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# Pollution Status of Heavy Metals in Surface Sediments Collected from West Coast of Peninsular Malaysia

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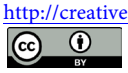



## Abstract

The present study examined status of heavy metals (Cu, Zn, Pb, Cd and Ni) pollution in the surface sediments of west coast of Peninsular Malaysia. Heavy metals concentrations were determined by using air acetylene flame atomic absorption spectrophotometer (AAS) Perkin Elmer Analyst 800. The results of particle size analysis of the surface sediments indicate high metals concentrations at sampling sites with high content of sand and clay particles. The results of Pearson's correlation analysis based on the relationship between particle size distribution and total heavy metals, and the relationship between the studied metals (Cu, Zn, Pb, Cd and Ni) revealed significant correlations at ($p < 0.05$ or $p < 0.01$) except few cases were non-significant correlations were observed. The I*geo* values ranged between I*geo* class 1 (unpolluted to moderately polluted) to Igeo class 5 (strongly to very strongly polluted) and the EF of no enrichment (EF < 1) to severe enrichment (EF = 10 - 25) depending on the sampling sites and the heavy metal. Lead showed the highest I*geo* values at all the sampling sites with exception of Bg. Lalang (class 1) and (EF = 10 - 25) at two sampling sites which might suggest the metal was significantly impacted by anthropogenic sources at the sampling sites. The pollution load index (PLI) indicates strong signs of pollution deterioration by the studied metals at all the sampling sites. The order of sampling sites from highest to lowest PLI values was; 2 > 5 > 3 > 1 > 6 > 4. The I*geo*, EF and PLI values indicate heavy metals contamination in the study area, particularly Pb. Considerable attention should be made at sampling sites that showed higher metals contamination in order to monitor metals pollution and save its biological components from deterioration.

## Subject Areas

Environmental Sciences

**Keywords**

Heavy Metals, Surface Sediments, Pollution Indices, West Coast of Peninsular Malaysia

## 1. Introduction

The continuous discharge of heavy metals into the coastal environment has attracted global attention and concern due to their toxicity, persistent and accumulation in sediment and living organisms. Sediments act as a sink of organic as well as inorganic pollutants (heavy metals) and provide a history of anthropogenic pollutant input [1] and environmental changes [2].

The pollution status of marine sediments has often been used as an important criterion to evaluate the condition of coastal environment and to understand the possible environmental changes caused by anthropogenic activities [3]. Generally, in unpolluted environment most of the heavy metals are in low levels. However, high metal concentrations in sediment do not automatically imply that contamination has occurred, but may simply reflect the natural mineralogical composition of the parent geological material and the grain size and organic matter content of the host sediment [4]. Naturally occurring elements as a result of pedogenesis are rarely noxious and are present in trace concentrations [5]. Heavy metals and related pollutants released from agricultural, industrial and domestic activities, ports, shipping boating and recreational use of water bodies, land-reclamation, tourism, chemical spills, sewage treatment plants, leaching from domestic garbage dumps and mining tend to accumulate in water, sediments and biota. Pollution of the natural environment by heavy metals is a worldwide problem as these metals are indestructible and have toxic effects on living organisms when they exceed a certain concentration limit [6].

Coastal ecosystems are among the most valuable on earth due to their provisioning of ecosystem services [7] [8]. Despite their value, coastal ecosystems are being rapidly degraded globally by human activities [9] [10] [11] such as eutrophication, toxic substances, heavy metals, acidification and siltation [12], which might have accelerated damage to the coast [13]. In coastal ecosystem, metals exist in either dissolved state in the water column or get deposited on the sediment bed, depending upon the nature of the chemical species and physicochemical factors like pH, conductivity, salinity and organic matter [14] [15] [16]. Despite different sources of heavy metals, the coastal and marine sediments are considered as the main fate for heavy metals presence in aquatic systems [17].

Research efforts have focused on coastal environments since these highly productive and sensitive areas are often directly and most seriously affected and exposed to this problem because of their proximity to sources of pollution [18] [19] from industries, shipping, agriculture domestic and other related activities. Heavy metals are regarded as a main anthropogenic pollutant in coastal and ma-

rine environments globally [20]. The enrichment of coastal environments with anthropogenic contaminants particularly heavy metals remains a topic of discussion in ecotoxicology. Several researches on heavy metals contamination in the coastal environments were reported from different part of the world by many authors; [21] [22] [23] [24] [25].

Recently, great development has occurred around Malaysia, especially Peninsular Malaysia as industrialization, urbanization, the advancement of agriculture and other activities related to the modern era are occurring rapidly, which have directly influenced the coastal ecosystems that contain aquatic resources, for example, fish, which are also used by humans [26]. The significant and benefit of the west coast of Peninsular Malaysia can not to be overemphasized as such many studies on distribution, enrichment and accumulation of heavy metals in the sediment of this area were reported by [27] and [28]. The present study was aimed to assess pollution status of heavy metal (Cu, Zn, Pb, Cd, and Ni) in the surface sediments of some selected areas in the west coast of Peninsular Malaysia using I*geo*, EF and PLI indices.

## 2. Materials and Methods

### 2.1. Study Area

Samplings were conducted in August 2008 to June 2010. A total of six sampling sites were selected for surface sediments in the west coast of Peninsular Malaysia (**Figure 1**). The sampling sites were chosen based on the past researches and human activities in the selected sampling sites in order to justify the status of heavy metals in the surface sediment and look for possibility of low or high metals input. The coordinates of sampling sites were recorded with Global positioning system (Garmin OREGON 45OT 850 MB waterproof GPS). Description of each sampling site was given in **Table 1**.

### 2.2. Sample Preparation and Acid Digestion

Surface sediment were collected in triplicate from top 3 - 5 cm [29] [30] at each sampling site (**Figure 1**), using plastic scoop and placed in separate labelled polyethylene plastic bags. Each sediment sample was instantly placed in ice and transported to the laboratory until further analysis.

**Table 1.** Description of sampling sites in the west coast of Peninsular Malaysia.

| No. | Sampling sites | Coordinates | Site description |
|---|---|---|---|
| 1. | S. Garam | N05˚39.552' E100˚23.983' | Jetty, aqua cultural area and paddy field |
| 2. | K. Juru | N05˚19.683' E100˚22.949' | Industrial area, urbanization and aquaculture |
| 3. | Sg. Puluh | N03˚04.786' E101˚23.903' | Jetty receiving domestic wastes and industrial area |
| 4. | Bg. Lalang | N02˚36.669' E101˚41.100' | Recreational and agricultural areas |
| 5. | M. Beku | N01˚47.746' E102˚53.395' | Jetty receiving domestic wastes and shipping activities |
| 6. | Sg. Tiga | N01˚25.841' E104˚00.281' | Jetty, agricultural and oil plantation |

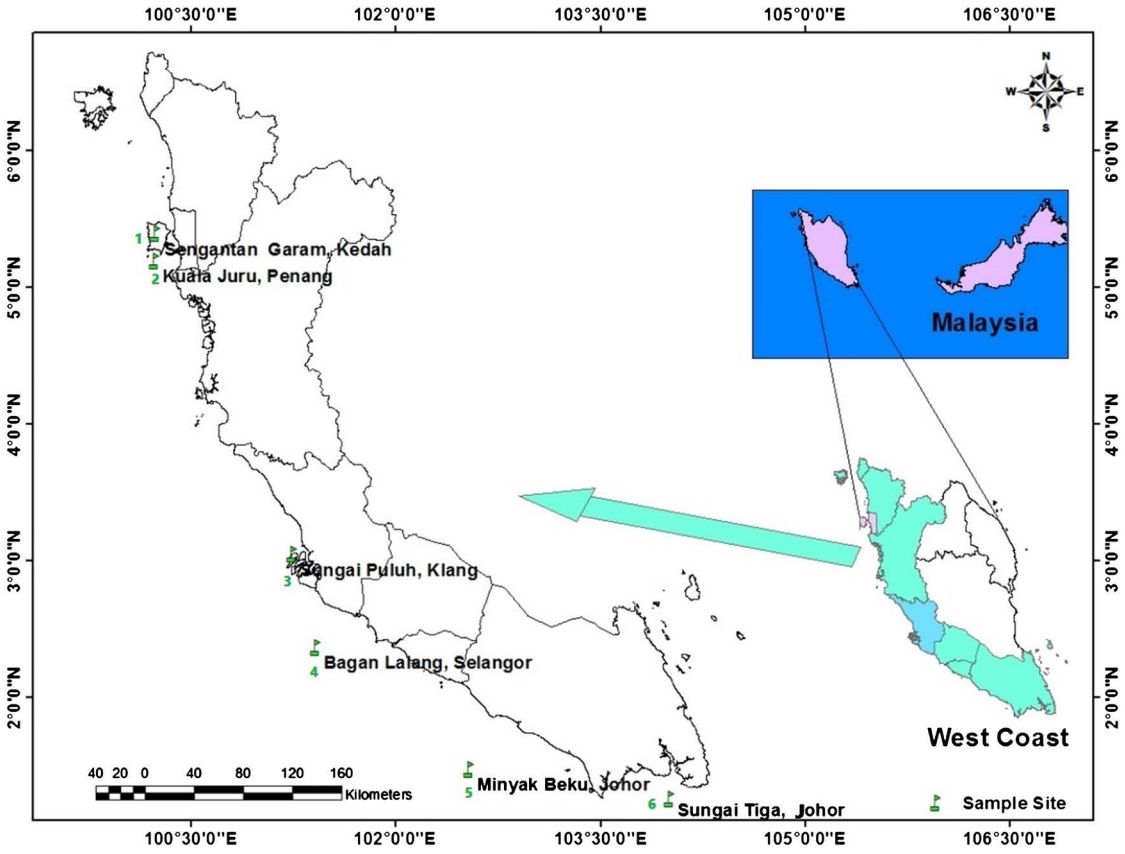


Figure 1. Map of west of Peninsular Malaysia showing six sampling sites (1, 2, 3, 4, 5 and 6).

Sediment samples were dried in the laboratory using an air-circulating oven to a constant dry weight at 80˚C. The dried sediment samples were crushed to powder by using a porcelain mortar and pestle then sieved vigorously to produce homogeneity [29], through a 63 μm stainless steel aperture sieve. For analyses of total heavy metals concentrations (Cu, Zn, Pb, Cd and Ni) in the sediment sample, direct aqua-regia method was employed as described by [29].

All samples were placed in a hot block digester at low temperature (40˚C) for 1 h and were then fully digested at high temperature (140˚C) for 3 h. The digested samples were diluted to a volume of 40 mL using double distilled water (DDW). The samples were filtered using Whatman No.1filter paper and the filtrate was stored in a refrigerator (4˚C) until metal determination. After filtration, the prepared samples were determined for Cu, Zn, Pb, Cd and Ni using an air-acetylene flame Atomic Spectrophotometer (AAS) Perkin-Elmer Model A Analyst 800. The data were presented in μg/g dry weight.

During the period of AAS metal analysis, a quality control sample was routinely included for every 5 - 10 samples. Procedural blanks and quality control samples made from standard solutions for Cu, Zn, Pb, Cd and Ni were analysed after every 5 - 10 samples to ensure the sensitivity and recovery of the instrument used. The procedures of quality assurance (QA) and quality control (QC) were employed to ensure the validity of the analytical data [31]. All plastics and

glassware used were washed with detergent, Deacon 90, rinsed with double-distilled water and soaked in 10% $HNO_3$ for at least 24 h, then rinsed with double-distilled water and allowed to dry at room temperature. The QA and QC were controlled by procedural blanks, sample replicates and certified reference material (CRM). The quality of the method was checked with a certified reference material (CRM) for Soil from International Atomic Energy Agency (IAEA), Soil-5, Vienna; Austria and Dogfish liver DOLT-3 from National Research Council Canada (NRCC) were analysed. These were checked to accuracy of the digestion method with the certified values supplied by the IAEA and NRCC. To ensure the sensitivity of the Atomic Absorption Spectrophotometer (AAS) and generate calibration curves against which sample concentrations were calculated. The results of similar digested samples analysed for Cu, Zn, Pb, Cd and Ni by the flame AAS Perkin Elmer A Analyst 800 showed acceptable recoveries of the metals. About 94% - 107% for soil and 87 - 110 for dogfish liver, recoveries of these metals were listed in Table 2. The percentages of recoveries for the heavy metal analyses were 97% - 107% (Table 2).

## 2.3. Surface Sediments Analyses

### 2.3.1. Particle Size Analysis

Particle size analysis is the measurement of the size of individual particles in the soil/sediment. Particle-size analysis of sediment samples was performed by pipette method following the procedure of [32]. The percentage of clay, silt and sand was classified by using United States Department of Agriculture (USDA) sediments classification scheme [33].

### 2.3.2. Assessment of Metals Contamination

Metals concentrations were calculated using world average shale [34] and lowest concentration in the present study as background concentrations (Table 3).

Table 2. Comparison of analytical result of CRM Soil-5 and Dolt-3 with certified concentrations using AAS Perkin Elmer A Analyst 800 (n = 3).

| Heavy metals | CRM | Certified values (µg/g) | Measured values (µg/g) | Recovery (%) |
|---|---|---|---|---|
| Cu | Soil-5 | 77.1 ± 4.7 | 75.62 ± 5.0 | 99 |
| | DOLT-3 | 31.2 ± 1.0 | 29.8 ± 2.7 | 96 |
| Zn | Soil-5 | 368.0 ± 8.20 | 362.9 ± 15.6 | 100 |
| | DOLT-3 | 86.6 ± 2.4 | 83.8 ± 3.5 | 97 |
| Pb | Soil-5 | 129.0 ± 26.0 | 124.8 ± 22.0 | 97 |
| | DOLT-3 | 0.32 ± 0.05 | 0.38 ± 0.09 | 109 |
| Cd | Soil-5 | 1.5 ± 0.0 | 1.56 ± 0.03 | 101 |
| | DOLT-3 | 19.4 ± 0.6 | 20.5 ± 0.4 | 105 |
| Ni | Soil-5 | 13 ± 0.00 | 12.85 ± 1.02 | 107 |
| | DOLT-3 | 2.72 ± 0.35 | 2.77 ± 0.74 | 101.8 |

1) Index of Geo accumulation (I*geo*)

The I*geo* accumulation index is a quantitative measure of the degree of pollution in aquatic sediments [35]. I*geo*, proposed by [36], calculate metal concentration in sediments by comparing current concentration with pre-industrial, undisturbed or crustal sediments levels, was used to quantitatively estimate the metal pollution status of sediments. Enrichment of metal concentration above baseline concentrations was calculated using (Equation (1)) the method proposed by [37], termed the geo accumulation index (I*geo*). I*geo* is mathematically expressed as:

$$\text{I}geo = \text{Log}_2\left(\text{Cn}/1.5\text{Bn}\right) \quad (1)$$

where Cn is the measured concentration of the examined metal "n" in the sediment and Bn is the geochemical background concentration of the metal "n". The factor 1.5 was used to analyse natural fluctuations in the content of a given substance in the environment and to detect very small anthropogenic influences [38]. The factor 1.5 was introduced in the equation to minimise the effect of possible variations in the background values, which may be attributed to lithogenic variations in soils. I*geo* assesses the metal pollution in terms of seven enrichment classes (0 to 6) ranging from background concentration to very heavily polluted, as shown in Table 4.

2) Enrichment Factor (EF)

Enrichment factor is a common approach to estimate how much the sediment is impacted (naturally and anthropogenically) with heavy metal [40], and is used to identify the precise origin(s) of the elements [41] [42]. Enrichment factor is computed relative to the abundance of species in source material to that found in the earth crust [43]. EF is generally defined as the observed metal to Fe ratio in a sample divided by the background metal/Fe ratio. EF is calculated using

Table 3. Background concentrations of sediment used in the calculation of pollution indices.

| Background concentrations | Cu | Zn | Pb | Cd | Ni | Fe | References |
|---|---|---|---|---|---|---|---|
| World average shale | 45 | 95 | 20 | 0.30 | 68 | 4.70 | [34] |
| Lowest concentration in the present study | 9.66 | 66.61 | 2.57 | 0.58 | 11.37 | 1.89 | Present study |

Table 4. I*geo* classes in relation to sediment quality [39].

| I*geo* | I*geo* classes | Pollution intensity |
|---|---|---|
| <0 | 0 | Unpolluted |
| 0 - 1 | 1 | Unpolluted to moderately polluted |
| 1 - 2 | 2 | Moderately polluted |
| 2 - 3 | 3 | Moderately to strongly polluted |
| 3 - 4 | 4 | Strongly polluted |
| 4 - 5 | 5 | Strongly to very strongly polluted |
| >5 | 6 | Very strongly polluted |

(Equation (2)) as follows;

$$EF = (Cn/Fe)\ \text{sample} / (Cn/Fe)\ \text{background} \quad (2)$$

where: Cn is the concentration of element "n".

To calculate the EF of a metal, the normalizer and the background levels of the metals should be determined. The geochemical normalization was obtained by using Fe as the reference element. Because Fe is one of the most abundant elements on Earth and usually poses no contamination concern, it is the frequent choice for normalization purposes. EF classes and sediment quality were shown in Table 5.

3) Pollution Load Index (PLI)

Pollution Load Index (PLI) is defined as an empirical index which provides a simple, comparative means for assessing the level of heavy metals pollution between different sites and at different times. It was employed by [45] to assess the extent of pollution by metals in estuarine sediments [46]. The PLI for a single site is the nth root of n number of multiplied contamination factor (CF) values as calculated using Equation (3).

$$\text{PLI for a site} = (CF1 \times CF2 \times \cdots \times CFn) 1/n \quad (3)$$

where: CF = C metal concentration/C background concentration of the same metal.

A PLI value of zero indicates perfection, a value of one that only baseline levels of pollutants are present and values above one would indicate progressive deterioration of the site and estuarine quality [45]. The PLI value of >1 indicates polluted, whereas PLI value < 1 indicates no pollution [47] [48].

## 2.4. Statistical Analyses

All statistical analyses of data were carried out using SPSS statistical package programs version 17. Data were tested for the basic assumptions of normality and homogeneity of variance in exploratory data analysis in SPSS 17. ANOVA was calculated, post host comparison was made using Duncan's multiple range test at 0.05 confidence level. The strength and significance of the relationships between metal concentrations Cu, Zn, Pb, Cd and Ni in the sediments were

Table 5. EF classes in relation to sediment quality [44].

| EF Classes | Sediment quality |
|---|---|
| <1 | Indicates no enrichment |
| <3 | Presence of minor enrichment |
| 3 - 5 | Moderate enrichment |
| 5 - 10 | Moderately to severe enrichment |
| 10 - 25 | Severe enrichment |
| 25 - 50 | Very severe enrichment |
| >50 | Extremely severe enrichment |

tested by the Pearson's correlation coefficients (r).

## 3. Results and Discussion

### 3.1. Results

#### 3.1.1. Heavy Metals Concentrations in the Surface Sediments

The means heavy metals concentrations in the surface sediments were presented in Table 6. The highest concentrations of Cu, Zn and Ni were observed at sampling site 2 as 65.39 ± 0.70, 442.19 ± 4.13 and 29.25 ± 0.08 respectively and that of Pb and Cd were observed at sampling site 5 as 53.73 ± 0.18 and 1.65 ± 0.11 respectively. Statistical analysis showed that metal concentrations Cu, Zn, Pb, Cd and Ni were significantly different ($p < 0.05$) between the sampling sites. Iron was used as reference element for the normalization purposes in the calculation of EF.

#### 3.1.2. Particle Size Analysis of the Surface Sediments

The result of particle size analysis was presented in Table 7. Four classes of grain size sediments were identified; sandy clay loam, silt clay, clay and sandy loam. The result revealed higher percentage of sand (52.08% - 72.66%) at sampling sites 1, 2, 5 and 6 and the sediments were classified as sandy clay loam. High percentage of clay particles (75.81%) dominated surface sediment from sampling site 4 while sampling site 3 showed mixture of clay (47.74%) and silt (40.09%) sediments at these sites and were classified as clay and silty clay respectively.

Table 6. Mean heavy metal concentrations (μg/gd/w ± SE, except Fe in percentage %) in the surface sediments.

| Sampling sites | Cu | Zn | Pb | Cd | Ni | Fe |
|---|---|---|---|---|---|---|
| 1 | 34.79 ± 0.19 | 60.83 ± 1.74 | 27.78 ± 1.41 | 1.25 ± 0.04 | 13.11 ± 0.16 | 1.98 ± 0.00 |
| 2 | 65.39 ± 0.70 | 442.19 ± 4.13 | 29.97 ± 0.36 | 1.24 ± 0.08 | 29.25 ± 0.08 | 2.78 ± 0.04 |
| 3 | 35.48 ± 0.64 | 256.50 ± 2.40 | 34.22 ± 0.23 | 1.37 ± 0.06 | 26.42 ± 0.24 | 2.14 ± 0.01 |
| 4 | 12.79 ± 0.57 | 75.38 ± 1.63 | 8.46 ± 0.83 | 0.60 ± 0.05 | 10.09 ± 0.54 | 1.89 ± 0.00 |
| 5 | 37.64 ± 0.95 | 241.87 ± 0.78 | 53.73 ± 0.18 | 1.65 ± 0.11 | 21.18 ± 0.48 | 3.31 ± 0.09 |
| 6 | 13.90 ± 0.03 | 117.38 ± 0.99 | 28.28 ± 0.11 | 1.40 ± 0.14 | 11.89 ± 0.29 | 3.67 ± 0.02 |

Table 7. Percentage grain size distribution and type of surface sediments.

| Sampling sites | Clay (%) | Silt (%) | Sand (%) | Sediment type |
|---|---|---|---|---|
| 1 | 22.67 | 11.68 | 65.57 | sandy clay loam |
| 2 | 24.9 | 22.88 | 52.08 | sandy clay loam |
| 3 | 47.74 | 40.09 | 12.08 | silty clay |
| 4 | 75.81 | 23.94 | 0.22 | clay |
| 5 | 17.44 | 9.71 | 72.66 | sandy loam |
| 6 | 22.73 | 19.73 | 57.41 | sandy clay loam |

### 3.1.3. Correlation between Particle Size Distribution and Total Metals

The correlations coefficients based on the relationship between particle size distribution and total heavy metals (Cu, Zn, Pb, Cd and Ni) were presented in Table 8. The Pearson's correlation analysis shows negative correlations between clay particle with; TLCu ($r = -0.564$), TLCd ($r = -0.528$) at $p < 0.05$ and TLPb ($r = -0.792$; $p < 0.01$). Sand particle shows positive correlations with TLPb ($r = 0.671$; $p < 0.01$) and TLCd (0.479; $p < 0.05$). Negative correlations were observed between fine grain particle with TLPb ($r = -0.673$; $p < 0.01$) and TLCd ($r = -0.489$; $p < 0.05$) while non-significant correlations were observed between silt particle with all the six heavy metals.

### 3.1.4. Correlation between Heavy Metals Concentrations

Pearson's correlation analysis was used to test the relationship between the heavy metals analysed in the present study. The correlation matrix showed that Cu, Zn, Pb, Cd and Ni were correlated with each other, showing a strong positive associations ($r = 0.753$ - $0.805$; $p < 0.01$) and a positive correlations of lesser confidence (0.519 - 0.588; $p < 0.05$) among the heavy metals.

Non-significant correlations ($p > 0.05$) were observed between Cu and Cd ($r = 0.312$), Ni also shows a non-significant correlation with Pb ($r = 0.414$) and Cd ($r = 0.074$) at $p > 0.05$ (Table 9).

### 3.1.5. Index of Geo Accumulation (*Igeo*)

The means I*geo* based on world average shale and lowest concentration in the present study were presented in Table 10. The I*geo* index based on world average shale [34], showed that 100% of Cu, Zn, Pb, Ni and 96% of Cd falls into class 1 (unpolluted), and 4% of Cd falls into class 2 (moderately polluted).

The I*geo* index calculated based on the lowest concentration showed 100% of

Table 8. Correlations coefficients between particle size distribution and total heavy metals.

| Particle size/Total heavy metals | TLCu | TLZn | TLPb | TLCd | TLNi |
|---|---|---|---|---|---|
| Clay | −0.564* | −0.236 | −0.792** | −0.528* | −0.277 |
| Silt | −0.048 | 0.38 | −0.202 | −0.247 | 0.392 |
| Sand | 0.441 | 0.025 | 0.671** | 0.479* | 0.057 |
| Finegrain | −0.445 | −0.04 | −0.673** | −0.489* | −0.067 |

*Correlation is significant at the 0.05 level (2 tailed). **Correlation is significant at the 0.01 level (2-tailed).

Table 9. Relationships between heavy metals concentrations in the surface sediments.

| Metals | Cu | Zn | Pb | Cd | Ni |
|---|---|---|---|---|---|
| Zn | 0.576* | 1 | | | |
| Pb | 0.558* | 0.588* | 1 | | |
| Cd | 0.312 | 0.519* | 0.753** | 1 | |
| Ni | 0.805** | 0.799** | 0.414 | 0.074 | 1 |

**Correlation is significant at the 0.01 level (2-tailed). *Correlation is significant at the 0.05 level (2-tailed).

**Table 10.** Means I*geo* Index and their classes indicated by superscript numbers.

| Sampling sites | Background Concentrations | I*geo* values and classes | | | | |
|---|---|---|---|---|---|---|
| | | Cu | Zn | Pb | Cd | Ni |
| 1 | World average shale | 0.13[a] | 0.14[a] | 0.27[a] | 0.73[a] | 0.05[a] |
| 2 | | 0.32[a] | 0.85[a] | 0.29[a] | 0.86[a] | 0.06[a] |
| 3 | | 0.19[a] | 0.51[a] | 0.41[a] | 0.76[a] | 0.09[a] |
| 4 | | 0.06[a] | 0.14[a] | 0.10[a] | 0.39[a] | 0.10[a] |
| 5 | | 0.17[a] | 0.52[a] | 0.61[a] | 1.90[b] | 0.07[a] |
| 6 | | 0.07[a] | 0.26[a] | 0.33[a] | 0.84[a] | 0.04[a] |
| 1 | Present study | 0.59[a] | 0.20[a] | 2.07[c] | 0.38[a] | 0.27[a] |
| 2 | | 1.48[b] | 1.21[b] | 2.22[c] | 0.45[a] | 0.62[a] |
| 3 | | 0.87[a] | 0.73[a] | 3.15[d] | 0.39[a] | 0.55[a] |
| 4 | | 0.28[a] | 0.20[a] | 0.79[a] | 0.20[a] | 0.20[a] |
| 5 | | 0.78[a] | 0.74[a] | 4.75[e] | 0.46[a] | 0.40[a] |
| 6 | | 0.32[a] | 0.37[a] | 2.56[c] | 0.43[a] | 0.24[a] |

I*geo*: [a]unpolluted to moderately polluted; [b]moderately polluted; [c]strongly to moderately polluted; [d]strongly polluted; [e]strongly to very strongly polluted.

Cd, Ni and 96% of Cu, Zn falls into class 1 (unpolluted). Cu and Zn were found to be moderately polluted (class 2) at sampling g site 2. The I*geo* index of Pb shows that 50% of Pb falls into class 3 (moderately to strongly polluted) and 16.66% of Pb fall into; class 5 (strongly to very strongly polluted), class 4 (strongly polluted) and class 1 (unpolluted) at sampling sites 5, 3 and 4 respectively.

### 3.1.6. Enrichment Factor (EF)

An enrichment factor was used to differentiate metals from anthropogenic (non-crustal) and geogenic (crustal) sources and to further evaluate the magnitude of contamination in the environment (Feng *et al.*, 2004). Table 11 shows mean EF values of heavy metals. The mean EF of Cu mostly shows minor enrichment (EF < 3), and moderately severe enrichment (EF = 5.02) at sampling site 2. The EF value for Cu at sampling sites 4 and 6 showed no enrichment (EF < 1) in respect to world average shale, while minor enrichment (EF = 1.40) and no enrichment (EF < 1) were observed at sampling sites 4 and 6 respectively for the EF of Cu calculated from the lowest concentration. Ni showed no enrichment (EF < 1) at all the sampling sites in respect to world average shale except at sampling site 3 were minor enrichment (EF < 3 was observed in respect to the lowest concentration. Minor enrichment (EF < 3) of about 67% was observed for Ni from the lowest concentration in the present study. The EF values for Zn showed no enrichment (EF < 1) to moderately severe enrichment (EF = 5 - 10). EF values of Pb showed minor enrichment (EF < 3) to severe enrichment (EF = 10 - 25). The mean EF values of Cd calculated based on lowest concentration showed minor enrichment (EF < 3) at all the sampling sites. The highest EF values

Table 11. Means EF of heavy metals and their classes indicated by superscript letters.

| Sampling sites | Background Concentrations | Enrichment Factor (EF) | | | | |
|---|---|---|---|---|---|---|
| | | Cu | Zn | Pb | Cd | Ni |
| 1 | World average shale | 1.50[b] | 1.68[b] | 3.16[c] | 9.23[d] | 0.54[a] |
| 2 | | 2.68[b] | 7.19[d] | 2.41[b] | 7.74[d] | 0.87[a] |
| 3 | | 2.06[b] | 5.62[d] | 4.46[c] | 8.91[d] | 1.00[b] |
| 4 | | 0.75[a] | 1.74[b] | 1.25[b] | 5.15[d] | 0.42[a] |
| 5 | | 1.20[b] | 3.70[c] | 4.33[c] | 6.79[d] | 0.47[a] |
| 6 | | 0.44[a] | 1.64[b] | 2.10[b] | 5.72[d] | 0.26[a] |
| 1 | Present study | 2.81[b] | 0.96[a] | 9.89[d] | 1.79[b] | 1.29[b] |
| 2 | | 5.02[d] | 4.12[c] | 7.55[d] | 1.50[b] | 2.09[b] |
| 3 | | 3.85[c] | 3.23[c] | 13.96[e] | 1.72[b] | 2.41[b] |
| 4 | | 1.40[b] | 1.00[b] | 3.93[c] | 1.00[b] | 1.00[b] |
| 5 | | 2.24[b] | 2.12[b] | 13.55[e] | 1.31[b] | 1.13[b] |
| 6 | | 0.82[a] | 0.94[a] | 6.59[d] | 1.11[b] | 0.61[a] |

EF: [a]no enrichment; [b]minor enrichment; [c]moderate enrichment; [d]moderately severe enrichment; [e]severe enrichment; [f]very severe enrichment.

Table 12. Values of pollution load index in the surface sediments.

| Sampling sites | World average shale | Lowest concentration in the present study |
|---|---|---|
| 1 | 0.89 | 2.45 |
| 2 | 1.78 | 4.92 |
| 3 | 1.45 | 4.01 |
| 4 | 0.49 | 1.35 |
| 5 | 1.58 | 4.35 |
| 6 | 0.85 | 2.34 |

PLI: PLI value of >1 indicates polluted, whereas PLI value < 1 indicates no pollution.

for Cu, Zn, Pb, Cd and Ni were observed at sampling sites 2, 2, 3, 1 and 3 respectively for the world average shale and lowest concentration of the present study.

#### 3.1.7. Pollution Load Index (PLI)

The calculated PLI values ranged from 0.45 - 1.78 at sampling sites 3 and 2 respectively for world average shale and 1.35 - 4.92 at sampling sites 4 and 2 respectively in respect to lowest concentration in the present study (Table 12). The order of PLI from highest to lowest values was; 2 > 5 > 3 > 1 > 6 > 4 for world average shale and lowest concentration in the present study.

### 3.2. Discussion

Grain size is an important factor to evaluate heavy metals concentration in the sediment [49]. The particle size analysis of the surface sediments showed that

sand and clay content appeared to be the principal sediment types at sampling sites 1, 2, 5 and 6 while sampling sites 3 and 4 contained higher mixture of clay and silt content with low sand substrates. Many studies have shown that mangrove sediments act as a trap for chemical contaminants because such sediments contain high percentage of silt and clay that cause an increase in the metals adsorption [50] [51] [52] [53] and indicated that heavy metal concentrations decrease with the increase of particle size, and the highest content levels occur in the finest fractions [44] [54] [55] [56]. High metals concentrations were observed at sampling sites with high content of sand and clay particles. Coarser particles may as well show higher concentration of heavy metals than finer ones in some instances, and the presence of coarse particles is possibly the reason for higher metal content [57] [58].

Pearson's correlation analysis was applied to test the relationships among heavy metals in the surface sediments of the west coast of Peninsular Malaysia. The results of correlation matrix showed that Cu, Zn, Pb, Cd and Ni were correlated significantly with each other except for few cases (Cu and Cd; Ni and Pb; and Ni and Cd) in the surface sediments. This finding is in good agreement with the studies conducted by [59] who reported significant correlation ($p < 0.01$) between Cd and Pb from marine sediments of Jinzhou Bay, China and that of [60] between Cu and Zn, Cu and Pb, and Zn and Pb from urban soils in Nanjing, China.

The strong positive correlation coefficients between Cu and Ni ($r = 0.805$), Zn and Ni ($r = 0.799$) and Pb and Cd ($r = 0.753$) was significantly observed suggesting some relationship between the metals and indicating their similar source of origin [59] [61] [62]. The low correlation coefficient of Cu and Cd, Pb and Ni, and Cd and Ni in the sediments suggests these metals may originate from different sources or that they might have different sediment deposition characteristics [63].

The I*geo* indexing approach was used to quantify the degree of anthropogenic contamination, and to compare the different metals in aquatic sediments [36] [37] [64], by comparing current concentrations with pre-industrial levels [36]. The result of geo accumulation index (I*geo*) based on world average shale shows that all the sampling sites were considered as unpolluted to moderately polluted with Cu, Zn, Pb, and Ni. Cadmium was considered moderately polluted at sampling site 5 while the remaining 5 sampling sites were considered as unpolluted with Cd.

The calculated I*geo* values based on the lowest concentration in the present study shows that all the sampling sites were unpolluted with Cu, Zn, Cd and Ni except sampling site 2 which was moderately polluted with Cu and Zn. The pollution intensity of Pb shows that most of the sampling sites were moderately to strongly polluted with Pb while sampling site 5 was considered strongly to very strongly polluted with Pb, sampling site 4 was the only site considered unpolluted with lead. Among the metals analysed, Pb shows the highest I*geo* values at

the entire sampling site which might suggest Pb in the sediment was most significantly impacted by anthropogenic sources. The I*geo* values for Pb might be associated with fuel used in jetties and industries. This is in agreement with [65] that, the presence of Pb in sediment cores might have been originated from the anthropogenic fly-ashes derived from the combustion of fossil fuels in industries, boat, ship and other vehicles. According to the calculated I*geo* values, five sampling sites were strongly polluted with Pb, sampling site 2 was moderately polluted with Cu and Zn while sampling site 5 was moderately polluted with Cd.

In general, the index of geo accumulation (I*geo*) indicates that the surface sediments collected from the sampling sites were unpolluted to moderately polluted (Class 1) to strongly to very strongly polluted (Class 5). The I*geo* values for Cu, Zn, Pb and Ni calculated from the lowest concentration found in the present study were higher than those calculated from the world average shale which may suggest anthropogenic input of these metals in the sediment.

In order to distinguish anthropogenic pollutants from natural content in the sediment, enrichment factors (EF) were calculated. The enrichment factors in the surface sediments varied between no enrichment to severe enrichment depending on the sampling sites and heavy metal. The EF values for Cu showed no enrichment to moderately severe enrichment and the EF for Zn and Pb show no enrichment to severe enrichment. The highest enrichment of Cu and Zn were mostly found at sampling site 2, and that of Pb was found at sampling site 3 as 13.96 which might suggest high anthropogenic activities as a result of domestic and industrial input at these sampling sites. EFs greater than 10 are considered to be non-crustal source [66] [67].

The EF values of Cd showed minor enrichment to moderately severe enrichment while the EF calculated from the lowest concentration found in the present study showed minor enrichment of Cd with highest mean value at sampling site 1. Nickel shows lowest EF values among the metals studied, the EF values indicated no enrichment to minor enrichment. The highest mean EF value of Ni was found at sampling site 3, which was associated with influx of domestic sewage and industrial activities in the area. All the EF values calculated for Cu, Pb and Ni using the lowest concentrations found in the present study were higher than the EF values calculated from the world average shale background which indicates these metals were enriched at the sampling sites. The EF values of Zn and Cd were all higher from the world average shale when compared with EF values from the lowest concentrations in the present study. The concentration of crustal elements may vary by location [68] and could result in overestimation or underestimation of the EF [69].

The PLI values calculated based on world average shale and the lowest concentration found in the present study indicates strong signs of pollution deterioration by the metals studied at all the sampling sites. The PLI value with respect to world average shale indicates that the metals were within the baseline level at sampling sites 1, 6, and 4 which implied no appreciable input from anthropo-

genic sources. The lowest concentration found in the present study shows higher PLI values than the world average shale background concentrations, clearly demonstrating anthropogenic contribution of metals. The highest degree of pollution based on PLI value was observed at sampling site 2 followed by sampling site 5 and lowest at sampling site 4. The order of sampling sites from highest to lowest PLI values was; 2 > 5 > 3 > 1 > 6 > 4.

## 4. Conclusion

The results obtained from this study indicate heavy metals contamination in the study area, particularly Pb. Heavy metals concentrations in the sediments varied significantly among the sampling sites. Higher concentrations of heavy metals were generally observed at sampling sites close to anthropogenic inputs from domestic, industrial, shipping or other sources of pollution. The contamination of Pb in the study area could be associated with fuel and oil lubricants used in jetties and industries which might have been originated from paint flakes from boats, sewage sludge and anthropogenic fly-ashes derived from the combustion of fossil fuels in industries, boat, ship and other vehicles. Hence, management practices, monitoring and Government policy should be employed for regulating contamination of this coastal environment by controlling effluent discharge from anthropogenic sources which is required to protect the marine ecosystem from deterioration. Further study on the physico-chemical parameters of water and sediments that could affect bioavailability, accumulation and toxicity of these metals in fishes, shell fishes and other aquatic resources should further be investigated.

## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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