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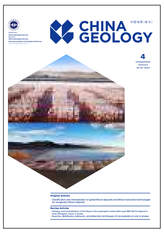

Heavy metal pollution assessment in marine sediments in the Northwest coast of Sabah, Malaysia

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ABSTRACT

Heavy metal contents along the Northwest coast of Sabah were determined to interpret the pollution level in the marine sediment. The metal abundance is regulated by the physico-chemical properties such as the average sediment pH (7.82, 9.00 and 8.99), organic matter (0.62%, 1.60%, and 2.27%), moisture content (25.00%, 29.70%, and 15.00%) and sandy texture in Kota Belud, Kudat and Mantanani Island, respectively. The major elements show Ca>Fe>Mg>Al>Mn for all study sites, while the heavy metals show Ni>Cr>Zn>Cu>Co>Pb, Cr>Ni>Zn>Cu>Pb>Co and Zn>Pb>Cr>Ni, for Kota Belud, Kudat and Mantanani Island, respectively. The pollution degree of heavy metals was evaluated by using the Sediment Quality Assessment (SQA). The SQA parameters indicated none to moderate pollution in Kota Belud that shows Class 0, Class 1 and Class 2 pollution. The parameters also indicated none to low pollution in Kudat and Mantanani Island that show only Class 0 pollution. The enrichment factor (EF) suggested minor to moderately severe metal enrichment by anthropogenic sources in Kota Belud, whereas only minor enrichment in Kudat and Mantanani Island. The modified pollution degree ($MCD<1.5$) and pollution load index ($0\leqslant PLI<1$) indicating only low pollution level in the marine sediments for all study sites. The objectives of this study are: (1) to determine the physico-chemical parameters of sediments, (2) interpret the heavy metal contents and (3) evaluate the sediment quality.

1. Introduction

Sediment pollution caused by heavy metals is a serious environmental problem due to their toxicity and irreversible effects. Heavy metals become pollutants when their concentrations exceed the permissible range of the risks appraisal established in sediment quality guidelines (SQGs) (Praveena SM et al., 2008; Sany SBT et al., 2013; Kumar V et al., 2019; Zhang Y et al., 2021). Their persistence, non-biodegradable and mutagenic properties in sediments may cause severe threats to human health, living organisms and the ecosystem (Jayamurali D et al., 2021). Heavy metals like Cu, Fe, Mn and Zn are essential while in low concentrations, whereas metals like Co, Cr, Pb and Hg are carcinogenic even at minute quantities (Shine JP et al., 1995; Abdu N et al., 2017).

The study areas were potential sites for socio-economic development such as Kota Belud for industrial and paddy agriculture (Tahir SH and Talip AM, 2020), Kudat for mariculture and fishery activities (Tsen HW et al., 2018) and Mantanani Island for tourism and fishery activities (Saleh E et al., 2021; Harun MA et al., 2021). Although lithogenic metals due to varying local geology contribute to the release of heavy metals in the study area, anthropogenic metals are the main environmental concern which are significant when assessing their distribution and pollution level (Vallius H et al., 2007; Wuana RA and Okeimen FE, 2011). These metals are naturally released during active weathering of the earth ’s crust, pedogenesis of the parent rocks and soil erosion, which are then transported via river discharge, surface run-offs, leachate or drainage structure into the marine sediments (Jaishankar M et al., 2014; Abdu N et al., 2017). Therefore, the sediments along coastal regions that act as a natural catchment reservoir for the deposition and accumulation of

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heavy metal burden were collected for the SQA study.

The bioavailability, distribution and potential impacts of the metal contaminants are often influenced by the physico-chemical properties of the marine sediments, such as the sediment pH, organic matter, moisture content, particle size distribution (*PSD*) and textural classification of the sediment (Hassan FM et al., 2010; Keshavarzifard M et al., 2019). Their mobility and intensity are also facilitated by metal speciation and metal binding through direct adsorption with clay or organic matter, coprecipitation of solid phases, metal complexes or ionic binding mechanisms (Zhou YF and Haynes RJ, 2010; Jaishankar M et al., 2014). The parameters used to monitor the extent of metal pollution are single pollution indices like geoaccumulation index (I_{geo}), enrichment factor (*EF*) and pollution factor (*CF*), as well as complex pollution indices like modified pollution degree (*MCD*) and pollution load index (*PLI*). The main objectives are to determine the physico-chemical parameters of sediments, estimate the heavy metal contents and evaluate the sediment quality. Therefore, this study is suitable as a geochemical baseline data to monitor and assess the environmental pollution caused by heavy metals in marine sediments for future socio-economic development of the study area.

2. Materials and Methods

2.1. Geological Setting

The samples were collected from the northwest coast of Sabah, stretching along Kudat and Kota Belud, including the offshore Mantanani Island. The sampling locations are bounded by latitudes 6°18'N to 7°00'N and longitudes 116°15'E to 116°45'E (Figs. 1 and 2), which are selected based on the pollution sources and geological background. The regional tropical climate promotes active weathering, while the heavy rainfall transports the weathered terrigenous materials from hilly reliefs or inland regions to the coastal shores. The study sites are made up of seven major formations, such as Ophiolite Complex, Chert-Spilite Formation, Crocker Formation, Kudat Formation, Wariu Formation (mélange), Timohing Formation and Quaternary alluvium which control the spatial distribution and abundance of heavy metals in the sediments (Idris MB and Kok KH, 1990; Graves JE et al., 2000; Lee CP et al., 2004; Mansor HE et al., 2021; Ling SY et al., 2022). Recent carbonate build-ups

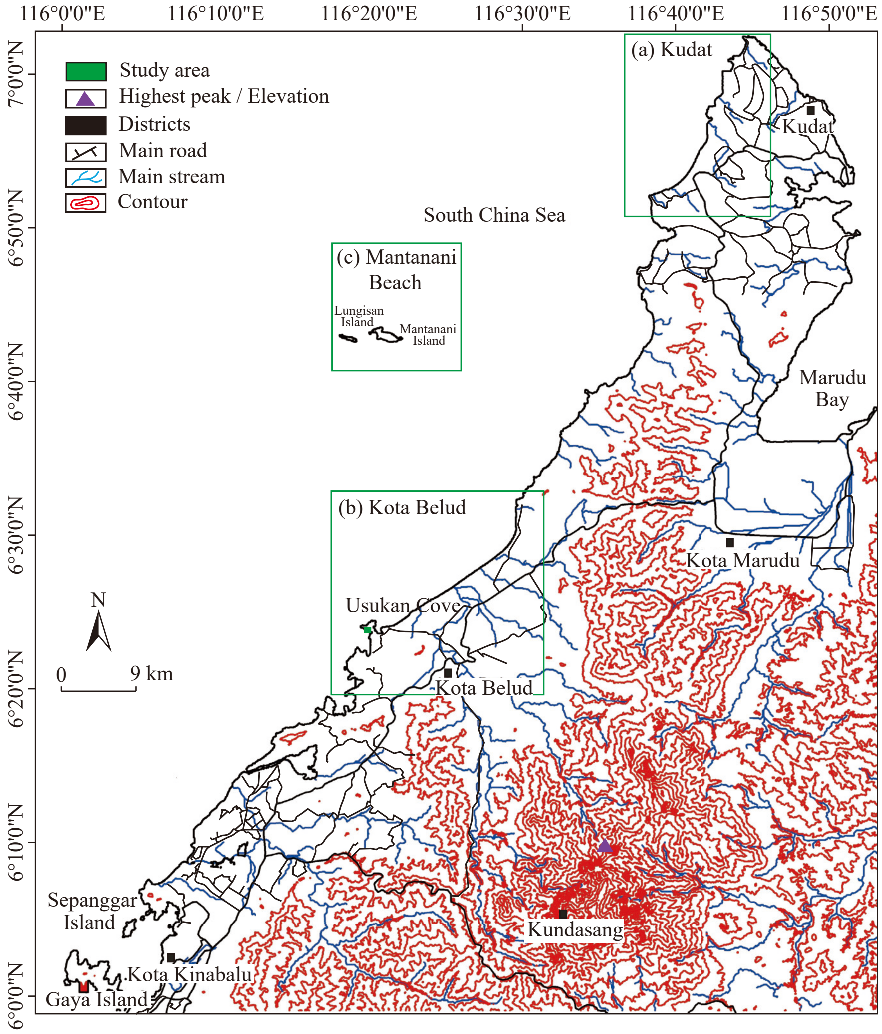

Fig. 1. Base map of the study sites (a)–Kudat, (b)–Kota Belud and (c)–Mantanani Island from the northwest coast of Sabah.

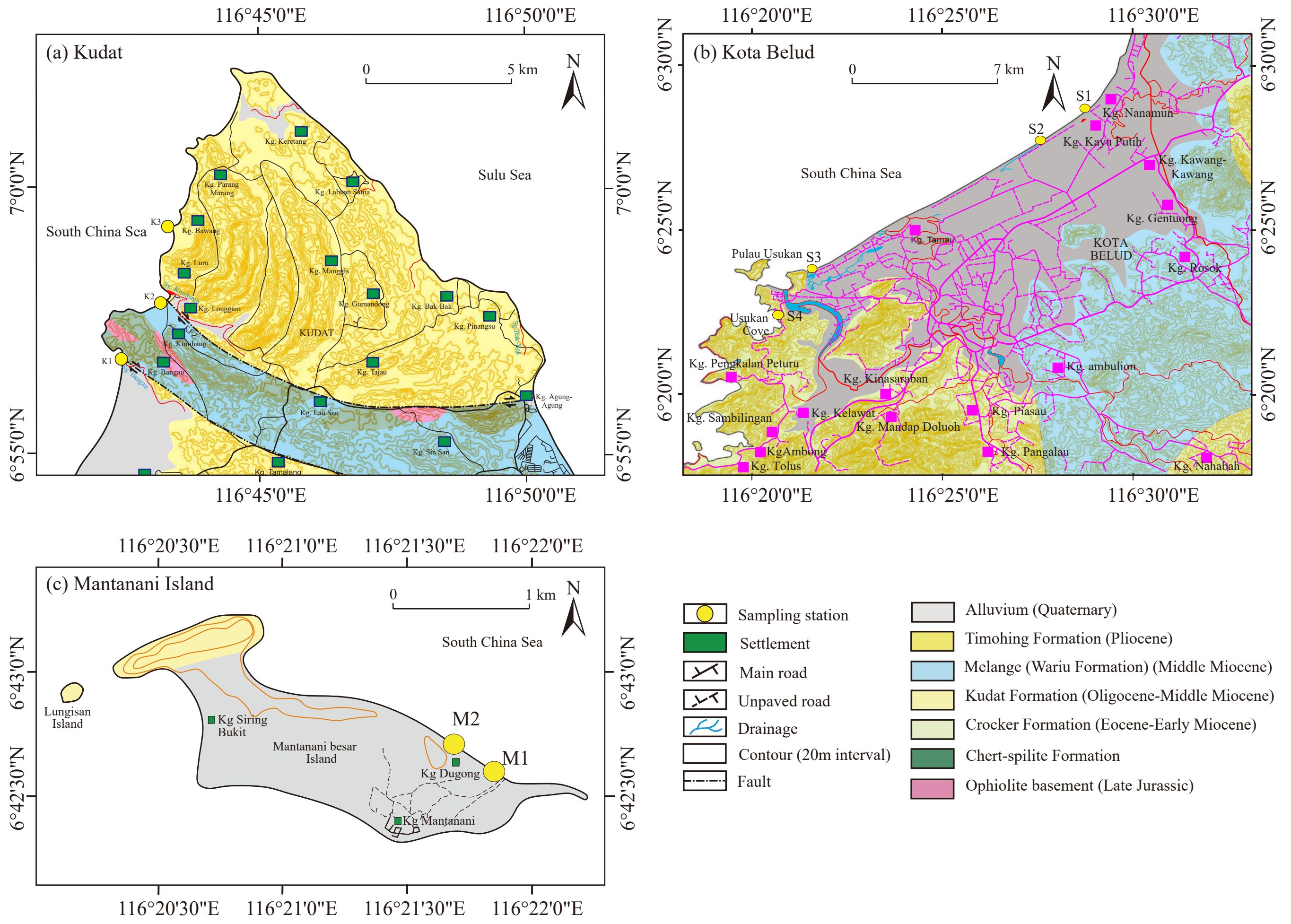

Fig. 2. Geological map of the study sites (a)–Kudat, (b)–Kota Belud and (c)–Mantanani Island, from the northwest coast of Sabah (modified from Department of Minerals and Geoscience of Malaysia, 2010).

from coral reefs and calcareous marine skeletons were also found distributed along the nearby Mantanani Island. Station 1 to 3 from Kota Belud are located in the alluvial region near the Wariu Formation, whereas Station 4 is located within the Crocker Formation. All sampling stations from Kudat and Mantanani Island are located in the recent alluvial deposits of carbonate sands, corals and skeletal fragments. The dismembered ophiolites and local geology affect the metal abundance and pollution level of the present study.

2.2. Sediment Sampling

Twenty-seven core samples were collected (up to 100 cm deep from the surface) along the coastal region using a core sampler attached to a PVC (Polyvinyl chloride) pipe and were tightly closed with PVC stopper. From the 27 core samples, ten were collected from Kota Belud, thirten 13 from Kudat and four from Mantanani Island as shown in Fig. 2. The core samples were then slowly extruded and subdivided into 10 cm cutting, that amount to a total of 153 sediment samples. However, only the average reading of each core samples was shown in Table 1. The samples were stored into airtight ziplock bags for preservation and were taken to the laboratory for further analysis.

2.3. Experimental

The pH test, moisture content (*MC*), organic matter (*OM*) and particle size distribution (*PSD*) analysis were performed based on the British Standard Method (BSI, 1990) to determine the physico-chemical parameters of the marine sediments. The extracted heavy metals from the carbonate coastal sediment were analyzed using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) model Perkin Elmer Optima 5300 DV. Method of determination using ICP-OES follows the USEPA 6010D method (USEPA, 2014). The aqua regia digestion method was performed by adding 1 g of air-dried fine-grained fractions (<63 μm) into the HCl : HNO_3 mixture in the ratio of 1:3 based on the USEPA 3050B method (USEPA, 1996). The major elements determined were Al, Ca, Fe, Mg and Mn, while the heavy metals determined were Co, Cr, Cu, Ni, Pb and Zn as shown in Table 2. The ecotoxicity of the heavy metals were also determined by comparing the heavy metals to the sediment quality guidelines (SQGs) (USEPA, 1977; Simpson SL et al., 2013) and background average of shale values (ASVs) (Turekian KK and Wedepohl KH, 1961) also as indicated in Table 2.

2.4. Statistical Analysis

The sediment quality analysis (SQA) was carried out using five main pollution indices, including the I_{geo} (Müller J, 1969), *EF* (Taylor SR, 1964), *CF*, *MCD* (Hakanson L, 1980) and *PLI* (Tomlinson DL et al., 1980) to interpret the intensity and extent of metal pollution in the marine sediment. Fe was selected as the reference element due to its high stability, lithogenic origin and low pollution in sediments. The association between the variables was also interpreted using the Pearson correlation matrix to interpret the potential sources of the variables via the IBM SPSS version 28 Software.

3. Results and Discussion

3.1. Physico-chemical Properties

Table 1 shows the physico-chemical disposition of marine sediments from the coastal regions of northwest Sabah. The geochemical parameters determined are the arithmetic average values for pH, moisture content (*MC*/%), organic matter (*OM*/%) and grain size based on the clay, silt and sand percentage in the sediment samples. The results show alkaline pH 7.82±0.46, 9.00±0.25 and 8.99±0.20 for Kota Belud, Kudat and Mantanani Island, respectively. The moisture content is the highest in Kudat (29.70%±4.09%), followed by Kota Belud (25.00%±4.56%) and Mantanani Island (15.00%±5.83%), whereas their organic matter is 1.60%±0.45%, 0.62%±0.62% and 2.27%±0.49%, for Kota Belud, Kudat and Mantanani Island, respectively. The overall sediment distribution pattern in the three regions were dominated by sandy textures (>95%), with low silt/clay fractions (<5%) along the coasts.

The spatial distribution and mobility of heavy metals from the source to various parts of marine sediment are influenced by the physico-chemical properties such as pH level, moisture, organic content and granulometric classification of the sediment along the coastal shores (Gwak YS and Kim SH, 2016; Ali H et al., 2019). These parameters also control the metal solubility, speciation, precipitation and sorption

Table 1. Average readings of each core samples collected for the physico-chemical parameters of the sediments.

Study Area	Station	Core ID	Physicochemical Properties					
			pH	*MC*/%	*OM*/%	Sand/%	Silt/%	Clay/%
Kota Belud	S1	TP1-1	7.09	25.01	0.25	99.14	0.46	0.40
		TP1-2	7.87	25.76	0.27	99.65	0.20	0.15
	S2	TP2-1	7.34	24.03	0.16	99.90	0.05	0.05
		TP2-2	7.81	23.82	0.24	99.55	0.30	0.15
		TP2-3	7.88	15.32	0.20	99.70	0.25	0.05
		TP2-4	7.51	25.30	0.12	99.59	0.25	0.15
	S3	TP3-1	7.25	26.16	0.32	98.37	1.02	0.61
		TP3-2	7.74	33.98	0.77	98.78	0.91	0.30
	S4	TP4-1	8.44	25.94	1.63	99.75	0.10	0.15
		TP4-2	8.48	27.89	1.78	98.73	0.91	0.36
	Range		7.09–8.48	15.32–33.98	0.12–1.78	98.37–99.75	0.05–1.02	0.05–0.61
	Mean±SD		7.82±0.46	25.00±4.56	0.62±0.62	99.24±0.48	0.45±0.36	0.24±0.18
Kudat	K1	S1	9.14	23.57	0.61	99.97	0	0.03
		S2	8.79	27.64	1.11	99.86	0.04	0.1
		S3	8.54	25.67	1.00	98.98	1.02	0
		S4	9.05	25.78	1.19	99.89	0.03	0.08
		S5	8.81	26.90	1.56	99.78	0.08	0.14
		S6	8.71	27.20	1.22	99.51	0.24	0.25
	K2	S7	9.24	31.26	1.61	98.24	0.96	0.8
		S8	8.75	34.58	1.70	98.20	1.58	0.22
		S9	9.16	32.10	1.71	98.24	1.71	0.05
		S10	8.77	33.81	1.61	98.45	1.37	0.18
		S11	9.10	35.11	1.99	98.53	1.29	0.18
		S12	9.20	32.52	1.76	98.69	1.16	0.15
	K3	S13	9.38	24.23	2.32	99.70	0.15	0.15
	Range		8.54–9.38	23.57–35.11	0.61–2.32	98.20–99.97	0–1.71	0–0.81
	Mean±SD		9.00±0.25	29.70±4.09	1.60±0.45	99.08±0.72	0.74±0.66	0.18±0.20
Mantanani Island	M1	M1.	9.2	10.19	1.92	94.91	5.09	0
		M2.	9.05	11.55	2.4	94.97	5.01	0
	M2	M3.	8.76	20.93	1.99	94.97	5.03	0
		M4.	8.8	20.92	2.98	94.99	5.01	0
	Range		8.76–9.20	10.19–20.93	1.92–2.98	94.91–94.99	5.01–5.09	0
	Mean±SD		8.99±0.20	15.00±5.83	2.27±0.49	94.97±0.04	5.03±0.04	0

processes (Mugwar AJ and Harbottle MJ, 2016). According to Singare PU *et al*. (2011), pH is a significant indicator to assess the pollution level in marine sediment, in which pH<6 or pH>10 is considered hazardous to the biogenic marine life. Extreme pH either too high or too low tend to increase the toxicity of heavy metals that can damage juvenile fish or directly kill adult fish, most corals and other invertebrates (Alabaster JS and Lloyd RS., 2013; Shi W et al., 2016). However, the present study shows the pH range is within 7.09–8.48, 8.54–9.38 and 8.76–9.20 for Kota Belud, Kudat and Mantanani Island, respectively which suggested that the current conditions are not hazardous.

The sorption behaviors of heavy metals to form weakly-bounded complexes on sediment surfaces are regulated by the pH level to facilitate coprecipitation, dissociation and remobilization of the metals from surficial sediments (Miranda LS et al., 2022). The alkalinity is mainly influenced by the seawater influx of South China Sea during high tides and the disintegration of carbonate-rich shelled organisms or corals washed ashore to the coastal regions of northwest Sabah (Yi LS et al., 2021). The biogenic carbonate sediment also facilitates the marine biogeochemical cycle, thus affecting the spatial distribution of metals (Zhou H et al., 2004; Praveena SM et al., 2008). The bicarbonate ions released during the weathering of silicate and carbonate minerals increase the capacity of carbonate-buffer system which resists fluctuation in the pH value and metal solubility in marine sediments (Zubir AA et al., 2018). The pedogenesis of serpentinized basement rocks originating from the intrusion of oceanic crusts in Kudat also contributes to the alkaline conditions of the shoreface sediments (Graves JE *et al.*, 2000; Luo JZ et al., 2020). The alkaline disposition further retains any labile or amorphous metals such as iron or manganese compounds, the carbonates and weakly bounded metals to the organic matter, which further reduce the bioavailability of heavy metals in the sediments (Loring DH and Rantala RT, 1992; Koukina SE et al., 2016).

Mantanani Island has higher major input of organic content due to the large amount of terrestrial plant remains near the sampling locations, as compared to Kota Belud and Kudat. The decomposed organic ligands and humus have active binding sites to form stable organo-metal complexes, thereby increasing the metal extraction from the source (Rieuwerts JS et al., 1998). Meanwhile, the sandy granulometric textures have higher porosity and permeability that allow further metal mobility and leachability to the catchment regions. Since metal contaminants are carried along effluent discharge and surface run-offs, sediments with higher moisture, organic and clay content have higher pollution degree. Heavy metal content is also higher in fine silt/clay fractions than sand, as clay contents have higher adsorption-desorption rate to accumulate the metals (Abrahim GMS et al., 2008; Nobi EP et al., 2010; Gu S et al., 2019).

3.2. Elemental concentrations

Table 2 shows the comparison of the elemental concentrations in marine sediments from Kota Belud, Kudat and Mantanani Island, with their average background values and the permissible range as suggested in SQGs by US EPA and ANZECC/ARMCANZ legislations. The ISQG-low is the trigger values, while ISQG-high represents high pollution that can adversely impact the sediment quality, marine organisms and the environment. Among the three study sites, Kota Belud has the highest concentrations for all elements, except Ca content. The major elements determined were Al, Ca, Fe, Mg and Mn, while the heavy metals determined were Co, Cr, Cu, Ni, Pb and Zn

The elemental concentrations from Kota Belud have a hierarchical order of major elements Ca>Fe>Mg>Al>Mn, and heavy metals Ni>Cr>Zn>Cu>Co>Pb. The highest concentration is Ca (61701±125338 mg/kg), followed by Fe (15995±6082 mg/kg), Mg (15334±5228 mg/kg), Al (6324±2036 mg/kg), Mn (304±127 mg/kg), Ni (152.3±79 mg/kg), Cr (77.9±38.2 mg/kg), Zn (37.5±10.7 mg/kg), Cu (17.2±10.7 mg/kg), Co (12.3±6.1 mg/kg) and Pb (6.9±2.1 mg/kg). All elements are within their respective average background values, except for Ca in Station 4, and Mg, Ni and Cr in several samples. The Ni and Cr contents have surpassed the standard limits of SQGs (in Table 2).

The elemental concentrations from Kudat have a hierarchical order of major elements Ca>Mg>Fe>Al>Mn, and heavy metals Cr>Ni>Zn>Cu>Pb>Co. The highest concentration is Ca (167838±69521mg/kg), followed by Mg (8371±2,924 mg/kg), Fe (2304±657 mg/kg), Al (1554±457 mg/kg), Mn (53±12 mg/kg), Cr (8.46±2.98 mg/kg), Ni (4.15±2.11 mg/kg), Zn (6.46±1.37 mg/kg), Cu (1.70±1.51 mg/kg), Pb (1.46±0.80 mg/kg) and Co (0.23±0.11 mg/kg). All elements are below their background values, except for Ca in all stations. As compared to the SQGs, the current study demonstrates all heavy metals are within the permissible range as shown in Table 2.

The elemental concentrations from Mantanani Island have a hierarchical order of major elements Ca>Mg>Fe>Al>Mn, and heavy metals Zn>Pb>Cr>Ni. The highest concentration is Ca (417577±12757 mg/kg), followed by Mg (6475±920 mg/kg), Fe (297±245 mg/kg), Al (200±209 mg/kg), Mn (18.54±5.79 mg/kg), Zn (20.09±14.36 mg/kg), Pb (8.32±10.54mg/kg), Cr (5.81±4.97 mg/kg) and Ni (2.35±2.75 mg/kg). All elements are below their background values, except for Ca in all stations. The current study demonstrates all heavy metals are within the permissible range as indicated in SQGs as shown in the following Table 2.

Natural and human-generated pressures affect the abundance and speciation of metal contaminants along the shoreface sediments. The elemental concentrations are evaluated from the fine-grained fractions of <63 μm by comparing to their respective average shale values (ASV) and sediment quality guidelines (SQGs) to monitor the ecotoxicity and pollution level in the marine sediments. The geochemical distribution and mobility of heavy metals are facilitated by the precipitation, ionic exchange and assimilation mechanisms by the inclusion of organic matter, clays, corals and shell

Table 2. Mean concentration of major elements and heavy metals in each core sample based on the ICP-OES analysis.

Study area	Station	Sample	Major elements (mg/kg)					Heavy metals (mg/kg)					
			Al	Ca	Fe	Mg	Mn	Co	Cr	Cu	Ni	Pb	Zn
Kota Belud	S1	TP1-1	6950	4009	19321	16729	375	13.94	95.49	9.89	185.5	8.17	43.52
		TP1-2	8524	3930	21040	19773	541	17.23	98.27	11.42	225.3	7.62	43.35
	S2	TP2-1	5803	2524	15248	15726	259	13.02	86.44	10.77	178.3	6.17	36.11
		TP2-2	6923	4710	18639	20287	376	15.56	114.5	10.78	208.3	8.08	40.33
		TP2-3	6242	3260	17277	18055	371	14.45	106.4	9.74	189.6	7.64	39.15
		TP2-4	6394	2768	17945	18270	318	15.21	94.18	10.81	197	7.35	40.57
	S3	TP3-1	7421	3100	19785	17958	246	17.34	96.14	15.20	200.6	8.09	51.38
		TP3-2	9043	6192	20847	14639	320	18.03	78.97	31.44	168.4	9.56	51.31
	S4	TP4-1	2563	353794	4476	6261	137	1.84	8.99	18.73	11.73	2.49	19.37
		TP4-2	3375	232720	5373	5641	103	1.64	8.17	40.70	9.11	4.34	22.11
Range			2563–9043	2524–353794	4476–21040	5641–20287	103–541	1.64–18.03	8.17–114.53	9.74–40.70	9.11–225.32	2.49–9.56	19.37–51.38
Average±SD			6324±2036	61701±125338	15995±6082	15334±5228	304±127	12.3±6.1	77.9±38.2	17.2±10.7	152.3±79	6.9±2.1	37.5±10.7
Kudat	K1	S1	1074	73793	1777	4546	38	bdl	4.95	5.18	2.12	2.56	6.81
		S2	1178	119690	1949	6429	48	bdl	5.90	1.48	2.66	1.26	6.80
		S3	870	76113	1574	4200	31	bdl	5.63	0.72	2.01	2.85	6.28
		S4	1267	106904	1806	5228	40	bdl	14.07	2.43	7.24	1.91	6.63
		S5	1615	160699	2663	7702	54	0.08	10.73	0.97	5.65	1.51	9.29
		S6	1449	113408	2700	5576	47	0.16	12.89	1.27	8.16	2.11	7.09
	K2	S7	1720	189569	1755	10810	59	0.42	7.99	4.61	3.65	0.71	7.66
		S8	1776	182458	2335	9930	60	0.19	8.57	0.66	4.21	1.30	5.19
		S9	2034	216336	3030	10707	68	0.29	8.12	1.27	3.89	0.40	5.86
		S10	2061	255997	3059	11263	69	0.12	7.46	0.64	3.63	0.28	7.29
		S11	1981	184967	3098	9830	67	0.29	9.73	1.07	5.15	1.53	5.80
		S12	2231	192805	2997	9407	63	0.27	9.76	0.41	4.92	1.12	5.80
	K3	S13	946	309152	1211	13192	45	bdl	4.11	1.40	0.65	bdl	3.50
Range			870-2231	73793–309152	1211–3098	4200–13192	31–69	0.08–0.42	4.11–14.07	0.41–5.18	0.65–8.16	0.28–2.85	3.50–9.29
Average±SD			1554±457	167838±69521	2304±657	8371±2924	53±12	0.23±0.11	8.46±2.98	1.70±1.51	4.15±2.11	1.46±0.80	6.46±1.37
Mantanani Island	M1	M1.	133	432325	236	6057	18.45	bdl	4.95	bdl	0.29	2.05	38.87
		M2.	95	417987	194	6729	17.06	bdl	5.90	bdl	0.13	2.55	7.98
	M2	M3.	104	405959	143	5598	12.54	bdl	5.63	bdl	1.31	8.95	9.28
		M4.	528	405106	674	7715	26.43	bdl	14.07	bdl	5.98	24.64	15.35
Range			92–528	405106–432325	143–674	5598–7715	12.5–26.43	–	3.40-13.69	–	0.13–5.98	2.05–24.64	7.98–38.87
Average±SD			200±209	417577±12757	297±245	6475±920	18.54±5.79	–	5.81±4.97	–	2.35±2.75	8.32±10.54	20.09±14.36
Background values (ASVs)			80000	22100	47200	15000	850	19	90	45	68	20	95
Sediment quality guidelines (SQGs)			USEPA (1977)		Non-Polluted			–	<25	<25	<20	<40	<90
					Moderately Polluted			–	25–75	25–50	20–50	40–60	90–200
					Highly Polluted			–	>75	>50	>50	>60	>200
					ISQG-Low			–	80	65	21	50	200
					ISQG-High			–	370	270	52	220	410

bdl: Below detection limit

fragments in the sediments (Dou Y et al., 2013). Besides clay and organic matter, major elements like Al, Ca, Fe, Mg and Mn also provide complexion sites for the adsorption and passivation of heavy metals (Elder JF, 1989; Zhang C et al., 2014). Major elements thus serve as natural repository that constitute a significant sink for other heavy metals in the marine sediments.

Heavy metals Co, Cr, Cu, Ni, Pb and Zn are also largely accumulated on aluminosilicate host minerals and major elements determined by the hydrodynamic conditions in the study sites. The elements are primarily derived from the oxidation-reduction process of weathered source rocks or terrigenous materials, in the form of metal oxides, hydroxides, oxyhydroxides and carbonates during the sediment load transport to the coastal regions (Graf DI, 1960; Bayon G et al., 2004; Ugwu IM and Igbokwe OA, 2019). Thus, the heavy metals such as Co, Cr, Ni, Pb and Zn are probably associated to the amorphous Fe-Mn sorbent phases, as shown in Station 1 to 3 from Kota Belud. The coastal sediments approaching the eastward of Kota Belud (Station 1 to 3) are composed mainly of detrital sediments from lithogenic origins, whereas the westward of Kota Belud (Station 4) are enriched by calcareous biogenic origins based on the major elements determined from the ICP-OES analysis. Table 2 showed the abundance of Al, Fe and Mn elements in Station 1 to 3 in Kota Belud. In contrast, the table showed the abundance of Ca in Station 4 in Kota Belud, as well as Kudat and Mantanani Island, which all similarly showed the biogenic calcareous sediment.

The high fluctuation of Ca content in the sediments indicate the natural enrichment of Ca from marine ecosystem which comprised mainly of corals, benthic foraminifera and shelled marine biota (Karageorgis AP et al., 2020; Yi LS et al., 2021). The Ca content contributed from the autochthonous biogenic groups had exceeded the background ASVs which form stable or insoluble complexes with the heavy metals, thus inhibiting the metals from remobilised back into the seawater column as secondary pollution (Ouhadi VR et al., 2010). A decreasing heavy metal trends were observed from sampling stations with higher Ca as shown in Table 2. All metals from Kudat and Mantanani Island show similar patterns, which are within the threshold values of SQGs, unlike Kota Belud.

Station 1 to 3 located in the alluvial region near the Wariu Formation in Kota Belud are influenced by the serpentinized basement rocks (Tashakor M *et al.*, 2017). The terrigenous materials, plant remains and land biota which also release heavy metals are transported via runoffs, leachate, channels or groundwater from the lithogenic crust, weathered parent materials or biogenic origin to the coastal environment (Martins MVA et al., 2018; Ling SY et al., 2022). The present study shows that the total heavy metal content in Kota Belud is higher than Kudat and Mantanani Island, in which the geochemical anomalies of Ni and Cr had exceeded the permissible range of SQGs (Table 2) and require extensive long-term monitoring due to their high mobility and toxicity in the marine sediment. The elevated metal concentrations in Kota Belud were also released from the ongoing paddy agriculture and industrial activities based on geological survey of the study area which eventually deposit and accumulate along the coastal region.

3.3. Sediment quality assessment

Table 3 and Table 4 summarize the sediment pollution indices that include I_{geo}, *EF*, *CF*, *MCD* and *PLI* based on the heavy metal contents. The present I_{geo} study shows Class 0 with $I_{geo}<0$ for all metals in the study areas, indicating the marine sediments are uncontaminated, except for Ni in Kota Belud. The Ni in Stations 1 to 3 in Kota Belud is within $0<I_{geo}<2$ which indicated uncontaminated to moderately contaminated sediment which falls within Class 1 and Class 2. The *EF* study of Kota Belud shows the average $1\leqslant EF<3$ (minor enrichment) for all heavy metals, except Ni with $5\leqslant EF<10$ (moderately severe enrichment). Kudat also shows $1\leqslant EF<3$ for all heavy metals, except Co with $EF<1$ (no enrichment). The present study of Mantanani Island shows all heavy metals has $EF<1$, except Pb with $1\leqslant EF<3.1$, except Pb with $1\leqslant EF<3$.

The *CF* study from Kota Belud shows average $CF<1$ (low pollution) for all heavy metals, except Ni showing $1\leqslant CF<3$ (moderate pollution). The moderate pollution factors are mostly obtained from Station 1 to 3 for Cr and Ni. On the other hand, the present study from Kudat and Mantanani Island both show average $CF<1$ for all heavy metals. Overall, the *MCD* and *PLI* of the marine sediments from Kota Belud, Kudat and Mantanani Island shows $MCD<1.5$ and $0\leqslant PLI<1$, indicating only low pollution level in the sediments due to low degree of metal pollution. Although all locations suggested the same classification index, Kota Belud has relatively higher *MCD* and *PLI* than Kudat and Mantanani Island.

The sediment quality assessment includes the single indices like I_{geo}, *EF* and *CF*, and multiple complex indices like *MCD* and *PLI* to interpret the extent of heavy metal pollution in the sediments due to lithogenic and anthropogenic factors. According to Herut B and Sandler A (2006), the *EF* values with $0.05\leqslant EF<1.5$ indicate enrichment from lithogenic origin, crustal materials, or natural processes, whereas $EF>1.5$ indicate enrichment by anthropogenic factors.

The interpretation of the average single indices for I_{geo} and *CF* from Kota Belud shows the hierarchical order of the metal pollution Ni>Cr>Co>Zn>Cu>Pb, in which Station 1 to 3 are moderately contaminated by Ni and slightly contaminated by Cr. All heavy metals from Station 4 show nil to low pollution factor that fall under Class 0 and are not hazardous to humans or marine ecology (Hossain S et al., 2019). The interpretation for Kudat and Mantanani Island show the ranking order of Cr>Zn>Pb>Ni>Cu>Co and Pb>Zn>Cr>Ni, respectively. The average I_{geo} and *CF* also show all heavy metals from Kudat and Mantanani Island are within Class 0, which signifies no pollution and are safe from

Table 3. Average I_{geo} and *EF* values of the heavy metals in marine sediment cores.

Study area	Station	Core ID	Geoaccumulation index (I_{geo})						Enrichment factor (*EF*)					
			Co	Cr	Cu	Ni	Pb	Zn	Co	Cr	Cu	Ni	Pb	Zn
Kota Belud	S1	TP1-1	−1.03	−0.50	−2.77	0.86	−1.88	−1.71	1.79	2.59	0.54	6.66	1.00	1.12
		TP1-2	−0.73	−0.46	−2.56	1.14	−1.98	−1.72	2.03	2.45	0.57	7.43	0.85	1.02
	S2	TP2-1	−1.13	−0.64	−2.65	0.81	−2.28	−1.98	2.12	2.97	0.74	8.11	0.95	1.18
		TP2-2	−0.87	−0.24	−2.65	1.03	−1.89	−1.82	2.07	3.22	0.61	7.76	1.02	1.08
		TP2-3	−0.98	−0.34	−2.79	0.89	−1.97	−1.86	2.08	3.23	0.59	7.62	1.04	1.13
		TP2-4	−0.91	−0.52	−2.64	0.95	−2.03	−1.81	2.11	2.75	0.63	7.62	0.97	1.12
	S3	TP3-1	−0.72	−0.49	−2.15	0.98	−1.89	−1.47	2.18	2.55	0.81	7.04	0.96	1.29
		TP3-2	−0.66	−0.77	−1.10	0.72	−1.65	−1.47	2.15	1.99	1.58	5.61	1.08	1.22
	S4	TP4-1	−3.95	−3.91	−1.85	−3.12	−3.59	−2.88	1.02	1.05	4.39	1.82	1.31	2.15
		TP4-2	−4.12	−4.05	−0.73	−3.48	−2.79	−2.69	0.76	0.80	7.95	1.18	1.91	2.04
	Mean		−1.51	−1.19	−2.19	0.08	−2.20	−1.94	1.83	2.36	1.84	6.08	1.11	1.34
Kudat	K1	S1	–	−4.77	−3.70	−5.59	−3.55	−4.39	–	1.46	3.06	0.83	3.40	1.90
		S2	–	−4.52	−5.51	−5.26	−4.57	−4.39	–	1.59	0.80	0.95	1.53	1.73
		S3	–	−4.58	−6.55	−5.67	−3.40	−4.50	–	1.88	0.48	0.89	4.27	1.98
		S4	–	−3.26	−4.80	−3.82	−3.97	−4.43	–	4.09	1.41	2.78	2.50	1.82
		S5	−8.48	−3.65	−6.12	−4.17	−4.31	−3.94	0.07	2.11	0.38	1.47	1.34	1.73
		S6	−7.48	−3.39	−5.73	−3.64	−3.83	−4.33	0.15	2.50	0.49	2.10	1.84	1.30
	K2	S7	−6.08	−4.08	−3.87	−4.80	−5.40	−4.22	0.59	2.39	2.76	1.44	0.95	2.17
		S8	−7.23	−3.98	−6.68	−4.60	−4.53	−4.78	0.20	1.92	0.30	1.25	1.31	1.10
		S9	−6.62	−4.06	−5.73	−4.71	−6.23	−4.60	0.24	1.41	0.44	0.89	0.31	0.96
		S10	−7.89	−4.18	−6.72	−4.81	−6.74	−4.29	0.10	1.28	0.22	0.82	0.22	1.18
		S11	−6.62	−3.79	−5.98	−4.31	−4.29	−4.62	0.23	1.65	0.36	1.15	1.17	0.93
		S12	−6.72	−3.79	−7.36	−4.37	−4.74	−4.62	0.22	1.71	0.14	1.14	0.88	0.96
	K3	S13	–	−5.04	−5.59	−7.29	–	−5.35	–	1.78	1.21	0.37	–	1.44
	Mean		−7.14	−4.08	−5.72	−4.85	−4.63	−4.50	0.23	1.98	0.93	1.24	1.64	1.48
Mantanani Island	M1	M1.	–	−4.77	–	−8.46	−3.87	−1.87	–	0.14	–	0.01	0.25	1.01
		M2.	–	−4.52	–	−9.62	−3.56	−4.16	–	0.15	–	0.00	0.28	0.19
	M2	M3.	–	−4.58	–	−6.28	−1.75	−3.94	–	0.17	–	0.05	1.20	0.26
		M4.	–	−3.26	–	−4.09	−0.28	−3.21	–	0.30	–	0.17	2.40	0.31
	Mean		–	−4.28	–	−7.11	−2.36	−3.30	–	0.19	–	0.06	1.03	0.44

metal pollution. The study shows the current heavy metals do not jeopardize human health or the marine environment. However, the present study serves as baseline data where the heavy metals may increase and has the possibility to pose risks of metal pollution in the future.

The overall *EF* values show heavy metals from Station 1 to 3 in Kota Belud are enriched by both natural and anthropogenic activities, whereas heavy metals in Station 4 are only enriched by natural sources. Geological processes such as pedogenesis of the parent rocks are significant here, whereby the disintegration of the sandstone or shale from Crocker Formation led to higher Cu, Pb and Zn content in Station 4, whereas disintegration of ultrabasic basement rocks lead to higher Cr and Ni content in Station 1 to 3 (Tashakor M et al., 2017). Station 1 to 3 also has higher enrichment than Station 3 due to the river transport of the sediment loads and heavy metals from the weathered source rocks or human activities to the coastal regions (Martins MVA et al., 2018). The tropical climates induce intense transport and discharge of the metal pollutants which also facilitate the enrichment of metals washed off from the land into the coastal sinks or water system (Pit IR et al., 2017).

The *EF* values for Kudat show minor enrichment of Pb, Zn, Cr and Ni by anthropogenic factors in Station 1 and 2, whereas Station 3 is only influenced by the lithogenic origins. The weathered parent materials are the products of sandstone and shale that formed the Kudat Formation (Gloaguen TV and Passe JJ, 2017; Yi LS et al., 2021), as well as cherts and ultrabasic rocks that formed the ophiolite complex of the study area also contribute to metal enrichment in the sediments (Kierczak J et al., 2021). The anthropogenic factors which contribute to metal enrichment in Kota Belud, Kudat and Mantanani Island include the disposal effluents from domestic waste, industry, agriculture and direct discharge of oil spill incidents to the marine environment (Adam AA et al., 2019; Pavoni E et al., 2021). Table 3 shows Kota Belud has relatively the highest enrichment factors due to the high discharge of pesticides and fertilizers from the paddy agriculture that release heavy metals which are carried along the channels or runoffs to the ocean basin (Tahir SH and Mustapa AT, 2020).

The multiple complex indices of *MCD* and *PLI* show that the marine sediments along the coasts had only mild pollution due to very low degree metal pollution. This suggests that the current status of heavy metals does not deteriorate the sediment quality or cause ecotoxicity to the marine ecosystem

Table 4. Average *CF*, *MCD* and *PLI* values of the heavy metals in marine sediment cores.

Study area	Station	Sample	Pollution factor (*CF*)						Σ*CF*	*MCD*	*PLI*
			Co	Cr	Cu	Ni	Pb	Zn			
Kota Belud	S1	TP1-1	0.73	1.06	0.22	2.73	0.41	0.46	5.61	0.93	0.67
		TP1-2	0.91	1.09	0.25	3.31	0.38	0.46	6.40	1.07	0.72
	S2	TP2-1	0.69	0.96	0.24	2.62	0.31	0.38	5.19	0.87	0.60
		TP2-2	0.82	1.27	0.24	3.06	0.40	0.42	6.22	1.04	0.71
		TP2-3	0.76	1.18	0.22	2.79	0.38	0.41	5.74	0.96	0.66
		TP2-4	0.80	1.05	0.24	2.90	0.37	0.43	5.78	0.96	0.67
	S3	TP3-1	0.91	1.07	0.34	2.95	0.40	0.54	6.21	1.04	0.77
		TP3-2	0.95	0.88	0.70	2.48	0.48	0.54	6.02	1.00	0.85
	S4	TP4-1	0.10	0.10	0.42	0.17	0.12	0.20	1.11	0.19	0.16
		TP4-2	0.09	0.09	0.90	0.13	0.22	0.23	1.67	0.28	0.19
	Mean		0.68	0.88	0.38	2.31	0.35	0.41	5.00	0.83	0.60
Kudat	K1	S1	–	0.06	0.12	0.03	0.13	0.07	0.40	0.08	0.07
		S2	–	0.07	0.03	0.04	0.06	0.07	0.27	0.05	0.05
		S3	–	0.06	0.02	0.03	0.14	0.07	0.32	0.06	0.05
		S4	–	0.16	0.05	0.11	0.10	0.07	0.48	0.10	0.09
		S5	0.00	0.12	0.02	0.08	0.08	0.10	0.40	0.07	0.04
		S6	0.01	0.14	0.03	0.12	0.11	0.07	0.48	0.08	0.06
	K2	S7	0.02	0.09	0.10	0.05	0.04	0.08	0.38	0.06	0.06
		S8	0.01	0.10	0.01	0.06	0.07	0.05	0.30	0.05	0.04
		S9	0.02	0.09	0.03	0.06	0.02	0.06	0.27	0.05	0.04
		S10	0.01	0.08	0.01	0.05	0.01	0.08	0.25	0.04	0.03
		S11	0.02	0.11	0.02	0.08	0.08	0.06	0.36	0.06	0.05
		S12	0.01	0.11	0.01	0.07	0.06	0.06	0.32	0.05	0.04
	K3	S13	–	0.05	0.03	0.01	–	0.04	0.12	0.03	0.03
	Mean		0.01	0.09	0.04	0.06	0.07	0.07	0.34	0.06	0.05
Mantanani Island	M1	M1.	–	0.06	–	0.00	0.10	0.41	0.57	0.14	0.06
		M2.	–	0.07	–	0.00	0.13	0.08	0.28	0.07	0.03
	M2	M3.	–	0.06	–	0.02	0.45	0.10	0.63	0.16	0.09
		M4.	–	0.16	–	0.09	1.23	0.16	1.64	0.41	0.23
	Mean		–	0.08	–	0.03	0.48	0.19	0.78	0.19	0.10

(Tomlinson DL et al., 1980). Nevertheless, the *MCD* and *PLI* values of Kota Belud are higher than Kudat and Mantanani Island, thus a long-term supervision of the study area is required to assess the pollution level in the marine sediments along the coasts.

3.4. Pearson Correlation Matrix

The major elements and heavy metals are selected as the variables for this study (Table 5) using the IBM SPSS Statistics v.28 software to determine the association of the elements and interpret their potential sources at the significance level of $p<0.01$ and $p<0.05$ for two-tailed test. The marine sediment from Kota Belud (N=53) shows Al and Fe are strongly positive correlated ($0.7\leqslant r<1$) to Mg, Mn, Co, Cr, Ni, Pb and Zn. Ca shows only strong negative correlation ($-0.7\leqslant r<-1$) with other variables, whereas Mg and Mn are strongly positive correlated to Co, Cr, Ni, Pb and Zn, except Cu. Cu is the only heavy metal that shows strong negative correlation with Mg, Cr and Ni. Other heavy metals like Co, Cr, Ni, Pb and Zn are strongly positive correlated to one another.

Marine sediment from Kudat (N=76) shows Al has strong positive correlation with Fe and Mn, but strong negative correlation with Pb. Ca is positively correlated to Mg and Mn, whereas Fe and Mg are positively correlated to Mn. Among the heavy metals, only Pb shows strong negative correlation with Al, Ca, Mg and Mn. Cr is also strongly positive correlated to Ni. The other variables do not show any significance with the elements. Marine sediment from Mantanani Island (N=24) shows Al has a strong positive correlation with Fe, Cr, Ni and Pb, whereas Ca has negative moderate correlation ($-0.4\leqslant r<-0.7$) with Al, Fe, Ni and Pb. Fe is strongly positive correlated to Cr, Ni and Pb, whereas Mg shows strong negative correlation with Zn. Mn only a has moderate positive correlation with other variables like Al, Fe, Mg, Cr, Ni and Pb. Heavy metals like Cr, Ni and Pb are strongly positive correlated to one another.

The Pearson correlation analysis shows the association between the major elements and heavy metals that reflect their potential sources, where positive signs indicate the variables are directly related while negative signs indicate the variables are inversely related to one another (Siddiqui AS and Saher NU, 2021). A strong correlation between the heavy metals suggest that they originate from a common natural or anthropogenic source, whereas a weak or no significant correlation indicate they originate from different pollution

Table 5. Pearson’s correlation matrix of elemental concentrations in the marine sediment cores.

Kota Belud (N=53)											
Element	Al	Ca	Fe	Mg	Mn	Co	Cr	Cu	Ni	Pb	Zn
Al	1										
Ca	−0.864**	1									
Fe	0.962**	−0.940**	1								
Mg	0.805**	−0.915**	0.919**	1							
Mn	0.786**	−0.731*	0.818**	0.839**	1						
Co	0.950**	−0.937**	0.988**	0.922**	0.775**	1					
Cr	0.799**	−0.941**	0.922**	0.986**	0.800**	0.922**	1				
Cu	−0.287	0.507	−0.502	−0.728*	−0.597	−0.506	−0.712*	1			
Ni	0.863**	−0.953**	0.958**	0.987**	0.834**	0.960**	0.980**	−0.683*	1		
Pb	0.943**	−0.913**	0.951**	0.813**	0.695*	0.934**	0.847**	−0.279	0.857**	1	
Zn	0.948**	−0.876**	0.955**	0.800**	0.636*	0.955**	0.815**	−0.328	0.860**	0.950**	1
Kudat (N=76)											
Element	Al	Ca	Fe	Mg	Mn	Co	Cr	Cu	Ni	Pb	Zn
Al	1										
Ca	0.424	1									
Fe	0.868**	0.195	1								
Mg	0.517	0.968**	0.224	1							
Mn	0.935**	0.626*	0.782**	0.720**	1						
Co	0.260	0.127	−0.397	0.455	0.283	1					
Cr	0.366	−0.225	0.448	−0.227	0.163	−0.367	1				
Cu	−0.311	−0.313	−0.474	−0.221	−0.308	0.697	−0.181	1			
Ni	0.381	−0.276	0.518	−0.273	0.181	−0.389	0.980**	−0.174	1		
Pb	−0.796**	−0.921**	−0.557	−.904**	−0.877**	−0.287	−0.010	0.188	0.013	1	
Zn	0.134	−0.431	0.256	−0.405	0.021	−0.361	0.377	0.220	0.425	−0.018	1
Mantanani Island (N=24)											
Element	Al	Ca	Fe	Mg	Mn	Co	Cr	Cu	Ni	Pb	Zn
Al	1										
Ca	−0.440*	1									
Fe	0.985**	−0.444*	1								
Mg	0.171	−0.068	0.150	1							
Mn	0.666**	−0.204	0.668**	0.623**	1						
Co	–	–	–	–	–	–					
Cr	0.990**	−0.498*	0.973**	0.143	0.610**	–	1				
Cu	–	–	–	–	–	–	–	–			
Ni	0.989**	−0.565*	0.977**	0.005	0.570*	–	0.999**	–	1		
Pb	0.916**	−0.437*	0.867**	0.108	0.477*	–	0.931**	–	0.918**	1	
Zn	−0.061	0.174	−0.008	−0.742**	−0.146	–	−0.080	–	0.014	−0.104	1

*Correlation is significant at the 0.05 level; **Correlation is significant at the 0.01 level; N=No. of samples

sources (Tao W et al., 2021; Dey M et al., 2021). Major elements are also included into the variables to interpret the affinity to form metal complexes which also influence the leachability, mobility and abundance of the heavy metals in marine sediment.

Based on the results, the strong association of major elements Al, Fe and Mn in Kota Belud, Kudat and Mantanani Island suggest they are released from similar source rocks, such as the clastic sedimentary rocks or ophiolitic rocks from the redox reaction during weathering process (Dalai TK et al., 2004). Major elements also constitute the primary sink to transport heavy metal burden from the source via direct bonding, ion exchange mechanisms and formation of metal complexion on the sediment surface (Churchman GJ, 2006). Besides the Al-Fe-Mn association, the strong Pb-Zn correlation in Kota Belud also suggest they originate from the clastic sedimentary rocks that made up the Crocker Formation, including sandstone, shale and calcareous siltstone, as well as Chert-Spilite Formation like cherts rich in quartz arenites (Clement JF and Keij J, 1958; Li et al., 2010; Gloaguen TV and Passe JJ, 2017; Ling SY et al., 2022). On the other hand, the negative correlation between Ca with other variables show that Ca tends to form stable metal compounds which are trapped, retained and accumulated in surficial sediments along the coasts (Madrid L and Diaz BE, 1992; Shahbazi K and Beheshti M, 2019).

The negative correlation suggest that Ca has a high buffering capacity that prevents the metals from remobilized and re-entering into the marine water as secondary pollutants (Korfali SI and Davies BE, 2004). There is no association between Ca with other major elements in Kota Belud and Mantanani Island, thus suggesting Ca originated from a different source, such as coral debris or shelled organisms from the marine biota (Yi LS et al., 2021). However, the strong positive Ca-Mg association from Kudat indicated that they are also released from weathered limestone in Kudat Formation (Suggate SM and Hall R, 2014). The natural emission of Mg, Fe and Mn from the hot plumes of oceanic hydrothermal vents also influence the metal anomalies which either circulate in the ocean water or eventually accumulate in bottom marine sediment (Lough AJ et al., 2019; White WM, 2020). The present study shows the major elements and heavy metals from Kudat and Mantanani Island have fewer association as compared to Kota Belud, which further suggested they are more likely to originate from the enrichment of marine system than the terrestrial parent materials.

The strong association between Co, Cr and Ni from Kota Belud also suggest that the elements originate from the pedogenesis of ultrabasic basement rock from the ophiolite complex or Wariu Formation (Tashakor M et al., 2011, 2017; Kierczak J et al., 2021). Cr, Ni and Zn also have higher adsorption and complexation capacity than Co and Pb, thereby allowing higher leachability and mobility to be transported over further distance from the source to the natural catchment near the coast (Dai L et al., 2019). Other heavy metals that show only weak or no significant correlation suggest that they are contributed from anthropogenic inputs, such as untreated sewage sludge, wastewater and effluents from industrial, agriculture or domestic activities (Martins MVA et al., 2018; Agoro MA et al., 2020; Pavoni E et al., 2021).

The anthropogenic sources were also based on the fieldwork observation that showed the developing agricultural and industrial activities in Kota Belud, mariculture or fishery activities in Kudat and tourism or fishery activities in Mantanani Island. Besides the untreated industrial sewage or effluents, the excessive use of pesticide and fertilizers in paddy fields cause heavy metal leaching in Kota Belud that led to the higher pollution level than Kudat and Mantanani Island (Satpathy D et al., 2014; Yi LS et al., 2021; Ling SY et al., 2022). Heavy metals enriched and released by the human factors can deteriorate the sediment quality or jeopardize the marine environment when the metal content surpassed the allowable range of SQGs.

4. Conclusions

The key findings of this study are:

(i) The abundance, distribution and mobility of heavy metals are controlled by the physico-chemical measurements such as sediment pH, organic matter, moisture content and sandy texture that fluctuates within the marine sediments in Kota Belud, Kudat and Mantanani Island, respectively.

(ii) Heavy metal content in marine sediments along the northwest coasts of Sabah were compared to the SQGs where all heavy metals are within the permissible range, except for Ni and Cr metals. The pollution level of heavy metals in marine sediment is evaluated using the SQA, including the I_{geo}, EF, CF, MCD and PLI parameters in Kota Belud, Kudat and Mantanani Island.

(iii) The present study shows Class 0, Class 1 and Class 2 pollution level in Kota Belud, in which the heavy metals are minor to moderately severe enriched by anthropogenic sources. The study also shows Class 0 pollution level in Kudat and Mantanani Island, and the metals are not or only minor enriched by anthropogenic sources. The overall $MCD<1.5$ and $0\leqslant PLI<1$ indicating only low pollution level in the marine sediments from all study areas.

(iv) Although the current status of the sediment quality is safe from pollution, the higher heavy metal content of Ni and Cr suggest that a continual observation and long-term monitoring are significant for the study areas in the northwest coast of Sabah, Malaysia.

CRediT authorship contribution

Baba Musta conceived of the presented idea. All authors participated in the fieldwork and sampling activities. Ling Sin Yi conducted the laboratory analysis and statistical analysis. All authors discussed the results and contributed to the final manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

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