the outer sea area, in the south of the study area. Zoning III was distributed in zone DIII for the partition of the sedimentary dynamic environment, which was in a weak depositional dynamic environment. The concentration of As and Hg in this zone was high, while the contents and pollution levels of the other heavy metal elements were relatively low. With reference to the above discussion on the impact of human activities, the source of As and Hg in Zoning III could be attributed to human sources such as mariculture pollution and shipping. In this context, it is worth noting that reducing the use of chemical fertilizers and pesticides in agriculture while eliminating the use of As-containing baits in mariculture could be effective means to minimize heavy metal pollution in this sea area.

Taking the research on the coastal waters of Fujian Province as an example, and based on previous researches, we comprehensively discussed the controlling factors of heavy metal elements in a typical BIES (Fig.8). The transport of heavy metals in coastal waters is mainly affected by natural and human factors (Xu *et al*., 2018), with the former including mainly the characteristics of the sources, the river inflow, the regional background, the sediment grain size, the physical and chemical environment, the tidal current, the ocean circulation and the atmospheric precipitation (Liu *et al*., 2019b). Human factors, on the other hand, mainly include industrial manufacturing, urban domestic wastewater, agricultural production, mariculture, port shipping and engineering construction (Wang *et al*., 2020a).

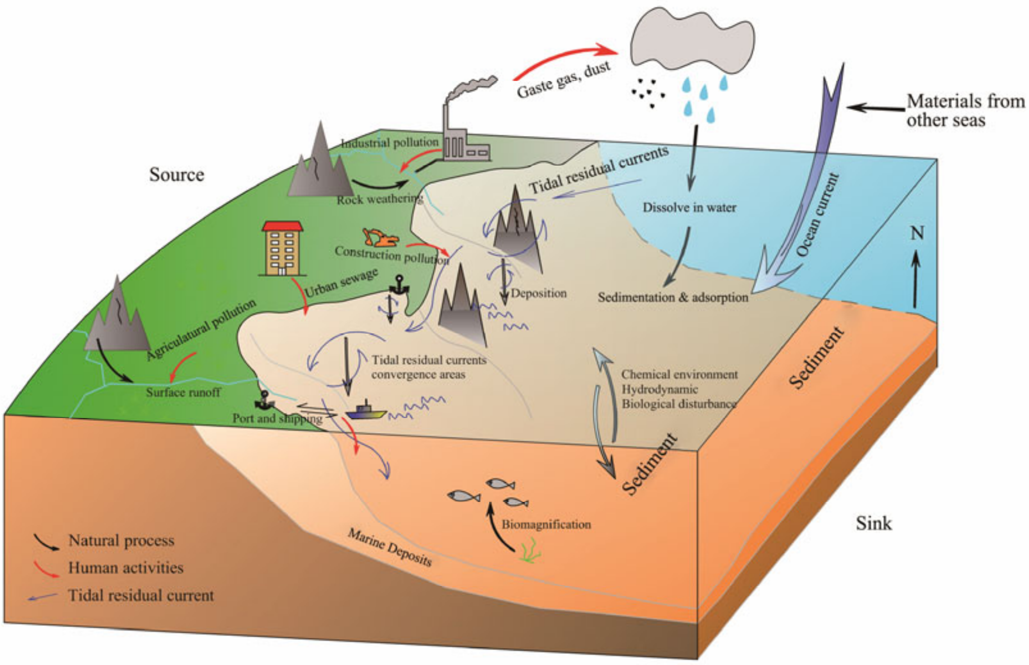

Fig.8 Model diagram of source to sink system for heavy metal elements in BIES surface sediments.

From this study, it was found that the complex hydrodynamic conditions caused by bays and islands had a significant effect on the transport of heavy metals in the BIES. The interior of the bay is usually the convergence area of tidal residual currents, and the sediments, carried by the tidal residual currents, therefore, deposit and accumulate in that region. Furthermore, due to the blocking effect of islands and their wake effect, islands have a significant trapping effect on the suspended matter (Pingree and Maddock, 1980; Rissik *et al*., 1997; Dong *et al*., 2009). Ocean circulations such as coastal currents, wind-ocean currents and thermohaline circulations are characterized by long transportation distances and large time scales (Chiri *et al*., 2019) and as such, they can also carry sediments from other sea areas to deposit in the bays or around the islands with weaker hydrodynamic conditions. Finally, there is no doubt that the river inflow has important influence on the accumulation of heavy metals in the BIES. Indeed, rivers (especially large ones) carry heavy metal elements, generated by parent rock erosion and human activities, from the 'source' area into the coastal waters, thus affecting the enrichment of heavy metal elements in the 'sink' area (Zhang and Liu, 2002). At the same time, rivers can also change the sedimentary dynamic environment of estuaries and thus affect the distribution of elements in estuarine waters (de Souza Machado *et al*., 2016).

Heavy metal elements contained in dust particles and exhaust gases on land areas can also enter the ocean water through atmospheric processes, thereby accumulating in sediments. However, it is generally believed that atmospheric deposition contributes less to the heavy metal elements in the surface sediments from the estuarine seas compared with other factors (McKee *et al*., 2004; Viers *et al*., 2009). Through the study of Cd, Cu, Pb and Zn in the Scheldt Estuary, Zwolsman (1993) found that the contribution of atmospheric deposition to the total amount of heavy metals in estuarine sediment was negligible. Similarly, even though Hg also enters the ocean through the atmosphere, the amount collected from atmospheric deposition in the nearshore ocean is relatively small compared to the open ocean (Yin *et al*., 2015; Sun *et al*., 2020). These studies indicate that atmospheric deposition contributes little to heavy metal elements in estuarine surface sediments.

According to the research in this paper, we suggest that the hydrodynamic conditions, which control the transportation of sediments from every source and regulate the sediment particle size compositions, might have a significant influence on the distribution of heavy metals in the BIES, whereas the chemical environment (TOC, pH, Eh, sulfide, *etc*.) do not work clearly.

5 Conclusions

Based on the contents of six heavy metals in surface sediments from coastal areas of Fujian Province, this paper focuses on the influence of the hydrodynamic environment, and systematically discusses how grain size compositions, chemical environment, tidal current, ocean circulation and human activities influence the distribution and transport of the heavy metals. The distribution characteristics of Cu, Pb, Zn and Cr in the study area are similar, but they are different from those of As and Hg. The distribution and migration of Cu, Pb, Zn and Cr were mainly controlled by natural factors including regional background, sediment grain size compositions, and tidal residual currents. By the correlation analysis, we found that the chemical properties of the environment exerted limited influences on the distribution of heavy metals in this BIES. The enrichment of As was mainly controlled by human factors such as agricultural production and marine aquaculture, and Hg was influenced by industrial manufacturing and shipping. large amounts of pollutants carried by rivers are settled or precipitated in the estuaries, thereby aggravating the heavy metal pollution. In addition, zones around bays and islands are depositional center of fine sediments, resulting in the enrichment of heavy metals. The weak hydrodynamic regions and tidal residual currents convergence areas are of great significance for the trapping and enrichment of heavy metals. For a typical BIES such as the area investigated in this study, the source, transport and distribution of heavy metals are affected by both human activities and natural factors, especially the hydrodynamic environment. In future studies, we will further explore the response of heavy metal pollution to sedimentary dynamic environment under extreme events and over long time scales.

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