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# Accumulation of Heavy Metals Lead (Pb) and Copper (Cu) in Mangrove Area of *Avicennia marina* in Manyar Subdistrict, Gresik District, East Java

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Atomic Absorption Spectrophotometer (AAS)

**Abstract** Human and industrial activities in the area of Manyar Subdistrict Mangrove Waters, Gresik District can increase pollution and the presence of heavy metals in aquatic environments and aquatic organisms. This triggers input of heavy metals, especially Pb and Cu. Mangroves can accumulate and have a high tolerance for heavy metals so they can be used as plants for phytoremidiation purposes (phytostabilization). The mangrove ecosystem can be used as a control of heavy metal pollution and can also be used as a pollutant trap. This study was conducted to determine the potential of phytoremidiation based on the accumulation and translocation of heavy metals in *Avicennia marina* mangroves in absorbing heavy metals Pb and Cu from their environment. Based on research results MacFarlane *et al.,* (2007) mangrove *Avicennia marina* is one of the plants that can effectively accumulate heavy metals due to its root system. The sample was analyzed using AAS (Atomic Absorption Spectrophotometer). The research parameters include levels of Pb and Cu metals in sediments, mangrove roots and leaves. The results of Pb heavy metal concentrations ranged from 0.30 - 4.84 ppm and Cu ranged from 0.27 - 11.42 ppm with the highest values found in sediment stations 2. *Avicennia marina* at the study site was excluder because the BCF value < 1 which ranged from 0.02 - 0.18 ppm and phyto-extraction because the value of TF > 1 ranged from 0.35 to 1.69 ppm. The results of this study indicate that *Avicennia marina* mangroves can be developed into phytoremidiation agents because they are able to absorb and transfer heavy metals from the environment to other body tissues.

## Introduction

In the current era of globalization, it cannot be denied that coastal areas have experienced a very high level of utilization, starting from the fields of trade, tourism, industry, settlements, and capture fisheries. Pollution is a condition which has changed from its initial condition which leads to a worse level. Port activities can also be a source of heavy metal pollution in the surrounding waters. One type of pollutant produced by industry is Toxic Waste (B3 Waste). Such conditions are caused by the inclusion of pollutants. This change will have an impact on existing organisms and the environment and settled in the environment or region. Mangrove ecosystems are affected by human activities in the form of various types of contaminants such as heavy metals (Kulkarni, Deobagkar and Zinjarde, 2018). (de Almeida Duarte et al., 2017) metals are one of the pollutants that have the highest toxicity because they have potential risks to organism.

The toxicity caused by heavy metals is considered a serious threat to mangrove ecosystems that have the potential to affect human health and other organisms (Wu et al., 2014). Metal pollutants which most often contaminate mangrove ecosystems originate from industry (MacFarlane, Koller and Blomberg, 2007). Mangrove ecosystems for

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humans also benefit directly and indirectly from the socio-economic conditions of the surrounding population. In addition, the mangrove ecosystem also functions as a trap for sediments and prevents erosion as well as stabilizing land forms in the estuary region (Hamzah and Setiawan, 2010). Based on the research of Pramudji (2000), mangrove plants have a unique root that plays a role in capturing all material originating from the sea or land, the pollutants and pollutants that were caused by human activities will be trapped in the mangrove forest area. Whereas (Pinheiro *et al.,* 2012) reiterated that the roots of mangrove plants have an important role in purifying water and sediments that have been contaminated by heavy metals carried by tides.

One of the coastal areas in Indonesia, namely the Mangrove area in Gresik District, East Java, is an area that is densely populated with human activities which results in pollution of the environment around the coast. Based on data from the Gresik District Central Bureau of Statistics in 2013 there were 402 large and medium industrial units spread across several sub-districts in Gresik. The three sub-districts that have increased growth in activity and pollution due to the number of industries, namely Gresik District, Kebomas District and Manyar District. Some large industries located in Gresik District include PT. Petrokimia Gresik, Gresik International Port Area, PT. Semen Gresik and Maspion V. Industrial Estate This industry has a positive impact, which is able to absorb labor and increase regional income. But on the other hand, it also causes negative impacts from human activities and industries produce waste that can pollute the environment. Ni’am (2018), industries in Gresik District produce B3 waste as much as 66.4%, which is around 12.9 million tons per year or 1.1 million tons per month.

In general, heavy metals for plant growth and development are divided into two, namely essential and non-essential metals. Cu is an essential metal, while Pb is a non-essential metal for plants (Hamzah and Setiawan, 2010). One type of heavy metal that is often encountered is lead (Pb) and copper (Cu). Lead is one of the heavy metals produced from motor vehicle emissions and industrial activities. In industry, lead is used in the fields of automotive, ceramic manufacturing, PVC and solder plastic industries. Pb heavy metals are also commonly used in a mixture of paints, pesticides and mixtures in vehicle fuels. Whereas the presence of Cu in the environment can accumulate in the waters or be deposited in sediments (Ni'am, 2018; Ida and Purwiyanto, 2013). The resulting heavy metal waste can cause physical disturbances in mangroves, in the form of yellowing and falling leaves, the worst level can cause death. Continuous events that last for a long time will cause the existence of the mangrove ecosystem to decline and even disappear (Prasetyo, Santoso and Lilik, 2017).

Based on the above problems in this study, we will discuss the accumulation of Pb and Cu in the mangrove *Avicennia marina*. Mangrove forest through its root system that reaches the ground and spreads widely is expected to be able to function to absorb pollutant contents, especially heavy metals in the surrounding aquatic environment, so that the pollutant's toxicity is mangrove forests can be reduced. The purpose of this study was to analyze the accumulation of Pb and Cu in the mangrove area of *Avicennia marina*. This research is important because if organisms, both biota and mangrove, are exposed to high amounts, they will cause disruption in metabolism and inhibition of growth, even death.

## Materials and methods

This research was conducted from February to March 2019 in the Manyar Mangrove District of Gresik District, East Java. The number of sample points taken is 2 points that are considered to represent the mangrove


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area. Furthermore, water samples, sediments and mangroves were analyzed in the Chemistry Laboratory of the Faculty of Mathematics and Natural Sciences Universitas Brawijaya, Malang. The research equipment used included knives, polyethylene bottles, cool boxes, GPS, scales, sample containers, volumetric flasks, Whatman filter paper no. 41, DO meter (Lutron PDO-520 type), pH meter (HANNA), Erlenmeyer, AAS (Atomic Absorption Spectrophotometry). The materials used in this study were distilled water, concentrated $HNO_3$, aluminum foil.

*Station Point Determination*

The observation station is determined by "purposive sampling" based on the characteristics or conditions of the environment around the mangrove ecosystem and determined as many as 2 observation stations which are polluted areas. Station 1 is located on the river mouth which is close to the residential area at the coordinate point 7°6’33” S and 112°35’48’ E. While Station 2 is located close to the industry at the coordinate point 7°7’26” S and 112°38’10’ E. The point of sampling location can be seen in **Figure 1**.


**Figure 1.** Research Location (Sub-district of Manyar, Gresik District) (Google Earth, 2019).

*Water Sampling Analysis*

Measurement and sampling of water was carried out in an in-situ parameter measured including temperature parameters, DO (Dissolved Oxygen), salinity and pH using DO meter, pH meter and refractometer. Sampling was carried out in February-March 2019. Water quality data analysis was carried out by descriptive method with a quantitative approach.

*Sediment Sampling Analysis*

Sediment samples were taken using Ekman grab and put into polyethylene bottles (Hamzah and Pancawati, 2013). Sediment samples were taken at each station. The sediment samples taken are bottom waters which have a depth of less than 20 cm. The sediment is dried in an oven with a temperature of 105°C for 12 hours to remove the water content. Sediment samples were weighed


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as much as 5 grams, then dissolved by adding 20 ml of concentrated $HNO_3$ and 10 ml of $HClO_4$, then added distilled water until the volume became 50 ml. Heat in the hot plate until the volume is reduced by 30 ml. If no mist has occurred, repeat adding 20 ml of concentrated $HNO_3$ and 10 ml of $HClO_4$, then reheating until fog occurs. Add the solution again with distilled water until the volume becomes 50 ml, then precipitate. The precipitated solution is filtered by the water phase with filter paper. The solution obtained is ready to be analyzed using Grafic-Furnice AAS.

*Mangrove Sampling Techniques for Avicennia marina*

Parts taken from the *Avicennia marina* include:

1. Roots
   The root sample taken is the breath root (*pneumatophora*) which is outside and inside the sediment (wire root), collected as heavy as 5 grams in 1 tree with a diameter of 0.4 - 0.6 cm. Root samples taken are mangrove roots that enter into the sediment (Hamzah and Setiawan, 2010).
2. Leaves
   The leaves taken are old green dark leaves 4-8 cm long which are located at the base of the branch Taking leaves of about 30 leave sheet and directly put into the cool box are then taken to the laboratory to be tested for heavy metals (Hamzah and Setiawan, 2010).

The root and leave sample were each weighed as much as 5 grams, then put in the furnace at a temperature of 600-650°C (ignition) for 3-4 hours. After finishing the process of ignoring the sample was dissolved by adding 20 ml of concentrated $HNO_3$ and 10 ml of $HClO_4$, then added distilled water until the volume became 50 ml. Heat in the hot plate until the volume is reduced by 30 ml. If no mist has occurred, repeat adding 20 ml of concentrated $HNO_3$ and 10 ml of $HClO_4$, then reheating until fog occurs. Add the solution again with distilled water until the volume becomes 50 ml, then precipitate. The precipitated solution is filtered by the water phase with filter paper. The solution obtained is ready to be analyzed using Grafic-Furnice AAS (Hamzah and Pancawati, 2013).

*Data Analysis*

To find out the metal accumulation in mangroves is done by calculating metal concentrations in sediments, roots and leaves. Comparison between metal concentrations in roots / leaves and concentrations in sediments is known as bio-concentration factor (BCF) (Hamzah and Setiawan, 2010). BCF in leaves and roots is calculated to determine how much metal concentration in leaves and roots originates from the environment (MacFarlane, Koller and Blomberg, 2007). The algorithm of BCF as below:

$$\mathrm{BCF} = \frac{[\text{Heavy metals}]_{(\text{roots/leaves})}}{[\text{Heavy metals}]_{\text{sediment}}}$$

In addition, a comparison between metal concentrations in leaves and roots is also calculated, known as translocation factors (TF). TF values were calculated to determine the displacement of metal accumulation from root to shoot (MacFarlane, Koller and Blomberg, 2007). The difference between BCF and TF values is used to calculate phytoremediation. The algorithm of TF as below:

$$\mathrm{TF} = \frac{[\text{Heavy metals}]_{\text{leaves}}}{[\text{Heavy metals}]_{\text{roots}}}$$


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## Results and discussions

### *Water Quality Data*

Water quality data collection was carried out in 2 stations with 3 repetitions in Manyar District, Gresik District. Water quality measurements are carried out in-situ at each observation station. The average results of water quality measurements at 2 stations can be seen in **Table 1.**

**Table 1. Results of Average Water Quality Measurements**

| No | Parameter | Unit | Station | |
|---|---|---|---|---|
| | | | 1 | 2 |
| 1 | DO | mg/L | 3.4 | 4.6 |
| 2 | Temperature | °C | 31.7 | 31.5 |
| 3 | pH | - | 6.7 | 7.1 |
| 4 | Salinity | ‰ | 13 | 19 |

Station 1 characteristics close to river estuary which is a discharge from household waste while station 2 is close to industry and human activities and ports. Based on the results of measurements of water quality parameters at the observation station shows that in general the results of pH measurements do not have different values ranging from 6.7 to 7. For temperature, the values obtained are around 31.5 - 31.7°C. Temperature has an important influence on the solubility of heavy metals. Increasing water temperature leads to an increase in the solubility of toxic heavy metals (Hertika *et al.,* 2019). Where the pH value and temperature based on seawater quality standards for marine biota in mangrove areas are still below the quality standard (PERMEN LH No. 5 of 2014, 2014). The salinity value and dissolved oxygen value have different values between station 1 and station 2. Station 1 is the station that has the lowest DO and salinity value (3.4 mg / L & 13 ‰). The results of dissolved oxygen were almost similar to previous studies (Hertika *et al.,* 2019). This low level of dissolved oxygen also affects heavy metal poisoning, because low concentrations of dissolved oxygen encourage increased toxicity of heavy metals in water (Hertika *et al.,* 2019). The low salinity at station 1 is assumed that Station 1 is close to the river mouth, so that it gets the influence of freshwater input. Anaerobic substrate condition is one of the characteristics of the estuary region. This indicates that in general dissolved oxygen in the estuary region is indeed very low and anaerobic. Dissolved oxygen from water is absorbed into the sediment and used for respiratory activities by bacteria (Hamzah and Setiawan, 2010). This low oxygen level can also be caused by waste disposal from industrial activities that are around sampling so as to inhibit phytoplankton or aquatic plants for photosynthesis.

### *Data on Heavy Metal Content of Pb and Cu in Sediments, Mangrove Roots and Leaves*

The results of the study showed the presence of Pb and Cu metals in mangroves and in their abiotic environments, namely in sediment and water. The ability to accumulate Pb and Cu metals by mangroves is certainly not free from the abiotic environment as a place of life, namely sediment and waters (substrate). The Pb metal content in sediments ranges from 3.78 to 4.84 ppm. The value of the Pb metal concentration range is still below the normal threshold range according to the determination of the normal concentration of Pb in sediments according to the National Oceanic and Atmospheric Administration (NOAA) which is equal to 46.7 ppm. Whereas for Cu metals ranged from 2.72 - 11.42 ppm. The high levels of Cu heavy metals at Station 2 are suspected to occur contaminants in coastal areas often


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concentrated in lagoon sediments and ports because of river transportation, human activities, and input from industrial activities (Hamzah and Setiawan, 2010). The high levels of Cu in station 2 are due to the nature of Cu which has a low solubility and is easy to settle, thus triggering the presence of Cu in the sediment (Awaliyah, Yona and Pratiwi, 2019). When compared between Pb and Cu, the metal with the greatest presence can be absorbed is Cu. The metal content of Pb which is a group of non-essential heavy metals accumulates less in sediments (de Almeida Duarte *et al.,* 2017; Wu *et al.,* 2014). Metal pollution in mangrove sediments is limited to total metal concentrations (Kariada and Irsadi, 2014).

**Table 2. Heavy Metal Content of Pb and Cu in Sediments, Mangrove Roots and Leaves**

| No | Parameter | Unit | Station | |
|---|---|---|---|---|
| | | | 1 | 2 |
| | **Sediment** | | | |
| 1 | Pb | ppm | 4.84 | 3.78 |
| 2 | Cu | ppm | 2.72 | 11.42 |
| | **Roots** | | | |
| 3 | Pb | ppm | 0.86 | 0.32 |
| 4 | Cu | ppm | 0.36 | 0.31 |
| | **Leaves** | | | |
| 5 | Pb | ppm | 0.30 | 0.54 |
| 6 | Cu | ppm | 0.48 | 0.27 |

The average concentration of Pb heavy metals is higher than the concentration of heavy metals Cu in the root tissue. The Pb metal content in roots ranges from 0.32 - 0.86 ppm with the highest concentration found at Station 1. The content of Cu in the root's ranges from 0.31 - 0.36 ppm. While the highest Pb metal content in leaves is at Station 2 which is 0.54 ppm. The highest Cu content in leaves is found in Station 1. Cu content ranges from 0.27 - 0.48 ppm. Based on the distribution of Pb and Cu heavy metals in the roots and leaves of *Avicennia marina* in this study showed that the average content of Pb and Cu in the roots was greater than in the leaves so that the ability to accumulate was higher in the root tissue. The mangrove root is able to lift and store heavy Pb and Cu metals found in the waters of Manyar Subdistrict, Gresik District. This is thought to be root tissue that interacts directly with sediment and contaminated water so that the heavy metal content of Pb at the root is higher than the leaves. This is similar to the results of the study (Hamzah and Setiawan, 2010) where the highest average metal content is found in root tissue.

*Avicennia marina* is a mangrove species that is very tight in absorbing metals. Based on physiological mechanisms, mangroves actively reduce the absorption of heavy metals when the concentration of heavy metals in sediments is high. Absorption is still done, but in limited amounts and accumulates in the roots. In addition, there are endodermic cells in the roots which become a filter in the process of absorption of heavy metals. From the roots, the metal will be translocated to other tissues such as stems and leaves and undergo a process of complexation with other substances such as phytochelatin (MacFarlane, Pulkownik and Burchett, 2003). The process of entering heavy metals into plants is in the form of cations or anions. Pb metal is a cation because it is positively charged. Metal absorption is carried out by the root tip, absorption occurs in the root epidermis. Then the ions move towards xylem, through the cytoplasmic system (simplas) moving from cell to cell. The absorption process then takes place in two processes, which are ionized directly into the meristem


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cells of the leaves which later functions as a support for plant growth, then immobilized ions are absorbed on the old leaves cells to be aborted later. The process of taking Pb and Cu metals in mangroves is a passive transport system. Passive transport systems are those transported by physical forces, namely high to low concentrations that occur in cells. After being absorbed by roots, heavy metals will be translocation to all parts of the plant (Wulandari, Budihastuti and Hastuti, 2018; Usman, Alkredaa and Al-Wabel, 2013).

*Accumulation and Translocation of Pb and Cu Metals by Avicennia marina*

The results of the calculation of BCF and TF values are varied (**Table 3**). The presence of heavy metal Pb in the sediment around the *Avicennia marina* root caused bioaccumulation in the root section at Station 1 to have the highest value. Bioaccumulation is a process of increasing the concentration of heavy metals in the body of an organism. To find out the bioaccumulation of heavy metals Pb and Cu at the root, Biocentration Factor (BCF) is needed. Calculation of the Pb BCF value at the root at Station 1 is the highest than the BCF Cu value (0.18 ppm). Unlike the BCF value in leaves tissue, it ranges from 0.02 - 0.18 ppm where the highest value is found in Cu metal at Station 1 (0.18 ppm). It is suspected that the *Avicennia marina* at this station has localized heavy metals Cu in roots to other parts such as leaves. The mechanism carried out by *Avicennia marina* on high concentration of pollutants is by amelioration and tolerance techniques. An important part in the process of translocation of heavy metals into the plant's body from the root to the leaves is the xylem vessels in the roots. Heavy metals Pb and Cu are translocated from the roots to all mangroves, including leaves through xylem vessels (Supriyantini *et al.,* 2017).

**Table 3. Calculation of Bioconcentration Factor (BCF) and Translocation Factor (TF) Heavy Metals Pb and Cu**

| Station | Parameter | BCF | | TF |
|---|---|---|---|---|
| | | Roots | Leaves | |
| 1 | Pb | 0.18 | 0.06 | 0.35 |
| | Cu | 0.13 | 0.18 | 1.33 |
| 2 | Pb | 0.08 | 0.14 | 1.69 |
| | Cu | 0.02 | 0.02 | 0.87 |

Based on the calculation of the BCF value of less than 1, *Avicennia marina* in the research location is an excluder plant that can prevent the entry of heavy metals from sediments to mangrove roots. This is evidenced even though heavy metals are high in waters but in tissues, especially roots are quite low. But it does not rule out the possibility when large concentrations in the sediment cause absorption by root tissue. Based on bioconcentration factors, the ability of *Avicennia marina* in accumulating Pb and Cu metals is relatively low because it has a BCF value of < 250 (MacFarlane, Koller and Blomberg, 2007).

The average value of *Avicennia marina* TF from root to leaves for essential metals (Cu) was higher when compared to non-essential metals (Pb) of 1.02 and 1.10. The low TF value in Pb shows that mangroves use the metal for metabolic activity and growth, whereas in Cu the metal mobility process from root to leaves is very high. Based on the results of these calculations, *Avicennia marina* against Pb and Cu heavy metals in the mangrove ecosystem of Manyar Subdistrict, Gresik District has phyto-extraction ability because TF is more than 1. Plants with TF more than 1 are classified as high-efficiency plants for translocation of heavy metals from roots to parts plants above the ground,


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especially at station 2 for heavy metals Pb with the highest TF value of 1.69, and station 1 for metals heavy Cu of 1.33 because the concentration of heavy metals in leaves is much higher than the concentration of heavy metals in root. But the high concentration in the leaves which causes high translocation values cannot be believed to be pure from the roots because the leaves can also absorb heavy metals from the air. Broadly speaking, based on the calculation of BCF and TF it was found that *Avicennia marina* was able to reduce heavy metal pollutants from the environment by absorption through the roots and translocation towards the leaves. The high BCF root value for all metals is supported by high concentrations of all metals in the roots and low sedimentsproduce high root BCF values (Hamzah and Setiawan, 2010).

According to Kulkarni, Deobagkar and Zinjarde (2018), mangroves can actively avoid excess heavy metal inputs and have a special treatment power through root organs. Heavy metals that enter plants depend on absorption (absorption) at the roots. The metals that have entered the plant through the roots will be distributed to the leaves and stored in the leaves. Apart from the translocation process, heavy metals from leaves can also be derived from atmospheric transport through attaching heavy metals from leaves and then entering the stomata (Hamzah and Pancawati, 2013; Usman, Alkredaa and Al-Wabel, 2013; Analuddin *et al.,* 2017). Higher concentrations of heavy metals in the mangrove tissue recorded in this study are a good indication of the ability of mangroves to act as phytoremediation agents by taking heavy metals and collecting them in their tissues. This is very important to prevent the spread of heavy metal pollution in coastal areas and to maintain the health of the coastal zone.

## Conclusions and suggestions

In general, heavy metal concentrations in Manyar Subdistrict, Gresik District in sediments are higher than those of mangrove roots and leaves because of the nature of sediments that accumulate heavy metals from the water column. The highest metal content is Cu in sediments at Station 2 (11.42 ppm). *Avicennia marina* in Manyar Subdistrict is excluder because BCF value is less than 1 and phytoextraction because TF value is more than 1. *Avicennia marina* is able to absorb pollutants from the environment through roots and distribute to other parts, especially leaves. Expected to know the benefits of the *A. marina* plant, can motivate the community to preserve the forest mangrove. Disposal of domestic and industrial waste is necessary monitored, so as not to exceed the standard standards quality. On the basis of it is expected that further research will be carried out about how wide the minimum plant is mangroves needed in one region, so that it can anticipate environmental changes are mainly a result heavy metal waste. Therefore, utilization is expected to be realized coastal estuary ecosystem area of the river which guarantees sustainability and power use continuously.

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