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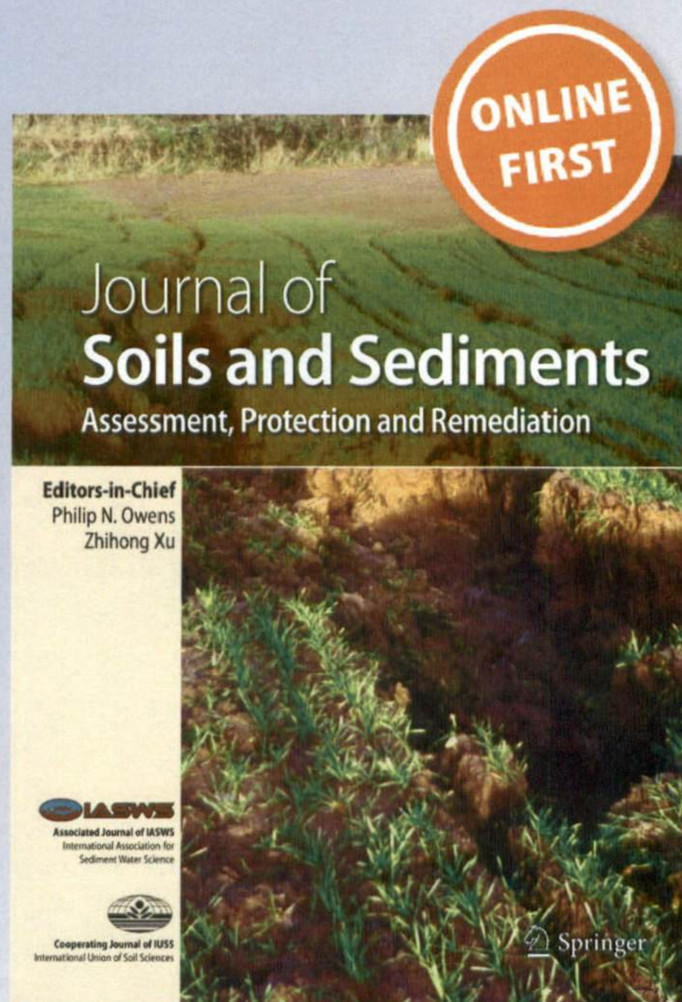


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SEDIMENTS, SEC 4 • SEDIMENT-ECOLOGY INTERACTIONS • SHORT ORIGINAL COMMUNICATION

# Phytoremediation of Pb in the sediment of a mangrove ecosystem

**Annie Melinda Paz-Alberto · Arnel B. Celestino · Gilbert C. Sigua**

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**Abstract**

*Purpose* Coal-fuelled power plants can discharge hazardous materials, particularly heavy metals such as lead (Pb). An alternative way of reducing Pb concentration from contaminated sediments is through phytoremediation. Presently, there are few research findings on the phytoremediation potential of mangroves on metals like Pb. The study was conducted to survey and identify mangroves that thrive near the coal-fired power plant and to assess the phytoremediation potential of mangroves on Pb in sediment.

*Materials and methods* The study sites were located in the mangrove ecosystems of Sitio Oyon and Sitio Asinan in Masinloc, Zambales, Philippines. The first stage of our study was to survey and identify the mangrove species. The second stage was to assess the levels of Pb in the sediments, water, and tissues of mangrove trees. The diversity assessment of the mangrove species was done through the use of $10\times12$ m quadrat technique. Water and sediment samples from each mangrove ecosystem were collected using composite sampling methods.

*Results and discussion* Three mangrove species were identified in the study sites: *Avicennia marina*, *Rhizophora stylosa*, and *Sonneratia alba*. The order of importance of the mangrove trees in the two sampling locations, based on an importance value index (IVI), were as follows: SA (IVI=171.20)>AM (77.79)>RS (51.01). The total uptake of Pb from sediments near the power plants varied significantly ($p\leq0.001$) among the three mangrove species. *S. alba* had the highest Pb uptake of 48.4 kg ha$^{-1}$ followed by *A. marina* (23.1 kg ha$^{-1}$), and *R. stylosa* (2.4 kg ha$^{-1}$). These three mangrove species have the potential to phytoremediate Pb in the sediment.

*Conclusions* The three mangrove species present in the coastal ecosystem near the electric power plant—*A. marina*, *R. stylosa*, and *S. alba*—were potential phytoremediators of sediment Pb. The present study indicated that the mangroves possess beneficial characteristics that remove Pb from contaminated sediments in areas directly affected by coal-fired power plants, and thus have potential phytoremediation properties.

**Keywords** Coal-fired power plant · Heavy metals · Mangroves · Pb · Phytoremediation

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## 1 Introduction

Today, the world is facing greater risks because of heavy emissions from industries that include coal and ore mining activities, mineral smelting, production of chemical-related products such as fertilizers, pesticides, batteries, and paints, manufacturing, and the building of coal-powered electricity plants. Increase accumulation of heavy metals above the normal level found in substrates could result in detrimental impacts on human health as well as the environment (Sigua 2003, 2005, 2010; Sigua et al. 2003). Coal-fired power plants are toxic polluters (Greenpeace Southeast Asia 2005) because they discharge hazardous materials, particularly heavy metals, in the waste stream of the plant that possibly lead to contamination in the neighboring surroundings. Scientific remediation strategies are being established to mitigate the heavy metal pollution in soils, sediments, and waters. Remediation strategies may include dilution techniques, chemical stabilization, soil washing, thermal desorption, and phytoremediation (FFTC 2009).

Currently, the central region of the Philippines accounts for 8.9% of total energy share of the country, and the electricity comes from hydropower, wind power, geothermal, and coal- and diesel-fuelled power plants (EMB and ICETT 2008). Barangay Bani, Masinloc, Zambales has a large coal-fired power plant that was built near the coastal region, where there are marine sanctuaries and coral reserves (Villanueva 2010). The Masinloc coal-fired power plant is one of 12 coal-fuelled power plants actively distributing electricity, and it can supply up to 600 MW of electricity (Alabastro and Jimenez 2008). The Masinloc power plant is reported to have heavy metals and metalloid emissions such as fly ash. The heavy metals detected include arsenic, cadmium, chromium, copper, lead (Pb), mercury, nickel, and zinc. The concentrations of these elements in the fly ash are not significantly higher than those elements found in uncontaminated soil (Alloway 1990, as cited by Brigden and Santillo 2002). However, large quantities of ashes produced in the power plant can cause an environmental threat because of the potential toxic components in the ashes that could leach into the environment (Brigden and Santillo 2002). In recent years, bioremediation technologies are being utilized for soil and water contamination by reducing or eliminating lethal environmental hazards through the accumulation of contaminants and dangerous waste by-products using live organisms. Currently, these methods are used for treating organic and petroleum hydrocarbons because they are cost effective in converting hydrocarbon compounds into safe by-products (Adriano et al. 1999).

Phytoremediation is a particular type of bioremediation that uses plants for the removal or degradation of harmful organic pollutants and toxic metals from soils, sediments, and waters (Schwitzguébel 2001; Paz-Alberto et al. 2007, 2011; Uera et al. 2007). It includes the accumulation of chemicals in plants, degradation of organic molecules, aids in the stimulation of microorganisms in the decomposition, absorption and volatilization, and reduction of the mobility and bioavailability of contaminants in the environment (Adriano et al. 1999). Phytoextraction is one of the basic processes of phytoremediation that uses metal-accumulating plants to take up pollutants from contaminated soils and transfer them to the aboveground parts of plant (Schwitzguébel 2001; Paz-Alberto et al. 2007, 2011; Uera et al. 2007).

Plants such as mangroves, which thrive in marshy ecosystems, have potential phytoremediation properties (Sari et al. 2004; Defew et al. 2005; Pahalawattaarachchi et al. 2009; Nazli and Hashim 2010; Kamaruzzaman et al. 2011; Nirmal et al. 2011). These are salt-tolerant trees that are found mostly in the intertidal areas of tropical and subtropical shorelines. The mangrove community can grow in a broad range of harsh environmental conditions and share unique adaptive features to cope with such situations, including salt excreting leaves, gas exchanging pneumatophores, and the production of viviparous propagules (Primavera et al. 2004). Mangrove ecosystems have important ecological roles such as the prevention of coastal erosion, providing breeding sites for marine organisms, and the reduction of the impacts of tropical storms and tsunamis (Long and Giri 2011). Mangroves have many traditional uses and ecological services; however, the potential of mangroves to take up excessive metals, such as Pb, in the substrate is still unknown. The objectives of this study were (1) to survey and identify mangroves that thrive near a coal-fired power plant and (2) to assess the phytoremediation potential of mangroves on the uptake of Pb contained in the sediment.

## 2 Materials and methods

### 2.1 Survey of plants in the mangrove ecosystem

The study was conducted in the mangrove ecosystems of Zambales, Philippines, during the dry season in January 2012. Study sites were situated in the mangrove ecosystems of Sitio Oyon and Sitio Asinan in Masinloc, Zambales, Philippines (Fig. 1); these areas were selected because of their proximity to the Masinloc Power Plant (Fig. 2). At each site, ten quadrats of $10 \times 12$ m were laid following a completely randomized design, and plant samples were collected from each quadrat. Some selected ecological parameters for the mangrove species (e.g., frequency, relative frequency, density, relative density, dominance, relative dominance, and importance value index) were used to determine the abundance and importance of each mangrove species (Smith and Smith 1998; Paz-Alberto 2005).

### 2.2 Determination of Pb in water

#### *2.2.1 Sampling method*

Water samples in each mangrove ecosystem were collected using a composite sampling technique. To accomplish this, the bottle submersion method was adopted from the protocol of CRL Environmental Corporation Laboratory, Clarkfield, Pampanga, Philippines. Collection of the samples below the water surface was carefully undertaken to prevent disturbance of the underlying sediments. About 100 ml of water were collected. Ten sample bottles were used to randomly collect water samples from each mangrove area. The water sample from each composite location was transferred into a plastic bottle and sealed.

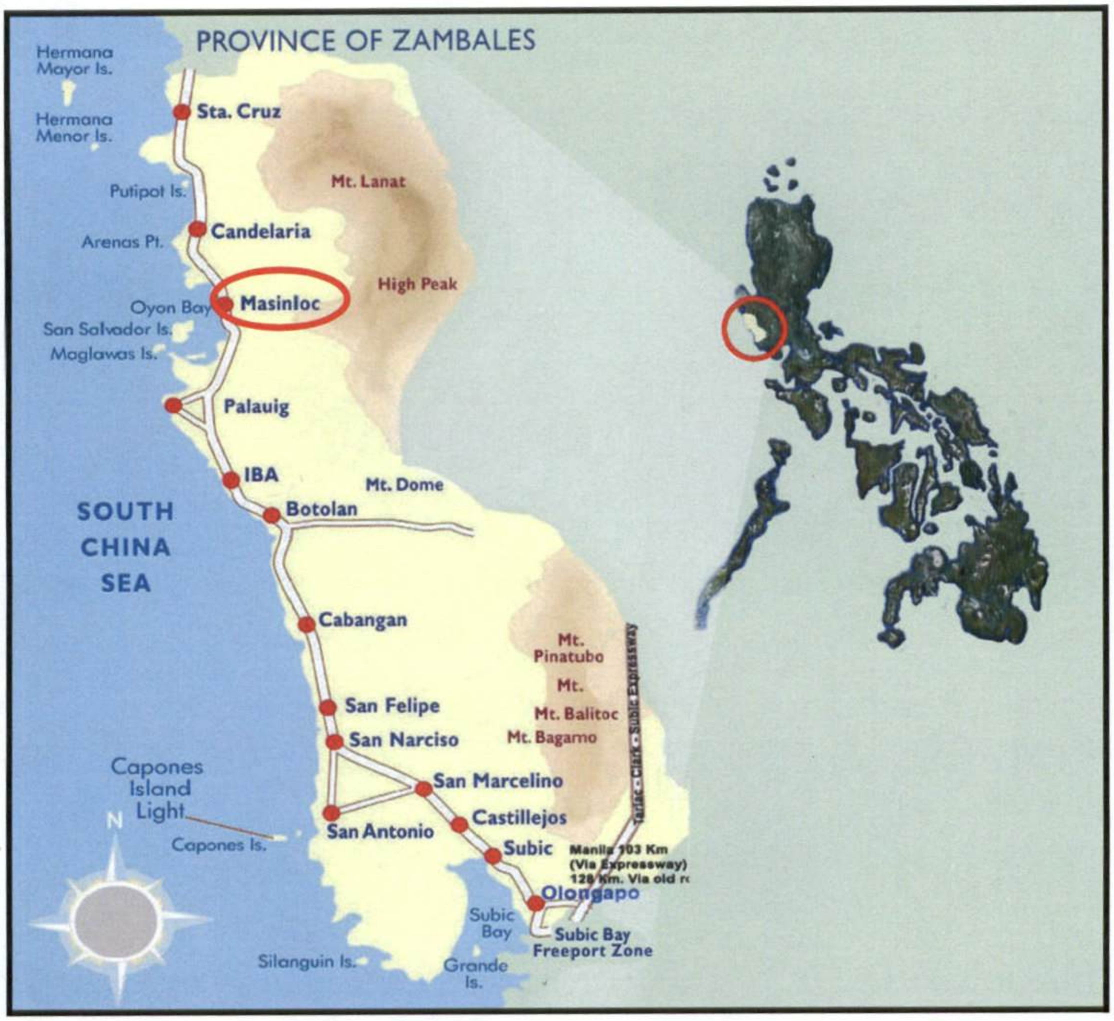


Fig. 1 Location of the study in Zambales, Philippines

### *2.2.2 Preparation and analysis of the water*

Plastic bottles containing marine water samples were stored in an ice chest filled with bagged ice. Concentrations of Pb in water samples were determined using flame atomic absorption spectrophotometer (AAS) at CRL Environmental Corporation, Clarkfield, Pampanga, Philippines.

## 2.3 Determination of Pb in sediments

### *2.3.1 Sampling method*

Sediment samples were collected using a composite sampling technique. The dipper method adopted from CRL Environmental Corporation Laboratory protocol was followed to collect the sediment samples. A stainless steel dipper was used to collect the sediment samples by pushing it firmly downward into the sediment and then quickly lifting it upwards to reduce loss of fine-grained sediment due to the flowing water. One scoop of sediment was collected every quadrat during the survey, totaling to ten scoops per station. Sediment samples from each composite location were transferred into a stainless steel container.

### *2.3.2 Preparation and analysis of the sediment*

Sediment samples were stored in an ice chest filled with bagged ice immediately after the collection. Sediment samples were homogenized prior to analysis. The concentration of Pb was determined using a flame AAS at CRL Environmental Corporation (as above).

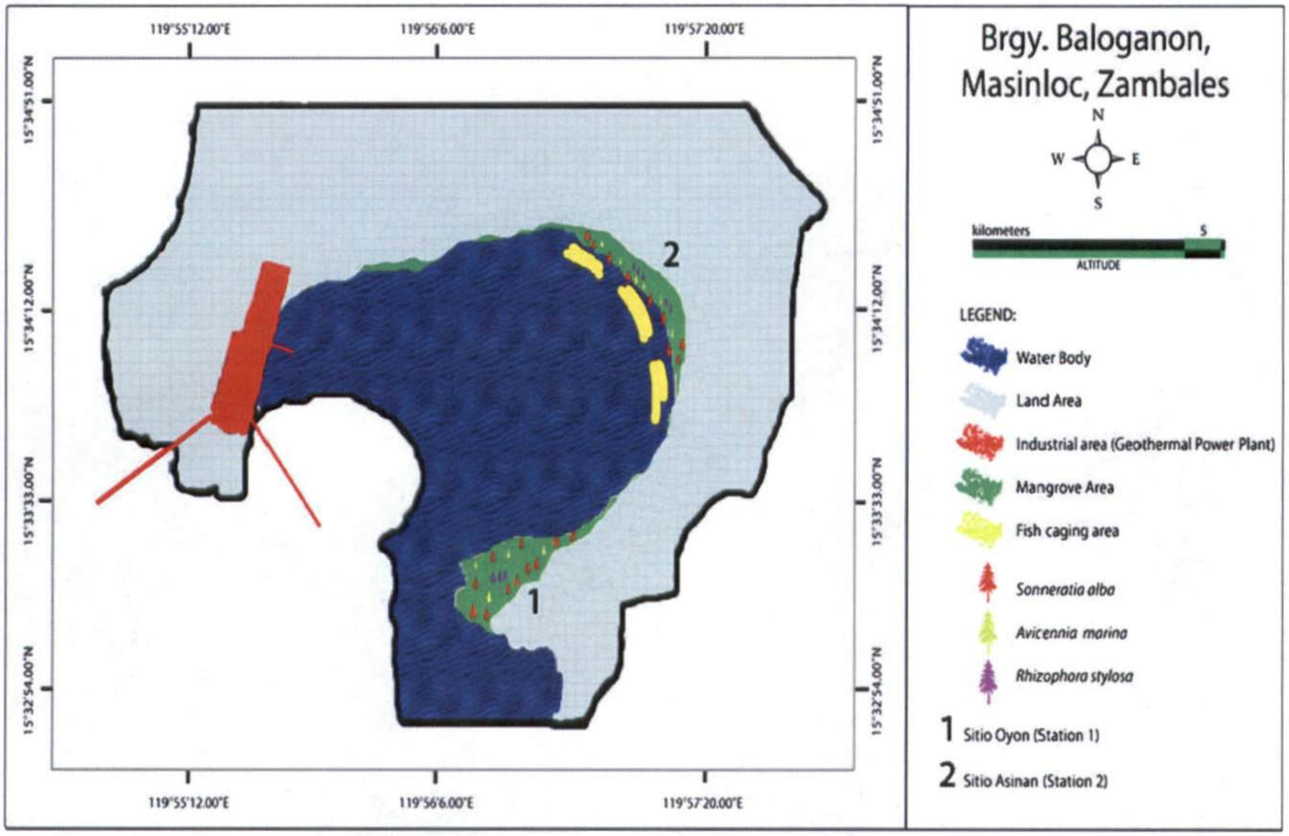


**Fig. 2** Mangrove ecosystems in Sitio Oyon and Sitio Asinan, Zambales, Philippines

### 2.4 Lead analysis of mangrove tissues

#### *2.4.1 Collection of plant parts*

For each of the mangrove species, plant parts—specifically prop roots or pneumatophores with a length of 12 to 18 cm and matured leaves—were gathered randomly, with a total weight of about 200 g per species. The roots were carefully cut and handpicked from above the sediment. The leaves were collected using a pair of scissor (Defew et al. 2005; Nazli and Hashim 2010). The roots and leaves of each mangrove species were washed with tap water (Pahalawattaarachchi et al. 2009). About three to five drops of phosphate-free soap were added and were rinsed with tap water twice followed by deionized water. The plant samples were stored in clean plastic bags.

#### *2.4.2 Plant sample preparation and Pb analysis*

The different plant parts were oven-dried at 80 °C for 4 h. Leaves of the same species were homogenized and pulverized into powder (Pahalawattaarachchi et al. 2009; Nirmal et al. 2011) and were placed in a clean ziplock plastic bag. The same procedures were followed for the root samples. Concentrations of Pb in plant tissues were determined using a flame AAS at CRL Environmental Corporation (as above).

### 2.5 Statistical analyses

Concentrations of Pb in water, plants, and sediments were analyzed by the analysis of variance using PROC GLM model (Statistical Analysis System 2000). Where the F-test indicated a significant ($p \leq 0.05$) effect, means were separated following the method of Duncan's multiple range test, using the appropriate error mean square (SAS 2000).

## 3 Results and discussion

### 3.1 Concentration of Pb in marine waters and sediments

The analyses of marine waters and surface sediments from the two mangrove sites, Sitios Oyon and Asinan, in Barangay Baloganon showed undetectable levels of Pb. Although, the movement of waves near Masinloc Power Plant was in an eastly direction, Pb did not accumulate in the waters and sediments of the mangrove ecosystems. Results of Pb analysis in water and sediment samples from the two sites corroborate with the studies of Zheng et al. (1997), Pahalawattaarachchi et al. (2009), Nazli and Hashim (2010), Parvaresh et al. (2010) and Qui et al. (2011). These researchers reported relatively low concentrations of Pb, and no heavy metal pollution was found in the waters and sediments of the mangrove ecosystems in their study areas. However, those studies indicated that mangrove species such as *Avicennia marina* (Parvaresh et al. 2010), *Rhizophora stylosa* (Zheng et al. 1997), *Rhizophora mucronata* (Pahalawattaarachchi et al. 2009), and *Sonneratia caseolaris* (Nazli and Hashim 2010) possessed the ability to absorb Pb, and that mangroves are able to filter pollutants in the environment.

### 3.2 Concentrations of Pb in plant tissues and total Pb uptake of mangrove species

All mangrove species had accumulated Pb in their plant tissues. Table 1 shows the presence or absence of Pb in the leaves and roots of the different mangrove species, particularly pneumatophores in *A. marina* and *Sonneratia alba* and prop roots in *R. stylosa*. Lead was present in the leaves of *A. marina* and *R. stylosa*, but was absent in the roots. On the contrary, Pb was present in the pneumatophores of *S. alba*, but not in the leaves (Table 1). Table 2 shows the mean Pb concentration in the leaves and roots in the mangrove ecosystems in Sitios Oyon and Asinan. The results show that *A. marina* can accumulate Pb in their leaves, and this finding is similar to the studies reported by MacFarlane and Burchett (2002), Sari et al. (2004), Shete et al. (2007), Parvaresh et al. (2010), and Nirmal et al. (2011). On the other hand, studies by Zheng et al. (1997) and Pahalawattaarachchi et al. (2009) showed that Pb was present in the leaves of *R. stylosa* and *R. mucronata*. Other species of *Sonneratia*, like *S. caseolaris*, revealed that Pb accumulated in the roots and pneumatophores (Nazli and Hashim 2010; Sing 2010), while Pb accumulated in the roots of *Sonneratia apetala* (Shete et al. 2007).

Results revealed that there were significant differences among the plant parts of the different mangrove species from the two sites. The pneumatophores of *S. alba* in Sitio Asinan

**Table 1** Presence or absence of lead (Pb) in different plant parts of mangrove species near the Masinloc power plant, Philippines

| Mangrove Species | Presence or absence of Pb | |
|---|---|---|
| | Leaves | Roots |
| *Avicennia marina* | + | – |
| *Rhizophora stylosa* | + | – |
| *Sonneratia alba* | – | + |

\+ indicates the presence of lead

– indicates the absence of lead

exhibited the highest mean concentration of Pb with 98.5±5.0 mg $kg^{-1}$. The leaves of *A. marina* (63.0±2.8 mg $kg^{-1}$) and *R. stylosa* (20.5±3.5 mg $kg^{-1}$) had the second and third highest mean concentrations of Pb, respectively. The results of Pb accumulation in the pneumatophores of *S. alba* were consistent with the study of Shete et al. (2007). Also, Sing (2010) reported that roots and pneumatophores of *Sonneratia* can immobilize heavy metals, particularly Pb. The Pb accumulation potential of *A. marina* leaves was supported by the study of Parvaresh et al. (2010) and Nirmal et al. (2011). Sari et al. (2004) found that the mobility of Pb in the aerial part, specifically in the leaves, is related to the mechanism of salt accumulation in the salt glands of the leaves. The presence of special salt glands found in the leaf cells of *A. marina* may play an important function in metal translocation because salt glands could stimulate the pumping of ions to the leaf tissues (Drennan and Pammenter 1982; Sari and Din 2012). The salts and ions may have accumulated in the vacuoles and collecting cells before secretion (Drennan and Pammenter 1982; Vasquez et al. 1999).

MacFarlane and Burchett (2000) reported that heavy metals can accumulate in the cell wall of the *A. marina* leaves. The outcome of Pb accumulation in the leaves of *R. stylosa* (Table 2) is corroborated by Zheng et al. (1997), who reported that the absorption of Pb was higher in the mature and senescing leaves of *R. stylosa*. Moreover, Pahalawattaarachchi et al. (2009) confirmed that Pb accumulated in the leaves and barks in *R. mucronata*, where Pb was translocated in the mature and senescing leaves. In terms of the overall Pb absorption, *A. marina* obtained higher Pb concentrations in the leaves than *R. stylosa* (see Table 2). This finding was similar to the studies of Cheng (2003). The location of the mangrove species could be a factor in Pb absorption. Results showed that the mangrove species near the source of pollution tended to accumulate a much higher concentration of Pb. In Sitio Asinan, *S. alba* and *A. marina* absorbed greater amounts of Pb due to their proximity to nearby major roads and households. Cheng (2003) stated that heavy metal can be absorbed by plants using their roots, stems, and leaves, which were later stored in the different plant parts. Moreover, the distribution and accumulation of heavy metals depend on plant species, heavy metal concentration, and exposure duration.

The total uptake of Pb from sediments near the power plants varied significantly among the three mangrove species (Table 2). The highest average uptake of 48.4 kg $ha^{-1}$ was observed for *S. alba* followed by *A. marina* with an average uptake of 23.1 kg $ha^{-1}$. *R. stylosa* had the lowest average Pb uptake of 2.4 kg $ha^{-1}$. Mangrove plants develop highly specialized root system for adaptation to littoral habitats. The architecture of plant root systems affects the numerous functions carried out by the roots. Studies have shown close

**Table 2** Importance value index, mean concentration of lead (Pb), and Pb uptake in the mangrove ecosystem

| Mangrove Species | Importance value index (RF+RD+RDo)** | Mean concentration of Pb in plant tissue (mg $kg^{-1}$) | Mean uptake of Pb*** (kg $ha^{-1}$) |
|---|---|---|---|
| *Avicennia marina* | 77.79b* | 63.0±2.8b (leaves); 0 (roots) | 23.1±2.5b |
| *Rhizophora stylosa* | 51.01c | 20.5±3.5c (leaves); 0 (roots) | 2.4±0.1c |
| *Sonneratia alba* | 171.20a | 98.5±5.0a (roots); 0 (leaves) | 48.4±3.0a |

*Means with the same letter superscript are not significantly different at 5% level by DMRT

***RF* relative frequency, *RD* relative density, *RDo* relative dominance)

***Uptake of Pb=(mean concentration of Pb, mg $kg^{-1}$) × (average dry matter yield per quadrat, 120 $m^2$) × (1 kg/1,000,000) × (10,000 $m^2$ $ha^{-1}$)

relationships between root architecture and functions, such as anchorage and acquisition of water and minerals (Stokes et al. 1996). *S. alba* (Sonneratiaceae) has quite complicated root systems that are composed of four types of cable roots, pneumatophores, feeding roots, and anchor roots (Purnobasuki and Suzuki 2004). Pneumatophores of *Sonneratia* species have well-developed root systems which undergo secondary growth (Sun et al. 2004) and have major physiological functions for water and nutrient conduction (Suzuki et al. 2002; Sun et al. 2004). Sun et al. (2004) studied the role of the pneumatophores of *S. alba*, and found that the vessels of secondary xylem were capable of water conduction. Another role of xylem ray is for storage of ergastic substances (Sun et al. 2004). Therefore, the complicated structure of the root system of *S. alba* suggests that it can absorb and accumulate chemical compounds such as Pb in its root tissues.

The root structure is of special interest in the absorption of heavy metals. Roots can absorb, store, and/or metabolize heavy metals contaminants (Tangahu et al. 2011). *A. marina* also develops a fairly complex root system that is quite similar to *S. alba* (Purnobasuki and Suzuki 2005), but due to its unique characteristics of having salt glands on its leaf cells, it has the ability to accumulate Pb in its leaves rather than in its roots and pneumatophores. The mobility of Pb in the leaves may be due to an increase in transpiration because of exposure to high salinity, leading to a higher flux of metals in the leaves (Otte 1991). Moreover, Pb accumulation in the leaves of *A. marina* may also be attributed to the physiological mechanism on the maintenance of osmosis potential in the cells for its adaptation to saline conditions (Sari and Din 2012). The system and process of ion uptake and transport, as well as salt secretion by the special glands, allow the mangrove plants to cope with high salinity levels while retaining water levels and cell turgor (Clough et al. 1982; Suarez et al. 1998). On the other hand, *S. alba* leaves did not absorb Pb because of the absence of salt glands. This could also be due to the brittle and succulent structure of its leaves as a coping mechanism to retain large quantities of water in the leaves in order to avoid the toxic effects of salts. Mobility and accumulation of Pb in the roots and leaves depend on the mangrove plant species and could be influenced by the physiological mechanism in relation to the adaptation of mangroves to saline environment. Results of this study clearly show that *S. alba*, *A. marina*, and *R. stylosa* are potential phytoremediators of Pb in the mangrove ecosystems.

## 4 Conclusions

Results showed that the three mangrove species present in the coastal ecosystem near the Masinloc electric power plant—*A. marina*, *R. stylosa*, and *S. alba*—were potential phytoremediators of Pb contained in the sediment. Mangroves thriving in marshy ecosystems have potential phytoremediation properties because they act as pollutant eradicators, which are critical for the protection of adjacent coastal ecosystems and nearby communities from the potential buildup of Pb in sediments. The present study indicated that mangroves possess many beneficial characteristics to remove Pb from contaminated sediments in areas directly affected by coal-powered electric plants, and thus local and regional policies on the conservation of mangroves should be formulated.

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