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The Quality Of Kayeli Bay Waters: PB And HG Accumulation In Water, Sediments, And Seagrass (Enhalus Acoroides) Of Buru Island In Maluku

Abstract: The difference in accumulation of lead (Pb) and mercury (Hg) in water, sediments, and seagrass (Enhalus acoroides) organs (roots, rhizomes, and leaves) was found in the waters of Kayeli Bay, Buru Island, Maluku Province. Samples were collected from ten sampling points (Kayeli Estuary, Suket Estuary, Anahoni Estuary, Waelata I Estuary, Waelata II Estuary, Waeapo Estuary, Sanleko Estuary, Marlosso Estuary, Nametek Beach, and Jikumerasa Beach). The concentration of Pb and Hg was analyzed using Atomic Absorption Spectrophotometer. The result showed that Pb and Hg contained in sediments were higher than in water. The highest Pb and Hg content was found in the seagrass root, followed by the rhizomes and leaves (roots> rhizomes> leaves). Seagrass (*Enhalus acoroides)* is one of the living organisms that can be used as a bioindicator of heavy metal pollution in the aquatic environment.

Keywords: Enhalus acoroides; Pb; Hg; bioaccumulation; bioindicator

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1. INTRODUCTION

Marine pollution, as a result of domestic and industrial activities, has become a major issue, especially in the developing countries. The intensity and volume of human activities, such as mining, agriculture, and tourism produce solid and liquid waste that directly or indirectly impacts the environment and the sustainability of marine natural resources. One of the waste produced is heavy metals. Heavy metal pollution have the potential to occur around mining locations. Heavy metals, including hazardous waste that acts as a source of pollution, are generally toxic. Considering their adverse effects on the environment and human health, heavy metals are becoming very popular to be studied (Rainbow, 2007; Roberts et al., 2001). Heavy metal waste is very dangerous because it has toxic effects on humans (Boran and Altinok, 2010). Heavy metals that infiltrate the aquatic environment will dissolve and accumulate in sediments or biota and the concentration of the metals can increase over time, depending on the environmental conditions of these waters (Wulandari et al., 2012). Bioaccumulation of metals in a marine organism initiates the response of the organism to contaminants and the geochemical cycle (Fisher, 2003). The chemical process of bioaccumulation of heavy metals is a reaction to the formation of complex compounds between heavy metals and the cells of organisms that function as ligands. This process is explained through the Ligand Biotic Model where free metal ions or their derivatives are designed to predict how these heavy metals interact with aquatic organisms (Campbell, 2002). Seagrass is a single seeded (monocot) plant that belongs to the angiospermae class. Seagrass is able to live and thrive in the intertidal zone and adapt to tidal dynamics. Seagrass plays very important ecological and physical roles. It provides habitats for several species of marine animals, such as: benthic macrofauna (snails, shellfish, sea cucumbers, starfish, feathers pigs, and crabs), turtles, and dugongs.

Seagrass is usually present in large quantities and can form a dense seagrass bed, covering a vast coastal area in subtropical and tropical regions. Seagrass is a primary producer in the sea if compared to other ecosystems (Azkab, 2006). The leaves, which are generally ribbon-shaped, act as traps of suspended materials carried by currents to the seagrass area. Its rhizomes and root system can stabilize sediments so as to prevent erosion, especially during storms, rain, and floods (Björk et al. 2008). The ability of aquatic vegetation, including seagrass and macro algae, to accumulate heavy metals has been studied by (Endang, 2008; Ahmad et al., 2015; Ambo-Rappe et.al., 2011; Sudharsan et al., 2012; Supriyantini et al., 2016; Thangaradjou et al., 2010; Tupan and Azrianingsih, 2016). These researchers found that seagrass belongs to one of the aquatic organisms that are quite sensitive in predicting the level of sea water contamination. This is possible, because throughout their lives, seagrass never moves and lives in a wet habitat or substrate (water and sediment). Therefore, seagrass is often used as an indicator of accumulation of metals in waters. Direct interactions of seagrass with water bodies and soils (substrate) occur through its root and leaf systems, which are for uptake of ions. Seagrass can also reflect the overall health status of waters (Ahmad et al., 2015; Supriyantini et al., 2016). For the past few years, Kayeli Bay waters have been potentially polluted by heavy metals. This is caused by traditional gold mining activities operating in the Waeapo River basin that directly leads to the bay. Uncontrolled illegal mining in this area will also threaten environmental sustainability. The environment polluted by mining chemical waste will have an impact on agricultural land, coastal, and marine ecosystems as well as long-term impacts on the health of the local population through the consumption of contaminated water and food (Tuaputty, 2014). Agus et al. (2005) and Ning et al. (2011) reported that traditional gold mining was one of the ways of heavy metals to enter aquatic environment. Research on the status of heavy metal pollution in the waters of Kayeli Bay in Buru Regency, thus, should be carried out. Therefore, the current research aimed to analyze the content of heavy metals, Pb (lead) and Hg (mercury) in Kayeli Bay waters, sediments, and seagrass (*Enhalus acoroides*) organs (roots, rhizomes and leaves).

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2. RESEARCH METHODS AND MATERIALS

The Determination of Sampling Points and Sample Collection

Sampling points were determined using a Garmin Geographic Positioning System (GPS) and based on the potential of the areas to be the targets of the pollutants. There were nine adjacent sampling stations and one station representing areas far from the pollutant source. On Station 1 (Kayeli Estuary), there were docks, settlements, and mining activities; On Station 2 (Suket Estuary), there were mangroves and no land activities; On Station 3 (Anahoni Estuary), there were no on-land activities but the station was positioned within a gold mining waste disposal stream (pollutant source); On Stations 4 and 5 (Waelata I and Waelata II Estuaries), there were activities on land (settlement, mining); On Station 6 (Waeapo Estuary), there were mangroves and land activities (settlements, mining), and On Stations 7, 8, and 9 (Sanleko Estuary, Marlosso Estuary, and Nametek Beach) there were seagrass and on-land activities (mining using drum, residential areas, docks). The physical-chemical parameters of water at each section were measured. Water samples were collected using polyethylene bottles (according to SNI 6989.57: 2008). Before sampling, physical chemical parameters such as temperature, pH, salinity, depth, turbidity and brightness were measured. Water samples were collected using sample bottles at a depth of ± 30 cm from each sampling point/station. To measure the concentration of lead (Pb) and mercury (Hg), the samples were added with HNO3 as a preservative until pH <2 then stored in a cool box at 5^{0}C. Then the samples were taken to the Basic Chemistry Laboratory of Universitas Muhamadiyah in Malang for analysis. Sediment samples were obtained using ekman grab. Three hundred grams of sediment was collected and then put into polyethylene plastic and transferred to the laboratory in a cool box. Seagrass sampling was done by using a scope. The seagrass plants were pulled out slowly to avoid the plants from being damaged. Samples of the seagrass (*Enhalus acoroides*) were taken around the waters of Kayeli Bay containing seagrass. Seagrass samples were put into plastic bags, then stored in cool boxes and taken to the laboratory for preparation before analyzing the concentration of lead and mercury (Pb and Hg) in the roots, rhizomes, and leaves.

Analysis of the Heavy Metals (Pb and Hg) Concentration

Seagrass (*Enhalus acoroides)* samples were collected only from two observation stations that contained seagrass (Marlosso and Jikumerasa). Seagrass was pulled out slowly from the soil in order to keep the roots, rhizomes, and leaves intact. Then, the organs were separated from the plants for analysis. Ten to twenty five milliliters of water (or with assumed magnesium concentration) and 2-10 g of sediment (or with assumed mercury concentration) were collected and extracted. The samples were inserted into a digestion tube, added with 5 ml of HNO3 p.a. and 0.5 ml of HClO4 p.a. and left overnight. The day after, the samples were heated in digestion blocks at 100 °C for an hour. After that, the temperature was raised to 50 °C. After the yellow steam was gone, the temperature of the digestion blocks was turned up until 200 °C. Destruction was completed after white smoke rose and 0.5 ml extract remained. The tube was lifted and let cool. The extract was diluted with ion free water to a precise volume of 50 ml and whipped with a tube shaker until homogeneous. This extract can be used for measurement of macro and micro elements. For standard measurement, the extract did not have to go through the graying process. Samples were analyzed using an Atomic Absorption Spectrophotometer (AAS). The tools and methods used for the extraction and analysis of the lead and mercury concentration in Kayeli Bay waters and sediments were similar to those used for the analysis of the lead and mercury concentration in seagrass organs.

Data Analysis

Data on the metal (Pb) and mercury (Hg) concentration were displayed in tables, graphs, and histograms then discussed descriptively. The result of the analysis of Pb and Hg concentration in water samples was compared with the standard value of Sea Water Quality Standards for marine biota (Decree of the Minister of Environment No. 51 of 2004), while the content of Pb and Hg in sediments was compared with the CCME, ANZECC 2002 and NOAA 1999 quality standards. The levels of Pb and Hg contained in seagrass organs were compared with the ISO 7387: 2009 quality standard on the Maximum Limits of Metal Contamination in Food Materials. Analysis of Multifactorial ANOVA (Two Way ANOVA, $p < 0.05$) was also used to find out the difference in heavy metal (Pb and Hg) accumulation at each sampling station. In addition, Two Way ANOVA ($p < 0.05$) was used to determine the difference in the accumulation of Pb and Hg in seagrass roots, rhizomes, and leaves.

3. RESULTS

Description of the Research Location

Kayeli Bay is a body of water that is located in Buru Regency, Maluku. This area lays in 3^{0} 15.55" 3^{0} 22.50" South and 127^{0} 01.35" 127^{0}01.35" East. Kayeli Bay is a semi-enclosed water that receives input of river flow from several estuaries including Kayeli estuary, Anahoni estuary, Waelata I estuary, Waelata II estuary, Waeapo estuary, Sanleko estuary, Marlosso estuary, Nametek Beach, and Jikumerasa Beach. The traditional gold mining activity in Botak Mountain has been going on since 2012 until now. To separate gold from chunks of rock and land, people use mercury and cyanide. The results of field observations indicate that almost 90% of the Botak Mountain area has been utilized for gold mining. Since 2013, the government has attempted to issue several regulations regarding the closure of mining activities and the handling of the restructuring of Mount Botak in Buru Regency. However, given the condition that the people still utilize a traditional approach to exploiting these gold resources, there is a high possibility that the pollutants can flow from land to river and end up at sea.

The Concentration of Lead (Pb) and Mercury (Hg) in Kayeli Bay Waters and Sediments

The result of the Two-way ANOVA analysis showed that there was a significant difference in lead (Pb) content between Kayeli Bay waters and sediments ($P < 0.05$). The average Pb content in the waters (1.2818 mg/Kg) was higher than that found in the sediments (1.2186 mg / l). The levels of Pb contained in Kayeli Bay waters (10 sampling stations) ranged between 0.005 ± 0.0005 mg/L and 0.023 ± 0.0005 mg/L. The highest Pb content was observed in Station 3, followed by Station 5 and Station 6 (Figure 1).

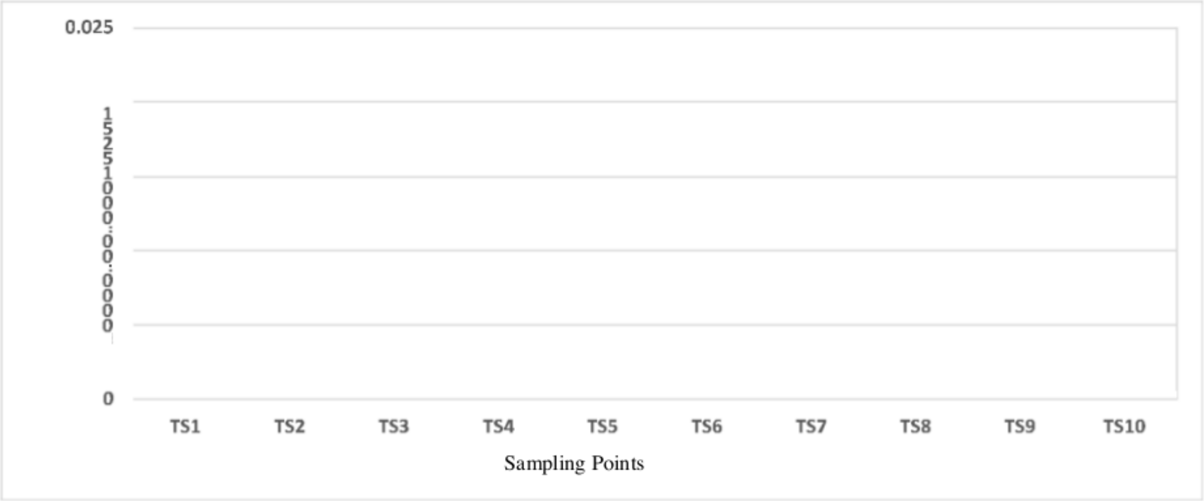

Figure 1. The Concentration of Lead (Pb) in Kayeli Bay Waters

The analysis showed that the concentration of Lead (Pb) in Kayeli Bay sediments ranged from 0.4789 ± 0.01913 1.8514 ± 0.07471 mg/Kg (Figure 2).

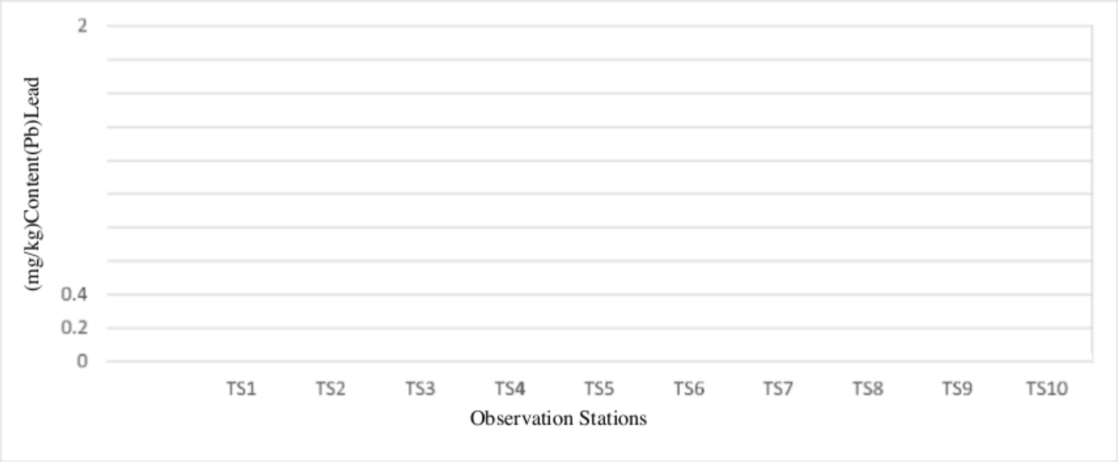

Figure 2. The Concentration of Lead (Pb) in Kayeli Bay Sediments

The result of the two-way ANOVA analysis indicated that there was a significant difference between Mercury (Hg) concentration found in the Kayeli Bay sediments (0.501 mg/L) and that found in the waters (0.003 mb/L) with $p<0.05$). The analysis has indicated that the ten sampling stations contained mercury (Hg) that has been beyond the threshold (0.001 mg/L) (Decree of the Minister of Environment No. 51/2004). The analysis showed that the concentration of Mercury (Hg) in Kayeli Bay waters ranged from 0.001±0.00012 0.005±0.00012 mg/L (Figure 3).

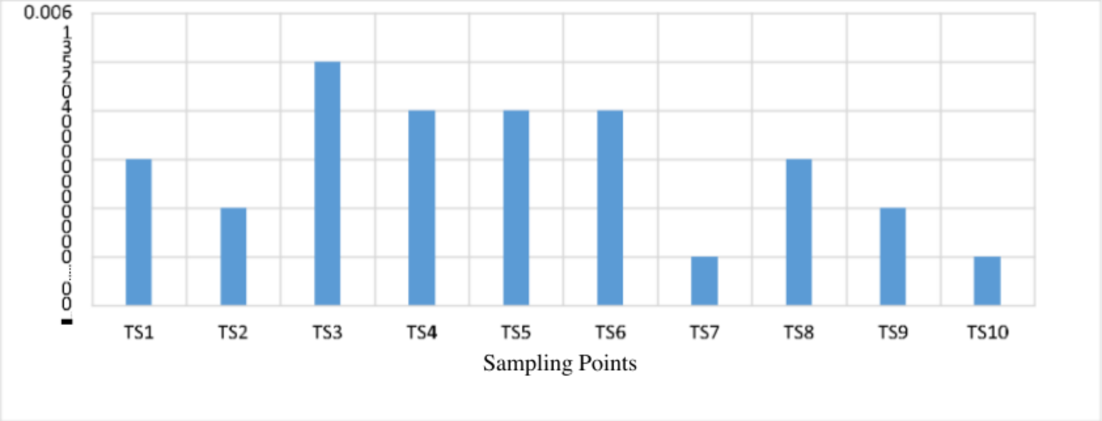

Figure 3. The Concentration of Mercury (Hg) in Kayeli Bay Waters

The result of the analysis showed that Mercury (Hg) contained in the Kayeli Bay waters was between 0.001±0.023 1.835±0.16828 mg/kg. The standard quality of the mercury concentration was then compared CCME from Canada, which was 0.13 mg/kg (Figure 4).

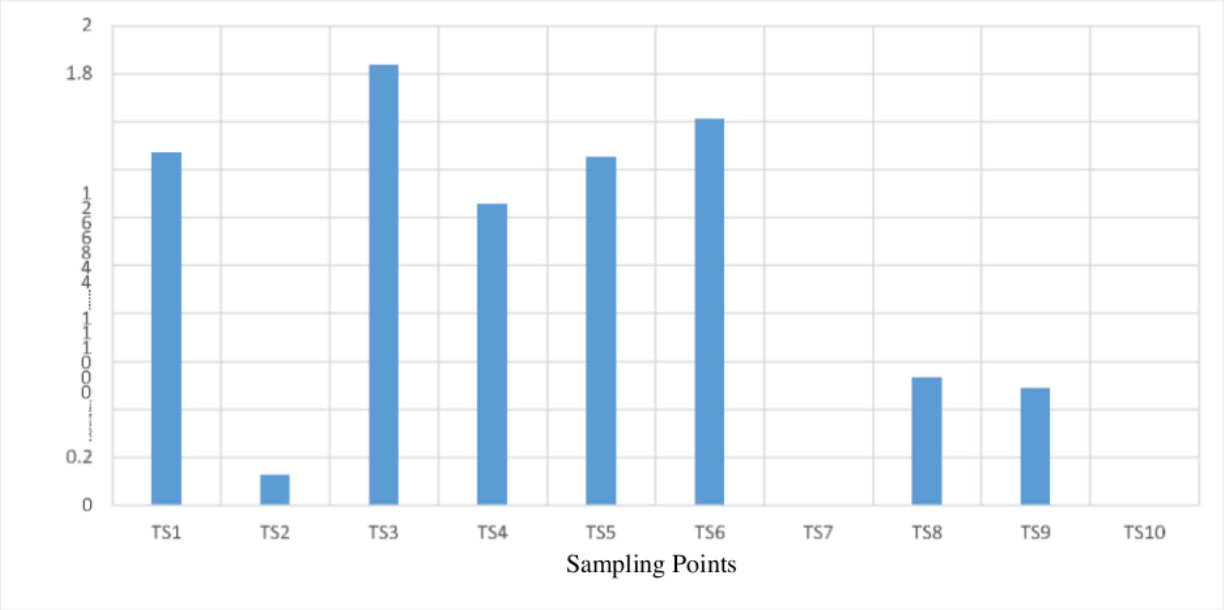

Figure 4. The Concentration of Mercury (Hg) in Kayeli Bay Sediments

The Concentration of Heavy Metals in Seagrass *(Enhalus acoroides)* Roots, Rhizomes, and Leaves

Seagrass (*Enhalus acoroides)* community was only found in Station 8 (Marlosso Estuary) and Station 10 (Jikumerasa Beach). Table 1 presents the difference between lead (Pb) and mercury (Mg) concentration found in the seagrass rots, rhizomes, and leaves.

Table 1. The Concentration of Lead (Pb) and Mercury (Hg) in Seagrass (Enhalus acoroides) Roots, Rhizomes, and Leaves at Station 8 (Marlosso Estuary) and Station 10 (Jikumerasa Beach)

Observation Station	Location	Root		Rhizome		Leaf	
		Pb (mg/Kg)	Hg (mg/Kg)	Pb (mg/Kg)	Hg (mg/Kg)	Pb (mg/Kg)	Hg (mg/Kg)
8	Marlosso Estuary	[b]5.3266±0.0558	[b]0.6687±0.0069	[b]3.5809±.0223	0,4772± 0,0080	[b]1.7981±0.0360	0.2717±0.0088
10	Jikumerasa Beach	[b]2.7449±0.0271	0.1782± 0.00867	[b]1.9271±0.0363	0,1099± 0,0105	[b]0.7985±0.0351	0.0576±0.0040
Average		[b]4.03375	0.4235	[b]2.754	0,2935	[b]1.2983	0.165
[a]Quality Standard		0.3	0.5	0.3	0.5	0.3	0.5

Note: [a]SNI/National Standard 7387:2009; [b]Beyond the Quality Standard

4. DISCUSSION

The Concentration of Lead (Pb) and Mercury (Hg) in Kayeli Bay Waters and Sediments

The analysis showed that there was a significant difference between the levels of lead (Pb) contained in the Kayeli Bay waters and those contained in the sediments. It was found that the average content of Pb in the sediments (1.2818 mg / Kg) was higher than that in the water (1.2186 mg / l). Similar results were also reported by Tupan, et al. (2014) who mentioned that Pb was higher in sediments than in waters and that Pb content had a significant correlation with seagrass (*Thalassia hemprichii*) roots. Some of heavy metals can penetrate into the soil (sediment) and some others will enter the river flow system and be carried out to the sea. Heavy metals that infiltrate marine ecosystem settle to the bottom of the water and be absorbed by the sediments (Jaibet, 2007). Lead (Pb) content found in the Kayeli Bay waters has exceeded the a water quality standard for marine biota, which is 0.008 mg/L (Decree of the Minister of Environment No. 51 of 2004). High lead content in the waters will potentially bring an impact on the disruption of the aquatic biota life. Heavy metals in these waters are subsequently absorbed in aquatic plants and animals including plankton and other microorganisms (Riani, 2012). Sediment and soil are known as the "storage systems" of pollutants that can deposit heavy metals, including lead (Pb). On the other hand, the concentration of Pb found in the Kayeli Bay sediments was still below the safe tolerance limit when compared to the CCME (2002) quality standard (30.2 mg /kg). Even though the levels of Pb contained in the sediments is considered safe, continued heavy metal deposition in Kayeli Bay can result in an increase of excess lead in the sediments. As the number of gold mining activities performed along Kayeli Bay improves, the potential of lead (Pb) contamination also grows. The results of the study also showed that there was a significant difference between Mercury (Hg) content in the Kayeli Bay waters and Hg content in the sediments. The highest Hg content was found in the sediments (0.501 mg / Kg), while the average Hg content in the waters was - 1.226 mg/l. Mercury (Hg) has stable chemical properties, especially in the sedimentary environment. Soon after Mercury infiltrate the waters, some of it will be carried out by the flow and the rest is absorbed by aquatic plants through the roots and transformed in the metabolic system of the plants. Thomas and Young (1998) argue that sediments are potential to store B3 in the coastal area. The levels of Mercury (Hg) found in the Kayeli Bay waters have exceeded the quality standard value for aquatic organisms (Table 5). The quality standard value of Mercury content in seawater is regulated in the Decree of the Minister of Environment No. 51 of 2004 concerning sea water quality standards for marine biota (0.001 mg/L). High Mercury content in the Kayeli Bay waters may be the result of traditional gold mining activities done in the upper reaches of Waeapo river. People around the area usually process gold using mercury liquid. Therefore, the gold mining activity has the potential to pollute the environment if it is not managed properly. An illegal use of Mercury in gold ore processing (tromol/tailing) will adversely affect the environment, especially the land around the industry area. This finding is in line with the result of the research conducted by (Mirdat et al. 2013) who found high concentration of Mercury (Hg) contained in the soil (0.57 ppm to 8.19 ppm) and in the waste (84.15 ppm to 575.16 ppm) around the drum/tailings area in Poboya Village. Gold processing that involves the use of mercury liquid will result in hazardous sludge. Sludge that is only accommodated in a small reservoir can get washed away by rain water and carried into the river and flow to the sea. The rest of the sludge will evaporate into the air and fall down to earth together with the rain. This is thought to be the main cause of the increasing mercury content in the Kayeli Bay sediments. Besides gold mining activity, high Mercury contamination has also resulted from human domestic activities and agricultural activities that generate a lot of waste. Household waste contains Hg that can accumulate in sediments. Mercury that evaporates can survive in the atmosphere for a year (Anonymous, 2011). Mercury (Hg) contained in waste is changed by microorganisms into methyl-mercury (Me-Hg) which has toxic properties and a strong binding capacity despite its high solubility. Therefore, Mercury can be easily accumulated in sediments through food chain's bioaccumulation and biomagnification processes (Purnawan et al, 2013).

The Concentration of Heavy Metals in Seagrass *(Enhalus acoroides)* Roots, Rhizomes, and Leaves

The results of the research showed that there was a significant difference between lead (Pb) and mercury (Hg) in seagrass roots, rhizomes, and leaves found at Station 8 (Marlosso Estuary) and Station 10 (Jikumerasa Beach) (Table 1). The concentration of Pb in seagrass roots was higher at station 8 (5.3266 ± 0.05579 mg / Kg) than station 10 (2.7449 ± 0.02713 mg / Kg). Compared to the quality standard values for heavy metal lead (Pb) based on the National Standard of SNI 7387: 2009 (0.3 mg /kg) and Food and Drug Administration/BPOM Indonesia (0.4 mg/kg), Pb content in Kayeli Bay is considered high. High concentrations of lead in seagrass roots suggest that Pb metals are easily absorbed in the form of ions. Lead can dissolve in fat and be able to penetrate the cell membrane, so metal ions will accumulate in cells and tissues. Similarly, the concentration of Pb in seagrass rhizomes was higher at station 8 (3.5809 ± 0.02233 mg/Kg) than station 10 (1.9271 ± 0.03634mg/Kg). Both of them have also exceeded the quality standard values. This may happen due to the absorption of Pb by the seagrass plants through the roots. Seagrass rhizomes have the ability to stabilize the seabed and vegetative reproduction of the seagrass. Therefore, high content of Pb in seagrass rhizomes can adversely affect the growth and distribution of seagrass. The concentration of Pb in leaves at station 8 (1.7981 ± 0.03602 mg/Kg) was also higher than that at station 10 (0.7985 ± 0.03515 mg/Kg). Compared to the National Standard quality SNI values and Food and Drug Administration/BPOM Indonesia, these figures are considered excessive. The transfer of Pb from the soil to plants depends on the composition and pH of the soil. Soil pH that is suitable for plant growth is 6-7.5 where nutrients are easily absorbed by the plant. The presence of heavy metals in the soil will reduce the soil pH so that it becomes acidic and is toxic to plants. The role of microorganisms as decomposers of organic matter will also be disrupted. Muhammadi, et al (2005) reported that soil pH decreased several days after application of heavy metals and microorganisms into the soil. Microorganisms also participate in the process of mobilizing and immobilizing heavy metals in the soil by lowering the pH around the soil. Furthermore, high Pb concentrations will have a toxic effect on plant photosynthesis and growth. Plants can easily absorb Pb when soil are low in fertility and organic matter. Compared to Station 10 (0.1782 ± 0.0087 mg / kg), Station 8 contained higher levels of mercury (Hg) (0.6687 ± 0.0069 mg / kg). However, according to the National Standard of SNI 7387: 2009 (0.5 mg/kg) and Food and Drug Administration/BPOM Indonesia (1.0 mg/kg), these values are considered normal or within the tolerance limits for plants.

Mercury is transported through the xylem and phloem transport network to other plant parts. In improving transport efficiency in plant organs, metals are bound by chelate molecules $(HO_2CCH_2)_2NCH_2CH_2N(CH_2CO_2H)_2$. Various types of chelate molecules that function to bind metals produced by plants such as histidine ($C_6H_9N_3O_2$) can bind Cr. In preventing metal poisoning to cells, plants have a detoxification mechanism. High Mercury (Hg) concentration in seagrass roots at Station 8 is thought to be the result of nearby gold mining activities that have first contaminated the surrounding soil and river water. The plant's first response to toxic is located in the root. Besides, there is also a series of physiological processes that play a role in the accumulation of metals throughout the plant life cycle that starts from the root. The concentration of Hg in seagrass rhizomes was also higher at station 8 (0.4772 ± 0.0080 mg/Kg) compared to station 10 (0.1099 ± 0.0105 mg/Kg). Even though these figures are still very low, compared to the national standard quality, the increase in metal content, especially mercury, needs to be watched out because plants play a crucial role as the main producer in the food chain system. Mercury (Hg) concentration will likely increase (highly accumulated) in biota with higher trophic levels. Similarly, the content of mercury (Hg) in seagrass leaves was higher at Station 8 (0.2717 ± 0.0088 mg/Kg) than at Station 10 (0.0576 ± 0.0040 mg/Kg). These values are still within the Mercury normal limit, however, the Hg content in Marlosso Estuary is quite concerning since it approaches the critical limit. If it is not handled carefully, there is a possibility that mercury pollutants from human activities can enter the waters periodically and damage the nearby aquatic environment in the near future.

5. CONCLUSION

The results of the study indicate that the accumulation of lead (Pb) and mercury (Hg) is more abundant in the Kayeli Bay sediments and seagrass roots. Seagrass (*Enhalus acoroides)* can be used as a bioindicator of metal pollution levels in aquatic environment.

6. ACKNOWLEDGMENTS

We would like to thank the Head and staff of the Basic Chemistry Laboratory at Universitas Muhammadiyah Malang for their kind assistance in analyzing the concentration of Pb and Hg in water, sediments, and seagrass.

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