

## Characteristics of heavy metals in surface sediments of the Van Don-Tra Coast, northeast Vietnam

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### ABSTRACT

Eighteen surface sediment samples were collected from the Van Don-Tra Coast, northwestern Gulf of Tonkin, for analyzing the concentration of heavy metals (Zn, Cr, As, Cu, Ni, Pb, Co, and Cd) and grain size. The statistical and contamination indices of heavy metals were calculated to evaluate the extent of pollution caused due to heavy metals and their potential harm to the environment. According to the analysis results, the contamination of heavy metals in the study area were higher than the background concentrations, except for Pb. The heavy metal concentrations in the study area followed the descending order of Zn > Cr > As > Cu > Ni > Pb > Co > Cd. The concentrations of heavy metals do not correlate with clay content, and almost heavy metals were non-correlated with silt and sand contents, except for Cr, implying that the grain size does not control heavy metals concentration in surface sediments in the region. The results of pollution indices imply that the contamination in the environment was due to excess accumulation of Cd, in contrast to that of Zn. The ecological pollution risk shows an increased risk of heavy metal pollution. According to the statistical index, the contamination of heavy metals in surface sediments in the study area could come from the same sources under the same environmental conditions and is related to the source of sedimentary material from the mainland near the study area.

### 1. Introduction

In recent decades, pollution of the marine environment by heavy metals has become a global issue. In general, the increase in pollutants results from the lack of defined maritime environmental legislation and the rise in pollution from human activities such as industry, agriculture, and urbanization (Ardila et al., 2023). Along with economic development, protecting the marine environment is one of the most critical tasks of coastal territories. Anthropogenic pollution and the observed high

magnetic remanence may be related to the enrichment of heavy metals in the uppermost sedimentary layer of the seafloor (Chan et al., 2001). The heavy metal pollutants in the aquatic environment significantly affect marine species and the food chain. In particular, heavy metals in surface sediments can affect human health due to the consumption of marine organisms, especially benthos (Gao et al., 2016; Romano et al., 2021). A substantial portion of the heavy metal influx accumulates in the estuary zone and continental shelf over time. Consequently, coastal areas are crucial for assessing the condition of heavy metal

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contamination (Harikrishnan et al., 2017). Sediments have been shown to harbor several pollutants of various origins, including organic substances. It is also a significant deposition site for hazardous materials. This deposition may originate from the adjacent parent rock or industrial emissions. In recent years, anthropogenic outputs have contributed significantly to the aquatic ecology. After a certain amount of absorption, pollutants might become a source of contamination in the surrounding environment (Gkaragkouni et al., 2021). Thus, the heavy metal pollution problem has attracted different researchers from all over the world (Al-Mutairi et al., 2021; Ardila et al., 2023; Gargouri et al., 2011; Leung et al., 2021; S et al., 2013; Tanjung et al., 2019; Wang et al., 2022). Gargouri et al. (2011) showed that on Sfax coast of Tunisia, the main sources of heavy metals in surface sediments were due to industrial and municipal discharges.

The release of contaminants from these polluted places is determined by the element's bioavailability and natural environmental circumstances. Even in high quantities, a poisonous metal may not be fatal if its bioavailability is poor, and vice versa (Yushin et al., 2023). Future research may find the concentration of trace elements in coastal sediment valuable for establishing baselines and evaluating sediment quality (Harikrishnan et al., 2017). Due to their chemical properties and behaviors, metals engage in numerous biogeochemical cycles. The distribution of heavy metals varies with grain size and tends to concentrate in the sediments' fine portion (Gkaragkouni et al., 2021)

Additionally, higher concentration of heavy metals was observed in the surface sediments in coastal areas of Galle Harbor, Sri Lanka and in the nearby locations of city canal outlet (S et al., 2013). Results of Wang et al. (2022) revealed that Cu, Hg, and Pb could be originated from anthropogenic activities, whereas As might be derived from natural sources. (Leung et al., 2021) indicated that Cd was the element that significantly affected the soil/sediment quality in the study area of Coles Bay Area, Svalbard, and Greater Bay Area, China. Recently, Ardila et al. (2023) investigated the heavy metal pollution in marine sediments on Mallorca Island, Spain, and reported that the concentration of heavy metals depends significantly on the distance to the outfall. Based on previous studies, it can be seen that the main sources of heavy metal pollution in surface sediments are due to anthropogenic activities (i.e., industrial and municipal activities) and natural sources. In addition, the concentrations of heavy metals in surface sediments tend to increase as the distance to the coastline decreases.

In Vietnam, the Gulf of Tonkin, known as "Vinh Bac Bo", is a relatively shallow area, with an average depth of 75 m and a distance covering of about 126.250 km<sup>2</sup>. The Gulf of Tonkin plays an important role in the economic development of some coastal provinces of Vietnam like Quang Ninh and Hai Phong. In this Gulf, there are lots of fishing ground areas that provide a large amount of seafood for North Vietnam. Along the coastal areas of this Gulf, industrial activities have been developed significantly with the establishment of lot of factories, industrial zones, and ports. Besides this, the Red River, one of the largest rivers in the world, also discharges to the Gulf of Tonkin. Thus, this region has faced significant problems associated with the possibility of heavy metal contamination in surface sediments. There are some works to assess the heavy metal contamination in the Gulf of Tonkin (Dang Hoai et al., 2022; Hieu Ho et al., 2010; Nguyen et al., 2012). Van Don-Tra Co is currently an area with a very fast construction pace. A series of construction projects have been carried out such as the construction of Van Don international airport, Ha Long - Van Don - Mong Cai Highway, Ao Tien international seaport and Van Don City. Parallel to the rapid pace of infrastructure development, activities of exploiting construction materials, leveling land, minerals, etc are developing rapidly. This has significantly affected the marine environment in the Van Don - Tra Co area. However, there is no assessment of heavy metal contamination in surface sediments of the Van Don-Tra Co coastal area, northwestern Gulf of Tonkin. The aims of the present investigation are as follows: i) To determine the concentration of heavy metals in surface sediments along the Van Don-Tra Co coast; and ii) To calculate the ecological pollution

risk parameters Igeo, CF, PLI, Er, and PER in relation to the heavy metal concentrations in surface sediments. The present investigation is the sole one that has been assessed in the specific area of interest, as far as we are aware. By evaluating the surface sediments, the current manuscript has assessed the attributes of heavy metal contamination and prevalence in surface sediments of the Van Don-Tra Co coast.

## 2. Materials and methods

### 2.1. Study area and sampling collection

The study area is located along the coastal area of Van Don-Tra Co, Quang Ninh province, northwestern Gulf of Tonkin (Fig. 1). Industrial activities onshore, fishing, port ship and tourist activities and residents living along the Van Don-Tra Co coastal area are the primary sources of environmental pollution in this sea area (Huong et al., 2022). The sea depth is relatively shallow, varying from a few meters to 20 m of water. The small island system distributes widely parallel to the coastline. The river system includes Ba Che, Tien Yen, Dan Ha, Ha Coi and Ka Long rivers. The most significant are Tien Yen and Ka Long rivers with basin areas of 1070 and 773 km<sup>2</sup>, respectively. In the current study, the surface sediments were collected at 18 stations, as shown in Fig. 1. The sample locations are arranged in two lines parallel to the coastline, one between the shoreline and island range and the other outside the island range. The most distant sampling station is about 20 km from the coast, and the closest is about 3 km. Sediment samples were taken by the gravity corer, with a mass of 100 kg and a sampling tube with diameter of 80 mm. A single-beam echo sounder measured the surface depth of the sampling station before releasing the gravity corer. Sediment samples are taken in the field to ensure their original shape, and the sediment sample columns are always placed vertically. The bottom of the sample tube and the two sides are perforated with small holes for water drainage. The estimated time for seawater to escape from the tube is about 180 minutes. The time to take samples from the field to the laboratory for processing is 36 hours. The samples were cut for 3 cm of surface sediment which reflect with the material during recent sources. The surface sediments were then dried. The sediments in the study area predominantly contain terrigenous materials, biological carbonates, and silicon.

### 2.2. Sediment grain size analysis

The grain size was measured by the LA-960 instrument. This device can measure the particle size ranging from 10 nm to 5 mm in both suspensions and dry powders. The samples were treated to remove impurities and organic matter before taking measurements. The organic matter was removed by soaking each 1 g of dry sample into 20 ml of Hydrogen Peroxide 6% (H<sub>2</sub>O<sub>2</sub>) at room temperature until there were no bubbles (for about 24 hours). The sample was dried again and weighed to determine the current weight and then soaked into 5 ml HCl (10%) for 4 hours (Ogoyi et al., 2011) The samples were washed with distilled water thrice to remove the HCl. The LA-960 device was then measured for processing the samples. The indices of, mean, median, mode, D (15, 50, 90), and percent composition of each grain level were determined by LA960 software.

### 2.3. Heavy metals analysis

The heavy metal (Cr, Co, Ni, Cu, Zn, As, Cd, and Pb) concentrations in surface sediment samples were analyzed using inductively coupled plasma mass spectrometry (ICAP Q ICP-MS). The ICP-Multi element standard solution of Merck™ with certified code number 1094980001 was used as the calibration standard sample of ICP-MS. To ensure the accuracy of the analysis results, parallel analyses were repeated three times for all the data, and the average values were taken. The analysis accuracies of the repeated samples were within ±5%. Recovery

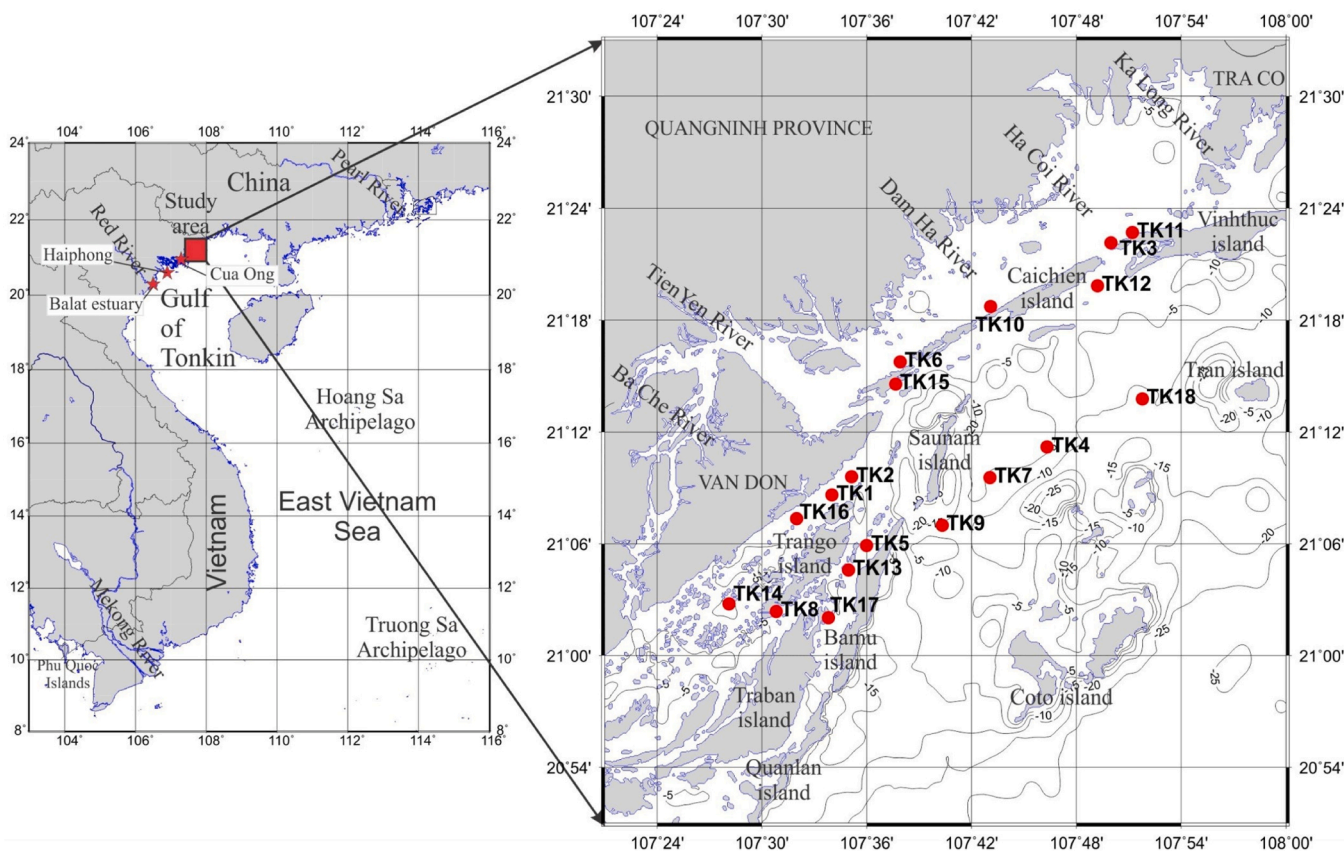


Fig. 1. The study area and sample stations.

efficiency greater than 90% was determined using standard reference materials.

2.4. Sediment quality assessment

To evaluate the heavy metal contamination of the surface sediment, the following indices were used:

2.4.1. \*Geo-accumulation index

To assess the pollution of heavy metals in sediments, Muller (1969) proposed the geo-accumulation index, which was estimated based on the following formula:

$$I_{geo} = \log_2 \frac{C_i}{1.5 \times B_i}$$

Where  $C_i$  is the measured concentration of heavy metal  $i$  in the sediment sample;  $B_i$  is geochemical baseline concentration of heavy metal  $i$ . The value of  $B_i = 57.4, 10, 20, 28.6, 103, 5.21, 0.30, 27.6$  mg/kg for Cr, Co, Ni, Cu, Zn, As, Cd, Pb, respectively (Zhang and Du, 2005). It may reflect the most suitable background values for study area. Based on  $I_{geo}$  values, the pollution levels are divided into 7 categories, including: unpolluted level ( $<0$ ); unpolluted to moderate level (0–1); moderate level (1–2); moderate to strong level (2–3); strong level (3–4); strong to extremely strong level (4–5); extreme level ( $>5$ ).

2.4.2. \*Pollution load index (PLI)

The pollution load index (PLI) provides a simple, comparative means for assessing the level of heavy metal pollution. PLI is used to evaluate the overall metal pollution level and pollution status in sediments. It is calculated based on the contamination factor (CF) with the following formula (Angulo, 1996; Tomlinson et al., 1980).

$$PLI = (CF_1 \times CF_2 \times CF_3 \times CF_4 \times \dots \times CF_n)^{1/n}$$

$$CF = C_i/B_i$$

where  $C_i$  is the measured concentration of heavy metal in the sediment sample;  $B_i$  is the baseline concentration of heavy metal  $i$ . The pollution level is also evaluated based on the CF value, accordingly if  $CF < 1$ : low pollution;  $1 < CF < 3$ : moderate pollution;  $3 < CF < 6$ : Serious pollution; and  $CF > 6$ : high pollution.  $PLI > 1$  indicates that sediment is polluted by heavy metals;  $PLI < 1$  shows that there is no sediment pollution (Benhadhya and M Hadjel, 2013; Varol, 2011)

2.4.3. \*Potential ecological risk index (PERI)

The potential Ecological Risk Index Method (PERI) was proposed by Swedish scientist Hakanson in 1980 and has been applied to evaluate the harm of heavy metals in sediments. The method was used widely and had significant influence internationally. (PER) is used to assess the potential harm of heavy metals to the environment. This index is calculated as follows:

$$PERI = \sum E_r^i$$

$$E_r^i = T_r^i \times \frac{C_i}{B_i}$$

where  $C_i$ : Measured concentration of heavy metal  $i$  in the sediment sample

$B_i$ : Geochemical concentration of heavy metal  $i$

$T_r^i$ : Toxic-response factor for heavy metal  $i$  which is as follows: Cr = 2, Co = Ni = Cu = Pb = 5, Zn = 1, As = 10, and Cd = 30.

$E_r^i$ : Potential ecological risk index for heavy metal  $i$

PER: Comprehensive potential ecological risk index. It is clarified into 5 levels as follows: low risk ( $E_r^i < 40$ ;  $PER < 150$ ); moderate risk ( $40 \leq E_r^i < 80$ ;  $150 \leq PER < 300$ ); considerable risk ( $80 \leq E_r^i < 160$ ;  $300 \leq PER < 600$ ); very high risk ( $160 \leq E_r^i < 320$ ;  $600 \leq PER < 1200$ ); and extremely high risk ( $E_r^i \geq 320$ ;  $PER \geq 1200$ ).

PER <600); high risk ( $160 \leq E_r^I < 320$ ); and very high risk ( $E_r^I \geq 320$ ; PER  $\geq 600$ ) (Benhaddya and M Hadjel (2013))

### 2.5. Statistical analysis

The results of heavy metal concentration were analyzed for standard deviation (SD), Kurtosis, and Skewness. Heavy metal concentration statistical analysis was performed using SPSS software version 24.

## 3. Results and discussions

### 3.1. Characteristics of heavy metal concentrations

The concentration of heavy metals in the studied sediments is summarized in Table 1. The minimum, maximum, and average values of heavy metal contamination in surface sediments are presented in Fig. 4. It can be seen that the highest concentration of heavy metals is found for Zn, with an average concentration of 165.3 mg/kg, while the lowest one is observed for Cd, with an average concentration of 3.9 mg/kg. Heavy metal concentrations in surface sediments are generally followed by the descending order of Zn, Cr, As, Cu, Ni, Pb, Co, and Cd. The average concentrations of heavy metals in the surface sediments of the study area are higher than their corresponding background values, indicating that the heavy metals pollute the surface sediments to different degrees.

The spatial distribution of heavy metals in surface sediments in the study area is shown in Fig. 6. We find that the concentration of heavy metals varies with location. Outside the Tien Yen estuary (stations KT6 and KT15), the highest concentration of heavy metals was more evaluated in KT6 and KT15 than in the surrounding area, except for Cd, implying that the Tien Yen may supply most Cd to this studied area. This is due to the presence of a high contamination factor value suggests that the cadmium heavy metals in this region are the result of human activities along the river's headwaters and along the coast. However, the lowest concentration of Cr, Co, Ni, Cu, Zn, and Pb in spatial distribution occurs in KT10 (Outside the Dam Ha estuary) than in the surrounding area, suggesting that most heavy metals may not be transported by this river. This implies that heavy metals in the study sediments are related to the source of sedimentary material from the mainland near the study area through estuary areas.

**Table 1**  
Content of heavy metals of surface sediment samples in the study area.

Points	Content of heavy metals mg/kg							
	Zn	As	Cd	Pb	Cr	Co	Ni	Cu
TK1	132.2	93.4	4.99	16.5	227.0	25.20	20.52	83.7
TK2	166.8	123.4	4.89	31.7	117.6	22.24	38.79	51.6
TK3	116.3	96.1	4.44	23.0	132.8	20.77	25.19	44.2
TK4	237.3	107.9	4.83	36.8	94.7	20.90	47.80	61.0
TK5	169.5	112.3	4.02	17.0	135.5	21.82	28.22	60.6
TK6	292.8	148.4	1.47	19.2	175.3	25.77	42.69	87.3
TK7	108.5	77.7	4.28	16.5	157.9	22.62	16.88	60.9
TK8	159.0	91.3	3.96	26.9	113.9	23.23	24.30	53.7
TK9	121.7	70.0	4.43	27.3	59.15	13.76	21.10	36.1
TK10	134.9	80.1	4.35	20.5	90.6	18.73	25.01	40.9
TK11	131.1	92.8	4.58	22.8	108.9	17.72	41.67	46.1
TK12	107.8	82.6	4.56	17.2	76.4	17.00	19.88	42.2
TK13	288.6	115.6	3.74	13.1	128.6	21.82	32.88	34.4
TK14	113.3	102.2	4.16	11.5	228.5	27.76	20.04	85.6
TK15	296.2	176.6	1.17	17.0	217.8	31.22	32.26	94.3
TK16	118.8	121.6	4.95	15.7	140.3	23.04	21.61	57.8
TK17	77.7	38.1	2.08	12.8	91.81	15.11	15.13	32.7
TK18	203.0	33.3	3.47	47.7	112.2	22.07	38.53	21.6
Min.	77.7	33.3	1.2	11.5	59.2	13.8	15.1	21.6
Max.	296.2	176.6	5.0	47.7	228.5	31.2	47.8	94.3
Aver.	165.3	98.0	3.9	21.8	133.8	21.7	28.5	55.3
Median	133.5	94.8	4.3	18.2	123.1	21.9	25.1	52.7
Standard Deviation	67.4	33.5	1.1	9.1	48.8	4.2	9.6	20.3
Kurtosis	-0.27	0.96	1.36	2.28	-0.18	0.42	-0.93	-0.60
Skewness	0.91	0.19	-1.40	1.37	0.71	0.16	0.51	0.47

The average values of the current investigation on heavy metals are 165.3(Zn), 98.0(As), 3.9(Cd), 21.8(Pb), 133.8(Cr), 21.7(Co), 28.5(Ni), 55.3(Cu). According to (Buccolieri et al., 2006) the mean values of Zn, Cr, Ni, and Cu were lower in the marine sediments of Taranto Gulf when compared to our current investigation. Similarly, Cu, Pb, Zn & Cd values were high in the sediments of the Tamaki Estuary, Auckland, New Zealand (Abraham et al., 2007). The obtained grain sizes of the study samples are shown in Fig. 2. The sediment samples consist of sand, silt, and clay contents. According to Folk's triangle diagram (Folk, 1974), the sediments in the study area are classified into three types of silty sand, sandy silt, and silt (Fig. 3), indicating that surface sediments in this region contains mostly silt and sand-fraction sediments, with minor clay-fraction sediments.

If skewness equals zero, the series is symmetrically distributed. The series gets asymmetrically proportional to the value's deviation from zero. The series is not symmetrical because it is right-skewed (positive) when the skewness > 0 and left-skewed (negative) when the skewness < 0. Typically, the arithmetic mean of right-skewed data will be bigger than the mode and median values. In the case of left-skewed data, however, the arithmetic mean is typically smaller than the mode and median values (Yalcin et al., 2016). The asymmetric distribution curves of heavy metals are plotted in Fig. 5. The calculated results of standard deviation (SD), Kurtosis, and Skewness are shown in Table 1. In the present study, heavy metals showed the light right-skewed (positive skew) distribution, except for Cd. In particular, Zn, As, Cr, Co, Ni, and Cu distribution curves are nearly generally distributed with Skewness values of 0.91, 0.19, 0.71, 0.16, 0.51, and 0.47, respectively. This indicates that the contamination of almost all heavy metals in surface sediments in the study area comes from the same sources under the same environmental conditions. Additionally, surface sediments in this region are provided mainly by surrounding rivers in the northeastern region of Vietnam (Nguyen Nhu et al., 2023), indicating that most heavy metals in surface sediments principally originate from Northeastern Vietnam too. The flow rate of the estuary is exceptionally high. Subsequently, the tidal forces withdrew the water, causing it to settle more evenly on both shores of the estuary. The alternative hypothesis relates to aquaculture and other human activities.

The Pearson correlation coefficients among heavy metals in surface sediments in the study area are listed in Table 2. The results in Table 2



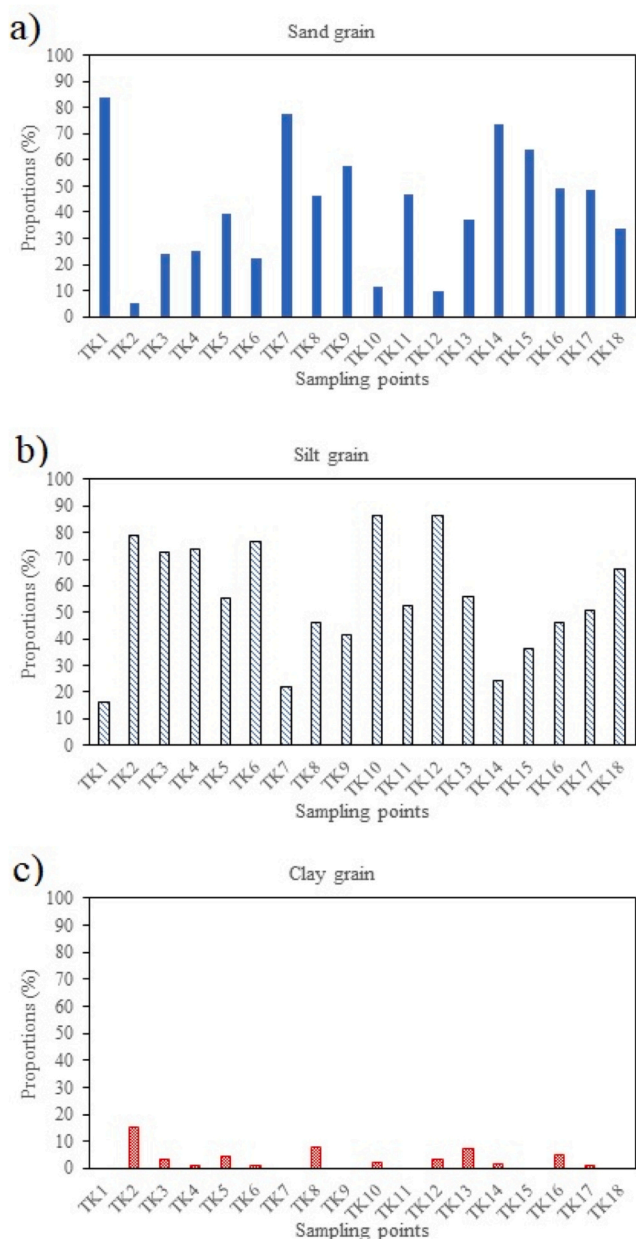


Fig. 2. Proportions of sand, silt, and clay in sediment samples. a) Sand grain proportion; b) Silt proportion; and c) clay grain proportion.

show that Cr exhibits a significant positive correlated with Co with a Pearson correlation coefficient of 0.883 and a value of  $p < 0.01$ . This result is like that of (Liu et al., 2021). As presented in this table, a significant positive correlation is also found between Zn and As, Zn and Ni, As and Co, Cu and As, Cu and Cr and Cu and Co, except for Cd. The positive correlation among heavy metals presents that these heavy metals may originate from the same sources. The correlations between heavy metals and different types of grain are also evaluated and presented in Table 3. As shown in Table 3, heavy metals do not correlate with clay content. Additionally, most heavy metals do not correlate with silt and sand contents, except for Cr. It significantly correlates negatively with silt and sand concentrations ( $p$  values of less than 0.01). Metal and organic matter distribution did not show a strongly marked dependence of its content on particle size, while the distribution character of some metals has a clear similarity. This implies that, although the principal sources of heavy metal pollution in the study area are related to the source of sedimentary material from the mainland near the study area,

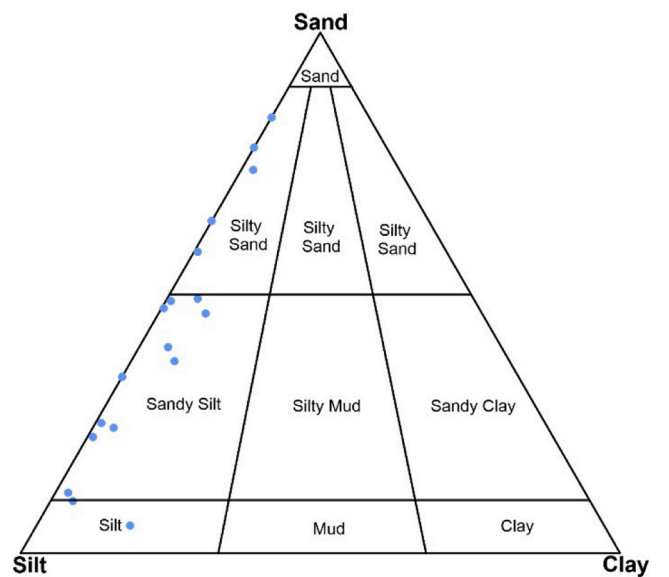


Fig. 3. Ternary diagram of surface sediments in this region following Folk (1974).

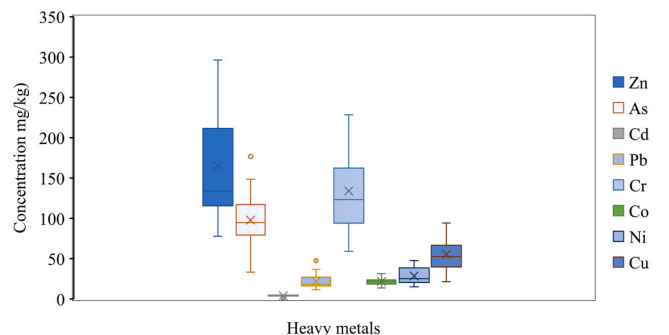


Fig. 4. The minimum, maximum, and average values of the concentration of heavy metals.

the grain size does not control heavy metals in surface sediments in the study area. This may be due to fine sediments (i.e., clay) generally containing high heavy metals than coarse sediments (i.e., silt and sand), but surface sediments in this region have very low clay-fraction sediments and high silt and sand-fraction sediments (Fig. 3). This indicates not strong correlations between heavy metals and size fraction sediments of most surface sediments in this region. However, only Cr shows strong positive correlation with sand-fraction sediments and strong negative correlation with silt-fraction sediments, implying that Cr is mostly found in sand-fraction sediments. This also suggests that Cr is mainly supplied by nearby sources in this region.

The concentration of heavy metals in surface sediments in the study area is compared with that in other sea areas, as shown in Table 4. Contamination of heavy metals in surface sediments in the study indicates that they are higher than the baseline concentrations, except for Pb (Zhang and Du, 2005). The concentration of As is 18.8 times higher than the baseline concentration, and Cd is ten times. The other elements have concentrations 1.6–2.9 times higher than the baseline concentration. The concentration of heavy metals in the present study is also higher than other seas in the world, as mentioned in Table 4 (Alharbi et al., 2017; Ardila et al., 2023; Delshab et al., 2017; Dou et al., 2013; Jahan and Strezov, 2018; Liang and Wong, 2003; Zhao et al., 2016; Zhu et al., 2011), except for Pb and Ni in Truong Sa archipelago, East Vietnam Sea. The concentration of heavy metals with the most significant difference in the study area compared to the Truong Sa archipelago is

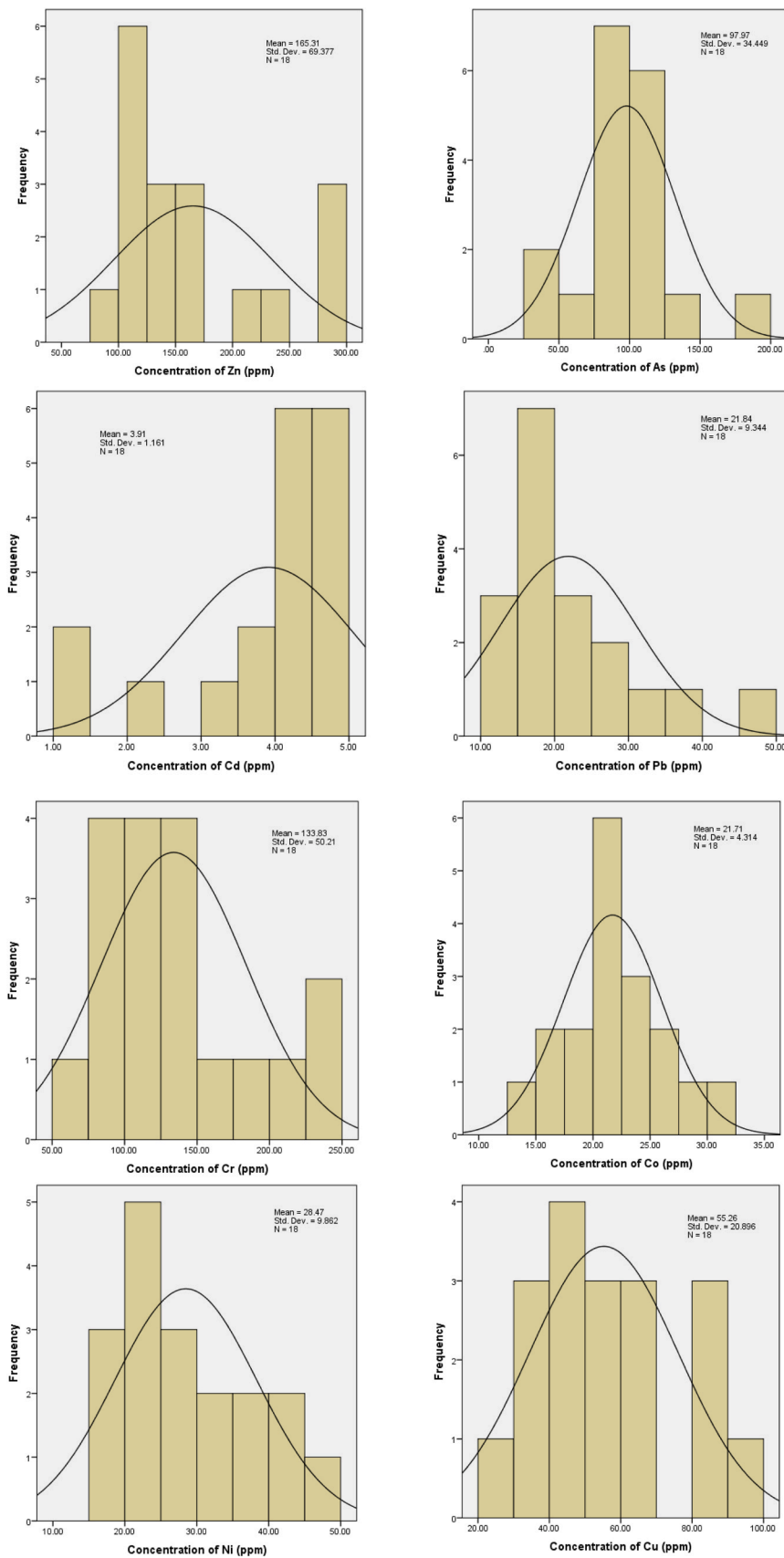


Fig. 5. Distribution curves of heavy metal concentrations in surface sediments in the study area.

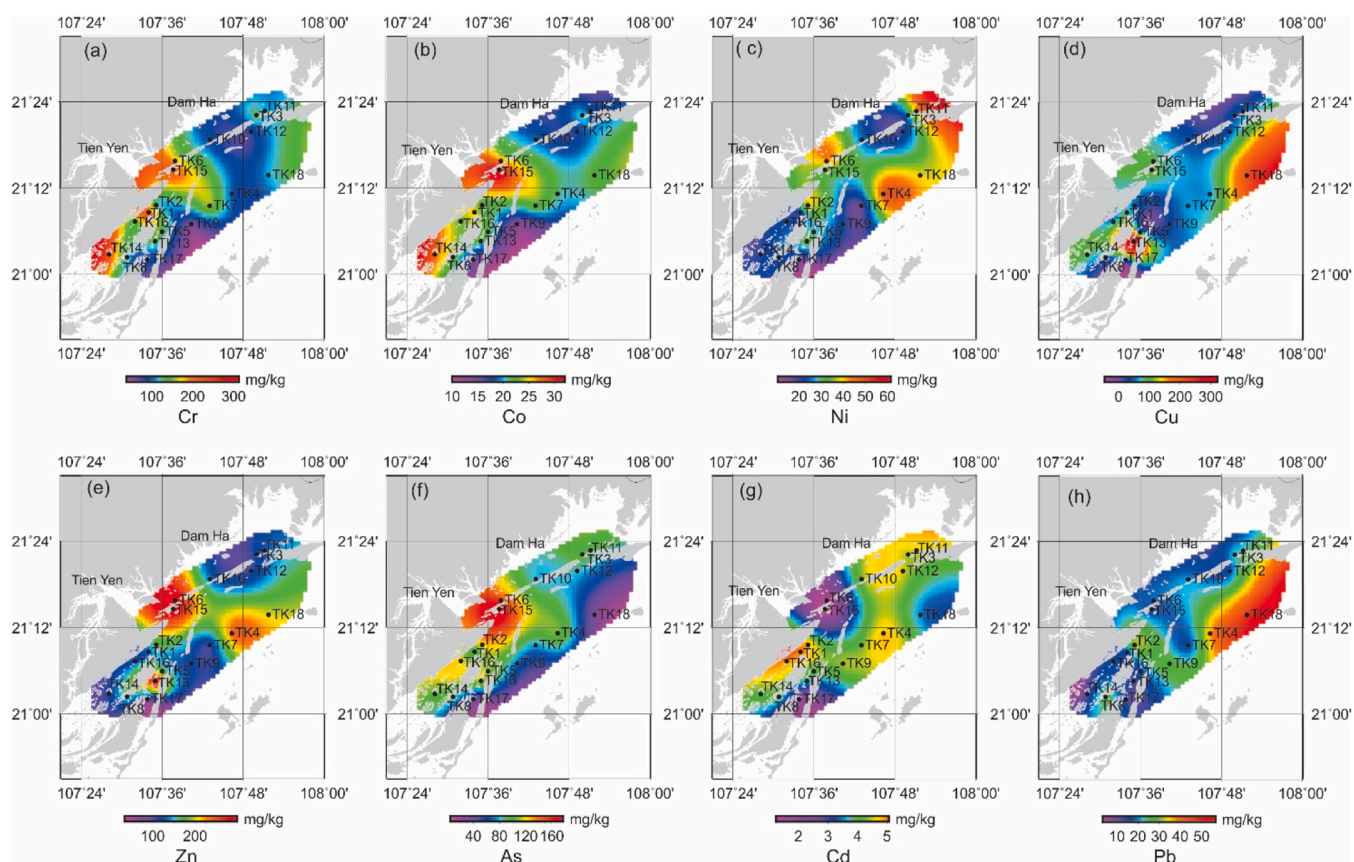


Fig. 6. The spatial distribution of concentration of heavy metals in surface sediments in the study area.

Table 2

The Pearson correlation coefficients among different heavy metals in the study area (\*\*  $p < 0.01$ ; \*  $p < 0.05$ ).

Elements	Zn	As	Cd	Pb	Cr	Co	Ni	Cu
Zn	1							
As	0.627**	1						
Cd	-0.525*	-0.282	1					
Pb	0.185	-0.297	0.180	1				
Cr	0.251	0.523*	-0.272	-0.408	1			
Co	0.520*	0.679**	-0.339	-0.153	0.883**	1		
Ni	0.70**	0.369	-0.124	0.577*	-0.072	0.181	1	
Cu	0.312	0.727**	-0.284	-0.391	0.840**	0.798**	0.059	1

Table 3

Pearson correlation between heavy metal concentration and grain contents in the study area (\*\*  $p < 0.01$ ; \*  $p < 0.05$ ).

Elements	Zn	As	Cd	Pb	Cr	Co	Ni	Cu
Clay	0.034	0.235	0.300	0.075	-0.181	0.014	0.115	0.427
Silt	0.192	-0.011	0.013	0.387	-0.601**	-0.361	0.468	-0.427
Sand	-0.184	-0.030	-0.063	-0.373	0.590**	0.334	-0.456	-0.168

the element Cd, which is about ten times higher. The concentration of other elements is about 1.2–4.4 times higher. Comparing the heavy metal concentration in the study area with some other regions of the Gulf of Tonkin, such as Ba Lat estuary, Hai Phong and Cua Ong coast (see Fig. 1), we see that the concentrations of Zn, Co, and Cu in the study area are not much different from those of Ba Lat estuary, but significantly higher than those of Cua Ong and Hai Phong coasts. The As, Cd, and Cr concentration in the study area is much higher than in these three regions. The concentration of Pb in the study area is lower than that of the Ba Lat estuary and Hai Phong coast but higher than that of the Cua Ong

coast. The difference in heavy metal concentrations of the above regions could be due to the various sediment sources and environmental characteristics. The high concentration of heavy metals in the study area, in comparison with the neighboring regions, such as Hoang Sa and Truong Sa islands or Cua Ong, Hai Phong, and Ba Lat coast areas, could be associated with the type of sediment sources and the difference in human activities.

**Table 4**  
Comparison of heavy metals in surface sediments in the study area and other seas.

Study areas	Zn mg/kg	As mg/kg	Cd mg/kg	Pb mg/kg	Cr mg/kg	Co mg/kg	Ni mg/kg	Cu mg/kg	References
Van Don –Tra Co, Northeast Gulf of Tokin, Vietnam	165.3	98.0	3.9	21.8	133.8	21.7	28.5	55.3	This study
Truong Sa archipelago, East Vietnam Sea	132.6	80.6	0.36	31.42	29.8	14.9	45.4	25.7	(Liu et al., 2021)
Mallorca Island, Spain	8.87	4.98	0.50	10.10	8.77	-	1.69	1.16	(Ardila et al., 2023)
Arabian Gulf, Saudi Arabia	5.70	54.0	-	1.80	5.16	1.72	26.6	4.23	(Alharbi et al., 2017)
Persian Gulf, Iran	21.08	3.70	-	3.39	16.13	2.20	19.0	15.4	(Delshab et al., 2017)
Port Kembla, Australia	235	18.5	0.6	74	11	2	20	59	(Jahan and Strezov, 2018)
Daya Bay, China	87.81	8.16	0.07	37.0	59.0	-	-	16.5	(Zhao et al., 2016)
Zhein Bay, South China	74.9	ND	0.063	35.7	23.1	-	-	7.95	(Zhu et al., 2011)
Hong Kong	149	ND	1.05	52.6	22.4	-	-	42.8	(Liang and Wong, 2003)
Beibu Bay, South China	67.28	9.53	0.16	27.99	53.6	-	-	58.2	(Dou et al., 2013)
Cua Ong coast, Gulf of Tonkin	40	26	0.09	16	-	-	-	20	(Hieu HO et al., 2010)
Hai Phong coast, Gulf of Tonkin	82.34	17.62	0.12	43.85	34.5	13.6	-	37.39	(Dang Hoai et al., 2022)
Ba Lat Estuary Gulf of Tonkin	154.5	-	0.419	76.57	66.25	17.5	-	80.72	Nguyen T.T., et al., 2012
Baseline concentration	103	5.21	0.3	27.6	57.4	10	20	28.6	Zhang and Du (2005)

**3.2. Contamination and ecological risk assessment of heavy metals in sediments**

An ecological risk assessment evaluates the likeliness of an environment to be impacted due to exposure to one or more environmental stressors. The Igeo, CF, PLI, Er, and PER indices were calculated and listed in Table 5. Based on the values of Igeo, it can be seen that the contamination levels of heavy metals in surface sediments in the study area ranged from unpolluted to high levels of contamination status. The studied sediments are strongly polluted due to As and Cd, whereas there was no pollution due to Pb and Ni. On the other hand, the sediments were highly polluted due to contamination of the heavy metals As and Cd. As shown in this table, the CF values range from 0.8 (for Pb) to 18.8 (for As) and follow the descending order: As > Cd > Cu > Cr > Co > Zn > Ni > Pb. Based on the CF value, it can be seen that the contamination of heavy metals in surface sediments exhibit a wide range. The highest pollution level is found for As, whereas the lowest one is observed for Pb. This may indicate that surrounding rivers can supply high As to this area, while they contain low Pb because most of heavy metals mainly come from nearby rivers in this region. The pollution load index (PLI) is 2.84, showing that heavy metals pollute surface sediment. As shown in Table 5, the potential ecological risk index (Er) for heavy metals in the sediment samples varies from 1.6 to 390.9 and follows the order: Zn < Pb < Cr < Ni < Cu < Co < As < Cd. The pollution risk is evaluated with a wide range also, with the highest pollution risk for Cd and the lowest for Zn. High pollution of Cadmium is due to occurrence in the environment from both natural and anthropogenic sources. The existing industrial operations greatly enhance environmental levels as Cd is commonly used as a pigment in paint, plastics, ceramics, and glass manufacturing companies. The comprehensive PERI is 616.8 and is assessed as a high

risk for heavy metal pollution. The recommendation to reduce the potential pollution of heavy metals in marine sediments is to proper disposal of water, use of alternative materials, regular monitoring, cleanup operations and reduce the use of heavy metals (Perumal et al., 2021).

**4. Conclusions**

The heavy metal concentrations in surface sediments were found to be greatest in Zn, with the following elements listed in descending order: Zn > Cr > As > Cu > Ni > Pb > Co > Cd. Except for Pb, the concentrations of heavy metals in the surface sediments surface of the current study area are more than in several other regions and the baseline concentration. In relation to the levels of pollution and the associated risks, the pollutants As and Cd exhibited the greatest values, while Pb and Zn displayed the lowest. The ecological risk index classifies the current study area as having a significant susceptibility to heavy metal contamination. Based on statistical association indicators, we are persuaded that the origin of heavy metal pollution is consistent with the source of sedimentary material from the mainland in close proximity to the research area. Future proposals include routine monitoring of marine sediments in order to facilitate the implementation of preventative measures against more pollution.

**CRedit authorship contribution statement**

**Nam Bui Van:** Data curation, Formal analysis, Validation, Writing – original draft. **Duc Anh Le:** Data curation, Formal analysis, Validation, Writing – original draft. **Trung Nguyen Nhu:** Conceptualization, Data curation, Funding acquisition, Investigation, Project administration,

**Table 5**  
The value of CF, PLI, Er, and PER indices in the study area.

Metals Index	Zn	As	Cd	Pb	Cr	Co	Ni	Cu
C <sub>i</sub>	165.3	98.0	3.9	21.8	133.8	21.7	28.5	55.3
Bi	103	5.21	0.3	27.6	57.4	10	20	28.6
Igeo	0.1	3.6	3.1	-0.9	0.6	0.5	-0.1	0.4
Evaluation	Unpolluted to moderate	Strong	Strong	Unpolluted	Unpolluted to moderate	Unpolluted to moderate	Unpolluted	Unpolluted to moderate
CF	1.6	18.8	13.0	0.8	2.3	2.2	1.4	1.9
Evaluation	Moderate	High	High	Low	Moderate	Moderate	Moderate	Moderate
PLI	2.84							
T <sub>r</sub> <sup>i</sup>	1	10	30	5	2	5	5	5
E <sub>r</sub> <sup>i</sup>	1.6	188.0	390.9	4.0	4.7	10.9	7.1	9.7
Evaluation	Low	High	High	Low	Low	Low	Low	Low
PER	621.7							



Validation, Writing – original draft, Writing – review & editing. **Van-Hao Duong**: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing. **Sang Pham Nhu**: Data curation, Investigation, Writing – original draft, Writing – review & editing. **Mohamed Saiyad Musthafa**: Data curation, Formal analysis, Validation, Writing – original draft, Writing – review & editing. **Tung Dang Xuan**: Data curation, Formal analysis. **Hoai-Nam Tran**: Data curation, Writing – original draft, Writing – review & editing. **Thuy Huong Tran Thi**: Data curation, Formal analysis. **Thanh-Duong Nguyen**: Conceptualization, Data curation, Formal analysis, Writing – original draft, Writing – review & editing.

## Declaration of Competing Interest

The undersigned hereby declare that:

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4. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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## References

- Abraham, G., assessment, R.P.-E. monitoring and, 2008, , 2007. Assessment of heavy metal enrichment factors and the degree of contamination in marine sediments from Tamaki Estuary, Auckland, New Zealand. SpringerGMS Abraham, RJ ParkerEnvironmental monitoring and assessment, 2008•Springer 136, 227–238. <https://doi.org/10.1007/s10661-007-9678-2>.
- Alharbi, T., Alfaifi, H., Almadani, S.A., El-Sorogy, A., 2017. Spatial distribution and metal contamination in the coastal sediments of Al-Khafji area, Arabian Gulf, Saudi Arabia. *Environ. Monit. Assess.* 189 <https://doi.org/10.1007/s10661-017-6352-1>.
- Al-Mutairi, K., ... C.Y. of environmental research and public, 2021, , 2021. A review of heavy metals in coastal surface sediments from the red sea: Health-ecological risk assessments. *mdpi.comKA Al-Mutairi, CK YapInternational journal of environmental research and public health*, 2021•mdpi.com 18, 2798. <https://doi.org/10.3390/ijerph18062798>.
- Angulo, E., 1996. The Tomlinson Pollution Load Index applied to heavy metal, 'Mussel-Watch' data: a useful index to assess coastal pollution. *Sci. Total Environ.* 187, 19–56. [https://doi.org/10.1016/0048-9697\(96\)05128-5](https://doi.org/10.1016/0048-9697(96)05128-5).
- Ardila, P.A.R., Alonso, R.Á., Valsero, J.J.D., García, R.M., Cabrera, F.Á., Cosío, E.L., Laforet, S.D., 2023. Assessment of heavy metal pollution in marine sediments from southwest of Mallorca island, Spain. *Environ. Sci. Pollut. Res.* 30, 16852–16866. <https://doi.org/10.1007/s11356-022-25014-0/FIGURES/10>.
- Benhaddya, M., M Hadjel, 2013. Spatial distribution and contamination assessment of heavy metals in surface soils of Hassi Messaoud, Algeria. SpringerML Benhaddya, M HadjelEnvironmental Earth Sciences, 2014•Springer 71, 1473–1486. <https://doi.org/10.1007/s12665-013-2552-3>.
- Buccolieri, A., Buccolieri, G., Cardellicchio, N., Dell'Atti, A., Di Leo, A., Maci, A., 2006. Heavy metals in marine sediments of Taranto Gulf (Ionian Sea, Southern Italy). *Mar. Chem.* 99, 227–235. <https://doi.org/10.1016/J.MARCHEM.2005.09.009>.
- Chan, L.S., Ng, S.L., Davis, A.M., Yim, W.W.S., Yeung, C.H., 2001. Magnetic Properties and Heavy-metal Contents of Contaminated Surface Sediments of Penny's Bay, Hong Kong. *Mar. Pollut. Bull.* 42, 569–583. [https://doi.org/10.1016/S0025-326X\(00\)00203-4](https://doi.org/10.1016/S0025-326X(00)00203-4).
- Dang Hoai, N., Thanh Dương, N., Thi Thu Do, H., Hoang, C., 2022. Distribution and ecological risk of heavy metal (loid) s in surface sediments of the Hai Phong coastal area, North Vietnam. Taylor & FrancisDH Nhon, ND Thanh, HN Manh, L Nguyen Thi Mai, H Do Thi Thu, C Hoang Thi, L Van NamChemistry and Ecology, 2022•Taylor & Francis 38, 27–47. <https://doi.org/10.1080/02757540.2021.2017901>.
- Delshab, H., Farshchi, P., Keshavarzi, B., 2017. Geochemical distribution, fractionation and contamination assessment of heavy metals in marine sediments of the Asaluyeh port, Persian Gulf. *Mar. Pollut. Bull.* 115, 401–411. <https://doi.org/10.1016/J.MARPOLBUL.2016.11.033>.
- Dou, Y., Li, J., Zhao, J., Hu, B., Yang, S., 2013. Distribution, enrichment and source of heavy metals in surface sediments of the eastern Beibu Bay, South China Sea. *Mar. Pollut. Bull.* 67, 137–145. <https://doi.org/10.1016/J.MARPOLBUL.2012.11.022>.
- Gao, W., Du, Y., Gao, S., Ingels, J., Wang, D., 2016. Heavy metal accumulation reflecting natural sedimentary processes and anthropogenic activities in two contrasting coastal wetland ecosystems, eastern China. *J. Soils Sediment.* 16, 1093–1108. <https://doi.org/10.1007/S11368-015-1314-0>.
- Gargouri, D., Azri, C., Serbaji, M.M., Jedoui, Y., Montacer, M., 2011. Heavy metal concentrations in the surface marine sediments of Sfax Coast, Tunisia. *Environ. Monit. Assess.* 175, 519–530. <https://doi.org/10.1007/S10661-010-1548-7>.
- Gkaragkouni, A., Sergiou, S., Geraga, M., Papaefthymiou, H., Christodoulou, D., Papatheodorou, G., 2021. Heavy Metal Distribution, Sources and Contamination Assessment in Polluted Marine Sediments: Keratsini Outfall Sewer Area, Saronikos Gulf, Greece. *Water Air Soil Pollut.* 232, 1–22. <https://doi.org/10.1007/S11270-021-05400-Z/METRICS>.
- Harikrishnan, N., Ravisankar, R., Chandrasekaran, A., Suresh Gandhi, M., Kanagasabapathy, K.V., Prasad, M.V.R., Satapathy, K.K., 2017. Assessment of Heavy Metal Contamination in Marine Sediments of East Coast of Tamil Nadu Affected by Different Pollution Sources. *Mar. Pollut. Bull.* 121, 418–424. <https://doi.org/10.1016/J.MARPOLBUL.2017.05.047>.
- Hieu HO, H., SWENNEN, R., VAN DAMME, A., 2010. Distribution and contamination status of heavy metals in estuarine sediments near Cua Ong Harbor, Ha Long Bay, Vietnam. *Geol. Belg.* 13. <https://popups.uliege.be/1374-8505/index.php?id=2765>.
- Huong, D.T.T., Ha, N.T.T., Do Khanh, G., Van Thanh, N., Hens, L., 2022. Sustainability assessment of coastal ecosystems: DPSIR analysis for beaches at the Northeast Coast of Vietnam. *Environ. Dev. Sustain* 24, 5032–5051. <https://doi.org/10.1007/S10668-021-01648-X/METRICS>.
- Jahan, S., Strezov, V., 2018. Comparison of pollution indices for the assessment of heavy metals in the sediments of seaports of NSW, Australia. *Mar. Pollut. Bull.* 128, 295–306. <https://doi.org/10.1016/J.MARPOLBUL.2018.01.036>.
- Leung, H.M., Cheung, K.C., Au, C.K., Yung, K.K.L., Li, W.C., 2021. An assessment of heavy metal contamination in the marine soil/sediment of Coles Bay Area, Svalbard, and Greater Bay Area, China: a baseline survey from a rapidly developing bay. *Environ. Sci. Pollut. Res.* 28, 22170–22178. <https://doi.org/10.1007/S11356-021-13489-2/TABLES/4>.
- Liang, Y., Wong, M.H., 2003. Spatial and temporal organic and heavy metal pollution at Mai Po Marshes Nature Reserve, Hong Kong. *Chemosphere* 52, 1647–1658. [https://doi.org/10.1016/S0045-6535\(03\)00505-8](https://doi.org/10.1016/S0045-6535(03)00505-8).
- Liu, J., Peng, A., Deng, S., Liu, M., Liu, G., Li, C., 2021. Distribution of heavy metals and radionuclides in the sediments and their environmental impacts in Nansha Sea area, South China Sea. *Mar. Pollut. Bull.* 166, 112192 <https://doi.org/10.1016/J.MARPOLBUL.2021.112192>.
- Ogoyi, D.O., Mwita, C.J., Nguu, E.K., Shiundu, P.M., 2011. Determination of Heavy Metal Content in Water, Sediment and Microalgae from Lake Victoria, East Africa. *Open Environ. Eng. J.* 4, 156–161.
- Perumal, K., Antony, J., Muthuramalingam, S., 2021. Heavy metal pollutants and their spatial distribution in surface sediments from Thondi coast, Palk Bay, South India. *Environ. Sci. Eur.* 33, 1–20. <https://doi.org/10.1186/S12302-021-00501-2/TABLES/7>.
- Romano, E., Bergamin, L., Croudace, I.W., Pierfranceschi, G., Sesta, G., Ausili, A., 2021. Measuring anthropogenic impacts on an industrialised coastal marine area using chemical and textural signatures in sediments: A case study of Augusta Harbour (Sicily, Italy). *Sci. Total Environ.* 755, 142683 <https://doi.org/10.1016/J.SCITOTENV.2020.142683>.
- S, M., MNA, M., KMPAH, P., 2013. An Assessment of Heavy Metal Contamination in Marine Sediments Precautionary Measures for Environmental Impact Management at Harbor Development: Galle Harbor Sri Lanka. *J. Ecotechnol. Res.* 17, 29–33. <https://doi.org/10.11190/JER.17.29>.
- Tanjung, R.H.R., Hamuna, B., Yonas, M.N., 2019. Assessing Heavy Metal Contamination in Marine Sediments Around the Coastal Waters of Mimika Regency, Indonesia. *J. Ecol. Eng.* Vol. 20 <https://doi.org/10.12911/22998993/113411>.
- Tomlinson, D.L., Wilson, J.G., Harris, C.R., Jeffrey, D.W., 1980. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. *Helgol. änder Meeresunters.* 33, 566–575. <https://doi.org/10.1007/BF02414780/METRICS>.
- Varol, M., 2011. Assessment of heavy metal contamination in sediments of the Tigris River (Turkey) using pollution indices and multivariate statistical techniques. *J. Hazard Mater.* 195, 355–364. <https://doi.org/10.1016/J.JHAZMAT.2011.08.051>.
- Wang, Z., Luo, P., Zha, X., Xu, C., Kang, S., Zhou, M., Nover, D., Wang, Y., 2022. Overview assessment of risk evaluation and treatment technologies for heavy metal pollution of water and soil. *J. Clean. Prod.* 379, 134043 <https://doi.org/10.1016/J.JCLEPRO.2022.134043>.
- Yalcin, F., Nyamsari, D., Paksu, E., Filomat, M.Y., 2016. Statistical assessment of heavy metal distribution and contamination of beach sands of Antalya-Turkey: an approach to the multivariate analysis techniques. *JSTORF Yalcin, DG Nyamsari, E Paksu, MG YalcinFilomat*, 2016•JSTOR 30, 945–952. <https://doi.org/10.2298/FIL1604945Y>.

- Yushin, N., Jakhu, R., Chaligava, O., Grozdov, D., Zinicovscaia, I., 2023. Natural and anthropogenic radionuclides concentration with heavy metals analysis of the sediments collected around Novaya Zemlya. *Mar. Pollut. Bull.* 194, 115346 <https://doi.org/10.1016/J.MARPOLBUL.2023.115346>.
- Zhang, Y.H., Du, J.M., 2005. Background values of pollutants in sediments of the South China Sea. *Acta Oceanol. Sin.* 27161–27166.
- Zhao, G., Ye, S., Yuan, H., Ding, X., Wang, J., 2016. Distribution and contamination of heavy metals in surface sediments of the Daya Bay and adjacent shelf, China. *Mar. Pollut. Bull.* 112, 420–426. <https://doi.org/10.1016/J.MARPOLBUL.2016.07.043>.
- Zhu, L., Xu, J., Wang, F., Lee, B., 2011. An assessment of selected heavy metal contamination in the surface sediments from the South China Sea before 1998. *J. Geochem Explor* 108, 1–14. <https://doi.org/10.1016/J.GEXPLO.2010.08.002>.