Quality indicators of metals pollution in the coastal sediments in Sangihe Islands Regency, North Sulawesi Province, Indonesia

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Abstract. Metal pollution in coastal areas is one of the focused environmental concerns. Sediment quality in the coastal zone reflects the long-term environmental status because it keeps a record of the development in the area. Quality indicators for metals in surface sediments were evaluated at 18 points of sediment sampling sites of these three bays using the Enrichment factor (EF) and the geo accumulation index (Igeo). Research locations in the coastal areas of Sangihe Island, Talengan Bay, Manalu, and Dagho Bay have values below the metal concentration at the background value. The Igeo category at the three research sites can generally be divided into two criteria, namely: Background concentration and unpolluted. For the types of Cr and Mg metals that are categorized as experiencing enrichment Minimum enrichment (Less), with an EF value <2, Moderate enrichment (Moderate), with a value of 2≤EF <5, and Enough enrichment (significant), with a value of 5≤EF <20. Whereas Co metal experiences enrichment of Very High enrichment (Very High), with a value of 20≤ EF < 40, and Extremely High (extremely high), with an EF value of ≥40, but still has an Igeo value <1 which indicates that the metal Co is still in the category of no pollution.

1. Introduction
Anthropogenic inputs have caused metal pollution in coastal areas. The metal biogeochemical cycle is a complex system in a coastal environment that involves physical, chemical, and biological processes. The sources include mining, metal product fabrication, solid waste disposal, fossil fuel combustion, and municipal/industrial waste effluent [1, 2] Increased mobility of toxic metals from pollutants to the environment is caused by acid rain [3]. As a consequence of industrialization and urban development with population growth, high concentrations of toxic metals are often found in sediments in many urban areas [4-8]. Toxic metals upon entering the coastal environment will accumulate in sediments because of the particles scavenging and settling. It is well known that fine-grained sediments are the main carriers of the toxic metals because of their higher specific surface area. Many studies have shown that coastal sediments are the repository for metal pollutants and provide time-integrated records of pollution history [6, 9–13].

Sediments can be considered as pollutant storage areas because after they enter the water column, adsorption to finer particles and complexation can cause their accumulation through deposition into sediments. Therefore surface sediment contamination is one of the most important quality indicators known in the coastal marine ecosystem. Besides this, there are benthic habitats such as coastal zone sediments, which play an important role in the marine environment. Moreover, heavy metals may be biologically up taken by marine organisms, reaching the bottom sediments after their death [19,20].
Co, As, Cr, Cd, and Mg are important biological elements because they have a significant function in the growth and health of biological systems [20], but they become toxic at higher concentrations [21]. Metals with different speciation and natural oxidation states in the earth's crust [22]. Depending on the type of rock, geophysical conditions, and geographical location throughout the world, the concentration of metals in sediments, soil, and water that exists as natural weather products from the Earth's crust can differ from one place to another [23, 24]. The trace amount of metals is a micronutrient essential for the growth and metabolism of many organisms [25]. However, excessive amounts of metal can be toxic to organisms and cause ecological toxicity [25]. In the twentieth-century anthropogenic metal inputs related to urbanization and industrialization have increased dramatically in coastal areas and are attracting attention around the world because about 80% of pollutants from human activities are incorporated into coastal environments [22, 26]. Therefore, metal pollution in coastal sediments has become a major environmental problem because it threatens the economic and ecological value of coastal areas. This paper summarizes the quality of metals in coastal sediments in the Sangihe Islands District, North Sulawesi Province, Indonesia, to find out indicators of the quality of metal pollution.

2. Methodology

2.1. Sampling methodology and analyses

The research location is on the Sangihe Island coastal area, Talengan Bay (Central Tabukan District) at 6 sediment sampling points; Manalu Bay (South Tabukan District) at 4 points of sediment sampling; and Dagho Bay, Bebu Bay, and Mahumu Strait (Tamako District) and Soweang Bay (South Manganitu District), hereinafter referred to as Dagho Bay and its surroundings at 8 points of sediment sampling. The study was conducted in August and September 2015. The sampling time is instantaneous, regardless of time or season. The selection of study locations was based on a preliminary report conducted in February 2015 [27], the results of the desk study, and the results of the Focus Group Discussion conducted in Tahuna, the Capital of the Sangihe Islands District. The chosen research location is a bay where there have been mariculture activities and has the potential of land for mariculture development.

Sampling Sediments of about 0.5 kg were taken using Van Veen Grab made of stainless steel at each sampling point. Sediments taken are sediments that are not in direct contact with Van Veen Grab and put into containers made of polyethylene that are first soaked in a solution of HNO₃ acid for 24 hours then rinsed using distilled water. Metal analysis of sediment samples followed APHA [28], using acid extraction methods [29]. The magnitude of the concentration of metals in sediments was measured using AAS Spectra AA 50 variant. Analysis of metals in sediments was carried out at the Soil Laboratory Installation, South Sulawesi Agricultural Technology Study Center in Maros.

2.2. Calculation of metal enrichment in sediment samples

As described below, several different methods were used to express metal pollution in the studied sediments, due to the lack of relevant background data for the unpolluted sediment in the area under investigation.

2.2.1 Metal enrichment factor (EF). Excess amounts of metals in the environment can cause pollution problems in coastal areas. Metal enrichment factor (EF) has been widely used as the assessment criteria to screen sediment metal concentrations of environmental concern to evaluate metal pollution in the sediments (e.g., [5-6, 30]). Because EF values can be used to distinguish natural metal concentrations from those of anthropogenic origin, it is widely used for sediment quality assessment (e.g., [5, 6, 30]). Mathematically, it is expressed as (e.g., [31]):

$$EF = \frac{\left(\frac{Me}{Al\ or\ Fe}\right)_{sample}}{\left(\frac{Me}{Al\ or\ Fe}\right)_{background}}$$
Where Me is the metal concentration of concern, \( \frac{Me}{Al\ or\ Fe} \). The sample is the metal to Al or Fe ratio in the sample, and \( \frac{Me}{Al\ or\ Fe} \). The background is the metal to Al or Fe ratio in the background. Al and Fe are used here as geochemical normalization elements because both Al and Fe are the most abundant elements in the earth’s crust [23, 24]. The metal background concentration should be derived from the sampling site if available [32]. However, the background data are not readily available in most of the cases. For metal EF calculation, when the local metal background values are not available, the upper continental crust values can be used as the background values [33]. In this case, metal concentrations in the upper continental crust [24] were adopted as alternatives for the background values. Metal EF values indicate the extent of metal enrichment in the sediments and can be used as sediment assessment reference criteria [34, 35, 36]. As a simple guideline, EF≈1 indicates natural crustal origin, whereas EF>10 suggests anthropogenic sources [37]. Zhang and Liu [34] also recommend that 0.5<EF<1.5 suggests that the metals may be entirely from crust natural weathering processes, whereas EF>1.5 indicates that a significant portion of the metal is delivered from non-crustal materials.

### Table 1. Classification of enrichment factors (EF).

| Value EF | Criteria         |
|----------|-----------------|
| EF<2     | Less (L)        |
| 2≤EF<5   | moderate (M)    |
| 5≤EF<20  | significant (S) |
| 20≤EF<40 | very high (VH) |
| EF ≥40   | extremely high (EH) |

2.2.2. The geoaccumulation index (Igeo). To understand the current status of the environment and the metal contamination with respect to the natural environment for the Sangihe Island coastal area. Calculation In the application, Igeo is used to determine the extent of contamination from anthropogenic on sediments [36] by comparing concentration values, which is now with a condition in the background value [37]. The degree of metal geoaccumulation is calculated using the method introduced by Muller [38] using the equation:

\[
Igeo = \log_2 \left[ \frac{a}{1.5 \times b} \right]
\]

Where,

- \( a \) = value of metal concentrations in sediments (μg g)
- \( b \) = metal concentration value in the background value (μg/g)

Factor 1.5 is a matrix used in minimizing of Possible effects of variations in sediment type in the background value that are possible to occur due to variations in lithogenic types in sediments [39]. The value of the division of Igeo categories is based on Muller [38] of water can be seen in Table 2.
Table 2. Geoaccumulation index classification (Igeo).

| Igeo   | Pollution intensity                      |
|--------|------------------------------------------|
| <0     | Background concentration (BC)            |
| 0≤Igeo<1 | Unpolluted (U)                         |
| 1≤Igeo<2 | Moderately to unpolluted (MU)           |
| 2≤Igeo<3 | Moderately polluted (M)                |
| 3≤Igeo<4 | Moderately to highly polluted (MH)      |
| 4≤Igeo<5 | Highly polluted (H)                    |
| ≥5     | Very highly polluted (VH)               |

3. Results and discussion

The concentrations of five metals detected in the seawater at all three sampling locations, including the three different sites from the Sangihe Islands coastal area, are summarized in Table 3.

Table 3. Comparison of metal concentrations in sediments Sangihe Island coastal area

| Location      | As (µg/g) | Co (µg/g) | Cd (µg/g) | Cr (µg/g) | Mg (µg/g) |
|---------------|-----------|-----------|-----------|-----------|-----------|
| Talengan Bay | nd        | 6.58-10.77| nd        | nd-6.91   | 0.20-0.48 |
| Manalu Bay   | nd        | 2.10-8.42 | nd        | 3.08-17.68| 0.31-0.50 |
| Dagho Bay    | nd        | 1.84-12.61| nd        | nd        | 0.17-0.53 |
| Background   | 13        | 19        | 0.3       | 90        | 15,000    |

nd = not detected
* from sedimentary rocks shales [40]

In general, metal concentrations at the study site have a value below the metal concentration at the background value. Original state (pristine) is the state described by the reference value (background value). This type of comparison is done to see the extent of contamination of metal against natural conditions. This study uses reference values from sedimentary rocks shale (sedimentary rocks shales) follows Turekian and Wedepohl [40] because of the grain size of the Sangihe Island Sea sediments found in this study the size of the dominant clay is dominant grain size in shale.

3.1. Enrichment factor

The source of metal contamination in sediments in the Sangihe islands can be explained using the enrichment factor (EF) approach. The categories of the Enrichment Factor (EF) calculation results show different results for each metal at the study site (Table 4). The enrichment value shows an indication of the non-natural influence of the presence of the metal in the sediment.

Table 4. Results of the Enrichment Factor (EF) category of each metal in the three study sites.

| Location      | As | Co | Cd | Cr | Mg |
|---------------|----|----|----|----|----|
| Talengan Bay  | L  | EH | L  | S  | S  |
| Manalu Bay    | L  | VH | L  | S  | M  |
| Dagho Bay     | L  | VH | L  | L  | M  |

L: Minimal enrichment (Less), with an EF value <2, M: Moderate enrichment (Moderate), with a value of 2≤EF <5, S: Enough enrichment (significant), with a value of 5≤EF <20, VH: Very High enrichment (Very High), with a value of 20≤EF <40, EH: Extremely high (extremely high), with an
EF value of ≥40.

Minimal enrichment (EF <2) that occurs indicates the source of metal input to the dominant sediment from natural sources, and medium enrichment (EF <5) indicates the source of metal input in the sediment has been contaminated by non-natural (anthropogenic) inputs [41]. EF values > 20 and EF values > 40 indicate that metals in sediments in the Sangihe Island coastal area are thought to have originated from anthropogenic activities around the Sea.

Metal in the aquatic environment will be absorbed by particles and then accumulated in the sediment. Heavy metals have the property of binding to other particles and organic matter and then settle to the bottom of the waters and unite with other sediments. This causes the concentration of heavy metals in sediments is usually higher than in waters [42]

The sediment fraction that contains a lot of heavy metals is an area that has a large surface area making it easier to interact, with interactions rich in organic matter so as to allow the process of adsorption, precipitation, and ion exchange [43]. The presence of sediment fraction will support the binding of heavy metals so that in a long time, the concentration of heavy metals will continue to increase. The results showed that the EF value in Co metal was in the EH and VH categories, it was thought to originate from industrial waste and domestic waste from households around the location, which was the main source of the Cobalt.

3.2. Geoaccumulation index
Calculations on the value of the Geoaccumulation Index (Igeo) generally have the same function with EF, which is used to see whether sediment samples undergo metal contamination other than input from nature or experience contamination from non-natural (anthropogenic) inputs. This research uses Igeo with the assumption that Igeo is used as an amplifier of the EF values that have been obtained in previous calculations. The category results from the Geoaccumulation Index for each metal in all study locations can be seen in Table 5.

| Location     | As  | Co  | Cd  | Cr  | Mg  |
|--------------|-----|-----|-----|-----|-----|
| Talengan Bay | BC  | U   | BC  | U   | U   |
| Manalu Bay   | BC  | U   | BC  | U   | U   |
| Dagho Bay    | BC  | U   | BC  | BC  | U   |

BC: Background concentration, with an Igeo value <0, U: Not polluted (unpolluted), with an Igeo value <2, MU: Moderately polluted (moderately to unpolluted), with a value of 1≤Igeo <2

Sangihe Island coastal area sediments generally do not experience the accumulation of non-natural in the three study sites. The Igeo category at the three research sites can generally be divided into two criteria, namely: Background concentration and unpolluted with an Igeo value <2. Igeo value of less than 2 is obtained by the type of Co, Cr metals in Talengan Bay, and Manalu Bay and Mg in all research locations and included in the Unpolluted category, which shows that the metal type is still categorized as a metal that has not been polluted. For the types of Cr and Mg metals that are categorized as experiencing enrichment Minimum enrichment (Less), with an EF value <2, Moderate enrichment (Moderate), with a value of 2≤EF <5, and Enough enrichment (significant), with a value of 5≤EF <20. Whereas Co metal experiences enrichment of Very High enrichment (Very High), with a value of 20≤ EF < 40, and Extremely High (extremely high), with an EF value of ≥40, but still has an igeo value <1 which indicates that the metal Co is still in the category of no pollution. If the metal is included in the moderately to an unpolluted category, it means that the Co metal content in the sediment is already in the stage of contamination or pollution. These results indicate that the metal has experienced non-natural input in the form of human activities
(anthropogenic). Non-natural inputs that occur generally take the form of human activities on land and at Seas such as shipping, agriculture, mining, household waste, and industrial waste [44, 45].

4. Conclusions
Research locations in the coastal areas of Sangihe Island, Talengan Bay, Manalu, and Dagho Bay have values below the metal concentration at the background value. Sangihe Island coastal area sediments generally do not experience the accumulation of non-natural in the three study sites. The Igeo category at the three research sites can generally be divided into two criteria, namely: Background concentration and unpolluted with an Igeo value < 2, Co, Cr metals in Talengan Bay, and Manalu Bay and Mg in all research locations and included in the Unpolluted category. Cobalt, despite having EF values in the Extremely High (EH) and Very High (VH) categories are still in the category of no pollution. Based on the analysis of the summarized information collected in this study, coastal sediment metal contamination should continue to raise our concern. It is not new that metal contaminations are still present in the world’s estuaries and coastal areas. The current issues are how we can effectively exercise the contamination assessment, environmental protection, ecosystem restoration, and sustainable development of the coastal areas. In the future, more attention should be paid to develop more precise contamination evaluation and strategies for contaminated coastal sites. Although the choice of geochemical normalization element is critical when evaluating the enrichment level of metals based on EF, a more accurate assessment should be achieved by choosing the local background values from adjacent sites with less anthropogenic disturbance.

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