

## DEFENSE STRATEGY OF MANGROVE *AVICENNIA MARINA* FACING HEAVY METALS (PB, CD, AND CU) POLLUTION AT MANGROVE AREA, SEMARANG AND JEPARA COASTAL WATERS, CENTRAL JAVA INDONESIA: A PROSPECT TO PHYTOREMEDIATION

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### ABSTRACT

Mangroves *Avicennia marina* is known to absorb organic and inorganic materials, including heavy metals. This study was conducted at three estuaries (Tapak, Seringin, and Semat), situated at the Semarang and Jepara coastal waters, Central Java. This study aimed to know the capability of *Avicennia marina* to store heavy metals (Pb, Cd, Cu) in their roots and leaves, and to investigate their way to protect against metal pollutant and their prospect to be employed as phytoremediation plant. The research was conducted in March - October 2018 in the mangrove area at the estuary of Tapak (Semarang), Seringin River (Semarang), and Semat (Jepara). The research method used was the case study method. Samples taken from the mangrove field were sediment soil, water, roots, and leaves of *A. marina*. Metals content analysis carried out using Atomic Absorption Spectrophotometry (AAS). The results showed that Pb, Cd, and Cu content in roots and leaves: the highest was in the Seringin estuary (Pb: roots = 4.630 mg/kg, leaves = 2.098 mg/kg; Cd: roots = 2.307 mg/kg, leaves = 1.011 mg/kg; Cu: roots = 9.211 mg/kg, leaves = 5.895 mg/kg), and the lowest was in Semat estuary (Pb: roots = 2.993 mg/kg, leaves = 1.163 mg/kg; Cd: roots = 0.774 mg/kg, leaves = 0.268 mg/kg; Cu: roots = 4.554 mg/kg, leaves = 2.979 mg/kg), respectively. All metal concentrations accumulated in roots and leaves of *A. marina* were considerably lower than those in the surrounding sediment (BCF<1). The mentioned metals stored in leaves were lower than those in roots (TF<1). The BCF and TF of leaves and roots obtained for Pb, Cd, and Cu were < 1; thus, *A. marina* practiced defense mechanism against metals contamination by implementing an exclusion strategy.

**KEY WORDS :** Mangrove, *Avicennia marina*, Pb, Cd, Cu, Bioconcentration Factor, Translocation Factor

### INTRODUCTION

Coastal waters, straits, estuaries, and marine waters with high biological resources; however, this region also has a high risk of environmental changes caused by various human activity disturbances. Coastal waters also serve as a shipping activity, industry, tourism, and residential places. However, coastal waters can also be used as a waste disposal area of human activities from the mainland,

increasing the input of various hazardous chemicals and marine life, including may interfere with the growth of mangrove ecosystems (Agardy and Alder, 2005).

Mangroves are vegetation communities and shrubs, occupying intertidal wetland habitat in the tropical and subtropical coastal area, including estuaries and marine shorelines. Mangrove community has essential benefits to human beings and other living creatures since it has physical,

biological, social, and economic functions. They provide many ecosystem services, such as playing a pivotal role in stabilizing land in the face of changing sea level by trapping sediments, cycling nutrients, processing pollutants, supporting nursery habitats for marine organisms, and providing fuelwood, timber, fisheries resources. They consist of complex food chains or food webs. They also buffer land from storms and provide safe havens for humans in the coastal countries in which they occur. Mangroves have a high capacity to absorb and adsorb heavy metals and other toxic substances in effluents and exhibit high species diversity (Agardy and Alder, 2005; Yulianto *et al.*, 2020; Nagelkerken *et al.* 2020).

Estuaries region of Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) are located at Semarang and Jepara coastal waters. Those areas are influenced densely by various human activities. All of these estuary areas are significantly covered with mangrove vegetation, which plays an essential role in the environment. Adjacent areas along the Tapak and Seringin river are utilized for various activities, such as residential, agriculture, aquaculture, and industrial zones. Wijayakusuma Industrial Estate Semarang is located right next to the mangrove area of Tapak, with some industries, such as food, spices, soy sauce (ketchup), textiles, metallurgy, batteries, cardboard packaging, ceramics, and garment (Martuti *et al.*, 2016, 2017; DPMPTSP, 2020). The Terboyo Industrial Park located along the Seringin River with some industries such as textiles, paper, tapioca, aluminum, paint, fodder, and timber. Meanwhile, the estuary of Semat located in Jepara regency. This area is close to the residential, agricultural, and center of furniture industries (DPMPTSP, 2020).

Due to the increased discharge of waste pollutants as a result of rapid industrial development along the coast and rivers, causing pressure on estuaries and mangrove ecosystems increased (Ruaeny *et al.*, 2015). One of the pollutant wastes is heavy metals. All heavy metals have adverse effects at high concentrations; however, they are considered environmental contaminants (Yulianto *et al.*, 1995; Soegianto *et al.*, 2010; Usman *et al.*, 2013; Yulianto *et al.*, 2019). They become important pollutants entering the estuary ecosystem (Zhang *et al.* 2014). Mangrove, through their roots also known as nutrients trap, including contaminants such as heavy metals. Heavy metals captured by sediments accumulated in the

mangrove roots and stored in the stability of the sediment-metal bond. Through the root system, heavy metals in sediments of mangrove roots absorbed and distributed throughout the plant (Keshavarz *et al.*, 2012).

The plant has the high ability to store chemical elements from soils and metabolize them into molecules in their tissues, which seems very hopeful to eliminate contaminants in the environment; it is called phytoremediator plant (Gurbisu and Alkorta, 2003). Due to most plant roots are situated in the soil, they can function in removing metals through filtering, adsorption and cation exchange, and through chemical changes induced by plants in the rhizosphere (Nouri *et al.*, 2009). This capability is used as a plant strategy to anticipate intoxication by heavy metals. Baker and Walker (1990) classify vegetations into three categories corresponding to their strategy for living on metal-polluted soils, i.e., metal excluders, indicators, and accumulators (or hyperaccumulators). Plant with a strategy of metal accumulators (hyperaccumulators) is a plant species that translocates metals in their up-soil tissues to rates far beyond those concentrated in the soil. These plants are capable of uptaking heavy metals from substrates and store them in their shoots, and they are considerably practiced in phytoremediation. Metal excluders are plants which functionally restrict the rates of heavy metal translocation inside them and preserve to low degrees in their higher shoot over an extended interval of soil levels. However, they can still concentrate on high quantities of metals in their roots. Indicator vegetal species are a plant that holds the heavy metal levels the same as those stored in the peripheral milieu or soil.

The population of *A. marina* on the mangroves area at Tapak, Seringin, and Semat estuary in northern coastal waters of Central Java is relatively decreased. Little information is available on the study of heavy metal concentration in this mangrove species. Most studies have focused on mangrove community structure. This paper was to reveal the lead (Pb), cadmium (Cd), and copper (Cu) contamination on mangroves *A. marina* and deals with the content of these heavy metals in sediment, water, mangrove roots and leaves of *Avicennia marina* from three estuaries situated at Semarang and Jepara coastal waters, Central Java. It was also to know whether this vegetation can strategically regulate heavy metal contamination in their environment subjected to metals pollutions.

## MATERIALS AND METHODS

The study was conducted in March - October 2018 in the mangrove ecosystem at the estuary of Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) River, the northern coast of Central Java (Figure 1). Root and leaf samples taken from *Avicennia marina* trees with trunk sizes ranging from 28-30 cm and tree height varied from 2-3 m. The taken root was the cable root (the end of the root, which is in the sediment). While the leaves taken were old leaves at the base of the branches and young leaves at the end of the branches. Subsequently, samples brought to the laboratory for analysis of Pb, Cd, and Cu content.

Samples of water (600 mL) were taken at each study site, with a depth of  $\pm 30$  cm from surface water. Water samples were stored in polyethylene bottles and preserved by adding a 20 mL HNO<sub>3</sub> solution. Onekg of sediment samples taken using a grab at each location with a depth of  $\pm 30$  cm from the surface, then put in plastic bags.

Furthermore, water and sediment samples obtained were brought to the laboratory for analysis of metals using atomic absorption spectrophotometer (Shimadzu AA-700). At each location, samplings of water, sediment, leaves, and roots were done triplicates.

## Data Analysis

Data of metals (Pb, Cd, and Cu) content in roots, leaves, sediment, water, were descriptively analyzed. While the calculation of Bioconcentration Factor (BCF) and Translocation Factors (TF) was conducted to evaluate *A. marina*'s strategy facing and adapting a metal-contaminated environment. BCF is the ratio of root metal to sediment metal concentration. While TF is the ratio of leaf metal to root metal concentration. The formula can calculate BCF and TF are as follows:

$$BCF = \frac{\text{Heavy metal in Root or Leaf}}{\text{Heavy metal in Sedimen or Water}}$$

$$TF = \frac{\text{Heavy metal in Leaf}}{\text{Heavy metal in Root}}$$

Data on the BCF and TF were tabulated using Windows Excel and are presented in the form of the Table.

## RESULTS AND DISCUSSION

Semarang city and other surrounding areas are a center of industrial activity. The development of industrial location in Semarang city is strategically along the east part of the Semarang-Demak and west part of Semarang-Kendal. Tapak river located

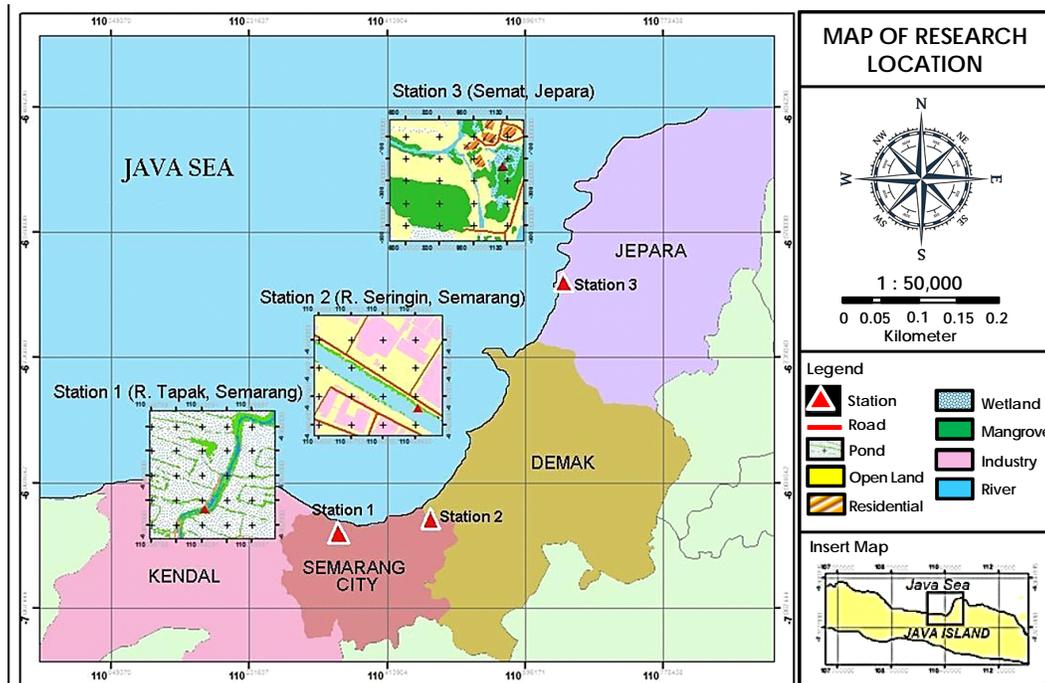


Fig. 1. Map of research location at estuary of Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) River, northern coastal of Central Java

in the western part of Semarang, close to Wijayakusuma Industrial Estate, and has an estuary in the Java Sea. Meanwhile, Seringin River is located in the eastern part of Semarang, through Terboyo industrial area, and has a river mouth in the Java Sea. And, the estuary of Semat located in Jepara regency. This area is close to the residential, agricultural, and center of furniture industries.

#### The lead, cadmium, and copper content in seawater and sediment

Heavy metals contents in water and sediment from the mangrove area at Station Tapak and Seringin (at Semarang) were adequately higher than Station Semat (at Jepara). Nevertheless, the seawater's metal concentrations were not far different in whether Station Tapak and Seringin or Station Semat. In sediment, at all research stations, the metal concentrations were in the order of magnitude  $Pb > Cu > Cd$ . Regarding the location, metal contents in seawater and sediment ranged in order of magnitude: Seringin > Tapak > Semat. And, the concentration of heavy metals in sediments was higher than in seawater (Table 1). In this present study, it was obtained the CF of Pb (Tapak: 5.89, Seringin: 9.9, and Semat: 6.84), Cd (Tapak: 174, Seringin: 520, and Semat: 306), and Cu (Tapak: 24.5, Seringin: 24.6, and Semat: 11). Martuti *et al.* (2017) exhibited more significant Cu content in sediments than in the water in the Tapak mangrove area (Semarang) with Cu in water and sediment ranged from 0.02 to 0.05 mg/L and 15.20 to 25.03 mg/kg, respectively, and the concentration factor (CF, the ratio of sediment/water) ranged from 500 to 898.

Whereas according to Pratama *et al.* (2012), Pb in water and sediments in the Tapak mangrove area (Semarang) ranged from 0.41-1.11 mg/L and 38.24-62.87 mg/kg, respectively, so that a CF ratio was obtained ranging from 57 to 93. Alisa *et al.*, (2020) found Cd content in seawater in Untung Island, Jakarta Bay, ranged from 0.067-0.079 mg/L. At the same time, the Cd in sediments reached 1,968-2,760 mg/kg, so the CF was from 29-35. Aminah *et al.*, (2016) investigated Cd level in the water at the Port of Pasuruan, East Java, and it ranged from 0.0271 to 0.0675 mg/L, and Cd in sediments ranged from 0.0343 to 0.1002 mg/kg, so the CF value was from 1.3-1.5.

#### The lead (Pb), cadmium (Cd) and copper (Cu) content in mangrove roots and leaves

All heavy metals in roots and leaves of *A. marina* from the mangrove community at Tapak and Seringin estuary were much higher than those in the Semat estuary area (Table 2). At all locations, the contents of Pb, Cd, and Cu in roots ranged from 2.699 to 4.893 mg/kg, 0.614 to 3.134 mg/kg, and from 3.679 to 10.022 mg/kg, respectively. The content of Pb, Cd, and Cu in leaves of *A. marina* was relatively lower than in roots, and it ranged from 1.097 to 1.968 mg/kg for Pb, 0.245 to 1.296 mg/kg for Cd, and 2.356 to 6.584 mg/kg for Cu (Table 2). Based order of magnitude, the root of *A. marina* gradually absorbed heavy metals:  $Cu > Pb > Cd$ . Pb and Cu in the roots and leaves of *A. marina* in the present study were lower compared to the mangroves of Muara Angke, Jakarta; in roots, it ranged from 20.98 to 59.16 mg/kg and 13.88 to 37.68

**Table 1.** Heavy metals concentration (Pb, Cd and Cu) in water and sediment from mangrove area at the Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) estuary, northern coastal waters of Central Java, Indonesia.

Sample	Station	Range/Average	Pb (mg/L) or (mg/kg)	Cd (mg/L) or (mg/kg)	Cu (mg/L) or (mg/kg)
Water	Tapak	Range	1.546-2.234	0.098-0.132	0.029-0.065
		Average $\pm$ SD	1.890 $\pm$ 0.344	0.116 $\pm$ 0.017	0.046 $\pm$ 0.018
	Seringin	Range	1.098-2.263	0.114-0.187	0.012-0.034
		Average $\pm$ SD	1.643 $\pm$ 0.586	0.148 $\pm$ 0.037	0.021 $\pm$ 0.012
	Semat	Range	0.779-2.212	0.109-0.176	0.011-0.033
		Average $\pm$ SD	1.442 $\pm$ 0.722	0.135 $\pm$ 0.036	0.020 $\pm$ 0.012
Sediment	Tapak	Range	9.677-12.211	1.908-3.477	5.677-10.354
		Average $\pm$ SD	11.131 $\pm$ 1.308	2.837 $\pm$ 0.823	8.002 $\pm$ 2.339
	Seringin	Range	12.698-18.223	14.556-18.589	8.756-12.111
		Average $\pm$ SD	16.271 $\pm$ 3.099	3.645 $\pm$ 2.083	10.911 $\pm$ 1.863
	Semat	Range	6.454-13.351	7.788-11.789	3.696-9.008
		Average $\pm$ SD	9.861 $\pm$ 3.449	1.478 $\pm$ 2.004	6.124 $\pm$ 2.685

mg/kg, respectively; and in leaves, it ranged from 54.31 to 64.32 mg/kg and 7.08 to 10.07 mg/kg, respectively (Hamzah and Setiawan, 2010). Martuti *et al.* (2017), in the Tapak mangrove ecosystem, Semarang found Cu content in roots ranged from 0.87 to 2.21 mg/kg, and Cu in leaves ranged from 0.88 to 2.64 mg/kg.

### The Bioconcentration and Translocation Factor

All metals were accumulated in roots to concentrations inferior to those of adjacent sediments with root bioconcentration factors (BCF) of  $< 1$  (Table 3). Root BCFs were not too varied across the research locations for all metals. However, root BCFs on essential metals were greater than the non-essential metals (BCF of Cu: 0.744-0.873; BCF of Pb: 0.284-0.344 and BCF of Cd: 0.422-0.633) (Table 3). Leaf BCFs and Translocation Factor (TF) on essential metals (BCF leaf/soil of Cu: 0.486 - 0.540 and TF shoot/root of Cu: 0.571 to 0.644) showed greater mobility than in non-essential metals (BCF of leaf/soil of Pb: 0.112-0.129 and BCF of leaf/soil of Cd: 0.181-0.277; TF of leaf/root of Pb: 0.389-0.454 and TF of leaf/root of Cd: 0.346-0.438) (Table 3). Metals accumulated higher in sediment and were absorbed and stored in a few levels by mangroves *A. marina*. Pb, Cd, and Cu were absorbed from soil to roots and translocated in the lower level from the roots to leaves. Roots tend to hold the metals remain outside the plant body so that metals do not penetrate greater inside the plant tissue. This mechanism indicated that an exclusion strategy for the metal defense was practiced widely by *A. marina*. Significantly higher metals concentration in sediments surrounding roots zones, but lower in

roots and shoots of the mangrove *A. marina* in the present study indicated that the roots act as a barrier for metal translocation and protect the above-ground sensitive parts of the plant from metal contamination. The mentioned mangrove species could limit relatively higher metal concentrations above the toxic concentration enter into the plant roots. With a low BCF ( $< 1$ ) and TF ( $< 1$ ), indicates the existence of an internal defense mechanism against metal intoxication; thus, they have the potential for phytostabilization. Root and leaf BCFs and TF for the essential metal Cu were higher compared to non-essential metals Pb and Cd. Those greater Cu BCFs and TFs indicate higher mobility of Cu from roots to shoots due to the important requirement of essential metal such as Cu by plant physiological processes. Martuti *et al.* (2017), in the Tapak mangrove ecosystem, Semarang obtained BCF value for Cu in *A. marina* ranged from 0.04-0.13, and TF ranged from 0.4-1.1 during six samplings of each two weeks. Those results are comparable with that of those present study. Considering the leaves and roots BCFs and TFs analysis for the essential (Cu) and non-essential metals (Pb and Cd), in the present study, *A. marina* can be classified as an excluded mangrove species. A plant holding elevated metal level in roots but has the shoots/roots ratio  $< 1$  is categorized as a heavy metal excluder (Baker and Walker, 1990; Boularbah *et al.*, 2006). The strategy used by *A. marina* to tolerate metals contaminated environment was by activating the roots that play a more significant role in holding the mobility of heavy metals from moving up towards the stems and leaves of the plant, thereby reducing the intoxication process in plants by metals. By those

**Table 2.** The range and average content  $\pm$  SD of Pb, Cd, and Cu in the roots and leaves of *Avicennia marina* from mangrove area at the Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) estuary, northern coastal waters of Central Java, Indonesia.

Sample	Location	Range / Average	Pb (mg/kg)	Cd (mg/kg)	Cu (mg/kg)
Roots	River Tapak	Range	2.774-3.594	0.844-1.565	5.896-7.566
		Average $\pm$ SD	3.163 $\pm$ 0.412	1.196 $\pm$ 0.361	6.986 $\pm$ 0.945
	River Seringin	Range	4.112-4.893	1.633-3.134	8.542-10.022
		Average $\pm$ SD	4.630 $\pm$ 0.449	2.307 $\pm$ 0.762	9.211 $\pm$ 0.750
	River Semat	Range	2.699-3.382	0.614-0.924	3.679-5.018
		Average $\pm$ SD	2.993 $\pm$ 0.351	0.774 $\pm$ 0.155	4.554 $\pm$ 0.758
Leaves	River Tapak	Range	1.099-1.432	0.245-0.693	3.741-4.443
		Average $\pm$ SD	1.243 $\pm$ 0.171	0.518 $\pm$ 0.240	3.988 $\pm$ 0.395
	River Seringin	Range	1.225-1.968	0.745-1.298	4.790-6.584
		Average $\pm$ SD	2.098 $\pm$ 0.945	1.011 $\pm$ 0.277	5.895 $\pm$ 0.967
	River Semat	Range	1.097-1.234	0.196-0.392	2.356-3.574
		Average $\pm$ SD	1.163 $\pm$ 0.069	0.268 $\pm$ 0.108	2.979 $\pm$ 0.609

**Table 3.** Bioconcentration Factor (BCF) of root metal and leaf metal to sediment metal concentration, and Translocation Factors (TF) of leaf metal to root metal concentration in *A. marina* from mangrove area at the Tapak (Semarang), Seringin (Semarang), and Semat (Jepara) estuary, northern coastal waters of Central Java, Indonesia.

Station	BCF						TF		
	Pb		Cd		Cu		Pb	Cd	Cu
	Root	Leaf	Root	Leaf	Root	Leaf	Leaf/Root	Leaf/Root	Leaf/Root
River Tapak	0.284	0.112	0.422	0.183	0.873	0.498	0.393	0.433	0.571
River Seringin	0.284	0.129	0.633	0.277	0.844	0.540	0.454	0.438	0.640
River Semat	0.304	0.118	0.524	0.181	0.744	0.486	0.389	0.346	0.654

strategies, *A. marina* can survive and continue their existence living in the metal-contaminated environment. For phytoremediation initiatives, mangrove *A. marina* is hopefully best employed as phytostabilizer, potentially aiding in the retention of toxic metals remain in sediments surrounding roots zones and thereby reducing heavy metals transport to adjacent estuarine and marine systems.

### CONCLUSION

*Avicennia marina* showed its strategy in protecting metals contamination by maintaining metals in the bottom-ground and limiting metals' translocation to aboveground towards sensitive parts such as stems and shoots. Since its strategy, *A. marina* is called as an excluder mangrove species. Mangrove *A. marina* is expected to become a phytoremediator species due to their ability to play as phytostabilizer of heavy metals, thereby reducing heavy metals transport to the adjacent estuarine and marine environment.

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