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

Endang Hilmi (dr.endanghilmi@gmail.com)
Universitas Jenderal Soedirman, Central Java
Rose Dewi
Universitas Jenderal Soedirman, Central Java
Eming Sudiana
Universitas Jenderal Soedirman, Central Java
Arif Mahdiana
Universitas Jenderal Soedirman, Central Java
Lilik Kartika Sari
Universitas Jenderal Soedirman, Central Java
Tri Nur Cahyo
Universitas Jenderal Soedirman, Central Java

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Abstract

Mangrove has ability to reduce impact of heavy metal contaminant which released from industry and domestic waste. Mangrove has ability to absorb, accumulate, and translocate heavy metal contaminant. Whereas the clustering and landscape of mangrove ecosystem show the grading and zoning of mangrove vegetation to accumulate heavymetal pollution. .Therefore, this study aimed to analyze distribution and clustering of heavy metal accumulation and traslocation in mangrove ecosystem.. The results showed the accumulation of Mercury were 0.1347 ± 0.0212 ppm (sediment), 0.0137 ± 0.0137 ppm (water), 0.0110–0.0640 ppm (stem and bark), 0.0020–0.0120 ppm (leaves), and 0.0260–0.0690 ppm (roots). Cadmium accumulations between 1.6048 ± 0.3595 ppm (sediment), 0.0292 ± 0.0047 ppm (water), 0.0790–0.2650 ppm (stem and bark), 0.0130–0.0480 ppm (leaves), and 0.1560–0.2300 ppm (roots). Furthermore, Zinc had score that were 35.1746 ± 5.4856 ppm (sediment), 0.0550 ± 0.0095 ppm (water), 10.3250–32.4490 ppm (stem and bark), 7.1200-23.4150 ppm (leaves), and 16.7170–39.1200 ppm (roots). However, bioaccumulation factors of Hg ranged between 0.0210–0.4751, Cd 0.0105–0.1541, and Zn 0.2807–1.2506 with the translocation factors of 0.0459–1.0547 (Hg), 0.0642–1.3855 (Cd), and 0.4086–1.3057 (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 industry (Syakti et al. 2013a; Hilmi et al. 2017b) sediments, garbage, and household. The adaptation pattern of mangrove vegetation to grow and survive in the polluted area can be conducted by the activity of specific root and metabolism system (Hilmi et al. 2017b, a). This activities can be used to reduce impact of heavy metal contaminant following nutrient absorption activity (Hilmi et al. 2017b, a; Wolswijk et al. 2020).

In Eastern Segara Anakan, mangrove ecosystem is categorized as a specific lagoon that takes freshwater supply from many rivers, including the Donan river (Syakti et al. 2013b; Hilmi 2018; Hilmi et al. 2021c, a, b) and seawater supply from the Indian Ocean. Eastern Segara Anakan Lagoon (E-SAL). Mangrove ecosystem in SAL can be used as an suitable area for waste disposal due to industrial, and antropogenic activities (Syakti et al. 2013a; Hilmi et al. 2017b; Prastyo et al. 2017). The disposal form industrial and anthropogenic activities in coastal ecosystem give impacts including heavy metal pollutants supply, specifically Mercury (Hg), Zinc (Zn), Lead (Pb), and Cadmium (Cd) (Hidayati et al. 2011; Xin et al. 2014; Kibria et al. 2016; Hilmi et al. 2017b; Cao et al. 2020).

Basically, heavy metal contaminants have specific characterize that are the non-degradable properties, the quickly accumulate in the substrates, sediment, and water and give negatively affecting of aquatic organisms life. Because the characteristics of their toxicity, abundance, persistence, and bioaccumulation (Jeong et al. 2021), heavy metal pollution also may cause death, litter fall, and stunted growth of trees (Melville et al. 2009; de Almeida Duarte et al. 2017; Costa-Böddeker et al. 2020; Shi et al. 2020). According to Marambio et al. (2020) and, Cao et al. (2020) the metal accumulation of Cu, Pb, Zn, Ni, and Cd give negatively affects organism growth and give high risks for fish farms, organism activity and coastal stabilization.

Mercury (Hg) especially methyl mercury (CH_3)-Hg as an industrial and laboratory waste that contains hazardous properties, strong binding capacity and high toxic levels (Li et al. 2016; de Almeida Duarte et al. 2017; Analuddin et al. 2017; Mapenzi et al. 2020; Shi et al. 2020; Wolswijk et al. 2020). Generally, Mercury is a liquid substance at room temperature (25^oC) with a freezing point of -39^oC and boils at 365,68^oC. Mangrove wetlands contribute up to 15% of the coastal sediment mercury (Hg) methylation, significantly impacting global Hg cycling (Li et al. 2016; Lei et al. 2019; 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 (Kibria et al. 2016; El-Amier et al. 2017; Zhang et al. 2019a; Costa-Böddeker et al. 2020) and has melting and boiling points of 321 °C and 67 °C, respectively (Ortega et al. 2017; Analuddin et al. 2017). And 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; Zhang et al. 2019a; Mapenzi et al. 2020).

Heavy metal accumulation and translocation (Hilmi et al. 2017b; Nadgórska–Socha et al. 2017; Shi et al. 2020; Jeong et al. 2021) of mangrove vegetation occur in the roots, stems, and mangrove leaves (Oo et al. 2009; Alzahrani et al. 2018; Costa-Böddeker et al. 2020). And then The bioaccumulation process shows absorption, accumulation, and utilization of contaminants from sediments in a root system and surface area (Ma et al. 2019; Zhang et al. 2019a; Shi et al. 2020). This activity reduces toxic effects using dilution and translocation to death organs (Alzahrani et al. 2018; Li et al. 2020) and organic absorption (Xiao et al. 2015; Nour et al. 2019). 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 (Xiao et al. 2015; Alzahrani et al. 2018; Chai et al. 2020).

The distribution and clustering of Zn, Hg, Cd accumulation explains the landscape and cluster of mangrove ability to reduce the impact of heavy metal contaminants (Syakti et al. 2013a; Hilmi et al. 2017b, 2021b) and describe the relationship and adaptation models of bioaccumulation and trasclocation using an index of Euclidian distance (Syakti et al. 2013a; Hilmi et al. 2015, 2017a, 2021a). This study aimed to analyze distribution 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 area (Hilmi et al. 2019a, 2021a, d) 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. 2015, 2021c, d, 2022). The samples of mangrove were collected from roots, barks, stems, and leaves (Monteiro et al. 2014; Xiong et al. 2018). 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 be labeled. The addition of 0.75 mL concentrated HNO_3 aims to reduce the pH to 2 (Hilmi et al. 2017b; Win et al. 2019; Mapenzi et al. 2020). 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 be labeled (Hilmi et al. 2017b; Prastyo et al. 2017; Mapenzi et al. 2020), 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, bark, roots and leaves were also collected using the destructive method. And pneumatophore roots were taken in a minimum water and soil depth of 10–20 cm. All of them were put into a plastic bag, be labeled, and then put into an icebox (Hilmi et al. 2017b; Alzahrani et al. 2018).

Heavy metal analysis

a. Hg (mercury)

Mercury was analyzed through the Spectrophotometric method, which involves the Shimatsu brand with an accuracy level of 2x10^{-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 be extracted using liquid tetra dhitizone. Hg was measured at a wavelength of 480 ηm and be analyzed using the mercury analyzer (SP-3D) method. The acid was intended to convert: Hg $^{2+}$ + $SnCl_2$ ◊ 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 2x10^{-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 2x10^{-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^+ + $SnCl_2$ → 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 (Syakti et al. 2013a; Hilmi et al. 2017b; Lin et al. 2021).

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

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

Bioaccumulation *Factor* (BAF) categories included (MacFarlane et al. 2003; Hilmi et al. 2017b; Marambio et al. 2020).

BAF ≤ 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 (MacFarlane et al. 2003; Hilmi et al. 2017b; Marambio et al. 2020).

TF ≤ 1 represents vegetations less or unable to translocate heavy metal pollution in other organs

TF > 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. 2021c, b).

Stage 1.

$$ED_{jk} = \sqrt{\sum_{i=1}^{s} (xij - xik)^2}$$

Stage 2.

$$D(j, k)h = \propto {}_1D(j, h) + \propto {}_2D(k, h) + \beta D(j, k)$$

Stations	2	3	4	...	22
1	ED_{12}	ED_{13}	ED_{14}		
2		ED_{23}	ED_{24}		
3			ED_{34}		
			...		
22			ED_{22}		

Notes:

Ed*jk* : Euclidean Distance

i : species

X*ij* : density in station- *j*

X*ik* : 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 of heavy metal contaminant in sediment and aquatic ecosystem

The potential of heavy metal in sediment and aquatic ecosystem come from domestic pollution, seawage treatment, pesticides, mining and pharmaceutical industries (Nour et al. 2019) which give negative impact for organism, community and environment (Jiang et al. 2017; Cao et al. 2020; Jeong et al. 2021). Table 2 showed the distribution of heavy metal contaminant on the sediment and aquatic ecosystem. Whereas Fig. 2 showed the environment factors in Segara Aanakan which receive supplies of heavymetal contaminant from industry and domestic waste

Table 2
The distribution of Heavy metal contaminant 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 (Stdev). 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 of deviation standard

While the interpolation of heavy metal accumulation in vegetation, sediment and water can be seen in Fig. 5. The data in Fig. 5 shows that Hg in vegetation and water < Hg in vegetation and sediment. The accumulation of Hg in vegetation and sediment has the potential to be moderate to high. Similar to the accumulation of Hg, the potential for Cd and Zn in vegetation and water < potential for Cd and Zn in sediments. However, the potential for Cd in vegetation and sediment is dominated by a very high accumulation potential

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

Bioaccumalation 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 BAF 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
Bioaccumalation factor (BAF) in Mangrove ecosystem

mangrove species	BAF with sedimen								
	Hg			Cd			Zn		
	stem	leaves	root	stem	leaves	Root	stem	leaves	root
Aegiceras corniculatum	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
Aegiceras floridum	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
Avicennia marina	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
Bruguiera gymnoriza	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
Bruguiera sexanggula.	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
Ceriops tagal	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
Excoecaria agallocha	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
Hibistus tiliaceus	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
Melaluca leucadendron	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
Nypa frutican.	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
Rhizophora apiculata	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
Rhizophora mucronata	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
Rhizophora stylosa	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
Sonneratia alba	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
Xylocarpus granatum	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 *Aviccenia 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) (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**
Aegiceras corniculatum	0.4276–0.5559	0.0855–0.1283	0.4589–0.5337	0.0898–0.1048	0.6170–0.9770	0.4491–0.7494
Aegiceras floridum	0.3506–0.4490	0.1710–0.1810	0.3342–0.4337	0.0898–0.1048	0.6710–0.7856	0.4481–0.5826
Avicennia marina	0.3860–0.8148	0.2090–0.3330	0.5991–0.8982	0.0838–0.1435	0.7114–0.8295	0.4257–0.7191
Bruguiera gymnoriza	0.3303–0.7486	0.0917–0.1102	0.4397-0.5200	0.0740–0.1630	0.6778–0.7511	0.4645–0.6004
Bruguiera sexanggula.	0.4128-0.7200	0.0688-0.1100	0.3420–0.4249	0.0939–0.1001	0.7650–0.8750	0.5210–0.6363
Ceriops tagal	0.3207–0.4320	0.0641–0.0893	0.3941–0.4001	0.0885–0.1147	0.7887–0.9445	0.5232–0.6313
Excoecaria agallocha	0.2616–0.3345	0.0576–0.1109	0.4210–0.5101	0.0978–0.2354	0.7090–0.7888	0.4519-0.5000
Hibistus tiliaceus	0.2000-0.2523	0.0534–0.1147	0.4100-0.4348	0.0642-0.0900	0.8210–0.9685	0.3201–0.4942
Melaluca leucadendron	0.3000-0.3889	0.0879–0.2064	0.5670–0.7164	0.0921–0.1482	0.8450–1.2339	0.9101–1.0627
Nypa frutican.	0.7985–1.0547	0.1100-0.2109	0.5761–0.6923	0.0767–0.1442	0.3419–0.4536	0.2358–0.3450
Rhizophora apiculata	0.2667–0.6923	0.1017–0.3462	0.7627–0.9557	0.0742–0.1626	0.7259–1.3883	0.5002–1.0427
Rhizophora mucronata	0.3261–0.3333	0.0580–0.0652	0.4973–1.3855	0.0909–0.2470	0.8193–0.8637	0.5610–0.6298
Rhizophora stylosa	0.3420–0.4128	0.0459–0.0655	0.4644–0.6760	0.0767–0.1038	0.8650–1.0043	0.5678–0.7836
Sonneratia alba	0.5859-1.5000	0.1406–0.28125	0.5769–0.6250	0.1058–0.1538	0.5388–0.6068	0.4196 –0.4086
Xylocarpus granatum	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**
Bruguiera gymnoriza	*Avicennia marina*	*Avicennia marina*
Sonneratia alba	*Rhizophora Mucronata*	*Rhizophora apiculata*
Nypa frutican.	*Rhizophora apiculata*	*Xylocarpus granatum*
zone 2	**zone 2**	**zone 2**
Aegiceras corniculatum	*Sonneratia alba*	*Aegiceras corniculatum*
Xylocarpus granatum	*Xylocarpus granatum*	*Bruguiera sexanggula.*
Avicennia marina	*Nypa frutican.*	*Melaluca leucadendron*
	Melaluca leucadendron	*Hibistus tiliaceus*
		Ceriops tagal
		Rhizophora stylosa
zone 3	**zone 3**	**zone 3**
Melaluca leucadendron	*Bruguiera sexanggula.*	*Bruguiera gymnoriza*
Bruguiera sexanggula.	*Hibistus tiliaceus*	*Sonneratia alba*
Rhizophora stylosa	*Bruguiera gymnoriza*	*Aegiceras floridum*
Rhizophora Mucronata	*Rhizophora stylosa*	*Rhizophora Mucronata*
Rhizophora apiculata	*Aegiceras corniculatum*	
Aegiceras floridum	*Excoecaria agallocha*	
zone 4	**zone 4**	**zone 4**
Hibistus tiliaceus	*Aegiceras floridum*	*Nypa frutican.*
Excoecaria agallocha	*Ceriops tagal*	*Excoecaria agallocha*
Ceriops tagal		

The Clustering of heavy metal accumulation in mangrove ecosystem

Mangrove clustering refers to grouping of mangrove ability to absorb and accumulate heavy metal (Hilmi et al. 2021c, a) using the *Hierarchical Clustering Methods* and *Nonhierarchical Clustering Method* (Rachmatin 2014; Hilmi et al. 2021c, b, 2022). The clustering of heavy metal accumulation (Zn, Hg and zinc accumulation) shows differences between mangrove species in accumulating pollution (Fig. 6**)**.

Discussion

The distribution 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 (Peraturan pemerintah no 82 2001), Hg, Cd, and Zn polluted the aquatic ecosystem in the Segara Anakan Lagon (SAL). 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. 2021a) 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 of 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 (Kayalvizhi and Kathiresan 2019; Wang et al. 2019; Cao et al. 2020). But mangrove has metabolism system to reduceimpact of heavy metal pollution. The mangrove roots can stop transport non-essential matter or to accumulate in other dead organs.

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). Basically, mangrove ecosystem as a fragile ecosystem will be influenced by potential of heavy metal contaminant in water and sediment (Alzahrani et al. 2018). But, mangrove species still have an excellent adaptation to reduce potential heavy metal using excretion, excluder and accumulation gland (Hilmi et al. 2017b; Dai 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 (Xiong et al. 2018; Wolswijk et al. 2020). 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 (Kibria et al. 2016; Analuddin et al. 2017; Liu et al. 2020b). 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 (Canadian Environmental Protection Act 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 (Canadian Council of Ministers of the Environment) 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 Peraturan pemerintah no 82 (2001) and Keputusan Menteri Negara Lingkungan Hidup NO 51 (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 et al. (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 Peraturan pemerintah no 82 (2001) and Keputusan Menteri Negara Lingkungan Hidup 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 (Ortega 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 pollution in Segara Anakan Cilacap relative different with Jeong et al. (2021), because had 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 *Lumnitzera 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 gymnorrhiza, 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. 2020a).

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, because mangrove vegetations have ability to absorb, accumulate and use heavy metals from water and sediments following nutrient absorption to support their growth and metabolic processes (Analuddin et al. 2017; Zhang et al. 2019a). This metabolisms are influenced by root activity to absorb, transfer and translocate to other parts (Xiao et al. 2015; Alzahrani et al. 2018). Basically, mangroves also 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 (Xiao et al. 2015; Nour et al. 2019). But, the heavy metals, such as Cd, Hg, and Zn,also 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 are 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 (Hilmi et al. 2017b; Alzahrani et al. 2018; Chai et al. 2020). Similarly, MacFarlane and Burchett (2002) and Zhang et al. (2019a) 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. 2017b, 2019b, 2021c, a) 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; Sitoe 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/lt (average 0.025 and stdev 0.045 mg/lt). The distribution of Cd accumulation between 0..095–0.132 mg/lt (average 0.1155 and stdev 0.090 mg/lt). The distribution of Zn accumulation between 15.88–25.06 mg/lt (average 19.43 and stdev 5.48 mg/lt)

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 (Mertens et al. 2006; Wang et al. 2013; Robson et al. 2014; Kibria et al. 2016). 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 m/kg (Mertens et al. 2006; Wang et al. 2013; Kibria et al. 2016; Costa-Böddeker et al. 2020). 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 and 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. 2018; Zhang et al. 2019a).

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

Bioaccumalation factor (BAF) of heavymetal contaminant

The Table 3 give data’s that mangrove species has abilities to reduce impact of heavymetal with bioaccumulation process, namely 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. 2019b), 3) phytodegradation or phytotransformation- the ability to absorb and destroy pollutants through the metabolism using enzymes or compounds (McCutcheon et al. 2002; Yang et al. 2008; Kagalkar et al. 2011), 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 (McCutcheon et al. 2002; de Oliveira et al. 2015; Yin et al. 2018)

According to Hilmi et al. (2017b), Analuddin et al. (2017) and Zhang et al. (2019a), 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 (Analuddin et al. 2017; Shi et al. 2020; Chen et al. 2021b).

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 (Hilmi et al. 2017b; Analuddin et al. 2017; Shi et al. 2020; Chai et al. 2020). 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 (Jiang et al. 2017; Analuddin et al. 2017; Alzahrani et al. 2018; Chai et al. 2020).

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 et al. 2020)

Mangrove landscape based on heavy metal accumulation

The mangrove landscape showed that mangrove pioneers, that were *Avicennia marina, Bruguiera gymnorrhiza, 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 (Syakti et al. 2013a; Costa-Böddeker et al. 2020).

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 (Ariani et al. 2016; Hilmi et al. 2017b; Analuddin 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 styllosas, Aegicera floridum, Rhizophora mucronat*a, Cluster 2 - *Ceriops tagal, Excoecaria agallocha, Rhizophora apiculata*, Cluster 3 **-** *Bruguiera gymnnorhiza, Nypa frutican*, and Cluster 4 **-***Avicennia marina, Heritiera littoralis, Aaegiceras corniculatum, Bruguiera sexangula, Sonneratia alba*. According Choi et al. (2020), Jeong et al. (2021), Yang et al. (2020) and Cao 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.

Conclussion

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). The activity to reduce impact of heavy metal pollution can be developed by planting system using *Sonneratia alba, Avicennia marina and Rhizophora mucronata* (Hg accumulation), *Rhizophora apiculata, Rhizophora mucronata* (Cd accumulation), *Avicennia marina, and Sonneratia alba* (Zn accumulation) as a pioneer species to be planted in pollution area.

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

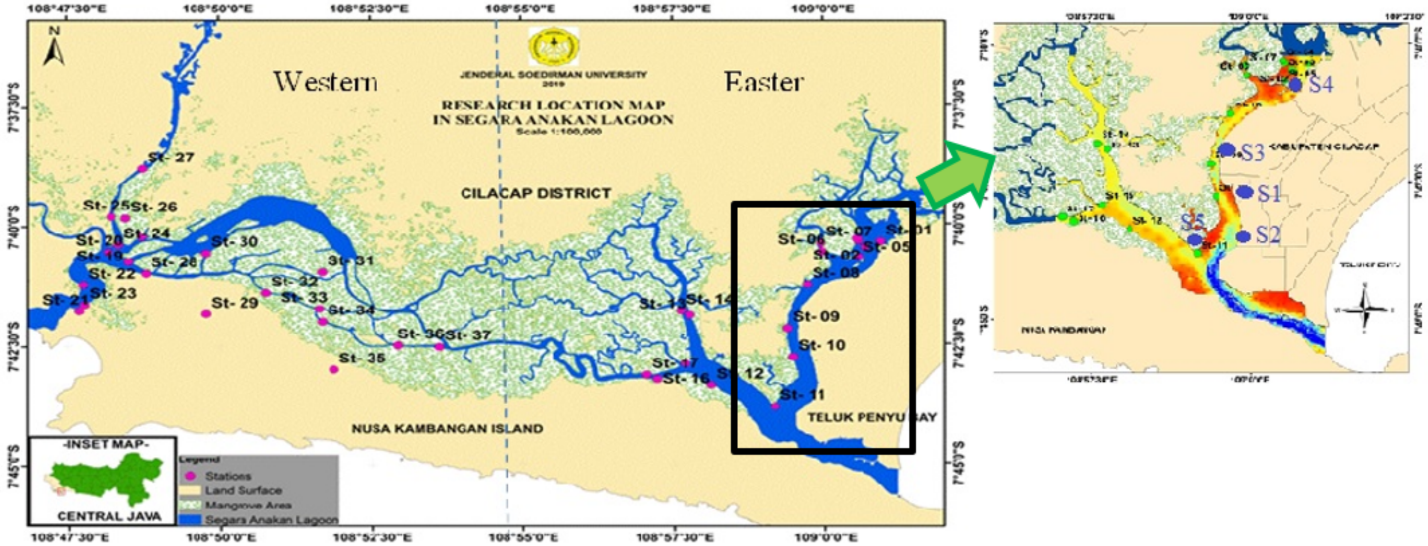

Figure 1

Study area

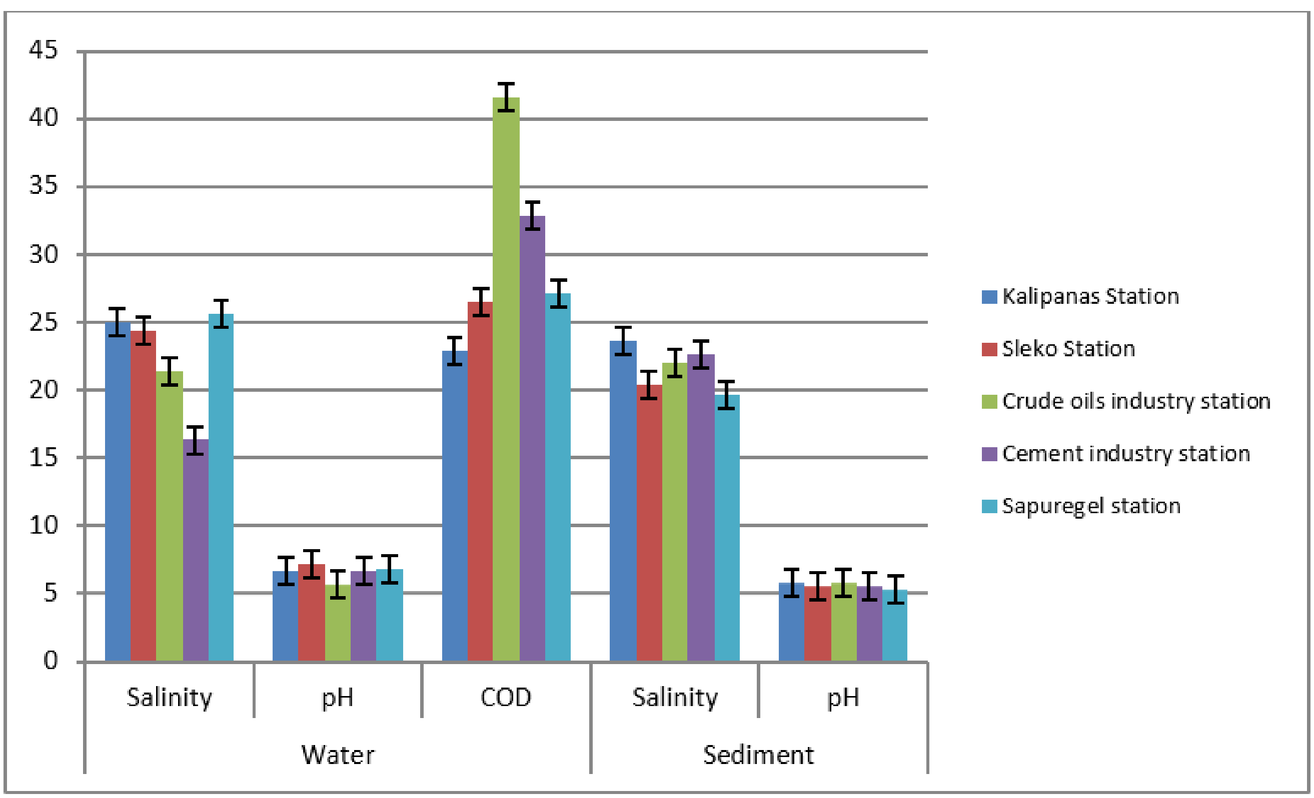

Figure 2

Environment factor of heavymetal accumulation

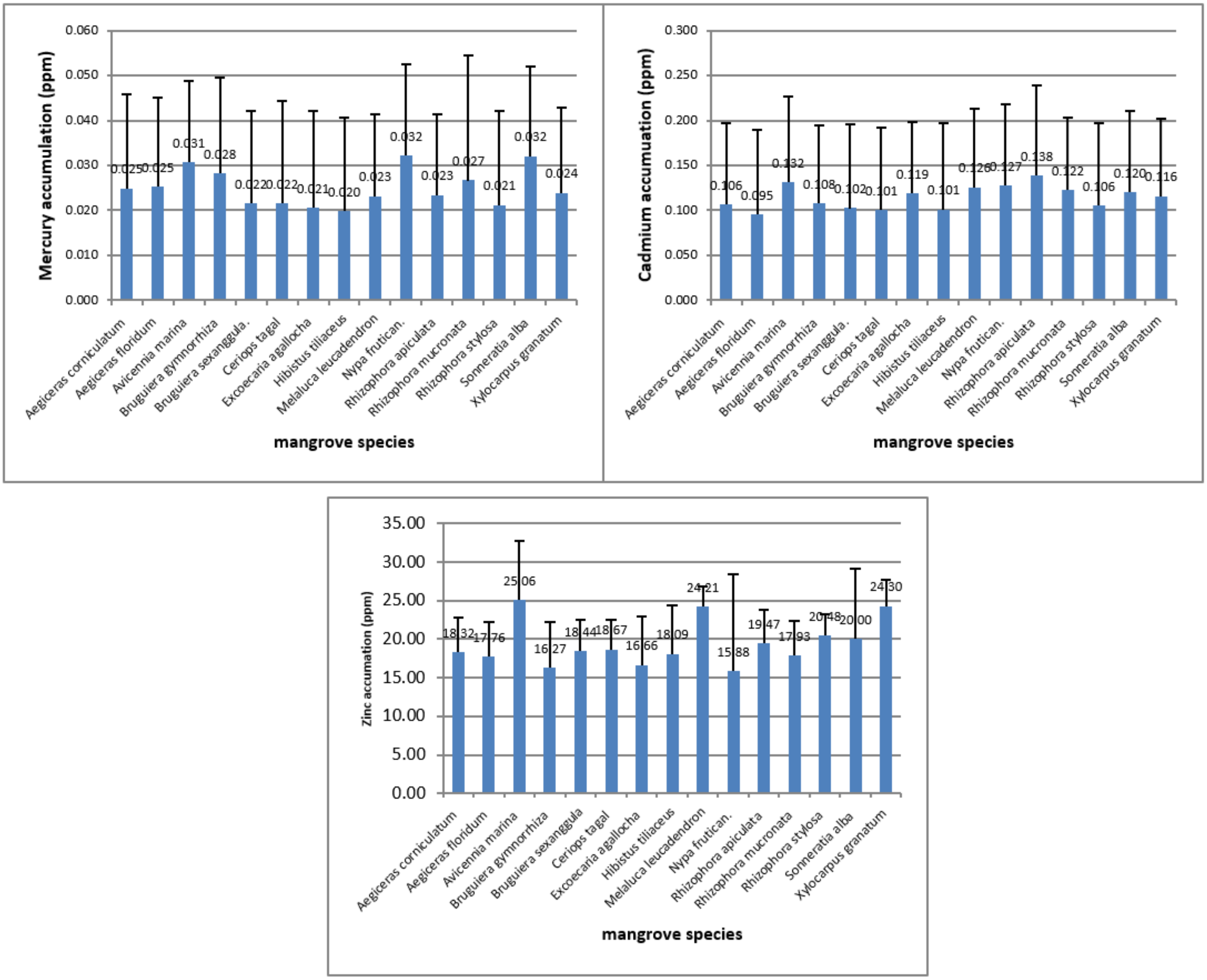

Figure 3

The potential of heavy metal accumulation on mangrove species

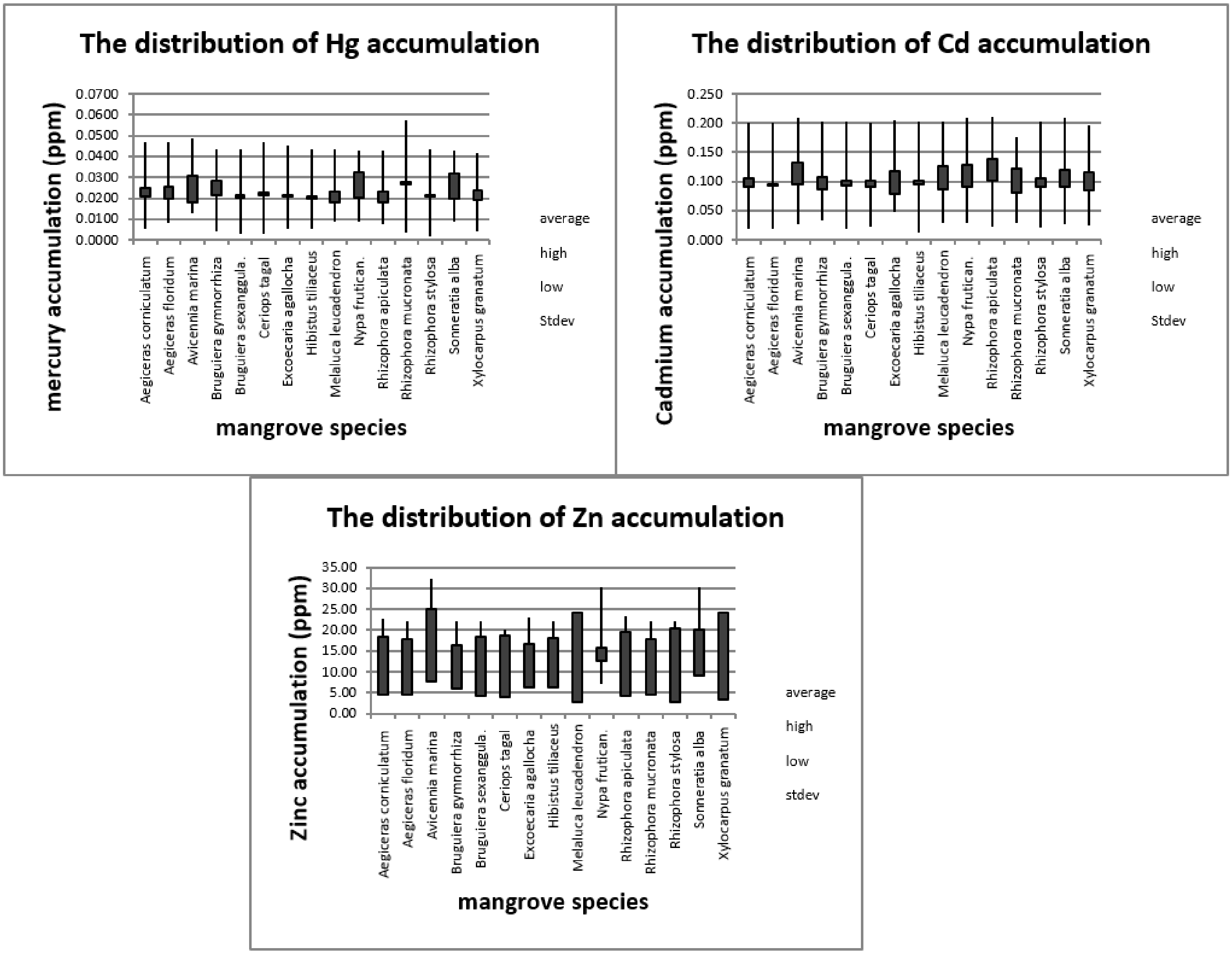

Figure 4

Distribution of heavy metal accumulation on mangrove species

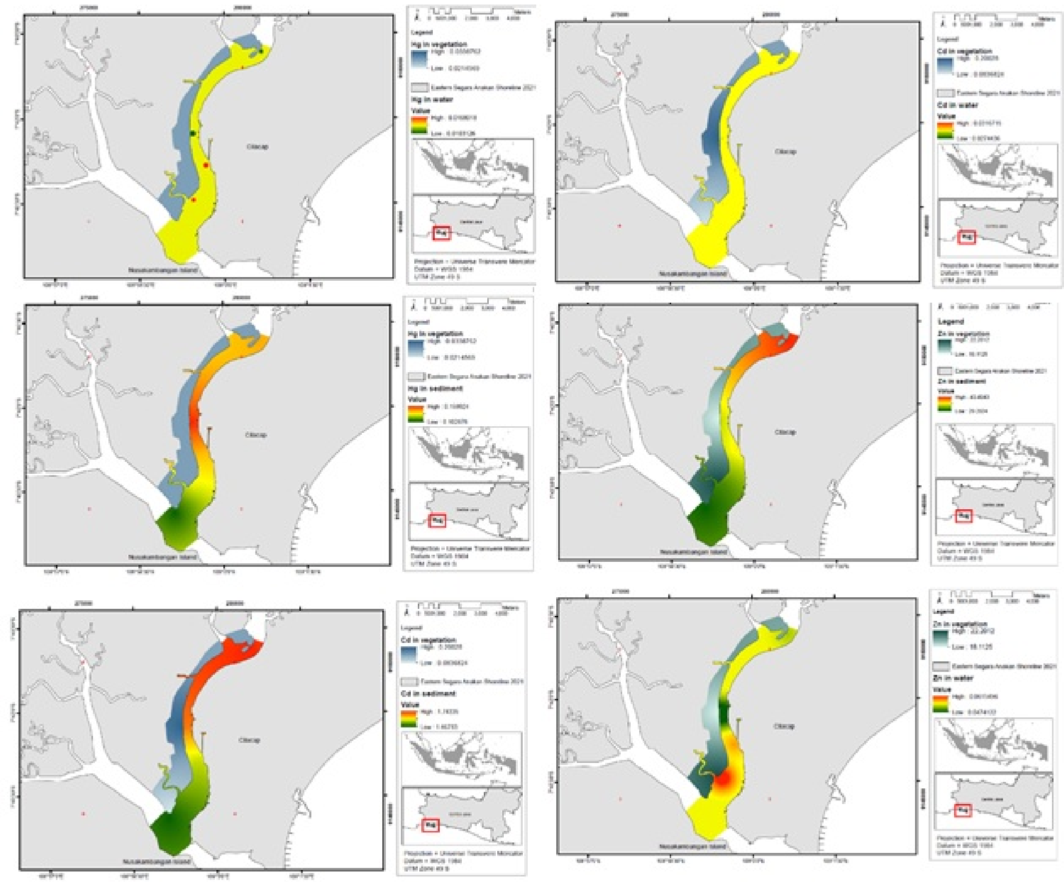

Figure 5

Interpolation of heavymetal contaminant distribution

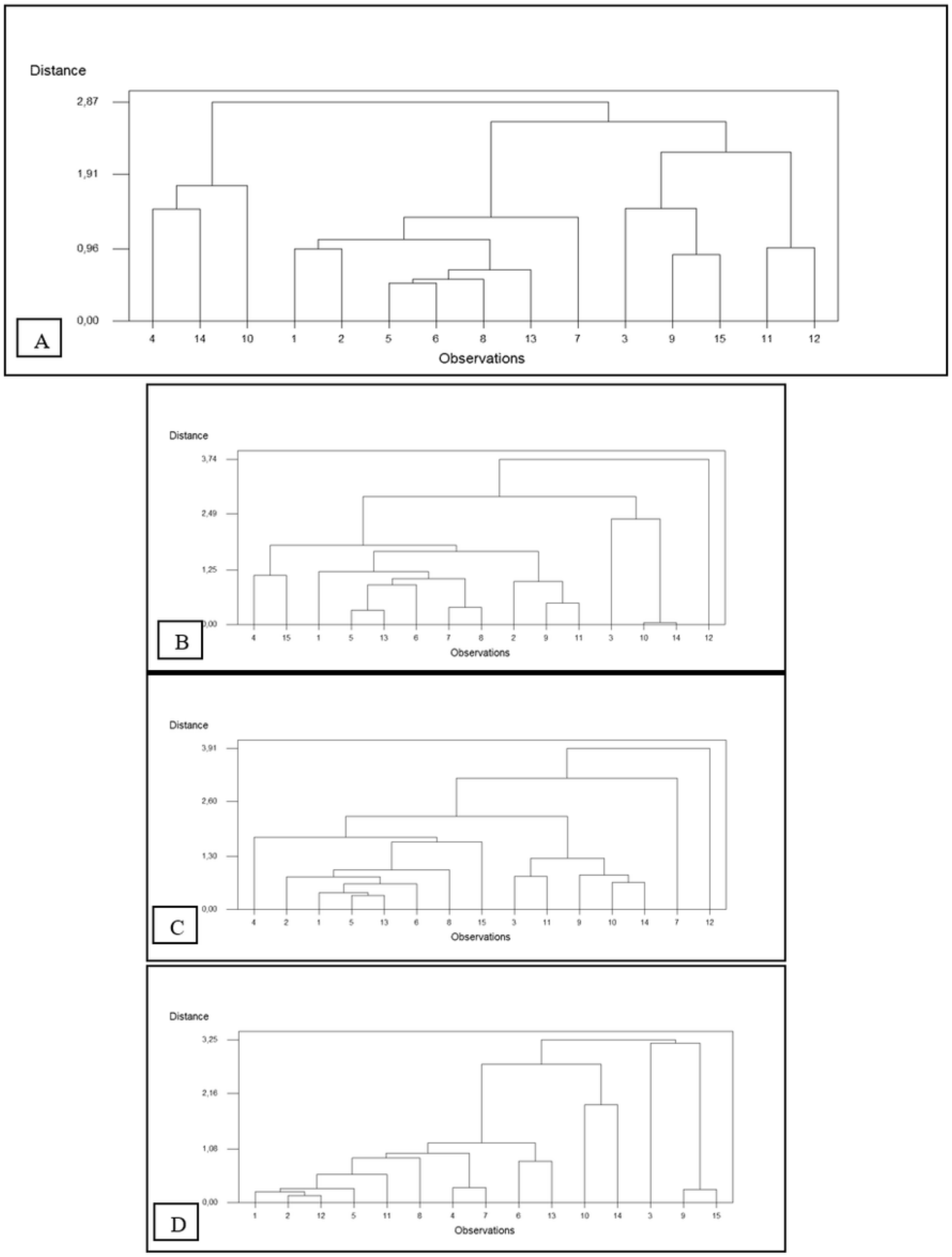

Figure 6

The clustering of heavy metal accumulation in mangrove vegetation

Note :

A (Heavy metal accumulation), B (Hg accumulation), C (Cd Accumulation), D(Zn accumulation)

1 = Aegiceras corniculatum, 2 = Aegiceras floridum , 3 = Avicennia marina , 4 = Bruguiera gymnorrhiza , 5 = Bruguiera sexanggula. 6 = Ceriops tagal , 7 =Excoecaria agallocha, 8 = Hibistus tiliaceus, 9 = Melaluca Leucadendron, 10 = Nypa frutican, 11 = Rhizophora apiculate, 12 = Rhizophora mucronata , 13 = Rhizophora stylosa, 14 = Sonneratia alba, 15 = Xylocarpus granatum