Accumulation of Heavy Metals of Cooper (Cu) and Lead (Pb) on *Rhizophora mucronata* in Mangrove Forest, Nelayan Village Sub Medan Labuhan Subdistrict and Jaring Halus Village, Secanggang Subdistrict, North Sumatra, Indonesia

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Abstract: This study aimed to analyzes the content of heavy metals Cu and Pb in the roots, leaves and barks of *R. mucronata* and analyze the ability of *R. mucronata* in accumulating heavy metals. Sampling was carried out at Medan Labuhan Sub-District Nelayan Village and Jaring Halus Village, Secanggang District. The analysis of Cu and Pb heavy metals was carried out at the Research Laboratory, Faculty of Pharmacy, University of North Sumatera. Using the Atomic Absorption Spectrophotometer (AAS) method.The results of this study indicate that the heavy metal content of Cu on the barks and leaves in Jaring Halus Village are higher than Nelayan Village. The content of Cu in roots are higher in Nelayan Village. The content of Pb Metal in the barks and roots in Jaring Halus Village are higher than Nelayan Village. The content of Pb metal in the leaves are higher in the Nelayan Village. Based on bio concentration factors, *R. mucronata*'s ability to accumulate Cu heavy metals are categorized as medium, whereas in accumulating Pb heavy metals are categorized as low.

1 INTRODUCTION

1.1 Background

Mangroves are one of several coastal ecosystems that have an important role. Mangrove ecosystems have the highest level of productivity compared to other coastal ecosystems. The large number of businesses using mangroves have caused the area of mangrove decreasing from year to year. These activities include coastal reclamation, land clearing for agriculture and aquaculture, industry and housing development in coastal areas. The direct impact caused by above activities is the entry of waste into the mangrove ecosystem, paricullary the waste which contains heavy metals (Hamzah and Setiawan, 2010). Mangrove community often gets supplies of pollutants such as heavy metals from industrial, household and agricultural waste. Mangrove plants include types of aquatic plants that have a very high ability to accumulate heavy metals in the waters. The process of absorbtion in plants occurs as various diffusion processes and the term used is translocation in animals. This transport occurs from cell to cell to vascular tissue so that it can be distributed to all parts of the body.

Soemirat (2003) states that the absorption process can occur through several parts of plants, namely:

1 Roots, especially for inorganic substances and hydrophilic substances.
2 Leaves for substances that are lipophilic.

Based on Panjaitan's, et al., (2009) data obtained on the content of heavy metals Pb and Cu in the mangrove forest of the Nelayan Village in Medan Labuhan District. In the water, obtained Cu content of 0.1198 mg / L and Pb content of 0.4522 mg / L. In sediments, obtained Cu content of 9.0735 mg / L and PB content of 9.9500 mg / L. From the data, it was found that sea water in the Mangrove Forest of Medan Labuhan Subdistrict was contaminated with heavy metals Cu and Pb because it exceeded the limit set by the LPM No.51 of 2004 KEPMEN, which was 0.05 mg / L.

Based on Handayani's research, (2006) data obtained on accumulation of Cu heavy metals at the root of R. mucronata trees amounted to 24.431 ppm.

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This shows that *R. mucronata* trees can be used as bioaccumulators of Cu heavy metals in mangrove forests.

1.2 Research Purpose

1. Determine the heavy metals content of Cu and Pb in the roots, leaves and barks of the tree *Rhizophora mucronata*.
2. Analyze *R. mucronata*'s ability to accumulate heavy metals Cu and Pb in the Mangrove Forest of Jaring Halus Village, Secanggang District and in the Nelayan Village of Medan Labuhan Sub-Village, so that it can be used as an accumulator of heavy metal pollution in the mangrove forest area.

2 MATERIAL AND METHOD

2.1 Time and Location

This research was conducted in two observation stations, station I was in the coastal area of Belawan, namely Mangrove Forest of Nelayan Village as an area suspected of being polluted because it was close to industry and station II in Mangrove Forest Jaring Halus Village which was suspected of being uncontaminated (control) because it was far from the industry. Heavy metal analysis was carried out at the Research Laboratory, Faculty of Pharmacy Universitas Sumatra Utara from April to August 2018.

2.2 Materials

The equipment used in this study consists of: knife, measuring tape, camera, compass, mortal and pastle, aquadest bottle, 250 ml Erlenmeyer flask, drip pipette, furnace (furnace), oven, funnel, Whatman filter paper size 42, universal pH, porcelain crucible, measuring cup, beaker glass, 100 ml and 25 ml measuring flask, thermometer, hand refractometer, hot plate, sample container, analytical scale, and atomic absorption spectrophotometry.

The materials used for this research are: sampling tally sheet, raffia, concentrated HNO3 solution, aqua bides, standard Cu and Pb solution, *R. muconata* roots sample consist of taproot, *R. muconata* leaves consist of old leaves and young leaves, *R. mucronata* barks affected by tides, sediment samples and seawater samples.

2.3 Sampling Procedure

Sampling in both locations were carried out by purposively following the transect path along the coastline. The roots, leaves, and barks samples were taken from *R. muconata* tree. Roots taken are taproots that are above the limit exposed to the tidal boundaries of the sea, while for the leaves taken are young leaves on the shoots and old leaves at the base of the twigs, the barks of the *R. mucronata* tree taken are tidal bark sea water. From the transect path, 3 sample points were taken at each location with a sample distance of 50 meters. Sample taken at each point with three replications. Data taken in the form of roots, leaves, and stem barks of *R. mucronata*. As supporting data, measurements of heavy metals in surface and sediment water (± 30 cm depth) as well as measurements of water quality parameters, such as air temperature, water temperature, water pH, and salinity at the six points were also measured.

2.4 Preparation of Roots, Leaves, Barks and Sediment Samples

Roots, leaves and barks samples are homogenized by compiling samples taken from three extraction points at each station. For the preparation of roots, leaves, and barks, the samples are cut into small pieces before smoothing. Likewise, sediment samples can be smoothed directly. After that they sre dried in an oven at a temperature of 105º C until a constant weight is obtained.

Samples of roots, leaves, barks and sediments are each weighed as much as 5 grams, placed on a hot plate to become charcoal. To speed up the occurrence of charcoal, a small amount of HNO3 can be dripped slowly. Samples that have become charcoal are added to the furnace at a temperature of 700º C (ignition) until become ash. After the ash process, roots, leaves and sediment samples dissolved by adding 10 ml of concentrated HNO3.

The solution mixture was crushed in porcelain crucible and then filtered into a 25 ml volumetric flask using whatman filter paper size 42. Crushed crisps were rinsed using twice aqua bides so that the metal content still attached to the crucible dissolved. After the filter is filtered, add aqua bides to the boundary line on the measuring flask. The solution obtained can be tested using AAS.

2.5 Water Sample Preparation

Seawater measured 100 ml, then added 10 ml of concentrated HNO3. Heat in an Erlenmeyer container on a hot plate until the volume becomes 30 ml. Drop the aqua bides solution until the volume becomes 100 ml then deposited. The precipitated solution is filtered by the water phase with whatman filter paper size 42. The solution obtained is ready to be analyzed using AAS.

2.6 Principle of Atomic Absorpsion Spectrophotometer (AAS)

AAS is set in advance according to the instructions in the manual tool. Then calibrated with a standard curve of each Cu and Pb metal with a concentration of 0; 0.2; 0.4; 0.6; 0.8 and 1 ppm. The absorbance and concentration of each sample were measured.

2.7 Data analysis

2.7.1 Real Concentration

To get the actual heavy metal concentration on the roots, barks, leaves and sediments in accordance with the standard operating procedures at the Research Laboratory, Faculty of Pharmacy, Universitas Sumater Utara, the formula used is:

$$\text{Real consentration (mg/L)} = \frac{\text{Consentration AAS (mg/l)} \times \text{Solvent volume (l)}}{\text{Sample weight (mg)}} \quad (1)$$

To get the actual concentration of heavy metals in water, the formula used is:

$$\text{Real consentration (mg/L)} = \frac{\text{Consentration AAS (mg/l)} \times \text{Sample solution (l)}}{\text{Sample weight (mg)}} \quad (2)$$

2.7.2 Bio concentration factor (BCF)

After the heavy metals content in the water have known, the data is used to calculate the ability of *R. mucronata* to accumulate heavy metals Cu and Pb through the level of bio concentration factor (BCF) using the formula:

$$\text{BCF Cu / Pb} = \frac{\text{(Heavy metal Cu / Pb) Plant)}}{\text{(Heavy metal Cu / Pb) Water}} \quad (3)$$

Information :

BCF> 1000	= High Ability
1000> BCF> 250	= Moderate Ability
BCF <250	= Low Ability

2.8 Descriptive Analysis

The data obtained was analyzed descriptively according to the environmental quality standards mentioned in the Decree of the Ministry of Environment No. 51 of 2004 for water quality. Quality standard for heavy metals in mud or sediment in Indonesia have not yet been established, so that as a reference, IADC / CEDA (1997) issued quality standards regarding metal content that can be tolerated.

3 RESULT

3.1 Aquatic Environment Conditions (temperature, water temperature, water pH, and salinity)

The condition of the aquatic environment results from in-situ measurements in the field, showing the different results between observation points. The temperature and the highest water temperature are found in the Mangrove Forest of Jaring Halus Village as well as the pH of the water. The highest salinity was obtained in the Mangrove Forest of the Nelayan Village (Table1).

Temperature at station II is higher than temperature at station I. This can be caused by the geographical location of the two observation stations. Temperature of the two observation stations can be categorized as high, this can occur due to the high intensity of the sun during sampling process.

Water temperature at station I is lower because the presence of *R. mucronata* trees in the Mangrove Forest of Nelayan Village is in a fairly close water surface closure. Whereas at the second station the existence of the *R.mucronata* tree is at the edge of the bay so that the closure of the water surface by the canopy is quite tenuous.

From the results of salinity measurements at both stations, the salinity range at station I is around 20-30 ppt with an average value of 23.4 ppt. While at station II around 20-30 ppt with an average value of 21.1 ppt. In station I, there are many mangrove forests that have been converted into fish ponds. The management of fish ponds there are pumps that take sea water and put into ponds which affect salinity in the area.

According to Hutagalung (1991) a decrease of salinity and pH as well as an increase of temperature caused a greater bioaccumulation rate because of the increasing availability of metals.

Table 1: Aquatic Environment Conditions Analyze.

Parameter	Station	
	I	II
pH Water	7	7.3
Salinity (ppt)	21,1	23.4
Water temp (°C)	27,3	28.1
Air temp (°C)	31.6	33.23

3.2 Heavy Metal Content of Cu and Pb in the Roots, Leaves and Barks of *R. mucronata*

Based on the results of measurements of heavy metals Cu and Pb on the barks, roots and leaves of the *R.mucronata* tree, showed that the barks, roots and leaves were higher accumulating Cu than in Pb metal (Table 2).

The results of measurements of heavy metals Cu and Pb on the roots of *R. mucronata* trees showed lower results compared to the content of the barks and leaves. At station I the average content of Cu in the roots of *R. mucronata* tree is around 5,033 mg/kg. The average content of Pb is around 0.884 mg/kg. At station II, content of Cu in the roots of *R. mucronata* tree is around 2,740 mg/kg. The average content of Pb is around 0.899 mg/kg. This is because the roots do not store the substances that have been absorbed from the soil for a long time. Though, it translocated to the stem, leaves, and fruits (Priyanto and Prayitno, 2004)

Based on the measurements of heavy metals Cu and Pb on the leaves of *R.mucronata* tree, the results were quite high. At station I, the average content of Cu in the leaves of *R. mucronata* trees is 7.697 mg/kg. The content of Pb is 1,160 mg/kg. At station II, the average metal content of Cu is 12,951 mg/kg. The average content of Pb is 1.138 mg/kg. The content of Cu at station II is higher than it is at station I.

The heavy metal content of Cu on the bark of station I is less than at station II. This is caused by differences in tree diameter at both stations. Diameter range at station I is 10.2 cm to 13.8 cm. While the range of trees at station II is 17.5 cm to 22.6 cm. The difference in tree diameter determines the amount of heavy metals and other substances that accumulate in the tree. The bigger the diameter of the tree, the bigger ability of the tree to accumulate heavy metals and other substances.

Table 2: Average Analysis of Heavy Metal Content of Cu and Pb in roots, leaves and barks of *R.mucronata*.

Sample	Station	Cu (mg/kg)	Pb (mg/kg)
Roots	I	5.033	0.884
Roots	II	2.740	0.899
Leafs	I	7.697	1.160
Leafs	II	12.951	1.138
Barks	I	8.357	1.115
Barks	II	21.734	2.480

3.3 Content of Heavy Metals Cu and Pb in the Water and Sediment

The content of heavy metals Cu and Pb in water and sediment in the water in the mangrove forest area of Jaring Halus Village is higher than the ones in the Nelayan Village Mangrove Forest. The average content of Cu and Pb in sediments in the Mangrove Forest Village of the Fishermen area is higher than those in Jaring Halus Mangrove Forest (table 3).

By the results of measurements of Cu and Pb heavy metals in water at both sampling stations, it can be seen that the content of Cu heavy metal has a higher concentration than the content of Pb metal. This is due to the origin of Cu metal pollution which is the main industrial waste that is above the sampling location at station I, the waste area of private oil palm plantations and community-owned agriculture at station II. Sea transportation activities contribute to Cu heavy metals in the environment but in doses that are not too huge. At station II, there is an oil palm management industry and sea transportation activities to connect the Jaring Halus Village at Secanggang District. In addition, Pb heavy metals in the environment are generally obtained from transportation activities and industrial activities. At the station I, there are a few sea transportation activities but there are numerous industrial activities in the Medan Industrial Area which is above the sampling location of station I.

From the results of measurement of Cu heavy metals at station I, the average data obtained as 0.0439 L/kg. While at station II the average data was obtained as 0.0496 L/kg. the content of Cu heavy metal at station II is higher than station I. This can be caused due to differences in sampling. At the time of sampling at station I, it was carried out when the dead tide (a small amount of tide entered the location). At station II the sampling was carried out during high tides (high tide entered at that location). From the measurement content of Pb heavy metal at station I, the data average obtained as 0.0137 L/kg, while in station II the average data obtained as 0.02457 L/kg. The content of Cu and Pb heavy metals in both stations have exceeded the limit set for seawater quality standards, 0.008 L / kg. (KLPM KEPMEN No. 51 of 2004) .

From the results of measurements content of Cu and Pb heavy metal in sediments at both sampling stations obtained the content of Cu heavy metal with average of 0,9003 mg/kg. While in station II, the content of Cu heavy metal obtained with an average of 0.776 mg/kg. the content of Cu heavy metal at station I is higher than station II because at station I there are many industrial activities and at station II it is likened to control. Cu pollution at both stations can still be tolerated (IADC/CEDA 1997).

From the measurement content of Pb heavy metal at station I, the average data obtained as 2.7588 mg/kg, while in station II the average data obtained as 0.9003 mg/kg and still included the tolerance limit (IADC / CEDA 1997).

The content of heavy metal in sediments is higher than it is in water. This can occur because of sedimentation in sediments during heavy metal content in high water. Heavy metals have properties that easily bind organic matter and settle in the bottom of the water and bind to sedimentary particles. So that, the content of heavy metal in sediments is higher it is in water.

Table 3: Average Analysis content of Heavy Metal of Cu and Pb in Water and Sediments.

Sample	Station	Cu (mg/kg)	Pb (mg/kg)	Quality Standart
Water	I	0.0439	0.0137	KEPMEN KLH No. 51, 2004 (0,008 mg/l).
Water	II	0.0496	0.02457	
Sediments	I	0.9003	2,75883	IADC/CEDA 1997, Cu (600 mg/kg) and Pb (1000 mg/kg).
Sediments	II	0.776	0.9003333	

3.4 Bioconcentration Factor (BCF) To Analyze the Ability of *R. mucronata* in Accumulating Heavy Metal Cu and Pb.

Based on the calculation of the value of bio concentration factor (BCF), it is known that the highest BCF value is for Cu metal which is 754,524 and the lowest BCF value is 188,527 for Pb metal. The value of the Cu and Pb bio concentration factors in two stations can be seen in Table 4.

From the results of the calculation of the bio concentration factor values for heavy metals Cu at the first station it can be concluded that the ability of *R. mucronata* to accumulate Cu metal is better than Pb metal. For station I Cu metal BCF values amount to 480,357 and for Pb metals amount to 230,533. At station II, the BCF value of Cu metal is 754,524 and for Pb metal is 188,527. In accumulating *Cu R. mucronata* metals are categorized as medium while in accumulating Pb it is categorized as low.

Table 4: Value of Cu and Pb Bio concentration (BCF) Factors in Nelayan Village and Jaring Halus Village.

Station	Cu Consentration		Bcf Cu (L/Kg)	Pb Consentration		Bcf Pb (L/Kg)
	Plant = Total Root, Bark And Leaves (Mg/Kg)	Water (L/Kg)		Plant = Total Root, Bark And Leaves (Mg/Kg)	Water (L/Kg)	
I	21.0877	0.0439	480.357	3.1583	0.0137	230.533
Ii	37.4244	0.0496	754.524	4.6321	0.02457	188.527

4 CONCLUSION

The content of heavy metal Cu in *R. mucronata* roots in Nelayan Village (5,033 mg/kg) are higher than Jaring Halus Village (2,740 mg / kg), while for the content of Pb in Jaring Halus Village (0,899 mg/kg) are higher than Nelayan Village (0.884 mg/kg). The content of Cu in *R. mucronata* leaves are higher in jaring Halus Village (12,951 mg/kg) than in Nelayan Village (7,697 mg/kg), while for the content of Pb are higher in Nelayan Village (1,160 mg/kg) than in Jaring Halus Village (1,138 mg/kg). The content of Cu in *R. mucronata* barks in Jaring Halus Village (21.734 mg/kg) are higher than Nelayan Village (8.35 mg/kg), Pb content in Jaring Halus Village (2.480 mg/kg) are higher than in Nelayan Village (1.115 mg/kg).

The ability of *R. mucronata* in accumulating Cu heavy metals in Nelayan Village and Jaring Halus Village is categorized as medium with BCF values of 480.357 and 754.524, whereas in accumulating Pb heavy metals in Nelayan Village and Jaring

Halus Village is categorized as low with BCF values of 230,533 and 188,527.

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