Spatial distribution and potential ecological risk assessment of heavy metals in the North-West Coast of Kundur Island, Kepulauan Riau Province, Indonesia

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Abstract. The objective of the present study was to analyze spatial distribution potential ecological risk of heavy metals in North-West Coast of Kundur Island. Samples of seawater and sediment were taken from five stations and were analysed by using AAS Perkin Elmer 3110 in Marine Chemistry Laboratory Faculty of Fisheries and Marine Science, Universitas Riau. The arrangement of the metals studied from higher to lower mean concentrations are: Fe>Ni>Zn>Mn>Cr>Sn>Pb>Cu in seawater and Fe>Mn>Zn>Ni>Cr>Pb>Sn>Cu in sediments, respectively. Distribution of heavy metal concentrations in seawater and sediment were found to have almost similar trends where higher concentrations were found in stations closed to the mining activities. Concentration of all metals in seawater were positively correlated with that in sediment, except for Mn. Contamination factor, geoaccumulation index and enrichment factor demonstrated that most of the sediment samples were moderately to heavily contaminated by Cr, Ni and Zn but not yet for other metals. However, PERI value indicated that the studied area is still has low ecological risk from the mining activity in the surrounding area.

1. Introduction

Coastal pollution has been increasing significantly over the recent years and is found compounding environmental problems in many developing countries. Pollution of coastal waters usually attributable to various human activities, not only on land but also in the sea, especially industries which may introduce a number of wastes containing heavy metals into the marine environment. Coastal and estuarine regions are the important sinks for many persistent pollutants and they accumulate in the organisms and bottom sediments [1, 2, 3, 4, 5, 6]. Pollution of aquatic ecosystems by heavy metals is an important environmental problem because of its permanent disturbances in marine ecosystems, leading to environmental and ecological degradation.

Heavy metals are accumulated in marine sediments, where they are incorporated in several biological and chemical cycles, affecting the water column and biota. Anthropogenic impacts including mining and industrial discharges, domestic sewage, non-point source, runoff and atmospheric precipitation are the main sources of toxic heavy metals that enter aquatic ecosystems [7, 8, 9]. Trace metal pollution occurs in coastal sediments which are near actively operated industries, mining, urban centers and agriculture [7, 10, 11, 12, 13, 14]. Previous studies in the coastal waters in the proximity of tin mining activities in Karimun Island showed the heavy metal concentrations were elevated significantly [15, 16]. Kundur Island is an island within the Riau Archipelago, part of the Kepulauan Riau Province of Indonesia. It lies at about 80 kilometres southwest of Singapore, 76 kilometres southwest of Batam and has an area of about 304 square kilometres. This island has been used as mining areas especially for granite and tin mining, and recently for sand by some (legal and illegal) mining companies. Other anthropogenic activities such as shipping, port, transportation and municipal are thought to contribute pollutants including heavy metals to its coastal waters. Dangers which may arise from the mining activity is the accumulation of heavy metals by sediment, water and biota that will
adversely affect the organisms that accumulate them either directly or indirectly. This study aimed to determine the concentrations of selected heavy metals (Fe, Cu, Cr, Mn, Ni, Pb, Sn and Zn) in seawater and sediment collected from the north-west part of Kundur Island coastal water.

2. Materials and Methods
This study was conducted in April 2018 by taking samples of seawater and sediment from the coastal waters of Kundur Island Kepulauan Riau Province, especially in its north-west part. Sampling of seawater and sediment was conducted in five stations (figure 1). Seawater samples were taken with 500 ml polyethylene sample bottles at 0-30 cm depth, the surface sediment samples (0-5 cm) were taken by using Ekman grab from each station and they were then treated by following the procedures of Yap et al. [17] for sediment and procedure of Hutagalung [18] for seawater analysis. All statistical analysis of data were carried out using SPSS statistical package programs version 17 and graphs were plotted with Microsoft EXCEL 2010. All data were also tested in exploratory data analysis in SPSS 17 for the basic assumptions of normality and homogeneity of variance. Water quality parameters measurement was done in triplicates at each sampling station at the same time with seawater and sediment sampling for heavy metal analysis.

Figure 1. Map of sampling locations in the North-west coast of Kundur Island

2.1 Enrichment factor and index of geoaccumulation
The extent of sediment contamination could be assessed by Enrichment factor (EF) and Geoaccumulation index (I-geo). EF represents the actual contamination level in the sediment [19] and is a good tool to differentiate the metal source between anthropogenic and naturally occurring [20, 21, 22]. The following equation was used in the present study to estimate the EF of metals in sediment from each station using Fe as a normalizer to correct for differences in sediments grain size and mineralogy:

$$EF_S = \frac{(C_x/C_{Fe})_{sample}}{(C_x/C_{Fe})_{shale}}$$

Where, $(C_x/C_{Fe})_{sample}$ and $(C_x/C_{Fe})_{shale}$ average shale value are, respectively, the metal concentration (µg/g d.w) in relation to Fe levels (% d.w) in sediment samples, and average shale values [23]. EF values were interpreted as suggested by Birch [24]; where EF < 1 indicates no enrichment, EF < 3 is minor enrichment, EF = 3–5 is moderate enrichment, EF = 5–10 is moderately severe enrichment, EF = 10–25 is severe enrichment, EF = 25–50 is very severe enrichment, and EF > 50 is extremely severe enrichment.

The I-geo values for the metals studied were calculated using the Muller’s [25] expression as follows:

$$I_{geo} = \log_2 \left( \frac{C_x}{1.5B_n} \right)$$
Where, \( C_n \) is the measured concentration of the examined metal ‘n’ in the sediment, and \( B_n \) is the geochemical background concentration of the metal ‘n’ of average shale. Muller [25] has distinguished seven classes of geoaccumulation index in relation to pollution extent as seen in table 1.

| I-geo  | I-geo class | Pollution intensity       |
|--------|-------------|---------------------------|
| >5     | 6           | very strongly polluted    |
| 4 – 5  | 5           | strongly to very strongly polluted |
| 3 – 4  | 4           | strongly polluted         |
| 2 – 3  | 3           | moderately to strongly polluted |
| 1 – 2  | 2           | moderately polluted       |
| 0 – 1  | 1           | unpolluted to moderately polluted |
| < 0    | 0           | unpolluted                |

2.2 Contamination factor (CF)

The level of metal contamination can be expressed by the contamination factor (CF). CF is the ratio between the metal content in the sediment to the background value of the metal [26]. It is an effective tool for monitoring the pollution over a period of time and it is calculated as follows

\[
CF_{metal} = \frac{C_{metal}}{C_{background}}
\]

CF<1 indicates low contamination; 1<CF<3 is moderate contamination; 3<CF<6 is considerable contamination; and CF>6 is very high contamination [27].

2.3 Potential ecological risk

The potential ecological risk index (RI) is calculated as the sum of all risk factors for heavy metals in sediments and the formula of the potential ecological risk index is given as:

\[
RI = \sum \frac{E_i}{C_i} \times CF
\]

The following terminologies are suggested for the Er and RI values: (1) Er <40, low ecological risk; 40 < Er ≤80, moderate ecological risk; 80 < Er ≤160, appreciable ecological risk; 160 < Er ≤320, high ecological risk; and >320, serious ecological risk; (2) RI <150, low ecological risk; 150 < RI <300, moderate ecological risk; 300 < RI <600, high ecological risk; and RI ≥ 600, significantly high ecological risk. RI method covers a variety of researching domains, i.e., biological toxicology, environmental chemistry as well as ecology, and can evaluate ecological risks caused by heavy metals comprehensively [27].

3. Results and Discussion

3.1 Water Quality Parameters

The results of water quality parameter measurements from each station (table 2) showed relatively no significant difference. The pH value of 7.6 - 8.0 can be considered still in the range of tolerable level by aquatic biota, while the temperature (27.4 – 28.9 °C) were also in the range for the life of the marine organism [28]. The water temperature will determine the biological activity and their activeness on marine biota in the water [29]. When compared with the water quality standards for marine biota (Kep. No.51/MENLH/2004) [30], the average of all environmental parameters in coastal waters of Kundur island are generally still in the range of tolerable level for marine organisms.
Table 2. Seawater quality parameter in the north-west coast of Kundur island, Indonesia

| Station | Temp. (°C) | pH  | Salinity (%) | Trans. (cm) | Current (m/s) | Depth (m) |
|---------|------------|-----|--------------|-------------|---------------|-----------|
| 1       | 27.5       | 7.7 | 32           | 25          | 0.10          | 11.3      |
| 2       | 28.0       | 8.0 | 32           | 20          | 0.10          | 5.2       |
| 3       | 27.4       | 7.9 | 31           | 21          | 0.10          | 3.2       |
| 4       | 28.4       | 8.0 | 31           | 21          | 0.20          | 4.3       |
| 5       | 28.9       | 7.6 | 30           | 20          | 0.21          | 13.0      |

3.2 Concentrations of heavy metals in seawater

Seawaters are naturally contain heavy metals in small amounts, but when the concentration in the water increases due to certain sources, the amount can be toxic to the organisms or humans who consume them. In general, all heavy metals can cause negative effect to aquatic organisms at certain concentration limits. The influence varies depending on the type of metal, the species of organisms, the permeability and the detoxication mechanism. Heavy metals in the water easily absorbed and accumulate in phytoplankton which is the starting point of the food chain, further higher up through the food chain to other organisms.

The mean concentrations of Cu, Cr, Mn, Ni, Sn, Zn, Pb and Fe in sea water at each station can be seen in Table 3. Highest mean metal concentrations in sea water was found for Fe, Mn, Pb, Cu, Cr and Ni in Station 3, whilst highest concentration for Zn was found in Station 1 and Sn in Station 5. Heavy metal concentration in the waters is always changing depending on the time of disposal of waste and the degree of aquatic environment perfection. Increased concentrations of heavy metals in seawater will usually be followed by its increase in the body of organisms, so that sea water pollution will be followed by the elevated concentrations of the metals in the living biota and sediment in the environment.

Table 3. Mean concentration of heavy metals in seawater

| Station | Cu   | Cr   | Mn   | Ni   | Sn   | Zn   | Pb   | Fe   |
|---------|------|------|------|------|------|------|------|------|
| 1       | 0.04 | 0.08 | 0.04 | 0.96 | 0.05 | 0.11 | 0.04 | 0.62 |
| 2       | 0.03 | 0.06 | 0.04 | 0.90 | 0.05 | 0.08 | 0.03 | 0.69 |
| 3       | 0.04 | 0.08 | 0.14 | 1.08 | 0.04 | 0.07 | 0.05 | 1.73 |
| 4       | 0.03 | 0.07 | 0.12 | 0.82 | 0.05 | 0.09 | 0.03 | 1.27 |
| 5       | 0.04 | 0.06 | 0.07 | 0.87 | 0.06 | 0.09 | 0.04 | 0.81 |
| Mean    | 0.04 | 0.07 | 0.08 | 0.93 | 0.05 | 0.09 | 0.04 | 1.03 |

Station 3 was found to have the highest concentration for most of the metals analyzed and on the other hand the lowest concentrations for Zn were also found in Station 3, Station 2 (Cu, Cr, Mn, Pb), Station 4 (Ni) and Station 1 (Fe). This is presumably due to Station 3 is an area located in the nearest mining activity where many dredgers are in their operation in both sides of Station 3, while other stations were relatively in a distance from the mining area as well as recreational area that is relatively not affected by the mining activities. Increase metal concentrations in sediment in a nearby mining areas [31, 32, 33]

3.3 Concentrations of heavy metals in sediment

Once heavy metals enter the seawater it will experience precipitation, dilution and dispersion. Heavy metal that settles on the sea floor will be accumulated into the sediments [34]. Unlike in sea water, the concentrations of metals in the sediment at Station 5 has the highest concentration of most metals and it was assumed that the metals were derived from the surrounding mining activities. The high
concentration of metals in this region is also thought to come from other anthropogenic activities inland, both from Karimun and Rangsang island which has both industrial area and mining activities. The coastal waters of those two islands were also used as human settlement, housing and most of fishermen always moored their boats in this area. The mean concentrations of metals in sediment samples from Station 5 was higher than that in other stations (table 4). Muddy sand sediment in Station 5 was also assumed to be the cause of higher metal concentrations. Sandy sediment would accumulate relatively lower metal concentration in comparison to muddy sediments. This might explain the lower concentration of metals studied in other stations [35].

To determine the level of pollution in the coastal waters of Kundur island, the concentration of heavy metals in the sediment were compared to ERL (Effects Range Low) and ERM (Effects Range Median) standards [36, 37]. Comparison of metal concentrations in the present study with the standard quality guidelines can be seen in Table 4. Concentrations of Ni in all stations were exceeded the ERL and ERM values. This might give indication that the Ni concentrations might have a negative impact on organisms living in the studied environment. As for Ni, Zn concentration at Station 1 has exceeded both ERL and ERM values, concentrations in other stations have also exceeded ERL but still below ERM values. Other metals in all stations were still below both ERL and ERM values. The value of the concentration of heavy metals was passed ERL values but remained below the ERM value means that there could be negative effects on aquatic organisms, and when the concentration of heavy metals exceeded the ERM value then the concentration of heavy metals have a negative effect on aquatic organisms [36, 37].

Table 4. Comparison of heavy metal concentrations in the sediments with SQG Standard

| Station | Cu    | Cr    | Mn    | Ni    | Sn    | Zn    | Pb    | Fe    |
|---------|-------|-------|-------|-------|-------|-------|-------|-------|
| 1       | 1.78  | 331.43| 639.30| 264.25| 4.43  | 423.63| 11.78 | 3106.20|
| 2       | 1.40  | 264.91| 372.81| 237.25| 5.48  | 238.38| 6.85  | 2948.40|
| 3       | 1.23  | 334.60| 640.60| 300.08| 1.50  | 344.48| 14.88 | 3085.00|
| 4       | 0.86  | 118.66| 273.35| 238.75| 6.25  | 140.71| 7.33  | 3449.27|
| 5       | 3.24  | 275.90| 779.82| 333.00| 8.78  | 313.53| 16.75 | 3058.20|
| Mean    | 1.70  | 265.10| 541.18| 274.67| 5.29  | 292.14| 11.52 | 3129.41|
| Aver Shalea | 45 | 90 | 950 | 68 | - | 95 | 20 | 4.6 |
| Aver Sdminb | 33 | - | - | 52 | - | 95 | 19 | 4.1 |
| ERLc   | 34   | 81   | -     | 20.9 | -   | 150  | 46.7 | -    |
| ERMd   | 270  | 370  | -     | 51.6 | -   | 410  | 218  | -    |

aTurekian and Wedephol [26]  bSalomon and Forstner [38]  cLong et al. [36, 37]

3.4 Relationships between metal concentration in seawater and sediment

Relationships between concentrations of metals in the seawater with the metal concentrations in the sediment from coastal waters of Kundur island can be seen in figure 2. Linear regression analysis showed positive correlations for all metals, except for Mn. Although all correlation coefficient values were considered having weak correlation, relationships between Sn concentrations in seawater and sediment has stronger correlation coefficient when compared to other metals. Aquatic sediments absorb persistent and toxic chemicals to levels of many times higher than the water column concentration [8].
Table 5 showed the mean EF values of the metals studied with respected to average shale [26]. Based on the interpretation suggested by Birch [24], EF values for Cu (0.58) was < 1 indicated no enrichment and this metal was originated from natural weathering process, but Ni showed extremely severe enrichment of this metal and Sn has severe enrichment. Moderately severe enrichment is shown for average EF values for Pb and Mn, whilst Cr and Zn EF values indicated very severe enrichment which according to Zhang and Liu [39] suggesting environmental contamination by these metals. Previous
study had demonstrated that municipal and/or industrial wastewater discharges into coastal zones are the most important sources for contamination of water and sediment with heavy metals [40].

I-geo of sediment was calculated and the results showed that overall average of I-geo values of all metals in sediments from Kundur island coastal water fall into class 1 which categorized as unpolluted to moderately polluted environment (table 6). However it can also be seen that Ni has the highest I-geo value followed by Zn and Cr and the lowest values were found for Cu and Fe. I-geo value for most of the metals studied was found at Station 5 and 1 when compared to other stations.

| Station | EF         | Cu   | Cr   | Mn   | Ni   | Sn   | Zn   | Pb   |
|---------|------------|------|------|------|------|------|------|------|
| 1       | 0.60       | 55.96| 11.43| 59.05| 11.23| 67.76| 8.95 |
| 2       | 0.50       | 47.12| 7.02 | 55.85| 14.63| 40.17| 5.48 |
| 3       | 0.42       | 56.88| 11.53| 67.52| 3.82 | 55.48| 11.38|
| 4       | 0.26       | 18.04| 4.40 | 48.05| 14.25| 20.27| 5.01 |
| 5       | 1.11       | 47.31| 14.16| 75.58| 22.59| 50.94| 12.93|
| Mean    | 0.58       | 45.06| 9.71 | 61.21| 13.31| 46.92| 8.75 |

| Station | I-geo Step | Cu   | Cr   | Mn   | Ni   | Sn   | Zn   | Pb   | Fe   |
|---------|------------|------|------|------|------|------|------|------|------|
| 1       | 1          | 0.01 | 0.74 | 0.15 | 0.78 | 0.15 | 0.89 | 0.12 | 0.01 |
| 2       | 2          | 0.01 | 0.59 | 0.09 | 0.70 | 0.18 | 0.50 | 0.07 | 0.01 |
| 3       | 3          | 0.01 | 0.75 | 0.15 | 0.89 | 0.05 | 0.73 | 0.15 | 0.01 |
| 4       | 4          | 0.00 | 0.26 | 0.06 | 0.70 | 0.21 | 0.30 | 0.07 | 0.01 |
| 5       | 5          | 0.01 | 0.62 | 0.18 | 0.98 | 0.29 | 0.66 | 0.17 | 0.01 |
| Mean    | 1          | 0.01 | 0.59 | 0.13 | 0.81 | 0.18 | 0.62 | 0.12 | 0.01 |

3.6 Potential ecological risk
In order to quantify the overall potential ecological risk of heavy metals in sediments, the values of PERI were calculated. The results of evaluation on potential ecological risk factor (Er) and the potential ecological risk index (RI) are summarized in table 7. The values in five sampling stations ranged from 23.93 to 39.39, with an average of 32.87. The higher and the lower PERI values were observed at Stations 5 and 4, respectively. The order of potential ecological risk coefficient (Er) of heavy metals in sediments of Kundur island coast was Ni > Cr > Zn > Pb > Mn > Cu. The mean potential ecological risk coefficient of those metals were all lower than 40, which categorized as low ecological risk. All the sampling sites were at low risk level where the RI values were much lower than 150 and therefore all those stations were considered having low ecological risk to the environment from heavy metals.

The potential ecological risk index was built by Hakanson [27] which integrated the concentration of heavy metals with ecological effect, environmental effect, toxicology, and used to assess the heavy metals pollution and ecological risk for sedimentology. Spatial distribution of single risk indices (Er,) is shown in Table 7 and it was found that the single risk indices of heavy metals were ranked in the order of Ni > Cr > Zn and the lowest was Cu. The result was consistent with that based on geoaccumulation index. The average ecological risk of Ni in the studied area was 20.20, indicating that Ni still posed a low risk to the local aquatic ecosystem. The mining activities and other anthropogenic sources of Ni may cause the relative enrichment of this metals in the study area. Tin and granite mining and their processing as well as living residents are thought to be the source of this metal in the sediments. The low ecological risk of all metals in the present study should be considered as save for the environment at the present time, but it should be of concerned in the future if the mining activities is still in ‘ongoing’
processes without proper arrangement and management to minimize their waste and if more licence mining permits are continuously being given by government.

### Table 7. PERI values of heavy metal in the sediments of Kundur Island coastal waters

| Station | Cu   | Cr    | Mn   | Ni   | Zn   | Pb    | RI   | Risk grade |
|---------|------|-------|------|------|------|-------|------|------------|
| 1       | 0.20 | 7.37  | 0.75 | 19.43| 4.46 | 2.94  | 35.15| Low        |
| 2       | 0.16 | 5.89  | 0.44 | 17.44| 2.51 | 1.71  | 28.15| Low        |
| 3       | 0.14 | 7.44  | 0.75 | 22.06| 3.63 | 3.72  | 37.74| Low        |
| 4       | 0.10 | 2.64  | 0.32 | 17.56| 1.48 | 1.83  | 23.93| Low        |
| 5       | 0.36 | 6.13  | 0.92 | 24.49| 3.30 | 4.19  | 39.39| Low        |
| Mean    | 0.19 | 5.89  | 0.64 | 20.20| 3.08 | 2.88  | 32.87| Low        |
| Min     | 0.10 | 2.64  | 0.32 | 17.44| 1.48 | 1.71  | 23.93| Low        |
| Max     | 0.36 | 7.44  | 0.92 | 24.49| 4.46 | 4.19  | 39.39| Low        |

4. Conclusion
Heavy metal concentrations in sediment of north-west coast of Kundur Island were found to be different for each station, where higher concentrations found in the proximity of mining activities. Concentrations of most heavy metals in seawater were positively correlated with those concentrations in sediment, except for Mn. The concentrations of analyzed metals in coastal waters around Kundur island still have low ecological risk and have no negative impacts on organisms living in those waters. However, continuous monitoring of the condition of this coastal waters is needed in line with the increasing mining activity along the coast and also infrastructure developments inland.

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