Chemical characteristics of the surface sediment in the Java Sea

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Abstract. Sediments have been used as an indicator of environmental pollution because of its role as a final sink for most of materials. This study aims to describe the chemical characteristics of 13 metal contents in surface sediments and to predict the source of metal using enrichment factor (EF) approach. During the Java-Makassar-Flores (JMF) Triangle experiment in August 2015, about 11 surface sediment samples were collected using Multicore Octopus. Here we analyzed only three sediment samples at sampling station close to the coast of Java (MC-09), at mid-Java Sea (MC-11) and close to the coast of Kalimantan (MC-13). The result show that surface sediments consist of olive loose sand to silty clay containing organic materials, foraminifera shells, fragments of litic and quartz with dominant color of olive. The result of total metal analysis yields Al as the most dominant element in all stations. The enrichment factor (EF) revealed that the surface sediments of Java Sea enriched in As, Zn and Co at MC-09, with Zn and Ni MC-11 and with Cu, As, Zn, Ni and Cr at MC-13. All surface sediments have indicated medium contamination of Zn.

1. Introduction

Marine sediment is an accumulation of particulate, organic or inorganic materials that settles in the bottom of water body [1]. Marine sediment serves as a sink for material deposited from the seawater and produces varying characteristics that reflect their sources and sedimentation processes. Deposition of marine sediment involves weathering, erosion and transportation from the land by wind, water and ice into the sea before they accumulate on the bottom of the sea. As a result, marine sediments show distinct physical, chemical and biological characteristics that differ from basin to basin. Physical characteristics of marine sediments include grain size that ranges from fine to coarse grain, while biological characteristics comprise of marine organisms and terrestrial organic matter that are accumulated in the sediments. Chemical characteristics of marine sediments include pH, potential of reduction-oxidation (redox) (Eh), organic matter (OM) content and metal content [2].

The Java Sea that composes 7% of Indonesian Seas is a shelf sea that is affected by monsoon winds and its dimension can be described as rectangles with an average depth of 50 m [3] and [4]. Java Sea is located between Kalimantan, Java and Sumatra and it is connected to the Natuna Sea in the northwest through Karimata Strait, to the Indian Ocean in the south through Sunda and Bali Straits, and is bordered by Flores Sea in the east [5]. Human activities in surrounding area of Java Sea would affect water quality of the sea which then would be recorded in marine sediments.
Marine sediments play important role in understanding sources of pollutants due to their function as sink for heavy metals. Previous studies show that marine sediments provide information on the source of heavy metals suspected as natural and non-natural pollutants [6, 7, 8, 9, 10], bioaccumulation [11] and benthic habitat quality monitoring [12].

To date, information on chemical characteristics of Java Sea surface sediments is limited to studies that focus on coastal sediments. One such studies is conducted by [13] in estuaries and coasts in the northern part of East Java province and the information on the chemical characteristics of the surface sediment in terms of metal content in these locations are still limited. Considering Java Sea is also an important fishing ground for surrounding population, including sediment feeder mollusks and crustaceans, it is imperative to understand the characteristics of heavy metals in Java Sea sediments. This study aims to identify chemical characteristics of metals within the Java Sea sediments using of enrichment factor (EF) approach.

2. Materials and methods

2.1. Materials

This research is a part of multi-disciplinary study titled “JMF Triangle Experiment” in Java Sea, Makassar Strait and Flores Sea using R.V. Geomarin III. Thanks to fruitful collaboration between Marine Geological Institute (PPPGL) of the Ministry of Energy and Mineral Resources (ESDM), Department of Marine Science and Technology (ITK), Bogor Agricultural University (IPB) and Marine Fisheries Research Institute (BRPL) of the Ministry of Marine Affairs and Fisheries (KKP). The cruise involved field sampling campaign in Java Sea using multicore Octopus as well as gravity corer including the three sampling sites used in this study (figure 1): MC-09 (-6.565°S and 114.404°E, 32 m water depth) that represents mainland Java; MC-11 (-5.034°S and 112.849°E, 58.6 m water depth) that represents the middle part of Java Sea; and MC-13 (-4.241°S and 114.358°E, 34.4 m water depth) that represents Kalimantan. Those samples were acquired on 7th and 8th August 2015. The samples were obtained by cutting the top 10 cm of the sediments and covered with aluminum foil before stored in 4°C cold storage. Metal analysis was conducted from February to April 2016 at the Laboratory of Marine Chemistry, Natural Resources and Environment, Center of Isotope and Radiation Application, National Agency for Nuclear Power (BATAN), Jakarta.

![Figure 1. Map of sediment sampling at three locations of the research in the Java Sea.](image-url)
2.2. Methods

Sample preparation started by subsampling the middle part of the sample to avoid aluminum contamination. The subsamples then were dried in the oven at 100°C for 24 hours followed by grounding the samples. The powder then weighed before being destructed following [14]. Sample destruction involved dissolving samples with strong acid and heated them in a closed system using microwave digestion system to accelerate the dissolution process. The destruction steps used approximately 0.2 grams of samples that were put into Teflon vial and dissolving reagents composed of 9 ml HNO3 65% and 3 ml HCl 37%. Samples were then put into Milestone Start D Microwave labstation at 150° C and 700 watts for 30 minutes. After cooling the samples into 40°C, samples were transferred into 50 ml vial to be centrifuged. An aliquot of 2 ml was than obtained from the resulting filtrate and diluted by aquabidest by a factor of 5. After homogenization, the samples were measured using the Inductively Couple Plasma-Optical Emission Spectrometry (ICP-OES) instrument following [15] procedures. The measurement result (C) is then converted into metal concentration in sediment sample (Cs) by taking into account the total volume of solution in ml (V), dilution factor (D, which is 5) and weight of dry sediment sample in grams (W) with the following equation:

\[ Cs = \frac{C \times V \times D}{W} \]  

Measurement results were calculated using spreadsheet program to obtain elemental ratio as well as enrichment factor (EF). Elemental ratios was calculated to understand the influence terrigenous material and clay minerals to metal content in the sediments. The ratio used in this study is ratio of each element to Al, Fe and Ti. The determination of these three elements aimed to illustrate the influence of some indicators, such as Al can be used as a clay minerals indicator [16] as well as to determine the environmental conditions of sediment deposition [17]; Fe can be used as an indicator of material origin of terrigenous and minerals that formed in situ [18] as well as describes the condition of redox in marine sediments [19]; and Ti as an indicator terrigenous material influx [18, 20].

The enrichment factor (EF) is an indicator to assess the influence of anthropogenic sources of contaminants on the surface of soil or sediment. Enrichment Factor can be calculated by normalizing the metal concentrations in sediment layers to reference metal concentration [21]. The reference is usually element, preferable metal, that is stable in the soil or sediment and essentially linked to sediment grain (fine particles) and its concentration is unaffected by anthropogenic influences [21]. The metals that are often used in previous studies include Al, Fe and Mn. In this study, we used Al because it is conservative, thus its concentration is stable or not affected by chemical and biological processes [16] [22]. The calculation of EF value for this study follows [21]:

\[ EF = \frac{X(s)/Al(s)}{X(b)/Al(b)} \]

Where:

- \( X(s) \) : concentration of \( X \) metal in this study (μg/g),
- \( Al(s) \) : concentration of Al in this study (μg/g),
- \( X(b) \) : concentration of \( X \) metal in the background value (μg/g),
- \( Al(b) \) : concentration of Al in the background value (μg/g)

Background value is natural elemental concentration in soil or sediment that depends on mineral characteristics and composition of soil and sediment source. The value that is usually applied as background value is based on elemental concentration in the Earth's crust or soil. This study used elemental concentration in marine mud as background value following Wedepohl (1960) in [16] as Java Sea sediments consists mostly of mud. Classification of elemental enrichment in sediments follow [21] as presented in table 1.
Table 1. Classification of EF values, after [21].

| EF value | Classification       |
|----------|----------------------|
| EF <2    | Minimal enrichment   |
| $2 < EF <5$ | Medium enrichment   |
| $5 < EF <20$ | Moderate enrichment |
| $20 < EF <40$ | High enrichment     |
| EF > 40  | Very high enrichment |

3. Result and discussion

3.1. Characteristic of surface sediments
Java Sea surface sediments are dominated by soft, olive silty clay to loose sand compose of shell fragments, organic matter, lithic fragments and quartz. Sediment of MC-13 contains coarser grained material compares to the other two sites with sediment of MC-09 that was acquired off Rembang, Central Java, exhibits the finest-grained sediment among the three samples. Detail descriptions of each samples are given in table 2.

The northeastward trend of coarsening grain size is considered to be related to the distance of each sample locations to their source. Particle size of sediment is influenced among other by ocean current while settling of particular fragments with larger size would be faster than smaller ones. Generally, larger particles would be deposited near sediment source while finer particles would be deposited far from the source [23]. Thus, the source of MC-13 sediments is closer than the sources of MC-09 and MC-11 sediments.

Shell fragment contents in MC-09 and MC-13 that are closer to Java and Kalimantan are relatively higher than MC-11 that indicate higher productivity off Java and Kalimantan compare to the middle part of Java Sea. One of productivity indicator is organic matter content in sediments that might be originated from the land as well as the sea [24] [25]. Quartz and lithic fragments in marine surface sediment signify terrigenous input in sample location. Highest quartz content in MC-13 points to higher terrigenous input to the site compare to MC-09 and MC-11.

Table 2. Characteristics of Java Sea Surface Sediments.

| Locations | Depth | Characteristics of Java Sea Surface Sediment | Microscopic photos |
|-----------|-------|---------------------------------------------|--------------------|
| MC-09     | 32 m  | Silt to silty clay, olive 5Y, soft. Sediment consistency is higher in lower part which is composed of fine sands containing angular shell fragments. | ![Microscopic photo] |
| MC-11     | 58.6 m| Olive sandy silt, soft, composed of silt to fine sand grained shell fragments and organic matter. Sediment consistency is higher in the lower part, while grain size show coarsening upward and shell fragment content is decreasing upward. | ![Microscopic photo] |
| MC-13     | 34.4 m| Loose medium to fine sand, olive, soft with high water content, composed of shell fragments including foraminifera >50%, quartz 25%, and lithic fragments 25%. Grain size becomes finer downward to form silty sand to clayey sand, olive, soft and relatively drier than the upper part. The sand fraction is composed of coarse to very fine sand, that consist of shell fragments including foraminifera 50-60%, lithic fragments 25-30% and quartz 15-20%. Shell fragments decrease upward while quartz fragments increase upward. | ![Microscopic photo] |

Source: [26]
*Magnification: 40x - 1000x
3.2. Metal content in sediment

The result of ICP-OES measurement show that in general the area close to Java (MC-09) and Kalimantan (MC-13) have relatively higher metal contents compare to MC-11 that was acquired from the middle Java Sea (figure 2). The result yields Al as the element with the highest concentration in the measured Java Sea sediment samples with concentration ranges of 15184.73-28799.50 µg/g. High concentration of Al is considered to be related to the fact that Al is one of major rock forming elements which natural abundance in nature is 8.1% and ranked third after oxygen (47%) and silicon (28%) [27].

A closer look on figure 2 show that metal contents in the site off Java (MC-09) are higher than in the site off Kalimantan (MC-13). The high level of metal concentration in the surface sediments in this study was thought to be due to several factors, including metallic characteristics and surface sediments observed, the distance of the metal source to the study site and oceanographic conditions. The physical characteristics of sediment in the form of fine grain particle (MC-09: silt to silty clay) have greater metal binding ability than coarser grain particle (MC-11: sandy silt and MC-13: sand) due to larger surface area of fine grain particle in metal binding [28, 29, 30]. It can be said that the high metal content in the MC-09 was suspected to be associated with fine grain particle (clay).

Surface circulation in Java Sea plays a major role in metal distribution. Java Sea is influenced by monsoon cycle that is generated by different air pressure cells over Asia and Australia [31] that initiates surface current flows in the Java Sea [32, 33]. During west season, monsoonal currents flow eastward from Karimata Strait to the Java Sea while weaker east monsoonal currents flow from the Flores Sea to the Java Sea [34]. Those currents influence the distribution of metal-bearing particles from their source to study locations and also control particle size that are deposited in the seafloor. Friction due to near-
bottom current between surface sediment and water mass would release metals from surface sediment into the water column and resulted in lower metal content in surface sediment and higher metal content in water column.

The average current vectors obtained from INDESO model in 2015 show little seasonal variations (figure 3). During west season (February), the current flows eastward with stronger current flowing off Java and weaker currents flows in the middle of Java Sea and off Kalimantan (figure 3a). Transitional I season in May (figure 3b) is indicated by westward flowing current with stronger current in the middle part of Java and weaker currents off Java and Kalimantan (figure 3b). East season (August) is also characterised by westward flowing current although relatively weaker current is observed in the middle Java Sea compared to Transitional I season (figure 3c). Transitional II season shows similar eastward flowing currents as west season (figure 3d) with weaker currents flowing off Java.

![Figure 3](image-url)

**Figure 3.** Average current speed during (a) west season-February, (b) the transitional I-May, (c) east season-August and (d) the transitional II-November 2015 close to Java (●: MC-09), the middle of Java Sea (▲: MC-11) and close to Kalimantan (■: MC-13).

Metal concentration in marine sediments is partly determined by sediment source as well as current pattern. According to [35] and [36], high accumulation of metals in the sediments is a result of weak current which in the study sites occur during east season (figure 3c). Higher metal concentration in sample sites off Java and Kalimantan relative to middle Java Sea site suggest that there are other factors controlling low metal content in MC-11. One of the factor is resuspension of surface sediment by bottom which reduces metal contents in residual surface sediment. High metal contents that was detected in off Java and Kalimantan sites are suspected to be related to anthropogenic activities that include both land-based and marine-based activities such as shipping, mining, agriculture, household and industrial wastes [37, 38, 10].
Even though human activities in Java and Kalimantan might release heavy metal into Java Sea, the scope of this study is too broad to understand the source of metal input. Nevertheless, it is worth mentioning that anthropogenic activities in study area include fishing, shipping, ports, oil fields and power plant on northern coast of Java and mining activity in Kalimantan. In order to understand the impact of anthropogenic activities to metal content in marine sediments, we have to determine natural metal content in marine sediments that are mostly influenced by terrigenous material derived from weathering of rock formations in Java, Kalimantan, Sumatra and Sulawesi. Metal contents measured from study area are compared to background value of nearshore mud presented by Wedepohl (1960) in [16] as well as previous studies from around the world to identify the level of contamination in study area (table 3).

Table 3. Comparison of heavy metals concentrations in surface sediments of the Java Sea with the background value and other studies (μg/g).

| Metal | Java Sea | Background Value | Other studies |
|-------|----------|------------------|---------------|
| Cu    | 33.73 - 34.65 | 56 | 9.6 - 35.6 a |
| Pb    | 4.97 - 7.18 | 22 | 22.6 - 43.7 a |
| As    | 2.48 - 4.70 | 5 | 3.4 - 13.6 a |
| Zn    | 52.71 - 100.50 | 92 | 106 b |
| Ba    | 12.15 - 34.90 | - | 42 - 596 c |
| Ni    | 13.43 - 19.06 | 35 | 3.5 - 35.8 a |
| Cr    | 20.64 - 29.02 | 60 | 11.6 - 76.2 a |
| Co    | 4.48 - 10.15 | 13 | 11.2 b |
| Cd    | 1.24 - 1.98 | - | 0.02 - 0.24 a |
| Ti    | 44.89 - 109.41 | 5000 | 1700 - 5600 c |
| Mn    | 246.53 - 538.61 | 850 | 168 b |
| Fe    | 14276.98 - 24893.56 | 65000 | 29000 b |
| Al    | 15184.73 - 28799.50 | 84000 | 26000-108000 c |

a nearshore mud, Ba and Cd undetected Wedepohl (1960) in [16]
Luanhe River Estuary [39]
Vigo Ria [40]
Messolonghi Lagoon Complex [41]

Table 3 shows that the contents of all metals, except Zn, measured from Java Sea sediments are below background value. Zn concentration in Java Sea sediments (52.71-100.5 μg/g) is slightly higher than the background value (92 μg/g) indicating that Java Sea sediments received excessive Zn from surrounding landmass. The source of Zn is believed to be from anthropogenic activities in islands surrounding Java Sea.

Comparison of metal contents in Java Sea sediments to previous studies reveals that Cu, Cd and Mn measured in this study exceed the concentrations of corresponding metals in Luanhe River estuary [39] and Vigo Ria [40]. That condition indicates that Java Sea sediments receive higher level of Cu, Cd and Mn compare to Luanhe River and Vigo Ria. The higher concentrations of Cu, Cd and Mn are considered to be related to the particle size of Java Sea sediments (table 2). Fine-grained sediments containing organic matter that predominate Java Sea sediments would affect absorption of Cu, Cd and Mn due to larger surface area of fine-grained particle to bind metal or organic matter [29, 30]. Another factor that might influence higher concentration of Cu, Cd and Mn in Java Sea sediments is anthropogenic activity from surrounding area.
3.3. Ratio of metal elements in sediments
Elemental ratios of each metal show slight variation between sample sites (table 4). Ratios of elements to Ti yield higher values than ratios to Al and Fe that suggest higher terrigenous influx in sample sites, particularly MC-13. Titanium in this study is used as terrigenous indicator following [18] and [20]. The site off Kalimantan (MC-13) has the highest ratios except Ba/Ti which highest value is observed from MC-09. In general, elemental Ti ratios among sample sites are observed to be higher in MC-13 followed by MC-11 and MC-09. This result indicate that terrigenous influx from Kalimantan is higher than from Java and the influx from Kalimantan might reach MC-11. Maximum value of Ba/Ti that is observed off Java suggests high productivity in MC-09 that might be related to terrigenous nutrient supply considering Ba/Ti can be used as an indicator of productivity [19].

Table 4. Comparison of elemental ratios to Al, Fe and Ti measured from Java Sea sediments to the background value of nearshore mud presented by Wedepohl (1960) in [16].

| Elements | MC-09 | MC-11 | MC-13 | Background value |
|----------|-------|-------|-------|-----------------|
| Cu       | 0.0012| 0.0011| 0.0019| 0.00066         |
| Pb       | 0.00024| 0.00032| 0.00029| 0.00026         |
| As       | 163 x 10^-6| 98 x 10^-6| 141 x 10^-6| 59.5 x 10^-6 |
| Zn       | 0.00348| 0.00347| 0.00322| 0.00109         |
| Ba       | 0.0012| 0.0008| 0.0069| -               |
| Ni       | 0.00066| 0.00088| 0.00091| 0.00042         |
| Cr       | 0.00098| 0.00135| 0.00165| 0.00071         |
| Co       | 0.00035| 0.00029| 0.00026| 0.00015         |
| Cd       | 69 x 10^-6| 82 x 10^-6| 85 x 10^-6| -         |
| Ti       | 0.0037| 0.0035| 0.0025| 0.0595          |
| Mn       | 0.018702| 0.018781| 0.014049| 0.010119       |
| Fe       | 0.8643| 0.9402| 0.9895| 0.77            |

| Elements | MC-09 | MC-11 | MC-13 | Background value |
|----------|-------|-------|-------|-----------------|
| Cu       | 0.0013| 0.0012| 0.0019| 0.0008          |
| Pb       | 0.00028| 0.00034| 0.0003| 0.00033         |
| As       | 189 x 10^-6| 104 x 10^-6| 143 x 10^-6| 76.9 x 10^-6 |
| Zn       | 0.004| 0.0036| 0.0032| 0.0014          |
| Ba       | 0.0014| 0.0008| 0.0007| -               |
| Ni       | 0.00076| 0.00094| 0.00092| 0.0005         |
| Cr       | 0.0011| 0.0014| 0.0016| 0.0009          |
| Co       | 0.000408| 0.000313| 0.000271| 0.000402      |
| Cd       | 80 x 10^-6| 87 x 10^-6| 86 x 10^-6| -             |
| Ti       | 0.0043| 0.0037| 0.0025| 0.076           |
| Mn       | 0.0216| 0.0199| 0.0141| 0.0013          |
| Al       | 1.1569| 1.0635| 1.0105| 1.2923          |
The ratios of elements to Al and Fe are distributed evenly in the three sites (table 3). This study uses Al as an indicator of clay minerals following [16] and Fe as an indicator of terrigenous material, in situ mineral [18] as well as reduction-oxidation condition of the sediments [19]. The ratios show that elemental to Al ratios are slightly higher than elemental to Fe ratios in sites off Java and Kalimantan. These results indicate higher terrigenous influence associated with clay minerals in MC-09 and MC-13 in comparison to MC-11. Higher elemental to Fe ratios in MC-11 might suggest different redox condition in the middle part of Java Sea as oppose to the sites of Java and Kalimantan.

3.4. Enrichment factor

Enrichment factor (EF) is calculated to understand the source of metal contamination. Calculation was performed for all elements except Ba and Cd which background values are below detection limit. The calculated EF of Java Sea sediments yields the following EF range: off Java site (MC-09) 0.06 – 3.18; central part of Java Sea (MC-11) 0.05 – 3.16 and off Kalimantan (MC-13) 0.04 - 2.94 (figure 4).

Generally, Java Sea sediments can be classified into minimal enrichment (EF<2) and medium enrichment (2<EF<5). Minimal enrichment is observed for Cu, Pb, Ni, Cr, Ti, Mn and Fe in MC-09; Cu, Pb, As, Cr, Co, Ti, Mn and Fe in MC-11; and Pb, Co, Ti, Mn and Fe in MC-13. Medium enrichment is observed for As, Zn, Co in MC-09; Zn and Ni in MC-11; and Cu, As, Zn, Ni and Cr in MC-13.

EF values (EF<5) from the three sites suggest that most of metal contents in Java Sea sediments are derived from natural sources. This interpretation follows [42] that classified metals with EF<2 are predominantly derived from natural sources, while EF > 5 are considered to be contaminated by anthropogenic activities. Because most of metals measured from Java Sea sediments yield EF values between 2 and 5, there is a possibility that metal enrichments in study area are mixture of natural and anthropogenic sources.

The highest EF values (2.94-3.18) are observed in Zn in all sites ranging from, while the lowest EF values are observed in Ti (0.04-0.06) in all sites. The results point to a mixture of natural and anthropogenic sources of Zn, particularly in MC-09 site which is located off northern coast of Java that is home to various industries. This interpretation is supported by ratios of Zn to Ti (table 4) and comparison between Zn from off Java to background value (table 3). Ratios Zn/Ti of all samples suggest that Zn is derived from terrigenous material. The fact that Zn slightly exceed background value which is interpreted as the result of anthropogenic input to sample site.
4. Conclusion

The study of chemical characteristics of Java Sea sediments was conducted in three sites: off Java Sea (MC-09), central part of Java Sea (MC-11) and off Kalimantan (MC-13). Java Sea sediments are characterized by olive silty clay to sand containing lithic fragment, quartz, organic matter and shell fragments. Thirteen metals (Al, Fe, Mn, Ti, Ba, Cu, Pb, Ni, As, Zn, Cr, Co and Cd) were measured using ICP-OES and the result is calculated to obtain ratios of elements to Al, Ti and Fe and enrichment factor (EF). The EF values show minimal enrichment on Cu, Pb, Ni, Cr, Ti, Mn and Fe in sample from off Java (MC-09); Cu, Pb, As, Cr, Co, Ti, Mn and Fe in sample from central part of Java Sea (MC-11); and Pb, Co, Ti, Mn and Fe in sample from off Kalimantan (MC-13). Medium enrichment is observed in As, Zn and Co in MC-09; Zn and Ni in MC-11; and Cu, As, Zn, Ni and Cr in MC-13. The highest EF values are observed in Zn from all sites that are interpreted as derived from natural sources with additional input from anthropogenic activities.

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