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*Research Article*

# Spatial Distribution of Selected Heavy Metals in Surface Sediments of the EEZ of the East Coast of Peninsular Malaysia

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Spatial distribution of selected metals (Al, Fe, Mn, Zn, Cu, and Co) in surface sediments in the EEZ of the east coast of Peninsular Malaysia was investigated. The aim of this paper is to determine the distribution pattern and pollution status of heavy metals in tropical shelf sediments since limited information is available. Heavy metal concentrations ranged between 207.58 and 491.33 $\mu g \cdot g^{-1}$ for Mn, 36.13 and 125.93 $\mu g \cdot g^{-1}$ for Zn, 14.49 and 22.33 $\mu g \cdot g^{-1}$ for Cu, 2.00 and 11.12 $\mu g \cdot g^{-1}$ for Co, 6.20 and 8.95% for Fe, and 0.94 and 6.62% for Al. The mean concentrations of heavy metals are in decreasing order as follows: Fe > Al > Mn > Zn > Cu > Co. Most metals registered low concentrations at the nearshore areas. Pearson correlation indicates that most of the metals are derived from the miscellaneous sources. Based on the EFs and Igeo, it is implied that the surface sediment trace metal levels in the study area might be enriched by anthropogenic sources. However, the PLI suggests that this area is not contaminated from the measured heavy metals. This work is important to register the current levels of metals so that any change in concentration can be monitored and managed.

## 1. Introduction

The exclusive economic zone (EEZ) of the east coast of Peninsular Malaysia that lies in the Sunda Shelf receives enormous sediment input from rivers under the monsoon type tropical climate. Heavy metals in marine sediments originate primarily from natural (riverine discharge) and anthropogenic (coastal human settlements) sources and are subject to both continental and marine control. When heavy metals enter the marine environment, the distribution is influenced by various physicochemical factors (e.g., sedimentary processes, mineralogical composition, hydrodynamic transport, redox conditions, and biological uptake). Significant amounts of pollutants are received by marine ecosystem every year via several pathways such as heavy industry processes, agriculture, aquaculture, untreated domestic waste, atmospheric emission, and shipping activities. Heavy metals are considered to be contaminants when human activity raises their concentrations in the environment exceeding natural levels [1].

Studies dealing with heavy metals in the EEZ of the east coast of Peninsular Malaysia are very limited [2–5]. Unfortunately, there is only one study that covered the area until the edge of the east coast of Peninsular Malaysia EEZ [2]. Most of the works have focused on specific area such as coastal waters [3–6], mangrove forest [7–9], and river-estuary [10–12]. Hence, it is important to understand the spatial distribution and concentration of metals in the study area. In this study, authors investigated the distribution of selected heavy metals and the pollution status and source of the heavy metals in the study area using environmental assessment indexes: pollution load index (PLI) [13, 14], enrichment factor (EF) [15, 16], and index of geoaccumulation (Igeo) [17, 18]. This study will be useful in environmental management by enlightening policy makers about the heavy metal issues in this crucial region.

## 2. Materials and Methods

*2.1. Environmental Setting.* The EEZ of the east coast of Peninsular Malaysia covers an area from 1°14.047′ to 7°48.92′N

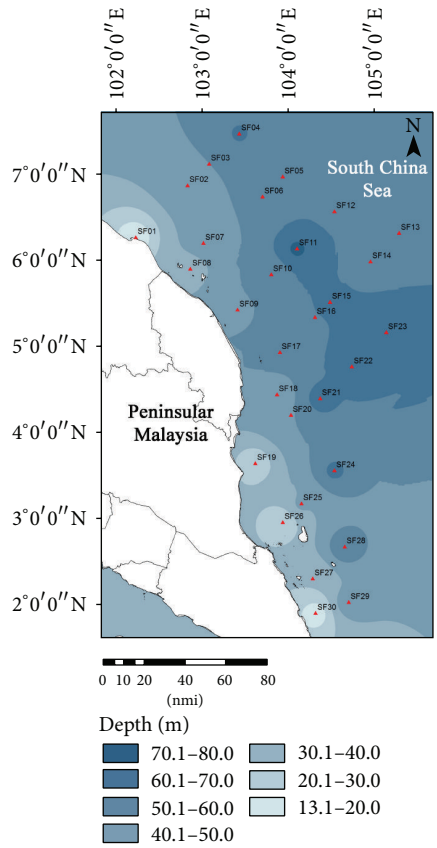


Figure 1: Map showing the location of sampling stations in the study area.

latitude and 102°5.03′ to 105°48.77′E longitude and is about 130,000 km$^2$ [19] located at the edge of the Indo-West Pacific (Figure 1). During the northeast monsoon, lower temperature and salinity are recorded due to the heavy rainfall and lower solar radiation [20]. Strong current flows close to the Peninsular Malaysia landmass in the northeast and southwest monsoons [21] and, therefore, controls sediment movement [22]. The water circulation is predominantly cyclonic during the northeast monsoon in the winter and anticyclonic during the southeast monsoon in summer [21]. The nearshore areas are influenced by the influx of several rivers (i.e., Kelantan River, Terengganu River, Kemaman River, Pahang River, and Pontian River).

*2.2. Sample Collection and Preservation.* Surface sediments from 30 sampling stations were collected during the cruise in June 2008 aboard the Department of Fisheries, Malaysia, KL Paus using Smith McIntyre Grab sampler (Figure 1). The water depths of the sampling locations ranged from 13.0 to 72.0 meters (Table 1). Sediments of the upper 5 cm were collected with a plastic spoon and stored in clean vinyl bags to prevent possible contamination. The sediment samples were oven-dried at 50°C and passed through a 2 mm sieve. From the bulk sediment, a representative subsample was powdered and homogenized in an agate mortar with a pestle for further analysis.

Table 1: The coordinates of each sampling station.

| Station | Latitude | Longitude | Depth (m) |
|---|---|---|---|
| SF01 | 06°13.99′N | 102°19.00′E | 13 |
| SF02 | 06°50.04′N | 102°47.04′E | 46.5 |
| SF03 | 07°05.03′N | 103°04.99′E | 50 |
| SF04 | 07°25.98′N | 103°26.01′E | 61 |
| SF05 | 06°56.09′N | 103°56.04′E | 52 |
| SF06 | 06°42.14′N | 103°35.17′E | 52 |
| SF07 | 06°10.00′N | 103°01.00′E | 45 |
| SF08 | 05°52.10′N | 102°51.92′E | 34 |
| SF09 | 05°22.06′N | 102°21.97′E | 47 |
| SF10 | 05°48.20′N | 103°48.98′E | 55 |
| SF11 | 06°06.16′N | 104°09.11′E | 72 |
| SF12 | 06°32.01′N | 104°22.11′E | 59 |
| SF13 | 06°16.98′N | 105°16.99′E | 55 |
| SF14 | 05°57.15′N | 104°58.13′E | 56 |
| SE15 | 05°29.08′N | 104°29.02′E | 60 |
| SF16 | 05°18.50′N | 104°12.60′E | 60 |
| SF17 | 04°54.12′N | 103°42.98′E | 54 |
| SF18 | 04°28.14′N | 103°49.98′E | 40 |
| SF19 | 03°37.07′N | 103°41.08′E | 23 |
| SF20 | 03°55.10′N | 104°00.05′E | 50 |
| SF21 | 04°22.16′N | 104°22.07′E | 65 |
| SF22 | 04°44.19′N | 104°38.44′E | 66 |
| SF23 | 05°08.10′N | 105°12.90′E | 67 |
| SF24 | 03°32.08′N | 104°36.00′E | 62 |
| SF25 | 03°09.14′N | 104°09.04′E | 41 |
| SF26 | 02°56.13′N | 103°49.97′E | 20 |
| SF27 | 02°16.94′N | 104°16.97′E | 30 |
| SF28 | 02°39.18′N | 104°38.91′E | 58 |
| SF29 | 02°00.55′N | 104°41.97′E | 46 |
| SF30 | 01°48.04′N | 104°15.03′E | 14 |

*2.3. Analytical Procedure.* Approximately 0.05 g of homogenized sample was digested in a sealed Teflon vessel with 1.5 mL mixed concentrated acids (2 HF : 3 $HNO_3$ : 3 HCl) [23, 24] with some modifications. The Teflon vessels were heated at 150°C for 7 hours. A clear solution with no residue was obtained at this stage. After cooling to room temperature, the digested solution in the Teflon vessel was transferred into a 15 mL polypropylene test tube and diluted with 10 mL high purity deionized water. An inductively coupled plasma mass spectrometer (ICP-MS; model 6200 Perkin Elmer Ltd.) was used for the determinations of Al, Fe, Cu, Mn, Zn, and Co. The accuracy was examined by analyzing in duplicate a Canadian Certified Reference Materials Project standard (NBS 1646a). All glassware and Teflon sample cups in this study were soaked in nitric acid of 5% overnight, rinsed with distilled water, and oven-dried to eliminate potential contamination as part of the QA/QC program.

*2.4. Enrichment Factor (EF).* This method normalises the measured metal concentration with respect to a reference metal and is frequently used as an indicator for pollution [15, 16]. The continental shale abundance metal concentrations [1] were used as the background metal contents. Aluminium is

Table 2: Classification of EF.

| EF value | Classification |
|---|---|
| EF < 1 | No enrichment |
| EF < 3 | Minor enrichment |
| EF = 3–5 | Moderate enrichment |
| EF = 5–10 | Moderately severe enrichment |
| EF = 10–25, | Severe enrichment |
| EF = 25–50 | Very severe enrichment |
| EF > 50 | Extremely severe enrichment |

used as a reference element because of its conservative nature [25–28]. Also, Al is a major constituent of clay mineral [29–31] and has been used as a reference element to assess the status of heavy metals pollution for some environments in Malaysia [4, 6, 32]. The EF was calculated using the relation proposed by Sutherland [15] and expressed as

$$\mathrm{EF} = \frac{[\mathrm{Cn/Cref}]}{[\mathrm{Bn/Bref}]}, \quad (1)$$

where Cn is content of the examined element in the examined environment, Cref is content of the examined element in the reference environment, Bn is content of the reference element in the examined environment, and Bref is content of the reference element in the reference environment. The categories of EF were proposed by Sutherland [15] as described in Table 2.

*2.5. Index of Geoaccumulation Index (Igeo).* Igeo is generally used to compare the status of heavy metal concentration with the background values. Igeo can describe the relationship between the measured element in the sediment fraction and the geochemical value in fossil argillaceous sediment or average shales [1]. This index was calculated using the formula suggested by Müller [33]:

$$\mathrm{Igeo} = \log_2\left[\frac{\mathrm{Cn}}{1.5\mathrm{Bn}}\right], \quad (2)$$

where Cn is the measured concentration of the element in soil or sediment and Bn is the geochemical background value. The constant value, 1.5, is background matrix correction factor due to the lithological variations. Müller [17] proposed the following descriptive classes for the Igeo values (Table 3).

*2.6. Pollution Load Index (PLI).* This index is widely used as a simple and comparative way to evaluate the degree of heavy metal pollution in marine sediment [13, 14]. This index is derived from the contamination factor (CF) that was proposed by Müller [17]. The background values of metals in this study are the average values of continental shale [1]. The CF ratio was estimated by dividing the concentration of each metal in the soil by the baseline/background value [14]:

$$\mathrm{CF} = \frac{\left[C_{\text{heavy metal}}\right]}{\left[C_{\text{background}}\right]}. \quad (3)$$

The PLI was determined as the root of the product of the $n$ CF:

$$\mathrm{PLI} = \left(\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \times \cdots \times \mathrm{CF}_n\right)^{1/n}. \quad (4)$$

Table 3: Classification of geoaccumulation index.

| Igeo | Igeo class | Description of sediment quality |
|---|---|---|
| <0 | 0 | Uncontaminated |
| 0-1 | 1 | Uncontaminated to moderately contaminated |
| 1-2 | 2 | Moderately contaminated |
| 2-3 | 3 | Moderately to strongly contaminated |
| 3-4 | 4 | Strongly contaminated |
| 4-5 | 5 | Strongly to extremely strongly contaminated |
| >5 | 6 | Extremely contaminated |

Table 4: CF and PLI classification metals.

| CF | Classification |
|---|---|
| 0 | None |
| 1 | None to medium |
| 2 | Moderate |
| 3 | Moderate to strong |
| 4 | Strongly polluted |
| 5 | Strong to very strong |
| 6 | Very strong |
| PLI | Classification |
| >1 | Polluted |
| <1 | No pollution |

Table 5: The value of accuracy of analysis for standard reference.

| Metals | Value of SRM ($\mu$g/g) | Value recorded ($\mu$g/g) | Accuracy of analysis (%) |
|---|---|---|---|
| Al | 2.297 | 2.633 | 114.63 |
| Fe | 2.008 | 2.303 | 114.69 |
| Cu | 10.01 | 9.8 | 97.03 |
| Mn | 234.5 | 244.7 | 104.36 |
| Zn | 48.9 | 43.2 | 88.34 |
| Co | 5 | 5.280 | 105.60 |

This empirical index provides a simple, comparative means for assessing the level of heavy metal pollution [13]. The contamination classes based on CF and PLI proposed by Tomlinson et al. [13] are shown in Table 4.

## 3. Results and Discussion

The recovery test coincided with the certified values of NBS 1646a. The recovery percentage of measured metals was found to be acceptable, ranging between 88.34% and 114.69% (Table 5).

The selected heavy metal concentrations in the surface sediments from 30 stations varied from 207.58 to 491.33 $\mu g \cdot g^{-1}$ for Mn, 36.13 to 125.93 $\mu g \cdot g^{-1}$ for Zn, 14.49 to 22.33 $\mu g \cdot g^{-1}$ for Cu, and 2.00 to 11.12 $\mu g \cdot g^{-1}$ for Co (Table 6). The average concentrations of Mn, Zn, Cu, and Co are

Table 6: Heavy metals concentration in each station.

| Station | Al | Fe | Cu | Mn | Zn | Co |
|---|---|---|---|---|---|---|
| SF01 | 5.23 | 7.84 | 21.98 | 338.61 | 61.19 | 9.50 |
| SF02 | 5.28 | 7.23 | 22.05 | 332.68 | 62.20 | 9.72 |
| SF03 | 4.77 | 7.36 | 21.07 | 316.10 | 69.58 | 9.52 |
| SF04 | 6.62 | 7.86 | 22.33 | 300.97 | 80.39 | 10.14 |
| SF05 | 4.47 | 7.22 | 19.75 | 346.04 | 62.56 | 9.66 |
| SF06 | 4.78 | 7.74 | 18.57 | 288.24 | 75.69 | 10.08 |
| SF07 | 5.16 | 7.31 | 19.04 | 274.51 | 65.29 | 9.35 |
| SF08 | 4.08 | 7.18 | 18.92 | 272.00 | 63.00 | 9.98 |
| SF09 | 3.86 | 7.51 | 17.41 | 256.86 | 62.94 | 9.24 |
| SF10 | 3.25 | 7.15 | 17.15 | 258.06 | 52.88 | 8.92 |
| SF11 | 5.00 | 7.30 | 17.19 | 296.44 | 62.06 | 9.41 |
| SF12 | 3.76 | 7.32 | 17.09 | 491.33 | 57.03 | 9.19 |
| SF13 | 4.75 | 7.48 | 17.12 | 372.78 | 63.51 | 10.10 |
| SF14 | 3.87 | 7.60 | 17.20 | 248.53 | 73.78 | 11.00 |
| SE15 | 3.27 | 7.31 | 16.41 | 250.00 | 58.73 | 9.43 |
| SF10 | 3.25 | 7.15 | 17.15 | 258.06 | 52.88 | 8.92 |
| SF11 | 5.00 | 7.30 | 17.19 | 296.44 | 62.06 | 9.41 |
| SF12 | 3.76 | 7.32 | 17.09 | 491.33 | 57.03 | 9.19 |
| SF13 | 4.75 | 7.48 | 17.12 | 372.78 | 63.51 | 10.10 |
| SF14 | 3.87 | 7.60 | 17.20 | 248.53 | 73.78 | 11.00 |
| SE15 | 3.27 | 7.31 | 16.41 | 250.00 | 58.73 | 9.43 |
| SF16 | 3.55 | 7.21 | 15.90 | 264.71 | 50.59 | 9.69 |
| SF17 | 4.12 | 7.90 | 16.14 | 239.04 | 62.95 | 9.02 |
| SF18 | 3.30 | 7.05 | 15.51 | 234.38 | 50.78 | 9.12 |
| SF19 | 0.94 | 6.20 | 14.49 | 226.56 | 39.45 | 8.13 |
| SF20 | 3.98 | 7.56 | 15.46 | 248.78 | 57.79 | 9.75 |
| SF21 | 3.66 | 7.39 | 16.13 | 236.79 | 51.27 | 8.86 |
| SF22 | 4.27 | 8.04 | 16.12 | 247.06 | 68.43 | 9.71 |
| SF23 | 5.39 | 8.05 | 16.20 | 240.16 | 79.13 | 10.14 |
| SF24 | 3.98 | 7.92 | 17.63 | 247.06 | 64.31 | 9.39 |
| SF25 | 3.35 | 7.71 | 16.34 | 238.19 | 65.26 | 11.12 |
| SF26 | 4.17 | 7.21 | 14.65 | 207.58 | 36.13 | 8.88 |
| SF27 | 4.15 | 7.94 | 17.08 | 232.14 | 54.37 | 2.00 |
| SF28 | 4.05 | 8.19 | 15.26 | 233.20 | 59.09 | 9.01 |
| SF29 | 4.13 | 8.95 | 17.96 | 223.97 | 125.93 | 9.47 |
| SF30 | 5.59 | 8.09 | 16.40 | 239.92 | 53.98 | 9.44 |
| **Average** | **4.23 ± 1.0** | **7.56 ± 0.49** | **17.48 ± 2.14** | **273.42 ± 57.92** | **63.01 ± 15.49** | **9.30 ± 1.50** |

The concentration of Al and Fe in percentage (%).
The other elements are in $\mu g \cdot g^{-1}$.

$273.42 \pm 57.92\ \mu g \cdot g^{-1}$, $63.01 \pm 15.49\ \mu g \cdot g^{-1}$, $17.48 \pm 2.14\ \mu g \cdot g^{-1}$, and $9.1 \pm 1.5\ \mu g \cdot g^{-1}$. Fe and Al contents vary, respectively, from 6.20 to 8.95% and from 0.94 to 6.62%, with an average of 7.56 ± 0.49% and 4.23 ± 1.0% (Table 6). The average concentrations of the heavy metals in this study followed the order of Fe > Al > Mn > Zn > Cu > Co.

Pearson correlation coefficient matrix was used to distinguish the relationships between observed heavy metals (Table 7). Only two moderate correlations were shown between elements Zn-Fe ($r = 0.71$, $p < 0.05$) and Al-Cu ($r = 0.63$, $p < 0.05$). The positive correlations indicate that

Table 7: Correlation between heavy metals and organic carbon in the study area.

| Elements | Al | Fe | Cu | Mn | Zn | Co |
|---|---|---|---|---|---|---|
| Al | 1.00 | | | | | |
| Fe | 0.49 | 1.00 | | | | |
| Cu | 0.63 | 0.10 | 1.00 | | | |
| Mn | 0.27 | −0.18 | 0.48 | 1.00 | | |
| Zn | 0.38 | 0.71 | 0.37 | 0.02 | 1.00 | |
| Co | 0.17 | 0.00 | 0.15 | 0.17 | 0.27 | 1.00 |

One-way ANOVA; $p < 0.05$.

the contents of these metals in the surface sediments probably originated from similar sources [34]. Most of the metals showed a weak correlation between each other ($0 < r < 0.49$, $p < 0.05$) indicating different sources. Meanwhile, Fe shows a negative correlation with Mn ($r = -0.18$, $p < 0.05$) suggesting that both metals maybe originated from different sources [35] or due to the pH-redox sensitive nature of these two major elements their postdepositional concentrations are subject to alteration by the complex biochemical processes. The statistical analysis of one-way ANOVA shows a significant difference in concentration between studied heavy metals.

Sediment Fe levels were higher in the southern sampling locations especially at SF28, SF29, and SF30 (Johor waters). The sources of Fe input in the area are unknown and, therefore, require further research. However, a huge amount of iron source may originate from the iron ore mining activity [36, 37] and other anthropogenic sources. The Fe contents in the other sites were also slightly higher than shale average [1]. This is owing to high estimated iron ore reserve in the east coast of Peninsular Malaysia. The iron-rich sedimentary rocks in the mining area of the east coast of Peninsular Malaysia are estimated to be transported via rivers run-off and land run-off [38]. Denton et al. [39] stated that the waste discharges from industrial processes are the potential sources of elevated heavy metals in the environment. Unfortunately, there is no available data of Fe from the river that linked to mining activity to substantiate this idea.

It is noted that concentration of Al was slightly higher in Kelantan and Johor waters than the other sites. The higher concentration of Al in those particular areas might be originated from both natural and anthropogenic sources. In the Kelantan area, a higher value of Al is assigned to the transports of aluminium-rich sediment by the river flow. This metal is commonly found in soil, minerals (e.g., sapphires, rubies, and turquoise), rocks (especially igneous rocks), and clays [40]. Logging and reforestation activities in the upstream areas such as Gua Musang and Lojing may expose the aluminum-rich sedimentary rocks or soils to weathering process and rainwater. The high energy of Kelantan River flow plays a major role in carrying the suspended particles towards the ocean. Meanwhile, the high content of Al in Johor waters might result from the bauxite mining activity in Ramunia Bay. Bauxite is a rock consisting of aluminum hydrate or hydroxide minerals, and it is the principal raw material that is used in the aluminum industry [41]. The aluminium-rich

Table 8: The comparison of heavy metal concentration in this study with other studies.

| Number | Area | Al | Fe | Cu | Zn | Mn | Co | Reference |
|---|---|---|---|---|---|---|---|---|
| (1) | EEZ of the east coast of Peninsular Malaysia | 4.23 ± 1.0 | 7.56 ± 0.49 | 17.48 ± 2.14 | 63.01 ± 15.47 | 273.42 ± 57.92 | 9.30 ± 1.5 | This study |
| (2) | EEZ of the east coast of Peninsular Malaysia (premonsoon) | 3.04 ± 1.4 | 2.03 ± 0.45 | 16.0 ± 7.2 | 76.3 ± 8.3 | n.a. | n.a. | [2] |
| (3) | EEZ of the east coast of Peninsular Malaysia (postmonsoon) | 4.57 ± 1.34 | 1.36 ± 0.38 | 15.1 ± 2.7 | 56.1 ± 17.1 | 269 ± 80 | n.a. | [2] |
| (4) | Strait of Johor | 8.25 ± 2.49 | 3.04 ± 0.67 | 30.7 ± 22.5 | 132.5 ± 52.6 | 265 ± 152 | 5.8 ± 1.5 | [52] |
| (5) | Strait of Malacca | n.a. | 2.15 ± 0.59 | 17.46 ± 8.08 | 63.68 ± 21.93 | 421 ± 209 | n.a. | [53] |
| (6) | Terengganu River | n.a. | n.a. | n.a. | n.a. | 517.9 ± 161.8 | 15.1 ± 7.4 | [10] |
| (7) | Kemaman River | n.a. | n.a. | 48.8 | n.a. | 597.8 | 16.0 | [4] |
| (8) | Pahang River | n.a | n.a | 18.65 ± 7.65 | n.a. | n.a. | n.a. | [12] |
| (9) | Upper crust, North China | n.a. | n.a. | 25 | 61 | n.a. | 18 | [54] |
| (10) | Upper continental crust | 7.7 | 3.1 | 14.3 | 52 | 527 | 11.6 | [55] |
| (11) | Shale average | 8.4 | 4.7 | 45 | 95 | 850 | 19 | [1] |

The concentration of Al and Fe in percentage (%).
The other elements are in $\mu g \cdot g^{-1}$.
n.a.: not analyzed.

minerals are easier to be transported to the study area via land run-off due to the mining area that is located close to the shoreline [38].

The higher concentration of Mn was observed at the offshore area of Terengganu waters. The distribution pattern of Mn as determined in the present paper was consistent with that in the previous study [2]. Salomons and Forstner [42] claimed that the chemical composition and diagenesis of sediment could affect metal accumulation process. The concentrations of Mn at all sites in the study area were relatively lower than the value of river-estuary sediments [10, 11], suggesting that the river discharge may flush the manganese-rich sediment from terrestrial into the coastal area. Then, the current dynamic may play a role in dispersing it toward the north and the offshore area. The other major factors that may control the metal distribution are the oxidation of accumulated metals and bioturbation by the benthic organisms [43].

The average Zn concentration was found to be comparable to the previous reports of Shazili et al. [2] in the sediments of this area. The weathering effects may change those materials into soluble form of Zn which is released into aquatic environment [44, 45]. The concentration of Zn was found to be higher in the surface sediment at site SF29 (Johor waters). The concentrations of this metal in the other sampling sites were almost uniform. We assume that the distribution pattern of Zn is closely related to the iron-rich deposits due to high adsorption between Zn and Fe ($r = 0.79$). According to Salomons and Forstner [42], iron hydroxides are able to adsorb large quantities of metals through cation exchange processes, and iron oxides also play an important role in trapping metals in aquatic sediments [46]. The other possible sources of Zn are from motor oil, grease, phosphate fertilizers, sewage sludge, transmission fluid, undercoating, and concrete [47].

Higher concentration of Cu was determined in Kelantan waters than the other sampling sites. High correlation between Cu and Al is probably due to the same origin and transport and the chemical affinities between them. Naturally, copper can be discharged into the environment from forest fires, weathering process of exposed soil, and decaying vegetation, while anthropogenic source of copper may originate from domestic use of copper-based chemicals, municipal untreated sewage sludge, and corrosion of copper materials. Kelantan River that flows via high-populated urban areas (e.g., Kuala Krai, Tanah Merah, and Kota Bharu) may carry these substances downstream. The other potential source of Cu in Kelantan waters might be from the leachate of Sabak beach landfill area. The produced leachate that contains soluble form of Cu [48–50] may permeate into the coastal area.

The concentration of Co was slightly high at SF13, SF14, and SF23 (Terengganu offshore), and the lowest concentration was identified at SF27 (Johor waters). Cobalt may enter the aquatic environment from both natural sources and human activities [51]. Under natural conditions, Co is found in most rocks, soil, water, plants, and animals. The association of Co with Mn is most likely due to the adsorption on to the Mn-(oxyhydr)oxides and consequent coprecipitation as marine sediments. The anthropogenic sources of cobalt are derived from soils near ore deposits and ore smelting facilities and soils contaminated by airport traffic, highway traffic, or other industrial pollution source [51]. Cobalt may enter aquatic environment via run-off and leaching when rainwater washes through the substances containing cobalt. Thus, the presence of this metal in the studied area possibly derived from the river discharge [48]. This is due to high concentration of Co as reported in the Kerteh mangrove [11], Terengganu River [10], and Pahang River [8].

The comparison of heavy metal compositions was made between the study area and other studies in the region and the crustal values of metals (Table 8). The average concentrations of metals (Al, Cu, Mn, Zn, and Co) are almost similar to initial report by Shazili et al. [2] except for Fe. On average,

Fe concentration was higher than the regional studies [52, 53], upper continental shelf [54], and shale average [1]. The concentration of Al, Cu, Mn, and Zn seemed to be lower and/or similar with respect to the regional studies [52, 53], upper continental crust [54, 55], and shale average [1]. In the meantime, Co was comparatively higher than the value of Strait of Johor [52], but it was still lower compared to the river-estuary values [8, 10], upper continental crust [54, 55], and shale average [1].

The distribution patterns of studied metals significantly varied spatially in the study area, but most of them were consistent with low concentration in nearshore area of Terengganu and Pahang (Figure 2). It seems that current dynamic is not the major factor affecting the distribution of studied metals in this study. But, we are unable to discuss this factor in more detail owing to the absence of current dynamic data such as wave, current, and sediment budget.

*Evaluation of Pollution*. For a better estimation of the heavy metals status in the surface sediments, the pollution load index (PLI), enrichment factor (EF), and index of geoaccumulation (Igeo) were calculated and discussed. The combination of these indices has been recognized as a powerful tool for assessing the anthropogenic input of heavy metals in different environments [56–58].

The EF was calculated for a better assessment of anthropogenic input for each metal (Table 9). The average EF for Fe (3.51) indicates that the surface sediments in the study area were moderately enriched with this metal [59]. The EF value above 1.5 indicates an anthropogenic contribution [60]. The EF values of Co (1.1), Al (1.0), Cu (0.8), Mn (0.7), Zn (0.7), and Pb (0.4) suggest that the sediments have no enrichment with those metals. According to Zhang and Liu [60], if EF is less than 1.5, the metal concentration is considered crustal or natural weathering origin. EF value close to 1 reflected a crustal origin, while those with a factor more than 10 are considered to have noncrustal sources [7]. The higher the EF values, the more severe the anthropogenic contribution. The highest EF value was recorded at SF19 sampling location for Fe (11.8). But, the calculated value for Fe at that site might be argued due to the remarkably low Al content (0.94%). In this study, the studied heavy metals were proven to be between no enrichment and moderately enrichment.

The values of Igeo in the sediment samples are shown in Table 10. The Igeo of Al (0.6) indicates that it remains in class 1 in all stations, suggesting that the study area is in background value with respect to this metal [17, 33]. The average value of Igeo for Co (0.7), Mn (0.8), Zn (0.8), Cu (0.8), and Fe (1.0) attains class 1 which indicates that sediments were unpolluted to moderately polluted with these metals [13].

The mean CF values for the metals in the study area follow the decreasing order as Fe > Zn > Al > Co > Cu > Mn (Table 11). As similar to EF, the value of Fe (1.6) was slightly higher than the other elements which may probably indicate the elevated Fe from anthropogenic sources into the study area. However, in terms of the total metals contamination, PLI value <0.5 implies that the EEZ of the east coast of Peninsular Malaysia was not polluted with the studied heavy metals. Some have proposed that the combination of low CF (C < 2) and PLI (<0.1) could be categorized as less polluted and/or not polluted [13, 17].

Table 9: Enrichment factor for each heavy metal in study area.

| Station | Elements | | | | | |
|---|---|---|---|---|---|---|
| | Al | Fe | Cu | Mn | Zn | Co |
| SF01 | 1.0 | 2.7 | 0.8 | 0.6 | 0.7 | 0.8 |
| SF02 | 1.0 | 2.5 | 0.8 | 0.6 | 0.8 | 0.8 |
| SF03 | 1.0 | 2.8 | 0.8 | 0.7 | 0.8 | 0.9 |
| SF04 | 1.0 | 2.1 | 0.6 | 0.4 | 0.9 | 0.7 |
| SF05 | 1.0 | 2.9 | 0.8 | 0.8 | 0.8 | 1.0 |
| SF06 | 1.0 | 2.9 | 0.7 | 0.6 | 0.9 | 0.9 |
| SF07 | 1.0 | 2.5 | 0.7 | 0.5 | 0.8 | 0.8 |
| SF08 | 1.0 | 3.1 | 0.9 | 0.7 | 0.8 | 1.1 |
| SF09 | 1.0 | 3.5 | 0.8 | 0.7 | 0.7 | 1.1 |
| SF10 | 1.0 | 3.9 | 1.0 | 0.8 | 0.7 | 1.2 |
| SF11 | 1.0 | 2.6 | 0.6 | 0.6 | 0.8 | 0.8 |
| SF12 | 1.0 | 3.5 | 0.8 | 1.3 | 0.7 | 1.1 |
| SF13 | 1.0 | 2.8 | 0.7 | 0.8 | 0.8 | 0.9 |
| SF14 | 1.0 | 3.5 | 0.8 | 0.6 | 0.9 | 1.3 |
| SF15 | 1.0 | 4.0 | 0.9 | 0.8 | 0.7 | 1.3 |
| SF16 | 1.0 | 3.6 | 0.8 | 0.7 | 0.6 | 1.2 |
| SF17 | 1.0 | 3.4 | 0.7 | 0.6 | 0.7 | 1.0 |
| SF18 | 1.0 | 3.8 | 0.9 | 0.7 | 0.6 | 1.2 |
| SF19 | 1.0 | 11.8 | 2.9 | 2.4 | 0.6 | 3.8 |
| SF20 | 1.0 | 3.4 | 0.7 | 0.6 | 0.7 | 1.1 |
| SF21 | 1.0 | 3.6 | 0.8 | 0.6 | 0.6 | 1.1 |
| SF22 | 1.0 | 3.4 | 0.7 | 0.6 | 0.8 | 1.0 |
| SF23 | 1.0 | 2.7 | 0.6 | 0.4 | 0.9 | 0.8 |
| SF24 | 1.0 | 3.6 | 0.8 | 0.6 | 0.7 | 1.0 |
| SF25 | 1.0 | 4.1 | 0.9 | 0.7 | 0.7 | 1.5 |
| SF26 | 1.0 | 3.1 | 0.7 | 0.5 | 0.4 | 0.9 |
| SF27 | 1.0 | 3.4 | 0.8 | 0.6 | 0.6 | 0.2 |
| SF28 | 1.0 | 3.6 | 0.7 | 0.6 | 0.6 | 1.0 |
| SF29 | 1.0 | 3.9 | 0.8 | 0.5 | 1.2 | 1.0 |
| SF30 | 1.0 | 2.6 | 0.5 | 0.4 | 0.6 | 0.7 |
| **Average** | **1.0** | **3.5** | **0.9** | **0.7** | **0.7** | **1.1** |

## 4. Conclusion

The distribution patterns of investigated metals (Al, Fe, Cu, Mn, Zn, and Co) varied in the study area, but most of them were consistent with low concentration in nearshore area of Terengganu and Pahang area. The removal of fine sediments away from the nearshore locations seems to be the controlling factor of metal concentrations. The spatial distribution of selected heavy metals varied significantly in surface sediments of the EEZ of the east coast of Peninsular Malaysia. The heavy metals are added to the sediments in the study area by two main pathways, via river discharge and land run-off. Various indices applied suggested that the sources of most measured heavy metals are merely natural in origin with exception of Fe which registered slightly higher levels. The higher content of Fe in the study area is probably derived

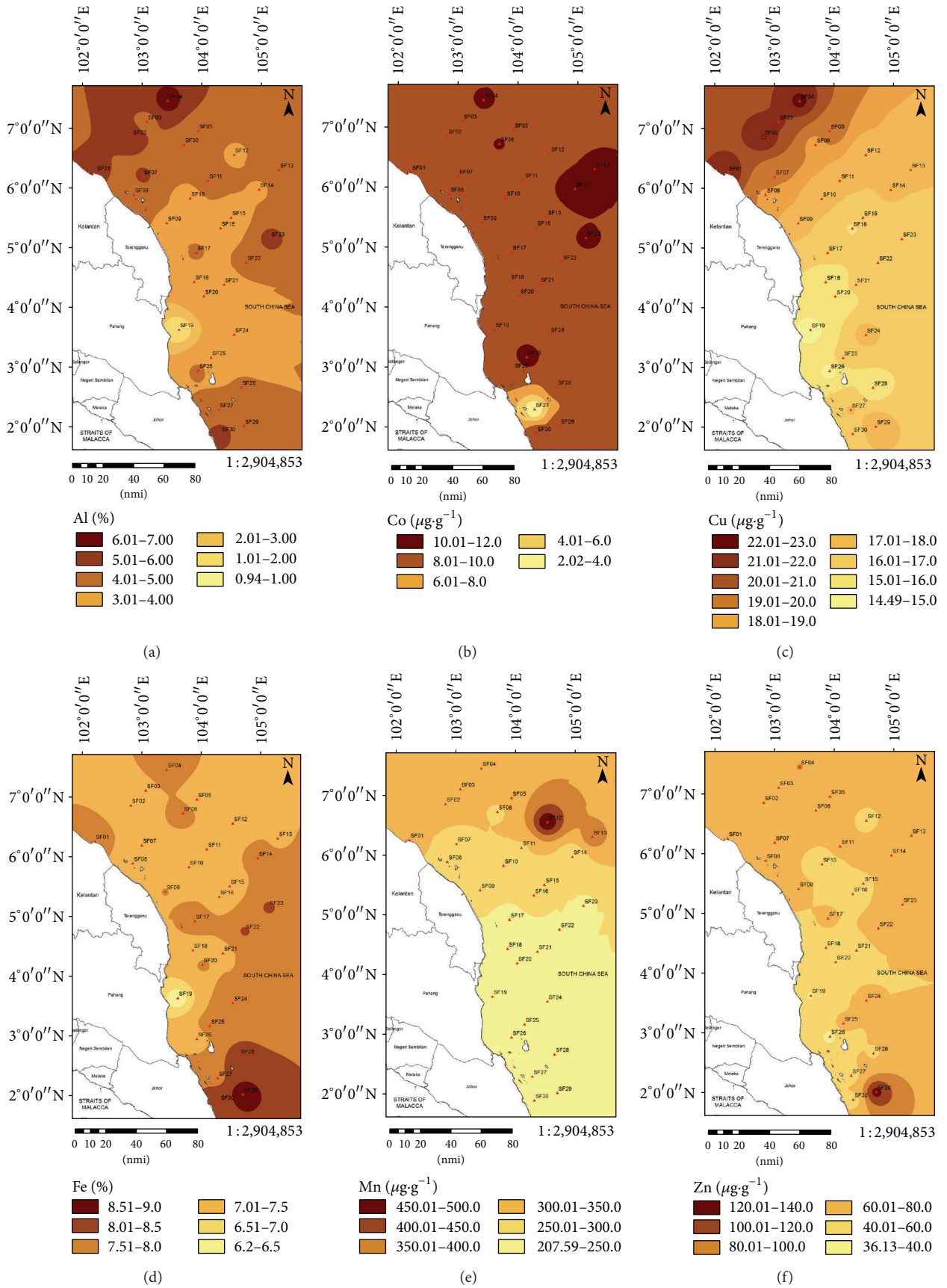


Figure 2: Distribution patterns of heavy metals in the EEZ of the east coast of Peninsular Malaysia.

Table 10: Index of geoaccumulation of heavy metals in the study area.

| Station | Elements | | | | | |
|---|---|---|---|---|---|---|
| | Al | Fe | Cu | Mn | Zn | Co |
| SF01 | 0.7 | 1.1 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF02 | 0.7 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF03 | 0.6 | 1.0 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF04 | 0.7 | 1.1 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF05 | 0.6 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF06 | 0.6 | 1.0 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF07 | 0.6 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF08 | 0.6 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF09 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF10 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF11 | 0.6 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF12 | 0.5 | 1.0 | 0.7 | 0.9 | 0.8 | 0.7 |
| SF13 | 0.6 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF14 | 0.5 | 1.0 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF15 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF16 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF17 | 0.6 | 1.1 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF18 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF19 | 0.0 | 0.9 | 0.6 | 0.8 | 0.7 | 0.6 |
| SF20 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF21 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF22 | 0.6 | 1.1 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF23 | 0.7 | 1.1 | 0.7 | 0.8 | 0.9 | 0.7 |
| SF24 | 0.5 | 1.1 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF25 | 0.5 | 1.0 | 0.7 | 0.8 | 0.8 | 0.7 |
| SF26 | 0.6 | 1.0 | 0.6 | 0.7 | 0.7 | 0.7 |
| SF27 | 0.6 | 1.1 | 0.7 | 0.8 | 0.8 | 0.2 |
| SF28 | 0.6 | 1.1 | 0.6 | 0.8 | 0.8 | 0.7 |
| SF29 | 0.6 | 1.1 | 0.7 | 0.8 | 1.0 | 0.7 |
| SF30 | 0.7 | 1.1 | 0.7 | 0.8 | 0.8 | 0.7 |
| **Average** | **0.6** | **1.0** | **0.7** | **0.8** | **0.8** | **0.7** |

Table 11: Contamination factors (CFs) and pollution load indices (PLIs) of sediment heavy metals in the study area.

| Station | Elements | | | | | | |
|---|---|---|---|---|---|---|---|
| | Al | Fe | Cu | Mn | Zn | Co | PLI |
| SF1 | 0.6 | 1.7 | 0.5 | 0.4 | 0.6 | 0.5 | 0.5 |
| SF2 | 0.6 | 1.5 | 0.5 | 0.4 | 0.7 | 0.5 | 0.5 |
| SF3 | 0.6 | 1.6 | 0.5 | 0.4 | 0.7 | 0.5 | 0.5 |
| SF4 | 0.8 | 1.7 | 0.5 | 0.4 | 0.8 | 0.5 | 0.5 |
| SF5 | 0.5 | 1.5 | 0.4 | 0.4 | 0.7 | 0.5 | 0.5 |
| SF6 | 0.6 | 1.6 | 0.4 | 0.3 | 0.8 | 0.5 | 0.5 |
| SF7 | 0.6 | 1.6 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF8 | 0.5 | 1.5 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF9 | 0.5 | 1.6 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF10 | 0.4 | 1.5 | 0.4 | 0.3 | 0.6 | 0.5 | 0.4 |
| SF11 | 0.6 | 1.6 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF12 | 0.4 | 1.6 | 0.4 | 0.6 | 0.6 | 0.5 | 0.5 |
| SF13 | 0.6 | 1.6 | 0.4 | 0.4 | 0.7 | 0.5 | 0.5 |
| SF14 | 0.5 | 1.6 | 0.4 | 0.3 | 0.8 | 0.6 | 0.4 |
| SF15 | 0.4 | 1.6 | 0.4 | 0.3 | 0.6 | 0.5 | 0.4 |
| SF16 | 0.4 | 1.5 | 0.4 | 0.3 | 0.5 | 0.5 | 0.4 |
| SF17 | 0.5 | 1.7 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF18 | 0.4 | 1.5 | 0.3 | 0.3 | 0.5 | 0.5 | 0.4 |
| SF19 | 0.1 | 1.3 | 0.3 | 0.3 | 0.4 | 0.4 | 0.3 |
| SF20 | 0.5 | 1.6 | 0.3 | 0.3 | 0.6 | 0.5 | 0.4 |
| SF21 | 0.4 | 1.6 | 0.4 | 0.3 | 0.5 | 0.5 | 0.4 |
| SF22 | 0.5 | 1.7 | 0.4 | 0.3 | 0.7 | 0.5 | 0.5 |
| SF23 | 0.6 | 1.7 | 0.4 | 0.3 | 0.8 | 0.5 | 0.5 |
| SF24 | 0.5 | 1.7 | 0.4 | 0.3 | 0.7 | 0.5 | 0.4 |
| SF25 | 0.4 | 1.6 | 0.4 | 0.3 | 0.7 | 0.6 | 0.4 |
| SF26 | 0.5 | 1.5 | 0.3 | 0.2 | 0.4 | 0.5 | 0.4 |
| SF27 | 0.5 | 1.7 | 0.4 | 0.3 | 0.6 | 0.1 | 0.3 |
| SF28 | 0.5 | 1.7 | 0.3 | 0.3 | 0.6 | 0.5 | 0.4 |
| SF29 | 0.5 | 1.9 | 0.4 | 0.3 | 1.3 | 0.5 | 0.5 |
| SF30 | 0.7 | 1.7 | 0.4 | 0.3 | 0.6 | 0.5 | 0.4 |
| **Average** | **0.5** | **1.6** | **0.4** | **0.3** | **0.7** | **0.5** | **0.4** |

from iron ore mining activities in the east coast of Peninsular Malaysia. Although the study area receives significant input by the anthropogenic sources of Fe, it is negligible compared with polluted areas. The other metals which registered lower concentration than the upper Earth's crustal and shale average value pointing to background levels are trace metals. Thus, we assume that there is not any significant pollutant in the EEZ of the east coast of Peninsular Malaysia. The data from this study will provide valuable information about the current status of heavy metal pollution and as baseline data for the future research in the region as part of continued assessment of metal levels when development along the east coast of Peninsular Malaysia is happening rapidly.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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