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Accumulation of lead and copper in *Rhizophora apiculata* from Setiu mangrove forest, Terengganu, Malaysia

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Abstract: *The accumulative partitioning of Pb and Cu in the Rhizophora apiculata was studied randomly in the Setiu mangrove forest, Terengganu. Samples of leaves, barks and roots were collected randomly from the selected studied species. Sediments between the roots of the sampled mangrove plants were also collected. The results from analysis for Rhizophora apiculata shows that the concentration of Pb and Cu were accumulated higher in root tissue compared to bark and leaf tissue but lower than surrounding sediment level. The average concentration of Cu for Rhizophora apiculata in leaf, bark, root and sediment was 2.73 , 3.94 , 5.21 and 9.42 mg l^{-1}, respectively. Meanwhile, the average concentration of Pb in leaf, bark, root and sediment was 1.43 , 1.38 , 2.05 and 11.66 mg l^{-1}, respectively. Results of concentration factors (CF) show that the overall the concentration of Pb and Cu were accumulated much higher in roots system of Rhizophora apiculata.*

Key words: *Setiu mangrove forest, Rhizophora apiculata, Lead, Copper*
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Introduction

Mangrove ecosystems are highly productive and play a vital role as a major primary producer within estuarine systems. The uniqueness of *Rhizophora apiculata* root systems serve as habitat and nursery area for many juvenile fish and crustaceans, which have both direct and indirect socio-economic importance and are of great importance to many scientific studies. They also provide erosion mitigation and stabilization for adjacent coastal landforms (Harty, 1997). In a plant-soil system, strong absorption and fixation of heavy metals by soil can easily cause residual accumulation in the soil, resulting in over-absorption of heavy metals by growing plants (Lian *et al*., 1999; Rajkumar *et al.,* 2007). These plant products are harmful to the health of humans (Defew *et al*., 2005). For this reason, it is important and necessary to study the relation between the content of heavy metals in soil and the absorption and accumulation by plants (Lian *et al*., 1999).

Little is known about the specific effects and bioaccumulation mechanisms of heavy metals in mangroves, although it is generally considered that mangroves show the ability to accumulate metals and possess a certain tolerance to relatively high levels of heavy metal pollution (Thomas and Eong, 1984). In Malaysia, pyroligneous acid of *Rhizophora apiculata* species has been used for ages as sterilizing agent, deodorizer, fertilizer, antimicrobial and growth promoting agent (Loo *et al*., 2006). Cu are essential plant micronutrients, and their uptake and allocation to plant organs such as photosynthetic leaf tissue is high and active. Conversely, Pb is both non-essentials to plant growth and of very low solubility. Its uptake is passive, and its translocation from roots to other plant organs is generally low (Baker and Walker, 1990; De Lacerda and Abrao, 1986).

Generally, accumulation does occur at the root level, with restricted transport to aerial portions of the plants. These indicate that plants actively avoid the uptake of trace metals. Many studies had been carried out on various plants to determine its heavy metal accumulation capability (Silva *et al*., 2006; Yu *et al*., 2007). They play an important role as a filter and natural pollution treatment centre because of the specialty of its root system thant manage to control the water quality and trap the sediments as well as particulates which are transported by the current into the oceans from the estuaries (Clark *et al*., 1998). In view of the importance of the mangrove to various aspects of the environment, research on the concentration of lead (Pb) and copper (Cu) in leaf, bark and root in mangrove plants of *Rhizophora apiculata* species as well as distribution of two heavy metals in sediment was carried out.

Materials and Methods

Sampling sites: Sampling was conducted at Setiu mangrove forest, which is located 20 km to north of Kuala Terengganu, Malaysia and facing the south China Sea. The study area has a diverse ecosystem with various natural resources, vast array of biological diversity and coastal and riverine fishing activities (Fig. 1). Samples were collected at the identified area where the species of interest can be found. Coordinates of the location are to be recorded prior to the sample collection. The samples are to be taken at the same time and

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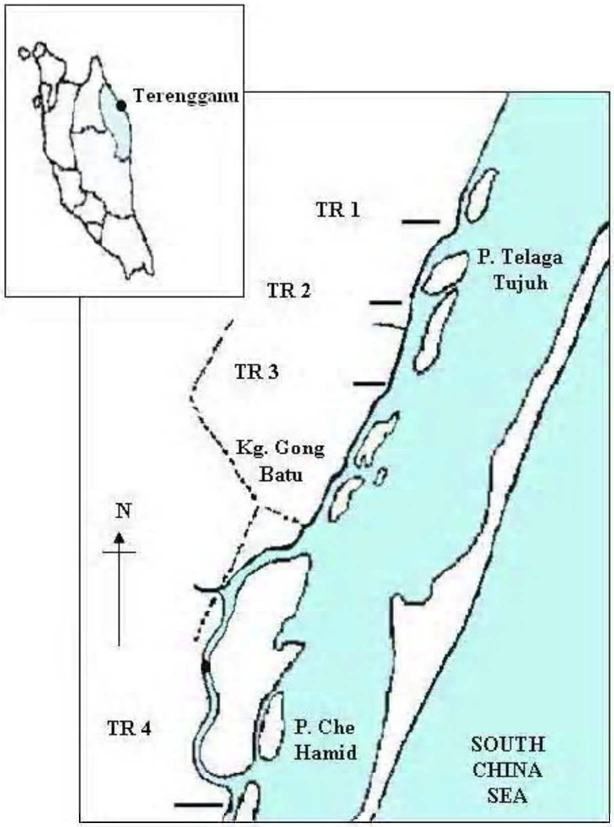


Fig. 1: Location of the study area of Balok mangrove (TR1, TR2, TR3 and TR4) where leaves, barks, roots and sediment were collected randomly from *Rhizophora apiculata*

be picked randomly to get variety of results (TR1, TR2, TR3 and TR4). Samples of the leaves, roots and barks are then put into plastic container before being transported to the lab. Root tissues of approximately 1000 g are sampled from what was considered to be nutritive roots for absorption while larger anchoring roots were avoided. Sediments between the roots of the sampled mangrove plants were also collected. Root, bark and leaves samples were washed with mili-Q grade water before storage. All samples were packed into plastic bags, label and kept frozen until analysis.

Lead (Pb) and copper (Cu) analysis in plant and sediment: Roots, barks and leaves samples was dried at 60°C to constant weight and homogenised. The plant parts were then grounded to a fine powder using a porcelain mortar and pestle. Precautions to prevent contamination will be given a priority. Samples then will be stored in plastic vials with labels and will be kept in a desiccators. Digestion and analytical procedures for elements Pb and Cu were analyzed using the published methods (Cioni *et al*., 1976; Noriki *et al*., 1980). Two replicates of 200 mg of each tissue samples was ashed at 450°C for 3 hr. The ashed sample then dissolved in 5 ml of 5M Hydrochloric acid and 1 ml of concentrated nitric acid. A clear solution with no residue should be obtained at this stage were analyzed for heavy metals using Inductive Couple Plasma-Mass Spectrometry (ICP-MS). Blank and Laboratory standard sample (SRM 1547/peach leaves) were also subjected to same procedure. The recovery test for plants was done using standard analysis and the percentage of recovery ranged from 95 to 105%.

The sediment samples were digested according to the published methods (Kamaruzzaman *et al*., 2001), with some modifications. An inductively-couple plasma mass spectrometry (ICP-MS) was used for the quick and precise determinations of Pb in the digested marine sediment. Briefly, the digestion method involved the heating of 50 mg of a < 63 µm size sample in a sealed teflon vessel with a mixed concentrated acids of HF, HNO_3 and HCl in the ratio of 2.5 : 3.5 : 3.5. The teflon vessels were kept at 150°C for 3-5 hr. After cooling, a mixed solution of boric acid and ethylene diamine tetraacetic acid (EDTA) was added and the vessel was again heated at 150°C for at least 5 hr. After cooling to room temperature, the content of the vessel was thoroughly transferred into a 10 ml polypropylene test tube and was diluted to 10 ml with deionized water. A clear solution with no residue should be obtained at this stage. Blank sample (sample without sediment) and laboratory standard sample (SRM 1646 "Estuarine Sediment") were also subjected to same procedure. The precision assessed by replicate analyses was within 3%. The accuracy was also examined by analyzing, in duplicate a Canadian Certified Reference Materials Project standard (DL-1a) and the results coincided with the certified values within a difference of ± 3%.

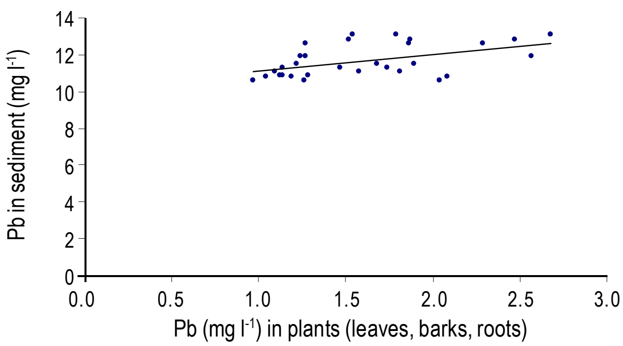
Statistical analysis: In order to compare the degree of storage of the metal, concentration factors (CF) were calculated as concentration of metal g^{-1} tissue over the concentration of metal g^{-1} sediment (Baker and Walker, 1990; De Lacerda and Abrao, 1986). Statistically significant differences between the four transects were also assesed using two-way ANOVA. Pearson product correlation coefficients between heavy metal in plants tissue and in sediments were also computed.

Results and Discussion

Cu and Pb accumulation detected in both *Rhizophora apiculata* varies in the study area of Setiu, Terengganu. Heavy metals concentrations were detected in the mangrove parts prove that Cu and Pb were distributed in all mangrove parts. From the observation, *Rhizophora apiculata* accumulated higher concentration of heavy metals in the root system compared to the other parts of plant. The results from analysis of *Rhizophora apiculata* shows that Cu was accumulated higher in root tissue compared to bark and leaf tissue but lower than surrounding sediment level. The average concentration of Cu in leaf, bark, root and sediment were 2.73, 3.94, 5.21 and 9.42 mg l^{-1}, respectively. It is interesting to note that the concentration of Pb was also higher in the root tissue compared to bark and leaf tissue but lower than surrounding sediment level. For *Rhizophora apiculata*, the average concentration of Pb in leaf, bark, root and sediment were 1.43, 1.38, 2.05 and 11.66 mg l^{-1} respectively. Pb and Cu concentration in each plant parts and surrounding sediment were shown in Table 1. In this study, it was observed that there is a positive correlation

Table - 1: Average concentration of Pb and Cu of *Rizhophora apiculata* in leaves barks, roots and surrounding sediment

Sample	Cu (mg l^{-1})	Pb (mg l^{-1})
Leaves	2.73	1.43
Barks	3.94	1.38
Roots	5.21	2.05
Sediment	9.42	11.66



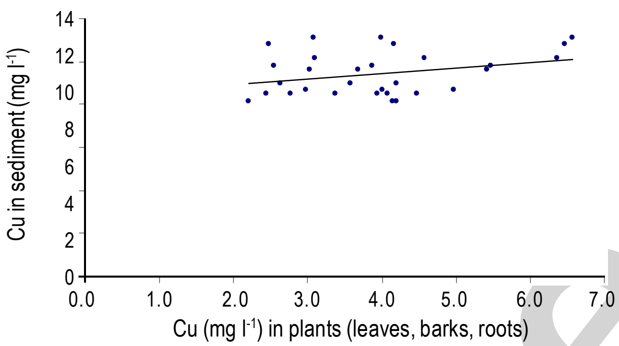


Fig. 2: A positive correlation between sediment with leaves, barks and roots in both Cu and Pb

between heavy metals in sediment and the concentrations of Pb and Cu in the plant parts (Fig. 2), suggesting the influence of the heavy metals in the plant parts. However, the concentration distribution in plant parts like leaves, bark and roots may vary depending on the concentration of heavy metals in the sediment, the types of heavy metals and also the tolerance of the species and its parts towards the heavy metals (Baker and Walker, 1990; De Lacerda and Abrao, 1986).

Overall, in Setiu mangrove areas, Pb and Cu are found to have higher concentration in the sediments. The increasing trend in their concentrations especially near the estuary reflects their predominant lithogenous origin. Furthermore, fine-grained sediments in the mangrove and nearby area are characterized by higher concentrations of these trace elements compared to the coarse-grained sediments of the inner part of the rivers. As reported by many scientists (Kamaruzzaman *et al.*, 2008; Kamaruzzaman and Ong, 2008), there is a positive correlation between grain size and the concentrations of Pb and Cu, suggesting the influence of the fine fraction in their incorporation into the sediments. For the sediments that were further away from the estuary, are characterized by the lower ratios of Pb and Cu, with the higher mean sediment size.

Results of concentration factors (CF) show that *Rhizophora apiculata* contained significantly less Pb, containing only 0.96 times sediment Pb levels. The increased accumulation of metals to plant tissues is thought to be achieved through the translocation of air absorbed through lenticels in pneumatophores to underground roots. This creates oxidized rhizospheres within the anaerobic soil environment, a reduction in complexing sulphides, a lowered stability of iron plaques, and a consequent higher trace metal concentration in the exchangeable form (MacFarlane *et al.*, 2003; Marchese *et al.*, 2008). Spatially, ANOVA test shows no significant differences were found in accumulation of Pb and Cu ($p>0.05$). Pearson product correlation coefficients shows increases in Pb and Cu in the sediment may increase the accumulation of Pb and Cu level in the plant tissues ($r = 0.4911$ and $r = 0.3215$, respectively).

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