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Distribution and Contamination of Trace Elements in Mangrove Sediments Collected from West Coast of Peninsular Malaysia

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Abstract In tropical and subtropical latitudes, woody plants known as mangroves flourish in slimy, anaerobic soils on the boundary between land and sea. They can withstand extreme weather conditions like high temperatures, unreliable tides, strong winds, and salt. The objective of this study was to determine the concentration of nine trace elements (Hf, Ga, Ba, V, Cs, Sc, Sb, Ta, and Co) in the sediment of the mangrove forest along the west coast of Peninsular Malaysia. Instrumental neutron activation analysis (INAA) was employed to determine the distribution of the elements within the sediments. The concentration of trace elements in the sediment in descending order are Sb < Co < Ta < Sc < Hf < Cs < Ga < V < Ba. The degree of elemental contamination in the sediments was assessed using the enrichment factor (EF) and geoaccumulation index (Igeo). The EF and Igeo of trace element ranged from 0.41 to 20.76 and -3.56 to 1.41 respectively. The EF and Igeo values of a few trace elements in the sediments at Kampung Panchor (L6) suggested that the area had probably deteriorated.

Keywords Enrichment, Geoaccumulation, Pollution, Neutron Activation

1. Introduction

The term "trace elements" has been used in a variety of contexts to describe common pollutants, which are widely distributed in the environment and are mostly brought on by the weathering of minerals and soil. The periodic chart defines trace elements as metallic elements low on the list with an atomic weight > 100 or a relative density > 5 [1,2]. While some trace metals are known to have practically detrimental nutritional effects, others may be crucial in nutrition of humans, animals, and plants. The environment contains trace metals from both anthropogenic (human activities) and natural sources (Fergusson, 1990). There are many different types of contamination brought about by the buildup of trace metals in both agricultural and seafood products [3,4].

Mangroves are woody plants that thrive in slimy, anaerobic soils on the edge of land and sea in tropical and subtropical climates. Extreme weather conditions including high temperatures, erratic tides, powerful winds, and salt can be endured by them. Human activity is the primary cause of anthropogenic sources in Malaysia, including mining, agriculture, aquaculture, tourism, and the usage of metal in industry, which results in higher concentrations of heavy metals that contaminate the mangrove zones [5-9]. The metals from the polluted sediment and water could be absorbed by marine living organisms such as plankton, which feeds on the detritus, and constitute a substantial pathway for toxins to enter its predators (including human being as a consumer).

All types of pollutants generated by human activity eventually find their way to surface sediment, where they can pose a range of issues for the environment. On the other hand, top sediments frequently interact with suspended components, influencing the release of metals into the water below [10,11]. The top few centimeters of the sediments show the current level of contamination, which is constantly changing. Because sediment can provide useful information about marine pollution, it is necessary to use samples of sediment to evaluate the level of elements pollution throughout the Peninsular Malaysia's west coast. The enrichment factor and geo-accumulation index will be used to determine the pollution level of sediment.

2. Materials and Methods

Sampling and Sample Preparation

The sediment samples used in this study were collected from river estuaries and mangrove forests throughout west coast of Peninsular Malaysia (Figure 1). These locations included latitudes of N 05° 20' 24.7" to N 01° 15' 58" and longitudes of E 100°24'25.2" to E 103°30'39".

Tourism, industrial areas, agriculture, aquaculture, shipping, and hydroelectric power plants are some of the local activities of the ten study locations. These activities are summarised in Table 1.

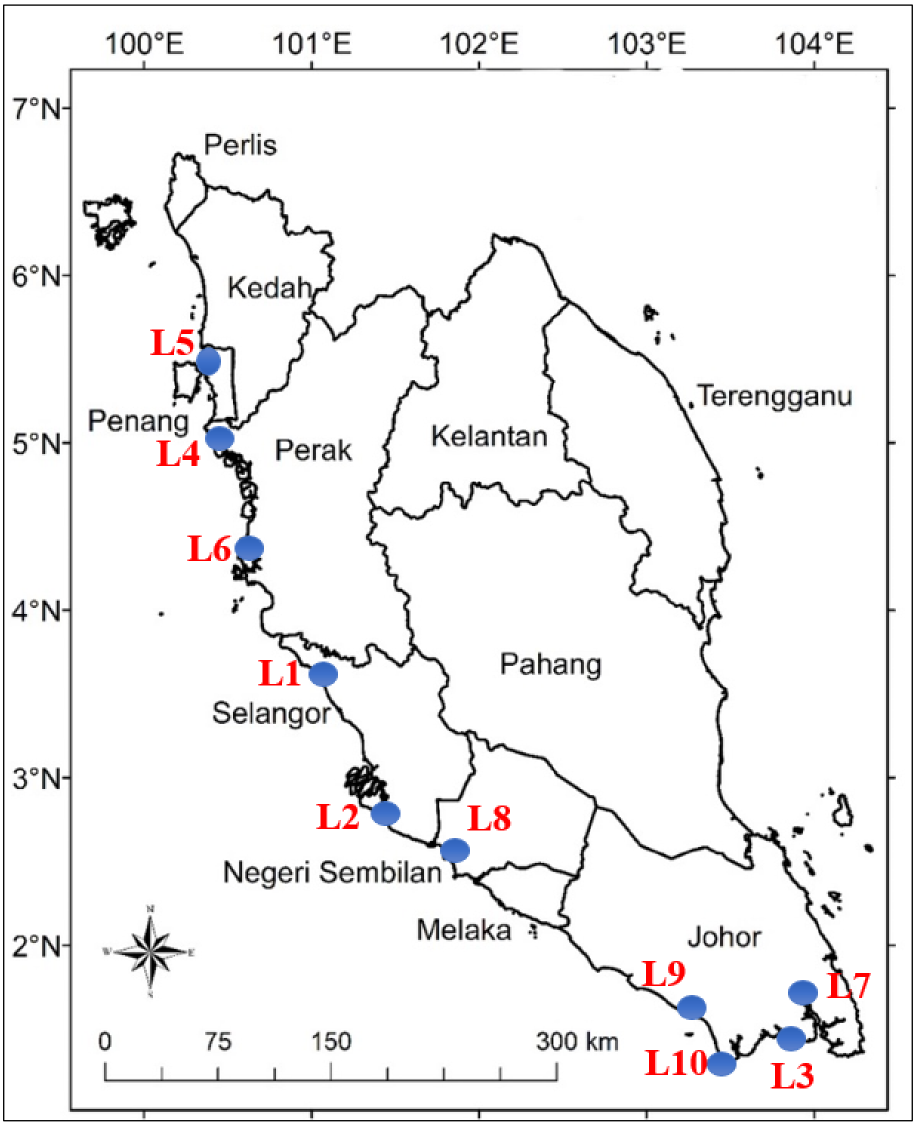

Figure 1. Sampling stations

Table 1. The description of sampling location

ID	Location Name and GPS Coordinate	Description of nearby activities
L1	Tok Muda, Kapar, Selangor N 03º 7' 30.9" E 101º 20' 27.7"	Hydroelectric power plant and residential area
L2	Sungai Besar Sepang, Selangor N 02º 56' 16.9" E 101º 45' 9.4"	Intertidal Area and residential area
L3	Sungai Pasir Gudang, Johor E 103º 57' 26" N 01º 24' 3.99"	Shipping, intertidal and residential area
L4	Kuala Gula, Perak N 04º 55' 58" E 100º 27' 33.6"	Fishing village, tourism Spot, fish cage and shrimp pond
L5	Juru, Penang N 05º 20' 24.7" E 100º 24' 25.2"	Industrial, residential and fishing village
L6	Kampung Panchor, Pantai Remis, Perak N 04º 31' 33.4" E 100º 39' 17.5"	Fishing village, tourism Spot, fish cage and shrimp pond
L7	Sungai Kim Kim, Johor N 01º 26' 40.2" E 103º 58' 14.2"	Intertidal Area and residential area
L8	Lukut, Port Dickson, Negeri Sembilan N 02º 34' 49.4" E 101º 47' 53.9"	Shipping and residential area
L9	Pulau Kukup, Johor N 01º 19' 18.7" E 103º 25' 30.6"	Fishing village, tourism Spot, fish cage and shrimp pond
L10	Johor Tanjung Pia, N 01º 15' 58" E 103º 30' 39"	Residential, shipping and tourism spot

Table 2. Sample radiation exposure time and counting period

	Short radiation		**Long radiation**	
Duration time	**1 minute**		**6 hours**	
Cooling time	20 minutes	24 hours	3 - 4 days	21-28 days
Counting time	5 minutes	20 minutes	1 hour	1 hour

Five samples of surface sediment were collected at each location, 0.5 to 1 m apart, at the depth of approximately 3.0 to 5.0 cm with a clean plastic spoon [5,12,13]. The surface sediment samples were kept at a temperature of -5 ^{o}C in plastic bottles [14]. Each location's surface sediments were heated to a consistent dry weight over a period of at least 72 hours at a temperature of about 80 °C to remove the moisture. Dry materials were powdered and sifted through a 63 µm stainless steel aperture after being ground into a powder in a glass mortar. After being vigorously stirred, the components were stored in plastic pillboxes [15,16].

Sample Preparation for Instrumental Neutron Activation Analysis

Approximately 0.150 g and 0.200 g of the powdered sample from each location were stored separately in heat-sealed polyethylene vials for short and extended irradiation, respectively. The concentration of the components was calculated using the INAA comparative approach. Blank samples, standard reference material

(SRM) IAEA - SL-1 (Lake Sediment) were exposed to thermal neutron flux of 4.0 x 10^{12} $cm^{-2}s^{-1}$ using a pneumatic transport facility as part of calibration and quality control processes at the MINT TRIGA Mark II research reactor running at 750 kW. Table 2 summarizes the sample radiation exposure time and counting.

A multichannel analyzer (MCA) and a hyper-pure Ge (HPGe) detector with an energy resolution of 1.8 keV at 1,332 keV of ^{60}Co were used to count radioactivity by using the precise energy of delayed gamma rays, the element present in the sample was identified, and the concentration of the element is calculated from the strength of the gamma peak. A distance between the sample and detector was maintained at 12–14 cm for short irradiation and 1-2 cm for long irradiation, depending on the level of activity of the irradiated samples. A 10% dead time was kept throughout the entire counting process [1,5,17,18].

Evaluation of Pollution Levels

There are numerous helpful techniques available to assess metal enrichment in sediment. The enrichment factor and geo-accumulation index methods were utilised in this study to calculate the influence of the pollutants on the sediment.

Enrichment Factor (EF)

The following equation was used to calculate the EF of heavy metals [20]:

$$EF = [(M/Fe)_{exp}]/[(M/Fe)_{shall}]$$

Where M_{ref} or Fe_{ref} refers to the common abundant element in typical shale, M_{exp} or Fe_{exp} refers to the element concentration in the experimental sample [21].

Classification and sediment contamination status are shown in Table 3.

Geo-accumulation Index

As suggested by Müller [23], another method that many researchers use to assess the enrichment of the metal above baseline or background is the geoaccumulation index (Igeo). According to Müller [23], the Igeo may be used to categorise the level of metal contamination into seven enrichment groups. Geo calculated using:

$$I_{geo} = \log_2[C_n/(1.5 \times B_n)]$$

Where C_n is the element's concentration in the enhanced samples and B_n is the element's background or pristine value. To reduce the potential impact of differences between background values that could lead to lithologic variances in the sediments, the factor 1.5 was utilized. Table 4 indicates classification and sediment contamination status [24,25].

Table 3. EF Classification

Classification	EF < 2	2 < EF< 5	5 < EF< 20	20 < EF< 40	EF > 40
enrichment status	Low enrichment	Moderate enrichment	High enrichment	Very high enrichment	Extremely enrichment

Table 4. Igeo classification

Classification	I_{geo}< 0	0< I_{geo} <1	1< I_{geo} <2	2< I_{geo} <3	3< I_{geo} <4	4< I_{geo} <5	I_{geo} > 5
Sediment pollution status	Unpolluted	Unpolluted to moderately polluted	moderately polluted	moderately to strongly	Strongly	strongly to extremely strongly	extremely polluted

3. Results and Discussion

The Certified Reference Material SL-1 was used to verify the sediment's purity and analytical processes. The analytical results for the measured SL-1 trace element values and certified reference material are shown in Table 5. The range of the INAA method's recoveries for any trace element was between 83.51 and 119.88%. The recovery rates between the measured and certified data using the INAA and AAS methods were satisfactory.

The results of the trace element assessment for Hf, Ga, Ba, V, Cs, Sc, Sb, Ta, and Co in the surface sediments of Peninsular Malaysia's west coast are reported in Table 6. The concentrations of Sb, Co, Ga, Hf, Sc, Ta, Cs, V and Ba varied from, 0.55 to 1.72, 1.84 to 8.58, 9.41 to 23.20, 4.04 to 11.00, 3.98 to 24.38, 1.01 to 3.18, 5.36 to 15.03, 16.50 to 109.71, 121.40 to 199.56 respectively.

The average concentrations of Hf, Ga, Sc, Ba, Sb, Ta, and V. Co, Cs were 7.46, 17.46, 11.47, 170.38, 1.08, 72.58, 5.76 and 8.72mg/kg, respectively. In comparison to other studies of mangrove sediments from the Santos Estuarine Sediments in Brazil [19] and surface marine sediments from Peninsular Malaysia [17], relatively lower Ba and Co contents were discovered at all sampling locations. The average concentrations of these nine elements decreased in the following order: descending order are Sb < Co < Ta < Sc < Hf < Cs < Ga < V< Ba. The concentration of metals that collected in sediment varied depending on their geographic location. Moreover, the change in the pattern of accumulation may have been influenced by the discharge of varied amounts of sewage and municipal wastes.

Figure 2 shows the EF factor for each element by using Fe as the normalising element. The EF of Ba, Co, Cs, Ba, Ga, Hf, Sc, Sb, Ta, and V in the sediment samples ranged from, 0.41 to 0.97, 2.43 to 8.33, 0.46 to 1.17, 1.37 to 3.35, 1.20 to 7.33, 0.66 to 1.86, 0.79 to 2.42, 2.15 to 20.76, and 1.14 to 3.27, respectively. It was found that Co (0.41) and Ta (20.76) lowest and highest respectively. Trace elements, Co and Ba at all of the studied locations, the majority of the sampling sites displayed EF values higher than 1.0, which may be classified as an absence of direct anthropogenic pollution. On the other hand, EF for the trace elements Ta, Cs and Hf of all the studied locations fall between 2 and 5, which denotes considerable enrichment (except at L6). EF for Sb, V, Sc, and Ga were typically less than 2, indicating that all of the locations studied had minor enrichment. The majority of the elements in L6 were found to be enriched in Cs, Ba, and Ta (EF > 5), which at this level of chemistry is known as a moderate to severe enrichment. High enrichment factor at L6 may have been caused by the aquaculture projects where the excretion of fish waste products, fish farming mortality, and fish decomposition, all of which will affect the mangrove sediments. However, other activities such as loading and unloading fish, uneaten fish feed, fish waste and cleaning fishing boats are routinely carried out at this sampling point L6, and the result is an accumulation of blood and organic waste in the sediments. According to other studies, there are many factors that are linked to the degradation of mangrove area such as tourism, agriculture, aquaculture, and urban sprawl that cause mangrove habitats to deteriorate [26,27].

Based on the Figure 3, geo-accumulation index of all the trace elements (Ba, Co, Cs, Ga, Hf, Sc, Sb, Ta, and V) fall into classes 0 (unpolluted) and 1 (unpolluted to moderately polluted). However, the EF of the trace element Ta at L6 indicated that the sampling site was likely moderately contaminated.

Table 5. The percentage recovery between measured and certified value of SL-1

Element	SL-1		Recovery (%)
	Measured value	Certified value	
Ba	639	533.6	83.51
Co	19.8	20.19	101.95
Cs	7	6.96	99.47
Ga	23.7	28.41	119.88
Hf	4.2	4.78	113.79
Sc	17.3	17.16	99.17
Sb	1.31	1.31	100.07
Ta	1.58	1.32	83.51
V	170	157.5	92.62

Table 6. Concentrations of trace elements (average ± standard deviation) in mg/kg

Element	L1	L2	L3	L4	L5	L6	L7	L8	L9	L10
Ba	121.4±5.8	199.6±13.3	192.2±13.3	177.5±34.0	193.5±53.3	129.8±13.4	nd	135.6±30.7	197.6±45.5	186.3±26.8
Co	5.46±0.52	6.59±0.72	3.97±0.27	5.84±0.31	8.23±0.49	1.84±0.09	6.12±0.22	3.33±0.13	8.58±1.07	7.67±0.39
Cs	5.36±1.24	8.72±1.49	7.28±0.38	10.8±0.6	15.0±0.9	7.99±0.68	7.71±1.02	8.11±2.19	7.77±0.69	8.35±0.36
Ga	9.41±0.40	20.6±3.9	16.7±3.1	17.6±2.6	21.1±3.6	12.2±1.7	23.2±2.1	19.7±0.7	14.9±0.9	19.1±4.1
Hf	6.34±1.73	11.0±1.7	7.78±0.47	4.34±0.19	4.04±0.34	6.47±1.12	10.4±0.4	8.95±0.13	8.68±0.77	6.57±0.50
Sc	5.40±1.21	10.2±0.9	12.8±0.2	11.3±0.3	13.1±0.63	3.98±0.20	12.5±0.3	10.9±0.4	24.4±1.1	10.2±0.6
Sb	0.55±0.24	0.90±0.07	1.51±0.01	0.81±0.17	0.87±0.13	0.65±0.19	1.72±0.08	1.54±0.15	1.13±0.07	nd
Ta	1.01±0.29	1.56±0.30	1.66±0.12	1.68±0.22	2.84±0.23	3.18±0.66	2.10±0.09	2.24±0.06	nd	1.18±0.05
V	37.0±1.1	58.7±3.2	109.7±10.4	71.9±5.6	77.13±8.71	16.50±2.85	103.36±5.34	103.14±3.78	85.06±22.31	63.38±5.85

nd – Not detected

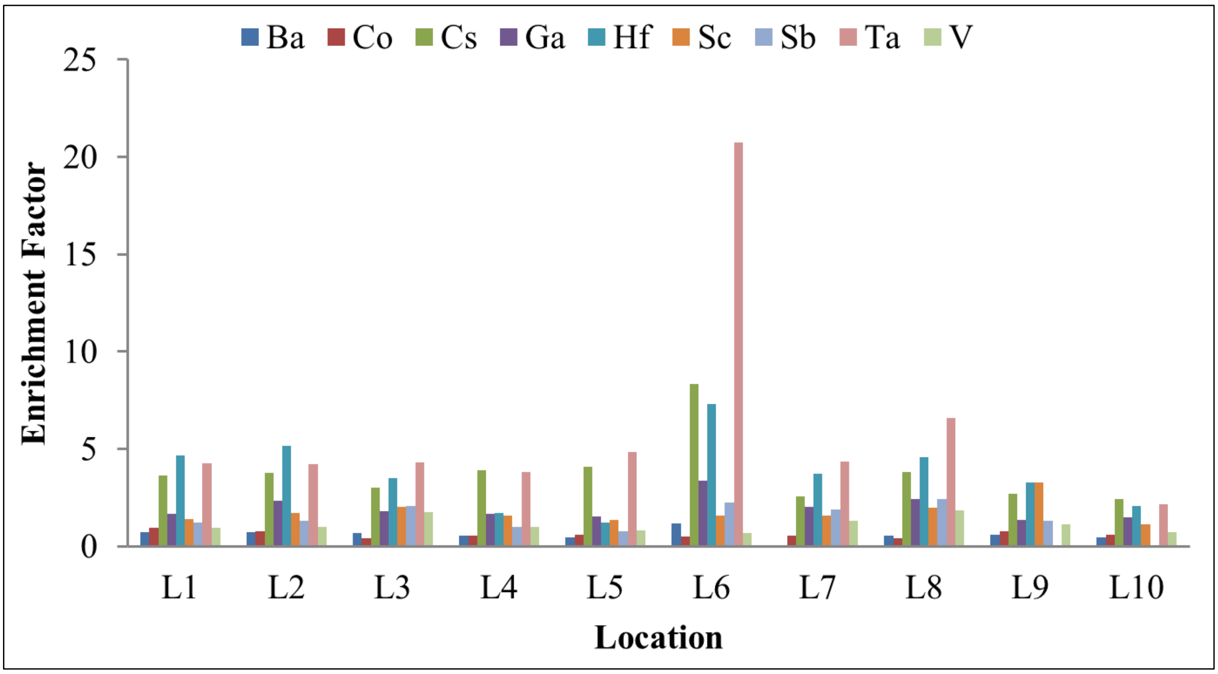

Figure 2. Enrichment factor (EF) of Trace elements in the sediment

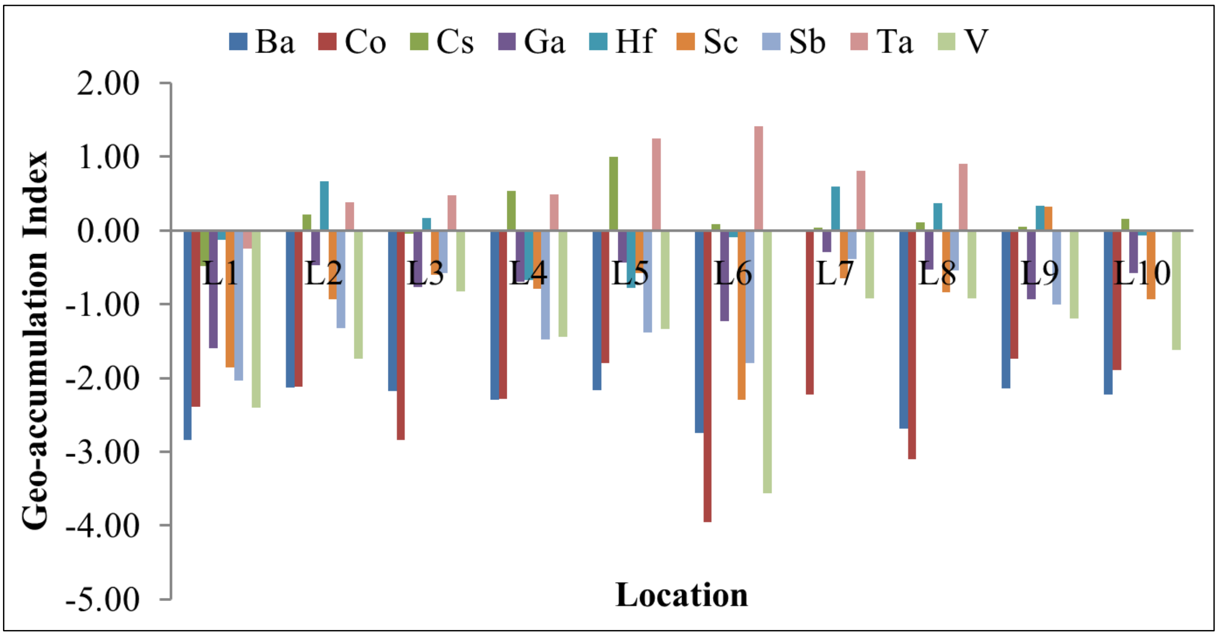

Figure 3. Geoaccumulation index (Igeo) of trace elements in the surface sediment

Table 7. Correlation regression of trace elements in the sediment

	Ba	Co	Cs	Ga	Hf	Sc	Sb	Ta	V
Ba	1	0.286	0.336	-0.139	-0.334	0.267	-0.412	-0.273	-0.095
Co		1	0.39	0.34	-0.063	0.627	-0.237	-0.542	0.2
Cs			1	0.499	-0.528	0.166	-0.084	0.444	0.108
Ga				1	0.314	0.279	0.38	0.241	0.629
Hf					1	0.211	0.516	-0.24	0.323
Sc						1	0.361	-0.579	0.607
Sb							1	0.126	0.742*
Ta								1	-0.162
V									1

*Correlation is significant at the 0.05 level (2-tailed)

The data set and statistical analysis were used to create correlation matrices that evaluated the relationships between the investigated metal concentrations. Table 7 lists the Pearson correlation coefficient values (r) between metal concentrations. The correlation coefficients between Co and Sc, Ga and V, Sc and V, and Sb and V are all significantly positive at 0.627, 0.607, 0.742, and 0.629, respectively. The significant association between Co and Sc, Ga and V, Sc and V, and Sb and V, is an indication of the complexity of trace elements as well as the significance of the role played by the diagenetic events triggered by their degradation, which in turn controls the behaviour of these trace metals. The strong association shows that the concentrations of these metals are consistent with the origins of the trace elements. Anthropogenic sources are the main contributing source of trace elements in places with mangroves [2,28].

4. Conclusions

Nine trace element concentrations in sediment from the mangrove forest along Peninsular Malaysia's west coast were studied. To ascertain how the elements were distributed within the sediments, instrumental neutron activation analysis (INAA) was used. The concentration of trace elements in the sediment in descending order are Sb < Co < Ta < Sc < Hf < Cs < Ga < V< Ba. The degree of elemental contamination in the sediments was assessed using the enrichment factor (EF) and geoaccumulation index (Igeo). The Results from EF and Igeo shows trace elements Cs, Hf, and Ta of all the studied locations fall between 2 and 5, which denotes considerable enrichment (except at L6). Ta is one of the most abundant elements in L6, which indicates that the area has likely deteriorated and could potentially have negative effects on the area's living resources. According to Pearson's correlation, the trace elements Co and Sc, Ga and V, Sc and V, and Sb and V have an anthropogenic influence on the mangrove sediments. The study's data will be used as a source of information for upcoming studies.

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