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# Distribution, origins, impact factors and ecological risks of metallic elements in the sediment from Changhua River estuary, Hainan Island, China

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Within the framework of global biogeochemical cycles, estuaries function as a critical interface that connects terrestrial and marine ecosystems. We assessed the heavy metal pollution risk in the Changhua River estuary area. The assessment focused on pollution characteristics, spatial distribution, and ecological risks, while also identifying potential origins of metallic elements through the application of correlation analysis, principal component analysis (PCA), and Positive Matrix Factorization (PMF) models. The results showed that: (1) Sediments were predominantly sandy and weakly alkaline properties, with significant spatial heterogeneity in metallic concentrations. The concentration of metals, compared to their background values (BVs), could be ranked as: Nearshore: Pb > As > Cd > Cu > Zn > Hg > Ni > Cr; Inshore: Cd > Hg > Pb > Zn > Cu > As > Ni > Cr; Estuary: Pb > Cd > Hg > Cu > Zn > Ni > Cr. Particularly, Cd and Hg in inshore sediments registered a mean concentration surpassing their BVs by 417.95% and 187.33%, respectively. (2) The sampling locations presented varying levels of ecological risk. Moderate ecological risk ( $150 \leq RI < 300$ ) accounted for 24.14% of the sites, primarily located in inshore and estuarine areas, while extreme ecological risk ( $300 \leq RI$ ) was observed in 6.90% of the sites, concentrated within inshore regions. Hg and Cd were identified as the most hazardous metals. (3) Regarding metallic sources, Hg mainly originated from coal burning, while Pb, Zn, and Cd were likely linked to anthropogenic activities associated with port operations. In contrast, Cr, Ni, and Cu were primarily derived from natural and agricultural sources, and As was mainly associated with the residual accumulation of fertilizers and herbicides. Overall, human activities, including agriculture, aquaculture, and port transport, are the primary origins of metallic contamination in the estuarine sediments. Effective management and

regular monitoring of port activities are essential for the governance of the estuarine environment. Such practices can yield valuable insights for continuous risk analysis, thereby supporting the ecological health and resilience of these vital environments.

#### KEYWORDS

sediments, metallic contamination, source apportionment, estuary area, Hainan Island

## 1 Introduction

Estuaries function as transitional zones between rivers and oceans and can provide essential ecosystem roles and benefits (Guen et al., 2019; Boicourt, 1983). These areas frequently accumulate and retain pollutants originating from human activities, such as nutrient compounds, polycyclic aromatic hydrocarbons, and metallic elements (Bantan et al., 2020; Guo et al., 2020; Kumar et al., 2015). As reported, substantial anthropogenic inputs have placed considerable pressure on estuaries and contributed to increased pollution levels (Negrin et al., 2021; Yin et al., 2016; Chapman and Wang, 2001). Estuarine systems are shaped by dual forces: river-derived elements like sediment transport and freshwater flow, as well as oceanic influences such as tidal patterns and saline mixtures (Cooper, 2001; Sherwood et al., 1990). The mixing of saline and freshwater induces the flocculation and deposition of fine-grained particles and heavy metals, elevating pollutant concentrations in the water column and sediments, making estuarine environments among the most contaminated ecosystems globally (Hamzeh et al., 2016; Sánchez-Moyano and García-Asencio, 2010; Zhang and Savenije, 2018; Lopez et al., 2020; Hu et al., 2024). The prevalence and enduring nature of metallic contaminants have emerged as a significant issue for environmental researchers, owing to their capacity for bioaccumulation, ecological harm, and resistance to natural degradation processes (Truchet et al., 2021; MaChado et al., 2016; Papagiannis et al., 2004). Metallic elements exist in various environmental media, including soil, suspended particulate matter, water, organisms, and sediment. The concentrations of metals dissolved in water remain relatively low (Kumar et al., 2020; Xie and Wang, 2020; Wang et al., 2012), while suspended particulate matter acts as a major carrier for these metals (Zeng et al., 2020; Yao et al., 2016).

Sedimentary layers exhibit a notable ability to retain metallic elements, frequently functioning as a repository or accumulation site. Additionally, it has the potential to release these elements into the water column, which can result in secondary pollution in the environment (Xiao et al., 2021; Ahmad et al., 2020; Yi et al., 2011; Pekey, 2006). The spatial patterns of metallic elements within sedimentary layers are strongly linked to sediment attributes, shaped by external influences such as absorption potential and hydrological linkages (Jia et al., 2021; Li et al., 2020; Bantan et al.,

2020). Therefore, gaining a comprehensive insight into sedimentary conditions is essential for understanding the buildup of metallic contaminants in estuarine sediments (Bi et al., 2017; Ye et al., 2012; Krupadam et al., 2007). Nevertheless, a knowledge gap exists regarding the influence of the sedimentary environment on the characteristics of metallic contaminants in estuaries. Further research is necessary to elucidate the movement of metallic contaminants from estuarine deposits into adjacent coastal marine areas.

Metallic elements can derive from natural processes, including the geochemical breakdown of soil and rock formations, as well as anthropogenic activities like farming practices, resource extraction, and manufacturing operations (Zhao et al., 2020a; Jia et al., 2020; Pekey, 2006). Various evaluation indexes have been applied to assess metallic contamination, such as the geo-accumulation index (Igeo) (Muller, 1969), the contamination factor (CF) (Hakanson, 1980), the enrichment factor (EF) (Sutherland, 2000), and the potential ecological risk index (Hakanson, 1980). The use of background values (BVs) is a fundamental requirement for accurately evaluating metallic contamination using these indices. Currently, the establishment of geochemical baselines, the assessment of pre-industrial sediment metallic concentrations, and the determination of soil BVs in surrounding areas emerge as the three dominant methods for establishing sediment metallic BVs (Wang et al., 2019; Chen et al., 2019). Nevertheless, these approaches possess inherent constraints. The geochemical baseline reflects a snapshot of concentrations at a particular moment, encompassing both natural and human-induced factors, which may lead to an underestimation of sediment metallic contamination (Wang et al., 2019). The use of the pre-industrial metallic concentrations is appropriate for small-scale studies, such as those conducted in lakes with specific sedimentation rates; however, inaccuracies can arise when analyzing extensive drainage basins, primarily due to variations in sedimentation rates (Gu et al., 2022). Applying soil BVs as proxies for sediment BVs is effective under specific geographical scenarios, like regions near riverbanks, but may introduce significant biases in other environments (Luo et al., 2022). Therefore, selecting an appropriate method to establish BVs will enhance the precision of assessments regarding metallic contamination in sediments. In addition to evaluating metallic contamination in sediments, identifying the origins of metallic pollutants is crucial for effective

pollution management. The positive matrix factorization (PMF) model has proven to be effective as a receptor-based method, widely utilized in numerous investigations to discern and measure the origins of metallic elements in both soil and sedimentary samples (Jiang et al., 2024; Proshad et al., 2022; Wu et al., 2021). In recent years, research on trace metals in Hainan has increased; however, most studies have primarily focused on individual river estuaries, coastal waters, or specific sections of estuarine environments, with limited attention given to the distribution and transformation of heavy metals (HMs) within an integrated river-estuary-ocean dynamic system. Moreover, existing studies are often constrained by insufficient sampling density, and assessments of pollution levels and potential ecological risks predominantly rely on single-index methods, limiting analytical depth. Currently, pollution source apportionment lacks comprehensive approaches such as multivariate statistical analysis, isotopic tracing, or model coupling. Additionally, the influence of hydrodynamic conditions on sedimentary heavy metal concentrations remains underexplored, necessitating more systematic and in-depth investigations (Xia et al., 2011; Fu et al., 2013; Hu et al., 2013; Kuang et al., 2021; Fu Z. et al., 2023; Fan et al., 2024).

To address existing research gaps, this investigation conducted an in-depth examination and source identification of metallic elements in surface sediments from the downstream and estuarine sections of the Changhua River. The study aimed to: (i) evaluate and contrast the accumulation of metallic pollutants, including As, Cd, Cr, Cu, Hg Ni, Pb, and Zn, in sediments using numerical calculations and index-based analytical methods; (ii) map the spatial distribution of these elements and identify high-concentration zones through spatial analysis techniques; and (iii)

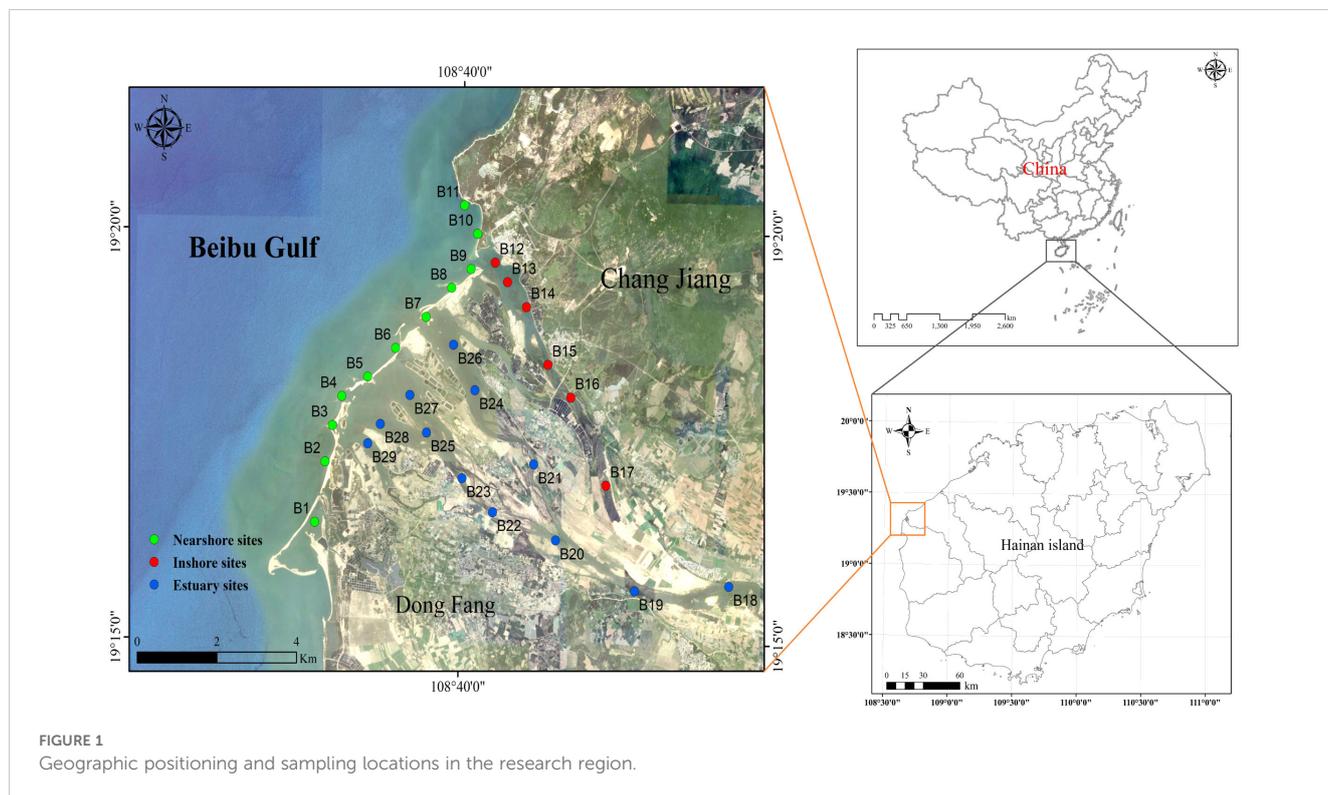
uncover the key drivers influencing the sources of metallic pollutants in sediments by integrating PMF modeling, correlation analysis, and principal component analysis (PCA). The findings of this study will enhance the understanding of the driving mechanisms of sedimentary metal pollution, particularly in economically developed regions characterized by complex geomorphology and dynamic hydrological conditions. Furthermore, the results provide a theoretical foundation and scientific support for sediment metal pollution management, contributing to the sustainable development of similar riverine environments.

## 2 Materials and methods

### 2.1 Sample collection, processing, and analysis

#### 2.1.1 Sample collection

Figure 1 displays a geographic representation of the research region, encompassing a wide array of sampling locations. The region was categorized into three distinct regions based on the region's human activity patterns (agriculture, industry, fisheries, and urban distribution), hydrodynamic conditions, and geographical characteristics (open rivers and semi-enclosed bays) (Wu et al., 2024, 2025), i.e., nearshore (B1-B11), inshore (B12-B17) and estuary (B18-B29). The estuarine area contains intensive aquaculture zones, while the inshore area is a semi-enclosed marine zone characterized by significant shipping activity and functions as an actively used harbor. Due to the semi-enclosed



nature of the inshore sea, substantial volumes of agricultural and household effluents are released into the Beibu Gulf through multiple upstream waterways, leading to local accumulation of pollutants (Wang and Wang, 2007).

### 2.1.2 Data sources and quality control

In July 2023, the project team collected surface sediment samples from 29 sites. All sampling locations were georeferenced using a Global Positioning System (GPS) receiver, and samples were collected from estuaries, bays, and nearshore areas to represent the entire estuarine region. At each sampling site, surface sediments (0–10 cm) were collected from three positions at 3-meter intervals. The procedures for sample collection, storage, and transportation of samples were carried out in strict accordance with the requirements of the code. All samples were kept clean during the sample collection, transportation, and storage.

The sediment samples collected in this study were analyzed at the Analytical Testing Laboratory of the Haikou Marine Geological Survey Center. Before measurement, the samples were air-dried, ground, and sieved through a 160-mesh sieve, followed by microwave digestion to prepare the test solutions. The concentrations of Ni, Cu, and Zn were measured using an X-ray fluorescence spectrometer (Axios Pw4400), while As and Hg concentrations were determined using a dual-channel atomic fluorescence spectrometer (AFS-8530). Additionally, Cr, Cd, and Pb were analyzed using an inductively coupled plasma mass spectrometer (ICP-Q). The grain size was determined according to the guidelines specified in the Specification for Marine Investigation (GB12763.1-2007, State Oceanic Administration, PR China), and sediment grain size analysis was conducted using a PSA 1190 LD laser diffraction particle size analyzer.

All analyses were performed in triplicate, and the accuracy and precision of the experiments were assessed using standard reference materials. The standard deviation was less than 10%, and the recovery rates of the standard materials ranged from 89% to 107%. The significance level was set to  $p \leq 0.05$ . The detection limits for Cu, Cr, Ni, Pb, Zn, As, Hg, and Cd were 1.2, 2.0, 1.5, 2.0, 2.0, 0.01, 0.002, and 0.07  $\text{mg}\cdot\text{kg}^{-1}$ , respectively.

## 2.2 Methods for ecological risk assessment

### 2.2.1 Geo-accumulation index

The  $I_{\text{geo}}$  index serves as a comparative metric for assessing metallic concentrations in soil samples relative to their BVs (Xu et al., 2019). Since the late 1960s, this index has been extensively utilized to assess contamination from metallic elements in soil. The formula for determining the  $I_{\text{geo}}$  is presented as below:

$$I_{\text{geo}} = \log_2 \left( \frac{C_n}{1.5 \times B_n} \right) \quad (1)$$

In Equation 1, where  $C_n$  represents the detected level of the metallic element in the collected samples, while  $B_n$  denotes the geochemical background concentration. The constant 1.5 accounts for potential natural variability in BVs. Generally,  $I_{\text{geo}}$  can be

TABLE 1  $I_{\text{geo}}$  grading standards (Wolswijk et al., 2020).

$I_{\text{geo}}$	Grade	Pollution Degree
$I_{\text{geo}} \leq 0$	0	No pollution
$0 < I_{\text{geo}} \leq 1$	1	Mild - moderate pollution
$1 < I_{\text{geo}} \leq 2$	2	Moderate pollution
$2 < I_{\text{geo}} \leq 3$	3	Moderate - severe pollution
$3 < I_{\text{geo}} \leq 4$	4	Severe pollution
$4 < I_{\text{geo}} \leq 5$	5	Severe-very severe pollution
$I_{\text{geo}} > 5$	6	Very severe pollution

categorized into seven levels, as indicated by the grading standards displayed in Table 1.

### 2.2.2 Single-factor pollution index

The PI index has emerged as an effective tool for long-term pollution monitoring. This index offers a straightforward yet robust method for quantifying metallic contamination and assessing the impacts of anthropogenic actions (Chai et al., 2021; Tian et al., 2017). Table 2 illustrates the contamination levels categorized according to the PI index (Wang et al., 2022; Cai et al., 2019a). The formula for calculating the PI index is presented as below:

$$PI = \frac{C_i}{S_i} \quad (2)$$

In Equation 2, where  $C_i$  denotes the detected level of a pollutant, and  $S_i$  indicates the BV of the pollutant.

### 2.2.3 Assessment framework for ecological risk

The RI index is frequently employed to assess possible environmental hazards in sediment (Hakanson, 1980). The evaluation formula is expressed as follows:

$$EI = T_i \times PI_i \quad (3)$$

$$RI = \sum_{i=1}^n EI = \sum_{i=1}^n (T_i \times PI_i) \quad (4)$$

In Equations 3 and 4, where RI refers to the overall ecological hazards posed by metallic elements, EI denotes the specific risk associated with individual metals, and  $T_i$  indicates their toxicity coefficients. The respective response coefficients for Ni, Cr, Cd, Hg, As, Pb, Zn and Cu are 5, 2, 30, 40, 10, 5, 1, and 5, as documented in

TABLE 2 Pollution gradation based on PI values.

Single-factor pollution index (PI)	Pollution gradation
$PI \leq 1$	Safe
$1 < PI \leq 2$	Warning threshold
$2 < PI \leq 3$	Slight contamination
$3 < PI \leq 5$	Moderate contamination
$PI > 5$	Severe contamination

studies by [Yüksel et al. \(2022\)](#) and [Fang et al. \(2019\)](#). Table 3 provides the updated classification of comprehensive RI levels ([Fei et al., 2019](#); [Hakanson, 1980](#)).

## 2.2.4 Source analysis of metallic contamination in sediment

### 2.2.4.1 Coefficient of variation

The CV can be used as a robust metric for assessing the spatial heterogeneity of metallic concentrations in sediment. According to the research by [Phil-Eze \(2010\)](#), CV values below 20% indicate low variability, values between 20~51% reflect moderate variability, and values ranging 51~100% denote high variability. Typically, a low CV suggests the predominance of diffuse or uniform pollution sources, whereas a high CV is indicative of notable differences in pollutant sources, often associated with discrete point-source inputs.

### 2.2.4.2 Positive matrix factorization

EPA PMF 5.0 is a positive definite short-matrix factorization tool developed by the United States Environmental Protection Agency, which is widely used in environmental data analysis, including atmospheric particulate matter, water quality and sediment. The model decomposes the observed environmental data into a factor contribution matrix and a factor spectrum matrix. Helps identify potential sources of pollution and quantify their contribution to pollutant concentrations in the environment. In addition, it is a multivariate factor analysis technique, employs the least iterative squares algorithm ([Vaccaro et al., 2007](#)). Unlike other methods, it does not necessitate predefined source or receptor component spectra. During factorization, error estimation is applied to individual data points, enhancing the treatment of missing or erroneous data. This method avoids generating negative values in matrix decomposition, thereby improving result reliability ([Yu et al., 2021](#)). The PMF model breaks down the original receptor data matrix (X) into three components: a factor score matrix (G), a factor loading matrix (F), and a residual matrix (E) ([Norris et al., 2014](#)). The equation for this decomposition can be expressed as follows:

$$X_{ij} = \sum_{k=1}^p G_{ik}F_{kj} + E_{ij} \tag{5}$$

In Equation 5, where  $p$  stands for the total number of factors, while  $i$  denotes a specific sample, and  $j$  indicates a particular element. The matrix  $X_{ij}$  is constructed from the concentration of element  $j$  in sample  $i$ .  $G_{ik}$  signifies the contribution of factor  $k$  to sample  $i$ ,  $F_{kj}$  describes the species distribution for each source and  $E_{ij}$  represents the residual matrix. All parameters in the above calculation process are unitless.

In the calculation process, the signal-to-noise ratio of the data needs to be analyzed first, If the signal-to-noise ratio (SNR) is greater than 2, the data are suitable for the model. The number of factors needs to be run and adjusted for many times, and the Q value, residual value and  $R^2$  are fully considered to make the calculation results stable. Then, the predicted value of the model is compared with the measured value. The fitting curve  $R^2$  is above 0.5, indicating that the measured value is highly correlated with the simulated value, and the representative factor number can be selected.

## 2.3 Data analysis and processing

Descriptive statistics were conducted using Microsoft Excel 2022. Origin 2024 was utilized to conduct correlation analysis and PCA modeling for determining the sources of metallic elements in the soil. The EPA PMF 5.0 tool was applied for source apportionment, while ArcGIS 10.8 was used to create maps of the research region and sampling locations. All other figures were generated using Origin 2024.

## 3 Results and discussion

### 3.1 Analysis of particle size distribution in the sediments

The influence of sediment grain size on metallic concentrations is multifaceted. The ternary diagram in [Figure 2](#) illustrates the distribution of sediment particle sizes. The data reveals that the surface sedimentary deposits are predominantly fine-grained and sandy in composition. Fine-grained sediments possess a larger specific surface area and a greater adsorption capacity, which typically leads to the immobilization of more metallic elements, thereby reducing their bioavailability and mobility ([Wang et al., 2013](#); [Chen et al., 2016](#); [Zhou et al., 2020](#)). In contrast, coarse-grained sediments often lead to the migration and bioaccumulation of metallic elements. Studies indicate that nearshore sediments are predominantly composed of sand, accounting for 70.64 - 98.26% of the total sediment composition, while fine-grained sediments contribute only 0.8 - 29.36%. Fine-grained sediments in the nearshore inner shelf of the study area are influenced by fluvial inputs, tidal forces, and wave energy, which collectively regulate sediment transport and the spatial distribution of metals. ([Wentworth, 1922](#); [Syvitski et al., 2005](#); [Xiao et al., 2016](#); [Wu](#)

TABLE 3 Ecological risk evaluation based on EI and RI values.

EI	Ecological risk level of an individual pollutant	RI	Potential ecological risk level
EI < 40	Clean	RI < 150	Slight ecological hazard
40 ≤ EI < 80	Slightly clean	150 ≤ RI < 300	Moderate ecological hazard
80 ≤ EI < 160	Strong	300 ≤ RI < 600	Strong ecological hazard
160 ≤ EI < 320	High Strong	RI ≥ 600	Extreme ecological hazard
EI ≥ 320	Extremely strong		

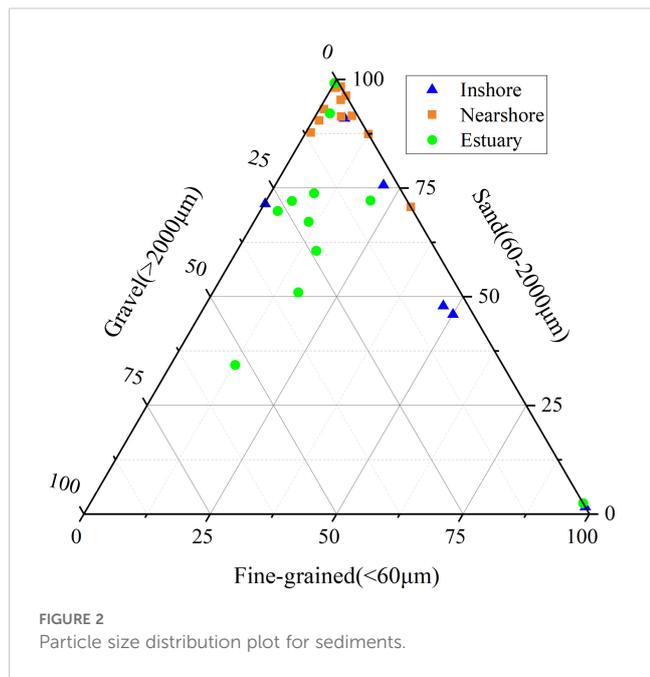


FIGURE 2 Particle size distribution plot for sediments.

et al., 2024, 2025). However, in estuarine and inshore areas, fine-grained sediments can reach maximum proportions of 98.39% and 97.52%, respectively. Notably, gravel is more prevalent in estuarine and inner bay regions, with a proportion up to 52.91%.

### 3.2 Spatial distribution of metallic elements in the sediments

This section aims to elucidate the spatial variation in metallic contamination across nearshore, inshore, and estuarine zones. Samples were classified according to field records and sampling locations for subsequent comparative analysis. In the Changhua

River estuary, the organic carbon content in the sediments ranged from 0.02% to 1.15%, with a mean value of 0.33%. The pH levels varied between 6.63 and 8.43, averaging 7.85, a value lower than those observed in the nearshore and inshore areas, indicating that the estuarine sediments are alkaline (Table 4). Metallic concentrations exhibited notable spatial heterogeneity across the research region (Figure 3). In nearshore sediments, average metallic concentrations remained below the corresponding BVs, which can be attributed to broad concentration ranges and prevailing hydrodynamic conditions. However, in inshore sediments, the metal elements including Cd, Hg, Pb, and Zn displayed average concentrations exceeding their BVs by approximately 417.95%, 187.33%, 67.41%, and 43.59%, respectively. Cr and Pb were identified as the most abundant elements in the inshore region, reaching peak concentrations up to 0.68 and 0.313 mg kg<sup>-1</sup>, respectively. Additionally, in the estuarine sediments, Ni, Cr, and Zn concentrations remained within acceptable limits, whereas Pb and Hg showed enrichment, registering 61.67% and 31.33% above their BVs, respectively. Across the entire research region, Cd and Pb emerged as the most accumulated elements, attaining peak concentrations up to 0.68 and 42.10 mg kg<sup>-1</sup>, respectively.

The coefficients of variation (CV) further reveal the spatial variability of metallic concentrations in the sediments. The CVs for Pb, As, and Cd ranged from 21.07% to 51.04%, indicating moderate variability. In contrast, the CVs for Ni, Cr, Hg, Zn, and Cu ranged from 53.18% to 92.91%, indicative of higher variability. Notably, exceptionally high concentrations of Cd and Pb were detected at locations B12, B16, B18, and B19, while higher Zn levels were recorded at location B12, suggesting pronounced point source pollution. These observations imply that anthropogenic activities have likely contributed to the enrichment of metallic elements in the sediments. Consistent with the findings of Pan et al. (2020) and Zhao et al. (2019), a higher CV indicates elevated human influence on soil metallic content, and thus it can act as a preliminary marker for anthropogenic impact.

TABLE 4 Statistics of metallic element parameters in the research region.

	Parameter	pH	Ni	Cr	Cd	Hg	As	Pb	Zn	Cu
Nearshore	Max	8.41	10.97	20	0.035	0.009	8.028	34.9	45.1	15.6
	Min	6.63	0.81	3.2	0.035	0.003	2.91	17	8.6	1
	Mean	8.03	4.07	7.05	0.035	0.006	4.684	23.61	19.67	5.13
Inshore	Max	8.43	24.62	70.2	0.68	0.313	10.3	40.2	163	27.4
	Min	8.13	7.04	14.9	0.12	0.015	2.84	26.1	33	6.4
	Mean	8.23	11.67	28.4	0.34	0.072	5.27	33.48	93.33	15.05
Estuary	Max	7.97	23.76	63	0.35	0.077	13	42.1	99.9	31.6
	Min	6.87	4.15	6.88	0.035	0.009	3.478	24	19.4	6.3
	Mean	7.5	9.67	19.76	0.097	0.0328	7.708	32.33	44.89	15.6
Region	BVs	-	24	61	0.065	0.025	7.7	20	65	15
Region	CV%	-	63.74	77.90	51.04	92.91	40.62	21.07	53.18	60.53

For values below the detection limit, half of the detection limit (LOD/2) is used as a substitute; Data description in mg.kg<sup>-1</sup>.

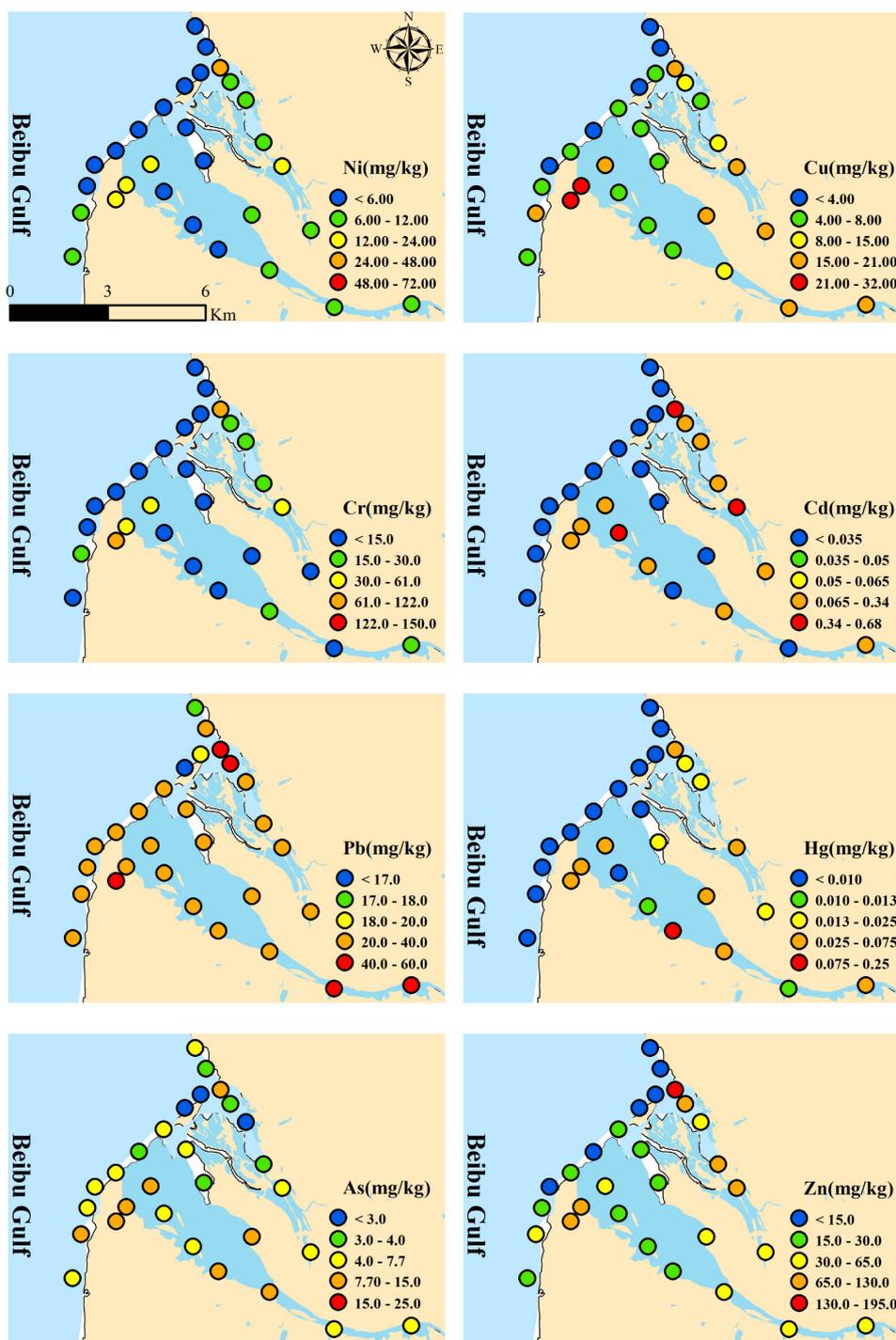


FIGURE 3 Spatial distribution of metallic elements in the sediments of the research region.

In summary, the pronounced variability in metallic concentrations across different regions may not reliably reflect true pollution levels due to inherent spatial heterogeneity and differences in BVs. Consequently, a comprehensive assessment of sediment contamination necessitates the integration of additional quantitative indices, such as  $I_{geo}$ , PI, EI, and RI, which explicitly account for these baseline variations.

### 3.3 Evaluation of ecological hazards and origins of metallic elements in the sediments

To assess metallic contamination in sediments more accurately, it is necessary to consider the BVs of sediments. Therefore, several quantitative indices, including the Geo-accumulation Index ( $I_{geo}$ ),

Pollution Index (PI), and Risk Index (RI), were employed to evaluate the metallic pollution levels in the sediments.

Figure 4 displays the  $I_{geo}$  values for metallic elements in the sedimentary samples collected from the research region. From the figure, while the inshore area has largely prevented widespread

pollution, certain sites in other regions exhibit severe contamination. The influence of anthropogenic activities surpasses that of geological weathering. The average  $I_{geo}$  values for As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the sediments across the entire region ranged from -1.36 to -0.68, -1.48 to 1.56, -3.93 to -1.94,

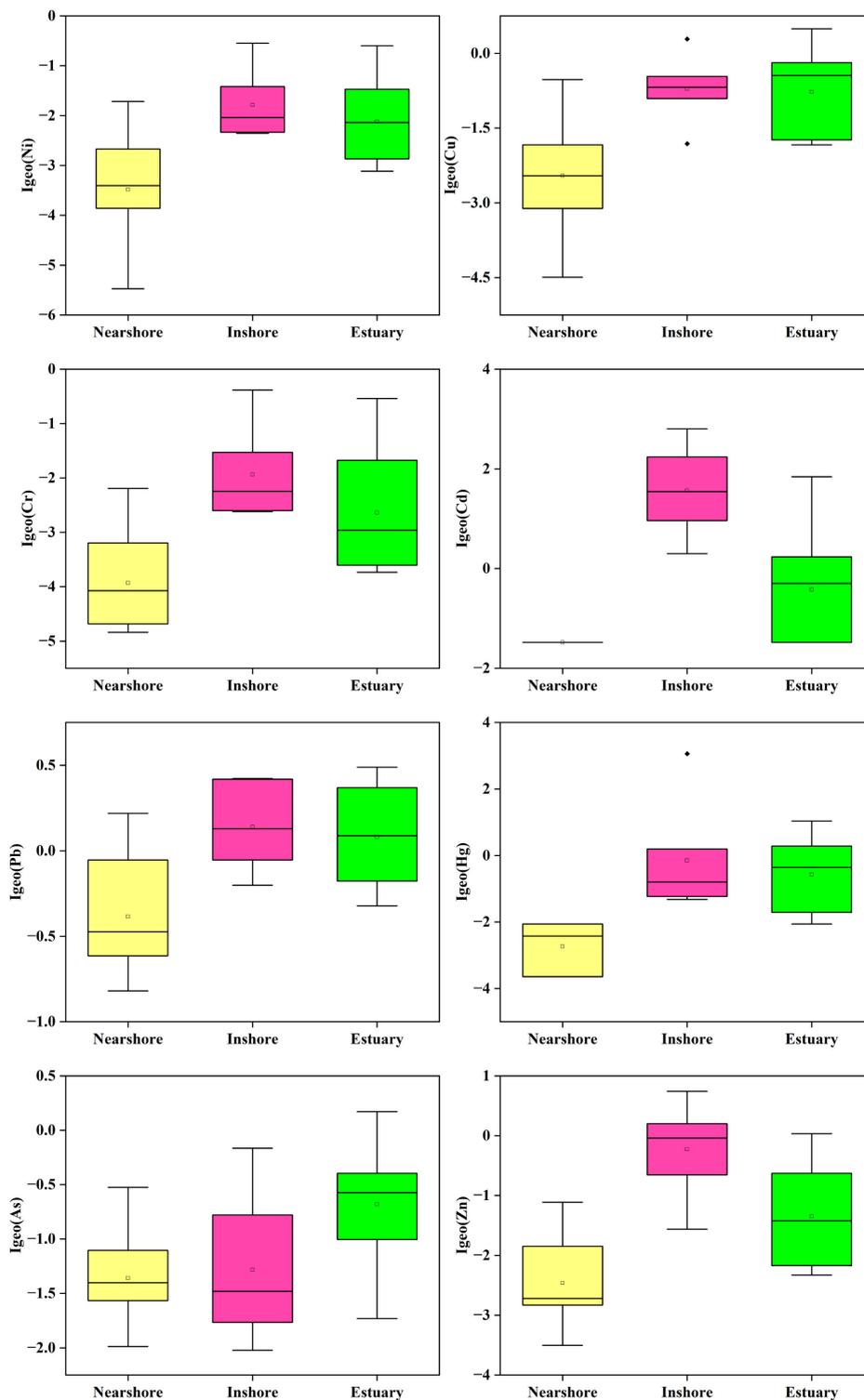


FIGURE 4  $I_{geo}$  index of metallic elements in the sediments across the research region.

-2.46 to -0.71, -2.74 to -0.15, -3.48 to -1.79, -0.38 to 0.14, and -2.46 to -0.23, respectively. The  $I_{geo}$  values for Cd and Pb in the inshore area reached maximum averages of 1.56 and 0.14, respectively, indicating moderate pollution for Cd and slight pollution for Pb. The  $I_{geo}$  results are comparable to those observed in other regions of Hainan Island, such as Doongzhaigang, Sibiwan, and Wenchang (Wang et al., 2024; Fu et al., 2024; Zhao et al., 2025). The average

$I_{geo}$  values for the other metallic elements remained below 0, suggesting no accumulation risk.

The PI values for the research region varied significantly, ranging from 0.37 to 1.69 for As, 0.54 to 10.46 for Cd, 0.05 to 1.15 for Cr, 0.07 to 2.51 for Cu, 0.12 to 12.52 for Hg, 0.03 to 1.03 for Ni, 0.85 to 2.11 for Pb, and 0.13 to 2.51 for Zn. Among these elements, Hg, Cd, Pb, Cu and Zn exhibited the greatest variability in

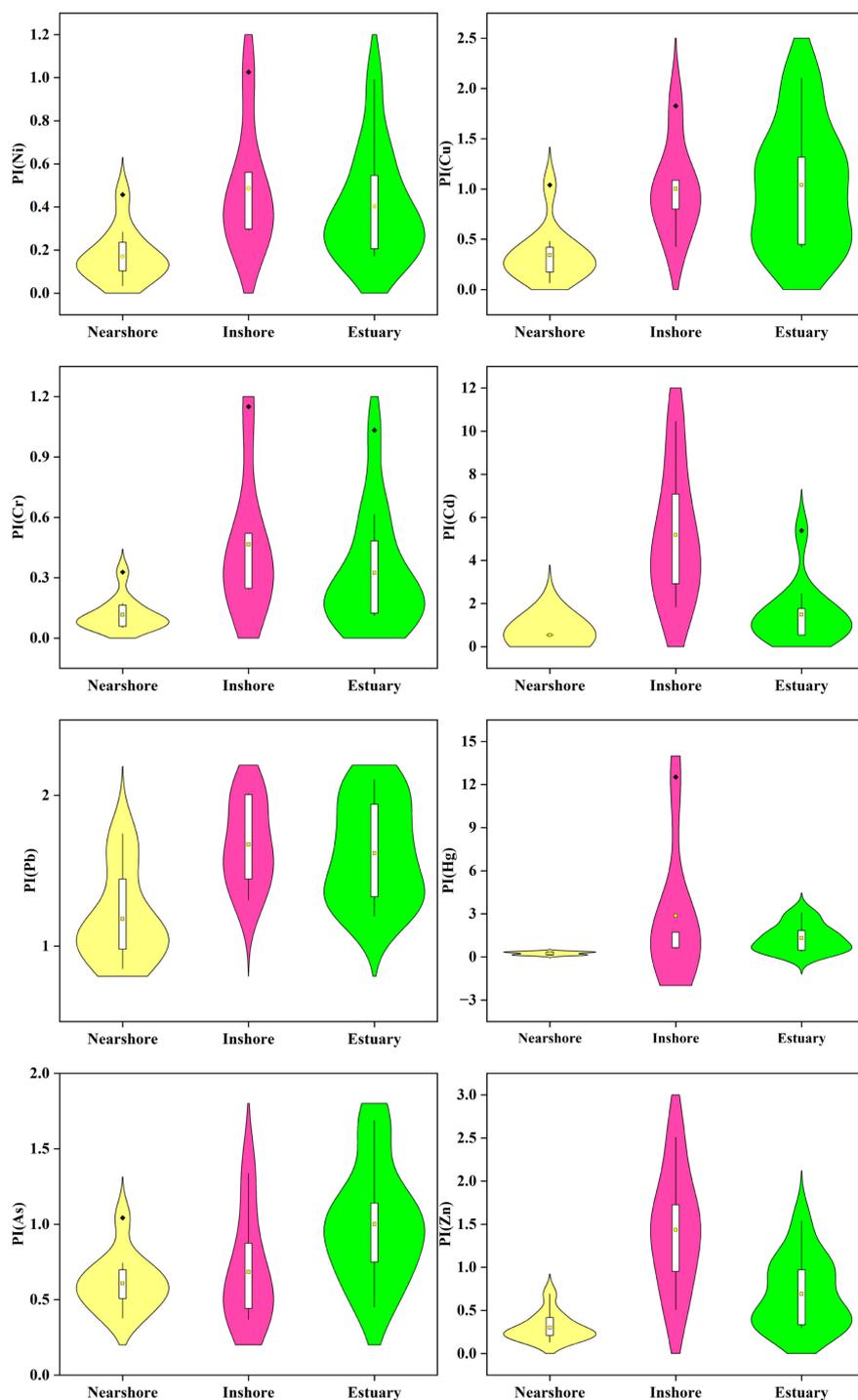


FIGURE 5 Violin and box plots of the PI index for metallic elements in the sediments.

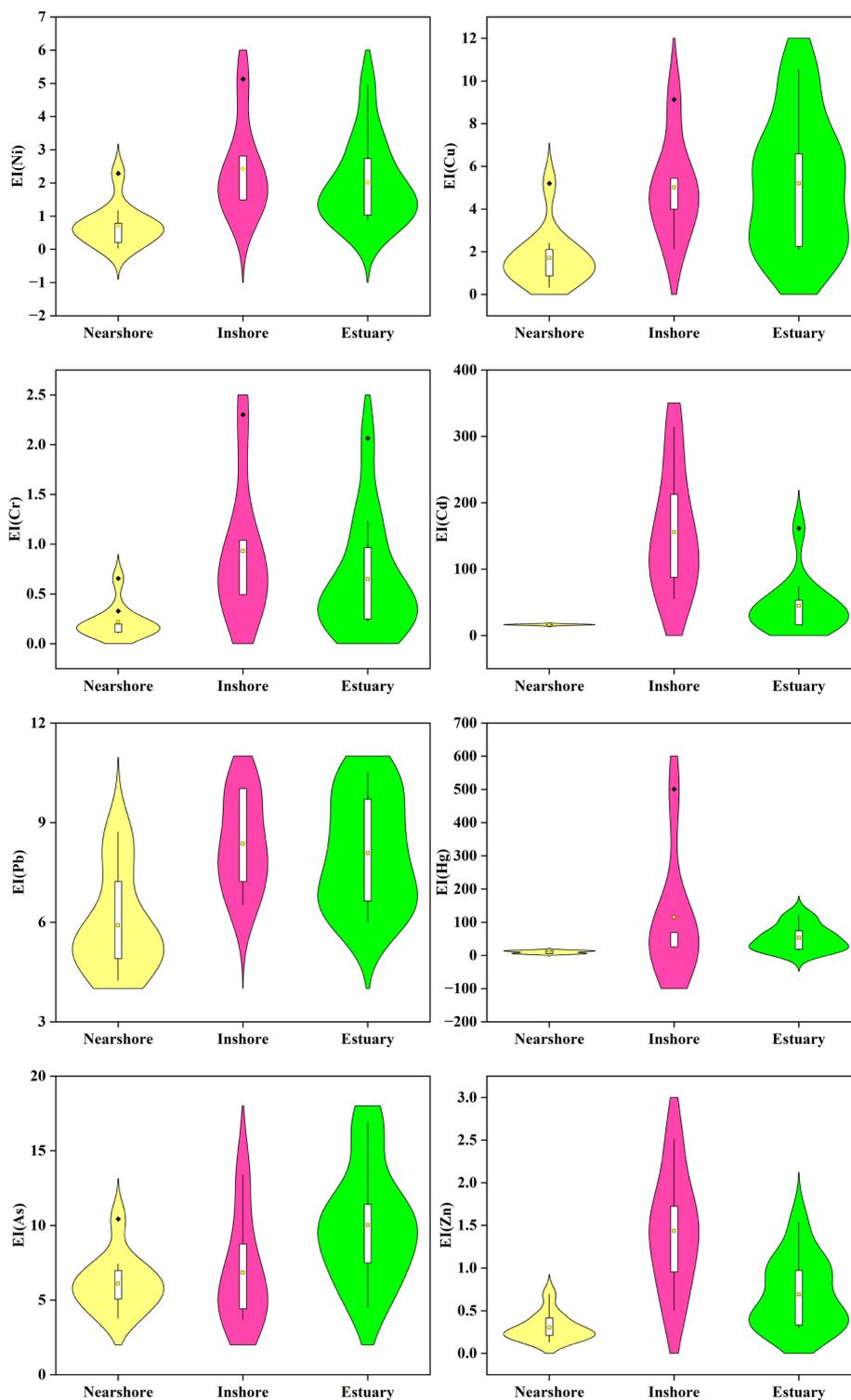


FIGURE 6 Violin and box plots of the EI index for metallic elements in the sediments.

PI values across the regions, and certain samples recorded a PI value greater than 2.0 (Figure 5). This observation indicates that the sediments in the research region are contaminated with these metallic elements.

Across the research region, the EI indices for metallic elements in the sediments were ranked as: Cd> Hg> As> Pb> Cu> Ni> Zn for the nearshore region, Cd> Hg> Pb> As> Cu> Ni> Zn for the inshore area, and Hg> Cd> As> Pb> Cu> Ni> Zn for the estuarine

area (Figure 6). In both the inshore and estuarine regions, Hg and Cd exhibited the greatest ecological threats, with mean EI values exceeding 160. Consistent with the findings from the Nandu River estuary, Cd and Hg exhibited the highest RI values. Furthermore, most sampling sites fell within the medium potential ecological hazard range (Ouyang et al., 2024).

The RI indices for metallic elements in the sediments ranged from 32.20 to 58.54 (mean of 41.13) in the nearshore region, from 103.36 to 665.22 (mean of 295.34) in the inshore region, and from 46.65 to 208.74 (mean of 123.99) in the estuarine region (Figure 7). Hg and Cd exhibited increased ecological risks in the inner and estuarine channel regions. From these RI values, it can be noted that a significant proportion of areas were anthropogenically affected. Specifically, 24.14% of the locations recorded RI values ranging from 150 to 300, reflecting moderate ecological risk, while 6.90% registered RI values exceeding 300, indicating high ecological risk. A notably high RI value of 665 was noted in the inner area, which represented a severe ecological risk.

### 3.4 Identification of the origins of metallic elements in the sediments

Principal Component Analysis (PCA) has proven effective in distinguishing the possible sources of metallic elements in sediments. Here, two principal components (PCs) accounted for a combined 70.6% of the overall variability in the sedimentary samples (Figure 8). The first principal component (PC1) represented 58.6% of this variability and exhibited strong positive correlations with Hg, Cd, Zn, Cr, and Ni. Additionally, PC 1 also showed a strong positive correlation with TOC, TN, and fine-

grained particles. The second principal component (PC2) accounted for 12.0% of the overall variability and displayed significant negative loadings for Pb, Cu and As. This pattern is likely due to weaker hydrodynamic conditions that favor the association of these elements with fine particles, localized pollution sources, and geochemical processes, such as the release of As in reducing environments and the enrichment of Pb and Cu in organic-rich sediments. As noted by Franco-Uría et al. (2009), the clustering of metallic elements often reflects shared sources, whether from human activities or natural processes. Herein, the strong loadings of Hg, Cd, Zn, Cr, and Ni on PC 1 imply a unified source for these elements. Research consistently highlights the substantial impact of human activities on the contamination levels of metallic elements in coastal sedimentary systems. For instance, Zhao et al. (2020b); Fu et al. (2024), and Yi et al. (2024) have attributed elevated concentrations of Pb, Cu, As, Cd, and Hg to anthropogenic contributions. The distribution of As, Cu, and Pb has been closely linked to the widespread application of phosphate-based fertilizers in both agriculture and mariculture (Li et al., 2010). Furthermore, agricultural practices have been identified as a significant contributor to Cd levels, particularly in farming regions (Wen et al., 2015). Agriculture in Changjiang County has developed rapidly, with an agricultural planting area of approximately 190,000 mu in 2022. Livestock farming includes 130,000 cattle, 112,500 pigs, and 2.1 million poultry. Aquaculture spans around 1,100 hectares, with pesticide usage of 120 tons and fertilizer use amounting to 47,500 tons. Hg, Cd, and Zn may also originate from ocean transportation, given the proximity of local fishing ports (Zhang et al., 2015). Commonly, the burning of fossil fuels, coal combustion, and industrial byproducts, whether in solid, liquid, or gaseous form, are identified as primary contributors to Hg

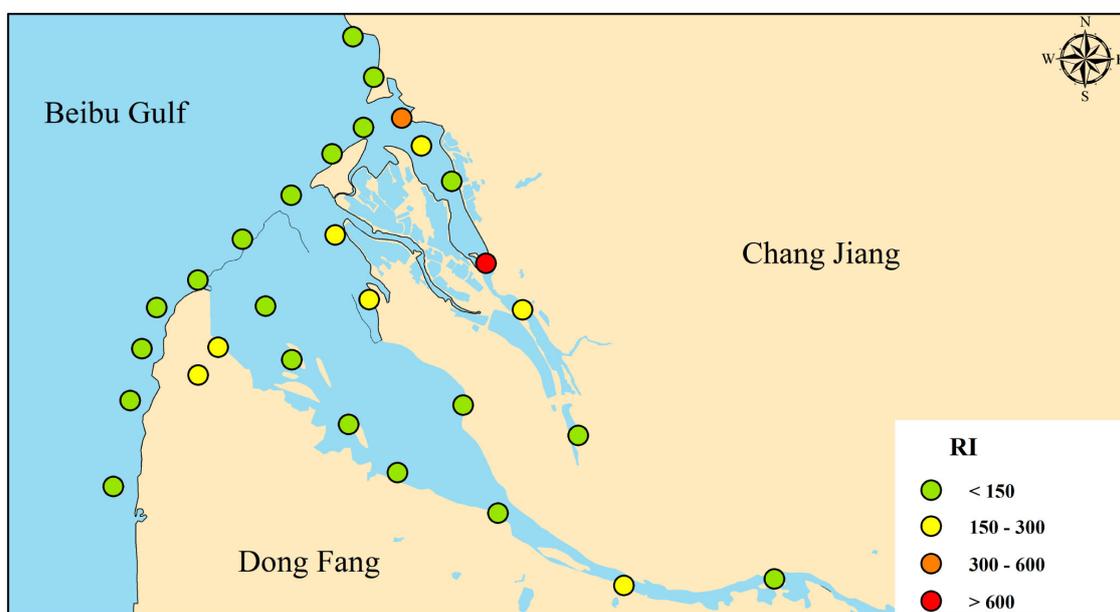


FIGURE 7  
Spatial distribution of the RI index for metallic elements in the sediments across the research region.



elements such as Cd, Hg, As, Pb, and Cu with TOC, TN, and fine-grained particles. This observation suggests that these elements are prone to adsorption onto organic matter and fine sediments, which enhances their accumulation in sedimentary environments. In contrast, negative correlations were observed between metallic elements and sand content, indicating that coarser sediments tend to dilute metallic concentrations due to their lower surface area and weaker adsorption capacity. These findings corroborate the results of the PCA analysis, thus highlighting the role of sediment texture and organic matter in controlling the distribution and enrichment of metallic elements in the research region.

PMF models provide a method for identifying and quantifying the sources of pollution in soils and other environmental media. This study utilized the EPA PMF 5.0 software to analyze the experimental data. It should be noted that the factor count in the model plays a crucial role in determining source allocation outcomes. To optimize the model, various scenarios with factor counts ranging from 2 to 5 were tested, with 20 iterations conducted to reduce Q values and manage scaling residuals. The model achieved its lowest Qrobust/Qture ratio when 4 factors were employed, establishing this as the optimal number for the analysis. Notably, most elements, excluding Cu and Cd, demonstrated  $r^2$  values exceeding 0.6, which underscored the reliability of the PMF model's source identification and affirmed that four factors adequately capture the data's underlying patterns. Figure 10 presents the source allocation of metallic elements in sediments derived from the PMF model, with the four factors contributing as follows: Factor 1 (17.6%), Factor 2 (14.6%), Factor 3 (63.4%), and Factor 4 (4.4%).

In Factor 1, Hg serves as the dominant contributing element, accounting for 84.9% of the factor, which is a high ecological risk as

indicated by the ecological risk assessments. Studies have shown that over 66% of Hg found in the natural ecosystems stems from anthropogenic sources, where coal burning and fossil fuels were identified as the principal source (Rashid et al., 2022). Our findings suggest that attention should be paid to Hg due to its potential ecological risks and high level of contamination. Other studies consistently reported the high ecological risks of Hg in some sediments (Zhang et al., 2024; Labenua et al., 2024; Liang et al., 2025). Indeed, coal burning and fossil fuels have been confirmed as the major contributor to Hg emissions in Hainan Province (Tian et al., 2012; Chen et al., 2024; Li et al., 2024; Fu K. et al., 2023). Based on this analysis, Factor 1 can be identified as originating from coal burning and fossil fuels.

In Factor 2, the primary contributing elements include Pb, Zn, and Cd, with contribution rates of 87.8%, 67.5%, and 46.3%, respectively. Studies indicate that Pb primarily originates from agricultural practices, livestock farming, and domestic wastewater discharge, while Zn and Cd are primarily associated with maritime traffic and port maintenance activities (Bai et al., 2011; Gu et al., 2014; Jiang and Guo, 2019; Korkanç et al., 2024). Therefore, Factor 2 can be identified as an anthropogenic mixed source associated with port activities.

In Factor 3, the dominant contributing elements include Cr, Ni, and Cu, with contribution rates of 76.9%, 76.2%, and 63.4%, respectively. The overall concentrations of Cr and Ni are relatively low, as most sampling points exhibit values below BVs. The CV for Cu is 60.53%, indicating high variability. Numerous studies have reported that  $CuSO_4$  is frequently used as an active ingredient in insecticide formulations (Fu K. et al., 2023; Weissengruber et al., 2018; Guan et al., 2018). Therefore, Cu can enter the soil through the

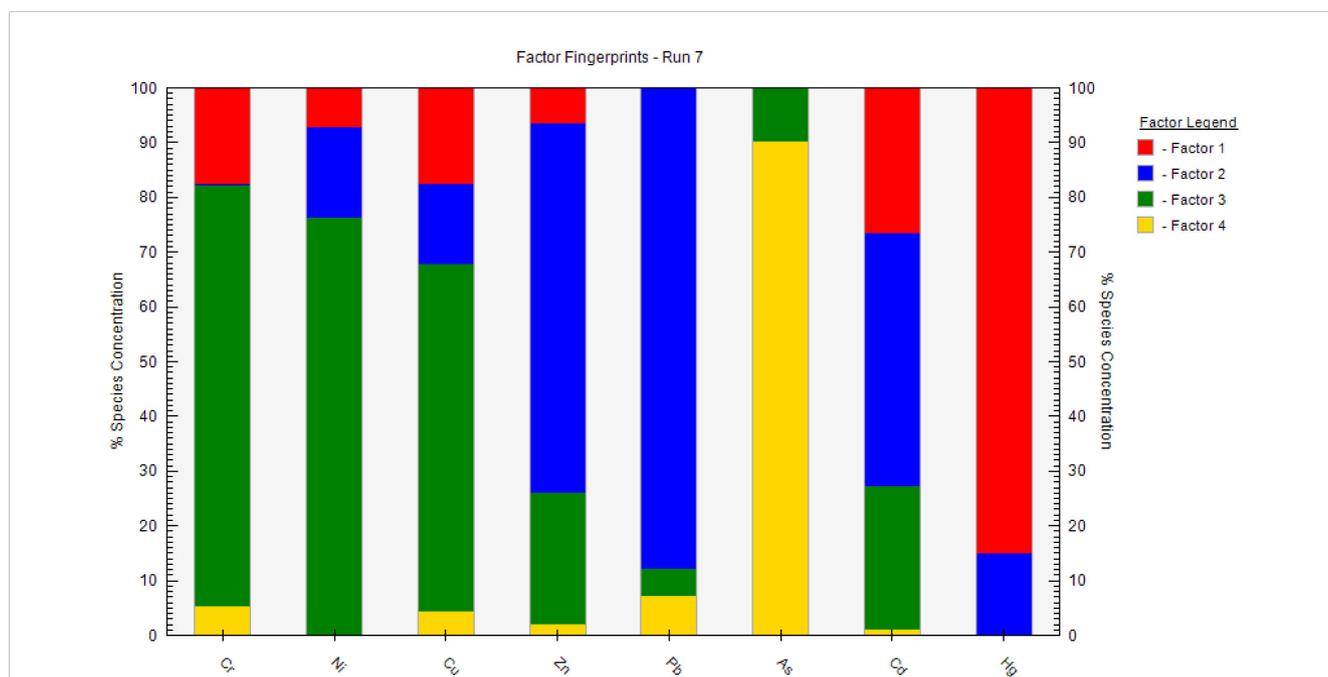


FIGURE 10 Source analysis of metallic elements in the sediments.

application of Cu-containing pesticides, such as the Bordeaux mixture (Cai et al., 2019b). This process leads to further accumulation in water bodies and sediments via precipitation. Our field investigation also revealed widespread application of Cu-based pesticides on crops such as mangoes, peanuts, and bananas in the research region. Based on these observations, it is hypothesized that Factor 3 is primarily influenced by natural and agricultural sources.

In Factor 4, As emerges as the primary loading element, contributing 90.4% to the factor. At some sampling locations, concentrations exceed the BV by 68.83%. The CV is 40.62, indicating moderate variability. As is commonly used as a feed additive and enters rivers through aquaculture wastewater, resulting in accumulation in sediments. Additionally, Calcium Arsenate and Sodium Arsenate find use in herbicides and pesticides (Huang et al., 2022). In agricultural practices, the prolonged and widespread application of synthetic fertilizers, manure, herbicides, and insecticides often leads to As contamination. Overall, Factor 4 is primarily attributed to agricultural sources.

### 3.5 Limitations

This study has certain limitations and uncertainties in assessing the pollution risk of heavy metals in the sediments of the Changhua River estuary. First, the limited number of sampling sites may not fully capture the spatial distribution characteristics of heavy metal contamination across the entire estuarine region. Second, the sampling was conducted in July, during the rainy season in Hainan Island, while no samples were collected during the dry season. As a result, the study does not account for the effects of seasonal and climatic variations on heavy metal pollution. Future research in this region should consider increasing the number of sampling sites and investigating variations in the types and concentrations of heavy metals in sediments under both dry and rainy season conditions. Additionally, when applying the PMF model for source apportionment, the limited sample size may introduce uncertainties, making the results less conclusive. To achieve a more accurate identification of heavy metal sources in sediments, further investigations using advanced techniques, such as isotopic tracing, are strongly recommended.

## 4 Conclusions

The present study conducted a thorough investigation into the spatial distributions and potential ecological risks of metallic contamination in the sediments along the entire coastal region of the Changhua River estuary on Hainan Island, including nearshore, inshore, and estuarine areas. It particularly focused on the metallic pollution across riverine, estuarine, and marine ecosystems. Findings revealed that concentrations of As, Cd, Cr, Cu, and Pb elements were markedly elevated in inshore zones compared to nearshore and estuarine areas. Based on  $I_{geo}$  and PI indices, sediments in inshore regions showed significant accumulations of Pb and Cd. With average RI values exceeding 150, inshore sediments were classified as

moderately contaminated by the analyzed metallic elements. Geogenic processes were identified as the primary sources of Cr and Ni, while Zn, As, Cd, Cu, Hg, and Pb were largely linked to human-induced discharges, as supported by PCA, Spearman's correlation, and PMF analyses. The alignment between PCA and PMF outcomes further strengthens the credibility of these conclusions. Overall, the risk associated with metallic elements was higher in the inshore and estuarine areas, while other regions exhibited lower risk levels. This pattern was primarily influenced by the presence of fine-grained materials and human activities. Additionally, hydrodynamic conditions contributed to a reduction in ecological risks within nearshore areas. To effectively mitigate the environmental hazards associated with metallic elements, it is essential to reduce emissions from agricultural, domestic, and transportation sources. Consequently, subsequent management strategies should incorporate targeted actions to reduce emissions of harmful substances, safeguarding and promoting the long-term health of marine ecosystems.

## Data availability statement

The original contributions presented in the study are included in the article/supplementary material. Further inquiries can be directed to the corresponding authors.

## Author contributions

JM: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. KF: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. ZC: Investigation, Supervision, Writing – review & editing. DW: Data curation, Methodology, Supervision, Writing – review & editing, Funding acquisition, Resources, Writing – original draft. DB: Investigation, Software, Writing – review & editing, Supervision. JG: Conceptualization, Investigation, Writing – review & editing, Supervision. SZ: Conceptualization, Investigation, Writing – review & editing, Data curation. ZX: Conceptualization, Writing – review & editing. DJ: Writing – review & editing. YG: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

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## Conflict of interest

Author YG was employed by Guiyang Baiyun District Municipal Development and Construction Co., Ltd.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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