The Ability of Mangrove Plant on Lead Phytoremediation at Wonorejo Estuary, Surabaya, Indonesia

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ABSTRACT

Lead (Pb) is a heavy metal often discovered to be polluting the water areas. One of the efforts made to overcome the heavy metal pollution in estuaries was phytoremediation technique using mangroves. The Wonorejo River was one of the rivers that received industrial waste loads. There were various types of mangrove plant species at the estuary of the Wonorejo River. The location of this research was divided into 3 monitoring stations (A, B, C). Station A was directly adjacent to the estuary, as location C was farther away and very close to the sea. However, station B was located between location A and B. This study aimed to determine the ability of mangrove in remediating and illustrating the distribution of Pb, at the Wonorejo River estuary. Moreover, it also aimed to determine the values of Bioconcentration (BCF) and Translocation (TF) Factors in the ability of *Avicennia alba, Avicennia marina, Sonneratia caseolaris, Avicennia lanata,* and *Rhizophora stylosa* to accumulate Pb. The samples were the roots, stems, and leaves of mangroves, with the water and sediment at the Wonorejo estuary, as all solid materials were also extracted. The samples were analyzed for heavy metal concentration, using an atomic absorption spectrophotometer (AAS). The results showed that the highest average Pb concentration for waters and sediments was obtained at station C and A, with values of 0.069 mg/L and 4.22 mg/kg, respectively. It was further observed that the Pb concentration in the water was lower than in sediment, indicating that the metal was accumulated in the sediments. On the basis of the BCF value, the most effective mangrove involved in the accumulation of Pb was *A. alba*. The highest values of TF for both root to stems and to leaves in the accumulation of Pb was also discovered in *A. lanata and A. alba* mangroves, respectively. On the basis of the Pb distribution mapping, the concentration of the metal was shown to increased, as the research location moved further away from the estuary. Conclusively, each type of mangrove had different ability to accumulate and translocate Pb in its body, with the potential of using those plants as phytoremediaton agents for the metal.

Keyword: BCF, Estuary, Lead, Mangrove, Phytoremediation, Sediment, TF, Water.

INTRODUCTION

Industrial waste has become a serious problem in the industrialization era (Setiawan, 2013). According to Yudo (2006), industrial waste is generally disposed of into rivers without treatment, which in turn causes a decrease in the quality of water bodies. Industrial waste is also known to be dangerous when it contains heavy metals (Nafie et al. 2019). Generally, heavy metals have dangerous properties to the environment and human health, due to the fact that they have toxic and carcinogenic characteristics (Alharbi et al., 2019), are non-biodegradable, and accumulate in the community (Sari, 2017).

The Wonorejo River is one of the water bodies in the city of Surabaya that receives industrial and household waste loads (Harnani nad Titah, 2017). The Wonorejo River empties into the east coast of Surabaya City, as the coastal zone has a high potential for heavy metal accumulation, due to being directly adjacent to land and

sea (Setiawan, 2013). The research conducted by Sakinah (2016), explained that the land-use change for settlements and industrial waste disposal in the Aveur river also affected the pollution in the waters of the Wonorejo estuary.

According to Munawar and Rina (2010), heavy metals such as mercury (Hg), lead (Pb), arsenic (As), cadmium (Cd), chromium (Cr), zinc (Zn), and nickel (Ni), were the inorganic materials often causing pollution in waters. When the industrial waste discharge containing heavy metals enters the water body, deposition occurs in the sediment (Febriana, 2017), as the elements that have settled within are bound and accumulated. The heavier the metals that are accumulated in the sediment, the higher and more toxic the concentration becomes.

The sediments in the Wonorejo River estuary are retained by the roots of mangrove plants. Mangrove plants have strong roots, as they have the abilities to hold sediments from entering the sea directly (Harnani and Titah, 2017). Mangroves have ecological functions, such as absorbing, transporting, and storing heavy metals within their environment of growth. The heavy metals entering the mangrove body are accumulated in the roots, stems, and leaves, due to the fact that they have translocation abilities (Setiawan, 2013). Each type of mangrove has the ability to accumulate different heavy metals (Harnani and Titah, 2017).

The research conducted by Febrina in 2017, showed that the highest concentration of Pb in the sediments of the Wonorejo River estuary was 6.42 mg/kg. This value was included in the category of heavy pollution, based on the USEPA. In addition, the research conducted by Khairuddin et al. (2018) showed that the average concentrations of Pb and Cd in the leaves of the *Sonneratia alba* and *Rhizophora apiculata* mangroves, were greater than those in the roots. This explained that

mangroves have a translocation function, where heavy metals absorbed are spread to other body tissues.

The study aimed to determine the Pb concentration in the Wonorejo River estuary area, and the water body before it. The second aim was to determine the Pb concentration in the sediments, roots, stems, and leaves of mangrove plants (*Avicennia alba, Avicennia marina, Rhizophora stylosa, Avicennia lanata,* and *Sonneratia caseolaris*), at the Estuary of the Wonorejo River area based on the remediating effectiveness and distribution description of Pb; the last purpose of this study was to determine the ability of mangroves in the coastal zone of the Wonorejo River estuary area.

MATERIAL AND METHOD

Sampling Location

The location of the study was at the Wonorejo Mangrove Ecoforest area, in Surabaya, Indonesia. On the basis on the consideration of mangrove thickness, the determination of sampling stations was observed visually, using the google earth application, with easy road access and safety to reach the study location. The research station was divided into 3 regions, namely Station A (7° 18'19.75" South Latitude and 112° 50'39.4" East Longitude), B (7° 18'20.19" South Latitude and 112° 50'40." East Longitude), and C (7° 18'21.90" South Latitude and 112° 50'40.56" East Longitude). The location of this research was also shown in Figure 1 below. The sampling of the water body before the Wonorejo coastal area was also conducted in three locations. The locations were near residential areas, middle of the Wonorejo River, and close to the estuary of the research location.
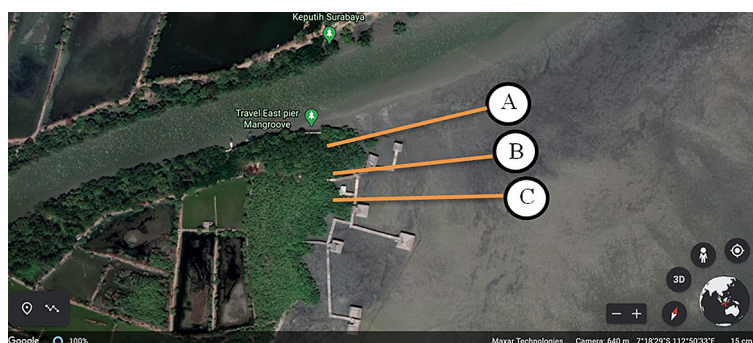

Figure 1. Location of this research

Sampling and Analysis Methods

Plotting was carried out to determine the sampling point, using the transect method. The selected tree samples had a diameter greater than 10 cm, which was related to the age and the ability of mangroves to accumulate heavy metals. According to Nurrahman et al. (2012), selected mangroves should have a diameter of 15–25 cm. This selection was based on the fact that the diameter was included in the tree category. The sampling point should also pay attention to the fact that the mangroves to be obtained should be exposed to the tides of the sea water. The coordinates of the sample points are shown in Table 1.

The sampling procedure in this study was based on Harnani and Titah 2017), which included collection, container, and storage. The samples obtained were further analyzed at the Sucofindo Surabaya Laboratory. The analysis of samples in the laboratory included the concentration of Pb. Composite sampling was also used for water and sediment samples. The samplings using the composite method were often carried out, in order to obtain accurate results (Munvika, 2018). This method was conducted by mixing several samples on a certain side into one. The water samples of approximately 500 ml were obtained, placed in a plastic bottle, and labeled for identity purposes, as the collection of sediment materials was also carried out, using a manual drill with a depth of 20–30 cm. Sediment sampling with a depth of ± 30 cm was assumed to be able to represent pollutants vertically (Rachmawati, 2018). In addition, the sediments obtained were positioned on the right and left sides of the selected mangroves. The distance between sediment sampling and mangrove stems was less than 30 cm. Moreover, sediment samples of about 500 gr were also obtained.

The mangrove plants having a trunk diameter of more than 10 cm had a strong root system. Strong roots support the ability of mangroves to absorb heavy metals effectively, as sampling of the mangrove root was carried out, using a manual drill and machete. Root sampling was referred to the research of Supriyantini et al., (2017), who stated that the mangrove root samples obtained, had a depth of 10–30 cm. The samples obtained weighed approximately 500 grams. The stem samples obtained were those of the mangrove plants, with leaves attached to them. The mangrove stems further obtained were in the range

of 500 gr. As regards the leaves, the samples obtained had green color, with no damages caused by pests or diseases. The size of the samples varied from small to large, which were also in the

Table 1. The coordinates of the sample points

No	Location	Code	Information	Coordinnate
			Station A	
1	Plot 1	SAP1 *A. alba*	Sample *A. alba* Stasiun A, Plot 1	SS 7°18'19.75"
				E 112°50'39,4"
2		SAP1 *S. caseolaris*	Sample *S. caseolaris* Stasiun A, Plot 1	SS 7°18'19.93"
				E 112°50'39.12"
3	Plot 2	SAP2 *A. alba*	Sample *A. alba* Stasiun A, Plot 2	SS 7°18'20.21"
				E 112°50'39.05"
4		SAP2 *S. caseolaris*	Sample *S. caseolaris* Stasiun A, Plot 2	SS 7°18'20.34"
				E 112°50'38.87"
5	Plot 3	SAP3 *A. alba*	Sample *A. alba* Stasiun A, Plot 3	SS 7°18'20.56"
				E 112°50'38.23"
6		SAP3 *S. caseolaris*	Sample *S. caseolaris* Stasiun A, Plot 3	SS 7°18'20.58"
				E 112°50'38.60"
			Station B	
7	Plot 1	SBP1 *A. alba*	Sample *A. alba* Stasiun B, Plot 1	SS 7°18'20.19"
				E 112°50'40."
8		SBP1 *S. caseolaris*	Sample *S. caseolaris* Stasiun B, Plot 1	SS 7°18'20.34"
				E 112°50'40.27"
9	Plot 2	SBP2 *A. alba*	Sample *A. alba* Stasiun B, Plot 2	SS 7°18'20.39"
				E 112°50'39.92"
10		SBP3 *A. marina*	Sample *A. marina* Stasiun B, Plot 3	SS 7°18'20.73"
				E 112°50'39.95"
11	Plot 3	SBP3 *A. alba*	Sample *A. alba* Stasiun B, Plot 3	SS 7°18'20.53"
				E 112°50'39.64"
12		SBP3 *S. caseolaris*	Sample *S. caseolaris* Stasiun B, Plot 3	SS 7°18'20,70"
				E 112°50'39.50"
13		SBP3 *A. lanata*	Sample *A. lanata* Stasiun B, Plot 3	SS 7°18'20.81"
				E 112°50'39.38"
			Station C C	
14	Plot 1	SCP1 *A. marina*	Sample *A. marina* Stasiun C, Plot 1	SS 7°18'21.90"
				E 112°50'40.56"
15		SCP1 *R. stylosa*	Sample *R. stylosa* Stasiun C, Plot 1	SS 7°18'21.83"
				E 112°50'40.32"
16	Plot 2	SCP2 *A. marina*	Sample *A. marina* Stasiun C, Plot 2	SS 7°18'22.05"
				E 112°50'40.19"
17	Plot 3	SCP3 *A. marina*	Sample *A. marina* Stasiun C, Plot 3	SS 7°18'22.12"
				E 112°50'39,89"
18		SCP3 *A. alba*	Sample *A. alba* Stasiun C, Plot 3	SS 7°18'21.98"
				E 112°50'39.52"
			Environmental	
19	River near residential areas	Near Residential	River near residential areas	SS 7°18'11.90"
				E 112°49'7.30"
20	Middle	Middle	The middle of the Wonorejo River	SS 7°18'28.8"
				E 112°49'44.80"
21	Estuary	Estuary	Near the estuary of the Wonorejo river	SS 7°18'21.30"
				E 112°50'36.60"

form of young and old leaves. The leaf samples obtained were about 100–200 gr.

All solid samples such as sediments and parts of mangrove (root, stems, leaves), were extracted based on method of USEPA SW-846–3050 B &. Afterwards, all samples were analyzed, using AAS to determine the Pb concentration.

Determination of BCF (Bioconcentration Factor) and TF (Translocation Factor)

The results of the analysis of the concentrations contained in the environment and mangrove plants were used to calculate the values of BCF (Bioconcentration Factor) and TF (Translocation Factor). The BCF and TF were used to determine the accumulation of heavy metals and translocation abilities, in parts of the mangroves.

Mapping of the Pb Distribution

The data from the analysis of heavy metals in mangrove plants, water, and surrounding sediments, were used as reference for making a distribution map of elements with the Surfer program. In addition, the method involved creating heavy metal distribution maps was the gridding technique. The grid was a network of rectangular dots, which were regularly distributed throughout the mapping area. Gridding is the process of using original data points (observational data) contained in the XYZ data file, to form additional parameters on a grid distributed out regularly. The results of the mapping obtained a visual description of the heavy metals distribution, with the influence of the mangrove forests presence in the Wonorejo coast.

RESULTS AND DISCUSSION

The concentration of Pb in the Waters of the Wonorejo River Estuary

The results of the Pb analysis in the waters of the Wonorejo River Estuary for the sampling point before entering the mangrove forest area, are described in Figure 2. The results of the metal analysis in the mangrove forest waters of the Wonorejo River Estuary are presented in Figure 3.

On the basis of Figure 2, the concentrations of Pb in the river near residential areas, middle of the water, and close to the estuary, were 0.042 mg/L, 0.04 mg/L, and 0.026 mg/L, respectively. This result further showed that the highest and lowest concentrations of the metal were discovered near residential areas and the estuary of the river, respectively. The high concentration of Pb near the residential areas was due to the fact that there was a source of the metal pollutants. The concentration of heavy metals in river was influenced by the amount of waste input into the water body. The concentration of heavy metals in river increased along with the discharged wastewater volume (Febriana, 2017). The trend lines further showed that the concentration of Pb from upstream to downstream declined, with this condition occurring due to the fact that Pb was deposited along the water body, until it reached the estuary of the Wonorejo River. Pb has a high density and low solubility, making it easier to be deposited in the bottom of the waters. The low concentration of heavy metals in waters was affected by some factors, specifically currents and waves, which cause the distribution of the elements on the surface of water in all directions (Febriana, 2017).
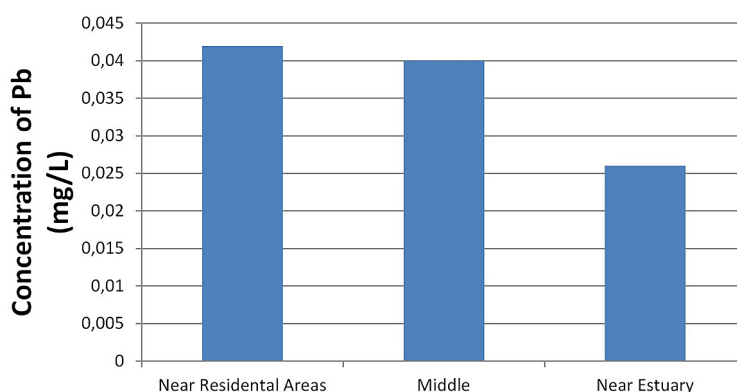

Figure 2. Concentration of Pb along the Wonorejo River

According to Palar (2004), the metals in waters occurred in ion form, both as single and in pairs. Pb in water bodies was discovered in the form of divalent (Pb^{2+}) or tetravalent (Pb^{4+}) ions. Moreover, according to Eastoe (1992), Pb in water were in complex types with organic groups, in a bid to create a colloidal solution or in the form of Pb^{2+}, $PbSO_4$, $PbCl_2$, and $Pb(OH)$ ions. However, Pb in the form of $PbCO_3$ was discovered to be present in the sea. Pb in waters was also discovered in dissolved and suspended forms, where the solubility of the metal was low enough for the concentration to be relatively small (Eckenfelder, 1989). The presence of metals in river estuaries was further affected by the adsorption process, which occurred in the water column. Adsorption is a process in which a fluid/liquid or gas is attached to a particle. The heavy metals in the dissolved phase were subsequently adsorbed by suspended sediments, as the materials were deposited to the bottom (Maslukhan, 2013). The concentration of Pb at the study location near the Wonorejo Mangrove Ecoforests, is described in Figure 3.

The average concentrations of Pb at each station (A, B and C) in the estuary waters of the Wonorejo River were 0.87 mg/L, 2.12 mg/L, and 2.94 mg/L, respectively. The concentration of Pb varied from < 0.1 mg/L to 8.7 mg/L. The highest and lowest average concentrations of Pb were at station C and A, respectively. This showed that the concentration of Pb from Station A to C had increased. This result was the same as in the study conducted by Febriana (2017). The increase of the Pb concentration was caused by the location of Station A, which was close to the estuary of the river. This condition caused the fluctuation of turbulence and dilution between river and sea water, with metal concentrations. The increase of the Pb concentration at Station B and C was also due to the elevation of salinity at both locations. According to Mance (1987), high and low salinity caused increased and decreased heavy metal contents in the waters, and vice versa. Besides, the stations did not experience seawater turbulence, and were only influenced by tides, as the concentration of the metal seemed to be constant. The high concentration of Pb at Station C was due to the presence of the heavy metal from sea pollution. This occurred due to the eastern coastal zone of the Surabaya City, which was often passed by vessels that contributed to the Pb pollution at sea.

On the basis of the Spatial Planning and Territory (RTRW) of the city, the Eastern Coastal area of Surabaya (Pamurbaya) is a protected/conservation area, as the water quality standards used in this research were due to the Decree of the Minister of Environment, Number 51 of 2004 concerning Marine Biota of 0.008 mg/L for Pb. The comparison of the concentration of Pb with quality standards are shown in Figure 4.

On the basis of Figure 4, it was observed that the concentration of Pb in the waters of the Wonorejo River Estuary had exceeded the established quality standards, both at the river location before the estuary area and the mangrove forest waters. According to Setiabudi (2005), when the heavy metal element in the environment exceeded the specified value of quality standard, it was
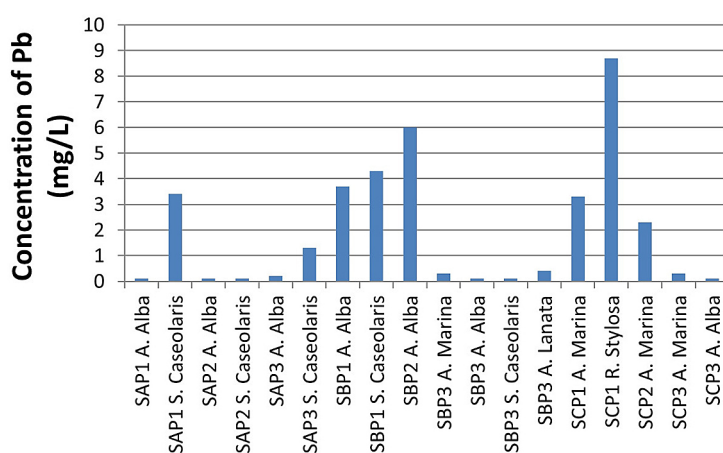

Figure 3. Concentration of Pb in the waters of the mangrove forest

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
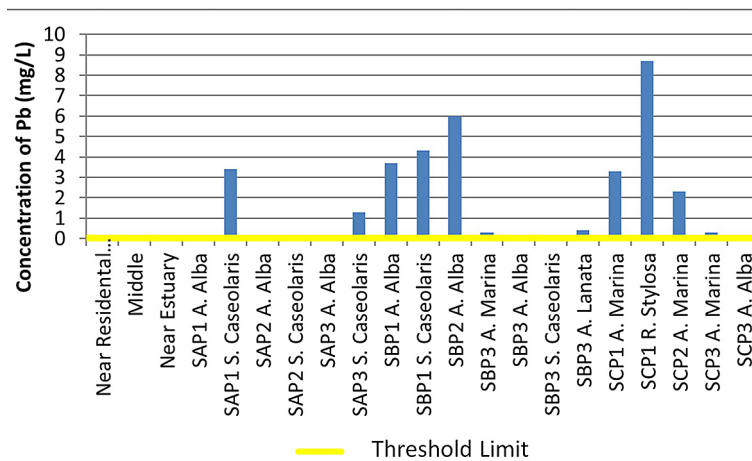SCP1–3 = the sampling points at station C, plot 1 to plot 3

Figure 4. The comparison of the concentration of Pb with quality standards
SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

considered toxic (poisoned). Pb is a highly toxic metal, the presence of which in small amounts is greatly poisonous. The exposure to Pb is dangerous to humans, and more deadly when it infiltrates into the ecosystem excessively. This is due to the fact that it damages the environment, and is highly dangerous for the surrounding biota (Rachmawati, 2018).

The concentration of Pb in the Sediments of the Wonorejo River Estuary

According to Alharbi et al. (2019), the sediments in water areas became the best indicators for tracking metal accumulation. Sedimentation of heavy metals in waters occurred due to the presence of carbonate, hydroxyl, and chloride anions (Hidayat, 2011). The sediments at the Wonorejo River Estuary were mixtures of mud and sand. The sediments in the form of mud are attached to more heavy metals, due to the presence of large number of organic substances. Heavy metals had the characteristics of being easily bonded by organic materials. A high percentage of the mud content tended to contain more heavy metals, as increased concentrations of the elements were often related to the sediments having small grain sizes. Therefore, it binds metals in sediments effectively (Purbonegoro et al., 2014). The results of the metal concentration analysis in the sediment at the Wonorejo River Estuary are described in Figure 5.

The average concentrations of Pb in the sediments at Station of A, B, and C were 2.87 mg/kg,

0.47 mg/kg, and 4.22 mg/kg, respectively. In addition, the highest concentration of Pb in the sediments at the Wonorejo River Estuary occurred at Station C. This was in accordance with the research conducted by Febriana (2017), which explained that the concentration of Pb in 2017 was 4.96 mg/kg to 6.42 mg/kg. On the basis of the analysis, it was discovered that there was an increase in Pb, from Station A to C, with a decrease observed at location B. The range of concentration of Pb in the sediments at the estuary of the Wonorejo River was below 0.1 to 10.1 mg/kg. Figure 6 further showed he contours of the Pb distribution in the sediments at the estuary of the Wonorejo River.

The highest concentration of Pb was within Station C, as a dark blue color was observed, with location A dominated by beige and light green colors. In addition, Station B was observed to be dominated by light green. These results indicated that the concentration of Pb at location B was the lowest among the three stations. The Pb distribution in the three stations showed that the farther the location from the estuary of the river and directly adjacent to the sea, the higher the concentration. The highest concentration of Pb in the sediment was at Station C, which was the farthest from the estuary of the river, while the lowest was at location B.

The high concentration of Pb at Station A occurred due to the nature of the heavy metal which had a high density, as it is easily accumulated in the sediment. Station A at an estuary area was a meeting of fresh and sea water, with high salinity
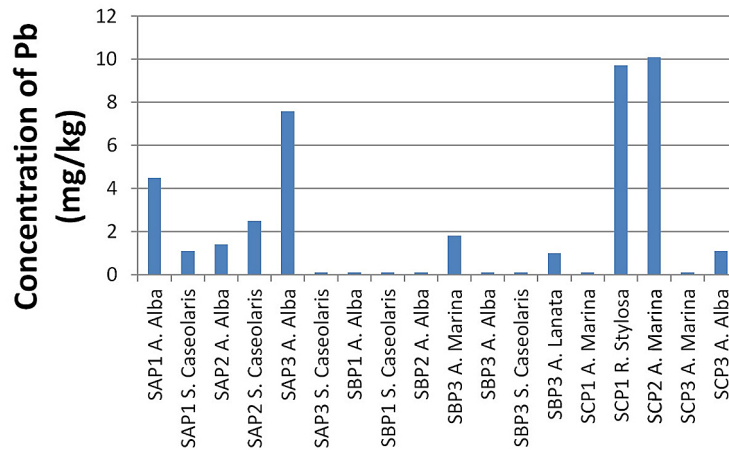
Figure 5. Concentration of Pb in the sediment of the mangrove forest

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
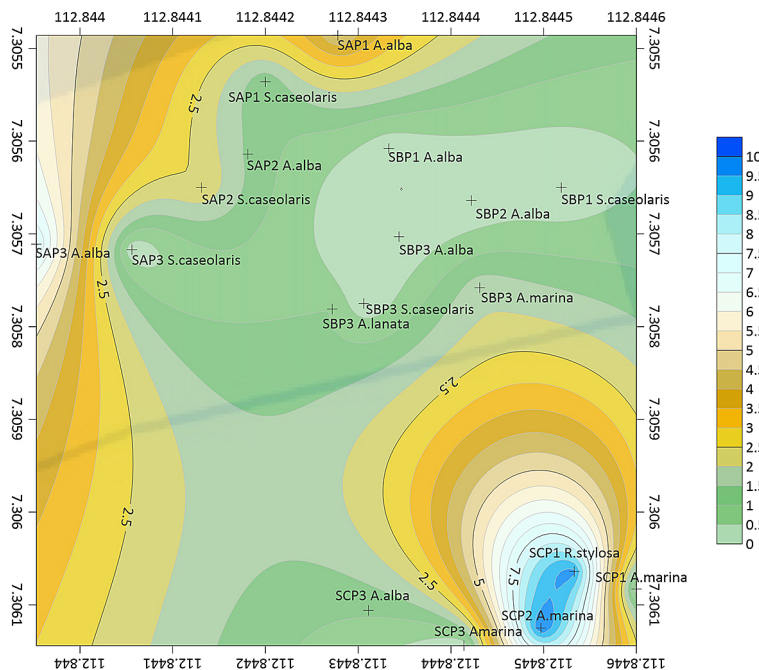SCP1–3 = the sampling points at station C, plot 1 to plot 3

Figure 6. Contours of the distribution of Pb in the sediments at the estuary of the Wonorejo River

and great ionic strength. It caused the destabilization of suspended solid particles, in order to form aggregations that occurred in sediments, due to gravity (Maslukah, 2013). Station A had a rapid mixing of fresh and sea water, with the mixture of the two different water masses causing a change in the salinity and pH, which subsequently triggers the desorption-adsorption process in estuary waters (Najamuddin et al., 2016). The rise in ionic strength also caused the force of attraction between the particles to be stronger, while triggering the accumulation of floc or clots, which

were then sedimented (Febriana, 2017). One of the factors that caused high concentration of Pb at Station C compared to locations A and B, was the high salinity. According to Mance (1987), high and low salinity caused increased and decreased heavy metal contents in the waters, and vice versa. In addition, it was suspected that there had been pollution in sea water by Pb, as it was carried by the waves towards the area of Station C, where it was deposited.

On the basis of the results of this study, the concentration of heavy metal in the sediment was

greater than that in the water. This was due to the heavy metals that had experienced deposition for accumulation in the sediment (Palar, 2004). According to Hutagalung (1991), heavy metal deposition occurred due to the element density being higher than the water population. Therefore, the concentrations of heavy metals were more regularly discovered in sediments, than in water. The density of the metal causes the floating of the element in the water to fall, which is then deposited in the sediment. The stable nature of sediments and the tendency to trap elements also caused heavy metal bonds to be higher.

The sediment quality standard used in this study referred to the Environmental Protection Agency (EPA) Sediment Quality, the Australian and New Zealand Environment and Conservation Council (ANZECC/ARMCANZ Guidelines), and the Canadian Council of Minister of Environment (CCME). Figure 7 further showed the comparison of the concentration of Pb in the sediments at the estuary of the Wonorejo River, with the quality standards.

The quality standard of Pb sediment, based on the EPA Sediment Quality (AS), was divided into three categories, including light, medium, and heavy pollutions. On the basis of Figure 7, the concentration of Pb in the sediments of all stations had been contaminated with the metal. The concentration of Pb in the sediments of the study area ranged from 0.1 to 10.1 mg/L. Another

quality standard based on the Australian and New Zeland Environment and Conservation Council (ANZECC/ARMCANZ Guidelines) stated that sediments were likely to be polluted when the concentration of Pb reaches 220 mg/L. However, according to the Canadian Council of Minister of Environment (CCME), sediments were likely to be polluted, when the concentration of Pb reaches 112 mg/L. On the basis of the reference from Australia and Canada, the sediments at the estuary of the Wonorejo River did not exceed the quality standard.

Mangrove ability to remediate Pb by determining BCF and TF

Mangrove plants were used as phytoremediation agents, due to being able to accumulate heavy metals around them naturally, which was also referred to as biosorption. Mangroves also filter, catch, and bind pollution within the environment, in the form of excess sediment, garbage, and other household wastes, which contributed greatly to improving water quality (Utami et al., 2018). The heavy metals in the aquatic environment were also accumulated in sediment, which enables them to increase with time (Amin, 2019). The heavy metals that were accumulated in the sediments at the estuary of the Wonorejo River were absorbed by the roots of the mangroves. The basic aspects of accumulation in plants included
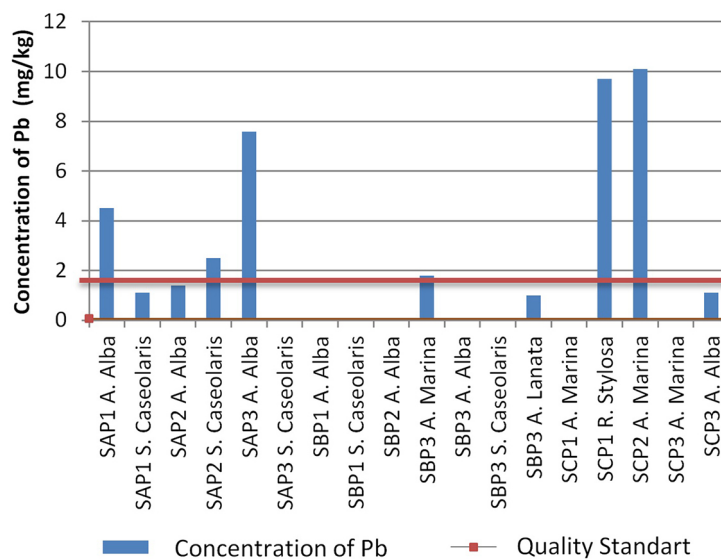

Figure 7. The comparison of the concentration of Pb in sediment with quality standards
SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

mobilization of heavy metals, translocation in plants, absorption of ions by plant roots and in tissues, as well as metal tolerance. According to Caroline et al. (2015), the absorption of heavy metals by plants had a principle that the higher the concentration of strong elements in the plant medium, the greater the metal being absorbed. The difference in concentration between the two types of media involved in plant tissues caused mass transfer by diffusion and osmosis. In this process, the mass of the substance with a higher concentration moved to a lower region, within the plant media.

According to Hardiani (2008), plants absorbed water-soluble metals through their roots. Plants were discovered to adjust the pH level, and developed a chelating substance in their roots. With a variety of amino acid compositions, they formed phytochelic compounds, which assisted the absorption of metals (Mansawan, 2016). The chelating agent that was formed bound the metal and transported it to the root cell, through active transport. Due to the exposure of plants to the metals, phytochelatins (PCs) were formed. Phytochelatin is a small-sized glutathione-derived enzymatic peptide synthesis, which binds metals. It is also the main part of the metal detoxification system in plants, with the general structure of (-glutamyl-cysteinyl)n-glycine, (n=2–11) (Ali et al., 2013). Moreover, Ali et al. (2013) described that phytochelatins (PCs) and metallotionin (MTs) were the significant peptides/proteins involved in the accumulation and tolerance of metals. Phytochelatins (PCs) and metallotionins (MTs) of plants were rich in cysteine sulfhydryl groups, which bind and absorb heavy metal ions. These metalotionins (MTs) have low molecular weight (4–10 kDa), and are rich in cysteine, which is protein that binds metals through the thiol group of its residues. Afterwards, the phytochelatins formed a reductase molecule in the membrane of the roots, in order to increase absorption. The function of this reductase was to decrease metals, which were then transported through special channels in the root membrane (Awad et al., 1994). Hermawati (2005) also reported that besides being affected by chelating compounds, the absorption of heavy metals by the roots was also influenced by the pH factor. The pH level has a crucial role in phytoremediation, because it affects the solubility of nutrients that allow plants to grow. Moreover, high pH inhibits nutrient solubility and plant growth. According to Tangio (2013), each type of metal

had a different pH to dissolve optimally, with Pb dissolving at a pH level of 5. Once the metal is brought into the cell of the roots, the plants carry out a detoxification mechanism, with the element being converted into a less toxic form, through chemical reactions or complex formation with secondary metabolites produced by plants, as they enter the tissues without poisoning the plants (Widyati, 2011). Plants did not only indicate the accumulation of heavy metals in roots, they also showed their accumulation in leaves and stems. Accumulation of heavy metals that have reached the leaves passes through plasmalemma, cytoplasm, and vacuoles, where the elements are unable to be associated with the physiological processes of the plant cells (Haryati, 2012).

The studies in the field of biochemistry and molecular genetics had also disclosed that hyperaccumulator plants have a large difference in the ability to accumulate and tolerate metals, compared to others, due to the differences in series of biochemical physiological processes and gene expressions, which had contributed greatly to the absorption, accumulation, and tolerance of plants towards the elements (Salt, 2006). The initial process in the accumulation of heavy metals was the rhizosphere interaction in the root zone. In this stage, the processing of elements in the plant medium, from unabsorbed to absorbable forms by involving some of the exudates produced by the roots, was carried out. Moreover, the hyperaccumulator plants accelerated the dissolution of metals in the rhizosphere, with the roots of these particular plants having a high selectivity to specific elements. The absorption of metal by roots was further determined by permeability, transpiration, pressure, and the presence of an enhanced uptake system, which is only being possessed by hyperaccumulator plants (Hidayati, 2013). The concentration of Pb in the mangrove roots was shown in Figure 8.

The species of mangrove grown at Station A included *A. alba and S. caseolaris*. The Pb concentration in the mangrove roots at Station A showed a range of 0.1 mg/kg to 3.4 m/kg. The average concentrations of Pb in the mangrove roots at Station A for *A. alba and S. caseolaris* were 0.13 mg/kg and 1.6 mg/kg, respectively. In addition, the species of mangroves discovered at Station B were *A. alba, S. caseolaris, A. marina, and A. lanata.* The average concentrations of Pb in roots of *A. alba*, *S. caseolaris, A. marina,* and *A. lanata* were 3.26 mg/kg, 2.2 mg/kg, 0.3 mg/kg,
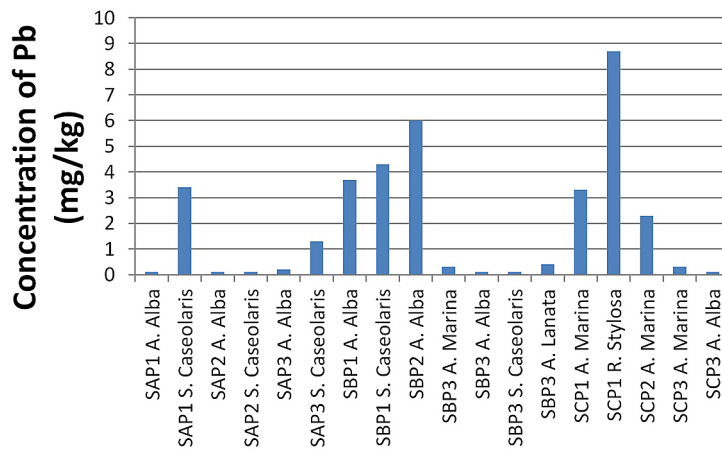
Figure 8. The concentration of Pb in the mangrove roots
SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

and 0.4 mg/kg, respectively. The range of Pb concentration in the mangrove roots at Station B was also between 0.1 mg/kg to 6 mg/kg. Moreover, the Pb concentration in the mangrove roots at Station C showed a range of 0.1 mg/kg to 8.7 mg/kg, as it reached 1.97 mg/kg, 8.7 mg/kg, and 0.1 mg/kg for *A. marina*, *R. stylosa,* and *A. alba*, respectively.

The highest and lowest average Pb concentrations in the mangrove roots at Station A, B, and C were observed in the species of *S. caseolaris & A. alba, A. alba & A. marina*, with *R. stylosa & A. alba,* respectively.

The concentration of Pb in *A. alba* mangroves at Station A and C were almost similar. However, *A. alba* at Station B was different from the other locations. The Pb concentration in the roots of *A. alba* at Station B was also higher than those at both A and C. This condition was caused by mangrove age and environmental factors.

Mangroves translocate heavy metals from the roots to other parts of their body. The ability of each mangrove to translocate heavy metals was different, as regards their physiological processes (Rachmawati, 2018). Hyperaccumulators undergo a metal translocation process from the roots to the canopy, which was confirmed to have a level far exceeding normal plants. This translocation was conducted by two primary processes, including the movement of ions and the volume of flux in the xylem, which is mediated by root pressure and transpiration (Gabbrielli, 1991).

The concentration of Pb in the mangrove stems was further shown in Figure 9. The average Pb concentration in the stems of *A. alba* and

S. caseolaris at Station A were 0.3 mg/kg and 0.18 mg/kg, respectively. The average Pb concentration in the stems of *A. alba* was greater, compared to the roots. This also indicated that there was a high mobility of Pb from the roots to the stems. However, this condition did not occur in the *S. caseolaris* mangrove, as the average Pb concentration in roots was greater than in stems.

The average Pb concentration in the stems of *A. alba, S. caseolaris, A. marina,* and *A. lanata* at Station B were 0.1 mg/kg, 1.9 mg/kg, 0.1 mg/kg, and 4.1 mg/kg, respectively. The highest and lowest average Pb concentrations were also discovered in the stems of *A. lanata with A. alba & A. marina*, respectively.

The average Pb concentration in stems of A. marina, R. stylosa, and A. alba at Station C were 3.2 mg/kg, 3 mg/kg, and 0.1 mg/kg, respectively. The highest and lowest average Pb concentrations were observed in the stems of *A. marina and A. alba*, respectively. The dominant high concentration of Pb was further observed in roots and stems. According to Heriyanto et al., (2011), greater Pb accumulation occurred in the roots and stems.

Each plant species had different mechanisms for absorbing and translating Pb. The Pb concentration in plant tissue was often around 10 mg/g, with hyperaccumulators accumulating the element in the canopy at about 1000 mg/g. Hyperaccumulators also had difficulties in accumulating more than 0.1% Pb in their crowns. Pb was tightly bound to minerals and soil organic matter, making it difficult for the plants to absorb the element through their roots. Once Pb was absorbed by the
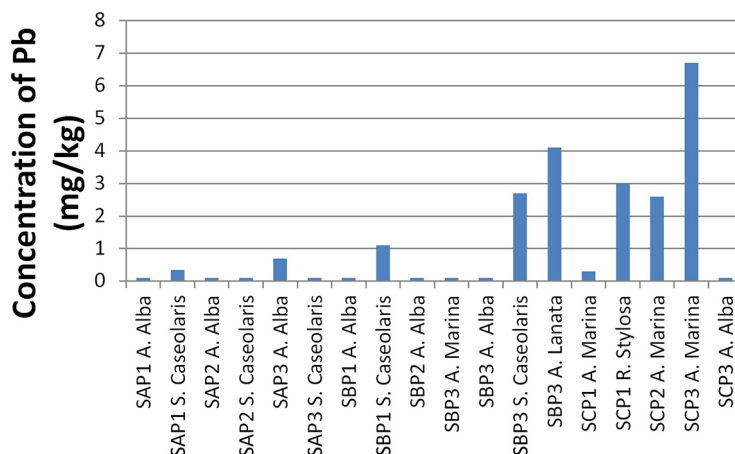
Figure 9 The concentration of Pb in the mangrove stems

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

roots, it easily formed complex bonds with the nutrients in the plants, limiting the plant's ability to translocate the element into the canopy (Hidayati, 2013).

Mangrove plants also absorbed and stored heavy metals in their body tissues, such as leaves, stems, and roots, which were transported in the sediment. Pb is mostly accumulated in plant organs, including their leaves, stems, and roots. The transfer of Pb from soil to plants relies on the composition and pH level of the land, with its cation exchange capacity (CEC). Plants also have the ability to absorb Pb, when soil fertility conditions and organic matter contents are low, whereas CEC is high (Sanadi et al, 2018).

The Pb concentration in leaves is further shown in Figure 10. The average Pb concentration in the leaves of *A. alba and S. caseolaris* at Station A, were 7.5 mg/kg and 1.96 mg/kg, respectively, which was in turn much higher than in the roots. Moreover, the average Pb concentration in the leaves of *A. alba, S. caseolaris, A. marina, and A. lanata* at Station B were 1.19 mg/kg, 0.51 mg/kg, 0.1 mg/kg, and 3.2 mg/kg, respectively. The average concentration of Pb in *A. lanata* leaves at Station B was also observed to be higher than in the roots. Moreover, the average concentration of Pb in the leaves of *A. marina, R. stylosa, and A. alba* at Station C were 0.84 mg/kg, 0.1 mg/kg, and 1.4 mg/kg, respectively.
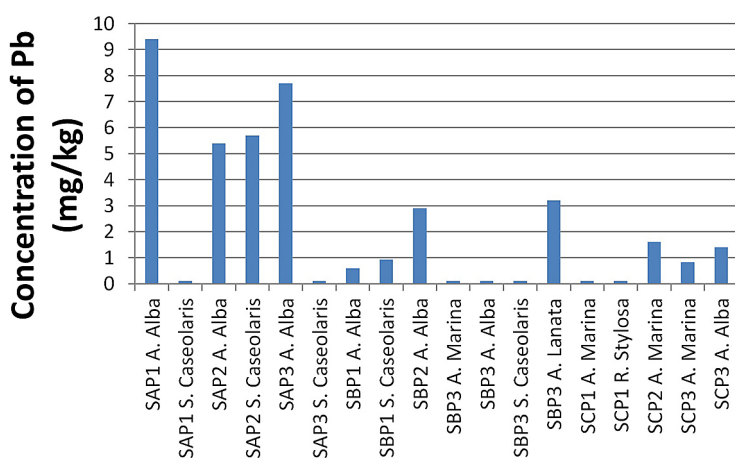

Figure 10. The concentration of Pb in mangrove leaves

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

According to Rachmawati et al. (2018), besides the high mobility of Pb from the roots to the leaves, it was also caused by the metal pollution in the air. The process of Pb entry into plant leaves was through the attachment of particles to the leaves and stomata, as the accumulation of the element contained within was high. The amount of Pb accumulated in the leaves was also one of the localization efforts performed by plants, through its accumulation into the plant organs (Heriyanto et al., 2011).

Determination of Bioconcentration Factor (BCF) and Translocation Factor (TF)

According to Sinha *et al.* (2004), plants acted as accumulators and excluders. The accumulator plants absorb pollutants and retain them in the body, while excluders limit the toxicants entering their tissues. Every plant has the tolerance capacity to accumulate heavy metals, as regards the purpose of phytostabilization. Bioconcentration (BCF) and translocation (TF) factors were used to predict which plants were used as phytoremediators. BCF is the ratio of metal concentrations in the environment, which is associated with plant tissue, while TF is the ability of plants to lift and distribute several elements through their bodies (Ganeshkumar et al., 2018). Generally, the BCF and TF values were greater and lesser than one, respectively. In addition, the BCF value was inverse to that of the TF, which showed that plants accumulated heavy metals, while possessing low ability to translocate them (Yoon et al 2006).

BCF was used to determine the ability of mangroves, in order to absorb heavy metals in the growing medium. The BCF was the ratio of heavy metals concentration in the roots, to the growing medium. According to Baker (1981), BCF was divided into three categories, namely the accumulator (BCF > 1), indicator (BCF = 1), and excluder (BCF < 1). The BCF value in this study was calculated by comparing the concentrations of Pb in the roots, which were then divided by the compositions in the sediment. The BCF calculation results are shown in Figure 11.

The BCF of *A. alba, A. marina, S. caseolaris, A. lanata,* and *R. stylosa* were 1.13–90, 0.28–164, 2.36–63.3, 7.70, and 1.22, respectively. The average BCF values of Pb were 21.84, 50.48, 22.63, 7.70, and 1.22 for *A. alba, A. marina, S. caseolaris, A. lanata, and R. stylosa* mangroves, respectively. The BCF values for all mangrove in this study were greater than one (> 1). Therefore, the mangrove types (*A. marina, A. alba, S. caseolaris, A. lanata and R.stylosa)* possessed the abilities to be hyperaccumulators of Pb.

On the basis of the research conducted by Febriana (2017), the mangrove type *Avicennia marina,* optimally absorbed Pb into its body. The accumulation factor was also influenced by the plant's age. However, the older the plant becomes, the more the concentration of heavy metals increases. Moreover, the concentration tends to decrease with the end of the plant's life cycle (Giller *et al*., 1993).

The BCF value was influenced by several factors, such as heavy metals, the organisms present,
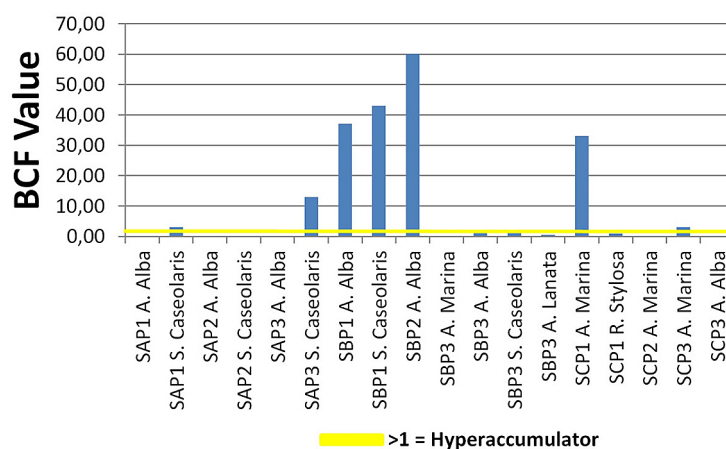
Figure 11. BCF Value

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

and water conditions (Hutagalung, 1985). A large BCF value also indicates that heavy metals accumulate in relatively huge quantities, from the sediment to the mangrove roots. Due to several factors, the difference in the BCF values occurred in similar mangroves. The factors that influenced the absorption of heavy metals by mangroves were concentration in sediment, age, metabolism, and density. According to Rosmarkam and Yuwono (2002), the plants' absorption rate was also influenced by the thickness of the cuticle layer, and nutrient status in the plant. Generally, the absorption of heavy metals decreases as the plant age increases. At high temperatures, the plant's ability to absorb heavy metals increases, due to the fact that its metabolism acts faster (Utami et al., 2018). The results of previous studies showed that the mangroves of *A. marina, A. alba, S. caseolaris, A.lanata,* and *R.stylosa* in the Wonorejo estuary, had a BCF value greater than 1 (BCF > 1) for Pb. It was then concluded that all mangroves in the study were accumulators. Therefore, the right growth zone for remediation of heavy metal pollutants was planting various mangrove species, as each plantation had different accumulation ability. Moreover, there were many pollutant substances in the estuary.

Translocation is the plant's ability to absorb and distribute various elements to its body parts (stems, leaves, and fruits) (Ganeshkumar et al., 2018). The value of the translocation factor was the ratio of the heavy metal substance in the stems or leaves, to the roots. According to (Majid et al., 2014), TF value has two categories, namely phytoextraction (TF > 1) and phytostabilization (TF < 1) mechanisms. The results of the TF calculation are shown in Figure 12.

On the basis of Figure 12, the TF from root to stem for *A. alba, S. caseolaris, A. marina, A. lanata, and R. stylosa* were 0.02–3.50, 0.09–22.33, 0.08–27, 10.25, and 0.34, respectively. The mangroves with a TF value greater than 1 (TF > 1), including *A. alba, A. marina, S. caseolaris, and A. lanata,* were the plants with phytoextraction mechanisms in their bodies . The mangroves with TF value less than 1 (TF < 1) had a phytostabilization mechanism, as the only plant in this category was *R. stylosa.*

On the basis of the table above, the TF range values of Pb from root to leaf for the *A. alba, A. marina, S. caseolaris, A. lanata, and R. stylosa* mangroves were 0.48–54, 1.14–1.49, 0.03–31.22, 8, and 0.01, respectively. The average TF values of Pb for the *A. alba, A. marina, S. caseolaris, A. lanata, and R. stylosa* mangroves were 40.31, 8.10, 11.03, 80, and 0.01, respectively. Due to the fact that the Pb translocation factor values of *A. alba, A. marina, S. caseolaris, and A. lanata* were greater than 1 (> 1), it was concluded that these types of mangrove had phytoextraction mechanisms. Phytoextraction is the situation where the roots absorb pollutants, which are then translocated to other organs to be reprocessed or disposed, at harvest or fall (Yoon *et al.*, 2006).

Therefore, since the *R. stylosa* mangrove had a TF value less than 1 (<1), it was concluded that it possessed a phytostabilization mechanism. Phytostabilizers are the plants that use their roots
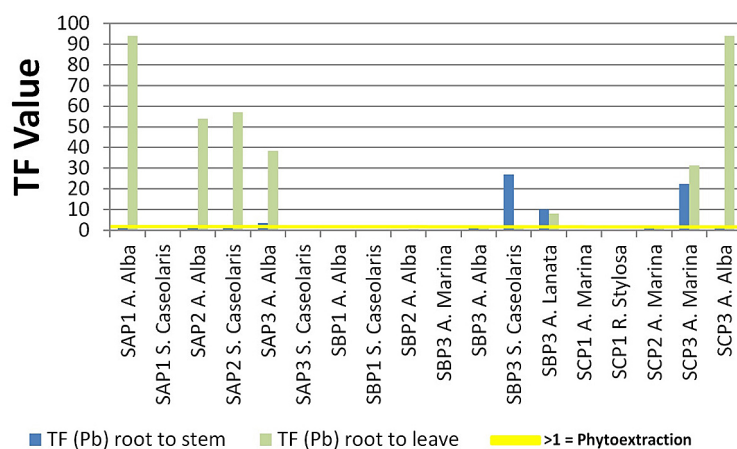

Figure 12. TF value from root to stem and TF value from root to leave

SAP1–3 = the sampling points at station A, plot 1 to plot 3,
SBP1–3 = the sampling points at station B, plot 1 to plot 3,
SCP1–3 = the sampling points at station C, plot 1 to plot 3

to change the environmental conditions. Plants stop the movement of metals that are absorbed and accumulated, through the use of the roots. Afterwards, they are absorbed and deposited in the rhizosphere, where plants limit the absorption of heavy metals from their environments (Hamzah and Pancawati, 2013).

The high TF value for non-essential metals (Pb) was due to high mobility of metals, from roots to leaves. Non-essential metals were not used in plant metabolism, as they were localized by plants in certain parts, in order for their toxicity to be degraded or reduced, through dilution (Rachmawati et al., 2018). Accumulation of heavy metals in mangrove bodies was also influenced by plant age. The older the plant becomes, the more the concentration of heavy metals increases. Moreover, the concentration tends to decrease with the end of the plant's life cycle (Giller *et al*., 1993).

The difference in TF values in similar mangroves occurred, due to several factors. One of the factors influenced the high TF value in leaf tissue. It was further suspected that the high concentration of heavy metals in leaves was due to the size and number of the stomata. The larger and more stomata on the leaves, the higher the absorption of heavy metals in leaf tissues tend to increase (Testi et al. 2019). According to Smith (1981), the high concentration of heavy metals in leaves increased the metal accumulation in leaf tissues over time. Moreover, this heavy metal was accumulated in the palisade tissue (fence network). The particles entered the through the stomata gap, settled in the leaf tissue, and accumulated between the spaces of fence/palisade cells.

CONCLUSIONS

The Pb concentration in the Wonorejo estuary had exceeded the quality standard, based on the Decree of the Minister of Environment, No. 51 of 2004, as it reached 0.008 mg/L. The Pb concentration in the Wonorejo estuary sediments had also exceeded the quality standard, based on the EPA Sediment Quality guidelines. Moreover, the highest Pb concentration occurred at Station C, as this location was directly next to the sea. The heavy metal density factors and pollutant sources also affected the distribution of heavy metals in the Wonorejo estuary. The Pb BCF values of *A.alba, A.marina, A.lanata, S. caseolaris* and *R. stylosa*

at the Wonorejo estuary, were observed to be greater than 1. This indicated that all mangroves involved were hyperaccumulators. However, the TF for Pb showed values greater than 1, which indicated that the mangroves involved were also phytoextractors. Conclusively, *A.alba, A.marina, A.lanata, S. caseolaris,* and *R. stylosa* should be used in Pb phytoremediation at coastal areas.

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