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Bioaccumulated Mercury by Several Types of Plants in Ex-Traditional Gold Processing Area, Gogorea Village, Buru Island

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Abstract

This study examines the accumulation of metallic mercury by several types of plants in the traditional gold processing area in the village of Gogore, Buru district. The plants that were sampled in this study were guava, lempuyang gajah, and harendong bulu. These three plant species were chosen because they dominate the vegetation in the gold processing area. The analysis results showed that the lempuyang gajah was the plant that accumulated the highest mercury metal, namely in the roots of 16.79 ppm and the leaves of 15.03 ppm. Guava plants accumulated metal mercury in the roots and leaves of 11.73 ppm and 9.90 ppm, respectively. Meanwhile, harendong plants accumulated mercury in the roots and leaves of 2.59 ppm and 10.39 ppm, respectively. The BCF values of guava, lempuyang gajah, and harendong bulu plants were 1.58, 0.41, and 0.39, respectively. Meanwhile, the TF values of the three plants were 0.84, 0.89, and 4.01, respectively. From these results, it can be concluded that the three plants can accumulate mercury in high enough concentrations, so these three types of plants are categorized as hyper tolerant plants and accumulators.

Keywords: Accumulation, Gogorea village, mercury metal, vegetation, hypertolerance, accumulator.

INTRODUCTION

Buru Regency, Maluku Province, has become the largest traditional gold mining area in Maluku since the end of 2011. Besides Mount Botak in Waelata sub-district, one place also the center of conventional gold mining on Buru Island is Gogorea village, the Waeapo sub-district. Thousands of traditional miners who once operated in Gogorea village also use mercury in the amalgamation process. The tailings produced from the process have the potential to pollute the environment around the place. The research results by Reichelt-Brushett et al. (2017) showed that the mercury content in soil samples from several traditional gold mining locations in Gogorea village was very high, namely 15.2 ppm, 4.23 ppm, and 6.80 ppm (Reichelt-Brushett et al., 2017).

Waste from the amalgamation process discharged into the environment around the mining area is then washed when it rains and enters the Gogorea river, which empties into Kayeli Bay. Mariwy et al. (2019) showed that sediments in several rivers that ended in Kayeli bay were contaminated with mercury(Mariwy, Male, & Manuhutu, 2019). Male et al. (2014) and Reichelt-Brushett, et al. (2017) showed that samples of several shellfish and fish and mangrove crabs taken from Kayeli bay were contaminated with heavy metal mercury (Male, Nanlohy, & Asriningsih, 2014; Reichelt-Brushett et al., 2017). This condition is very dangerous because mercury as gold mining waste distributed to rivers and seas in large quantities can also accumulate in humans through the food chain (Mariwy, H. Dulanlebit, & Yulianti, 2020; Rumatoras, Taipabu, Lesiela, & Male, 2016).

This condition requires serious handling because if it is not carried out, the mercury pollution process in Kayeli Bay will be increasingly dangerous for the people who live around the bay. One technology that is easy to apply and environmentally friendly in accumulating metallic mercury from polluted soil is phytoremediation by utilizing plants (Galal, Gharib, Ghazi, & Mansour, 2017). This technology is very useful for cleaning the contaminated environment from harmful contaminants (Tangahu et al., 2011). Plant species are needed to reduce metal accumulation on land with high metal content to increase environmental quality.

Based on the research team's observations on the former gold mining area in Gogorea village, three plants dominated the vegetation in the area. There are guava plants (*Psidium guajava* L.), lempuyang gajah (*Zingiber zerumbet* L.), and harendong bulu (*Clidemia* *hirta* L.). The ability of these three plants to tolerate mercury content during their growth needs to be studied to get an idea of the types of plants suitable for use as phytoremediation agents to reduce mercury content in the former traditional gold processing area Gogorea village.

This research studied the accumulation capacity of heavy metal mercury by three plants; there are guava plants (*Psidium guajava* L.), lempuyang gajah (*Zingiber zerumbet* L.), and harendong bulu (*Clidemia hirta* L.). This research is part of an effort to remediate the former gold processing land in Gogorea village using typical plants. The samples will be analyzed as part of a standard procedure to study the bioaccumulation of heavy metals by plants are the soil around the plant, plant roots, and leaves to see the ability of heavy metal accumulation by the plants sampled in the study (Siahaan, Utami, & Handayanto, 2014).

METHODOLOGY

Materials

Materials used are: plant samples of guava plants (*Psidium guajava* L.), lempuyang gajah (*Zingiber zerumbet* L.), and harendong bulu (*Clidemia hirta* L.), soil samples, aquades, filter paper, tissue, aluminium foil, ice cubes, mercury solution 1000 ppm, HNO_3 pa (E. Merck), $HClO_4$ pa (E. Merck)

Instrumentations

Mercury analyzer (NIC MA 300) LPPT UGM, Analytical balance (Cyberscan CON 110) Oven (memmert) Mortal and pestle, Glassware (pyrex), and Hot plate (Cimarec*)*

Methods

Sampling

The plant samples used in this study were guava plants (*Psidium guajava* L.), lempuyang gajah (*Zingiber zerumbet* L.), and harendong bulu (*Clidemia hirta* L.). Take each root and leaf, then put into a sample bag. Soil was taken using a trofol, separated from dirt, then put into a sample bag. The sample is then put into a cold box for laboratory analysis.

Standard curve

The mother liquor of 1000 mg/kg 0.1 mL was put in a 100 mL volumetric flask, adjusted with distilled water to the limit mark (Hg: 1000 g/kg). Mother solution Hg 1000 g/kg 0.1 mL, put in a 10 mL volumetric flask, adjusted with distilled water to the limit mark (Hg: 10 g/kg). Make a standard concentration with a range (μg/kg): 0.05; 0.1; 0.2; 0.4; 0.8; 1.6; 3.2 by: take each (mL) 0.05; 0.1; 0.2; 0.4; 0.8; 1.6; 3.2 was put in a 10 mL volumetric flask, adjusted to the mark with distilled water, poured into a vial tube and analyze with mercury analyzer.

Sample Preparation and Destruction

The sample was dried at 40 ^{o}C for 1 hour, then crushed and put into a glass beaker (Mariwy et al., 2020). Each was weighing 1 gram, pour in a 100 mL Erlenmeyer, then add 10 mL of HNO_3: $HClO_4$ (1:1) solution, after which the sample is heated on a hotplate until clear and white smoke comes out, filtered, and adjusted to 50 mL with a measuring flask. . Make blanks with the same treatment without the sample. The mercury content in the sample is then measured with a mercury analyzer.

Data analysis

Based on the results of serial measurements of Hg standard solution with a mercury analyzer, it is made to obtain a straight line on the graph between absorbent and concentration using linear regression Equation 1.

$$y = ax + b \tag{1}$$

Note: x = concentration
y = absorbance
a = slope/ gradient
b = intercept

The concentration obtained from this curve is the concentration in mg/L standard. While the samples analyzed were in mg/kg units (Equation 2).

$$\text{Total of Hg } (\mu g/kg) = \frac{\text{Hg analysis} - \text{Blank x Volume x Dillution factor}}{\text{Sample Weight}} \tag{2}$$

The potential of plants to accumulate mercury (Hg) in polluted soil will be analyzed by calculating the BCF (Bioaccumulation Concentration Factor) value determined by the ratio of metals in the roots to those in the soil (Equation 3).

$$BCF = \frac{\text{Concentration in leaves or roots of plants}}{\text{Mercury concentration in soil}} \tag{3}$$

In the translocation process, the ratio between the metal concentration in the canopy and the metal concentration in the plant's roots is determined by the TF (Translocation Factor) value (Equation 4).

$$\text{TF} = \frac{\text{Concentration in plant leaves}}{\text{Concentration in plant roots}} \tag{4}$$

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RESULTS AND DISCUSSION

Overview of sampling locations

The three plant samples used were taken from the area of the former amalgamation process in Gogorea village, Waeapo sub-district, Buru district. The Gogorea village map is shown in Figure 1.

Figure 1. Map of The Gogorea village, Waeapo sub-district. The red arrow indicates the sampling location.

In 2012-2020 the village of Gogorea became one of the traditional gold mining centres. Thousands of conventional miners who have operated in Gogorea village use mercury in the amalgamation process. The tailings produced from the process have the potential to pollute the environment around the place. Although this mining site was closed in 2020, the high mercury content in the area needs to be reduced not to cause serious impacts on public health.

Sample preparation and destruction processes

Root and leaf samples were cleaned of adhering soil debris. Meanwhile, soil samples were separated from impurities such as stone, wood, and fibrous roots. Then the sample was dried using an oven at 40 ^oC for 1 hour. Drying using a temperature of 40 ^oC because mercury metal has volatile properties, the temperature is maintained as much as possible so that the mercury in the sample does not evaporate. This drying method also aims to reduce the water content in the sample to extend the durability of the sample during storage. The dried samples were then ground. The purpose of grinding the sample is so that the destruction process is faster.

The samples were then weighed 1 gram each. The sample was dissolved using HNO_3 and $HClO_4$ (1:1). The addition of HNO_3 aims to separate the mercury from the compounds. At the same time, the function of $HClO_4$ as an oxidizing agent is to maximize the metal scission process from organic compounds contained in the sample. In this process, the organic compounds in the sample will turn into CO and CO_2 while HNO_3 at high temperatures is decomposed into various oxides such as NO, NO_2, HNO, and many more.

$$Hg(CHO)_x + HNO_3 + HClO_4 \longrightarrow Hg(NO_3)_2 + CO_2 + NO_x + HClO_3 + H_2O$$

In this reaction, $HClO_4$ is reduced to $HClO_3$. At the same time, the remaining HNO_3 becomes NO or NO_2. This process is repeated until all metals are entirely oxidized. The destruction process is carried out until the solution becomes clear and white smoke comes out.

Preparation of standard solutions and calibration curves

The standard solution was used for the preparation of the calibration curve. The standard solution series is as follows (µg/kg): 0.05; 0.10; 0.20; 0.40; 0.80; 1.60; 3.20. Then the standard solution was analyzed using a mercury analyzer. The data from the standard solution analysis can be seen in Table 1.

Table 1. Standard solution analysis data

No	Concentration (ppb)	Absorbance (A)
1	0.05	0.00046
2	0.10	0.00087
3	0.20	0.00194
4	0.40	0.00387
5	0.80	0.00877
6	1.60	0.01516
7	3.20	0.02957

Based on the results of the standard solution, it can be seen that the greater the concentration of the standard, the greater the value of the absorbance, or it can be said that the concentration is directly proportional to the value of the absorbance. The data is then used to create a calibration curve. A calibration curve is made to express the relationship between absorbance and concentration of a series of standard solutions. The calibration curve is shown in Figure 2.

Figure 2 shown that the regression equation y = 0.0092x + 0.0003 and the correlation coefficient (R^2) is 0.9977. This curve shows that the correlation coefficient value of concentration to absorbance has a relationship. The higher the concentration, the higher the absorbance value.

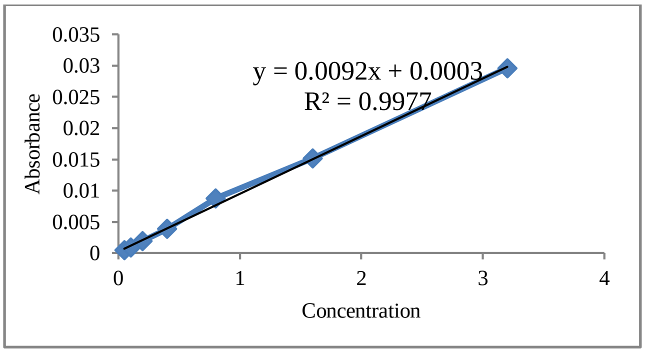

Figure 2. Mercury calibration curve

Mercury Analysis in samples

Mercury analysis in plant and soil samples is shown in Table 2. Based on Table 2, it can be seen that the concentration of mercury in the soil is relatively high, namely 13.69 ppm, 75.86 ppm, and 32.83 ppm. The concentration of mercury in the soil is very influential on mercury concentration in the plants that grow on it.

Table 2. The data of mercury content in roots, leaves, and soil

Plants	Code	Absorbance	Mercury content $(\mu g/kg)$	mercury final (mg/Kg)
Guava	Root	0.00262	253.790	11.73
	Leaves	0.01894	201.96	9.90
	Soil	0.00318	313.910	13.69
Lempuyang gajah	Root	0.00356	355.190	16.79
	Leaves	0.00182	333.62	15.03
	Soil	0.00812	1696.000	75.86
Harendong bulu	Root	0.01168	61.658	2.59
	Leaves	0.00126	213.310	10.39
	Soil	0.00363	724.320	32.83

This number is shown in the results of the mercury analysis in the roots and leaves of the three samples, where the plants contain metallic mercury. The graph of mercury concentration in plant samples can be seen in Figure 3.

However, it is not only the mercury concentration in the soil that is an essential factor in metal accumulation in plants. There are several other factors such as soil pH, chemical elements/complex compounds in the ground. The pH soil effect on the mercury absorption by plants can be explained that the higher the pH of the soil, the lower the mobility of the metal so that the absorption of metal by plants is small. A low soil pH indicates that the mobility of metals in the soil is higher so that the absorption of metals in plants is more significant. While the influence of complex compounds can stimulate metal accumulation in plants so that metals can be bound and absorbed by plants.

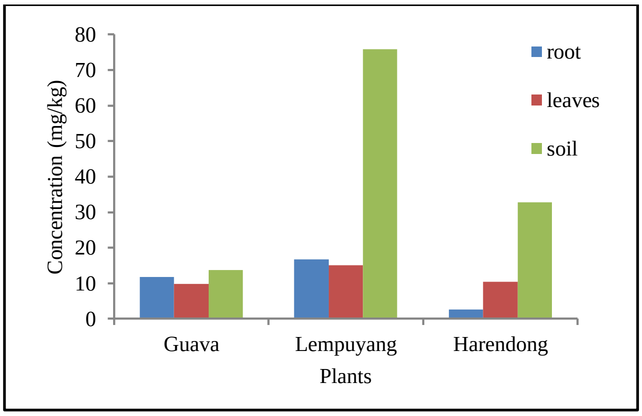

Figure 3. Graph of mercury concentration in plants

Each plant is sensitive to heavy metals and has a different ability to accumulate heavy metals. Based on the results of this study, it was shown that the three types of plants were able to accumulate metallic mercury with different concentrations. The lempuyang gajah is a plant that accumulates the highest metal mercury, namely in the roots of 16.79 ppm and the leaves of 15.03 ppm. Guava plants accumulated metal mercury in the roots and leaves of 11.73 ppm and 9.90 ppm, respectively. Meanwhile, harendong plants accumulated mercury in the roots and leaves of 2.59 ppm and 10.39 ppm, respectively.

According to Hardiani and Henggar (2009), the amount of metal accumulation in the roots is a detoxification mechanism carried out by plants to avoid metal poisoning in plant cells by accumulating metals in the roots (Hardiani & Henggar, 2009). The absorption of metallic mercury in plant roots is due to the presence of phytochelatins produced by the roots as a metallic mercury binder. When phytochelatin binds to Hg, it will form a bond at the sulfide end of cysteine and form a complex compound so that Hg will be translocated into plant tissues. The reaction between cysteine and Hg is shown in Figure 5.

The translocation process will decrease concentration in roots and increase metal concentration in leaves. The harendong bulu also shows this process. The mercury accumulation in harendong bulu leaves is higher than in the roots. This condition is also related to the BCF and TF values of harendong bulu. Harendong TF value is greater than one, where TF> 1 indicates the plant has a high ability to translocate mercury from roots to leaves.

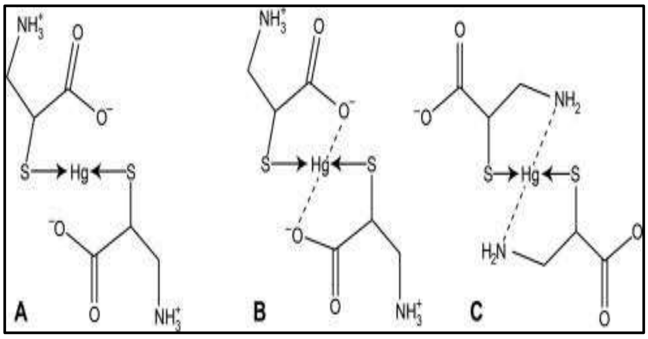

Figure 4. The reaction between Hg and cysteine (Ajsuvakova et al., 2020)

While the BCF value <1 suggests that the harendong is an excluder plant or does not absorb much metal from the soil to the roots. This value does not rule out the possibility of mercury absorption from the soil to the roots, although it is limited and directly translocated to other plant parts. However, the high concentration of mercury in the leaves that caused the high translocation value could not be believed to only come from the roots, because the leaves can absorb mercury from the air.

Based on the amount of mercury accumulation in the three plants, it can be said that these three plants have a tolerance to heavy metals because it can grow and develop well in soils that have a high toxicity. Tolerance can come from the ability of plants to store metals in vacuole cells or their ability to chelate metals (Chaney et al., 1997). The three plants can also be categorized as hyperaccumulator plants because they can accumulate mercury as much as 10 mg/kg (ppm) of their dry weight..

BCF and TF (Bioaccumulation Concentration Factor and Translocation Factor)

The ability of plants to accumulate metals can be seen from the BCF and TF values. The BCF value obtained will then be used to compare with standard BCF calculation. The BCF value calculation resulted from Yoon et al. (2006) (Yoon, Cao, Zhou, & Ma, 2006). The BCF value is more than one. This number is an accumulator plant, while a plant BCF value close to one is an indicator plant, and a plant BCF value less than one is an Excluder.

Table.3. BCF and TF values

Plants	BCF	TF
Guava	1.58	0.84
Lempuyang gajah	0.41	0.89
Harendong bulu	0.39	4.01

BCF is used to see the level of a metal accumulation from soil to plants, while the TF value is to see metal translocation from roots to leaves (Baker, 1981; Borolla, Mariwy, & Manuhutu, 2019). BCF and TF data from the three plants are shown in Table 3. The highest BCF value occurred in guava, which is 1.58, and the lowest in the harendong bulu (0.39). The guava BCF value is more than 1, indicating that it is a mercury accumulator plant, based on the ability to accumulate mercury from the soil to the plant parts well. Meanwhile, lempuyang gajah and harendong bulu have BCF < 1, which is categorized as an excluder or does not absorb much metal from the soil to the roots.

The results of the TF calculation show that the TF value of guava and lempuyang gajah <1 is 0.84 and 0.89, respectively. So that both plants experienced a phytostabilization mechanism or the ability of plants to transfer metals from roots to leaves was not good. Samar et al., (2019) have studied to calculate the TF value of the Kalopo Bean plant (*Calopogonium mucunoides*) (Samar, Mariwy, & Manuhutu, 2019). They found that the kapolo bean absorbs mercury for 50 days in the phytoremediation process. This process undergoes a phytostabilization mechanism.

Meanwhile, the harendong bulu has a TF value >1, which is 4.01, so that this plant undergoes a phytoextraction mechanism or has a high efficiency to translocate mercury from the roots to the leaves of the plant. This condition is the same as the research conducted by Yoon 2006 that generally, the BCF value is inversely proportional to the TF value. The BCF value will be greater than one, while the TF value will be less than one (Yoon et al., 2006).

Certainly, mercury in the three plants' leaves in the former traditional gold mining location comes from the bioaccumulation process. The plants' bioaccumulation process comes from contaminated mercury soil. This bioaccumulation is only a small part that comes from the air. This condition because anthropogenic mercury sources can potentially pollute the air on a large scale usually come from coal-burning activities for power generation, mining, oil refining, solid waste incineration, industrial processes, metal smelting, and cement production (United Nation Environment Programme (UNEP), 2013).

CONCLUSION

Based on the results of the study it can be concluded that The lempuyang gajah plant is a plant that accumulates the highest metal mercury, namely in the roots of 16.79 ppm and the leaves of 15.03 ppm.

Guava plants accumulate metals mercury in roots and leaves were 11.73 ppm and 9.90 ppm, respectively. Meanwhile, harendong plants accumulated mercury in the roots and leaves of 2.59 ppm and 10.39 ppm, respectively. The BCF values of guava, lempuyang gajah and harendong bulu are 1.58, 0.41, and 0.39, respectively. Meanwhile, the TF values of the three plants were 0.84, 0.89, and 4.01. respectively. These three plants are suitable to be used as phytoremediator agents to reduce mercury content in the former traditional gold processing land in the village of Gogorea, Buru island.

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