

THE QUALITY ASSESSMENT OF HEAVY METALS IN MARINE SEDIMENTS FROM USUKAN COASTAL BEACH, KOTA BELUD, SABAH.

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ABSTRACT. A total of fifty-three (53) sediment samples were collected from Usukan coastal beach to study the potential of pollution due to heavy metals in the marine ecosystem. The sediment samples were collected along the coastal beach using a core sampler. The ICP-OES analysis was used to identify the concentration of heavy metals in the marine sediment samples. The results of pH analysis showed the increase of pH from 5.69 to 8.48 from inland into the sea. The lowest moisture content was 4.99%, whereas the highest was 48.75%. The organic matter ranges from 0.30 to 6.73%. The sediment texture varies from sandy, sandy loam, and sandy clay loam texture. The decreasing ranking order of heavy metals concentration is Fe (4476-29829 ppm) followed by Al (5803-8524 ppm) and Mn (103-504 ppm), which are still within the background values and standard limits. The assessment of Fe, Al and Mn contamination in sediment samples was performed by comparing with the allowable range of average background values and the standard limits from Sediment Quality Guideline (SQG) in marine sediment. In conclusion, the results of quality assessment using the geoaccumulation index (Igeo), contamination factor (CF), modified degree of contamination (mCd), and pollution load index (PLI) showed that the sediment from Usukan beach has a very low contamination level that causes only mild pollution.

KEYWORDS. Geochemistry, Heavy Metal, Sediment Quality, Marine Environment

INTRODUCTION

The assessment of heavy metals quality in marine sediment is greatly affected by active chemical weathering or pedogenesis of the geological source rocks (Le'Pera *et al.*, 2001; Li *et al.*, 2019). Heavy metals found naturally in the Earth's crust and the rock-forming minerals are transported via rivers, surface run-offs, or any drainage system from the parent materials situated in areas with higher elevation and accumulate in the ocean basin. Besides topographical conditions, the tropical climate of the study area that experiences hot and wet seasons also lead to the transport and redistribution of heavy metals in marine sediments (Seaward and Richardson, 1989; Han *et al.*, 2001). When the heavy metal content exceeds the permissible level, the metals will increase the toxicity levels and contaminate the sediment (Zhang *et al.*, 2015; Jayamurali *et al.*, 2021). Heavy metals such as aluminium (Al), iron (Fe) and manganese (Mn) are released from lithogenic and anthropogenic sources that accumulate in the marine sediments, which also serve as a dynamic natural sink for the pollutants (Chuan and Yunus, 2019). Thus, the coastal regions are selected to assess the sediment quality due to metal contamination.

Heavy metal pollution is a global concern as the contaminants are toxic and non-biodegradable, but instead accumulate in sediments which are then released into the seawater column and enter the food chains of marine organisms (Yang *et al.*, 2021; Xiang *et al.*, 2021). The significant contribution of contaminants is from the terrestrial origin, such as natural processes and anthropogenic pollutant inputs via river discharge, land run-off, rapid industrialization, fishing or agricultural activities, and oil spills (Gopal *et al.*, 2017; Tchounwou *et al.*, 2012). All processes will lead to environmental or marine pollution and cause an imbalance in the global ecosystem. The concentrations of naturally occurring heavy metals vary according to the geological background (Vallius *et al.*, 2007; Wuana and Okeimen, 2011). Thus this has to be considered when assessing the degree of contamination in the study area. Therefore, Sediment Quality Guideline (SQG) is used as an appraisal to compare the concentrations of

these elements to determine the quality and the eco-toxic level in marine sediments (Abolfazl and Ahmad, 2012; Birch, 2018). The profile and distribution of elements in sediments provide a record of depositional history, pollution origins, and the migration of the heavy metals to various parts of the marine sediments. The main objective of this research is to assess the distribution of selected heavy metal contamination in Usukan coastal beach by comparing with the permissible range of average background values and the standard limits established in the Sediment Quality Guideline (SQG).

DESCRIPTION OF STUDY AREA

The study area is located in Kota Belud, bounded by a latitude $6^{\circ} 17' 40''$ N to $6^{\circ} 36' 50''$ N and longitude $116^{\circ} 16' 50''$ E to $116^{\circ} 33' 0''$ E (Diagram 1). Kota Belud is formed from three major formations namely the Crocker Formation, which aged from Eocene to Early Miocene, Wariu Formation, which aged from Middle Miocene, and Quaternary Alluvium (Sanudin and Baba, 2007). The primary rock units underlying Kota Belud are interbedded sandstone and shale from Crocker Formation, and a mixture of fragmented rocks from different origins of Wariu Formation or melange. The Wariu Formation consists of argillite, breccia, metaclastics, micritic limestone, chert, spilite and ultramafic rocks from Wariu Formation (Collenette, 1958; Clement and Keij, 1958; Tongkul, 2006; Junaidi and Basir, 2012; Hall and Breitfeld, 2017). The Quaternary alluvium is originated from the weathered rocks from Crocker and Wariu Formation. The coral deposit aged from Pleistocene to Quaternary mainly occurred along the beach shoreline and nearby islands.

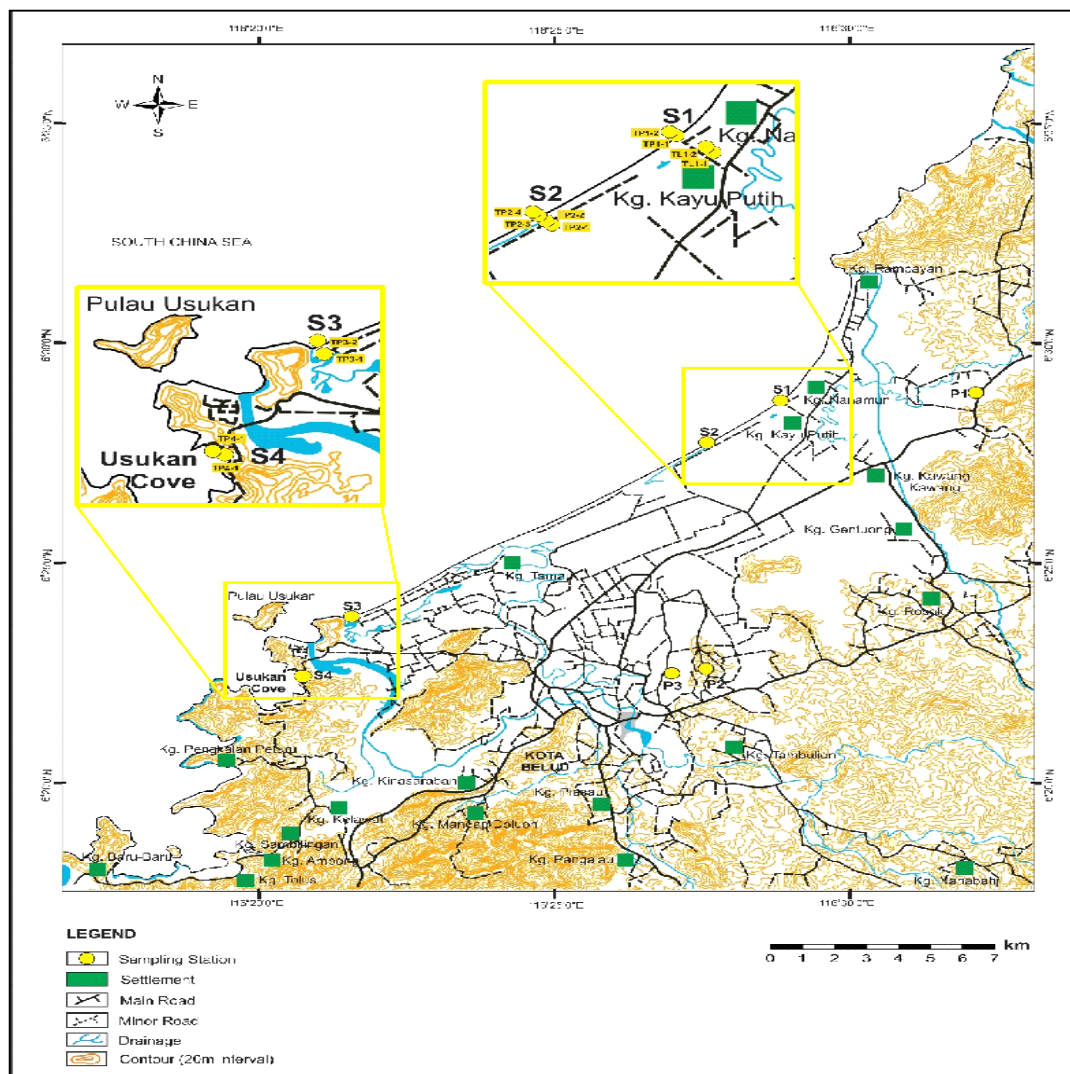


Diagram 1: Base map of the study area and sampling stations in Kota Belud, Sabah. (Source: Minerals and Geoscience Department of Malaysia, 2010)

MATERIALS AND METHODOLOGY

For this study, a total of fifty-three (53) sediment samples were collected from Usukan coastal beach. The sediments were collected as undisturbed samples in 100cm long PVC cores using the core sampler and were tightly closed with the PVC stopper. The sediments were then extruded from the PVC cores using the horizontal extrusion method before being segmented into every 10cm thick subsampling and kept in sealed ziplock bags for further laboratory analysis. The analyses conducted were pH, moisture content (MC), soil organic matter (SOM), and particle size distribution (PSD) to determine the physico-chemical characteristics of sediments based on the BSI technique (BSI, 1990). The pH analysis was conducted immediately once the samples were extruded. The samples for moisture content were dried at 105⁰C for 8 hours using the conventional oven method, whereas the samples for SOM analysis were dried in the furnace overnight at 400⁰C based on the dry combustion method. Air-dried samples were used for the particle size distribution (PSD) analysis using the dry sieving and pipette method. The heavy metal concentrations were determined using the aqua regia digestion method via Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) analysis using USEPA Method 6010D (Rev 4) (USEPA, 2014). The contamination levels of Al, Fe, and Mn metals in the sediments were also evaluated using the Igeo, CF, mCd, and PLI statistical parameters to assess the sediment quality and were compared with the SQG and average background values (Turekian and Wedepohl, 1961).

RESULTS

Physico-Chemical Properties of Marine Sediments

Table 1 shows the pH value, moisture content, and organic matter in the sediment samples for every 10cm interval depth. The average pH readings for all sediments are within the range of 5.6-8.5. The average moisture content shows the lowest (4.9-5.8%) in the marine sediment profile (SP) as the samples are collected the furthest into land areas as compared to the core sediments (TP) (15.3-39.9%) and mud soil (TL) (35.4-48.8%) which are collected nearer to the sea. The SOM percentage shows the lowest range of 0.30-0.32% in SP, followed by 0.12-1.78% in TP and 4.66-6.73% in TL. The USDA textural classification shows SP and TP have sandy texture while TL has sandy loam and sandy clay loam texture, in which the grain size composition is shown in Diagram 2.

Table 1: Average values of pH, moisture content, and organic matter of sediments.

Samples	SP2	SP3	TP1-1	TP1-2	TP2-1	TP2-2	TP2-3	TP2-4	TP3-1	TP3-2	TP4-1	TP4-2	TL1-1	TL1-2
pH	5.7	6.5	7.1	7.9	7.3	7.8	7.9	7.5	7.3	7.7	8.4	8.5	6.1	6.4
MC %	5.0	5.8	25.0	25.8	24.0	23.8	15.3	25.3	26.2	39.9	25.9	27.9	35.4	48.8
OM %	0.3	0.3	0.3	0.3	0.2	0.2	0.2	0.1	0.3	0.8	1.6	1.8	4.7	6.7
Texture	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy	Sandy loam	Sandy clay loam

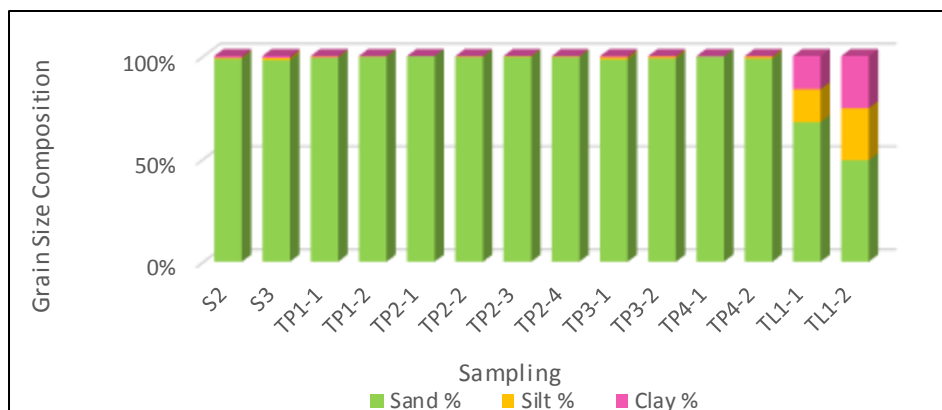


Diagram 2: Distribution of grain size composition in Usukan beach sediment.

Heavy Metal Concentration in Sediments

Table 2 shows the total concentration of heavy metals Fe, Al, and Mn in the fine-grain fraction with size $<63\mu\text{m}$. The total mean concentration and standard deviation of heavy metals determined are Al (mean $6,824\pm 2,025$ ppm), Fe (mean $17,804\pm 6,513$ ppm), and Mn (mean 300 ± 109 ppm), and their background values are based on the average shale values (ASV) (*Turekian and Wedepohl, 1961). The heavy metal contents are also compared with the standard background limits of SQG established by NOAA (1999), also known as Screening Quick Reference Tables (SQuiRTs).

Table 2: Total concentration of heavy metals in beach sediment (fine grain size $<63\mu\text{m}$).

Samples	Heavy Metal (ppm)								
	Al			Fe			Mn		
	Min.	Mean	Max.	Min.	Mean	Max.	Min.	Mean	Max.
SP2	5,101	7,607	10,162	5,101	20,051	10,162	200	325	455
SP3	2,108	6,228	9,311	2,108	16,291	9,311	76	228	321
TP1-1	6,490	6,950	7,437	6,490	17,399	7,437	365	375	397
TP1-2	7,563	8,524	9,610	7,563	22,076	9,610	462	541	679
TP2-1	4,716	5,803	6,891	13,444	16,130	18,816	217	259	301
TP2-2	4,801	6,923	8,903	14,069	18,810	22,330	286	376	455
TP2-3	4,975	6,242	7,821	14,292	17,060	20,616	291	371	464
TP2-4	4,777	6,394	7,339	13,874	17,841	20,147	224	318	371
TP3-1	7,311	7,421	7,531	19,227	19,678	20,129	245	246	246
TP3-2	8,442	9,043	9,579	18,114	20,745	23,320	277	320	375
TP4-1	1,355	2,563	4,089	2,300	4,476	8,073	87	137	204
TP4-2	1,127	3,375	4,686	1,811	5,373	7,921	29	103	187
TL1-1	8,134	9,926	13,124	25,595	29,829	36,407	266	345	458
TL1-2	4,597	8,533	12,732	15,422	23,496	31,400	126	255	337
Mean	6,824			17,804			300		
SD +/-	2,025			6,513			109		
SQG (SQuiRTs)	4.7%			1.8%			330		
*ASV	80,000			47,200			850		

Marine Sediment Quality Assessment

The geoaccumulation index (Igeo), contamination factor (CF), modified degree of contamination (mCd), and pollution load index (PLI) are the assessed parameters to determine the contamination level of heavy metals in the study area. The selected elements are Al, Fe, and Mn, which are the dominant elements from the parent materials and surrounding geological sources. Since there are no local background values for the study area, the average shale values (ASV) from the Earth's crust are selected as reference values for marine sediments (Turekian and Wedepohl, 1961). Table 3 shows the Igeo values

($I_{geo} < 0$) that suggest no contamination for all elements (Muller, 1969). The parameters $CF < 1$ and $mCd < 1.5$ both suggest none to a very low degree of contamination in the study area (Hakanson, 1980). The PLI shows $0 < PLI \leq 1$, which indicates only mild pollution due to the background levels of pollutants in the marine ecosystem (Tomlinson et al., 1980). This shows that the contaminants are more likely due to natural inputs of local geology instead of anthropogenic sources.

Table 3: Sediment quality assessment based on I_{geo} , CF, mCd and PLI parameters.

Parameter		SP2	SP3	TP1-1	TP1-2	TP2-1	TP2-2	TP2-3	TP2-4	TP3-1	TP3-2	TP4-1	TP4-2	TL1-1	TL1-2
I_{geo}	Al	-3.98	-4.27	-4.11	-3.82	-4.37	-4.12	-4.26	-4.23	-4.02	-3.73	-5.55	-5.15	-3.60	-3.81
	Fe	-1.82	-2.12	-2.02	-1.68	-2.13	-1.91	-1.91	-2.05	-1.99	-1.85	-3.98	-3.72	-1.25	-1.59
	Mn	-1.97	-2.48	-1.77	-1.24	-2.30	-1.76	-1.78	-2.00	-2.38	-2.00	-3.22	-3.63	-1.89	-2.32
CF	Al	0.10	0.08	0.09	0.11	0.07	0.09	0.08	0.08	0.09	0.11	0.03	0.04	0.12	0.11
	Fe	0.42	0.35	0.37	0.47	0.34	0.40	0.40	0.36	0.38	0.42	0.09	0.11	0.63	0.50
	Mn	0.38	0.27	0.44	0.64	0.31	0.44	0.44	0.37	0.29	0.38	0.16	0.12	0.41	0.30
ΣCF		0.90	0.69	0.90	1.21	0.72	0.93	0.91	0.82	0.76	0.91	0.29	0.28	1.16	0.90
mCd		0.30	0.23	0.30	0.40	0.24	0.31	0.30	0.27	0.25	0.30	0.10	0.09	0.39	0.30
PLI		0.25	0.19	0.24	0.32	0.20	0.25	0.24	0.22	0.22	0.26	0.08	0.08	0.32	0.25

Physico-Chemical Properties Against Sediment Quality

Table 4 shows the relationship between the physico-chemical parameters (pH, MC, SOM, PSD) and the sediment quality in terms of the sum of contamination factor, modified degree of contamination, and pollution load index. From Diagram 3, it is shown that moisture content, silt, and clay composition have a positive correlation with PLI whereas pH and sand composition show a negative correlation. OM is the only geochemical parameter that shows no correlation to the PLI. Therefore, this indicates that the physico-chemical properties of sediments regulate the heavy metal content and impact the marine sediment quality.

Table 4: Relationship between the physico-chemical properties and sediment quality

Samples	Physico-Chemical Properties						Sediment Quality		
	pH	MC %	SOM %	Sand %	Silt %	Clay %	ΣCF	mCd	PLI
SP2	5.69	4.99	0.30	98.69	0.55	0.76	0.48	0.24	0.19
SP3	6.53	5.76	0.32	98.01	1.07	0.92	0.35	0.17	0.14
TP1-1	7.09	25.01	0.25	99.14	0.46	0.40	0.53	0.26	0.20
TP1-2	7.87	25.76	0.27	99.65	0.20	0.15	0.74	0.37	0.26
TP2-1	7.34	24.03	0.16	99.10	0.05	0.05	0.38	0.19	0.15
TP2-2	7.81	23.82	0.24	99.55	0.30	0.15	0.53	0.26	0.20
TP2-3	7.88	15.32	0.20	99.70	0.25	0.05	0.51	0.26	0.18
TP2-4	7.51	25.30	0.12	99.59	0.25	0.15	0.45	0.23	0.17
TP3-1	7.25	26.16	0.32	98.37	1.02	0.61	0.38	0.19	0.16
TP3-2	7.74	39.85	0.83	98.78	0.91	0.30	0.49	0.24	0.21
TP4-1	8.44	25.94	1.63	99.75	0.10	0.15	0.19	0.10	0.07
TP4-2	8.48	27.89	1.78	98.73	0.91	0.36	0.16	0.08	0.07
TL1-1	6.08	35.43	4.66	67.80	16.10	16.10	0.53	0.26	0.22
TL1-2	6.37	48.75	6.73	49.40	25.30	25.30	0.41	0.20	0.18

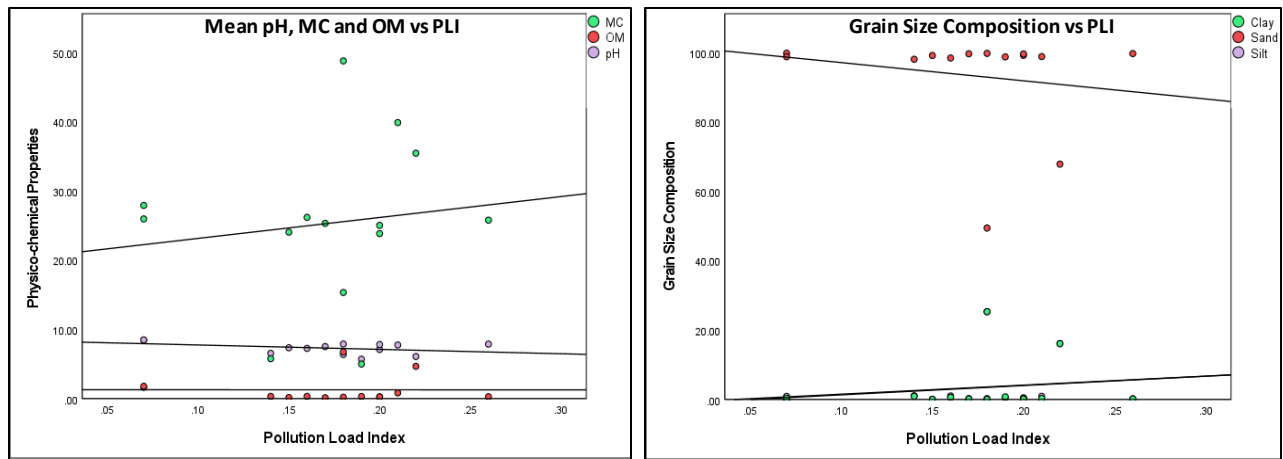


Diagram 3: Relationship between physico-chemical properties against PLI.

DISCUSSION

Physico-Chemical Properties in Marine Sediments

The sediment characteristics such as moisture content and water-holding capacity are affected by soil texture, structure, hydraulic conductivity, porosity, and permeability (Ball, 2011; Gwak and Kim, 2016). All sediments change from acidic to alkaline properties as the sampling stations are heading toward the seaward direction, probably due to the intrusion of seawater from the South China Sea, which may also impact the marine sediment quality (Ding *et al.*, 2020). Besides seawater intrusion, the dissolution of bicarbonate (HCO_3^-) yielded from the surrounding corals and shelled marine organisms further increased the alkaline properties of the sediments in the coastal region (Ahmed *et al.*, 2016). According to McCauley *et al.* (2017), the organic matter also affects pH values in sediments. The high SOM will release more H^+ ions into the sediments, which explains the results of Table 1. The SOM percentage also affects the moisture content. USDA (1975) also stated that sandy texture has lower SOM than clayey texture, leading to a lower buffer capacity and higher water infiltration rates in the sediments. Therefore, the fine-textured clay in mud soil has a higher affinity to hold SOM than the coarse-textured sand which contributed to the acidic properties of mud soil (Prasad and Power, 1997; Gui *et al.*, 2021). The clayey texture also influences the adsorption-desorption process of major elements such as Al in the sediments (Myung, 2008; Muli, 2017). The moisture content is inconsistent due to the difference in beach terrain, tidal cycle, storm-wave action, and evaporation or precipitation process from where the samples are collected (Scmutz and Namikas, 2018; Atherton *et al.*, 2001; Namikas *et al.*, 2010). According to Luo *et al.* (2020), soil pH has the ability to control the migration and redistribution of heavy metals in the beach sediments. The negatively-charged elements, including OH^- ions, can move freely in alkaline sediments and bind with other positively-charged metals such as Al, Fe and Mn from the fine-grain fraction of $<63\mu\text{m}$ as shown in the ICP-OES analysis (Allaway, 1957; Sparks, 2003). Generally, the spatial distribution of heavy metals in marine sediment is also dependent on the physico-chemical properties.

Sources Identification of Heavy Metals

Heavy metals are elements with a density of more than 5 g/cm^3 (Csuros and Csuros, 2002) or with an atomic weight of at least five times greater than the water density (Tchounwoo *et al.*, 2012). Sediments are sensitive indicators when assessing heavy metals. The metals are continuously added from different inputs, either terrestrial origins (parent materials, crustal earth, atmospheric deposition, marine water

column) or anthropogenic activities (industrial and agricultural activities, pesticide chemicals, domestic wastes). The accumulation of heavy metals in marine sediments that serve as a natural reservoir is thus selected for the geochemical study on heavy metal assessment (Förstner, 2006). In this study, the natural input sources of Fe and Mn elements in beach sediments are from atmospheric dust and direct volcanic origin, such as the hydrothermal fluids on the seafloor (Nicholson *et al.*, 1997; White, 2020). Although Fe is readily soluble in low oxygen levels and is essential to sustain the marine carbon cycle, it remains localized in sediments along the continental margins. The total Fe content in near-shore sediments is also high due to the high Fe^{2+}/Fe^{3+} ratio of beach sediment in aluminosilicate fractions, which were derived from the clay minerals or oxidised organic matter and accumulate in the marine sediment (Chen *et al.*, 1994; Dezileau *et al.*, 2007; Taylor and Mcquaker, 2011). The presence of Fe content in beach sediment also originated from the microbial reduction of Fe in near-shore marine sediment that formed the oxide minerals (Canfield, 1989; Grecco *et al.*, 2011). The alkaline properties also reduce the Fe solubility and restrict its mobility to build up in sediment (Lindsay, 1979; Tsai and Schmidt, 2020).

The Al content is also elevated in the sediment because it is the third most naturally occurring element in the earth's crust and from the weathering process of sedimentary rocks from both Crocker and Wariu Formation. The amphoteric properties of Al to form both organic and inorganic complexes and polymerise under a wide range of pH, temperature, dissolved substances, and long period of water exposure allows the element to exist abundantly in the marine environment (Dijkstra and Fitzhugh, 2003; Amir *et al.*, 2020). The alkaline properties also cause the Al element to become immobile and accumulate, which result in an elevated concentration in the sediments. The Mn element found in beach sediment along the shoreface originates from parent rocks, such as limestone and shale (Force and Cox, 1991; Govind *et al.*, 2021), cherts and basalts from the ophiolites (Sugisaki *et al.*, 1991; Narejo *et al.*, 2019). The calcareous foraminifera assemblages from the marine ecosystem also release Mn (Boyle, 1983, Bauer *et al.*, 2013; Li *et al.*, 2021). Besides Mn, the bioaccumulation of aquatic organisms is also enriched in Al in the seawater column, which ultimately also deposits in sediment (Moore and Bostrom, 1978; Neff, 2002). Other anthropogenic activities that may influence sediment quality are crop fertilization, pesticide chemicals, and other domestic or industrial wastes. In conclusion, the primary contributing factors for the heavy metal content in the sediments are natural sources such as the oceanographic setting, parent rock materials, and local geology. All the heavy metal burdens are transported from higher regions and accumulate in Usukan coastal beach that serves as a natural sink.

Marine Sediment Quality Assessment and Correlation

The Pollution Load Index (PLI) is a standardized system to assess the degree to which the sediment is associated with heavy metals and detect the pollution levels in sediments. The PLI values show very low pollution primarily due to the background level of pollutants in the study area. When compared with the background ASV, the average concentration of Al ($6,824 \pm 2,025$ ppm), Fe ($17,804 \pm 6,513$ ppm), and Mn (300 ± 109 ppm) metals are within the permissible background levels of 80,000 ppm, 47,200 ppm, and 850 ppm, respectively (Turekian and Wedepohl, 1961). The heavy metal concentrations are also below the standard limits established in the SQUIRTs for marine sediments (NOAA, 1999), which further justifies that the minor deterioration of pollutants in the study area is more likely from natural inputs than anthropogenic sources. The accumulation of metal pollutants in sediment loads are also greatly influenced by the fine granulometric fraction as the fine texture has greater pollutant retention and metal adsorption due to a higher specific surface area to volume ratio than the sand fraction (Abolfazl and Ahmad, 2012; Maher *et al.*, 1999; Hart, 1982; Durães *et al.*, 2018).

On the contrary, the sand fraction is coarser and has lower water retention but higher drainage, hence indicating a negative correlation with the metal pollutants in PLI. The slight negative correlation between pH against PLI also suggests that soil acidity increases the leaching of heavy metals to accumulate in sediments and affects the quality of marine sediment (Zhang *et al.*, 2018). Overall, the

sediment quality assessment does not show any significant impacts of metal pollution or severe threat to aquatic organisms and human health in the Usukan coastal beach.

CONCLUSION

The geochemical and statistical assessment of heavy metals indicates a very minimal degree of pollution in the marine sediments. The sediment quality indices show that the heavy metal contents of Al (6,824ppm), Fe (17,804ppm), and Mn (300ppm) are within the background values and standard limits of the Sediment Quality Guideline for marine sediments. The pollution load index is influenced by the physico-chemical properties of sediments such as the acidic to alkaline properties (pH 5.69-8.48), low to moderate moisture content (4.99-39.85%), very low to moderately high organic matter (0.12-6.73%) and fine to coarse texture which control the distribution of heavy metals to various parts of the marine sediments. The background pollutants are mainly from the geological source rocks comprising sedimentary rocks from Crocker Formation and limestone, shale, and ophiolitic rocks from Waru Formations that are transported by rivers, leachate, or run-offs to the marine sediments near the coastal regions. In short, the geochemical and statistical assessment implied that there are no significant impacts of metal pollution or severe threat to the marine sediment quality in Usukan coastal beach. Nevertheless, further investigations and ongoing monitoring are highly recommended to assess the long-term effects of their inputs into the Kota Belud marine ecosystem.

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