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SEDIMENTS QUALITY BASED ON GEO-ACCUMULATION INDEX IN HEAVY METALS (Pb, Cu, and Cd) OF CENGKOK COASTAL WATERS, BANTEN BAY

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ABSTRACT

This area, which is located near the Banten Bay, is quite active with anthropogenic activities such as industry, fisheries, settlements, and shipyards that have the potential to pollute the environment with heavy metals. This study aims to determine the chemical environmental conditions of the waters and the concentration of heavy metals in the sediments of Banten Bay. Sediment samples were taken at five stations consisting of river and bay sections in April, May, July, August, and September 2019. Heavy metal content in sediments was analyzed according to APHA 2012 using hydrochloric acid and nitric acid destruction methods. The results of heavy metal concentrations in sediments were compared with ANZECC 2000 regulations and sediment quality was determined using the Geo-accumulation Index. The sediment at the observation site is dominated by the silt fraction. The concentration of heavy metals in this study was obtained in the form of lead (Pb) ranging from <0.05 to 6.408 mg / Kg, copper (Cu) ranging from 0.059 to 8.791 mg / Kg, and cadmium (Cd) ranging from 0.042 to 0.605 mg / K. While based on the level of heavy metal contamination in sediments using the Geo-accumulation index (I Geo), the value of all metal types in each month has a value <0.000 at all observation stations. Thus it is concluded that the concentration of heavy metals in the sediment is lower than the threshold determined during the observation. So that the metal concentration weight in the sediment has no significant effect on the environment.

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Keywords: Banten Bay; heavy metals; sediments

INTRODUCTION

Banten Bay is one of the active areas with industrial activities such as the Suralaya power plant, plastic raw material, chemical, steel, sugar, and shipping transportation activities that emit waste such as lead (Pb), cadmium (Cd), and copper (Cu). The presence of heavy metals in aquatic environments is a serious threat to ecosystems because of their toxicity, which cannot be destroyed naturally, and the ability to accumulate biologically in food chains. Heavy metals that get into the water can be in the form of dissolved metals or in the form of particles. Heavy metals have low

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solubility, are easily absorbed, and accumulate in basic sediments (Wuana & Okieimen, 2011; Zahran et al., 2015).

The distribution of suspended and sedimentary heavy metals depends on physical and chemical interactions in the environment. When the heavy metal that is suspended in water has exceeded the carrying capacity, the heavy metal will be deposited into the sediment. However, in some conditions, heavy metals in sediments can be released into the water. Therefore, the sediment serves as a carrier as well as a good heavy metal reservoir. Sediments are rock particles, organic charts, and minerals formed due to the deposition process. Sediments are believed to act as filters for many metals originating from the mainland before settling on the seabed and accumulating.

An increase in heavy metal concentrations in sediments can be an indicator of pollution caused by human activities and naturally through geological weathering. Predictions of heavy metal enrichment in sediments can be analyzed in various ways such as using the Geo-accumulation Index (I_{res}) which is used to determine the level of metal contamination in sediments.

This study analyzed the chemical environmental conditions of the waters and the presence of Pb, Cu, and Cd metals in the sediments of Cengkok waters, Banten Bay in April, May, July, August, and September 2019. This study of heavy metals is needed because there are many industries in that area that are expected to produce waste in the waters and are deposited in the sediments. This study is also expected to provide information on the level of sediment quality in the Banten Bay and can be used as a reference by both local governments and interested parties. Studies on heavy metals in sediments had been conducted in various locations such as Jakarta Bay (Takarina, 2010; Kusuma et al., 2015; Manalu et al., 2017), Pelabuhan Ratu Bay (Afifah, 2019), Semarang Waters of Central Java (Maslukah, 2013), Segara Anakan of Central Java (Hidayati et al., 2014), Gresik Waters of East Java (Lestari & Budiyanto 2013), and Kalabat Bay of Bangka Belitung (Arifin, 2011). Most of the results of these studies indicated that waters close to community centers such as industrial activities or ports have been contaminated with heavy metals, so studies of heavy metal pollution in sediments are important to do.

METHODS

Time and Location

The study was conducted from April to December 2019. The sampling of sediments was carried out at five stations in April, May, July, August, and September 2019 and took several supporting parameters such as physical and chemical parameters of the waters. Sediment sampling locations consist of five stations which can be seen in Figure 1.

Figure 1. Sampling Locations in the Cengkok Coastal Waters, Banten Bay

The observation stations location determined at five stations starting from the river, river mouth, to the sea. Station determination was based on river flow and community activities. Stations 1 and 2 are the centers of community activities that are located in river bodies and river mouths, while Stations 3, 4, and 5 are stations in the sea of Banten Bay.

Field Data Measurement and Sampling

Measurement of water quality parameters, which were partly carried out in-situ including temperature, salinity, dissolved oxygen (DO), transparency, and pH of waters. Water samples were taken at a depth of 1 m using Van Dorn and the samples were stored in HDPE bottles for TSS analysis and turbidity. Sediment samples taken on the surface layer of sediments using the Grab Sampler were then put into plastic and stored in a cool box during transportation to the laboratory.

Analysis of Heavy Metals in Sediments

Analysis of heavy metals in sediments was carried out using the 2012 American Public Health Association (APHA) method by the destruction of nitric acid and perchloric acid. Sediment samples that had been dried were weighed as much as 0.5 grams and were degraded with 5

mL concentrated HNO₃ at 100 °C until the volume of the solution became 2 mL. Then, 5 mL of concentrated $HClO₄$ and 10 mL of concentrated $HNO₃$ were added. It was heated at 150 °C until the white steam disappears. After digestion, the solution was then diluted to exactly 50 mL and filtered with 0.45 µm filter paper. Furthermore, heavy metal concentrations were determined using the Atomic Absorption Spectrophotometer (AAS).

Analysis of Sediment Type

Sediment grain size analysis was carried out by 10 fractions. Sedimentary fraction is divided into two, namely the first five fractions which are considered as sand (coarse to fine sand), and the second five fractions considered as dust and clay (coarse to fine). Separation of grain size of sediments included in the sand fraction was carried out using stratified filters of sizes 1, 0.5, 0.25, 0.125, 0.063, and <0.063 mm (Nugroho & Basit, 2014). Clay fraction with grain size <0.063 mm was analyzed using the pipette method to determine the weight of coarse silt, medium silt, fine silt, coarse clay, and fine clay.

Data Analysis

Data from measurements of metal concentrations were analyzed using Geo-accumulation Index $(I_{\alpha}$. The Geo-accumulation Index (I_{∞}) was used as a measure of pollution and determines the condition of contamination by metals in sediments (Shafie et al., 2013). The Geoaccumulation Index was developed by Muller (1969) with equation (1) as follows.

$$
I_{\rm geo} = \log_2 \frac{c x}{1.5 \times B n}
$$

Where *Cx* is the metal concentration on sediment, *Bn* is background value as stated by Turekian & Wedepohl (1961), 1.5 is the constant of the differential probability of background value caused by an external effect such as anthropogenic activities.

RESULTS AND DISCUSSION

Aquatic Environmental Conditions

The observation results of the aquatic environmental condition such as temperature, salinity, dissolved oxygen (DO), pH, transparency, turbidity, and TSS are presented in Table 1.

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Table 1. Aquatic Environment Parameters Data Observed by Each Month

The temperature of the waters during observations in April to August ranged from 28 - 32.5 °C, except in September which was very high, ranging between 33.4 to 35.3 \degree C. In general, sea surface temperature in the territory of Indonesia has a fairly wide range of 26 - 31.5 °C (Syaifullah, 2015). This shows that the temperature

of the observations included in the normal range. According to the Ministry of Environment (Kep. Men. LH) No. 51 of 2004, the good temperature for marine biota ranges from 28.0 to 32.0 °C, so that the water temperature is still in the good category for aquatic biota in April - August.

The salinity of the waters varies according to the observation area for example at Stations 3, 4, and 5 which are coastal areas ranging from 26 - 37 ‰, whereas in the estuary region (Station 1) and estuary mouth (Station 2) range from 0 - 32 ‰ (Table 1). This salinity variability can be influenced by seasons which will affect the water supply coming from the river and also the circulation of the bay seawater which is influenced by the Java Sea to the Sunda Strait and vice versa. This may cause a large variability in salinity in the estuary, namely low salinity due to high freshwater flow, and conversely high salinity due to low freshwater flow and high influence of seawater.

Dissolved oxygen at the observation site has a considerable variation in value. In the beachfront area (Stations 3, 4, and 5) ranged from 3.7 to 7.8 mg/l, while in the estuary region (Station 1) and estuary mouth (Station 2) ranged from 3.2 to 7.6 mg/l which can be seen in Table 1. Dissolved oxygen is used by aquatic organisms for the process of respiration and breaks down organic matter into inorganic by microorganisms, so that the increasing of organic matter in the waters and this activity can reduce DO levels in the waters (Simanjuntak, 2007; Patty et al., 2019). According to Patty (2013), water conditions during the rainy season have low DO levels due to the presence of waste originating from land entering the waters through a freshwater flow so that much dissolved oxygen is needed for the decomposition of the material. In general, DO values at the observation site are still in the good category according to Kep. Men. LH No. 51 of 2004 with a minimum DO value of 5 mg/l, but the DO value was not good in September at several observation stations.

The degree of acidity (pH) generally indicates conditions that tend to be alkaline or alkaline with a pH of around 8 (Millero, 2013), which is different from the relatively lower river water conditions. The large supply of fresh water in the West Season and Transition Season I causes the pH value at the observation site in April and May to be lower in the estuary region (Station 1), whereas the pH value at the East Season and Transition Season II at the observation location has a higher value (Table 1). In general, the pH value at the observation site is still in a good category according to Kep. Men. LH No. 51 of 2004 with a pH ranging from 7.00 - 8.50. Changes in the pH value in waters will also affect the chemical properties of water including solubility of heavy metals, alkaline waters tend to occur metal chelation into particles so that it will reduce the solubility

of heavy metals in the water that has the potential to settle into sediments.

Transparency value at observation station varied on the seashore (Station 3, 4, and 5) within $0.44 - 2.00$ m. At estuary (Station 1) and mouth estuary (Station 2) within a range of $0.07 - 0.30$ m as seen on Table 1. Low transparency value at Station 1 and Station 2 are influenced by the river and has the potential to contain high sediment and dissolved particles, organic and inorganic through run off the stream from the land. It causes the sunlight obstructed into the water (Indrayana et al., 2014). These seasonal changes are also contributed to the turbidity of water at the observation station because of the higher value of turbidity, the transparency value decreases (Reynaldi, 2018). Turbidity at observation station ranging between 0.5 – 240 NTU on the seashore (Station 3, 4, and 5) and ranging between $29 -$ 180 NTU on the estuary (Station 1) and mouth estuary (Station 2) as seen on Table 2. Turbidity in the waters can be caused by suspended and dissolved organic and inorganic material such as plankton and other microorganisms (Effendi, 2003). In general, transparency value and turbidity on the observation station are categorized as not good based on Kep. Men. LH No. 51/2004 with the maximum value of transparency value at 3 m and turbidity at 5 NTU.

TSS value ranging between 0.4 – 196 mg/l on East Season (July – August), 9.2 – 40 mg/l on Transition Season II (September), and 2.9 – 325 mg/l on Transition Season I (April – May) as shown in Table 1. But in general, TSS value is higher at estuary and mouth estuary than decrease through the seashore. Variation of high TSS value at the river mouth is allegedly caused by transport material by beach oceanography condition that dominated by sediments such as sand, gravel, and clay (Purbani et al., 2010). On the West Season and Transition Season I (April – May) a lot of particulate material came from the river by erosion caused by high rainfall. Otherwise, at East Season and Transition Season II, supply decrease. Water quality data in both physical and chemical waters varied based on location and time of the observation. However, in general, the condition of the waters was still quite good in supporting the marine life.

Sediment Characteristics

The results of sediment fractionation analysis show that the composition of the sediment was dominated by mud (silt and clay) in the fifth month (Figure 2).

Figure 2. Sedimentary Fractions at the Study Site in April (a), May (b), July (c), August (d), and September (e) 2019

The difference in sediment fraction can be influenced by several factors such as waves, currents, and tides (Nugroho & Basit, 2014; Zhou et al., 2015). The increase in the mud fraction (silt and clay) shows that the current is calm enough to precipitate the fraction. The increase in the finer fraction will affect the concentration of heavy metals in the sediment. The smaller the grain size of the sediment (silt and clay), the greater the content of heavy metals. This is because fine sediments have a larger surface area making it easier to bind heavy metals (Liang et al., 2019). Nevertheless in general the content of heavy metals in sediments was smaller than the standard.

Heavy Metal Concentration in Sediments Lead Metal (Pb)

Pb metal is a nonessential metal that is very dangerous and toxic to living (Palar, 2012). The results of the total Pb analysis in sediments at the study site ranged from <0.005 (below the instrument detection limit) to 6.408 mg/kg (Figure 3).

Figure 3. Pb Metal Concentration (mg/kg) in Sediments at the Study Site

The total Pb concentrations were very low in April and September, which had values below the instrument detection limit at all stations, while in May, July, and August the total Pb concentration was detected in the sediment even though not at all observation locations. Pb variability in sediments is thought to be due to the influence of input from the source and also the circulation of seawater which affects the transportation and deposition of material that has been exposed to dissolved Pb. Pb can be transported according to flow conditions or water currents with different strengths depending on the season. Pb can originate from industrial activities in the western part of Banten Bay and Pb transportation can enter the waters through the water from industrial activities other than liquid waste. Pb may also be derived from natural material components that are in the earth's crust, especially in the galena mineral (PbS), cerussite (PbCO₃), anglesite (PbSO₄), and pyromorphite $(Pb_5(PO_4)_3Cl)$ (Cheng & Hu, 2010). Pb from anthropogenic activities can come from mining, smelting, industry, waste disposal, coal burning, and lead-containing fuels (Needleman, 2004).

Pb heavy metal concentrations at the study sites obtained have a lower value when compared to research conducted by Hapsari (2017) which was also carried out in the waters of the Banten Bay at 60.76 - 107.61 mg/kg. Pb concentration in this study also has a lower value compared to polluted areas such as in Jakarta Bay with concentrations ranging from 24.86 - 59.32 mg/kg (Kusuma et al., 2015). The Pb quality standard value in sediments according to the Australian and New Zealand Environment Conservation and Council (ANZECC, 2000) about 50 mg/kg. All stations in the five-month research location were below the quality standard which indicates that the heavy metal Pb is still safe and does not have a bad impact on the surrounding environment.

Copper Metal (Cu)

Concentrations of heavy metal Cu at the study site in the five months ranged from 0.059 to 8.791 mg/kg. The results of the analysis of heavy metal Cu at each station are shown in Figure 4.

Figure 4. Concentrations of Cu (mg / kg) in Sediments at the Study Site

Cu concentrations in April had the greatest value compared to concentrations in May, July, August, and September. In April, the highest Cu concentration was around the river body (Station 1) of 8.791 mg/kg and the lowest concentration was around the bay (Station 5) with a value of 5.353 mg/kg. The concentration of heavy metals Cu decreased in May around bodies and river mouths (Stations 1 and 2) having concentrations ranging from 0.059 to 1.037 mg/kg and in the bays (Stations 3, 4, and 5) having values ranging from 0.716 to 0.981 mg/kg. In July, Cu concentrations ranged from 0.813 to 2.839 mg/kg. In August, Cu concentrations ranged from 2.687 to 5.196 mg/kg. In September, the highest concentration of heavy metal Cu was at Station 5 of 2.887 mg/kg. Metals in sediments are strongly influenced by inputs from various sources and also the condition of water motion which is affected by seasons. Cu concentrations are quite consistently found in both sediments of the river or in the sea. Cu sources from human activities such as the shipyard industry, wood processing industry, tin-zinc smelting industry, and household waste (Wang et al., 2014). In addition, Cu is also commonly used in port activities, the use of anti-fouling paint, a mixture of a brass material for household appliances, and is used in machine parts (Palar, 2012). Other sources of heavy metals Cu are natural sources such as chalcopyrite (CuFeS_2) and copper sulfide (CuS_2) (Effendi, 2003).

The concentration of Cu in this study was lower value than in the Jakarta Bay area with a concentration of 11.42 to 67 mg/kg. The concentration of heavy metal Cu in this study was also below the quality standard according to AN-ZECC (2000) with a value of 65 mg/kg in the five months so it can be suspected that the presence of Cu in the sediment does not have a bad impact on the surrounding environment.

Cadmium Metal (Cd)

Cadmium (Cd) is a heavy metal that has high toxicity. Like Pb, the Cd concentrations at the study sites in April, July, and August had values below the instrument detection limit, and only four locations were detected with a range between 0.042 to 0.605 mg/kg, and in September there was only one station with a value 0.225 mg/ kg. The results of the analysis of heavy metal Cd at each station are shown in Figure 5.

a April **¤** May □ July **a** August **a** September

This indicates that the source of Cd supply at the study site is relatively low, such as Pb. Potential sources of Cd from anthropogenic activities are agriculture, mining, combustion, and industrial wastewater (Järup & Åkesson, 2009). Cd metal is commonly used as a color paint material in ships, the plastics industry, the alloy industry, electronic equipment, batteries, textiles, and pesticides (Hossen et al., 2015). In addition, Cd can also come from natural materials that form complex bonds with ligands both organic and inorganic with different solubility levels.

The concentration of heavy metal Cd at the study site was still lower than in the Jakarta Bay area with a concentration of 0.32 to 3.49 mg/kg (Kusuma et al., 2015). The standard value of Cd in sediment according to ANZECC (2000) is 1.5 mg/kg, and when compared with the results of this study it was found that the concentration of heavy metal Cd at all stations in the four months was below the quality standard.

Geo-accumulation Index

The level of heavy metal pollution in sedi-

ments can be calculated using the Geo-accumulation Index (Igeo) shown in Table 2.

Month	Metals	Station				
		1	$\overline{2}$	3	$\overline{\mathbf{4}}$	5
April	Pb	< 0.000	< 0.000	< 0.000	< 0.000	< 0.000
	Cu	-2.941	-3.177	-3.333	-3.505	-3.657
	Cd	< 0.000	< 0.000	< 0.000	< 0.000	< 0.000
May	Pb	-8.393	-9.927	< 0.000	-7.863	< 0.000
	Cu	-10.148	-6.024	-6.376	-6.104	-6.559
	Cd	-2.338	-3.860	-2.317	-1.803	< 0.000
July	Pb	-5.332	-6.858	-8.122	< 0.000	-6.207
	Cu	-5.208	-5.858	-4.571	-4.670	-5.377
	Cd	< 0.000	< 0.000	< 0.000	< 0.000	< 0.000
August	Pb	-5.261	-4.148	< 0.000	-5.924	-4.932
	Cu	-3.699	-3.820	-4.385	-4.405	-4.651
	C _d	< 0.000	< 0.000	< 0.000	< 0.000	< 0.000
September	Pb	< 0.000	< 0.000	< 0.000	< 0.000	< 0.000
	Cu	-6.042	-5.326	-5.012	-4.651	-4.547
	Cd	-1.000	< 0.000	< 0.000	< 0.000	< 0.000

Table 2. Geo-accumulation Index of Heavy Metals (Pb, Cu, Cd) in Sediments in April, May, July, August, and September

The Geo-accumulation Index in the heavy metals Pb, Cu, and Cd at the study site at five months had values <0.000 at all observation stations, indicating that the heavy metals Pb, Cu, and Cd are in the uncontaminated category. Pollution of the marine environment is caused by the results of the activity of living organisms that enter the marine environment. The content of heavy metals in sediments is strongly influenced by natural and anthropogenic sources (Rustam et al., 2018).

CONCLUSION

Variability occurs in Pb, Cu, and Cd metals in sediments which are not only influenced by coastal processes but also metal sources. Metal sources are thought to provide an important role in the presence of metals in the waters of the bay and Cu, which shows a dominant supply compared to Pb and Cd. The concentration of Pb, Cu, and Cd metals in sediments in April, May, July, August, and September is lower than the AN-ZECC 2000 quality standard. Sediment quality in the observation station had uncontaminated by heavy metals based on the Geo-accumulation Index.

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