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Spatial distribution of trace elements in coastal sediments of Ha Tinh province, Vietnam

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ABSTRACT

Sediment compositions and trace element (Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg) concentrations were analyzed to assess the sediment quality and the factors affecting the trace elements' concentration in Ha Tinh province's coastal sediments. Results revealed that the concentration of trace elements in the coastal sediments was generally lower than in other coastal areas in Vietnam. The significantly positive correlation of the fine sediment composition with Cu, Pb, Zn, Cr, and Fe indicated that the silt and clay sediments could absorb these trace elements. The highly positive correlation among Cu, Pb, Zn, Cr, and Fe suggested that the trace elements likely originated from similar sources. The sediment quality assessment showed that trace element concentrations were lower than the limited values in the sediment quality guideline (QCVN 43:2017/BTNMT). According to the Igeo index, the sediments were not polluted by Cu, Pb, Zn, Cr, Fe, and As but were likely polluted by Hg and Mn in some sampling sites. Therefore, it is necessary to invest the future studies to assess the Hg and Mn concentration levels in these sites to protect the marine environment.

Keywords: Trace element, sediment, marine environment, Ha Tinh; Vietnam.

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INTRODUCTION

Sediments in the coastal area act as sinks for various organic and inorganic pollutants transported by the river systems, falling out from the atmosphere and released from human activities along the coast. Industry, urbanization, tourism, and maritime development have rapidly released pollutants into the marine environment, including trace elements (TEs) [1]. The TEs such as Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg in marine sediments can be originated from natural or anthropogenic sources such as the lithology of the area and industrial and maritime activities. The TEs can be released from the marine sediments into the seawater through the disturbance processes caused by waves, tidal currents, and benthic activities. The high TE concentrations in marine sediments will cause negative impacts on the benthos and marine ecosystems [2]. TEs such as Hg, As, Pb, Cr, Mn, Cu, and Zn tend to accumulate and impact negatively marine organisms' growth [3]. The TEs will be transferred from the sedimentary environment into the organisms in the food chain. The bioaccumulation of the TEs can be the highest in the species at the top of the food chain and can be harmful to human health through consuming seafood [4]. The transferred processes of the TEs from the marine sediments into the benthos and fish have been observed in different coastal regions in the world, for example, in farmed scallops in coastal areas of China [5], bivalves (*Meretrix meretrix*) in Netravathi Estuary, India [3]. Therefore, assessing the concentration of TEs in coastal sediments is crucial for studying theese elements' toxicity, bioconcentration, and biomagnification in marine environments. In the present study, the concentration of eight trace elements (Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg) and sediment compositions were analyzed to assess the sediment quality and the natural and anthropogenic factors affecting the concentration of the TEs in coastal sediments of Ha Tinh province.

MATERIALS AND METHODS

Study area

The coastal area of Ha Tinh province is examined according to the Decree No. 40/2014/ND-CP of the Vietnam Government to implement the articles of the Law on Natural Resources and Environment of Sea and Islands [6, 7]. According to that, the coastal area of Ha Tinh province has internal and outer boundaries. The lowest tidal water lines examines in 18.6 years, and the outer boundary is extended from the lowest tidal water line to a distance of six nautical miles. The shoreline of the study area has a total of 137 km in length, with the main direction of northwest-southeast. The shoreline in the north from the Nghi Xuan district to the Cam Xuyen district has some main estuaries, consisting of Hoi Estuary, Sot Estuary, and Nhuong Estuary. Curves and steep slopes characterize the southern shoreline and have headlands such as Than Lan Cape and Ron Cape (Figure 1). The depth of the study area ranged from 0–40 m, being shallower in the northern parts and deeper in the southern part. The northeast and southwest monsoons, characteristics by the diurnal tidal regime, influenced by the oceanographic characteristics. The study area has coral reefs on Son Duong island and a high diversity of economically valuable species of bivalves, gastropods, crustaceans, and fishes. In this area, the marine economy is rapidly developing with different sectors in industry, maritime, tourism, and urbanization. Typically, Vung Ang economic zone has been investing in developing metallurgical industries, steel industries, thermal power, petrochemical refining, and seaports. The industrial activities of metallurgy, thermal power, and seaports in the coastal area are developing at a rapid rate and can cause impacts on the marine environment. For example, the study area seriously suffered impacts from the marine environmental incident in the year 2016. This severe incident caused mass fish deaths along the coastal area of four central provinces (Ha Tinh, Quang Binh, Quang Tri, and Thua Thien Hue) [8]. Therefore, the results from this study will provide baseline data for evaluating the impacts of anthropogenic activities on the marine environment.

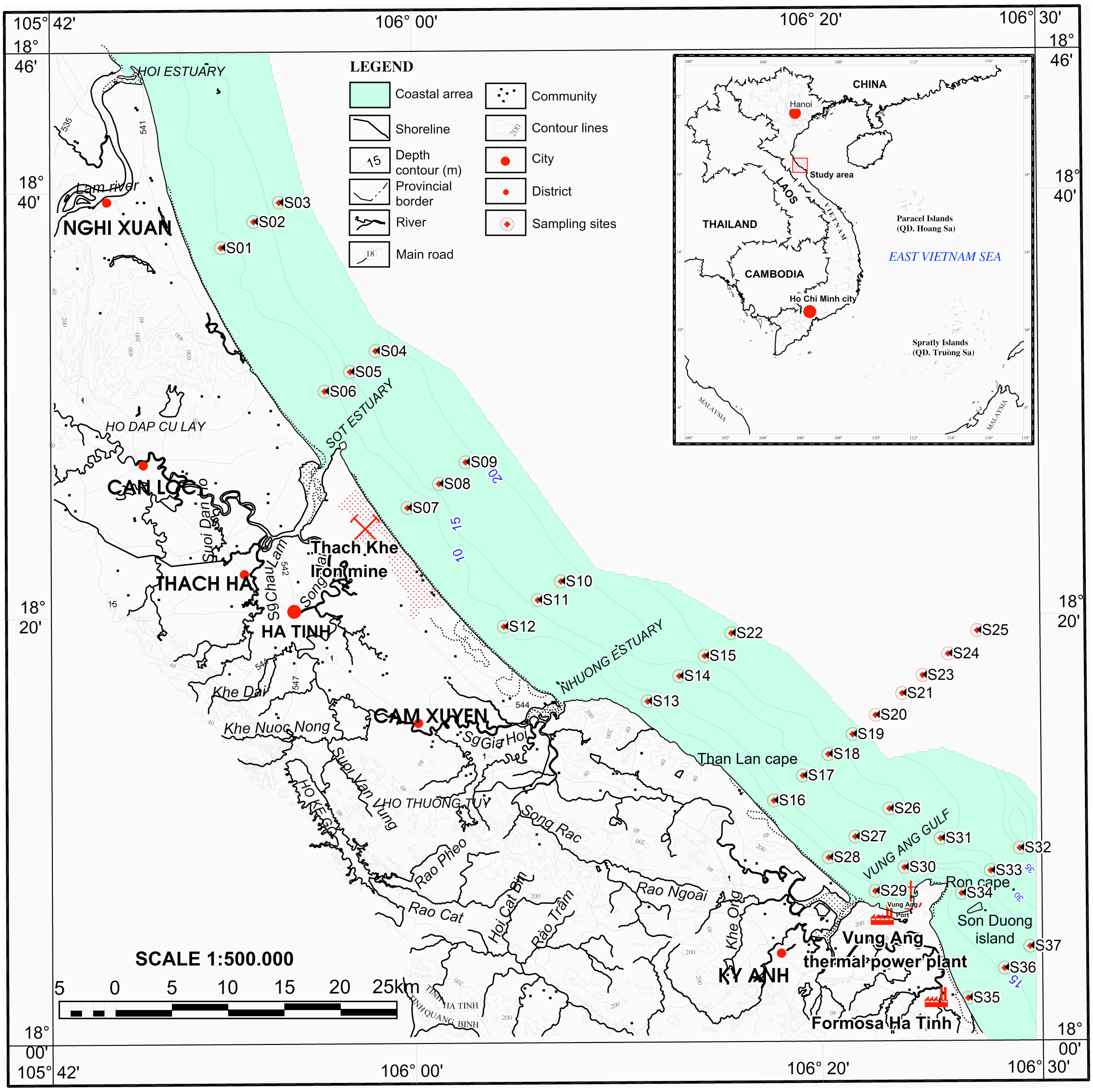

Figure 1. The study area and sampling sites in coastal area of Ha Tinh province

Sample collection and analysis

Thirty-seven surface sediment samples were collected using a Peterson grab sampler in November 2021. The sampling sites were spatially distributed in the coastal area of Ha Tinh province (Figure 1). The surface sediments at S20, S21, S23, S24, and S25 sampling sites were taken in the nearshore area with a maximum distance from the shoreline of 24.5 km and a depth of 45 m [9]. For studying the variation of TEs with sediment layers, four sediment cores were collected at the sampling sites S02, S17, S28 and S37 using a gravity corer. The sediment cores were sliced at 0–5 cm, 20–25 cm, and 40–45 cm. As a result, the total sediment samples were 49. Sediment samples were immediately put into the PE bags, an icebox at < 4 °C, and transported to the laboratory for processing [10].

Five grams of fresh sediments were put into a beaker for sediment composition analysis. The sediment sample was pretreated with an H_2O_2 solution of 10% and acid HCl 1 N for 24 h to remove organic matter and carbonates altogether. Then, 10 mL of distilled water was added and dispersed using an ultrasonic cleaner for 3 min. Sediment grain sizes were analyzed using an automatic laser diffraction particle size analyzer LA-950V2 (HORIBA Co.), with a measurement range of 0.01–3,000 μm. The sediment composition was calculated based on sand, silt, and clay percentage. The mud content was calculated using silt and clay [11].

For trace element analysis, sediment samples were dried entirely at 60 ^oC in an electric oven and pulverized using an agate mortar and pestle. The visible organic matter particles (roots and small branches) and shell carbonate fragments were manually removed from sediment samples using stainless steel forceps. A 0.1 mg fine powder sediment sample was treated in a microwave Teflon vessel with an acid mixture (1:5 HF: HNO_3). The mixture was heated in a microwave system (Multiwave PRO, Anton Paar, Australia). The HF acid residue was evaporated, then the samples were diluted with ultrapure MILLI-Q water to 50 mL for further analysis. The concentrations of eight trace elements (Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg) were analyzed with an Atomic Absorption Spectrometer (Agilent 240FS). The methods' accuracy and precision were assessed the certified marine sediment reference material PACS-2 (National Research Council Canada). The analytical precision for replicate samples was within ±10% [10].

Statistical analysis

The mean and standard deviation of TEs and sediment compositions were calculated. The concentration of TEs below the limit of detection (LOD) was calculated by dividing the LOD by the square root of 2 [12]. The spatial distribution of sediment contents and TEs was presented using Mapinfo software (Pitney Bowes Software Inc.). Pearson's correlation was used to examine the correlation among the TEs with sediment parameters (sand, silt, and clay). A significant level was accepted for all statistical tests at $p \leq 0.05$. Statistical tests were performed using SPSS 20.0 for Windows (SPSS Inc. 2011).

RESULTS AND DISCUSSION

Sedimentary characteristics in coastal area of Ha Tinh province

The marine sediments are mainly composed of sand, silt, and clay. An earlier report examined the study area's main sediment types, consisting of sand and mud sand (Figure 2). In the present study, the sand fraction ranged from 42.2–95.0%, with a mean of 70.51 ± 14.45%. The silt fraction varied in a small range from 3.0–35.8%, with an average of 16.39 ± 8.23%. The clay fraction ranged from 1.8–29.2%, with a mean of 13.1 ± 6.59%. The mud content was calculated by the sum of the silt and clay, ranging from 5.0–57.8%, with a mean of 29.49 ± 14.29% (Table 1). The sediment compositions in the present study were similar to a report on establishing the sediment grain size compositions in the coastal area of Ha Tinh province [13]. These authors showed that the sediments in the studied area were mainly composed of sand, which evenly distributed from the north to the south (Figure 2). The muddy sand is mainly distributed nearby the estuaries and at a depth of 10–20 m. The mud content was unevenly distributed on the seabed and formed 5–25% contours, primarily in muddy sand sediments (Figure 2). Results showed that mud contents tended to be high in the sampling sites nearby the Hoi estuary, Sot estuary, Nhuong estuary, Khau estuary, and south of Son Duong island. These areas could be directly received from the suspended matter transported from rivers and (or) have a weak hydrodynamic condition, suitable conditions to accumulate the fine grain size sediments. The silt and clay fractions tended to increase slightly with the depth of the sediment layers (Table 2) and gradually decrease to the coastline (Figure 2).

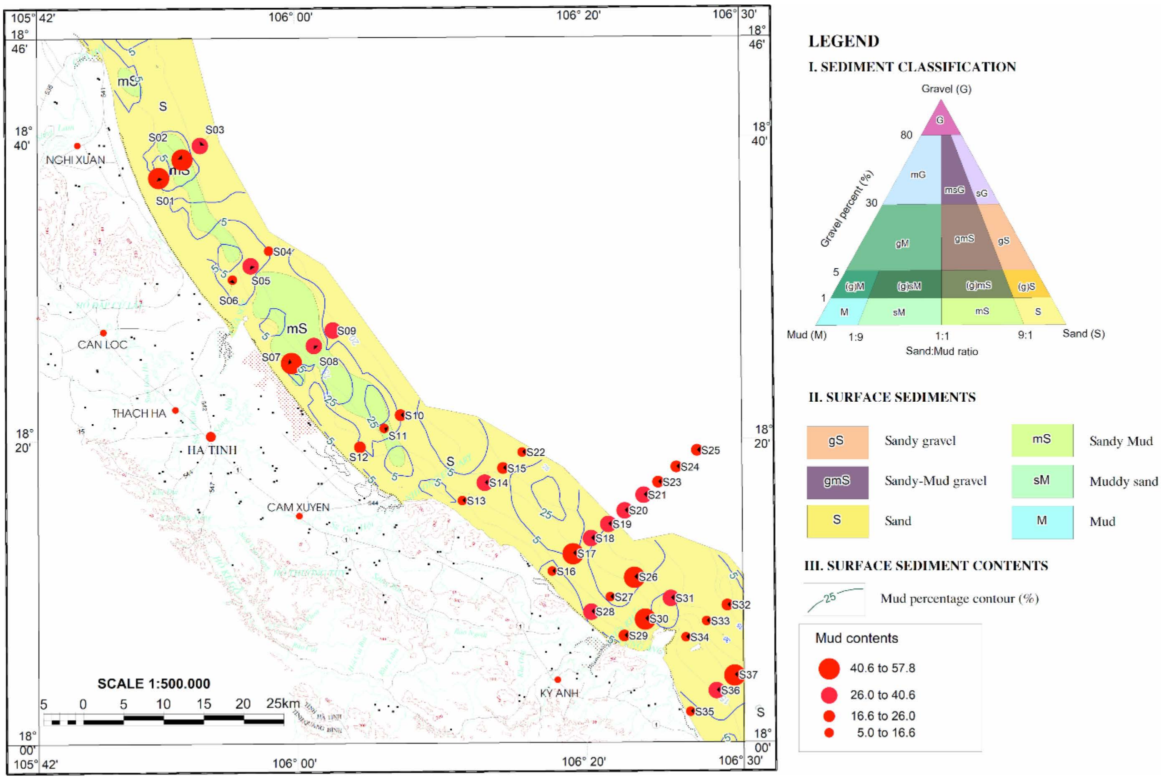

Figure 2. Distribution of surface sediments in coastal area of Ha Tinh province

Note: The spatial distribution of surface sediments is taken from Thanh [13]

Table 1. Statistical data of trace element concentrations and sediment composition in coastal area of Ha Tinh province

Parameter	Unit	Min.	Max.	Mean	SD	N	QCVN 43:2017/BTNMT
Cu	mg/kg	4.24	24.30	10.51	6.30	37	108
Pb		4.95	25.18	13.32	5.47	37	112
Zn		7.10	86.10	40.99	21.29	37	271
Cr		5.66	35.97	14.25	9.75	37	160
Fe		7,658	36,660	24,359.24	6,901.92	37	-
Mn		3.54	2,697.30	575.16	413.20	37	-
As		1.06	11.08	2.88	1.69	37	41.6
Hg		0.00	0.12	0.06	0.02	37	0.7
Sand	%	42.20	95	70.51	14.45	37	-
Silt		3.00	35.80	16.39	8.23	37	-
Clay		1.80	29.20	13.10	6.59	37	-
Mud		5.00	57.80	29.49	14.30	37	-

Spatial distribution of trace elements in coastal sediments of Ha Tinh province

Cu concentration ranged from 4.24–24.3 mg/kg, with a mean of 10.51 ± 6.3 mg/kg. Cu concentration tended to be higher in the sampling sites (S01, S02, S07, S13, S14, and S30) that were rich in the muddy sand sediments in the north of Sot estuary, reaching the highest value in the Vung Ang Gulf. Cu concentration decreased from the coastal area to the nearshore area (Figure 3a) and with the depth of sediment cores (Table 2). The average Cu concentration in the coastal sediments of Ha Tinh province was 10.51 ± 6.30 mg/kg, similar to that in Ha Long bay (14.53 ± 6.30 mg/kg) [14], Tien Yen gulf [15], sedimentary basin of the Red river, Central Vietnamese shelf and slope and Nam Kon Son basin area [16]. Cu concentration was 3–5 times lower than in the coastal sediments of Thanh Hoa province [17] and Cam estuary [18] (Table 3). Pb concentration in the coastal sediments of Ha Tinh province varied in an extensive range from 4.95–25.18 mg/kg, with a mean of 10.51 ± 6.3 mg/kg (Table 1). The spatial distribution of Pb concentration was higher in the muddy sand in the northern part and reaching to the maximum in the Vung Ang gulf. Pb concentration gradually decreased from coastal to nearshore areas (Figure 3b). Pb concentration markedly decreased in the deeper sediment layers (Table 2). Compared with other coastal regions in Vietnam, Pb concentration had a similar range with Ha Long bay [14], Tien Yen gulf [15], the sedimentary basin of the Red river, the Central Vietnamese shelf and slope, and Nam Kon Son basin area [16], but was 6–9 times lower than in the coastal sediments in Thanh Hoa province [17] and Cam estuary [18] (Table 3). Zn concentration ranged from 7.1–86.1 mg/kg, averaging 40.99 ± 21.29 mg/kg (Table 1). Zn concentration was the highest in the Vung Ang gulf. Zn concentration markedly decreased with the depth of sediment layers and from the coastal to the nearshore area (Table 2, Figure 3c). The mean concentration of Zn was approximately that in Ha Long bay [14], Tien Yen gulf [15], the sedimentary basin of the Red river, the Central Vietnamese shelf and slope, and the Nam Kon Son basin area [16], but was 3–5 times lower than coastal sediments in Thanh Hoa province [17] and Cam estuary [18]. The similar concentration of Cu, Pb, and Zn in sediments from the present study with those in sediments from other coastal areas referred that the human activities in coastal areas did not highly impact the marine sediments in Ha Tinh province. The range of Cr concentration was 5.66–35.97 mg/kg, with a mean of 14.25 ± 9.75 mg/kg. Cr concentration tended to be higher in the muddy sand in the northern area and did not show a clear reduction trend from the coastal area to the nearshore area (Fig. 3d). Cr concentration tended to increase with the depth of the sediment layers (Table 3). Fe concentration varied from 7,658–36,660 mg/kg, with a mean of 24,359.2 ± 6,901.9 mg/kg (Table 1). Fe concentration was higher in the muddy sand in the northern area and the Vung Ang gulf. Fe concentration gradually decreased from the coastal area to the nearshore area (Fig. 3e). Compared with other coastal areas in Vietnam, the mean concentration of Fe was similar to the sedimentary basin of the Red river [16] but was lower than that in other coastal areas (Table 3). Mn concentration ranged from 3.5–2,697.3 mg/kg, with a mean of 575.2 ± 413.2 mg/kg. The considerable variation in Mn concentration was due to some anomalies in the sampling sites (S16, S23, and S35) in the Southern and Northern Vung Ang gulf (Figure 3f). Mn concentration tended to decrease with the depth of sediment cores (Table 2). Mn concentration was two times higher than that in Tien Yen gulf [15] but lower than that in Ha Long bay [14], the sedimentary basin of the Red river, Central Vietnamese shelf and slope, and Nam Kon Son basin area [16], and the coastal area of Thanh Hoa province [17] and Cam estuary [18] (Table 3). Results showed that this study's higher Mn concentration in the coastal sediments could relate to steel production because manganese phosphate is commonly used to treat rust and prevent corrosion on steel. The As concentration varied from 1.06–11.08 mg/kg, with an average of 2.88 ± 1.69 mg/kg (Table 1), gradually increasing from the coastal area to the nearshore area (Figure 3g). The As concentration in the present study was 2–14 times lower than that in other coastal regions (Table 3). Hg concentration varied from below the limit of detection to 0.12 mg/kg (Table 1). Hg concentration was higher in the muddy sand and nearby the Vung Ang gulf. Hg concentration did not tend to change significantly from the coastal area to the nearshore area (Figure 3g).

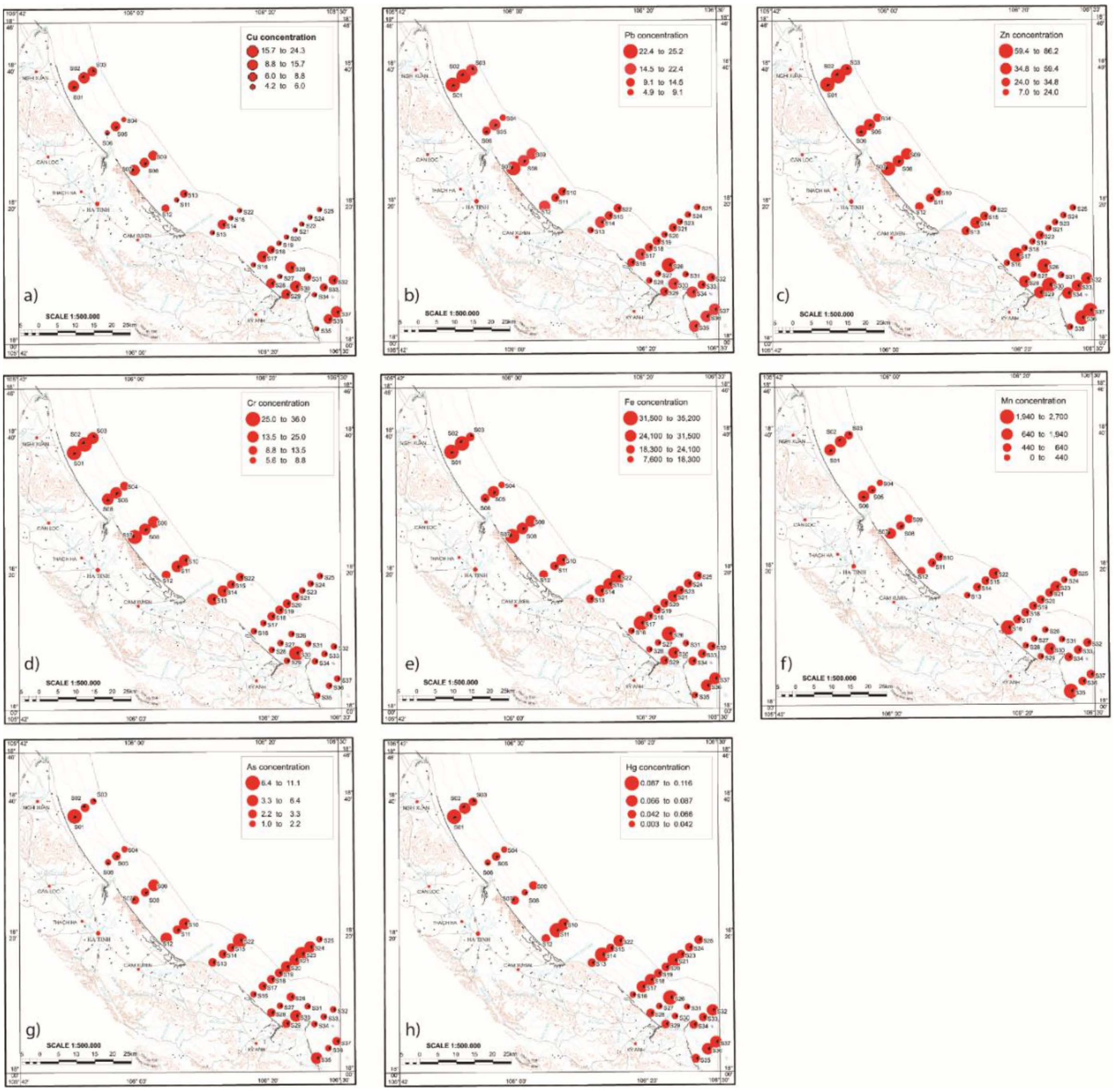

Figure 3. Spatial distribution of trace element concentrations in coastal sediments of Ha Tinh province

Table 2. Depth variation of trace element concentration (mg/kg) and sediment compositions (%) in sediment cores

Depth	Cu		Pb		Zn		Cr		Fe		Mn	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
0–5	16.90	5.10	17.24	6.66	64.00	17.20	13.23	15.15	27341.34	7258.68	517.95	108.44
20–25	13.21	5.03	13.17	3.83	48.25	11.76	24.66	6.96	29269.35	4491.49	467.27	62.69
40–45	13.32	5.95	12.09	3.92	45.55	13.92	23.70	7.96	28620.44	7370.82	385.45	88.68
Depth	As		Hg		Sand		Silt		Clay		Mud	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
0–5	2.81	0.32	0.07	0.02	60.05	9.79	21.60	5.63	18.35	4.25	39.95	9.79
20–25	1.97	0.41	0.05	0.01	59.26	10.27	23.02	6.44	17.73	4.02	40.75	10.27
40–45	2.21	0.22	0.05	0.01	59.96	8.76	24.10	5.90	15.95	2.86	40.05	8.76

Table 3. Comparison of trace element concentration (mg/kg) in coastal sediments of Ha Tinh province with other studies

No.	Locations	Year	Cu	Pb	Zn	Cr	Fe	Mn	As	Hg	Reference
1	Ha Tinh, Vietnam	2021	10.51 ± 6.30	13.32 ± 5.47	41.00 ± 21.29	14.25 ± 9.75	24,359.24 ± 901.92	575.30 ± 413.20	2.88 ± 1.69	0,06 ± 0,02	This study
2	Ha Long bay, Vietnam	2020	14.53 ± 8.13	30.39 ± 14.85	50.87 ± 22.89	-	-	-	6.06 ± 3.02	-	[14]
3	Tien Yen, Vietnam	2012	24.27	25	17	28.6	-	215.4	17.1	-	[15]
4	Sedimentary basin of the Red river	2021	17.25 ± 3.58	19.17 ± 3.47	72.17 ± 8.92	61.06 ± 13.77	25,000 ± 5,800	922 ± 200	6.63 ± 2.37	-	[16]
5	Central Vietnamese shelf and slope		25.91 ± 8.69	24.15 ± 3.34	95.25 ± 19.68	75.94 ± 15.44	36,300 ± 4,700	1,700 ±1,800	7.83 ± 2.33	-	
6	Nam Kon Son basin area		21.39 ± 6.71	21.41 ±4.13	80.09 ± 22.81	72.59 ± 21.96	33,400 ± 7,300	1,100 ± 1,800	9.43 ± 3.17	-	
7	Thanh Hoa, Vietnam	2021	56.51	66.76	142.37	52.29	49,777.65	1,167.34	24.7	-	[17]
8	Cua Cam, Vietnam	2013	82	92	178	90	36,200	827	42	-	[18]

Inter-trace element relationship

The correlation coefficient matrix among TEs and sediment composition (sand, silt, clays) is presented in Table 4. Trace elements (Cu, Pb, Zn, Cr, Fe) were positively and highly correlated, suggesting that the trace elements likely originated from similar sources [19]. As shown in Figure 3, the concentration of these elements all has the same spatial distribution patterns, being higher in the sampling sites near the shoreline and estuaries and tending to decrease in the nearshore area gradually, implying that these TEs could be originated from anthropogenic activities in the coastal zone and the transportation of pollutants from rivers [17]. Cu, Pb, Zn, Cr, and Fe concentrations were negatively correlated with sand grain sizes (Table 4). Results were similar to that found in the coastal sediments of Thanh Hoa province [17] and Ha Long bay [14].

Table 4. Correlation matrices of trace elements and sediment parameters in sediments in coastal area of Ha Tinh province

	Cu	Pb	Zn	Cr	Fe	Mn	As	Hg	Sand	Silt	Clay
Cu	1										
Pb	0.757**	1									
Zn	0.843**	0.795**	1								
Cr	0.425**	0.454**	0.398**	1							
Fe	0.700**	0.666**	0.622**	0.566**	1						
Mn	-0.045	0.176	-0.036	0.189	0.090	1					
As	0.052	0.143	-0.118	0.162	0.333*	0.399**	1				
Hg	0.185	0.216	0.179	-0.085	0.321*	0.021	0.245	1			
Sand	-0.810**	-0.582**	-0.635**	-0.401**	-0.707**	0.136	-0.096	-0.155	1		
Silt	0.782**	0.514**	0.595**	0.468**	0.759**	-0.165	0.152	0.175	-0.921**	1	
Clay	0.688**	0.383**	0.490**	0.347*	0.703**	-0.208	0.122	0.204	-0.874**	0.910**	1

Notes: *: Correlation is significant at the 0.05 level (2-tailed); **: Correlation is significant at the 0.01 level (2-tailed).

In contrast, the concentration of TEs was positively correlated with the fine-grained sediment (silt and clay) (Table 4). This result showed that marine sediments with high fine-grained composition would have the ability to accumulate high concentrations of TEs. Therefore, the coastal areas with lower hydrodynamic regimes, which favor the deposition of fine-grained sediments and/or receipt of a pollutant source, will have a significant risk of environmental pollution [20]. The characteristics of marine sediment grain sizes are well-known to influence the concentration and distribution of TEs. Fine-grained sediments such as clay and silt have a large surface area and high adsorption capacity of TEs [21]. This feature could explain the lower concentration of TEs in the study area compared to other coastal regions in Vietnam (Table 3). The reason for this pattern is that the mud contents in the coastal area of Ha Tinh province ranged from 5.0–57.8%, being lower than that of 17.3–99.5% in the coastal area of Thanh Hoa province [17], and 17.3–99.5% in Ha Long bay [14].

Sediment quality assessment

The marine sedimentary environment is essential for many benthic species, such as bivalves, snails, crabs, and benthic fishes. In the present study, the concentration of TEs (Cu, Pb, Zn, Cr, As, Hg) was much lower than the allowable limit in QCVN 43:2017/BTNMT [23] (Table 1), so the marine sediment environment was safe for benthic organisms. Pollutants in the sedimentary environment can accumulate into organisms through the three main mechanisms of food digestion, the uptake rate through respiration, and the penetration into the organism’s skin [3, 22]. Therefore, the sediment quality assessment will provide scientific information to protect marine ecosystems, mitigate the transfer of pollutants along the food chain and protect human health.

The geoaccumulation index (*Igeo*) was calculated for TEs elements to evaluate the increase in the concentration of TEs in the coastal sediments of Ha Tinh province compared with the background values. The *Igeo* index was proposed by [24] and calculated by the following equation:

$$Igeo = \log_2 \frac{C_n}{1.5C_n}$$

C_n is the analyzed concentration of TEs in the sediments; C_n is the geochemical background concentration of trace element; and the coefficient of 1.5 is the background matrix correction factor due to the lithogenic effect. In the present study, the C_n values of the TEs were adopted from Zhao, Jiang [25]: 15 mg/kg for Cu, 20 mg/kg for Pb, 65 mg/kg for Zn, 60 mg/kg for Cr; 31,000 mg/kg for Fe, 530 mg/kg for Mn, 7.7 mg/kg for As, and 0.025 mg/kg for Hg.

According to Muller [24], the *Igeo* values of the TEs (Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg) were divided into three classes, consisting of class 0 (*Igeo* < 0: unpolluted), class 1 (0 < *Igeo* < 1: unpolluted to moderately polluted), class 2 (1 < *Igeo* < 2: moderately polluted) (Figure 4). The *Igeo* values of Cu, Pb, Zn, Cr, Fe, and As were less than zero, suggesting that these elements did not pollute the sediments. The *Igeo* of Mn ranged from -7.8 to 1.76, in which most sediment samples were unpolluted. Three sediment samples in the sampling sites (S07, S22, S23) had the *Igeo* of class 1, classifying them as unpolluted to moderately polluted. Only the sediment samples in the sampling sites (S16, S35) had the *Igeo* of class 2, classifying as unpolluted to moderately polluted. Among TEs, *Igeo* values of Hg was relatively high, ranging from -3.41 to 1.62. Therefore, the marine sediments in the present study were classified from unpolluted, unpolluted to moderately polluted, and moderately polluted. The moderately polluted level of Hg were observed in the sampling sites (S01, S10, S11, S14, S17, S21, S26, S37). In general, the *Igeo* values of TEs Cu, Pb, Zn, Cr, and As in the present study were similar to the early report in the sedimentary basin of the Red river, Central Vietnamese shelf and slope and Nam Kon Son basin area [16]. The elevated level of Hg in marine sediments suggested that future studies should focus on examining the bioconcentration of Hg in benthos and the migrating of Hg in the food chain.

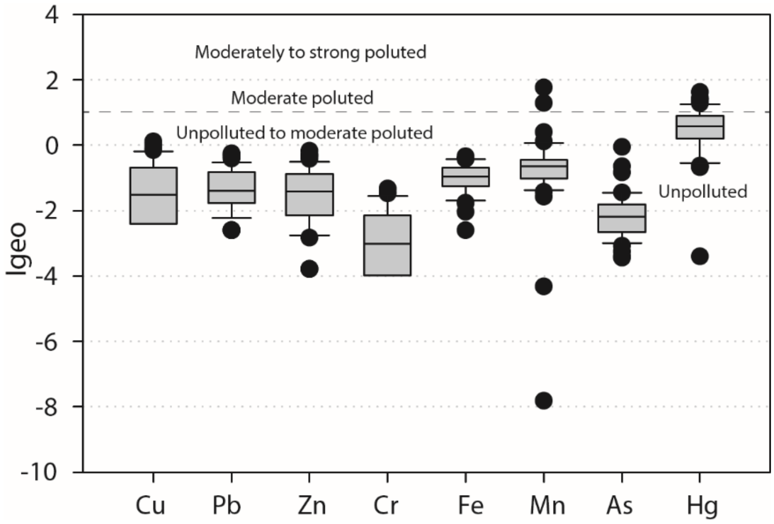

Figure 4. Geoaccumulation index of trace metals in sediments of the study area

CONCLUSIONS

Sediments in the coastal area of Ha Tinh province had a large sand composition, which had a lower ability to accumulate TEs than in other coastal areas in Vietnam. In general, the concentration of Cu, Pb, Zn, Cr, Fe, Mn, As, and Hg in the sediments of the study area was lower than those of other coastal areas in Vietnam. The concentration of TEs tended to be higher in the muddy sand, which is distributed near the estuaries and Vung Ang gulf, except for As and Hg, Cu, Pb, Zn, Cr, Fe, and Mn concentration gradually decreased the coastal area to the nearshore area.

The concentration levels of TEs in the coastal sediments of Ha Tinh province were lower than the allowable limit in QCVN 43:2017/BTNMT, so the marine sediment environment was safe for aquatic organisms.

Based on the *Igeo* index, the sediments were not polluted by Cu, Pb, Zn, Cr, Fe, and As but were likely polluted by Hg and Mn in some sampling sites. Therefore, it is necessary to invest the future studies to assess the Hg and Mn concentration levels in these sites to protect the marine environment.

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