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Assessing Heavy Metal Contamination in Marine Sediments Around the Coastal Waters of Mimika Regency, Indonesia

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ABSTRACT
The heavy metal contamination has become a serious problem in the aquatic environment, including marine sediments. This study was aimed at analyzing the content of heavy metals in marine sediments, then assessing and evaluating the level of heavy metal contamination and its ecological risks. The sediment samples were taken using a grab sampler at six sites in the coastal waters of Mimika Regency, Indonesia. The TCLP testing method was used to determine the content of heavy metals in marine sediments. The results showed that the concentrations of Pb, Cu, Cd, and Hg in marine sediments ranged from <0.25 to 0.59 ppm, <0.02 to 0.54 ppm, <0.005 to 0.03 ppm, and < 0.0007 ppm. Only the concentrations of Cu on the Meoga and Puriri sites exceeded the quality standard in the Government Regulation of the Republic of Indonesia No. 18 year 1999. The results of the assessment of heavy metal contamination and its ecological risk potential indicate that the coastal waters of Mimika Regency are included in the category of low contamination and low ecological risks. The concentration of heavy metals in this study shows that the coastal waters of Mimika Regency are not contaminated by Pb, Cu, Cd, and Hg heavy metals.

Keywords: marine sediments; heavy metals; contamination assessment; pollution load index; ecological risk; Mimika waters

INTRODUCTION

Heavy metals constitute one of the pollutants in the aquatic environment. The heavy metal pollution in the aquatic environment has become a serious problem in water columns, sediments, and aquatic organisms. The heavy metals in the aquatic environment are one of the important intermediary sources for pollution in aquatic ecosystems [Aly Salem et al., 2014; Armid et al., 2014; Ouali et al., 2018;]. The heavy metal pollutants are the most dangerous because they can be toxic compared to other pollutants in the waters [Boran and Altinok, 2010] and their high potential to accumulate in the food chain in the aquatic environment [Ubeid et al., 2018]. Humans can be contaminated by toxic heavy metals through the consumption of aquatic organisms [Yakinov et al., 2007]. Therefore, the assessment and evaluation of the level of heavy metal pollution in an aquatic environment is an important study [Feng et al., 2011; Gao and Chen, 2012].

The high activity in the mining industry, agricultural activities and other activities in the Mimika Regency are feared to have an impact on the pollution in the coastal and marine waters [Tanjung et al., 2019]. There are seventeen watersheds in the Mimika Regency that are connected to the coastal and marine waters [IFACS, 2014], which have the potential to contain heavy metals from waste disposal from the mining industries, as well as the agricultural and anthropogenic activity. The increase in wastewater discharges allows for increased concentrations of heavy metals in coastal and marine areas [Suryono and Rochaddi, 2013]. In addition, an increase in the population of coastal areas can also affect the pollution loads in the coastal and marine environments [Arifin, 2011]. Indirectly, all activities on land can have an impact on reducing the water quality, so that it can disrupt the natural ecosystems in the coastal and marine waters.

Heavy metals are the elements that are very dangerous and toxic, compared to other pollutants [Boran and Altinok, 2010]. Their danger level depends on their concentration and toxicity [Chen et al., 2016]. Increasing the concentration of heavy metals in marine waters will cause the heavy metals needed for metabolic processes to change into toxic for the marine organisms. Various results from the study indicate that the marine sediments in some Indonesian waters have been contaminated by heavy metals at low to high levels of contamination [Nugraha, 2009; Hariyadi et al., 2017; Tjahjono and Suwarno, 2018]. Especially in the Mimika Regency waters, the studies on the heavy metal content in marine sediments have not been reported to date. The results of studies that have been reported only pertain to the heavy metal concentrations at the seawater surface [Tanjung et al., 2019]. A comprehensive assessment of marine waters quality can provide the information on the condition of marine waters, whether it has the potential to pose a threat, especially for the aquatic biota life [Hamuna et al., 2018].

This study was aimed at analyzing the content of four types of heavy metals in marine sediments, namely lead (Pb), copper (Cu), cadmium (Cd), and mercury (Hg). Then, the level of heavy metal contamination and its ecological risk in marine sediments was assessed and evaluateed. The results of this study are very important as basic information and additional data on the quality of marine waters of the Mimika Regency, Papua Province, Indonesia.

MATERIALS AND METHODS

Study area

The Mimika Regency is located in the south of Papua Province, Indonesia. The coastal waters of the Mimika Regency are dynamic waters with a depth of about 50 to 80 meters and have very high potential fisheries resources such as several types of shrimp and demersal fish [Pranowo et al., 2013]. The rivers in the Mimika Regency range from the Mimika Barat District to Agimuga District, and several watershed areas such as the Ombo, Ajkwa, Minajerwi, Otakwa, Agimuga, and Cemara watersheds which all flow to the coastal waters of Mimika. The width of the river in the downstream ranges from 100 to 150 meters, while the depth reaches 36 meters in the dry season or 58 meters during the rainy season [Mimika Regency Government, 2014].

This study was conducted on October 2016. The location for the marine sediment sampling sites in the coastal waters of the Mimika Regency included the coastal waters of the Mimika Barat District to the Mimika Timur Jauh District. The number of sampling sites is six, namely (1) Meoga waters; (2) Puriri waters (3) Poumako waters (4) Atuka waters (5) Keakwa waters; and (6) Kokonao waters. The sampling sites in this study are presented in Figure 1.

Collection and analytical procedures of sediment samples

The marine sediment samples were taken using a grab sampler (size 20 x 20 cm). The grab sampler was arranged so that the open condition was lowered by stretching the rope to hit the sediment on the marine surface. When the rope was pulled back, the grab sampler automatically collected the surface sediment. A total of 500 grams of sediment was put into polyethylene plastic for analysis in the laboratory.

The sediment samples were analyzed at the BINALAB Laboratory in Bandung, West Java, Indonesia, which is a laboratory accredited by KAN (National Accreditation Committed). In this study, the Toxicity Characteristic Leaching Procedure (TCLP) was used as a testing method to determine the heavy metal content in the marine sediments. The use of this method refers to the USEPA (United States Environmental Protection Agency) 1311 year 1992 (USEPA, 1992). The TCLP testing method has been used as an official test for the content of hazardous and toxic waste in Indonesia, according to the Government Regulation of the Republic of Indonesia No. 18 year 1999 (Government of the Republic of Indonesia, 1999).

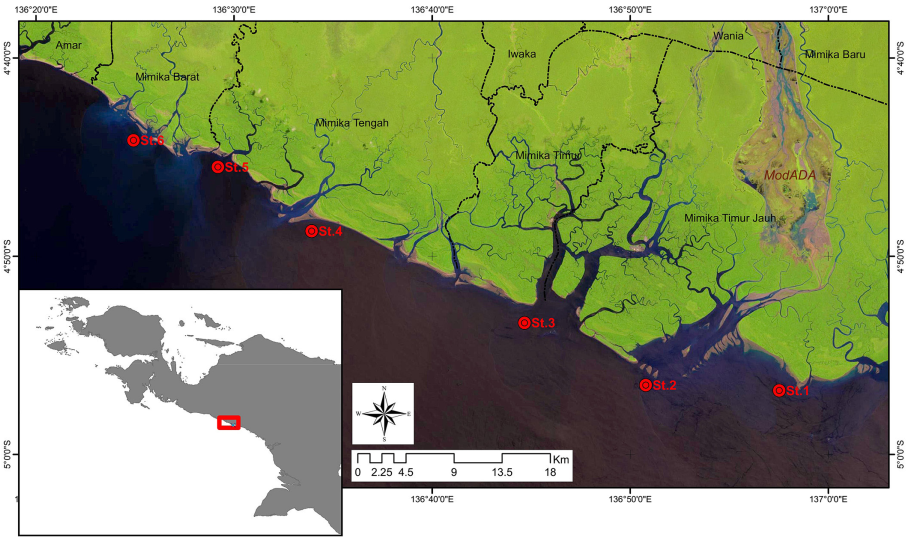

Figure 1. Location map of the sampling sites along the coastal waters of Mimika Regency, Indonesia; St.1 = Meoga site, St.2 = Puriri site, St.3 = Poumako site, St.4 = Atuka site, St.5 = Keakwa site, and St.6 = Kokonao site

Data analysis

Marine sediments quality

A descriptive analysis was used to determine the quality of marine sediments by comparing the concentration of heavy metals from the results of the laboratory analysis with the standard quality of the heavy metal concentrations in sediments. The quality standard refers to the Government Regulation of the Republic of Indonesia No. 18 of 1999 about the Management of Hazardous and Toxic Waste.

Contamination factor

Contamination factor (CF) is an indicator in the assessment of sediment contamination used to evaluate the level of contamination in the aquatic environment caused by toxic substances. [Hakanson, 1980]. The CF value for each heavy metal is determined by the following equation [Turekian and Wedepohl, 1961]:

$$CF = \frac{C_{metal}}{C_{background}} \quad (1)$$

where C_{metal} refers to the concentration of metals in sediment,
$C_{background}$ refers to the value of a reference metal, which is the value of the metal in the average shale.

Hakanson [1980] classified the level of sediment contamination based on the *CF* values into following four categories: low contamination ($CF < 1$), moderate contamination ($1 \geq CF < 3$), significant contamination ($3 \geq CF < 6$), and contamination that is very high ($CF \geq 6$).

Contamination degree

The contamination degree (*CD*) is the sum of the contamination factor (*CF*) in each sampling site [Hakanson, 1980]. *CD* values calculated according to the following formula:

$$CD_i = \sum CF_i \quad (2)$$

The *CD* values are classified into four categories as follows: low contamination ($CD < 6$), moderate contamination ($6 \leq CD < 12$), considerable contaminate ($12 \leq CD < 24$), and very high contamination ($CD \geq 24$) [Hakanson, 1980].

Pollution load index

The method proposed by Tomlinson et al. [1980] was used to determine the pollution load index (*PLI*) at each site. *PLI* for each study site was determined by the following equation:

$$PLI = (CF_1 \times CF_2 \times \ldots \times CF_n)^{1/n} \quad (3)$$

Tomlinson et al. [2009] classified *PLI* values into two categories: PLI > 1 indicates the polluted category, and PLI <1 indicates the unpolluted category.

Potential ecological risk index

The potential ecological risk index (*RI*) is commonly used to assess and evaluate the ecological risk of heavy metal pollution in sediments [Hakanson, 1980; Li et al., 2012]. The *RI* values were calculated according to the following formula:

$$RI = \sum E_i$$
$$E_i = T_i \times CF_i \quad (4)$$

where E_i is the monomial potential ecological risk factor for individual factors.
T_i refers to the toxic response factor of heavy metal (toxic response for Hg, Cd, Cu, and Pb were 40, 30, 5, and 5, respectively),
CF_i refers to the contamination factor for each heavy metal.

The potential ecological risks of marine sediments caused by the heavy metal contamination can be classified into five categories, namely: low risk ($E < 30$; $RI < 100$), moderate risk ($30 < E < 50$; $100 < RI < 150$), sufficient risk large ($50 < E < 100$; $150 < RI < 200$), very high risk ($100 < E < 150$; $200 < RI < 300$), and disaster risk ($E > 150$; $RI > 300$) [Gan et al., 2000].

RESULTS AND DISCUSSION

Concentrations of heavy metal in marine sediments

The concentrations of heavy metals in the marine sediments in the six study sites are presented in Table 1. The concentrations of heavy metals in the marine sediments were in the following order: Pb > Cu > Cd > Hg with concentrations range from <0.25 to 0.59 ppm, <0.02 to 0.54 ppm, <0.005 to 0.03 ppm, and <0.0007 ppm, respectively. The highest Cu concentrations were observed at Meoga site and the lowest in Poumako, Atuka, Keakwa, and Kokonao sites. The highest Pb concentration was found in Puriri site and the lowest in Poumako, Atuka, Keakwa, and Kokonao sites. The highest Cd concentration occurred at Meoga site and the lowest at Atuka and Keakwa sites. The concentrations of heavy metals in several sites, such as Hg in all sites, Cu and Pb in Poumako, Atuka, Keakwa, and Kokonao sites, and Cd at Atuka and Keakwa sites are not detected by the instruments used (below the detection limit). This indicates that the concentration of these heavy metals in marine sediments is very low.

On the basis of the quality standards of heavy metal concentration in the Government Regulation of the Republic of Indonesia No. 18 year 1999), only the concentrations of Cu at Meoga and Puriri sites have exceeded these standards. The high Cu concentration in the Meoga and Puriri sites occurred because the two sites are near the Ajkwa river estuary, which contains the deposits from the mining industry (Modified Ajkwa Deposition Area or ModADA). Although Cu is classified as an essential metal, it can be toxic to marine organisms, especially if the concentration is very high. For example, Bryan [1976] reported that aquatic organisms from the Crustaceans and Mollusks family can die within 96 hours if the Cu concentrations range from 0.17 to 100 ppm and 0.16 to 0.5 ppm, respectively. Besides Cu, the concentration of Pb in Meoga and Puriri sites is also quite high, compared to other sites.

Tanjung et al. [2019] reported the concentrations of heavy metals (Cu, Pb, and Cd) in seawater in the coastal waters of Mimika Regency. Compared to the results of this study, the concentration of heavy metals Cu, Pb, and Cd in the marine sediments is higher than the concentration

Table 1. Heavy metal concentration in marine sediment of coastal waters of Mimika Regency (in ppm)

Heavy metals	Standards (ppm)*	Sites					
		Meoga	Puriri	Poumako	Atuka	Keakwa	Kokonao
Cu	0.19	0.54	0.37	<0.02	<0.02	<0.02	<0.02
Pb	2.5	0.57	0.59	<0.25	<0.25	<0.25	<0.25
Cd	0.05	0.03	0.01	0.01	<0.005	<0.005	0.01
Hg	0.01	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007

Remarks: < are below the detection limit; *Government Regulation of the Republic of Indonesia No. 18 year 1999 about the management of hazardous and toxic waste in seawater. This is due to the dilution of heavy metals in seawater and there is no high accumulation due to the influence of ocean currents. In addition, the heavy metals in the water column can accumulate in the marine sediment [Effendi et al., 2016; Ubeid et al., 2018] because it has a low solubility [Effendi et al., 2016].

The results of previous studies showed variations in the concentration Cu, Pb, Cd, and Hg of heavy metals in the marine sediments in Indonesian waters (Table 2). The results of this study showed lower concentrations of Cu, Pb, Cd, and Hg than in Banten Bay [Suwandana et al., 2011], West Sumbawa waters [Akbar et al., 2015], coastal waters of Jambi [Susantoro et al., 2015], and Tangerang waters [Hariyadi et al., 2017]. Likewise, higher Cu, Pb, and Cd concentrations were reported by Arifin [2011], Permanawati et al. [2013], Suryono and Rochaddi [2013], Effendi et al. [2016]. In turn, the Cu, Pb, and Cd concentrations in the waters of Northeast Sulawesi [Ahmad, 2009], and the Pb concentrations in Popoh Bay waters [Yona et al., 2018] were lower than the results of this study. The Pb and Cd concentrations in Demak estuary [Tjahjono and Suwarno, 2018] and Hg in the coastal waters of Semarang [Suryono and Rochaddi, 2013] tended to show relatively the same heavy metal concentrations as the results of this study.

Sediments contamination assessment

The contamination factor (*CF*), contamination degree (*CD*) and pollution load index (*PLI*) values are presented in Table 3. The average *CF* values of heavy metals in marine sediments were in the order: Cd > Pb > Cu > Hg with an average value are 0.0389, 0.018, 0.0036, and 0.0018, respectively. The *CD* values for each study site ranged from 0.0314 to 0.1423, where the highest *CD* value occurred at Meoga site, while the lowest at Atuka and Keakwa sites. According to the category of *CF* and *CD* values by Hakanson [1980], CF for each heavy metal includes the low contamination category (*CF* < 1), and the *CD* for each study site also includes the low contamination category (*CD* < 6). The concentration of Cu which has exceeded the quality standards at Meoga and Puriri sites is considered not yet dangerous and is toxic to the aquatic organisms.

The *PLI* value of heavy metals in the marine sediments in the six study sites is very low (Table 3). The *PLI* value showed that the six study sites classified as unpolluted (*PLI* < 1) were considered to indicate a perfect state (*PLI* = 0) [Tomlinson et al., 1980]. The *PLI* value is very important because it can provide the data and understanding of the environmental quality and its changes, including providing the information and

Table 2. Comparison of the results of this study with the concentration of heavy metals from previous studies in several waters in Indonesia

Sites	Concentration (ppm)				References
	Cu	Pb	Cd	Hg	
Mimika	<0.02–0.54	<0.25–0.59	<0.005–0.03	<0.0007	Present study
Northeast Sulawesi	<0.001–0.004	<0.001–0.002	<0.001–0.001	–	Ahmad, 2009
Kelabat Bay, Jambi Island	0.18–6.43	1.04–22.01	0.47	–	Arifin, 2011
Banten Bay	3.29–10.14	<0.006–17.34	0.27–3.13	0.037–0.235	Suwandana et al., 2011
Jakarta Bay	15–169.5	14–58.1	0.012–0.75	–	Permanawati et al., 2013
Semarang coastal waters	1.7–11.91	3.94–10.45	–	<0.00084	Suryono and Rochaddi, 2013
West Sumbawa waters	16–101.7	<0.5–25.8	<0.1	< 0.02–0.03	Akbar et al., 2015
Jambi coastal waters	2–12	5–7	<0.5	<0.01	Susantoro et al., 2015
Mahakam delta	27.66	27.59	1.07	–	Effendi et al., 2016
Ambon Island	–	0.084–2.09	0.541–1.196	–	Tupan and Uneputty, 2017
Tangerang waters	4.73–45.73	<0.20–14.32	<0.10	0.94–14.10	Hariyadi et al., 2017
Tanjung Emas Port, Semarang	4.7–51.85	3.57–22.4	0.16–19.75	–	Tjahjono et al., 2017
Popoh Bay	–	0.05–0.44	0.02–0.12	–	Yona et al., 2018
Demak estuary	–	0.256–0.682	0.005–0.058	–	Tjahjono and Suwarno, 2018

Remarks: < are below the detection limit; – are not available

Table 3. The contamination factor (CF), contamination degree (CD) and pollution load index (*PLI*) of heavy metals in marine sediments

Sites	*CF*				*CD*	*PLI*
	Cu	Pb	Cd	Hg		
Meoga	0.012	0.0285	0.1	0.0018*	0.1423	1.49625E-08
Puriri	0.0082	0.0295	0.0333	0.0018*	0.0728	3.53727E-09
Poumako	0.0004*	0.0125*	0.0333	0.0018*	0.048	8.10185E-11
Atuka	0.0004*	0.0125*	0.0167*	0.0018*	0.0314	4.05093E-11
Keakwa	0.0004*	0.0125*	0.0167*	0.0018*	0.0314	4.05093E-11
Kokonao	0.0004*	0.0125*	0.0333	0.0018*	0.048	8.10185E-11
Total	0.0218	0.108	0.2333	0.0108	-	-
Average	0.0036	0.018	0.0389	0.0018	-	-

Remarks: * The detection limit is used as a reference for heavy metal concentration.

advice to the government for the management of environmental pollution [Mohiuddin et al., 2010].

Ecological risk assessment

Ecological risk assessment as used to identify the ecological risk level of heavy metals in the coastal waters of the Mimika Regency. The value of ecological risk consists of the potential ecological risk factor (*E*) of each heavy metal and the potential ecological risk index (*RI*) for each study site, as presented in Table 4. On the basis of the *E* value in this study, the ecological risk factor of heavy metals was in the following order: Cd > Pb > Hg > Cu with an average value of 1.1667, 0.09, 0.07, and 0.0183, respectively. The ecological risk index ranges from 0.6347 to 3.2725; the highest value occurred at the Meoga site, while the lowest at the Atuka and Keakwa sites.

According to *E* and *RI* value categories by Gan et al. [2000], the E value of each heavy metal is included in the low ecological risk category ($E < 30$). Likewise, the RI value of each study site is included in the low ecological risk category ($RI < 100$). The E and RI values indicate that the concentration of heavy metals in the marine sediments did not have a significant ecological impact on all study sites.

CONCLUSIONS

The results of this study have provided important information about the concentration of some heavy metals in the marine sediments in the coastal waters of the Mimika Regency, Indonesia. On the basis of the analysis, only the Cu concentration in the Meoga and Puriri sites have exceeded the quality standards, while the concentration of Pb, Cd, and Hg are still below the quality standard in the Government Regulation of the Republic of Indonesia No. 18 year 1999. Both sites are located near the estuary of the Ajkwa river, which is very much related to the disposal of tailings from the mining industry. The results of the assessment of sediment contamination indicate that the value of contamination factor (*CF*)

Table 4. The potential ecological risk factor (*E*) and the potential ecological risk index (*RI*) of heavy metals in marine sediments

Sites	*E*				*RI*
	Cu	Pb	Cd	Hg	
Meoga	0.0600	0.1425	3	0.07*	3.2725
Puriri	0.0411	0.1475	1	0.07*	1.2586
Poumako	0.0022*	0.0625*	1	0.07*	1.1347
Atuka	0.0022*	0.0625*	0.5*	0.07*	0.6347
Keakwa	0.0022*	0.0625*	0.5*	0.07*	0.6347
Kokonao	0.0022*	0.0625*	1	0.07*	1.1347
Total	0.1099	0.54	7	0.42	-
Average	0.0183	0.09	1.1667	0.07	-

Remarks: *The detection limit is used as a reference for heavy metal concentration and contamination degree (*CD*) was included in the low contamination category. Likewise, the value of the pollution load index (*PLI*) was included in the unpolluted category. The results of ecological risk assessment (*E* and *RI*) showed that the concentration of heavy metals in the marine sediments in all study sites had very low ecological risks. From the results in this study, it can be concluded that the high activity of the mining industry, agricultural activities and other activities have not significantly affected the heavy metal (Cu, Pb, Cd, and Hg) contamination in the coastal waters of the Mimika Regency.

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