

Potential Mangrove Species in Porong River Estuary as Inhibiting Agent of Heavy Metal (Pb, Cu and Zn) Pollution

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ABSTRACT

This study investigated the ability of the mangrove species to accumulate heavy metals such as lead (Pb), copper (Cu) and zinc (Zn). Concentrations of these metals in sediment, roots and leaves of *Avicennia alba*, *Sonneratia alba*, *Avicennia marina* and *Rhizophora mucronata* found in the mangrove ecosystem of the Porong River estuary, Sidoarjo, East Java, Indonesia were measured. The bio-concentration factor (BCF) and translocation factor (TF) were calculated. The results showed that Pb concentrations in the roots and leaves of mangroves were 0.0038-0.0505 mg/kg and 0.0042-0.0395 mg/kg, respectively, while Cu concentration in the roots of mangroves was 0.2658-0.3390 mg/kg and in the mangrove leaves was 0.0655-0.1490 mg/kg. The average concentration of Zn found in mangroves ranged from 1.327-3.1380 mg/kg in the roots and 1.767-3.674 mg/kg in the leaves. Among all the mangroves, the highest BCF values for Pb, Cu and Zn were found in *Avicennia alba*. However, the highest TF for Pb and Zn was found in *Sonneratia alba*. On the other hand, the highest TF for Cu was found in *Rhizophora mucronata*. The capability of *Avicennia alba* to accumulate Pb, Cu and Zn heavy metals compared with other species is highly important for determining a suitable species for phytoremediation.

Keywords: Bioaccumulation, bio-concentration, heavy metals, mangrove wetland, phytoremediation

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INTRODUCTION

Rising populations around the world the development of industry may result in anthropogenic discharges that pose a threat

to ecosystems. Industries listed as causing heavy-metal pollution are the battery, electronic, chemical, electroplating and paint industries (Marg, 2011). Sidoarjo Regency in Indonesia is one of the many cities that contribute to environmental pollution due to industrial activities such as cement, battery, chemical, electroplating and plastics manufacturing. Consequently, this area is prone to heavy-metal pollution. The strongest impact is experienced by the estuarine and coastal waters since heavy metal discharge from industries and urban areas from incoming freshwaters and tidal water is accumulated in the estuary (Tam & Wong, 2000). This situation is expected to occur in the Porong River estuary. The increasing concentrations of Pb, Cu and Zn in this estuary will have long-term implications to the biota.

Cu and Zn act as essential plant micronutrients, being involved in enzyme mechanisms, while Pb is a non-essential element that is toxic to the biota. Zinc is often found in contaminated sediment of estuarine areas at high concentrations of up to 800 $\mu\text{g/g}$ (Luoma, 1990). The presence of these heavy metals in high-level concentrations may cause harm to metabolic processing in the cellular level, inhibit enzyme reaction and the anti-oxidative process, delay growth and cause mortality of biota (Vangronsveld & Clijsters, 2008). Furthermore, these heavy metals are continuously found at high concentrations in contaminated estuarine areas, including the Porong River estuary. A previous study by Lapindo Mud Flow reported that discharge from the Porong River contained Cu and Pb at levels of

24.5 and 17.8 mg/L , respectively (United Nations Environment Program/Office for the Coordination of Humanitarian, 2006). In 2009, Pb was found in the estuary of the Porong River ranging from not detected to 0.0490 mg/L while Cu was determined to be 0.083-1.310 mg/L (Juniawan, Rumhayati, & Ismuyanto, 2012; Parawita, Insaftiri, & Nugraha, 2009). Zn reached 0.03 ± 0.01 mg/L in 2005 in pond waters in the Porong estuary (Kohar, Budiono, Indriany, & Wilujeng, 2005).

Mangrove estuary ecosystems have the ability to reduce heavy-metal contamination due to the ability of their roots to absorb and re-translocate heavy metals into other parts of the plant (Mejías, Musa, & Otero, 2013). In addition, it has been concluded by a previous study that mangroves have high tolerance to heavy metal pollution; the LC_{50} for Cu, Zn and Pb in mangroves was 566, 580 and 400 $\mu\text{g/g}$, respectively (MacFarlane, Koller, & Blomberg, 2007). The capability of mangroves to accumulate heavy metals from their environment makes them a suitable candidate for heavy-metal pollution remediation. This capability is applied in terms of the phytoremediation method. It is a method that employs plants for undergoing remediation of pollutants. By taking advantage of the ability of mangrove to absorb and accumulate heavy metals, phytoremediation is considered a cost effective and ecologically friendly technology (Etim, 2012).

However, despite its advantages, application of the phytoremediation technique using mangrove plants especially in the Porong River estuary faces difficulties.

The selection of the most appropriate mangrove species for remediation in this area is currently under discussion. Furthermore, studies regarding the ability of the mangrove species to inhibit Pb, Cu and Zn, particularly in the Porong River estuary, are limited.

This study tried to determine the concentration of heavy metals, Pb, Cu and Zn, in sediment and parts (roots and leaves) of the mangrove plant found in the Porong River estuary, and finally to reveal the potential of the mangrove species in the Porong River estuary as a suitable inhibiting agent of Pb, Cu and Zn pollution.

MATERIALS AND METHOD

The research was conducted in the mangrove ecosystem of the Porong River estuary in September 2014. Three sampling stations were chosen to represent the difference in heavy metal input from the river mouth (Stations 1 and 2) and close to the open sea (station 3). Four mangrove species were identified in this area: *Avicennia alba*, *Sonneratia alba*, *Avicennia marina* and *Rhizophora mucronata*. Mangroves more than 2 m in height and 15-20 cm in diameter, as well as their surface sediment, were collected. Sampling sites are presented in Figure 1 and Table 1.

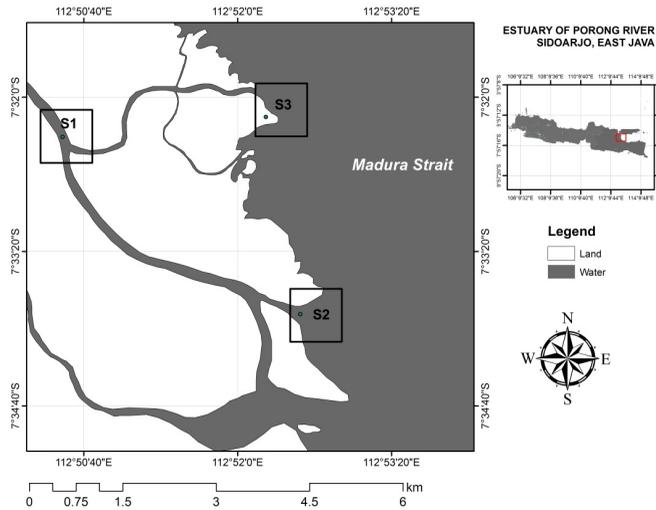


Figure 1. Location of sampling sites

Table 1
Sampling sites position and dominated mangrove species in mangrove ecosystem of Porong

Study Area	Mangrove Species
S 07° 53' 90" E 112° 84' 10"	<i>Avicennia alba</i> , <i>Avicennia marina</i> , and <i>Sonneratia alba</i>
S 07° 56' 40" E 112° 87' 60"	<i>Avicennia marina</i> and <i>Sonneratia alba</i>
S 07° 53' 60" E 112° 87' 10"	<i>Rhizophora mucronata</i> and <i>Avicennia marina</i>

Each species was sampled in duplicate for its sediments, roots and leaves from the same individual tree. A 10-cm diameter PVC pipe was used to sample the sediment at the mangrove roots at 5-10 cm deep from the surface layer. Next, about 20-30 pieces of the leaves were taken from middle-aged mangrove trees, while the roots were removed with a knife at the part buried in the sediment. Prior to laboratory analyses, all samples of roots, leaves and sediments were placed in plastic bags and stored in a cool box. The samples were then dried in an oven at 105 °C as the samples were measured for heavy metal content (Pb, Cu and Zn) based on dry weight calculation. Water quality parameters i.e. temperature, salinity, pH and DO were measured *in situ* as the supporting data for environmental conditions.

The analysis was conducted in the Water Quality Laboratory, Jasa Tirta Malang. Atomic Absorption Spectrophotometry (AAS) Shimidzu AA-6800 was used to measure the concentration of heavy metals in the sediments, roots and leaves of the mangrove. To monitor the performance of the instrument, blank and standard solutions were used to obtain data quality by developing calibration curves. Heavy metal in the sediments and mangrove parts (leaves and roots) were analysed using the APHA method (USEPA-311B and 3050B).

To determine the potential of mangrove species for accumulation of heavy metals Pb, Cu and Zn in the study area, BCF and TF were calculated. BCF is a ratio between

metal concentrations in the leaf to metal concentration in the sediments, while TF is generated from the ratio between metal concentration in the leaf to metal concentration in the roots (Etim, 2012). The statistical method was applied to determine the significance difference of the mean of heavy metal concentrations in the sediment among the stations using one-way ANOVA. SPSS 16.0 and Microsoft Excel 2007 software were utilised for the data analysis.

RESULTS AND DISCUSSIONS

Concentration of Pb, Cu and Zn in the Surface Sediment

The average concentration of Pb, Cu and Zn in the surface sediment of the Porong River estuary is presented in Figure 2. Pb was found between 0.059-0.068 mg/kg, which is below the Interim Sediment Quality Guideline of the Canadian Council of Ministers of the Environment (CCME) level of 35 mg/kg. The concentration of Pb in the surface sediment was considerably lower than the concentration of Pb in the sediment at the mangrove ecosystem of Jakarta Bay, which was between 18.640-29.570 mg/kg as reported by Hamzah and Setiawan (2010). Jakarta Bay is surrounded by significantly increasing anthropogenic activities. Waste, including heavy metal pollution, comes from five river mouths to Jakarta Bay (Takarina, 2011). While the Porong River estuary is also an urban area, environmental stress is lower than at Jakarta Bay.

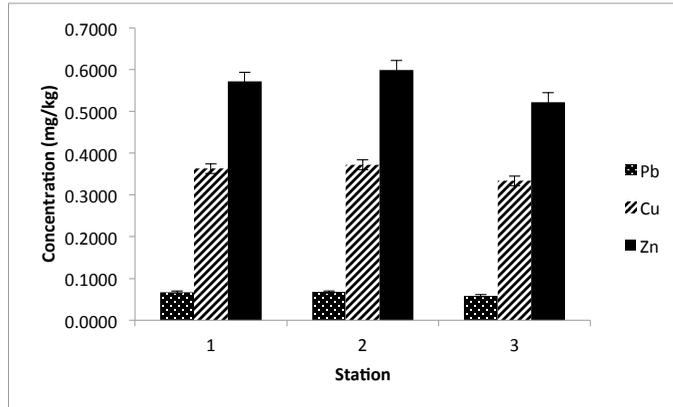


Figure 2. The concentration (mean and standard error) of Pb, Cu and Zn in mangrove sediments at Porong River estuary (n=4)

Concentration of Cu in surface sediment of the study area ranged from 0.3338-0.3723 mg/kg. Compared to that of the mangrove ecosystem in Tapak, Semarang with an average of 0.275 mg/kg (Kariada & Irsadi, 2014), Cu concentration in this study was higher. However, it did not exceed the Interim Sediment Quality Guideline of CCME (0.019 mg/kg).

Subsequently, the average concentration of total Zn in the Porong Estuary Mangrove Sediment was 0.584 mg/kg, which was significantly lower than the Interim Sediment Quality Standard. Zn of CCME (123 mg/kg) found in the mangrove sediment of the Porong River estuary was considerably less than at other locations such as Mai Po, Hong Kong (240 mg/kg) and Hawksbury, Australia (94 mg/kg) (MacFarlane, 2002). In Indonesia, Zn concentration of mangrove sediment in the Porong estuary was lower than in the Mahakam Delta (74.95 mg/kg) (Budiyanto & Lestari, 2013), Jakarta Bay (64.2-209.4 mg/kg) (Sindern et al., 2016)

and Dumai (31.490–87.110 mg/kg) (Amin, et al., 2009).

The average concentration of Cu and Zn was not significantly different among the stations ($F_{Cu}=1.14$ and $F_{Zn}=1.02$, p value >0.05 , $n=4$) in the Porong River estuary. However, there was a significant difference in Pb concentration among the stations ($F_{pb}=4.76$, p value >0.05 , $n=4$). The highest concentration of Pb was at Station 2, while the lowest concentration for Pb was found at Station 3. Mangrove at Station 2 was located near incoming water streams that may have been contaminated as a result of the development of industrial areas and harbour activities as well as increasing population around the upper water stream of the Porong River. Consequently, mangrove swamp sediment in these areas retain and accumulate more heavy metals compared to Station 3, which is located near the open sea. It has been noted by MacFarlane and Burchett (2002) that mangrove sediment has significant ability in trapping heavy metals

originating from tidal waters, downstream rivers and river run-off.

Furthermore, Zn was the most prevalent heavy metal in the sediment of this estuary, followed by Cu and Pb ($Pb < Cu < Zn$). Among the eight metals found there (Fe, Mn, Ni, Pb, Cu, Zn, Cr, Cd), Zn was present in abundance, that is at 105 ppm in the mangrove sediment, Punta Mala Bay, Pacific Panama (Defew, Mair, & Guzman, 2005). In aquatic environments, Zn and Cu are classified as nonpoint sources i.e. originating from anthropogenic activities; for example, urban and agricultural runoff and boating activities are common sources of Cu in water (Joseph & Kundig, 1998).

It is very important to measure Pb, Cu and Zn concentration in mangrove sediment as concentration of the heavy metals, Pb, Cu and Zn, influence their concentration in parts of the mangrove. A high concentration of metals in sediment due to high bioavailability is associated with the concentration of these metals in the mangrove plant (Marchand, Fernandez, & Moreton, 2016).

The concentration of heavy metals Pb, Cu and Zn in mangrove sediment is high in environments that have significant human-induced stressors. A high concentration of Pb, Cu and Zn was found in the sediment of the mangrove ecosystem near Yaibu City, the Red Sea Coast of Saudi Arabia, resulting from industrial effluent, domestic runoff and sewage (Abohassan, 2013). Maldonado-Román et al. (2012), who conducted a similar study in Peninsula La Esperanza in the northern coast of Puerto Rico, reported

that mangrove vegetation presence in coastal areas tended to accumulate high amounts of heavy metals including Pb, Cu and Zn in their sediment, predictably from clandestine dumpsites and thermo-electrical refineries. Furthermore, with regards to anthropogenic input, the concentration of Pb, Cu and Zn found in the surface sediment of Sundarban Mangrove Ecosystem, Bangladesh, in 2012 was significantly associated with non-point sources (agricultural activities) and point-sources (jetties and boat activities) (Kumar et al., 2016).

Concentration of Pb, Cu and Zn in Mangrove Parts

It has been proven that mangrove tissue has high capacity for absorbing and accumulating heavy metals from sediment. Excess metals in sediment tend to be distributed among mangrove tissue depending on mobility of heavy metals (Kumar, Sajish, Kumar, George, & Viyol, 2010). The roots of four studied mangrove species accumulate heavy metals Pb, Cu and Zn in varying concentration, in the proportion $Zn > Cu > Pb$. This pattern is likely due to the concentration of metals in the sediment near their roots. Moreover, the higher value of Zn and Cu in roots of all studied mangrove species found in this estuary was likely due to their function as an essential element of mangrove plants. Pb was found at low concentration in the roots, ranging from 0.004-0.051 mg/kg, because Pb content in the sediment (0.059-0.068 mg/kg) is also low. This is due to the fact that Pb is a non-essential element, and it is more

immobilised in the sediment. In addition, Pb has low solubility in acidic sediment such as the mangrove ecosystem (Usman, Alkredaa, & Al-Wabel, 2013). Concentration of Zn,

Cu and Pb in the roots and leaves of four mangrove species found in the Porong River estuary is listed in Table 2 and Table 3, respectively.

Table 2
Average of metal concentration (mg/kg dry weight) in roots of mangrove species at Porong River estuary

Species	Pb	Cu	Zn	Species	Pb	Cu	Zn
Mangrove Roots Tissue				Mangrove Roots Tissue			
<i>Avicennia alba</i>				<i>Avicennia marina</i>			
Mean	0.0505	0.3390	1.6385	Mean	0.0437	0.2917	3.1375
SD	0.0672	0.1428	0.0940	SD	0.0520	0.1918	3.9769
Maximum	0.0980	0.4400	1.7050	Maximum	0.1390	0.6100	11.2000
Minimum	0.0030	0.2380	1.5720	Minimum	0.0030	0.0700	1.0260
% CV	133.02	42.13	5.74	% CV	119.11	65.78	126.75
<i>Sonneratia alba</i>				<i>Rhizophora mucronata</i>			
Mean	0.0038	0.2658	1.3270	Mean	0.0320	0.0865	1.6385
SD	0.0010	0.0689	0.1859	SD	0.0240	0.0332	0.0799
Maximum	0.0050	0.3440	1.5990	Maximum	0.0490	0.1100	1.6950
Minimum	0.0030	0.1780	1.2060	Minimum	0.0150	0.0630	1.5820
% CV	25.53	25.92	14.01	% CV	75.13	38.42	4.88

Table 3
Average of metal concentration (mg/kg dry weight) in leaves of mangrove species at Porong River estuary

Species	Pb	Cu	Zn	Species	Pb	Cu	Zn
Mangrove Leaves Tissue				Mangrove Leaves Tissue			
<i>Avicennia alba</i>				<i>Avicennia marina</i>			
Mean	0.0395	0.1490	3.6735	Mean	0.0072	0.1908	1.9198
SD	0.0163	0.0325	0.9383	SD	0.0059	0.0988	0.9832
Maximum	0.0510	0.1720	4.3370	Maximum	0.0190	0.3180	3.3280
Minimum	0.0280	0.1260	3.0100	Minimum	0.0044	0.0400	0.8830
% CV	41.17	21.83	25.54	% CV	81.71	51.80	51.21
<i>Sonneratia alba</i>				<i>Rhizophora mucronata</i>			
Mean	0.0348	0.1145	3.0250	Mean	0.0042	0.0655	1.7665
SD	0.0197	0.0635	2.5536	SD	0.0003	0.0445	1.3739
Maximum	0.0620	0.2080	6.7630	Maximum	0.0044	0.0970	2.7380
Minimum	0.0150	0.0660	1.3020	Minimum	0.0040	0.0340	0.7950
% CV	56.60	55.50	84.42	% CV	6.73	68.01	77.78

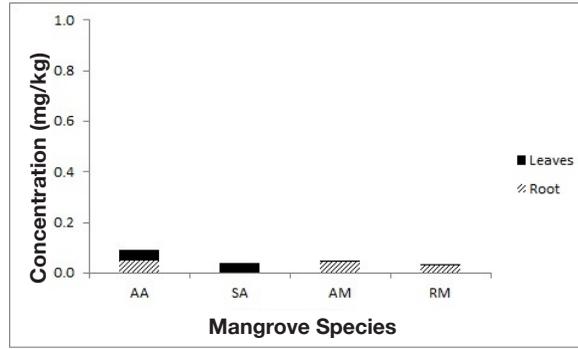
The concentration of Pb, Cu and Zn in the roots of *A. marina* in the Porong River estuary are significantly lower than those reported in the Sydney estuary (153, 189 and 378 mg/kg for Cu, Pb and Zn, respectively). The average concentration of these metals in sediment is 95, 42 and 196 mg/kg for Pb, Cu and Zn, respectively (Chaudhuri, Nath, & Birch, 2014). *A. alba* in Cochin, India was observed to accumulate Cu, Pb and Zn in its roots as much as 53.6, 3.3 and 1.0 mg/kg (Harish & Murugan, 2011). This concentration is also higher than for *A. alba* in the Porong River estuary. The heavy metal content in sediments in Cochin, India is also higher than in the Porong River estuary. It can be concluded that bioavailability of metals in sediments seem to be in line with their concentration in the mangrove plant.

As seen in Table 3, there are similar trends for accumulation of Pb, Cu and Zn in Porong River estuary sediments in mangrove roots and leaves. The highest concentration in leaves were of Zn, followed by Cu and Pb. This trend is similar to that reported in another study (Mn>Cr>Zn>Ni>Cu>Pb>Cd) (Lotfinasabasl & Gunale, 2012). The difference among concentration of the metals Pb, Cu and Zn in the roots and leaves of the mangrove species found in the Porong River estuary is illustrated in Figure 3.

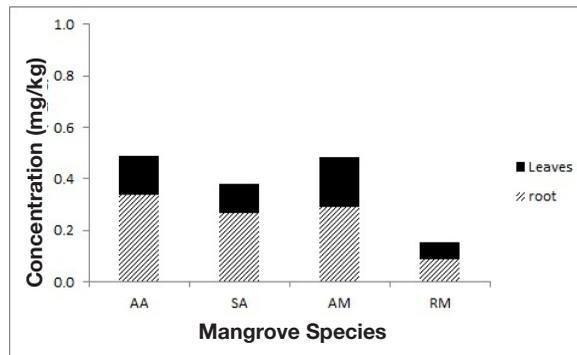
Among the four studied mangrove species, a greater amount of Pb is

accumulated in the roots of *A. alba*, *A. marina* and *R. mucronata*. The genus *Avicennia* is proven to accumulate a large amount of heavy metals including Pb, Cu and Zn (Harish & Murugan, 2011). Only the species *S. alba* distributes Pb among its leaves more than among other parts. Genus *Sonneratia* readily translocates Pb and Zn to the upper parts such as the leaves (Lotfinasabasl & Gunale, 2012). In contrast, Cu is primarily accumulated in the roots of all the species found in the Porong River estuary, while the concentration of this metal in the leaves is approximately 1.5-2 times lower than it is in the roots. Zn is accumulated more in the leaves of the species *A. alba*, *S. alba* and *R. mucronata*. However, *A. marina* preferred to accumulate Zn more in its roots compared to in its leaves. In line with this, another study found that the highest concentration of Zn in *A. marina* from mangroves in the Red Sea was also in the roots, approximately 36.8 mg/kg (Usman, Alkredaa & Al-Wabal, 2013).

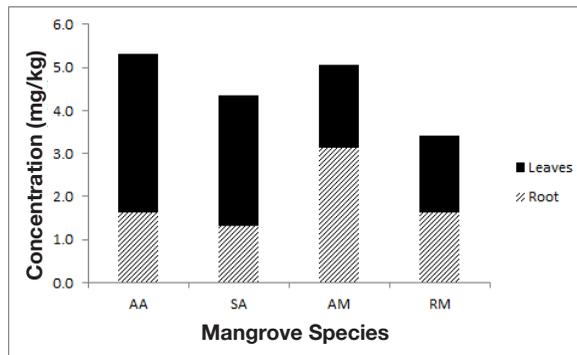
Variability of heavy metal absorption in different parts of mangrove might be influenced by several factors. A previous study reported that the factors of sampling time, tissue age, morphology and physiology may cause varying rates of accumulation in mangrove parts (Lewis, Pryor, & Wilking, 2011).



(a)



(b)



(c)

Figure 3. Comparison between concentration of heavy metals (a) Pb, (b) Cu and (c) Zn in mangrove parts in Porong River estuary (AA : *Avicennia alba*, SA: *Sonneratia alba*, AM: *Avicennia marina* and RM: *Rhizophora mucronata*)

The BCF and TF of Pb, Cu and Zn in Mangrove Plants

To evaluate pollutant absorption capability of mangroves, the bio-concentration factor,

BCF, was used (Wu et al., 2014a). The BCF of four mangrove species is presented in Figure 4. Genus *Avicennia* had the highest BCF in the roots for Pb and Cu. Specifically,

A. alba accumulated significantly higher levels of Pb in both its roots and leaves compared with the other mangrove species. Moreover, this study also revealed that the highest accumulation of Cu in the leaves

was also by *A. marina*. The presence of Cu in leaves is influenced by the metabolic need for essential micronutrients such as Cu (Qiu, et al., 2011). The *Avicennia* genus accumulates Cu in greater amount compared

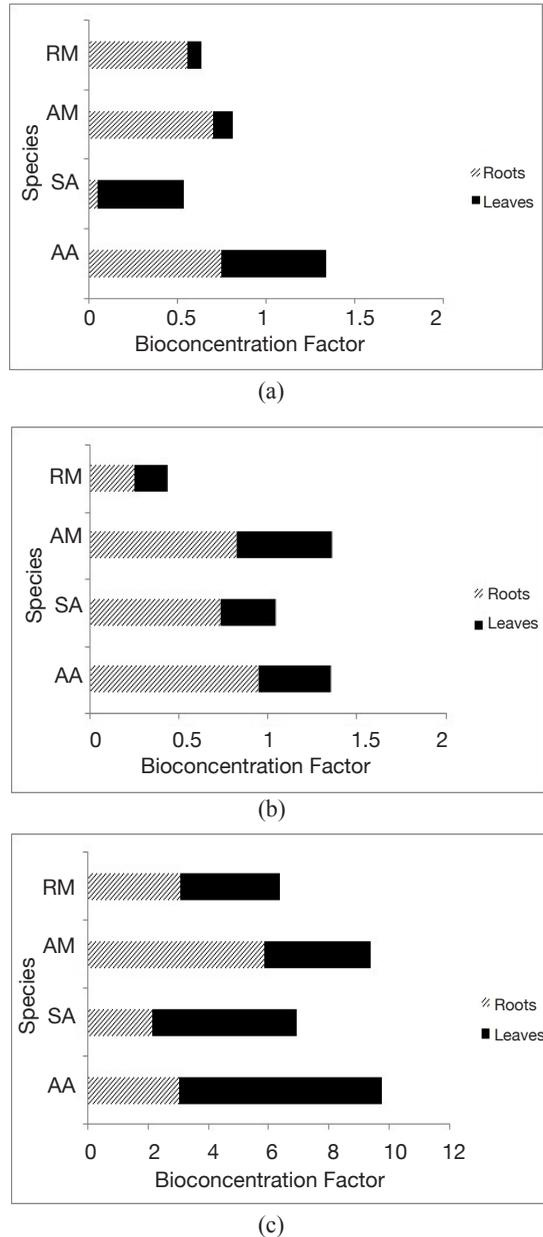


Figure 4. Comparison between bioconcentration of (a) Pb, (b) Cu and (c) Zn in mangrove parts in Porong River estuary (AA: *Avicennia alba*, SA: *Sonneratia alba*, AM: *Avicennia marina* and RM: *Rhizophora mucronata*)

to other mangrove species. This concurs with the findings of Parvaresh et al. (2011), who found that *A. marina* accumulated Cu to a greater extent than did the other species. For Zn absorption, the average BCF at the roots of *A. marina* was considerably higher than in the other species, while in the leaves, it was greater in *A. alba*. Hence, the BCF for Zn of *A. alba* was only slightly higher than the BCF of *A. marina*. However, the BCF for Zn in the mangrove species *R. mucronata* and *S. Alba* was lower than the BCF of genus *Avicennia*.

The BCF for Cu in the mangrove species *Rhizophora stylosa* from Dumbea, New Caledonia in the main roots and leaves was found to be 0.86 and 0.52, while the BCF for Zn was found to be 0.48 and 0.16 (Marchand, Fernandez & Mareto, 2016).

This number is lower than the BCF of Cu in the roots of the genus *Avicennia* from this study. Moreover, the leaves of *R. stylosa* from Dumbea have a BCF of Cu that is higher than the BCF of Cu in the leaves of *A. alba*, *S. alba* and *R. mucronata* in this study. However, the BCF of Zn in the roots and leaves of *R. stylosa* from Dumbea is lower than the BCF of Zn in the roots and leaves of all studied species in the Porong estuary.

TF values indicate heavy metal movement rate (from roots to leaves) throughout the plant. TF of the four Porong River estuary mangrove species for Pb, Cu, and Zn ranged from 0.1375-9.2667, 0.4309-0.7572 and 0.6119-2.2796, respectively (Figure 5). The highest TF for Pb and Zn belong to *S. alba*, while *R. mucronata* had the highest Cu TF value.

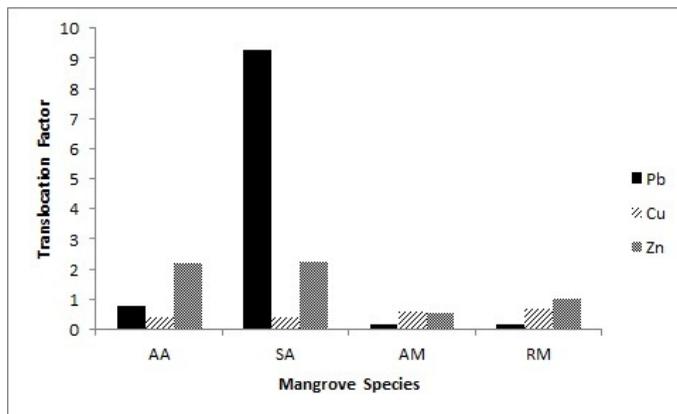


Figure 5. Translocation factor in mangrove species in Porong River estuary

TF in the mangrove species *Sonneratia apetala* and *Cyperus malaccensis* in Nansha Mangrove, South China is 0.205 and 0.110 for Cu and 0.364 and 0.049 for Pb (Wu et al., 2014b). TF for Cu in both mangrove species endorsed by Wu et al. (2014b) is lower than

that of all studied mangroves in this study. However, *S. alba* and *A. alba* have higher concentration of TF for Pb than the species *S. apetala* and *C. malaccensis* in Nansha Mangrove.

The high BCF and TF values indicate that these species are able to accumulate heavy metals effectively. The highest BCF for Pb and Cu was in the roots of *A. alba*. However, the BCF values of these metals were almost less than 1. Moreover, the highest BCF for Zn was in the leaves of the same plant. Thus, it can be concluded that Pb and Cu are not accumulated in the mangrove effectively, while Zn is confirmed to be the metal most accumulated in parts of the mangrove plant.

CONCLUSION

Pb, Cu and Zn concentration in sediment of the Porong River estuary was below the sediment quality guidelines. *A. alba* accumulated high concentration of these metals in both the roots and leaves, recording the highest BCF among all the mangrove species. However, the mobility of Pb and Cu was greater in *S. alba*, while Zn was mostly translocated in *R. mucronata*. The capability of *A. alba* of accumulating Pb, Cu and Zn was significantly higher compared to that of other species in the East Java Porong River estuary, indicating its suitability for phytoremediation of heavy-metal pollution in the study area.

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