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Accumulation of Heavy Metal Copper (Cu) in Mangrove Vegetation in River Flow of the Sea Coast

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ABSTRACT

Mangroves have ecological functions, one of which is being able to absorb and accumulate heavy metals which can pollute the environment. The aim of the research is to assess the ability of mangroves to accumulate the heavy metal Cu. The research location is on the East Surabaya River Coast. The research method is purposive sampling with a distance of 250 m from one point to another. The pH value is classified as neutral to slightly alkaline. The salinity value is classified as very high. The texture of mangrove sediments has fine particles consisting of clay and dust. Cu concentrations in sediments are still below quality standards according to the United States Environmental Protection Agency (US-EPA). The Cu concentration in the sediment was highest in the Tambak Oso River with an average of 50.50 mg/kg. The concentration of heavy metals in the roots is higher than in the leaves. The bioconcentration factor (BCF) value is classified as the excluder, while the translocation factor value is classified as the phytostabilization and phytoextraction classes.

1. INTRODUCTION

The East Coast of Surabaya is an area of Surabaya City which is an industrial city and densely populated with 8,633 km² (BPS Surabaya City, 2023). The negative impact of the rapid industry causes environmental pollution due to industrial waste and the presence of non-domestic waste due to the impact of dense settlements. Both of these wastes which are discharged directly into rivers mostly contain heavy metals including copper (Cu). The presence of copper heavy metals in industrial liquid waste is in the form of bivalent Cu (II) ions (Setiawan, 2008). In addition, household waste such as soap waste and floor cleaners also contain CuO which is discharged directly into rivers (T *et al*., 2013). The river flow will be carried to the river mouth.

Jagir River, Kebon Agung River and Tambak Oso River are rivers that are close to industrial and residential locations. Hemanto & Nawiyanto (2012) said that the Surabaya City area has changed into an industrial area so that companies in the area dump their waste into the river. In addition, the source of livelihood for the surrounding community is as fishermen or shipping activities carried out in the estuary/downstream areas. The use of ships usually involves painting activities, so that the heavy metal Cu acts as the main ingredient in giving blue and metallic colors to anti-rust paint (Santi *et al*., 2017). The source of Cu in waters comes more from air containing Cu in the form of particulates carried by rainwater.

Mangrove is a type of tree plant that grows and develops on the coast. Mangrove has a very important role both biologically, physically and chemically. Biologically, mangrove plays a role in balancing the ecosystem and the sustainability of marine biota. Physically, mangrove acts as a protector of abrasion in coastal areas. Chemically, mangrove acts as a biofilter because it has the ability to accumulate heavy metals that have the potential to pollute the environment. The socio-economic function of mangrove forests as objects of education and research. According to the book by (Rahim & Wahyuni, 2017) explains that the mangrove ecosystem is a unique ecosystem because it includes land and sea ecosystems with various types of land and water biota. This unique condition is what makes it attractive for education and research.

Mangroves are able to accumulate heavy metals and are very tolerant to heavy metals. This is in line with research Susilawati, *et al*. (2021) that mangroves act as phytoremediation agents for waters and have biofilter capabilities because they are able to filter, bind, and capture pollution. Mangrove trees can stop or reduce the absorption and accumulation of heavy metals through the roots. This process reduces the movement of metals and also dilutes them, thereby reducing the entry of metals into the food chain system (Hamzah, 2010). The absorption and accumulation of heavy metals by plants is divided into three processes, including metal absorption by roots, metal translocation from roots to other parts of plants, and metal localization in certain cell parts which aims to prevent inhibiting the metabolism of the plant (Hamzah & Priyadarshini, 2019).

Bioconcentration is the accumulation of heavy metal Cu absorbed by mangrove plant roots from sediment and absorbed by leaves from air pollution. Bioconcentration Factor (BCF) is used to calculate the ability of roots or leaves to accumulate heavy metals. Translocation is the movement of heavy metal Cu from plant roots to other plant parts such as stems. Translocation Factor (TF) is used to calculate the ability of plant roots to absorb heavy metals and translocate them to other plant parts.

Based on the results of previous studies, various types of mangroves are able to accumulate heavy metals because they have a root system that can absorb metals. The study (Othman *et al*., 2015) explains that the Sonneratia alba mangrove can accumulate copper (Cu) heavy metals, especially in the tissue. In addition, in the study, the (Setiawan, 2015) Sonneratia alba type can accumulate Pb heavy metals more than the *Avicennia marina* type by 35.3 ppm. The results of Dewi's study (2018) explain that different types of mangroves, namely the *Avicennia marina* and *Rhizophora mucronata* mangroves, can accumulate Cu heavy metals in sediments ranging from 0.685 – 1.545 mg kg^{-1}. The average sediment concentration of Cu heavy metals in *Avicennia marina* and *Avicennia alba* which dominates around the Jagir River estuary is 63.85 mg kg^{-1} and 24.85 mg kg^{-1} (Harnani & Titah, 2017).

Based on the background above, it is necessary to conduct a study that explains the content of heavy metal copper (Cu) in mangrove vegetation in the Jagir River, Kebon Agung and Tambak Oso. The purpose of the study was to examine the ability of mangroves in the Jagir River, Kebon Agung and Tambak Oso to accumulate heavy metal Cu in terms of bioconcentration factors (BCF) and translocation factors (TF) so that they can be used as accumulators of heavy metal pollutants in the mangrove area. This study includes the chemical and physical characteristics of mangrove sediments so that factors that affect the size of metal accumulation can be identified, in addition, this study was conducted in 3 locations at once so that a comparison of pollutant sources can be made.

2. RESEARCH LOCATION

The research was conducted in December 2023 – April 2024 at the Jagir River Estuary, Kebon Agung and Tambak Oso, Surabaya City, East Java (Figure 1). Laboratory analysis activities were carried out at the Faculty of Agriculture, National Development University "Veteran" East Java.

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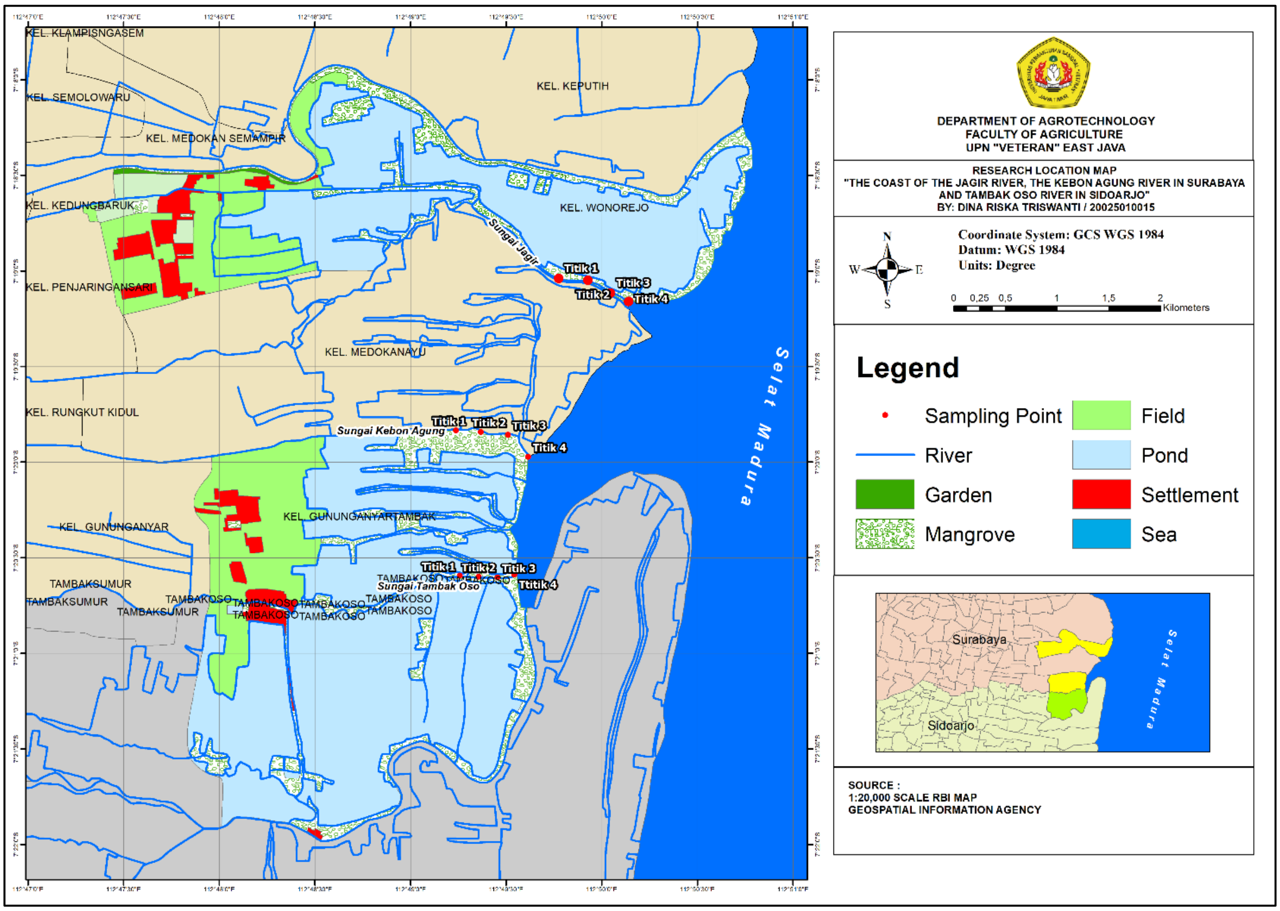

Figure 1. Research location

2.1. Data Collection

Determination of sampling points for leaves, roots, and sediments in mangrove plants was carried out based on the purposive sampling method. Determination of sampling points was along the river to the estuary because heavy metal pollution Cu could come from rivers and the sea. Sampling of soil, roots, leaves and sediments was taken on the right and left according to the location point, one river has 4 sampling locations, the distance between one point and another is 250 meters.

Sediment sampling was taken using a trowel to the root area of approximately 30 cm. The criteria for sampling in mangrove plants are mangroves that have a tree circumference of 20-25 cm and a height of approximately 3-5 m. The mangroves taken are mangroves that are neither young nor old. Root sampling used a saw on the part of the root that was submerged in the sediment of approximately 30 cm. Leaf sampling was carried out on young and old leaves, the leaves taken were leaves that came from the same tree as the sampling, the criteria for leaves were intact and healthy leaves. The physicochemical parameters of the sediment measured consisted of pH, salinity and soil texture. pH was measured using the conductometry method, salinity using the conductometry method, sediment texture using the pipette method and heavy metal Cu was measured using the wet ashing method of the AAS (Atomic Absorption Spectophotometer) tool.

2.2. Bioconcentration Factor (BCF)

According to (Baker, 1981; Rachmawati *et al*., 2018) bioconcentration factor analysis is used to calculate the ability of mangrove roots and leaves to accumulate metals. BCF can be calculated using the formula:

$$\text{BCF} = \frac{\textit{Heavy metal concentration in Root or Leaf}}{\textit{Heavy metal concentration in Sediment}} \quad (1)$$

There are 3 BCF categories, namely accumulator (high accumulation rate) if BCF value > 1, indicator (moderate accumulation level) if BCF value = 1, and excluder (low accumulation rate) if BCF value < 1.

2.3. Translocation Factor (TF)

According to (MacFarlane *et al*., 2003; Rachmawati *et al*., 2018) the heavy metal translocation factor is used to calculate the process of heavy metal translocation from roots to leaves. TF can be calculated using the formula:

$$\text{TF} = \frac{\textit{Heavy metal concentration in Leaf}}{\textit{Heavy metal concentration in Root}} \quad (2)$$

where TF value < 1 is called phytostabilization plants, and TF value > 1 is called phytoextraction plant.

2.4. Data Analysis

The observation data were analyzed using the Pearson correlation test using IBM SPSS Statistics 22 software. The correlation test was used to determine the closeness of the relationship between Cu heavy metals and physicochemical parameters. The correlation value is 0-1, the closer to 1 the stronger the relationship, the closer to 0 the weaker the relationship.

3. RESULTS AND DISCUSSION

The Jagir River is 9 km long, 73 m wide and has an area of 253 km², along the river there is land use for housing, shops, cosmetics and furniture industries. The color of the water in the Jagir River looks brown in general and the flow is calm but sometimes the current is moderate depending on the time. Along the flow towards the estuary there are several mangrove plants dominated by Rhizophora sp. mangroves, the density of mangroves is not only filled by *Rhizophora sp.,* there are also *Avicennia sp.* still does not dominate.

Kebon Agung River is 11 km long and 7–11 m wide, along the flow there are settlements and various industries. The color of the water in the Kebon Agung River looks brown in general and the flow is calm but sometimes the current is moderate depending on the time. Along the flow to the estuary there are several mangrove plants dominated by *Avicennia marina* and *Avicenia alba mangroves.*

Tambak Oso River is 34 km long, along the flow there are settlements and various industries. The research location area has dense settlements, mangrove plants that are dense with residential areas. Mangroves along the Tambak Oso River are very diverse, none dominate. The color of the water in the Tambak Oso River is murky brown and the flow is calm but sometimes the current is moderate depending on the time. The condition of the sediment used for the growth and development of mangrove plants is black and smells better than the two research locations and there is a lot of plastic waste, cloth and others.

3.1. Physical Characteristics

Physical characteristics are used as supporting factors for the presence of heavy metals, namely sediment texture as a place for mangrove plants to grow and develop. Soil texture is the ratio of dust, clay and sand fractions.

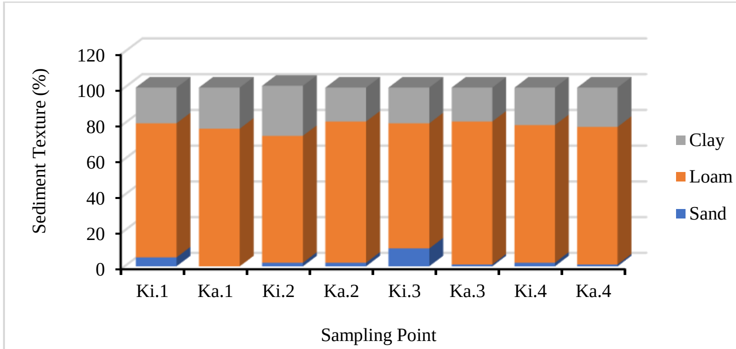

Figure 2. Composition of sediment texture in the Jagir River

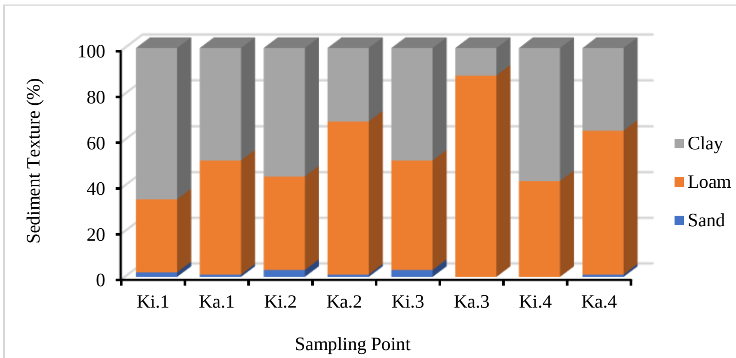

Figure 3. Composition of sediment texture in the Kebon Agung River

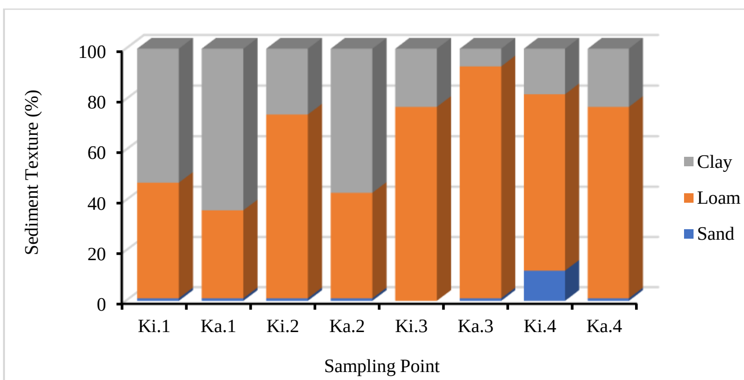

Figure 4. Composition of sediment texture in the Tambak Oso River. (Description: Ki1 = point 1 on the left and so on, Ka1 = point 1 on the right and so on)

The composition of sediment texture in the three research locations is dominated by dust and clay. The soil texture in the Jagir River is a class of dusty clay, in the Kebon Agung River is a class of dusty clay and in the Tambak Oso River is a class of dusty clay. Soil dominated by fine fractions causes the formation of many micropores so that the holding or absorption capacity is very strong. Based on the research results of Lestari & Rahadian (2017) Mangrove soil is characterized by alluvial hydromorph or marine clay formed from sediment. The sediment is formed in calm water and is soil that has not fully developed. The sediment contains many solid particles carried by water from the river flow to the sea which moves slowly. Soil aggregation that is easily decomposed or dissolved by water causes the soil to become muddy (Lestari & Rahadian, 2017). The existence of dust and clay textures is caused by the roots of mangrove vegetation binding dust and clay particles so that these particles will settle and form mud (Aini *et al.,* 2016).

3.2. Chemical Characteristics

Most of the pH in the three research locations are classified as neutral. According to Setyawan *et al*. (2002) in (Akhrianti *et al*., 2018) the pH of mangrove soil is usually slightly acidic to neutral due to the activity of sulfur-reducing bacteria and sedimentation of acidic clay soil, the activity of sulfur-reducing bacteria is characterized by dark, acidic and foul-smelling soil. The pH quality standard in the mangrove ecosystem based on the Decree of the Minister of Environment No. 51 of 2004 is 7-8.5 while mangrove plants can grow optimally at a pH range of 6-7.

The salinity value in the three rivers is classified as very high because it exceeds 4 mS/cm. This is because the mangrove sediment at the research location is inundated by seawater during high tide, the condition of point 4 at the Kebon Agung River location looks very high because it is directly adjacent to seawater. While at point 1, the average in each river is the lowest position because river 1 is an outer point far from seawater so that there is a mixture of seawater and fresh water which greatly affects the salinity value. Salinity affects mangrove growth, mangrove plants can live and grow in brackish water environments with varying salinity. (Basyuni *et al.,* 2019) said that generally mangrove plants are tolerant to high salinity levels, but tolerance between mangrove species to salinity varies.

Table 1. Chemical characteristics

No	River Name	pH	Salinity (mS cm^{-1})
1.	Jagir	7.33	12.56
2.	Kebon Agung	7.42	11.90
3.	Tambak Oso	7.23	13.05

3.3. Concentration of Heavy Metal Cu in Sediment, Roots and Leaves

The results of the study of heavy metals in sediments of the three highest rivers are the Tambak Oso River. This is because the Tambak Oso River is close to settlements that mostly use materials that produce Cu (copper) waste such as detergents, floor soaps and so on, the flow of the Tambak Oso River also comes from industrial areas. Azzahra (2015) said that along the flow of the Tambak Oso River there is a lot of garbage, from industrial and household waste and waste. The content of mangrove sediment in the three rivers is still below the standard quality but almost exceeds the standard quality limit. According to the United States Environmental Protection Agency (US-EPA) in 2004 the standard quality of heavy metals Cu sediment was 49.98 mg/kg. The standard quality according to *the Wisconsin Department of Natural Resources* (Doyle *et al.,* 2003) Cu content in the three rivers is included in the level of pollution two (moderate) with a quality standard below 91 mg/kg. Heavy metals that enter the water will be deposited in the sediment so that the sediment value tends to be high compared to the roots and leaves of mangroves. (Purwiyanto, 2013) stated that copper (Cu) heavy metal is a heavy metal that is easy to settle in sediment, so that the accumulation of Cu metal in the sediment will be higher. The heavy metal content in the roots is higher than in the leaves. Cu heavy metals that settle in various parts of the mangrove plant such as roots, leaves and stems, the ability of mangroves at the research location is able to act as phytostabilization and phytoextraction. Where, phytostabilization of Cu heavy metals settles in the roots but can prevent it from entering other parts of the plant, while phytoextraction is absorbing Cu heavy metals from the roots then translocating them to other parts of the plant such as roots, leaves and stems and then processed so as not to poison the body of the plant. This is reinforced by Utami *et al.* (2018) that mangrove plants can overcome hazardous materials through dilution by storing a lot of water to dilute the concentration of heavy metals in their bodies so that heavy metal toxicity can be reduced.

Table 2. Average concentration of heavy metal Cu

No	Cu Metal	Unit	River		
			1 (Jagir)	2 (Kebon Agung)	3 (Tambak Oso)
1.	Sediment	mg/kg	47.30	38.53	50.50
2.	Root	mg/kg	14.60	12.70	14.20
3.	Leaf	mg/kg	8.00	15.80	10.40

Description: 1 (Jagir River, 2 (Kebon Agung River), and 3 (Tambak Oso River)

3.4. Bioconcentration Factor (BCF) and Translocation Factor (TF)

The three research locations are included in the low accumulation level. Excluders are plants that can prevent heavy metals from entering the plant but the heavy metal content in the root area is still high. (Handayani *et al.,* 2018) explained that if the bioconcentration factor <1 then it is still in a safe condition, meaning that the plant does not accumulate much heavy metals in the soil. (Burges *et al.*, 2018) added that excluder plants selectively do not take up metals and instead can secure metals in the plant root area (*rhizosphere*) through deposition. Excluder plants limit the entry of metals into the roots and their translocation to the upper parts of the plant such as stems and leaves. The types of phytoremediation are phytoextraction, rhizofiltration, phytodegradation, phytostablization, and phytovolatilization.

Table 3. Bioconcentration Factors

No	River Name	BCF Root	BCF Leaf	BCF Plant	Class
1.	Jagir	0.31	0.17	0.48	Low
2.	Kebon Agung	0.33	0.42	0.74	Low
3.	Tambak Oso	0.28	0.21	0.49	Low

Table 4. Translocation Factors

No	River Name	TF Value	Class
1.	Jagir	0.57	Phytostabilization
2.	Kebon Agung	1.36	Phytoextraction
3.	Tambak Oso	0.75	Phytostabilization

Phytostabilization class means that mangrove plants in the research locations of Jagir and Tambak Oso Rivers have the ability to prevent the entry of metals into other parts of the plant through the roots by producing root exudates that can stabilize, demobilize, and bind metals, thereby reducing the availability of these metals. According to (Sabir *et al.*, 2014), phytostabilization plants accumulate heavy metals in the root area and limit the entry of heavy metals into other parts of the plant. Heavy metals cannot move in the rhizosphere so that they are not bioavailable and less toxic to plants, animals and humans. Plants are able to retain metals in the root area by limiting the translocation process to the upper part of the plant. Phytoextraction class means that heavy metals are absorbed by plant roots and translocated to the upper part of the plant for reprocessing. The TF value in the Kebon Agung River is phytoextraction because the heavy metal content of Cu at points 2, 3 and 4 in the leaves is higher than in the roots because the leaves not only absorb heavy metals through the roots but also from the air. This is reinforced by Nursagita & Hermin (2021), mangrove leaves have the ability to absorb water and pollutants including heavy metals from stomata and cuticles. The difference in TF and BCF values is influenced by plant factors such as differences in mangrove types, mangrove age and mangrove density. In addition, pollutant sources also affect the difference in these values. This is in line with the findings of Rachmawati *et al.*(2018) that differences in BCF values can occur due to differences in species so that their absorption is also different, TF values are different because even though in the same waters but if the types of mangroves are different then the TF value will be different.

3.5. Statistical Test

The pH *t*-test between Jagir River and Kebon Agung River obtained a t-count of 1.425, meaning there is no significant difference. This is because the conditions of the factors that affect pH are mostly the same. The pH *t*-test between Jagir River and Tambak Oso River obtained a t-count of 2.561, meaning there is a significant difference. This is because the number of macrofauna in Jagir River and Tambak Oso River is different, in addition, the sediment conditions of Tambak Oso River are not good for macrofauna life, thus affecting the pH value of the sediment. The pH *t*-test between Kebon Agung River and Tambak Oso River obtained a *t*-count of 3.618, meaning there is a significant difference. Kebon Agung River and Tambak Oso River are small in size but have different sources of pollution. Tambak Oso River is closer to settlements and home industries so that environmental pollution is higher. This is what causes the difference in pH between Kebon Agung River and Tambak Oso River. The *t*-table obtained is 2.145.

The *t*-test of salinity between Jagir River and Kebon Agung River obtained a *t*-count of 0.479, meaning there is no significant difference. The *t*-test of salinity between Jagir River and Tambak Oso River obtained a *t*-count of 0.392, meaning there is no significant difference. The *t*-test of salinity between Kebon Agung River and Tambak Oso River obtained a *t*-count of 0.719, meaning there is no significant difference. This is because the conditions that affect salinity, including the ebb and flow of sea water, are the same. The *t*-table obtained is 2.201.

The *t*-test of heavy metals between the Jagir River and the Kebon Agung River obtained a *t*-count of 3.689, meaning there is a significant difference. This is because the source of Cu from the Jagir River is greater than the Kebon Agung River because the Jagir River is closer to the highway and settlements and the size of the river is larger. The *t*-test of heavy metals between the Jagir River and the Tambak Oso River obtained a *t*-count of 1.109, meaning there is no significant difference. The source of Cu between the Jagir River and the Tambak Oso River is on average the same, originating from residential areas, because the area is densely populated. The *t*-test of heavy metals between the Kebon Agung River and the Tambak Oso River obtained a *t*-count of 4.901, meaning there is a significant difference. This is because the size of the Tambak Oso River is small but the source of pollution is greater than the Kebon Agung River. The *t*-table obtained was 2.447.

3.6. Correlation Between Parameters and Heavy Metal Cu

The relationship between dust fraction and heavy metal Cu is a real relationship, where the correlation value of 0.642 means high closeness. The more dust fractions, the more heavy metals. This is because the concentration of heavy metals is influenced by grain size, the finer the soil fraction, the stronger the binding heavy metals. The fine sediment fraction has a large surface area with a more stable ion density to bind metals than the larger sediment fraction (Sahara, 2009).

The relationship between pH and heavy metal Cu has a real relationship, where the correlation value of –0.771 means high closeness. (Mahasri *et al*., 2014) said that low pH conditions (<7) will increase the solubility of heavy metals and the toxicity of heavy metals. If the pH is low, the concentration of H^+ is large so that copper ions in the form of Cu^{2+} will compete with H^+ and there is repulsion between H^+ and Cu^{2+} ions to bind to the active site on the adsorbent. This causes the heavy metal copper to not be absorbed optimally by the roots of mangrove trees and settles in the sediment, so the heavy metal content is getting higher.

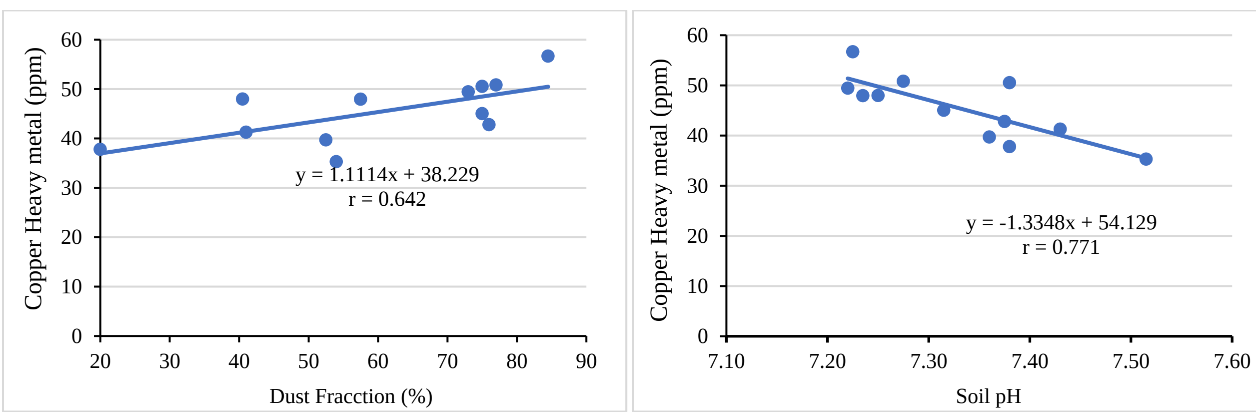

Figure 5. Correlation graph of heavy metal Cu with dust fraction (left), and heavy metal Cu with pH

4. CONCLUSIONS

The highest concentration of sediment heavy metals is in the Tambak Oso River at 50.50 mg/kg below the quality standard. The pH value between 7.22–7.51 is neutral. The salinity value between 8.44–13.85 is very high salinity. Mangroves in the Jagir River, Kebon Agung River and Tambak Oso River have low heavy metal accumulation capabilities, meaning that plants do not accumulate much heavy metals in the soil and are capable of phytostabilization and phytoextraction. Based on this result, it is recommende that the local government will provide socialization to the surrounding community to always maintain cleanliness so that the river is not polluted. Further research needs to be done to add parameters that support the presence of heavy metals such as macrofauna, organic materials and observations of water samples or environmental conditions and other heavy metals so that the results are maximized so that knowledge is conveyed completely to the reader.

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