

The Clustering, Distribution, Bioaccumulation and Translocation as an Ability of Mangrove Vegetation to Reduce Impact of Heavymetal (Hg, Cd and Zn) Pollution

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Abstract

Mangrove has ability to reduce impact of heavy metal contaminant released from industry and domestic waste. . Therefore, this study aimed to analyze interpolation and clustering of heavy metal accumulation and traslocation in mangrove ecosystem.. The results showed the accumulation of Mercury in water, stem and bark, leaves, and roots were 0.1347 ± 0.0212 ppm, 0.0137 ± 0.0137 ppm, $0.0110\text{--}0.0640$ ppm, $0.0020\text{--}0.0120$ ppm, and $0.0260\text{--}0.0690$ ppm. Cadmium accumulations between 1.6048 ± 0.3595 ppm, 0.0292 ± 0.0047 ppm, $0.0790\text{--}0.2650$ ppm, $0.0130\text{--}0.0480$ ppm, and $0.1560\text{--}0.2300$ ppm. Furthermore, Zinc had score that were 35.1746 ± 5.4856 ppm, 0.0550 ± 0.0095 ppm, $10.3250\text{--}32.4490$ ppm, $7.1200\text{--}23.4150$ ppm, and $16.7170\text{--}39.1200$ ppm. However, bioaccumulation factors of Hg ranged between $0.0210\text{--}0.4751$, Cd $0.0105\text{--}0.1541$, and Zn $0.2807\text{--}1.2506$ with the translocation factors of $0.0459\text{--}1.0547$ (Hg), $0.0642\text{--}1.3855$ (Cd), and $0.4086\text{--}1.3057$ (Zn). The mangrove landscape showed that of *Sonneratia alba* (best of Hg accumulation), *Rhizophora mucronata* (best of Cd accumulation), *Avicennia marina* (best of Zn accumulation)

Introduction

Mangrove ecosystem has ability to absorb, filter, bind and trap heavy metals from industrial (Hilmi et al. 2017, Syakti et al. 2013) sediments, garbage, and household waste (Utami et al. 2018). Mangrove also has adaptation pattern to grow and survive in the polluted area, including the activity of specific root and metabolism system (Hilmi et al. 2017, Hilmi et al. 2017). The metabolism of accumulation and translocation can be used to reduce impact of heavy metal contaminant following the activity of nutrient absorption metabolism (Hilmi et al. 2017 and Hilmi et al. 2017b, Deri and Alirman 2013, Wolswijk et al. 2020).

Mangrove ecosystem in Eastern Segara Anakan is a specific lagoon that takes freshwater supply from many rivers, including the Donan river (Fachrul et al. 2011, Hilmi et al. 2021, Hilmi 2018, Umroh et al. 2016) and seawater supply from the Indian Ocean. Eastern Segara Anakan Lagoon (E-SAL) also be used as an suitable area for waste disposal due to industrial, and antropogenic activities (Prastyo et al. 2017). The industrial and anthropogenic activities in coastal and estuary ecosystems, increase heavy metal pollutants supply, specifically Mercury (Hg), Zinc (Zn), Lead (Pb), and Cadmium (Cd) (Cao et al. 2020, Hidayati et al. 2011, 2014, Hilmi et al. 2017, Kibria et al. 2016, Xin et al. 2014). Basically, heavy metal contaminants have characterize the non-degradable properties, the quickly accumulate in the substrates, sediment, and water and give negatively affecting of aquatic organisms life. Heavy metal contaminant also may cause death, litter fall, and stunted growth in trees (Costa-Böddeker et al. 2020, de Almeida Duarte et al. 2017, Melville et al. 2009, Shi et al. 2020) because the characteristics of their toxicity, abundance, persistence, and bioaccumulation (Jeong et al. 2021). According to Marambio-Alfaro et al. (2020) the metal accumulation of Cu, Pb, Zn, Ni, and Cd give negatively affects organism growth. Similarly, Cao et al. (2020) also established that heavy metal pollution give high risks for fish farms and coastal stabilization.

The first is Mercury (Hg) especially methyl mercury (CH_3)-Hg as an industrial and laboratory waste that contains hazardous properties and high toxic levels (Analuddin et al. 2017, de Almeida Duarte et al. 2017, Li et al. 2016, Mapenzi et al. 2020, Shi et al. 2020, Wolswijk et al. 2020a). However, methyl mercury has higher toxic properties and a strong binding capacity than Mercury. Generally, Mercury is a liquid substance at room temperature (25°C) with a freezing point of -39°C and boils at $365,68^\circ\text{C}$. Mangrove wetlands contribute up to 15% of the coastal sediment mercury (Hg) methylation, significantly impacting global Hg cycling (Lei et al. 2019, Li et al. 2016, Shi et al. 2020). Cadmium (Cd) is a silver, soft and shiny white metal that is insoluble, easy to react and produces cadmium oxide. Cd is produced by the cement and coal loading industry (Costa-Böddeker et al. 2020, El-Amier et al. 2017, Kibria et al. 2016, Zhang et al. 2019) and has melting and boiling points of 321°C and 67°C , respectively (Analuddin et al. 2017, Ortega et al. 2017). The last is Zinc as an essential heavy metal is needed by an organism to grow, develop, and support hemocyanin development in blood and enzymatic systems (Machado et al. 2002, Mapenzi et al. 2020, Zhang et al. 2019).

Heavy metal accumulation and translocation (Chai et al. 2020, Hilmi et al. 2017, Jeong et al. 2021, Nadgórska-Socha et al. 2017) of mangrove vegetation occur in the roots, stems, and mangrove leaves (Alzahrani et al. 2018, Costa-Böddeker et al.

2020, Oo et al. 2009). The bioaccumulation process shows absorption, accumulation, and utilization of contaminants from sediments following a root system and surface area (Chai et al. 2020, Ma et al. 2019, Zhang et al. 2019). This activity reduces toxic effects using dilution and translocation to death organs (Alzahrani et al. 2018, Xie et al. 2020) and organic absorption (Nour et al. 2019, R. Xiao et al. 2015). However, translocation refers to moving contaminants between the cell and the vascular tissue to be distributed in the stem, leaves, and other organs. In general, these processes are passive transport systems based on nutrient absorption and distribution (Alzahrani et al. 2018, Chai et al. 2020, R. Xiao et al. 2015).

The interpolation and clustering of Zn, Hg, Cd accumulation explains the relation of mangrove ability to reduce the impact of heavy metal contaminants (Hilmi et al. 2021, Hilmi et al. 2017) and describe the relationship and similarity adaptation models of bioaccumulation and traslocation using an index of Euclidian distance (Hidayati et al. 2011, Hilmi et al. 2015, Syakti et al. 2013). this study aimed to analyze interpolation and clustering of heavy metal accumulation and traslocation in mangrove ecosystem

Material And Methods

Study Area and Time

The study was conducted on 2021 in Eastern Segara Anakan (E-SAL) as a waste disposal place (Hilmi et al. 2019) could be shown in Fig. 1 and Table 1. E-SAL was dominated by *Rhizophora spp*, *Sonneratia spp*, *Avicennia spp*, *Bruguiera spp*, *Aegiceras spp*, and *Ceriops spp* (Hilmi et al. 2017, Hilmi et al. 2021, Hilmi et al. 2021). The samples of mangrove were collected from roots, barks, stems, and leaves (Monteiro et al. 2014). The study area was divided into Kalipanas, Sleko port, crude oil processing, cement, and Sapuregel stations.

Table 1
Research stations

No	Location station	Geography coordinates	
		Latitude (S)	Longitude (E)
1.	Kalipanas	07°42'36,60" S	108°59'43,91" E
2.	Sleko Port	07°43'17,11" S	108°59'31,00" E
3.	Crude oil industry	07°41'48,64" S	108°59'34,98" E
4.	Cement industry	07°40'59,81" S	109°00'40,35" E
5.	Muara Pelawangan Timur	07°43'40.87"S	108°59'03.31"E

Research procedures

Sample collection s

a. Water and sediment sample's

A total of 600 mL of water samples from the lagoon were placed into a bottle and labeled. The addition of 0.75 mL concentrated HNO₃ to reduce the pH to 2 (Hilmi et al. 2017, Mapenzi et al. 2020, Prastyo et al. 2017). Furthermore, 250 g of sediments were collected using Eckman grab at a depth of 50 cm from the soil surface. They were then placed into a plastic bag and labeled (Hilmi et al. 2017, Mapenzi et al. 2020, Prastyo et al. 2017), then moved into an icebox.

b. Vegetations samples

A total ≥ 150 grams of mangrove leaf samples were extracted from dark green leaves. A similar amount of its stem and bark were also collected using the destructive method. However, the same number of samples were taken from pneumatophore

roots in a minimum water depth of 10–20 cm. All of them were put into a plastic bag, labeled, and then into an icebox (Hilmi et al. 2017).

Heavy metal analysis

a. Hg (mercury)

Mercury was analyzed through the Spectrophotometric method, which involves the Shimatsu brand with an accuracy level of 2×10^{-4} ppm. The mangrove leave stems and roots extraction filtrates were mixed with 10 ml of H_2SO_4 , 2 ml of $KMnO_4$ 2%, 1 ml of $K_2S_2O_8$, and 1 ml of stannous chloride ($SnCl_2$) 10%, then extracted using liquid tetra dithizone. Hg was measured at a wavelength of 480 nm and analyzed using the mercury analyzer (SP-3D) method. The acid was intended to convert: $Hg^{2+} + SnCl_2 \rightarrow HgO$ in cold vapor using the Hg Detector analyzer (SNI, 2009).

b. Cd (Cadmium)

The potentials of Cadmium were measured by the Flame Atomic Absorption Spectrometric method using a set of AAS tools with a precision level of 2×10^{-4} ppm. The extraction filtrates were sucked with a respirator tube of 20 ml and put into a nebulizer, followed by fogging and evaporation. The vapors formed were burned with an open flame and atomization, then irradiated by cathode rays of specific wavelengths. Cadmium was determined at a wavelength of 228.8 nm and 10 mA, where the detector captured the light absorption results. The absorbance scoring of the sample and the standard solution appeared on the AAS screen with the line equation (SNI, 2009)

c. Zn (Zinc)

Zinc was analyzed using the Spectrophotometry method (Shimatsu brand with a level of accuracy of 2×10^{-4} ppm). The extraction filtrate from mangrove leaves, stem, bark, and root samples were first reduced by mixing 10 mL H_2SO_4 , 2 mL 2% $KMnO_4$, 1 mL $K_2S_2O_8$, and 1 mL stannous chloride ($SnCl_2$) 10%, then extracted with tetra dithizone liquid. This was meant to change: $Zn^{2+} + SnCl_2 \rightarrow ZnO$ into cold vapor by the Zn detector analyzer (SNI, 2009).

Data analysis

Distribution of accumulation, bioaccumulation, and translocation factor

The distribution of a heavy metal accumulation and translocation from mangrove species, soil, and water was conducted using standard deviation and average. Bioaccumulation factor (BAF) and translocation factor (TF) of Hg, Cd, and Zn were constructed by the equation of (Hilmi et al. 2017, Lin et al. 2021, Syakti et al. 2013).

$$BAF = \frac{\text{heavymetalaccumulationofmangroveorgans} \left(mg kg^{-1} \right)}{\text{heavymetalaccumulationofmangrovesoil} \left(mg kg^{-1} \right)}$$

$$TF = \frac{\text{heavymetalaccumulationofmangroveorgans} \left(mg kg^{-1} \right)}{\text{heavymetalaccumulationofmangroveroots} \left(mg kg^{-1} \right)}$$

Bioaccumulation *Factor* (BAF) categories included (Ashraf et al. 2011, Hilmi et al. 2017, MacFarlane et al. 2003, Marambio-Alfaro et al. 2020).

$BAF \leq 1$ refers to vegetations less or unable to accumulate heavy metal pollution

$BAF > 1$ stands for vegetations that can accumulate heavy metal pollution.

Translocation *Factor* (TF) categories comprised (Ashraf et al. 2011, Hilmi et al. 2017, MacFarlane et al. 2003, Marambio-Alfaro et al. 2020).

$BAF \leq 1$ represents vegetations less or unable to translocate heavy metal pollution in other organs

$BAF > 1$ refers to vegetations that can translocate heavy metal pollution in other organs

Clustering Mangrove

In the following stages, cluster analysis was built using similarity and dissimilarity through euclidian distance analysis (Hilmi et al. 2021, Hilmi et al. 2021).

Stage 1.

$$ED_{jk} = \sqrt{\sum_{i=1}^s (x_{ij} - x_{ik})^2}$$

Stage 2.

$$D(j, k)h = \alpha_1 D(j, h) + \alpha_2 D(k, h) + \beta D(j, k)$$

Stations	2	3	4	...	22
1	ED ₁₂	ED ₁₃	ED ₁₄		
2		ED ₂₃	ED ₂₄		
3			ED ₃₄		
			...		
22			ED ₂₂		

Notes:

Edjk : Euclidean Distance

i : species

Xij : density in station- j

Xik : density in station- k

D : Distance

α1 : 0,625

α2 : 0,625

β : - 0, 25

Mangrove Landscape

A mangrove landscape was developed using accumulation of Hg, Cd, and Zn contaminants and translocate them in barks, stems, roots, and leaves. The mangrove landscape described the distribution and zonation of mangrove species to reduce

impact of heavy metal pollution.

Results

The accumulation of heavy metal

The distribution and interpolation of heavy metal in sediment and water

The accumulation of heavy metal in sediment and water from domestic pollution, sewage treatment, pesticides, mining and pharmaceutical industries (Nour et al. 2019) which give negative impact for population growth and environment (Cao et al. 2020, Choi et al. 2020, Liu et al. 2020). Table 2 and Fig. 2 show the accumulation of heavy metal pollution on the sediment and aquatic ecosystem.

Table 2
Heavy metal accumulation in Segara Anakan Lagoon

Environ ment	Hg (ppm)			Cd (ppm)			Zn (ppm)		
	average	Stdv	status	Avera ge	stdv	status	average	Stdv	status
Sediment	0.1347	0.0212	unpolluted	1.6048	0.3595	polluted	35.1746	5.4856	polluted
Water	0.0137	0.0029	polluted	0.0292	0.0047	polluted	0.0550	0.0095	polluted
Corelation	-0.42008			-0.57603			-0.26915		

The interpolation and distribution of total accumulation of heavy metal in mangrove ecosystem

The species distribution of heavy metal accumulation in mangrove ecosystem was shown in Fig. 3. The data's indicated that *Bruguiera gymnorrhiza*, *Sonneratia alba*, *Avicennia marina*, and *Rhizophora mucronata* had the highest Hg accumulation between 0.0110–0/0640 ppm (stem and bark), 0.0020–0.0120 ppm (leaves) and 0.0260–0.0690 ppm (roots). *Rhizophora apiculata* and *Rhizophora mucronata* had the highest Cd accumulation between 0.0790–0.2650 ppm (stem and bark), 0.0130–0.0480 ppm (leaves). However, *Avicennia marina* and *Sonneratia alba* had the highest Zn accumulation among 10.3250–32.4490 ppm (stem and bark), 7.1200–23.4150 ppm (leaves), and 16.7170–39.1200 ppm (roots).

The other conditions the distribution of heavy metal accumulation can be shown by the value of potential average and deviation standard (Stddev) of heavy metal accumulation. Using this parameters, this research show that the distribution of heavy metal accumulation in mangrove species,can be shown in Fig. 4. The data showed that the average score of heavymetal accumulation (both of Hg, Cd, Zn) higher than the distribution standard

However, the interpolation of heavymetal accumulation in vegetation, sediment and water could be showed on Fig. 5. The data on Fig. 5 showed that Hg in vegetation and water < Hg in vegetation and sediment. The Hg accumulation in vegetation and sediment had potency between moderate until high potential. Similar with Hg accumulation, the potential of Cd and Zn in vegetation and water have potency < the potential of Cd and Zn in sediment. But, the potential of Cd in vegetation and sediment was dominated by very high potential accumulation.

Bioaccumulation factor (BAF) and Translocation Factor (TF) of heavymetal contaminant

Bioaccumulation factor (BAF) of heavymetal contaminant

Bioaccumulation of heavy metal on Table 3 explained that Hg is between 0.0210–0.4751, Cd 0.0105–0.1541, and Zn 0.2807–1.2506. The BAF of Mercury was dominated by *Avicennia marina*, *Nypa frutican*, and *Sonneratia alba*. For cadmium were *Avicennia marina* and *Rhizophora apiculata* and for zinc was *Avicennia marina*. Alzahrani et al. (2018) stated that the mean BCFs values obtained for Cr, Cu, Ni, Pb, and Cd in the mangrove leaves had 0.43, 0.88, 0.47, 1.57, and 0.39. The values in aerial roots were 0.47 for Cr, 0.59 for Cu, 0.49 for Ni, 1.60 for Pb, and 0.23 for Cd.

Table 3
Bioaccumulation factor (BAF) in Mangrove ecosystem

mangrove species	BAF with sedimen								
	Hg			Cd			Zn		
	stem	leaves	root	stem	leaves	Root	stem	leaves	root
<i>Aegiceras corniculatum</i>	0.1399–0.2031	0.0313–0.0420	0.3271–0.3654	0.0566–0.0567	0.0111–0.0112	0.1061–0.1234	0.3507–0.7087	0.2347–0.5436	0.5227–0.7254
<i>Aegiceras floridum</i>	0.1215–0.1641	0.0244–0.0625	0.3323–0.3654	0.0411–0.0412	0.0111–0.1122	0.1100–0.1234	0.3341–0.5699	0.2322–0.4226	0.5561–0.7254
<i>Avicennia marina</i>	0.1538–0.4063	0.0629–0.1143	0.1888–0.5234	0.0677–0.1111	0.0104–0.0172	0.1130–0.1280	0.4893–1.0734	0.3517–0.8993	0.6878–1.2506
<i>Bruguiera gymnoriza</i>	0.2643–0.3321	0.0286–0.0787	0.1266–0.3114	0.0659–0.0962	0.0244–0.0322	0.1499–0.1687	0.4500–0.6765	0.2783–0.3421	0.5991–0.7677
<i>Bruguiera sexangula.</i>	0.1259–0.2355	0.0210–0.0323	0.3049–0.4111	0.0455–0.0676	0.0101–0.0322	0.1071–0.1521	0.4441–0.6222	0.3229–0.3554	0.5075–0.6776
<i>Ceriops tagal</i>	0.1460–0.2551	0.0292–0.0444	0.4552–0.5665	0.0687–0.0887	0.0200–0.0565	0.1743–0.1821	0.7319–0.8211	0.4892–0.5111	0.7749–0.8818
<i>Excoecaria agallocha</i>	0.0843–0.0992	0.0357–0.0555	0.3221–0.3555	0.0770–0.0923	0.0356–0.0565	0.151–0.1711	0.4459–0.5552	0.2843–0.3322	0.6290–0.7212
<i>Hibistus tiliaceus</i>	0.1071–0.1445	0.0487–0.0556	0.4243–0.5111	0.0765–0.0995	0.0113–0.0233	0.1760–0.2112	0.7288–0.8111	0.3719–0.4112	0.7525–0.8112
<i>Melaluca leucadendron</i>	0.2643–0.3112	0.0643–0.0811	0.3114–0.4221	0.1074–0.1233	0.0222–0.0432	0.1499–0.2001	0.7392–0.8333	0.6366–0.7111	0.5991–0.6671
<i>Nypa frutican.</i>	0.2787–0.3214	0.0643–0.1065	0.3048–0.4112	0.1067–0.1110	0.0211–0.0222	0.1541–0.1564	0.2807–0.5433	0.1936–0.2323	0.8211–0.8882
<i>Rhizophora apiculata</i>	0.0857–0.1875	0.0469–0.0571	0.1625–0.4609	0.0831–0.1319	0.0111–0.0175	0.1074–0.1543	0.4236–0.6287	0.2919–0.4739	0.4545–0.8689
<i>Rhizophora mucronata</i>	0.1071–0.1797	0.0214–0.0313	0.3286–0.5391	0.0572–0.1704	0.0105–0.0304	0.1151–0.1230	0.3947–0.7145	0.2564–0.5492	0.4570–0.8721
<i>Rhizophora stylosa</i>	0.1101–0.1406	0.0156–0.0302	0.1232–0.3406	0.0578–0.1001	0.0129–0.0232	0.1010–0.1246	0.4543–0.7075	0.4231–0.5520	0.5353–0.7044
<i>Sonneratia alba</i>	0.1953–0.4571	0.0469–0.0857	0.3048–0.3333	0.0738–0.0963	0.0163–0.0197	0.1280–0.1541	0.4424–0.5858	0.3355–0.4051	0.8211–0.9654
<i>Xylocarpus granatum</i>	0.1002–0.1857	0.0103–0.0286	0.1134–0.2984	0.0786–0.0941	0.0113–0.0185	0.1130–0.1448	0.5523–0.7664	0.2435–0.629	0.5565–0.5870

Translocation Factor (TF)

The translocation factor of heavy metal in mangrove ecosystem showed that Mercury ranged between 0.0459–1.0547, Cadmium 0.0642–1.3855, and Zinc 0.4086–1.3057 (Table 4). The ability of mangrove species in E-SAL showed that highest TF of Mercury were *Avicennia marina* *Sonneratia alba* and *Nypa frutican*. Similarly, the species with the highest TF Cadmium

was *Rhizophora apiculata*, while the highest in Zinc were *Rhizophora styles* and *Xylocarpus granatum*. Alzahrani et al. (2018) reported that the mean values of TF in aerial roots were Cd (2.72) > Cu (1.74) > Ni (1.42) > Pb (1.29) > Cr (0.90).

Table 4
Translocation factor (TF) in mangrove ecosystem

mangrove species	TF					
	Hg		Cd		Zn	
	stem	leaves	Stem	leaves	Stem	leaves
<i>Aegiceras corniculatum</i>	0.4276– 0.5559	0.0855– 0.1283	0.4589– 0.5337	0.0898– 0.1048	0.6170– 0.9770	0.4491– 0.7494
<i>Aegiceras floridum</i>	0.3506– 0.4490	0.1710– 0.1810	0.3342– 0.4337	0.0898– 0.1048	0.6710– 0.7856	0.4481– 0.5826
<i>Avicennia marina</i>	0.3860– 0.8148	0.2090– 0.3330	0.5991– 0.8982	0.0838– 0.1435	0.7114– 0.8295	0.4257– 0.7191
<i>Bruguiera gymnoriza</i>	0.3303– 0.7486	0.0917– 0.1102	0.4397– 0.5200	0.0740– 0.1630	0.6778– 0.7511	0.4645– 0.6004
<i>Bruguiera sexangula.</i>	0.4128– 0.7200	0.0688– 0.1100	0.3420– 0.4249	0.0939– 0.1001	0.7650– 0.8750	0.5210– 0.6363
<i>Ceriops tagal</i>	0.3207– 0.4320	0.0641– 0.0893	0.3941– 0.4001	0.0885– 0.1147	0.7887– 0.9445	0.5232– 0.6313
<i>Excoecaria agallocha</i>	0.2616– 0.3345	0.0576– 0.1109	0.4210– 0.5101	0.0978– 0.2354	0.7090– 0.7888	0.4519– 0.5000
<i>Hibistus tiliaceus</i>	0.2000– 0.2523	0.0534– 0.1147	0.4100– 0.4348	0.0642– 0.0900	0.8210– 0.9685	0.3201– 0.4942
<i>Melaluca leucadendron</i>	0.3000– 0.3889	0.0879– 0.2064	0.5670– 0.7164	0.0921– 0.1482	0.8450– 1.2339	0.9101– 1.0627
<i>Nypa frutican.</i>	0.7985– 1.0547	0.1100– 0.2109	0.5761– 0.6923	0.0767– 0.1442	0.3419– 0.4536	0.2358– 0.3450
<i>Rhizophora apiculata</i>	0.2667– 0.6923	0.1017– 0.3462	0.7627– 0.9557	0.0742– 0.1626	0.7259– 1.3883	0.5002– 1.0427
<i>Rhizophora mucronata</i>	0.3261– 0.3333	0.0580– 0.0652	0.4973– 1.3855	0.0909– 0.2470	0.8193– 0.8637	0.5610– 0.6298
<i>Rhizophora stylosa</i>	0.3420– 0.4128	0.0459– 0.0655	0.4644– 0.6760	0.0767– 0.1038	0.8650– 1.0043	0.5678– 0.7836
<i>Sonneratia alba</i>	0.5859– 1.5000	0.1406– 0.28125	0.5769– 0.6250	0.1058– 0.1538	0.5388– 0.6068	0.4196 – 0.4086
<i>Xylocarpus granatum</i>	0.5423– 0.6224	0.0958– 0.1001	0.4556– 0.6497	0.0712– 0.1279	0.8990– 1.3057	0.8998– 1.0716

Mangrove landscape based on heavy metal accumulation

The mangrove landscape represent the pattern of mangrove zone to accumulate Hg, Cd, and Zn pollution and also showing of mangroves adaption to grow and live in heavy metal polluted areas (Table 5).

Table 5
Mangrove zone base on heavy metal accumulation

Mangrove zone's		
Hg accumulation	Cd accumulation	Zn accumulation
zone 1	zone 1	zone 1
<i>Bruguiera gymnoriza</i>	<i>Avicennia marina</i>	<i>Avicennia marina</i>
<i>Sonneratia alba</i>	<i>Rhizophora Mucronata</i>	<i>Rhizophora apiculata</i>
<i>Nypa frutican.</i>	<i>Rhizophora apiculata</i>	<i>Xylocarpus granatum</i>
zone 2	zone 2	zone 2
<i>Aegiceras corniculatum</i>	<i>Sonneratia alba</i>	<i>Aegiceras corniculatum</i>
<i>Xylocarpus granatum</i>	<i>Xylocarpus granatum</i>	<i>Bruguiera sexanggula.</i>
<i>Avicennia marina</i>	<i>Nypa frutican.</i>	<i>Melaluca leucadendron</i>
	<i>Melaluca leucadendron</i>	<i>Hibistus tiliaceus</i>
		<i>Ceriops tagal</i>
		<i>Rhizophora stylosa</i>
zone 3	zone 3	zone 3
<i>Melaluca leucadendron</i>	<i>Bruguiera sexanggula.</i>	<i>Bruguiera gymnoriza</i>
<i>Bruguiera sexanggula.</i>	<i>Hibistus tiliaceus</i>	<i>Sonneratia alba</i>
<i>Rhizophora stylosa</i>	<i>Bruguiera gymnoriza</i>	<i>Aegiceras floridum</i>
<i>Rhizophora Mucronata</i>	<i>Rhizophora stylosa</i>	<i>Rhizophora Mucronata</i>
<i>Rhizophora apiculata</i>	<i>Aegiceras corniculatum</i>	
<i>Aegiceras floridum</i>	<i>Excoecaria agallocha</i>	
zone 4	zone 4	zone 4
<i>Hibistus tiliaceus</i>	<i>Aegiceras floridum</i>	<i>Nypa frutican.</i>
<i>Excoecaria agallocha</i>	<i>Ceriops tagal</i>	<i>Excoecaria agallocha</i>
<i>Ceriops tagal</i>		

The Clustering of heavy metal accumulation in mangrove ecosystem

Mangrove clustering refers to grouping of mangrove ability to absorb and accumulate heavy metal (Awaliah, 2018, Hilmi et al. 2021, Hilmi et al. 2021) using the *Hierarchical Clustering Methods* and *Nonhierarchical Clustering Method* (Hilmi et al. 2021, Hilmi et al. 2021, Rachmatin, 2014). The clustering of heavy metal accumulation (Zn, Hg and zinc accumulation) shows differences between mangrove species in accumulating pollution (Fig. 6).

Discussion

The distribution and interpolation of heavy metal in sediment and water

The data on Table 2 showed that the accumulations of (1) mercury were 0.1135–0.1559 ppm (sediment) and 0.0108–0.0166 ppm (water), (2) cadmium ranged from 1.25–1.96 ppm (sediment) and 0.0245–0.0339 ppm (water), and (3) zinc

were 29.67–40.66 ppm (sediment) – 0.0455–0.0645 ppm (water). Based on the pollution category (UU no 82, 2001), the Segara Anakan Lagon (SAL) was contaminated by Hg, Cd, and Zn. Nour et al. (2019) records the data of heavy metal accumulation on the Red Sea coast and Egypt which Zn accumulation between 14.94–134.22 ug/g, Cd 0.12–1.25 ug/g, and Pb 3.17–40.25 ug/g. Alzahrani et al. (2018) reports that the mean concentrations of heavy metals in sediments are Cr (46.14 mg g⁻¹ ± 18.48) > Cu (22.87 mg g⁻¹ ± 13.60) > Ni (21.11 mg g⁻¹ ± 3.2) > Pb (3.82 mg g⁻¹ ± 2.46) > Cd (0.75 mg g⁻¹ ± 0.87). According to Choi et al. (2020) potential heavy metal accumulation in Busan city has distribution that are Zn ≥ Pb > Cu > Cr ≥ As > Ni ≥ Cd > Hg. However, Hao et al. (2019) determine that the bioaccumulation of six heavy metals in China's Hainan and Zhoushan coastal regions including Cu, Pb, Zn, Cr, Cd, and Hg. Jeong et al. (2021) states that from the principal component analysis show that Cr, Ni, Cu, Zn, As, and Hg were derived naturally, while Cd and Pb were from anthropogenic sources..

Table 2 also showed that the potential heavy metal in sediment and water ecosystem had weak negatively correlation (a value < 0.5). However, the potential heavy metal in sediment still was higher than in the water ecosystem. According to Xin et al. (2014), 90% of heavy metals contaminants are high deposited in sediments. And Chen et al. (2021) and Barreto et al. (2016) also write that the organic materials and complex compounds were easily bound and deposited in sediments. Similarly, Cao et al. (2020) writes that heavy metal does not correlate with particle size, organic matter, or nutrients. Furthermore, heavy metal pollution give impact for water quality, including biological oxygen demand (BOD), chemical oxygen demand (COD), Oxygen demand (COD), pH, conductivity, total suspended solids (TSS), Kjeldahl nitrogen (TKN), ammonium (NH₄⁺-N), nitrate (NO₃⁻-N), and total phosphorus (TP) (Kibria et al. 2016, Xiao et al. 2019).

Basically, the highest heavymetal accumulation occurred in mangrove vegetation, not in water or sediment, because the mangrove vegetation is used as a pathway to accumulate heavy metal ions from water and sediment (Katipana, 2015). But mangrove has metabolism system to reduce heavy metal accumulation. The mangrove roots can stop transport non-essential matter or to accumulate in other dead organs. This system aims to reduce the transfer of metal concentration mangrove stem and leaves.

The heavy metal contaminant in the sediment and water body is influenced by salinity, chemical oxygen demand (COD), and pH (Fig. 2). The station 3 had the highest COD score than others and is defined as a polluted area (COD < 25 ppm). Mangrove ecosystem as a fragile ecosystem will be influenced by potential of heavy metal contaminant in water and sediment (Alzahrani et al. 2018, Cochard, 2017). But, mangrove species still have an excellent adaptation to reduce potential heavy metal using excretion, excluder and accumulation gland (Dai et al. 2017, Hilmi et al. 2017, Xie et al. 2020)

A disposal waste from oil refinery and cement industry is the main source of mercury contaminant. It is accumulated by a long-time deposition and binding process with organic matter (Sitepu et al. 2011, Wolswijk et al. 2020, Xiong et al. 2018). However, the accumulation in the sediment of E-SAL should be lower than the US EPA standard (< 0.2 mg/Kg). The largest cadmium accumulation in E-SAL comes from oil refinery activity. The oil refinery industry caused long cadmium deposition in the sediment and was distributed by tidal currents into the water column (Analuddin et al. 2017, Kibria et al. 2016, Liu et al. 2020). Other sources of high cadmium accumulation include port and shipping activities (Karar et al. 2019). The Cd accumulation of sediment had an average of 1.60 mg/kg, more than the standard of CCME (1999) (< 0.7 mg/ kg). Lin et al. (2021) established that Cd was the major contributor to health risk and social mortality. The greatest accumulation of Zinc (35.17 mg/kg) comes from the cement industry. However, its accumulation is lower than the CCME standard (2001) (< 124 m /kg). This is still higher than Riyanti et al. (2019) which focused on Payung Island South Sumatra (16.11 mg/kg).

In aquatic ecosystems (rivers and lagoon), mercury and cadmium accumulation had different significant distributions with Zinc accumulation. The decree of MENLH No. 51 of 2004 showed that their accumulation in the E-SAL was polluted because had exceeded the water quality standards for aquatic biota (Hg > 0.001 mg/L and Cadmium > 0.001mg/L). Ginting, Indris, and Syakti, (2019) suggest that the Tanjung unggat river estuary and Tanjung in Pinang City have Cd accumulation of < 0.001 mg/L, which was lower than in E-SAL (0.029 mg/L). However, Zinc accumulation in the aquatic ecosystem had less

score than MENLH No. 51, 2004 (Zinc < 0.1 mg/L), known as unpolluted. However, the heavy metal accumulation in E-SAL (0.055 mg/L) was still higher than Triantoro et al. (2017) with Zn accumulation of 0.003 mg/L.

The interpolation and distribution of total accumulation of heavy metal in mangrove ecosystem

The interpolation and distribution of total accumulation of heavy metal accumulation in Segara Anakan Cilacap relative different with Kabata dan Pendias, (2001) research (concentrations of heavy metal in vegetation were 27–150 ppm (Zn) and 20–10 µg/Kg (Cd)). Analuddin et al. (2017) demonstrated that potential high concentrations of Cu (83.85 µg g⁻¹) and Hg (0.52 µg g⁻¹) are found in the tissues of *L. racemose*, while high concentrations of Cd (10.81 µg g⁻¹), Zn (70.41 µg g⁻¹), and Pb (1.36 µg g⁻¹) are found in the tissues of *Bruguiera gymnorhiza*, *Bruguiera parviflora*, and *Ceriops Tagal*. Alzahrani et al. (2018) writes that the potential of heavy metals in *Avicennia marina* has range Cr > Cu > Ni > Pb > Cd (Analuddin et al. 2017, Liu et al. 2020).

The heavy metals such as Hg, Cd, and Zn accumulation of mangrove vegetation on roots, stems, or leaves are bigger than in the water body but smaller than in sediments. This condition shows that mangroves can absorb, accumulate and use heavy metals from water and sediments following nutrient absorption to support their growth and metabolic processes (Analuddin et al. 2017, Hidayati et al. 2014, Zhang et al. 2019). These accumulations are influenced by root activity to absorb, transfer and translocate to other parts (Alzahrani et al. 2018, R. Xiao et al. 2015). However, mangroves has ability to avoid death from heavy metal pollution, reducing toxic mechanisms using a dilution and translocation to dead or unnecessary tissues. Moreover, mangroves can increase organic matter absorption (Nour et al. 2019, Xiao et al. 2015). Heavy metals, such as Cd, Hg, and Zn, significantly increase malonaldehyde and proline contents, inhibit photosynthetic pigment, and non-protein thiols, glutathione, and phytochelatins (Dai et al. 2017). de Almeida Duarte et al. (2017) find that metal contamination of Cd, Cu, Pb, Cr, Mn, and Hg in water, sediment, red-mangrove vegetation (*Rhizophora mangle*), and tissues.

Heavy metal accumulation in roots is higher than in stems and leaves because the roots function as a direct contact and nutrient absorber, followed by heavy metal absorption from sediment and water column (Analuddin et al. 2017, Nour et al. 2019) and then translocated to other parts (Alzahrani et al. 2018, Chai et al. 2020, Hilmi et al. 2017). Similarly, MacFarlane and Burchett (2002) and Heriyanto (2011) established that the ion concentration of roots is higher than in the leaves and other parts. However, mangrove roots metabolize to avoid excessive heavy metal input, reducing the negative impacts on growth. The metal absorption is also influenced by roots' absorption activity depending on the root system and their lenticels size (Ma et al. 2019, Mapenzi et al. 2020)

The main mangrove species, such as *Sonneratia alba*, *Avicennia marina*, and *Nypa frutican*, had the highest Hg accumulation. In contrast, *Avicennia marina*, *Rhizophora apiculata*, and *Nypa frutican* had the greatest Cd. *Avicennia marina*, *Melaluca leucadendron* and *Xylocarpus granatum* had greatest ability to accumulate Zn. *Sonneratia alba*, *Avicennia marina*, *Nypa frutican*, and *Rhizophora apiculata* dominated the first zone of the Segara Anakan (Hilmi et al. 2019, Hilmi et al, 2017) and had both width spreading root (Hilmi et al. 2015) and a good respiratory system to grow in contaminant area. For example, the *Avicennia marina* had pneumatophore roots with a small diameter (< 0.9 cm) to absorb and reduce heavy metals pollution (Penha-Lopes et al. 2010, Siteo et al. 2014).

The distribution of heavy metal accumulation in the mangrove ecosystem of Segara Anakan in Fig. 4 showed that average of heavy metal accumulation > deviation standard. The distribution of Hg accumulation between 0.020–0.032 mg/lit (average 0.025 and stdev 0.045 mg/lit). The distribution of Cd accumulation between 0.095–0.132 mg/lit (average 0.1155 and stdev 0.090 mg/lit). The distribution of Zn accumulation between 15.88–25.06 mg/lit (average 19.43 and stdev 5.48 mg/lit)

Moreover, the Fig. 5 must be correlated by critical condition of heavy metal pollution. The Zinc has a critical concentration classification with the deficiency ranging from 10–20 mg/kg. The healthy plants contain an amount of Zinc between 10–100 mg/Kg with an average of 60 mg/kg and toxic concentrations between 100–1000 mg/kg (Kibria et al. 2016, Mertens et

al. 2006, Robson et al. 2014, Wang et al. 2013). Generally, Cadmium has a natural concentration in the soil between 0.1 mg/kg – 1 mg/kg, and human activities such as fertilization and industrial disposal can increase more than 0.1 mg/kg to 0.3 mg/kg (Costa-Böddeker et al. 2020, Kibria et al. 2016, Mertens et al. 2006, Wang et al. 2013). The critical and toxicity of cadmium concentration in plants ranges from 10 to 20 ppm dry weight. Jiang et al. (2017) stated that mangroves had a response of phenolic metabolism to reduce the impact of Cadmium in plants. Mercury is toxic for many organisms, either as a single element or a compound (Li et al. 2016, St. Gelais & Costa-Pierce, 2016). According to (Mapenzi et al. 2020), the standard concentration of Mercury (Hg) was less than 0.2 mg/Kg. The toxicity symptoms of mercury affect plants, specifically chlorophyll damage, growth limitation, reducing membranes of root cells, photosynthesis, respiration, uptake of water, nutrients, and chlorophyll synthesis (Alzahrani et al. 2018a, Zhang et al. 2019).

Bioaccumulation factor (BAF) and Translocation Factor (TF) of heavymetal contaminant

Bioaccumulation factor (BAF) of heavymetal contaminant

The Table 3 give information that mangrove species as bioaccumulation agent has abilities to reduce impact of heavymetal with bioaccumulation process that are 1) phytoextraction- the absorption ability of pollutants from water or soil through mangrove roots stored in plant canopy (Win et al. 2019, Yu et al. 2022), 2) Phytovolatilization- the absorption of pollutants using evaporative process and are transpired by mangrove leaves (McCutcheon et al. 2002, Zhang et al. 2020), 3) phytodegradation or phytotransformation- the ability to absorb and destroy pollutants through the metabolism using enzymes or compounds (Kagalkar et al. 2011, McCutcheon et al. 2002), 4) phytostabilization- a pollutant transforming process into non-toxic compounds without absorbing process or keep these pollutants in soil (McCutcheon et al. 2002, Radziemska et al. 2021, Zhang et al. 2021), 5) rhizofiltration- the pollutant absorbing process by mangrove root on low pollutant concentrations (de Oliveira et al. 2015, McCutcheon et al. 2002, Yin et al. 2018)

According to Ayub (2015), BAF value ranges between 0 to > 1. A value > 1 indicates that the plants could accumulate contaminants because BAF has a positive correlation with ability to accumulate contaminant. This research showed that presence of *Avicennia marina*, *Sonneratia alba*, *Nypa frutican*, *Rhizophora mucronata*, and *Rhizophora apiculata* in the E-SAL was quite effective to accumulate heavy metals from waters, but mangrove doesn't have the good ability to accumulate contaminant from sediments because mangroves still must have adaptation to live in pollution conditions (Nicolaus et al. 2015, Shi et al. 2020).

Translocation Factor (TF)

Translocation Factor (TF) is used to determine the metal transfer process and translocation from root to leaf. TF value > 1 shows that the plant can translocate the contaminants absorbed into the upper organs (Analuddin et al. 2017, Chai et al. 2020, Hilmi et al. 2017). Mangrove root has an important role in preventing and reducing of heavy metal contaminants. However, the mangrove roots have ability to prevent the metal transport process and increase the accumulation of heavy metal contaminants (Alzahrani et al. 2018, Z. J. Chen et al. 2021, Jiang et al. 2017)

The translocation factor correlates also showing the ability of heavy metal accumulation in stems and leaves. *Avicennia marina*, *Rhizophora styles*, *Xylocarpus granatum*, *Sonneratia alba*, *Nypa frutican*, *Rhizophora mucronate*, and *Rhizophora apiculata* have good hyperaccumulation system for reducing contaminants in water body and sediment. And give positive impact to reduce impacts of heavy metal on other living organisms (Alzahrani et al. 2018, Marambio-Alfaro et al. 2020)

Mangrove landscape based on heavy metal accumulation

The mangrove landscape showed that mangrove pioneers, that were *Avicennia marina*, *Bruguiera gymnorhiza*, *Sonneratia alba*, *Nypa frutican*, *Rhizophora mucronata*, *Rhizophora apiculata*, and *Xylocarpus granatum* had high ability to reducing impact of heavy metal contaminant, because this species had function as phytoextraction, Phytovolatilization,

phytodegradation or phytotransformation, phytostabilization, and rhizofiltration to eliminate lethal effects of heavy metal pollution (Costa-Böddeker et al. 2020, Hidayati et al. 2014, Syakti et al. 2013).

The mangrove landscape was developed to protect the water system from land-based sources of pollution in the marine and coastal ecosystem and reduce toxic mechanisms using the dilution and translocation process (Analuddin et al. 2017, Ariani et al. 2016, Hilmi et al. 2017). Additionally, zonation reduces the impact of malonaldehyde content, photosynthetic pigment, proline content, synthesis of non-protein thiols, glutathione, phytochelatins (Dai et al. 2017), yellow leaves (de Almeida Duarte et al. 2017), increase of lysosomal membrane integrity, cytogenetic and a pre-pathological condition (Shi et al. 2020)

The Clustering of heavy metal accumulation in mangrove ecosystem

This study showed that the mangrove ecosystem had 4 clusters, including Cluster 1- *Melaleuca leucadendron*, *Xylocarpus granatum*, *Rhizophora stylosas*, *Aegicera floridum*, *Rhizophora mucronata*, Cluster 2 - *Ceriops tagal*, *Excoecaria agallocha*, *Rhizophora apiculata*, Cluster 3 - *Bruguiera gymnorhiza*, *Nypa frutican*, and Cluster 4 - *Avicennia marina*, *Heritiera littoralis*, *Aegiceras corniculatum*, *Bruguiera sexangula*, *Sonneratia alba*. According Choi et al. (2020) and Yang et al. (2020) note the clustering of heavy metal accumulation be influenced by rivers' sites and surface sediment. The results showed that cluster 1 was dominated by silt and clay with high organic matter and nutrient concentrations. Cluster 2 contained rivers that were significantly affected by artificial pollution from heavy metals. Cluster 3 included sites with the lowest pollution effects caused by organic matter, nutrients, and heavy metals.

Conclusion

The lagoon and mangrove ecosystem were polluted by heavy metal pollution using the accumulation of mercury data between 0.01375 ppm (water) – 0.13475 ppm (sediment), Cadmium 0.0292 ppm (water) – 1.6048 ppm (sediment), and Zinc 0.0550 ppm (water) – 35.1746 ppm (sediment). Reducing the impact of heavy metal pollution can be affected by planting system of *Sonneratia alba*, *Avicennia marina* and *Rhizophora mucronata* (Hg accumulation), *Rhizophora apiculata*, *Rhizophora mucronata* (Cd accumulation), *Avicennia marina*, and *Sonneratia alba* (Zn accumulation).

Declarations

Declaration of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

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Figures

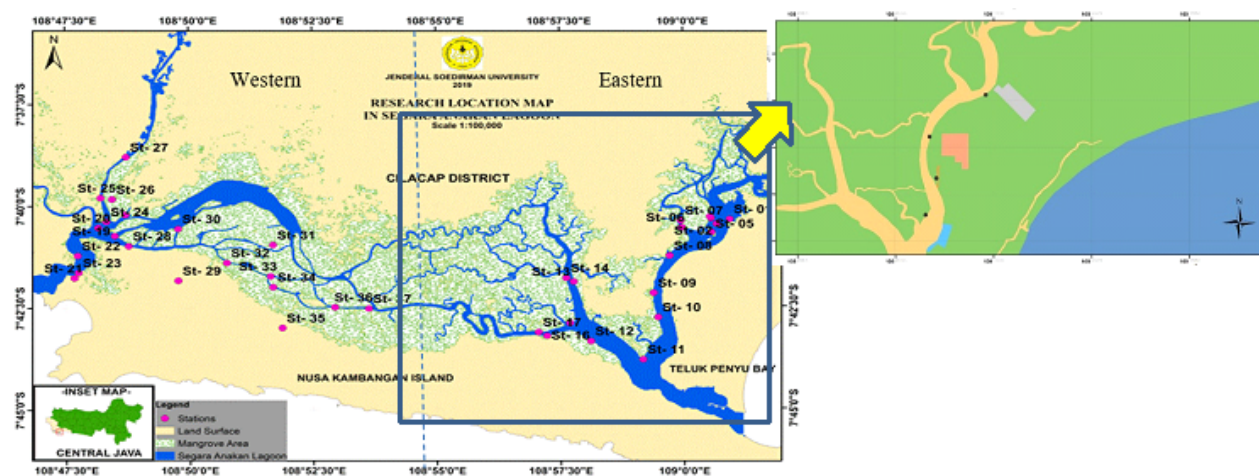


Figure 1

Study area

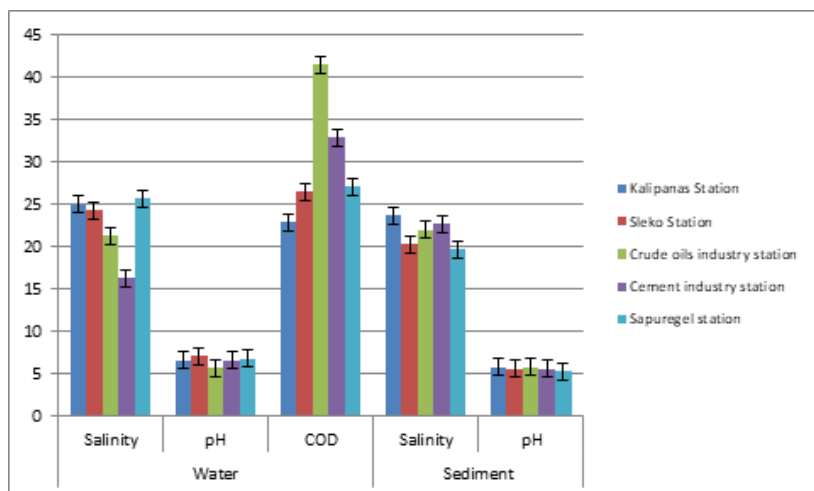


Figure 2

Environment factor of heavymetal accumulation

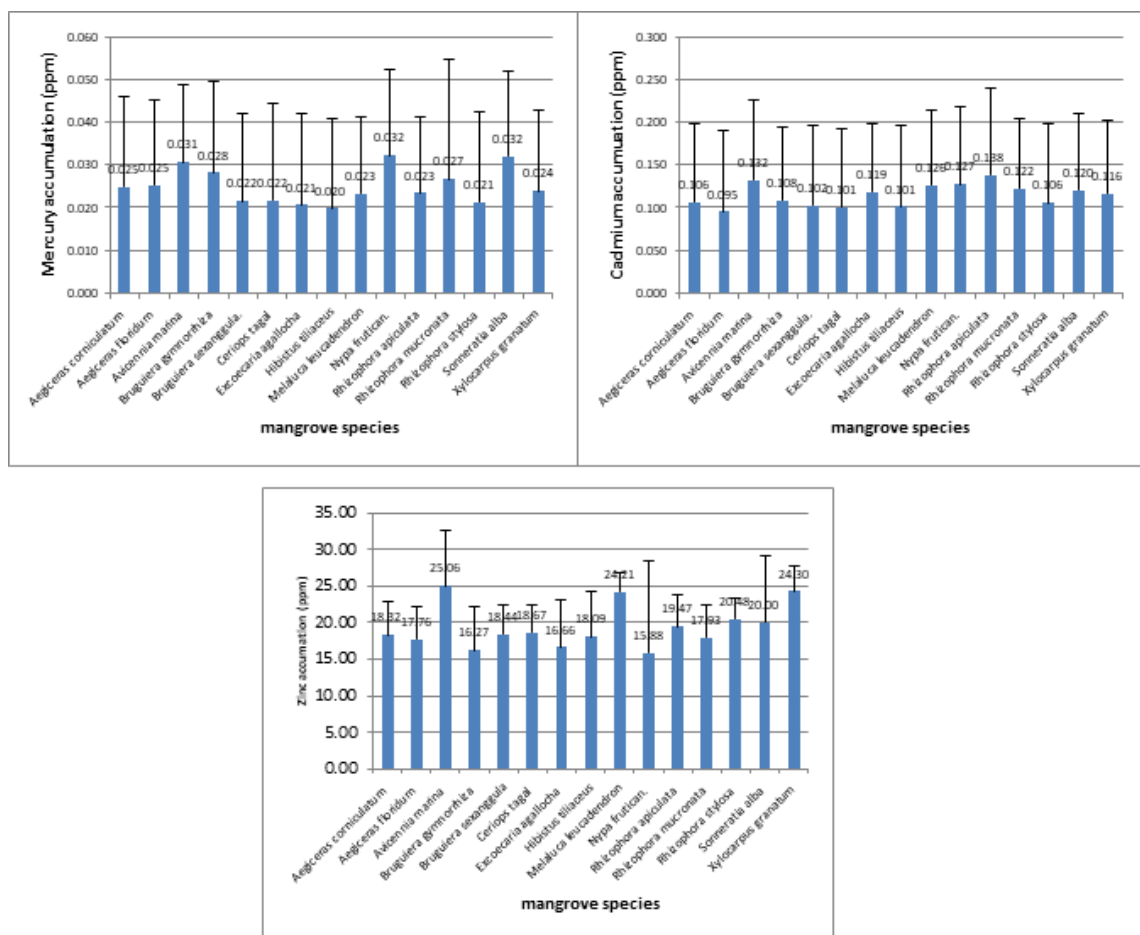


Figure 3

The potential of heavy metal accumulation on mangrove species

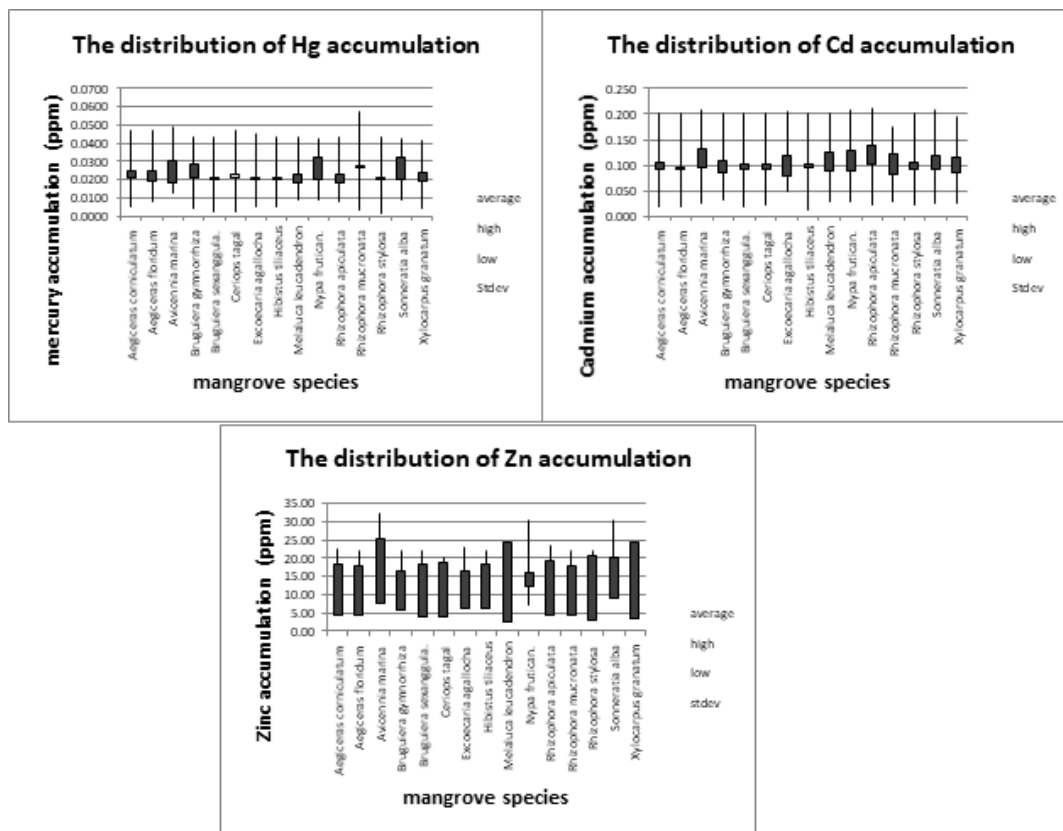


Figure 4

Distribution of heavy metal accumulation on mangrove species

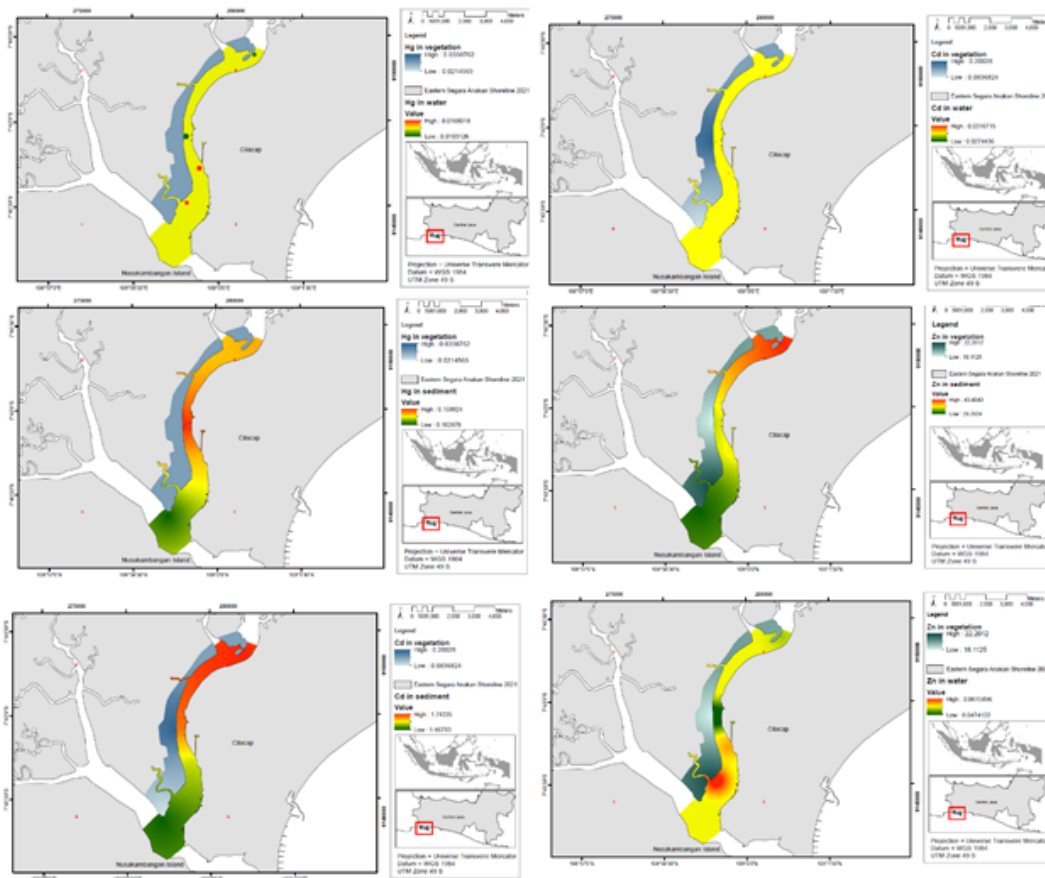


Figure 5

Interpolation of heavymetal contaminant distribution

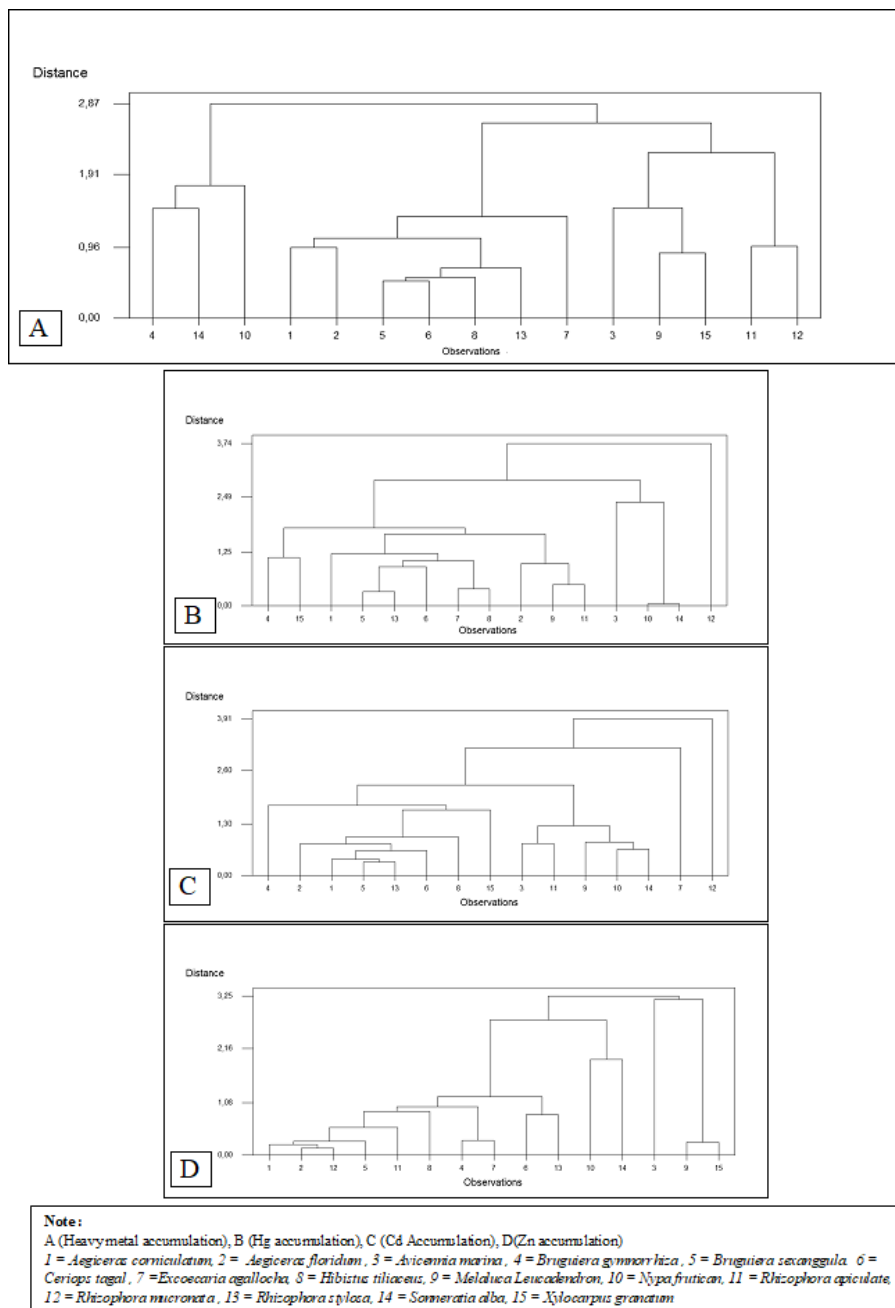


Figure 6

The clustering of heavy metal accumulation in mangrove vegetation