Source Identification, Bioavailability, and Risk Assessment of Heavy Metals Pb, Cu, and Zn in Surface Sediments of Kelabat Bay, Bangka Island

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Abstract

Although concentrations of many heavy metals has been measured totally, they may not give a good indicator for environmentally hazard to organism. The purpose of this study is to identify sources, determine bioavailability, and assess risk based on the geochemical fractionation of heavy metals Pb, Cu and Zn on the surface sediments of Kelabat Bay, Bangka Island. Fractionation of heavy metals was analyzed by sequential extraction. The concentrations of heavy metals Pb, Cu, and Zn in sediments ranged from 8.86-29.21 mg.kg^{-1} (average 16.85 mg.kg^{-1}), 0.16-9.54 mg.kg^{-1} (average 4.39 mg.kg^{-1}), and 25.58-237.24 mg.kg^{-1} (average 71.99 mg.kg^{-1}). Pb and Zn in Kelabat Bay are more bound to non-residual fractions (F1+F2+F3) or non-resistant with a range of 60.63-89.87% and 47.98-84.66% that are mainly come from anthropogenic activities. Cu tend to be stored or bound to the residual fraction (F4) with a proportion of 97.7-100% meaning that it comes from natural sources. Based on the Risk Assessment Code (RAC), Pb have a low to moderate risk in the environment and Zn heavy metals are not at risk to low. While Cu has no risk to the environment. These conditions indicate the potential for biological availability (bioavailability) of Pb and Zn in the inner bay waters.

Keywords: *marine pollution, risk assessment, geochemical, sediments,*

Introduction

Pollution in the marine environment has become a problem on a local, regional and global scale. Heavy metals have specifications that are difficult to degrade, accumulated in the body of organisms, high toxicity, and carcinogenic (Bastami *et al*., 2018). Water sedimens generally used as an indicator in monitoring water pollution for it can store most of the contaminants. The total heavy metal concentration can reflect the level of pollution in the marine environment, although they are not a good indicator for assessing bioavailability of heavy metals in organisms (Arifin and Fadhlina, 2009; Lu *et al*., 2014). Heavy metals that flow to the marine environments due to human activities show high mobility and are associated with carbonate, oxide, hydroxide and sulfide sedimentary phases (Bastami *et al*., 2017). Research on geochemical fractionation of heavy metal has been carried out widely in coastal and marine areas such as the Malacca Strait (Yap *et al*., 2003), Persian Gulf (Karbassi *et al*., 2005), Jakarta Bay (Takarina *et al*., 2008; Arifin and Fadhlina, 2009), Berau- East Kalimantan Delta (Situmorang *et al*., 2010), Rodrigo de Freitas-RJ / Brazil Lagoon (Fonseca *et al*., 2011), and Banten Coastal Waters (Takarina *et al*., 2013).

Kelabat Bay located in the northern part of Bangka Island, Bangka Belitung Islands Province. It is an estuary of several rivers and constantly receiving input of organic and inorganic materials. The rivers continuesly flow out the materials from the human activities, such as mining, agriculture, plantation, and community settlement. Offshore tin mining activity will result in increased distribution and concentration of heavy metals in the water column which then be adsorbed on suspended particles and deposited on the sediment. There are also fishing activities as a livelihoods of the community in Kelabat Bay. The input from these activities is thought to have ecological pressure due to anthropogenic input. Research on the geochemical of heavy metals in sediments from Kelabat Bay has not been carried out. The purpose of this study is to identify sources, determine bioavailability, and assess risk based on the geochemical fractionation of heavy metals Pb, Cu and Zn on the surface sediments of Kelabat Bay, Bangka Island.

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Materials and Methods

The research was carried out in the inner part of Kelabat Bay (Figure 1.) April 2018. Collection of sediment samples was carried out at 6 stations. The surface sediments (±2-4 cm) are collected using a grab sampler, then put in a plastic container that has been pre cleaned with acid solution. Furthermore, it is stored in a cool box at -4^{0}C (Takarina *et al*., 2013; Bastami *et al.,* 2017).

Sediment samples for geochemical analysis are dried first at 80°C, then mashed with mortar (Liu *et al*., 2015; Bastami *et al*., 2017). The sediment samples are then analyzed by sequential extraction (Takarina, 2014). F1 (exchangeable fraction) was extracted using 1g sample then continued with the addition of 5 mL glacial acetic acid. The sample was left to stand for 30 seconds then shaken 30 seconds. Samples were incubated at room temperature overnight. The samples were centrifuged 5-30 minutes (3000rpm) and decanted. The filtrate was analyzed with AAS.

F2 (reducible/Fe-Mn oxide fraction) was obtained by adding 10 mL 0.04 M $NH_2OH.HCL$ to the sample. The sample is heated 80^{0}C (24 hours) then cooled. The samples were centrifuged 5-30 minutes (300rpm) and decanted. The filtrate was analyzed with AAS.

F3 (oxidizable/organic fraction) is obtained by adding 3 mL 0.02M HNO3 and 2 mL H_2O_2. Sample was then heated for 2 hours and added 3 mL H_2O_2 and reheated for 3 hours. Added 5 mL 1.2M NH_4OAc in 10% HNO_3 to the samples then decanted and analyzed with AAS.

F4 (residual fraction), 0.5 g residual of F2 plus 4 mL aquaregia (HCL:HNO_3= 3:1). Samples plus 6 ml of HF. Next, the sample is heated to dry and added 4 mL aquaregia (HCl:HNO_3= 3:1). Samples plus 6 ml of HF. The sample was diluted with 10 mL 0.1N HCL. Next the samples were analyzed with AAS.

Results and Discussion

The concentrations of Pb in the sediments of the Kelabat Bay (Table 1.) ranged from 8.86-29.21 mg.kg^{-1} (average 16.85 mg.kg^{-1}). For Cu, concentrations ranged from 0.16-9.54 mg.kg^{-1} (an average of 4.39 mg.kg^{-1}). Furthermore, Zn concentrations ranged from 25.58-237.24 mg/kg (average 71.99 mg.kg^{-1}). These indicate concentration increase compared with those of previous studies in the same location (Arifin, 2011). This increase in concentration is thought to be due to ongoing mining activities, resulting in such heavy metals accumulation in the sediment. This tin mining activities produce liquid waste containing heavy

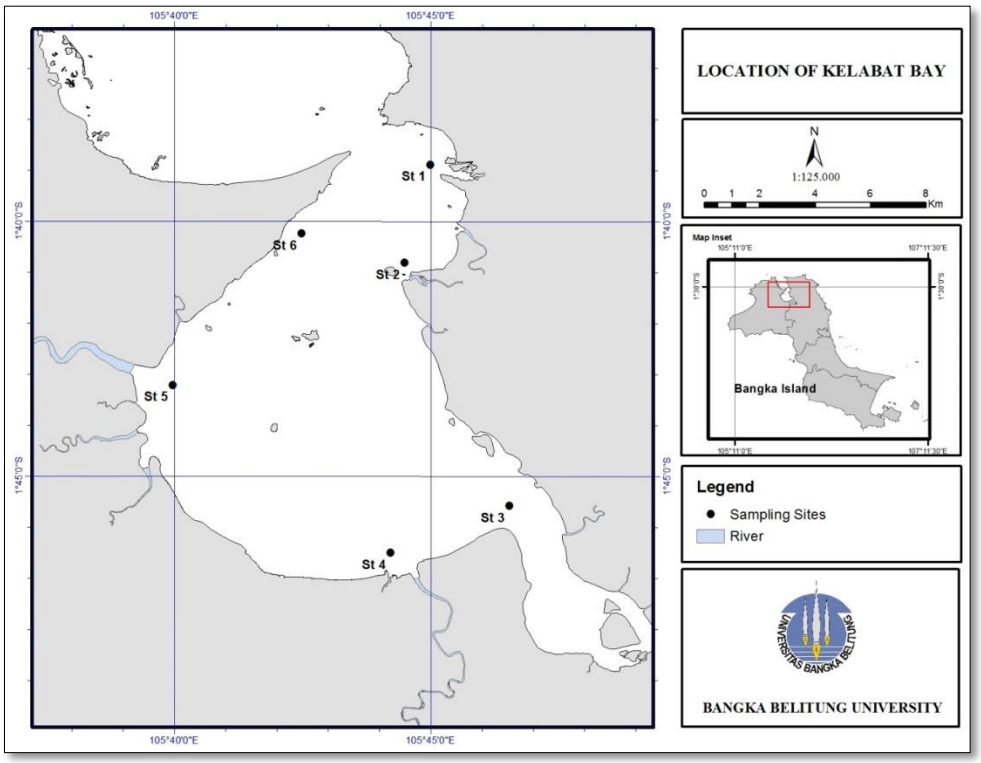

Figure 1. Research location in Inner Kelabat Bay Waters

metals such as Cr, Cd, Hg, Cu, Pb, Zn, and Sn (Prianto and Husnah, 2009; Arifin, 2011).

The concentrations of Pb is still higher (Table 1.) compared with those found in the waters of the Jambi Sea (Susantoro, 2015), Natuna waters (Sagala *et al*., 2014), coastal Gresik, East Java (Lestari and Budiyanto, 2013) and coastal waters of Semarang (Rositasari and Lestari, 2013). Conversely, the concentration of Pb heavy metals in Kelabat Bay is still lower compared to Jakarta Bay waters (Kusuma *et al*., 2015). The concentrations of Cu are relatively lower compared to those found in the waters of Jakarta Bay, Jambi Sea, Natuna and Gresik coast. The concentration of Zn heavy metals shows relatively lower concentrations compared to the waters of Jakarta Bay, the Gresik-Java Tmur coast, and the Semarang coast. However, they are still relatively higher compare to the Jambi Sea.

This result (Table 1.) indicates/shows that Pb and Cu are below to both those used in ISQG and PEL of CCME and the PEL ANZECC/ARMCANZ of Australia and New Zealand. The concentration of Zn observed generally lower than that of the two guidelines although some have exceeded.

In addition to the measurement of total concentration, the ratio of metals in different geochemical parts of the sediment (stable and unstable parts), can be measured to determine the origin of metals in the environment (Bastami *et al*., 2018). Those derived from human activities such as household, agricultural, mining, and industrial wastewater discharges tend to be labile, whereas those coming from natural sources (weathering of rocks). Fractionation of Pb, Cu and Zn in the sediment waters consisted of exchangeable fractions, reducible (Fe-Mn oxide) fractions, oxidizable (organic) fractions, and residual fractions (Figure 2.). The residual fraction (F4) of heavy metals is mainly composed of crystalline or lithogenous fraction minerals from sediments (Liang *et al*., 2018). The proportion of residual fraction (F4) in Pb, Cu and Zn is ranged between 10.13-39.37%, 97.7-100% and 15.34-52.02% respectively. Cu tends to be stored or bound to the residual fraction. In contrast, Pb and Zn tend to be labile fraction. This shows that Cu mainly come from natural sources. Heavy metals found in residual fractions (F4) are aquatic/non-bioavailable biota (Ramirez *et al*., 2005; Situmorang et al., 2010; Gu, 2018). The high percentage of residual fraction of heavy Cu metals in resistant, bind strongly to sedimentary minerals (Liu *et al*., 2015). In general, this residual fraction shows low toxicity and is not available to be absorbed by sediments was also found in the waters of the Berau Delta, East Kalimantan (Situmorang *et al*., 2010), Guangdong east coast, South China (Gu and Lin, 2016), North Persian Gulf (Neyestani *et al*., 2016) and the Caspian Sea (Bastami *et al*., 2018).

Non residual fraction/ labile fraction is the sum of F1 (exchangeable fraction), F2 (reducible fraction) and F3 (oxidizable fraction). Non-residual fractions (unstable parts) are easily absorbed in

Table 1. Comparison of heavy metals in Kelabat Bay sediments with the quality guidelines and other locations

Location	Time	Pb (mg.kg^{-1})	Cu (mg.kg^{-1})	Zn (mg.kg^{-1})	Reference
Kelabat Bay	April 2018	8,86-29,21 (16,85)	0,16-9,54 (4,39)	25,58-237,24 (71,99)	This research
Jakarta Bay	September 2014	24,86-59,32 (38,53)	11,42-67 (33,13)	26,14-241,01 (109,01)	Kusuma *et al*. (2015)
Jambi Sea	August 2012	6-7	2-11	18-26	Susantoro (2015)
Natuna Sea	November 2012	0,05-22,76	3,77-11,00	-	Sagala *et al.* (2014)
Gresik, East Java	February2012	1,7-12,7 (4,29)	23,7-234 (85,5)	77-405 (133)	Lestari dan Budiyanto (2013)
Semarang	August 2010	10,9-15,62 (13,69)	-	84,14-131,74 (97,11)	Rositasari dan Lestari (2013)
Kelabat Bay	March dan July 2006	1,0-22,0 (11,46)	0,2-6,4 (2,50)	2,3-34,4 (13,64)	Arifin (2011)
SQGs					
CCME (2002)	ISQG	30,2	18,7	124	
	PEL	112	108	271	
ANZECC/ ARMCANZ	Low	50	65	200	
Guidelines (2000)	High	220	270	410	

Note : - = The study were not measure in spesific parameter; SQGs = Sediment Quality Guidelines; ISQG = Interim Sediment Quality Guidelines; PEL = Probable Effect Level

sediment granules and have a high level of bioavailability (available in the body of the biota) (Bastami *et al*., 2018; Liu *et al*., 2018). The fractionation of Pb and Zn in inner Kelabat Bay is likely bound to the non-residual fraction (F1 + F2 + F3) or non-resistant with a range of 60.63-89.87% and 47.98-84.66% respectively (Figure 2.). The high percentage of non-residual or non-resistant fraction shows the input of Pb and Zn mainly from human/ anthropogenic activities rather than natural sources (Arifin and Fadhlina, 2009; Situmorang *et al*., 2010; Bastami *et al*., 2018). The concentration of Pb and Zn also show the potential (non residual fraction> 40%) of metal mobility and bioavailability on the surface of the sediment (Delshab *et al*., 2017; Gu, 2018).

Fraction 2 is a metal bound to Fe-Mn oxide seen in Pb and Zn heavy metals with a percentage of 11.85-34.88% and 32.83-72.73%. This indicates that Fe- Mn oxide is efficient as a scavengers in Pb and Zn metals (Liang *et al*., 2018). Fraction 3 is a metal that is bound to organic components seen in Pb and Zn heavy metals with a percentage of 14.67 - 55.32% and 3.71-44.24%. The Pb heavy metals (Figure 2.), the percentage between F2 and F3 tends to be inversely proportional, where the percentage of F3 tends to be greater. For Zn heavy metals (Figure 2.) also shows an inverse tendency between F2 and F3, where the percentage of F2 tends to be higher. This shows that the reducible (Fe-Mn oxide) fraction and oxidizable (organic) fraction can undergo transformation between fractions. In conditions of reduction, the metal in the reducible fraction can be released back into the water column and can be bound to organic matter in the oxidizable fraction (Shen *et al*., 2007). Under oxidation conditions, metals in oxidizable fractions can also be released again and bound by Fe-Mn oxide (Zhou *et al*., 2010). The Risk Assessment Code (RAC) is considered an appropriate criterion for investigating the effects of biological potential in sediments that have been

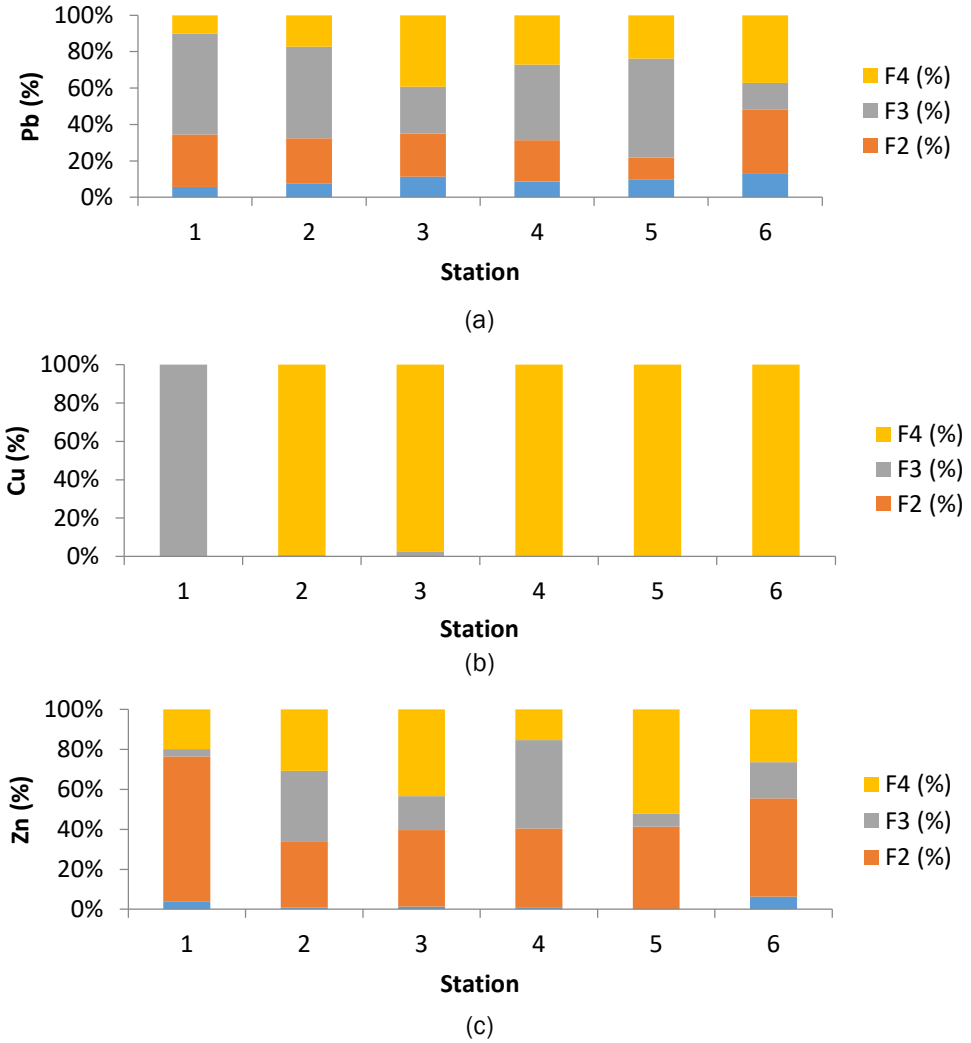

Figure 2. Fractionation of a) Pb, b) Cu and c) Zn in Kelabat Bay sediments (%) (F1 = exchangeable fraction; F2 = reducible (Fe-Mn oxide) fraction; F3 = oxidizable fraction (organic); F4 = residual fraction)

Table 2. Risk classification according to RAC

Category	Risk	Metals in exchangeable/ carbonate fraction (%)
1	No risk	< 1
2	Low risk	1-10
3	Medium risk	11-30
4	High risk	31-50
5	Very high risk	>50

Table 3. Comparison of RAC value (%) of heavy metals Pb, Cu and Zn in surface sediment of Kelabat Bay

Heavy metals	Station					
	1	2	3	4	5	6
Pb	5.72 (L)	7.48 (L)	11.27 (M)	8.72 (L)	9.96 (L)	13.21 (M)
Cu	0 (N)	0 (N)	0 (N)	0 (N)	0 (N)	0 (N)
Zn	3.71 (L)	0.81 (N)	1.32 (L)	1.05 (L)	0.04 (N)	6.33 (L)

Note: N = No risk; L = Low risk; M = Medium risk

contaminated and are widely used to evaluate metal mobility and bioavailability on the sediment surfaces (Bastami *et al*., 2018). In RAC, the percentage of the exchangeable/carbonate metal phase is applied to evaluate the environmental risks of heavy metals in the sediment (Neyestani *et al*., 2016). The RAC classification can be seen in Table 2. Risk evaluation (Table 3) of Pb namely low to moderate risk, Cu are not at risk and Zn are not at risk to low. These conditions indicate that Pb and Zn are likely to be harmful to the environment

Conclusions

Pb and Zn in Kelabat Bay are more bound to non-residual fractions (F1+F2+F3) or non-resistant with a range of 60.63-89.87% and 47.98-84.66% that are mainly come from anthropogenic activities. Cu tend to be stored or bound to the residual fraction (F4) with a proportion of 97.7-100% meaning that it comes from natural sources. Pb has a low to moderate risk and Zn has no to low risk, while Cu has no at risk in the environment. These suggest there is potential for biological availability of Pb and Zn in the inner bay waters.

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