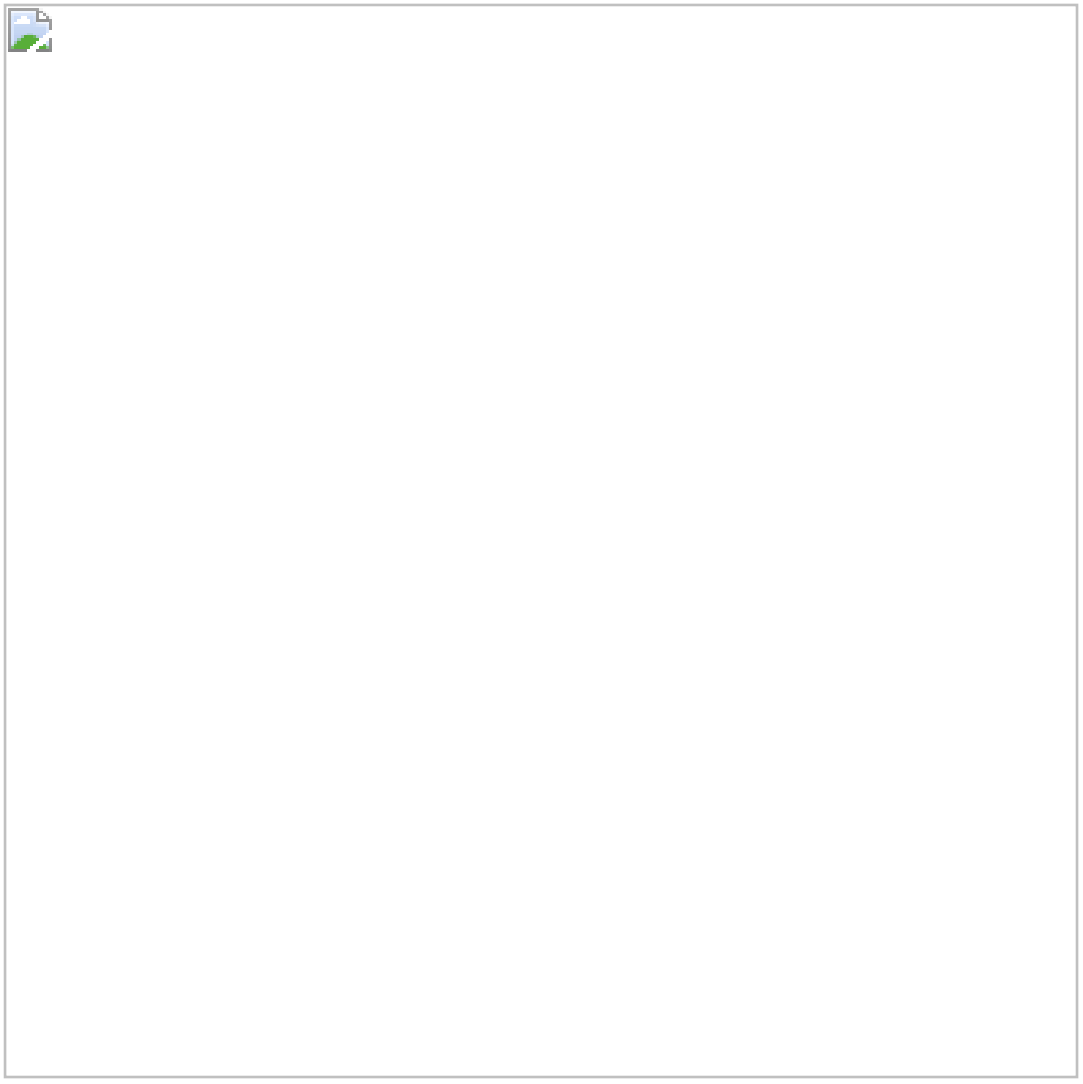

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Authors: Khan, Waseem Razzaq, Zulkifli, Syaizwan Zahmir, bin Mohamad Kasim, Mohamad Roslan, Zimmer, Martin, Pazi, Ahmad Mustapha, et al.

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*Research Article*

# Risk Assessment of Heavy Metal Concentrations in Sediments of Matang Mangrove Forest Reserve

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**Waseem Razzaq Khan[1,2], Syaizwan Zahmir Zulkifli[3,4], Mohamad Roslan bin Mohamad Kasim[1], Martin Zimmer[5], Ahmad Mustapha Pazi[1], Nur Amira Kamrudin[3,4], Fahad Rasheed[6], Zikria Zafar[6], Roslan Mostapa[7], and M. Nazre[1]**

## Abstract

Matang Mangrove Forest Reserve (MMFR) is one of the most productive and managed forests in the world. On the other hand, it has become a concern whether MMFR is being degraded as a result of exposure to industrial pollution. Industries located around MMFR dispose effluents contaminated by heavy metals. This study was conducted to analyze heavy metal contamination and risk assessment status in MMFR sediments. Sediment samples from six compartments were collected based on age and location of the mangrove plantation. Total metal digestion and modified sequential extraction were performed to estimate the concentration of heavy metals. Based on the estimation, risk assessment code, geo-accumulation index, pollution load index, and contamination factor were computed to classify the compartments according to their contamination and pollution levels. Organic matter and sediment texture (silt, clay, and sand content) were also analyzed to find its correlation with heavy metals. According to the results, high concentrations for Copper, Nickel, and Cadmium were observed in Compartment 42, while Compartment 18 and Compartment 74 showed higher concentrations for Zinc and Lead. Heavy metals showed weak positive correlation with clay and silt, but weak negative correlation with sand. For organic matter, only Zinc showed statistically significant but weak negative correlation. Risk assessment code, geo-accumulation index, pollution load index, and contamination factor categorized the compartments into unpolluted to moderately polluted. Based on the study outcomes, it can be concluded that MMFR, although acquiring industrial discharge, is not with a high risk of heavy metal contamination.

## Keywords

MMFR, heavy metals, risk assessment code, geo-accumulation index, contamination

[1]Faculty of Forestry and Environment, Universiti Putra Malaysia
[2]Institut Ekosains Borneo, Universiti Putra Malaysia Kampus Bintulu
[3]Department of Biology, Faculty of Science, Universiti Putra Malaysia
[4]International Institute of Aquaculture and Aquatic Sciences, Universiti Putra Malaysia
[5]Leibniz-Centre for Tropical Marine Research (ZMT), Fahrenheitstraße 6, Bremen, Germany
[6]Faculty 02 Biology/Chemistry, University Bremen
[7]Member of IUCN-SSC Mangrove Specialist Group, Gland, Switzerland

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**Corresponding Authors:**
Waseem Razzaq Khan, Department of Forest Management, Faculty of Forestry, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.
Email: waseemjatoi4@gmail.com

M. Nazre, Department of Forest Management, Faculty of Forestry, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.
Email: nazre@upm.edu.my

Mangroves are considered as one of most productive ecosystems in tropical and subtropical areas (Marchand et al., 2011). They can be classified as a transition zone between marine and freshwater environments (Udechukwu et al., 2015). In an estuarine environment, mangroves play an essential role in the food web as a primary producer to provide food for the local fauna. Despite all its ecological services, mangrove ecosystems are exploited by agriculture practices, land encroachment, and aquaculture practices (Macfarlane et al., 2007).

Role of mangroves in sedimentation process is important as they trap suspended particles and organic matters (OMs; Furukawa et al., 1997). Aquatic biological systems are also polluted by secondary sources in the form of heavy metals bound in OM of sediments (Bi et al., 2017; Chakraborty et al., 2015; Jafarabadi et al., 2017). Heavy metals uptake in mangrove trees is stimulated by their physiological and biochemical processes, owing to which mangrove trees are called "chemical reactors" (de Silva et al., 2006, p. 1). Meanwhile, discharge from industries, agriculture, and mining are primary contributors to heavy metal contamination in mangrove ecosystems (Marchand et al., 2011). Decomposition and littering also play a relevant role (Morales-Munoz et al., 2005).

Heavy metals are nonbiodegradable pollutants and thus bioaccumulate over time (Okocha & Adedeji, 2012). Chemical factors such as reactivity of metals, weathering, and composition of sediments (i.e., hydroxides/oxides, carbonates, silicates, and sulphates; Zhu et al., 2006) influence the concentration, bioavailability, and distribution of metals (Chakraborty et al., 2014; Liu et al., 2017). In addition, physical factors such as soil grain size and soil texture also effect heavy metal concentration (Maslennikova et al., 2012). Sediments have either fine or coarse particles. Coarser sediments usually harbor less heavy metals as compared with finer sediments. This is due to the larger ratio of surface area to volume for adsorption. In some cases, coarse sediments/particles also have high metals concentration (Chakraborty et al., 2014).

Matang Mangrove Forest Reserve (MMFR) is situated in Perak, Peninsular Malaysia. It expands over approximately 40,466 ha. MMFR is a productive mangrove forest surrounded by charcoal factories, fish net industries, aquaculture farms, and residential settlements. The risk evaluation of heavy metal contamination in the sediments of selected compartments in MMFR was investigated in this study. Furthermore, various contamination indices such as pollution load index (PLI), geo-accumulation index ($I_{geo}$), contamination factor (CF), and risk assessment codes (RACs) were computed to classify the sampled compartments according to their pollution levels.

## Methods

### Study Site

MMFR (4°45′ N, 100°35′ E), for management purpose, was divided into various compartments (Figure 1). In this study, six compartments (18, 31, 42, 55, 71, and 74) were selected for sediment collection (Table 1). Moreover, compartment selection was based on locality and three age groups, which were 15-year-old, 25-year-old, and virgin jungle reserve (VJR). The VJR compartments were those which have remained untouched for more than 80 years. Each age-group was represented by two selected compartments. Compartment 18, Compartment 31, and Compartment 42 were located near highly populated zones. Whereas, Compartment 55, Compartment 71, and Compartment 74 were not surrounded by any urbanization. Five samples were randomly taken from all the compartments, making up a total of 30 samples. The stream surface area was selected for the wet sediment test sample collection. To prevent samples from contamination, they were stored in an ice box and enclosed in ziplock plastic bags. Sediment samples were transported to the soil laboratory in the Faculty of Forestry, Universiti Putra Malaysia and were stored at −10°C for 24 hr. Samples were oven dried at 60 °C for 3 days and then subjected to a soil grinder and sieved through a 63 μm sieve.

**Table 1.** Overview of Samples Collected Within Each Compartment.

| Compartments | No. of sediment samples | Compartment age | Salinity (ppt) | pH | River name |
|---|---|---|---|---|---|
| 18 | 5 | 15 years | 20.3 | 4.6 | Menangis |
| 31 | 5 | 15 years | 18.9 | 5 | Sanga Besar |
| 71 | 5 | 25 years | 18.8 | 6.2 | Mongokok |
| 74 | 5 | 25 years | 23.2 | 4.5 | Tiram Dilam |
| 42 | 5 | VJR | 20.7 | 4.9 | Menangis |
| 55 | 5 | VJR | 19.8 | 4.8 | Trong |

*Note.* VJR = virgin jungle reserve.

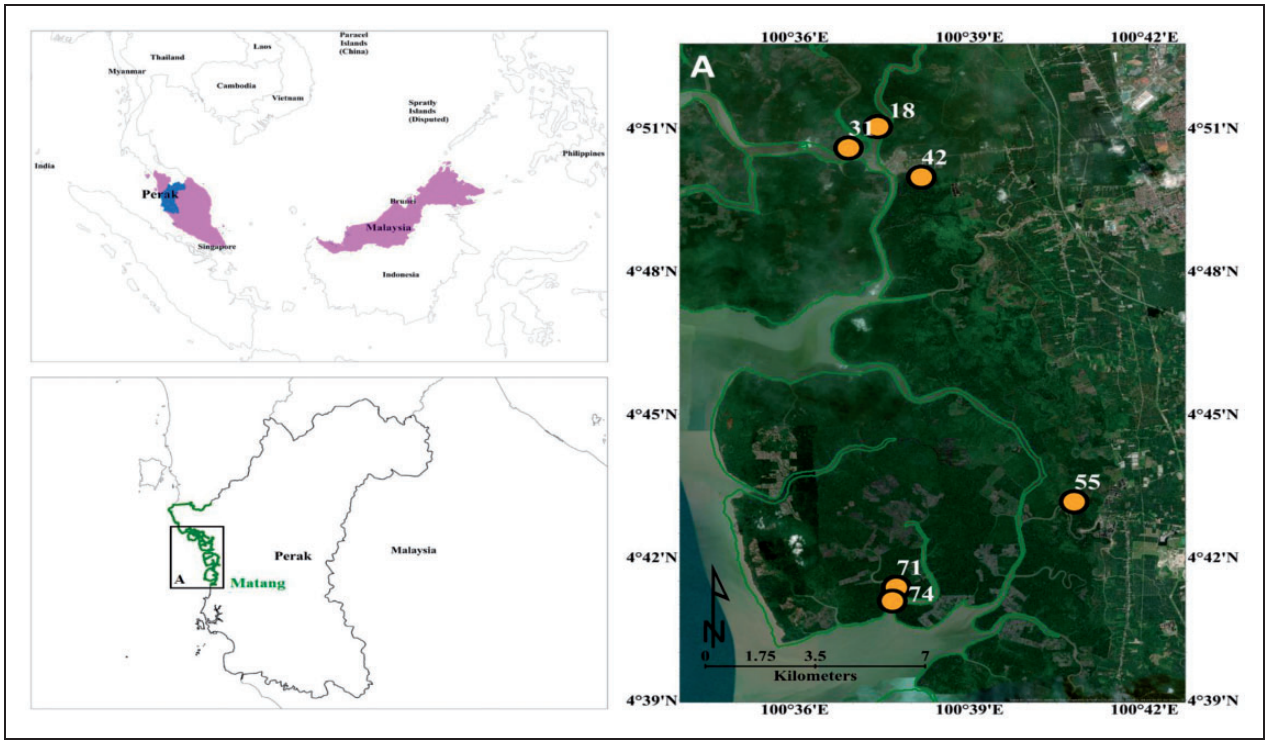


**Figure 1.** Study Site: Matang Mangrove Forest Reserve (A). Compartments for sampling are shown (yellow dots) by overlaying on imagery from Google Earth (Source: Google Earth Pro V 7.3.2.5776, April 9 2018, lat 4.72224410 lon 100.6738670, Eye alt 25.72 km, Maxar Technologies, CNES/Airbus).

## *Analytical Techniques*

*Total Metal Digestion.* Aqua regia-digestion method was adopted to estimate the total metal content in the sediments (Abubakar et al., 2018). Dried sediment samples of 1 g were incubated in 10 ml solution of concentrated nitric acid ($HNO_3$) AnalaR grade British Drug House (BDH) 69%, and perchloric acid ($HClO_4$) AnalaR grade BDH 60%, in a ratio of 4:1 on a hot block digester at 40 °C for 1 hr. Temperature was gradually increased to 140 °C for 3 hr. The samples were then cooled at room temperature and 40 ml of distilled water was added to make the solution reach 50 ml. This solution was filtered by using Whatman No. 1 filter paper and collected in clean plastic vials (Andrews et al., 1989). Since the total metal content could not provide accurate information on the distribution and bioavailability of heavy metals (Abubakar et al., 2018; Swati & Hait, 2017), the fractional methodology was adopted to assess bioavailability, mobility, and distribution of the investigated metals (Badri & Aston, 1983; Tessier & Campbell, 1987).

*Sequential Metal Extraction Procedure.* In Figure 2, the sequential metal extraction process is comprised of four steps (Badri & Aston, 1983; Tessier & Campbell, 1987). After each step, the obtained liquid was washed with 20 ml of doubled distilled water and filtered through a Whatman No. 1 filter paper. As a result, the liquid fraction and filtered out sediment residues were obtained. The residue from each step was dried and used in the subsequent step. To determine metal concentrations, filtrates from each phase were analyzed through the atomic absorption spectrometer (AAS Shimazdu AA6300).

*Total Organic Matter.* Loss on ignition method was applied to measure the total organic matter (Barillé-Boyer et al., 2003; Jia-Ping et al., 2013).

*Soil Texture.* For determination of soil texture, the pipette method was used. For the sediment classification, a procedure proposed by the United States Department for Agriculture was adopted (Skaggs et al., 2001).

*Risk Assessment Code.* Bioavailability and risk of heavy metals can be expressed by metal bonding strength in an aquatic system. To determine the RAC, Fractions 1 and 2 from the sequential extraction process were analyzed (González et al., 2013). Table 5 shows the criteria for RAC with results.

*Geoaccumulation Index.* $I_{geo}$ was developed by Muller (1969) to evaluate metal contamination in sediments. It is a comparison between current metal concentrations

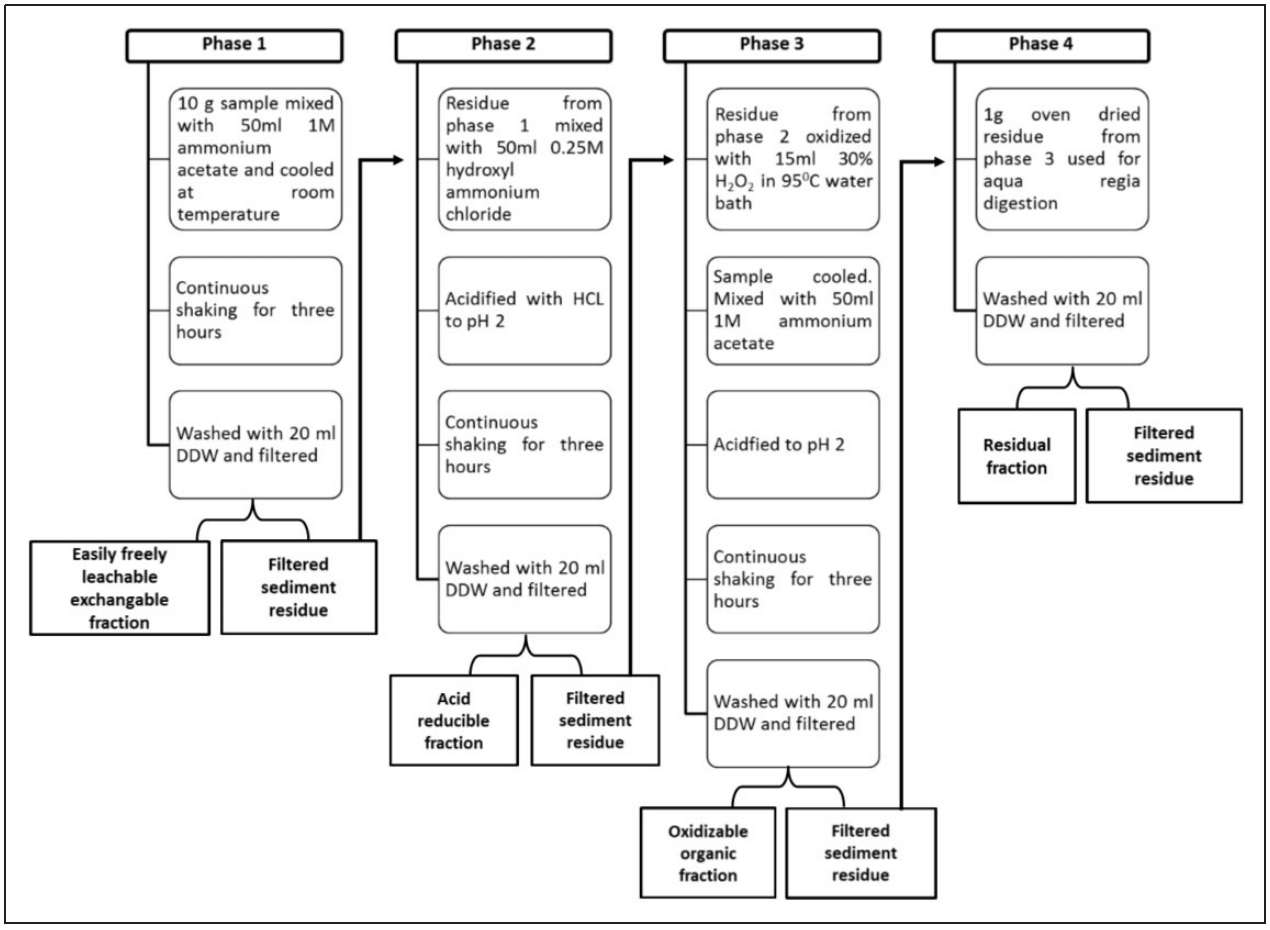


**Figure 2.** Sequential Metal Extraction Steps.

and unpolluted or preindustrial time. Global average shale, as proposed by Turekian and Wedepohl (1961), was used as reference in this study, since there were no data present for preindustrial time

$$I_{\text{geo}} = \log_2(C_n/1.5B_n),$$

where $I_{\text{geo}}$ = geoaccumulation, $C_n$ = concentration of the metal, 1.5 = correction factor, $B_n$ = background value of the metal (average shale in the case study).

$I_{\text{geo}}$ is categorized between 1 and 6 ($I_{\text{geo}} \leq 0$ = unpolluted, $I_{\text{geo}} < 1$ unpolluted to moderately polluted, $I_{\text{geo}} < 2$ = moderately polluted, $I_{\text{geo}} < 3$ moderately to strongly polluted, $I_{\text{geo}} < 4$ strongly polluted, $I_{\text{geo}} < 5$ strongly to very strongly polluted, $I_{\text{geo}} > 5$ very strongly polluted; Çevik et al., 2009).

*Contamination Factor.* The ratio of observed metal concentrations in sediments to the background value reported by Turekian and Wedepohl (1961) provided information on the extent of metal contamination. Background values for Copper (Cu), Lead (Pb), Zinc (Zn), Nickel (Ni), and Cadmium (Cd) were 45, 20, 95, 68, and 0.3, respectively. $\text{CF} < 1$ means *low contamination*, $1 < \text{CF} < 3$ means *moderate contamination*, $3 < \text{CF} < 6$ is *considerable contamination* and $\text{CF} > 6$ means *very high contamination*.

$$\text{CFn} = C_n/B_n,$$

where CF$n$ = concentration factor, $C_n$ = concentration of metal, and $B_n$ = background of a metal ($n$).

*Pollution Load Index.* A simple index can be used to describe the level of pollution (Tomlinson et al., 1980). When PLI = 1, there is no pollution. On the contrary PLI > 1 will classify the area as polluted (Cabrera et al., 1999).

$$\text{PLI} = (\text{CF1} \times \text{CF2} \times \text{CF3} \cdots \times \text{CF}n)^{1/n},$$

where CF = contamination factors and $n$ = number of metals.

**Table 2.** Certified Reference Material (CRM).

| Heavy Metal | CRM | Certified (μg/g) | Measured (μg/g) | Recovery (%) |
|---|---|---|---|---|
| Cd | Estuarine sediment 1646a | 2.11 | 1.84 | 87.65 |
| Cu | Estuarine sediment 1646a | 10.01 | 11.39 | 113.85 |
| Pb | Estuarine sediment 1646a | 11.70 | 11.39 | 97.41 |
| Zn | Estuarine sediment 1646a | 49.90 | 43.73 | 87.65 |
| Ni | Estuarine sediment 1646a | 39.50 | 31.79 | 80.48 |

To validate the quality of experiment, certified reference material was obtained for the comparison of results, as shown in Table 2.

### *Statistical Analysis*

The SPSS Version 25 software was used to carry out statistical analysis. Descriptive analysis was performed to determine metal concentrations. Principal component analysis (PCA) helped to reduce the wide range of variables. Pearson correlation coefficient was applied to measure the statistical relation between the variable.

## Results

### *Acid Digestion Total Metal Content*

Total metal concentrations for Cu, Zn, Pb, and Ni ranged from 0.3780 to 40.54, 20.98 to 80.08, 2.14 to 8.14, and 4.97 to 17.34 μg/g, respectively, for all six compartments (18, 31, 42, 71, 74, and 55). Concentrations of Cd were below detection limit of the instrument; hence, it was considered as not detected category (Table 3).

### *Fractional Sequential Extraction*

Highest concentration for Cu, Zn, Pb, Ni, and Cd was detected in Compartments 42 (F4–59.73), 18 (F4–41.11), 74 (F4–7.41), 42 (F3–10.41), and 42 (F4–0.918) μg/g, respectively. Meanwhile, the lowest concentration for Zn, Pb, Ni, and Cd was in Compartments 55 (F1–0.66), 18 (F2–0.22), 55 (F2–0.092), and 74 (F2–0.024) μg/g, respectively. Cu was found in not detected category (Table 3).

### *Principal Component Analysis*

Three components (Table 4) were extracted to observe similarity and behavior of metals. Components (C1, C2, and C3) were accounted as 37.51%, 23.87%, and 18.40%, respectively, making the total variance as 79.79%. Component 1 (C1) was dominated by Cu, Zn, Pb, and Ni and showed a relation with clay, silt, and OM. In Component 2 (C2), clay, sand, and OM were prominent. Finally, Component 3 (C3) was dominated by Cd and silt (Figure 3).

From the diagram, the variables were classified based on the distance between the values of variables. Value of each variable in the three components could be considered as dominant when the distance from the component line was farthest.

### *Risk Assessment Code*

RAC was computed by using the combination of F1 and F2 percentages from sequential extraction technique as input. Cu, Zn, Pb, Ni, and Cd were observed in the range of −2.261% to 0.428%, 6.161% to 26.351%, 6.166% to 20.504%, 6.22% to 13.585%, and 9.690% to 21.603%, respectively. The lowest concentration was observed for Cu (−2.261%) in Compartment 55 and the highest concentration was observed in Compartment 55 for Zn (26.351; Supplementary Material). For metal risk assessment, Cu was categorized as no risk in all the compartments, and Zn, Pb, Ni, and Cd were classified in low-to-medium risk range (Table 5).

### *Geo-Accumulation Index*

All elements showed have negative values that ranged from −0.84 to −6.07. Furthermore, Cd was detected in Compartment 42 and Compartment 74 only (Table 6). Negative values showed that compartments were uncontaminated.

### *Contamination Factor and Pollution Load Index*

Cd showed negative values for CF in contrast to other elements. The highest CF value was shown by Cu (0.90) in Compartment 42 and lowest by Cd (−5.45) in Compartment 55 (Table 7). PLI values were negative in all the compartments, except in Compartment 42, which showed a value of 0.168. However, all values were less than one (Table 7). According to the results of CF and PLI, all compartments were in uncontaminated range.

## Discussion

### *Fractional Sequential Extraction*

Concentration of heavy metals, except Pb, in Fraction 1 (F1) was less than in Fraction 2 (F2) and Fraction 3 (F3)

**Table 3.** Fractional Sequential Extraction.

| Compartment | Cu | Zn | Pb | Ni | Cd |
|---|---|---|---|---|---|
| 18 | | | | | |
| F1 | .265 ± .056 | 4.55 ± 1.45 | .559 ± .115 | .522 ± .157 | .030 ± .004 |
| F2 | −.112 ± .016 | 7.031 ± .731 | .225 ± .056 | .620 ± .075 | .129 ± .026 |
| F3 | 2.90 ± .877 | 22.76 ± 3.74 | 5.53 ± 1.06 | 6.02 ± .985 | .165 ± .063 |
| F4 | 32.76 ± 2.22 | 41.11 ± 2.54 | 6.40 ± .532 | 9.139 ± 1.89 | .536 ± .062 |
| Sum total | 35.72 | 75.451 | 12.714 | 16.301 | 0.86 |
| Digestion | 25.69 | 48.72 | 5.33 | 14.98 | −1.35 |
| Difference | 10.03 | 26.731 | 7.384 | 1.321 | 2.21 |
| 31 | | | | | |
| F1 | ND | .853 ± .054 | .550 ± .029 | .477 ± .024 | .037 ± .002 |
| F2 | −.170 ± .006 | 3.56 ± .226 | .328 ± .016 | .606 ± .035 | .165 ± .007 |
| F3 | 1.19 ± .071 | 29.57 ± .359 | 6.30 ± .646 | 8.61 ± .109 | .173 ± .015 |
| F4 | 33.85 ± 2.76 | 37.64 ± 1.02 | 6.85 ± .148 | 7.77 ± .291 | .894 ± .083 |
| Sum total | 34.87 | 71.623 | 14.028 | 17.463 | 1.269 |
| Digestion | 24.11 | 66.92 | 6.66 | 14.53 | −0.55 |
| Difference | 10.76 | 4.703 | 7.368 | 2.933 | 1.819 |
| 42 | | | | | |
| F1 | ND | 10.364 ± 1.76 | 1.038 ± .068 | 1.031 ± .139 | .053 ± .001 |
| F2 | −.209 ± .010 | 12.84 ± 1.54 | .476 ± .016 | .806 ± .20 | .166 ± .013 |
| F3 | 2.208 ± .109 | 34.75 ± 2.22 | 5.44 ± 1.42 | 10.41 ± .692 | .197 ± .014 |
| F4 | 59.73 ± 5.83 | 30.32 ± 1.56 | 7.36 ± .394 | 8.46 ± .361 | .918 ± .059 |
| Sum total | 61.72 | 88.274 | 14.314 | 20.707 | 1.334 |
| Digestion | 40.54 | 80.08 | 8.14 | 17.34 | −0.02 |
| Difference | 21.18 | 8.194 | 6.174 | 3.367 | 1.354 |
| 71 | | | | | |
| F1 | ND | 1.109 ± .156 | .796 ± .048 | .582 ± .038 | .0514 ± .001 |
| F2 | −.242 ± .005 | 5.67 ± .494 | .647 ± .026 | .257 ± .038 | .122 ±. 022 |
| F3 | .697 ± .087 | 22.70 ± 1.36 | 5.21 ± .473 | 6.64 ± .451 | .044 ± .014 |
| F4 | 22.85 ± 1.87 | 22.153 ± 1.76 | 7.136 ± .325 | 6 ± .501 | .751 ± .093 |
| Sum total | 23.3 | 51.632 | 13.785 | 13.48 | 0.9685 |
| Digestion | 10.96 | 36.02 | 6.81 | 10.8 | −0.64 |
| Difference | 12.34 | 15.612 | 6.975 | 2.68 | 1.6085 |
| 74 | | | | | |
| F1 | ND | 3.21 ± .453 | 1.042 ± .045 | .754 ± .062 | .051 ± .001 |
| F2 | −.240 ± .003 | 7.75 ± .584 | .385 ± .177 | .363 ± .050 | .0240 ± .014 |
| F3 | .651 ± .168 | 24.12 ± 6.06 | 5.55 ± 1.39 | 8.096 ± 2.02 | .085 ± .031 |
| F4 | 23.82 ± .569 | 28.90 ± .874 | 7.41 ± .104 | 7.92 ± .297 | .614 ± .120 |
| Sum total | 24.23 | 63.98 | 14.387 | 17.133 | 0.774 |
| Digestion | 10.56 | 48.41 | 8.03 | 13.26 | −0.04 |
| Difference | 13.67 | 15.57 | 6.357 | 3.873 | 0.814 |
| 55 | | | | | |
| F1 | ND | .661 ± .205 | .527 ± .020 | .354 ± .023 | .039 ± .003 |
| F2 | −.253 ± .002 | 3.45 ± .219 | .486 ± .019 | .092 ± .021 | .147 ± .006 |
| F3 | .693 ± .146 | 6.59 ± .848 | 2.139 ± .734 | 2.55 ± .322 | .085 ± .021 |
| F4 | 10.75 ± 5.56 | 4.81 ± .820 | 3.86 ± .234 | .287 ± .606 | .590 ± .022 |
| Sum total | 11.19 | 15.601 | 7.012 | 3.283 | 0.861 |
| Digestion | 0.378 | 20.98 | 2.14 | 4.97 | −1.633 |
| Difference | 10.812 | −5.379 | 4.872 | −1.687 | 2.494 |
| Overall mean | 31.83 | 60.30 | 12.70 | 14.72 | 1.006 |
| Digestion mean | 18.70 | 50.18 | 6.18 | 12.64 | −0.70 |

*Note.* F1 = Factor 1; F2 = Factor 2; F3 = Factor 3; F4 = Factor 4; ND = not detected.

in all sampled compartments (Table 3). In case of Cu, the concentration in F1 was not detected in all compartments other than Compartment 18. For F2, Cu was negative in all compartments. Negative value means that samples are having concentrations which are below the calibration curve limit of the standard. According to Abubakar et al. (2018), Cu and Cd mostly originated from lithogenic sources (>90% from residual fraction),

while Zn, Pb, and Ni were sourced from anthropogenic activities (>50% from nonresistant or mobile fraction).

In Fraction 4 (F4), the residual fraction, concentration of heavy metals was higher than in all the other three fractions. Heavy metals in F4 came from natural origin, that is, sediment surface and geological weathering (Swati & Hait, 2017). However, these were not easily available because of its bonding with crystal lattice (El-Azim & El-Moselhy, 2005). The other three fractions (F1, F2, and F3) were called mobile fraction. They were readily bioaccessible due to little change in physiochemical structure of the sediment (Passos et al., 2010). Extensive industrialization, agriculture/aquaculture, domestic wastes, mining, dumping of automobile scraps, and fluids significantly contribute to heavy metal pollution (Zn, Pb, and Ni) in F1, F2, and F3 (nonresistant fraction).

**Table 4.** Rotated Component Matrix.

| Variables | C1 | C2 | C3 |
|---|---|---|---|
| Cu | **.700** | −.082 | .234 |
| Zn | **.943** | −.178 | −.082 |
| Pb | **.874** | .068 | .192 |
| Ni | **.952** | −.144 | −.115 |
| Cd | −.126 | .068 | **.819** |
| Clay% | .133 | **−.925** | .185 |
| Sand% | −.319 | **.762** | −.498 |
| Silt% | .432 | −.228 | **.700** |
| OM% | .061 | **.770** | .317 |
| Total Variance % | 37.51 | 23.87 | 18.40 |

*Note.* C1 = Component 1; C2 = Component 2; C3 = Component 3; OM = organic matter. The values are bold which are greater than +.7 and −.7.

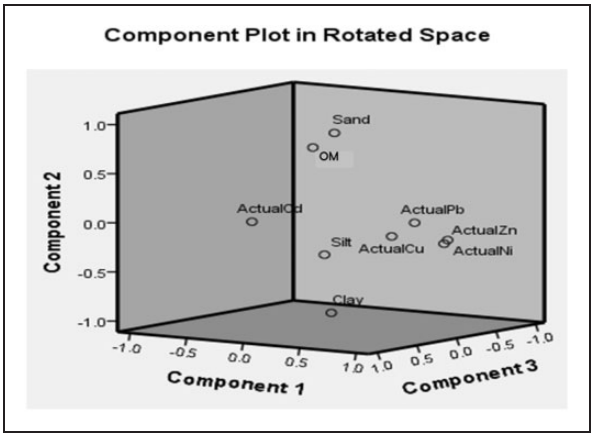


**Figure 3.** 3D Representation for PCA. OM = Organic matter.

## *Heavy Metals Concentration in the Sediments*

This study showed that the concentrations of trace metal for all elements were still lower as compared with the previous study in mangroves at Sungai Puloh, Malaysia (Udechukwu et al., 2015). The Sg. Puloh mangrove

**Table 6.** Geo-Accumulation Index ($I_{geo}$).

| Compartment | Cd | Cu | Pb | Zn | Ni |
|---|---|---|---|---|---|
| 18 | – | −1.39 | −2.51 | −1.55 | −2.78 |
| 31 | – | −1.51 | −2.17 | −1.09 | −2.81 |
| 42 | −2.17 | −0.7 | −1.88 | −0.84 | −2.56 |
| 55 | – | −6.07 | −3.88 | −2.77 | −4.38 |
| 71 | – | −2.63 | −2.12 | −1.98 | −3.24 |
| 74 | −1.38 | −2.68 | −1.90 | −1.55 | −2.94 |

**Table 7.** Contamination factor (CF) and Pollution load Index (PLI).

| Compartment | Cd | Cu | Pb | Zn | Ni | PLI |
|---|---|---|---|---|---|---|
| 18 | −4.52 | 0.57 | 0.26 | 0.51 | 0.22 | −0.59 |
| 31 | −1.86 | 0.53 | 0.33 | 0.70 | 0.21 | −.54 |
| 42 | −0.09 | 0.90 | 0.40 | 0.84 | 0.25 | 0.168 |
| 55 | −5.45 | 0.008 | 0.10 | 0.22 | 0.07 | −0.12 |
| 71 | −2.15 | 0.24 | 0.34 | 0.37 | 0.15 | −0.39 |
| 74 | −0.13 | 0.23 | 0.40 | 0.50 | 0.19 | 0.005 |

**Table 5.** Classification of RAC.

| | | Percentages | | | | |
|---|---|---|---|---|---|---|
| Risk | Metal in exchangeable and Carbonate Fraction (F1+F2)% | Cu | Zn | Pb | Ni | Cd |
| No risk | <1 | **(−2.261 to 0.428)** | | | | |
| Low risk | (1–10) | | **(6.161–26.351)** | **(6.166–20.504)** | **(6.224–13.585)** | **(9.690–21.603)** |
| Medium risk | (11–30) | | | | | |
| High risk | (31–50) | | | | | |
| Very high risk | >50 | | | | | |

*Note.* RAC = risk assessment code.

estuary received vast amounts of contamination of heavy metals from industrialization and urbanization. Even though trace metals were detected in MMFR, they were in minute concentrations since it was one of the most sustainable and reserved mangrove ecosystems in Malaysia. A study by Ismail et al. (1995) in Kuala Sepetang, Perak, Malaysia conducted between 1992 and 1993 also showed lower concentrations of Cd, Cu, Pb, and Zn (0.35 μg/g, 4.91 μg/g, 26.14 μg/g, and 33.91 μg/g).

Regardless of the observed low contamination by heavy metals, there was still a possibility of urbanization, industrialization, and tourism around the investigated area leading to accumulation of heavy metals over the years. The effect of consistent anthropogenic activities, as reported by Rahman et al. (2013), was evidenced in the decline of wild Milky Stork population from surface sediment samples in foraging areas. A study by Lomoljo et al. (2010) in Kuala Gula bird sanctuary showed lower concentrations of heavy metals due to minimal human activities around the study area (Table 8). However, findings by Lomoljo et al. (2010) claimed that even though the concentrations in certain areas might be low, as in the study area (MMFR), it might increase through bioaccumulation. Therefore, a follow-up study should be conducted to monitor the levels of contamination in the reserve forest.

## Soil Texture and OM

The OM content in the sediment was between 38% and 45% for all six compartments. Remarkably, lower values in the range of 7% to 16% (Idriss & Ahmad, 2013) and even 0.1% to 2.8% (Ahmad et al., 2009) were reported in other Malaysian mangroves. Similar to this study, Maslennikova et al. (2012) reported 50% to 33% of OM in Russian lake. This OM mostly originated from land and water sources, but litter decomposition and subsurface roots also had a contribution. Sediment texture in all compartments was characterized by high clay (37%–50%), silt (55%), and low sand (8%–38%) content.

In this study, correlation strength was categorized from 0 to 1. Positive significant values ranged from weak (0–0.3), intermediate (0.3–0.8) to strong (0.8–1). For negative significant value, the above-mentioned range can be applied to values between −1 and 0. A correlation between heavy metals, OM, and sediment texture, negative and positive relation was observed (Table 9). As a general rule, heavy metal pollution is reflected in the positive and strong correlation between

**Table 8.** Summary of Heavy Metals Concentration in the Sediments (μg/g).

| Location | Cu | Zn | Pb | Ni | Cd | References |
|---|---|---|---|---|---|---|
| Total heavy metals digestion | | | | | | |
| 18 | 25.69 | 48.72 | 5.33 | 14.98 | −1.35 | Present study |
| 31 | 24.11 | 66.92 | 6.66 | 14.53 | −0.55 | |
| 42 | 40.54 | 80.08 | 8.14 | 17.34 | −0.02 | |
| 71 | 10.96 | 36.02 | 6.81 | 10.8 | −0.64 | |
| 74 | 10.56 | 48.41 | 8.03 | 13.26 | −0.04 | |
| 55 | 0.378 | 20.98 | 2.14 | 4.97 | −1.633 | |
| Sequential Extraction Technique | | | | | | |
| 18 | 35.72 | 75.451 | 12.714 | 16.301 | 0.86 | Present study |
| 31 | 34.87 | 71.623 | 14.028 | 17.463 | 1.269 | |
| 42 | 61.72 | 88.274 | 14.314 | 20.707 | 1.334 | |
| 71 | 23.3 | 51.632 | 13.785 | 13.48 | 0.9685 | |
| 74 | 24.23 | 63.98 | 14.387 | 17.133 | 0.774 | |
| 55 | 11.19 | 15.601 | 7.012 | 3.283 | 0.861 | |
| West coast of Peninsular Malaysia | 6.0–50.0 | 600–900 | 15.0–30.0 | – | <1.0 | Ismail et al. (1993) |
| Malaysian Coast, Malaysia | 1.79–8.17 | 17.05–42.43 | 6.16–27.52 | – | 0.27–0.54 | Ismail et al. (1995) |
| Peninsular Malaysia, Malaysia | 1.63–150.81 | 23.70–609.20 | 7.97–93.11 | 2.41–36.29 | ND–1.06 | Zulkifli et al. (2010) |
| Kuala Gula, Perak, Malaysia | 9.7–57.0 | 71.0–120.0 | 28.0–47.0 | – | 0.9–1.7 | Rahman et al. (2013) |
| Sungai Puloh mangrove estuary, Malaysia | 46.89 | 1023.68 | 78.8 | 35.54 | 0.94 | Udechukwu et al. (2015) |
| Kuala Gula bird sanctuary | 4.39–7.9 | 29.0–53.2 | 12.1–28.9 | – | 0.4–1.6 | Lomoljo et al. (2010) |

**Table 9.** Correlation Between Heavy Metal, Soil Texture, and Organic Matter.

| | Cu | Zn | Pb | Ni | Cd | Clay% | Sand% | Silt% | OM% |
|---|---|---|---|---|---|---|---|---|---|
| Cu | 1 | | | | | | | | |
| Zn | .604** | 1 | | | | | | | |
| Pb | .615** | .787** | 1 | | | | | | |
| Ni | .556** | .948** | .819** | 1 | | | | | |
| Cd | .254** | −.163* | .137 | −.207* | 1 | | | | |
| Clay% | .166 | .271** | .183* | .262** | .052 | 1 | | | |
| Sand% | −.286** | −.361** | −.269** | −.336** | −.165* | −.875** | 1 | | |
| Silt% | .331** | .342** | .275** | .303** | .248** | .382** | −.781** | 1 | |
| OM% | −.155 | −.161* | .040 | −.106 | .060 | −.495** | .250** | .147 | 1 |

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

OM and heavy metals (Abubakar et al., 2018). In this study, Zn exhibited weak negative relation with OM. Clay and silt showed weak positive correlation with Zn, Pb, and Ni. Silt expressed intermediate positive relation with Cu, Zn, Ni, and weak positive relation with Pb and Cd, respectively (Table 9). The observed weak correlation between heavy metals and OM in the study evidenced that heavy metals came from natural origin. On the other hand, OM may be released in subsurface decay with water column movement.

### *Principal Component Analysis*

Cu, Zn, Ni, and Pb were dominated in PC1, exhibiting its source with distribution in sediments. Cd, Pb, Zn, and Cu were exposed to the environment because of pesticides and phosphate fertilizer applications (Wang et al., 2015). PC2 was influenced by OM, clay, and sand. At Ubatuba Bay Brazil, the same pattern was found in the surface sediments (Burone et al., 2003). PC3 showed Cd and silt were dominant (Table 4). Silt has the ability to bind metals. This was also observed in the sediments of the Shuangtaizi Estuary China in PC3 analysis (Li et al., 2017).

### *Risk Assessment Code*

RAC classified Zn, Pb, Ni, and Cd in the selected compartments in the range of low-to-medium risk. Cu was classified in no risk range. In this classification, it was observed that the percentage of Zn, Pb, Ni, and Cd was high in the VJR compartment as compared with other compartments. This could be because VJR has not been managed since the last 80 years. Therefore, the pollution from industrial and domestic waste had remained. For Cd risk assessment, all compartments were in moderate risk range, except for Compartment 71. Moreover, the sequential extraction technique also revealed that the source of Cd was natural. In comparison, RAC percentage was higher than 50 in the sediments of Sungai Puloh mangrove Malaysia (Abubakar et al., 2018) which meant that heavy metal content was in the high-risk range. A study conducted in Can Gio mangrove stand in Vietnam showed Ni and Cu exhibited less risk to the ecosystem (Thanh-Nho et al., 2019). Furthermore, a study was conducted in Yemen to assess the heavy metal enrichment in sediment samples. This area was also categorized as less polluted for heavy metals (Zn, Cu, Ni, and Pb), except for Cd (Al-Edresy et al., 2019).

### *Geo-Accumulation Index*

All the values for $I_{geo}$ were negative, which showed that these compartments were uncontaminated (Table 6). The same results were identified in Mengkabong Lagoon, Malaysia, and South-Eastern Baltic Sea (Remeikait_e-Nikiene et al., 2018; Praveena et al., 2007), whereby the heavy metal concentration was in the unpolluted range.

### *Contamination Factor and Pollution Load Index*

All the values calculated for CF were less than 1. This could mean that the area was less polluted or not enriched with metals (Table 7). In case of PLI, negative values were obtained in Compartment 18, Compartment 31, Compartment 55, and Compartment 71. This meant that sediments were not contaminated with the investigated heavy metals. Compartment 42 and Compartment 74 showed positive and PLI of less than 1. Based on that, it was concluded that all the compartments were unpolluted and did not release metals into the mangrove ecosystem (Table 8). In comparison, a study was conducted by Udechukwu et al. (2015) at Sg. Puloh mangrove estuary Malaysia. Based on the obtained PLI, it was concluded that Sg. Puloh mangrove estuary was under the effect of moderate pollution. On the contrary, another study was conducted in Cochin, India, whereby mangrove sites were stressed with heavy metals

contamination (CF = 66.8) and severe pollution effects ($I_{geo}$ = − 1.8–5.4; Joseph et al., 2019).

## Implications for Conservation

Signs for low-to-moderate heavy metal contaminations were observed in all six compartments by RAC. On the other hand, values of CFs, which were $I_{geo}$ and PLI, classified the compartments as unpolluted. This has also reinforced the outcome of previous studies (Ismail et al., 1995, Lomoljo et al., 2010). It could be said that anthropogenic sources also contributed to the observed concentrations of Zn, Pb, and Ni in all compartments. However, heavy metal pollution was only higher, whereby the source was from natural activities, that is, sediment surface and geological weathering (Swati & Hait, 2017). Since the pollution levels posed no risk, no remedial measures were required. However, it was suggested to monitor levels of heavy metals every 5 years. Percentage of clay and OM was very high in all compartments, showing richness and productivity of the mangrove ecosystem. No concrete conclusions could be drawn from the correlation between OM and heavy metals. Therefore, a new method could be investigated for OM extraction. PCA results exhibited that the whole study area was highly influenced by metal contents in sediments. It was recommended that a further study should be conducted to determine the behavior of other heavy metals and pollutants.

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### ORCID iDs

Waseem Razzaq Khan https://orcid.org/0000-0002-5981-2105
Syaizwan Zahmir Zulkifli https://orcid.org/0000-0001-5822-7273
Ahmad Mustapha Pazi https://orcid.org/0000-0001-6966-1497
Fahad Rasheed https://orcid.org/0000-0001-8807-5370
M. Nazre https://orcid.org/0000-0001-7685-1184

### Supplemental material

Supplemental material for this article is available online.

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