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DISTRIBUTION OF HEAVY METALS IN MANGROVE SEDIMENT AT THE THA CHIN ESTUARY, SAMUT SAKHON PROVINCE, THAILAND

การกระจายของโลหะหนักในดินตะกอนป่าชายเลน ณ ปากแม่น้ำท่าจีน จังหวัดสมุทรสาคร ประเทศไทย

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Abstract

The concentrations of selected heavy metals (Cd, Cu, Pb and Zn) at a depth of 0-30 cm sediments from 11 stations in a mangrove forest at Bang Ya Praeg, Tha Chin Estuary, Samut Sakhon Province, Thailand were examined. The ranges of heavy metals, in mg/kg dw, were as follows: Cd (0.035 to 0.070), Cu (7.90 to 21.91), Pb (11.91 to 25.74) and Zn (55.99 to 75.05). The dry season (April 2009) had higher levels of heavy metals than the wet season (August 2009). Heavy metals in the study area had the following order of prevalence: Zn > Pb > Cu > Cd. A significant relationship was observed between heavy metals and the chemical and physical properties of the sediment, indicating that complexation with organic materials may play an important role in the distribution patterns of these metals. Analyses of the metals in various grain-size fractions revealed that the finest fraction had significant relationship with heavy metal content, especially that of Cd and Zn.

Keywords: Mangrove Sediments, Heavy Metals, Tha Chin Estuary, Organic Matter, Samut Sakhon, Thailand

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บทคัดย่อ

ทำการตรวจวัดความเข้มข้นของโลหะหนัก (แคดเมียม, ทองแดง, ตะกั่ว และสังกะสี) ในตะกอนดินที่ระดับความลึก 0-30 ซม. จาก 11 สถานี ในบริเวณป่าชายเลน อำเภอบางหญ้าแพรก ปากแม่น้ำท่าจีน จังหวัดสมุทรสาคร ประเทศไทย ช่วงของโลหะหนักหน่วยเป็น มก./กก. น้ำหนักแห้งเป็นดังนี้ แคดเมียม 0.035 ถึง 0.070, ทองแดง 7.90 ถึง 21.91, ตะกั่ว 11.91 ถึง 25.74 และ สังกะสี 55.99 ถึง 75.05 ในฤดูแล้ง (เมษายน 2553) มีระดับความเข้มข้นของโลหะหนักสูงกว่าฤดูฝน (สิงหาคม 2553) โลหะหนักในพื้นที่ศึกษา เรียงลำดับการตรวจพบดังนี้ สังกะสี>ตะกั่ว>ทองแดง>แคดเมียม ความสัมพันธ์ระหว่างความเข้มข้นของโลหะหนักกับสมบัติของดินทั้งด้านกายภาพและเคมี มีนัยสำคัญพบว่า ในสัดส่วนความซับซ้อนของอินทรียวัตถุอาจมีบทบาทต่อรูปแบบกระจายตัวของโลหะหนัก การวิเคราะห์โลหะหนัก ขนาดของตะกอนดินต่าง พบว่า โลหะหนักมีความสัมพันธ์ในขนาดของตะกอนที่ละเอียดที่สุดอย่างมีนัยสำคัญ โดยเฉพาะแคดเมียม และสังกะสี

คำสำคัญ: ตะกอนดินป่าชายเลน, โลหะหนัก, ปากแม่น้ำท่าจีน, อินทรียวัตถุ, สมุทรสาคร, ไทย

Introduction

Mangrove forest ecosystems are very important not only to sustain environment but also to provide food sources to many living things. These ecosystems act as bridges between fresh water ecosystem and marine ecosystem, hence they serve as diverse habitats for a large amount of residents. Even though they are providing direct and indirect benefits to human being, it is being degraded in many areas. Tha Chin River is one of four main rivers in Thailand. Along the bank of Tha Chin River from Chinat Province to the Tha Chin Estuary at Samut Sakorn Province are crowded with various human activities, settlements, agriculture and industries. These are the causes of very much polluted at Tha Chin Estuary [1,2,3].

Analysis of pollutants in sediments has become an important tool for tracing anthropogenic water pollution[4] because some pollutants are absorbed by fine-grained particles and materials in suspension. After flocculation and sedimentation, bottom deposits are enriched in pollutants by a factor of 1000 or more[5]. The objective of this study was to determine the distributions of Cd (cadmium), Cu (copper), Pb (lead) and Zn (zinc) in mangrove sediments. Compounds containing these heavy metals are widely used in industrial and agricultural processes. The compounds can be unstable and can contaminate the environment. The chemical and physical properties of organic matter, percent organic matter (%OM), cation exchange capacity (CEC) and soil texture of the sediment were also analyzed.

Materials and Methods

Sampling Site and Sediment Collection

The sampling site for this study is located at N 13° 30′ 10.37′′ and E 100° 16′ 15.88′′ in a mangrove forest in the Gulf of Thailand. The mangrove forest at the

Mangrove Forest Learning and Development Center 2 is located at Bang Ya Praeg, Tha Chin Estuary, Samut Sakhon Province. The area is part of the National Conservation Forest on the west side of Tha Chin Estuary. Sediment samples were collected for determination of chemical and physical properties as well as for analysis of heavy metals. Transect lines were used in the study (Figure 1). Three transect lines were placed 100 m apart, perpendicular to the river bank. On each transect line, sample plots were placed every 50 m. Three core samples were collected from each plot at a depth of 0-30 cm. Sediment samples were collected during both the dry (20th April 2009) and wet (22nd August 2009) seasons. Sediment samples were kept in tightly closed plastic bags.

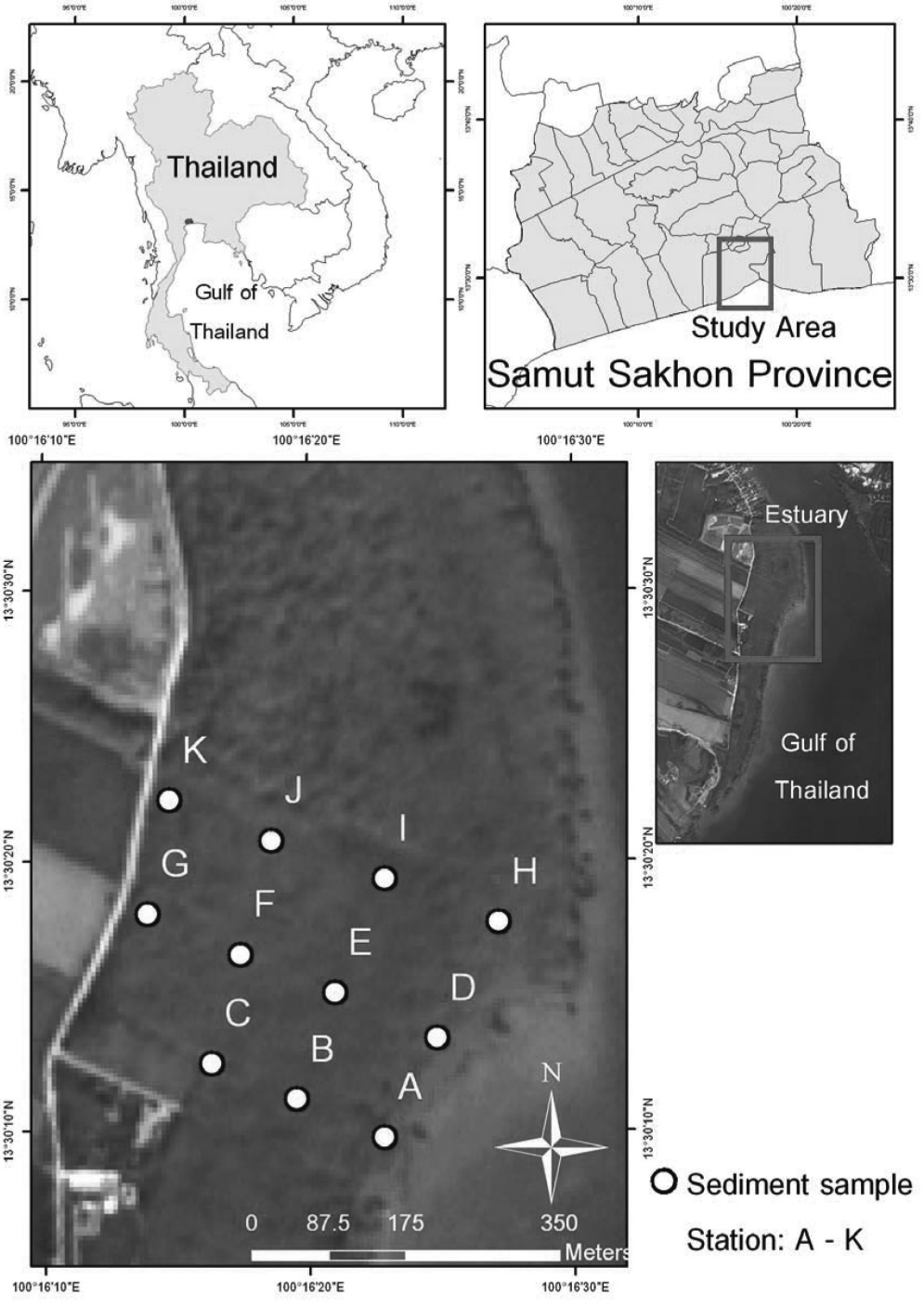

Figure 1 Map of the study area showing the location of the sample plots

Analytical Method

In the laboratory, the samples were dried at room temperature and passed through a 2 mm sieve to remove larger particles. The samples were then dried in an oven at 70ºC. Soil texture was determined with the hydrometer method[6]. CEC in me/100 g, was determined using a NaOAC 1N, pH 8.2, processed with a mechanical vacuum extractor[7]. The %OM was determined by the Walkley and Black Rapid Titration Method[8]. All samples for this study were analyzed in triplicate.

For heavy metal analysis, approximately 0.5 g of the dried, homogenized sediment sample was transferred into digestion vessels. The sediments were digested with HNO_3 and HCl using a milestone microwave digestion system, model MLS-1200 MEGA, followed by the addition of 50 ml of distilled water[9]. The concentrations of Cu and Zn in the filtered and digested samples were determined by flame atomic absorption spectrometry (FAAS) using a Varian atomic absorption spectrophotometer, model spectra AA-55. Pb and Cd concentrations were determined by graphite furnace atomic absorption spectrometry (GFAAS) using a Varian spectrAA 220Z graphite furnace atomic absorption spectrophotometer. Statistical analysis was performed using SPSS Version 11.5 for Windows. Pearson correlation was used in the study.

Results and Discussion

Heavy Metals Concentrations in Sediment

Heavy metals are discharged into the river from both point and non-point sources. The Tha Chin tributaries are occupied by agricultural features such as rice fields, vegetable farms, fruit orchards, pig farms, duck farms and fish farms, in addition to several industries such as chemical plants, paper factories and storage battery factories. Agricultural runoff, aquacultural effluent and domestic effluent enter the river through drains and tributaries. In addition, industries located in the vicinity of tributaries discharge effluent into the river. Pesticides such as endosulfan, betahexachlorocyclohexane (BHC), endrin, malathion, parathion, monocrotophos, carbofuran and carbaryl are used for pest control in the agricultural lands adjacent to the Tha Chin River [10]. Hence, the Tha Chin River is likely to be contaminated by pesticides and heavy metals.

Cd in sediment:

The distribution of Cd in sediments collected from 11 stations during the dry and wet seasons is presented in Figure 2.

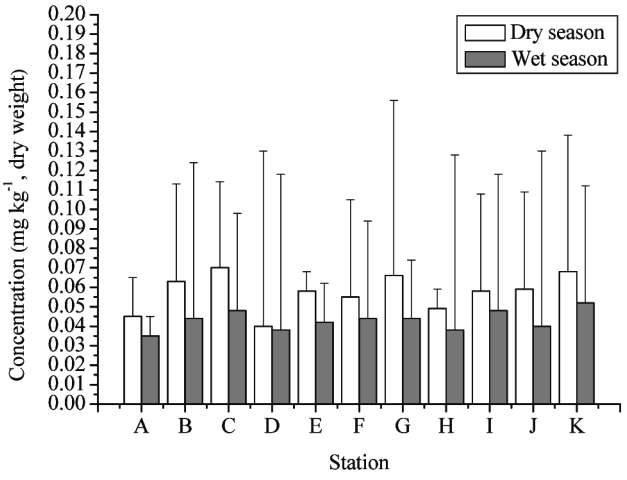

Figure 2 Comparison of Cd concentrations during the dry and wet seasons for each station.

Cadmium occurs naturally in the Earth's crust and is released into the environment from point sources, such as industrial discharges, and from non-point sources, such as agricultural runoff. The important source of Cd emission is the production of artificial phosphate fertilizer[11]. The concentration of Cd was higher during the dry seasons than the wet season. The concentration of Cd in the sediment was higher at landward locations and gradually decreased seaward.

Cu in sediment:

The distribution of Cu in sediment collected from 11 stations during dry and wet seasons is presented in Figure 3.

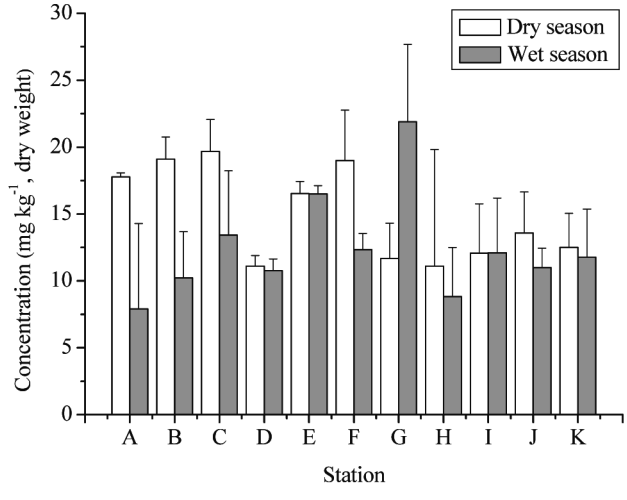

Figure 3 Comparison of Cu concentrations during the dry and wet seasons for each station.

When Cu ends up in soil, it attaches strongly to minerals and organic matter. As a result, it does not travel very far after release. In surface waters, Cu can travel great distances on suspended sludge particles or as free ions[12]. The concentration of Cu is higher during dry seasons than the wet season, except at Station G, where the concentration of Cu during wet season was higher than dry seasons. The sediment concentration of Cu did not show obvious variation between landward and seaward locations.

Pb in sediment:

The distribution of Pb in sediment collected from 11 stations during dry and wet seasons is presented in Figure 4.

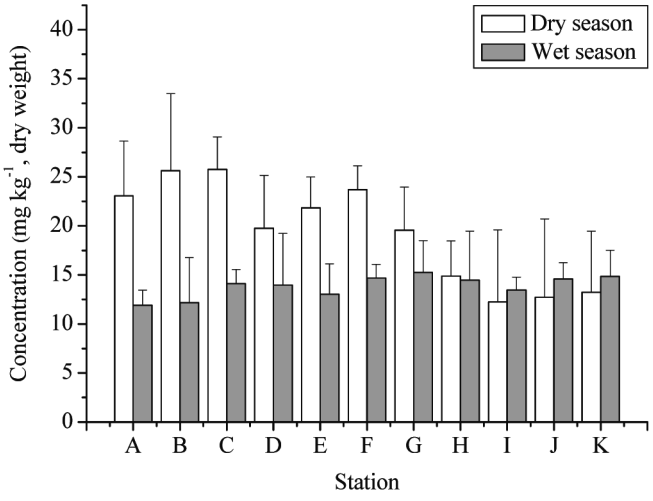

Figure 4 Comparison of Pb concentrations during the dry and wet seasons for each station.

The major source of Pb is gasoline because tetraethyl lead ($PbEt_4$) is used in some grades of petrol (gasoline). Pb is also a major component of the lead-acid extensively used in batteries[13]. The concentration of Pb is higher during the dry seasons than the wet season, except at Stations I, J and K, where the concentration of Pb during the wet season was higher than during the dry season. The sediment concentration of Pb during the dry season was lower at landward locations and gradually increased in the seaward direction. There was no difference in concentration for all landward stations during the wet season.

Zn in sediment

The distribution of Zn in sediment collected from 11 stations during dry and wet seasons is presented in Figure 5.

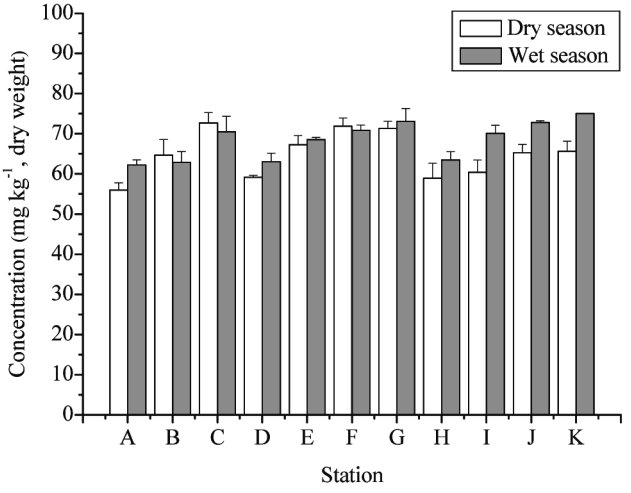

Figure 5 Comparison of Zn concentrations during the dry and wet seasons for each station.

The concentration of Zn was lower in the dry season than in the wet seasons, except at Stations B, C and F, where the concentration of Zn was higher in the dry season than in the wet season. The concentration of Zn in the sediment was higher at landward stations and gradually decreased seaward. This might be because the adsorption of zinc increases with an increasing amount of adsorbent and decreases with adsorbent particle size. The geochemical phases of iron and manganese oxides act as active support materials for the adsorption of zinc ions [14].

For finding difference between heavy metals concentration and both seasons using F – test found that Cd had significantly difference ($p < 0.05$) between dry season and wet season at Stations A, I and J. In the same way, the concentration of Cu at Station A had a significant variation ($p < 0.05$) between dry season and wet season. Stations G, I and J had a significantly ($p < 0.05$) contrary of Pb concentration. Finally, the concentration of Zn at Stations D and J had a significant difference ($p < 0.05$) between dry and wet season.

Table 1 shows the average concentrations of Cd, Cu, Pb and Zn in mangrove sediments.

The concentrations of the four heavy metals were higher at landward stations and gradually decreased seaward. Zn concentrations were higher at the back of the mangrove forest areas. The maximum concentration of Zn was observed at Station K (75.05±0.04 mg/kg, dw), collected during the wet season, and the minimum was at Station A (55.99±1.81 mg/kg, dw), collected during the dry season. The maximum concentration of Pb was observed at Station C (25.74±3.32mg/kg, dw), collected during the wet season, and the minimum was at Station A (11.91±1.55 mg/kg, dw), collected during the dry season. The highest concentration of Cu was observed at Station G (21.91±5.77 mg/kg, dw), and the lowest concentration of Cu was found at Station A (7.90±6.39 mg/kg, dw); both samples were collected during the wet season. The maximum concentration of Cd was observed at Station C (0.07±3.42 mg/kg, dw), collected during the wet season, and the minimum was found at Station A (0.035±3.42 mg/kg, dw), collected during the dry season.

Table 1 Heavy metals concentration (mg/kg) in sediments from the mangrove forest

Station	Cd		Cu		Pb		Zn	
	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season	Wet season
Line 1								
A	0.045±0.02	0.035±0.01	17.77±0.31	7.90±6.39	23.04±16.10	11.91±1.55	55.99±1.81	62.25±1.26
B	0.069±0.05	0.044±0.08	19.10±1.65	10.23±3.44	25.64±7.84	12.17±4.61	64.70±7.87	62.87±2.70
C	0.070±0.04	0.048±0.05	19.67±2.40	13.43±4.80	25.74±3.32	14.10±1.45	72.71±2.53	70.54±5.87
Line 2								
D	0.040±0.09	0.038±0.08	11.10±0.79	10.77±0.87	19.75±5.40	13.97±5.26	59.17±0.45	63.06±2.11
E	0.058±0.01	0.042±0.02	16.53±0.90	16.50±0.61	21.84±3.14	13.02±3.10	67.26±2.26	68.56±0.55
F	0.055±0.05	0.044±0.05	19.00±3.78	12.33±1.21	23.69±2.43	14.67±1.41	71.93±2.04	70.88±1.32
G	0.066±0.09	0.044±0.03	11.67±2.64	21.90±5.77	19.56±14.90	15.25±3.24	71.37±1.79	73.13±3.17
Line 3								
H	0.049±0.01	0.038±0.09	11.10±8.72	8.83±3.67	14.87±11.61	14.47±5.00	58.95±3.71	63.49±2.02
I	0.058±0.08	0.048±0.07	12.07±3.69	12.10±4.09	12.24±7.34	13.45±1.33	60.44±2.98	70.12±1.96
J	0.059±0.05	0.040±0.09	13.57±3.08	11.00±1.44	12.73±7.98	14.58±1.67	65.30±2.08	72.84±0.39
K	0.068±0.07	0.052±0.06	12.50±2.56	11.77±3.59	13.21±6.26	14.85±2.67	65.66±2.48	75.05±0.04
Soil Quality[15] Standards	Not exceed 37 mg/kg		-		Not exceed 400 mg/kg		-	
World median[16]	0.35 mg/kg		30 mg/kg		35 mg/kg		90 mg/kg	

Concentration expressed as mean ± SD (mg/kg dw, n = 3)

The order of the concentrations of all heavy metal averages for both seasons was: Zn (66.65±2.53 mg/kg, dw) > Pb (16.58±3.85 mg/kg, dw) > Cu (13.67±1.75 mg/kg, dw) > Cd (0.05±0.01 mg/kg, dw). These values are similar to those reported for the same metals in sediment samples from the Tha Chin Estuary, Thailand. The mean concentrations of Cd, Cu, Pb and Zn were 1.8, 58.3, 88.5 and 144.8 mg/g, respectively. Cd was most prevalent in the exchangeable fraction; Cu and Pb were dominantly found bound to the organic matter fraction, and the highest percentage of Zn was found bound to Mn and Fe oxide fractions [17].

Chemical and physical properties of sediment

The mangrove sediments had high to very high ranges (3.36 – 6.10) of %OM. These high levels in the mangrove sediments are due to decomposition of dead organisms as well as to mangrove detritus and anthropogenic inputs. Waves and currents can transport the organic matter to the low-energy mangrove forest, where it is deposited. Microbial degradation of the organic matter in mangrove sediments generally removes all oxygen from the subsurface, creating ideal conditions for bacterial sulfate reduction [18]. The mean percentage of OM in front of the mangrove forest (Stations A, D and H) was 4.35, 4.14 and 4.22, respectively, and landward of the mangrove forest (Stations B, C, E, F, G, I, J and K) the %OM was 4.40, 5.42, 5.04, 4.41, 4.76, 5.20, 4.90 and 5.06, respectively. Stations landward of the mangrove forest generally had higher %OM, while stations seaward of the forest had lower %OM. Wave and tidal frequency may therefore influence the movement of OM from land to sea.

CEC of the sediment in both dry and wet seasons was analyzed. The moderate to high CEC values ranged from 11.15 - 29.03 $cmol_c\ kg^{-1}$, correlating significantly ($r = 0.375$, $P < 0.01$) with the presence of clay. The content of “free” iron (dithionite-extractable) was considerably higher in clay-rich horizons[19].

For finding difference between chemical properties of sediment and both of seasons by using F – test statistical revealed that OM had significantly difference ($p < 0.05$) between dry season and wet season at Station A. In the same way, CEC level at Stations D and J had a significant variation ($p < 0.05$) between dry and wet season. The other station had not significantly different.

Sediment characteristics for each station are shown in Table 2. The percentage of sand at Station A was lower than at Stations D and H. On the other hand, the percentage of silt and clay at Station A was higher than at Stations D and H. These results reveal that Stations A, D and H influenced the deposition of sediment particles. Station H was located closer to the estuary than Stations A and D. Sediment was transported by sea water. Larger particles would have been deposited more readily than the smaller particles.

Table 2 Variation of sediment texture in dry (April 2009) and wet seasons (August 2009)

Station	% Sand (dry season)	% Silt + Clay (dry season)	% Sand (wet season)	% Silt + Clay (wet season)
Line 1				
A	26.80	73.20	39.20	60.80
B	43.72	56.28	44.34	55.66
C	32.08	67.92	47.30	52.70
Line 2				
D	32.54	67.46	44.87	55.13
E	42.58	57.42	41.76	58.24
F	42.80	57.20	41.80	58.20
G	40.18	59.82	44.52	55.48
Line 3				
H	48.16	51.84	47.12	52.88
I	43.94	56.06	44.52	55.48
J	39.94	60.06	48.24	51.76
K	48.80	51.20	45.45	54.55

Correlation of heavy metals concentration and physical-chemical properties of sediment

The correlation between four heavy metals concentration and the chemical and physical properties in the sediment were calculated using the Pearson correlation, as shown in Tables 3-4.

Table 3 Correlation coefficient matrix showing inter-element and element- physicochemical relationships in the sediment (dry season; April 2009)

	Sand	Silt	Clay	OM	CEC	Cd	Cu	Pb	Zn
Sand	1.000								
Silt	-.989**	1.000							
Clay	.771**	-.856**	1.000						
OM	.166	-.236	.440	1.000					
CEC	.032	-.122	.416	.153	1.000				
Cd	.463	-.552*	.764**	.701**	.518	1.000			
Cu	-.327	.345	-.346	-.068	.387	.128	1.000		
Pb	-.494	.509	-.472	-.211	.399	-.059	.809**	1.000	
Zn	.166	-.268	.579*	.431	.874**	.815**	.386	.344	1.000

* Correlation is significant at $p < 0.05$ ** Correlation is significant at $p < 0.01$ (n = 11)

Table 4 Correlation coefficient matrix showing inter-element and element- physicochemical relationships in the sediment (wet season; August 2009)

	Sand	Silt	Clay	OM	CEC	Cd	Cu	Pb	Zn
Sand	1.000								
Silt	-.666*	1.000							
Clay	-.197	-.600*	1.000						
OM	.179	-.355	.274	1.000					
CEC	.349	-.678*	.516	.374	1.000				
Cd	.308	-.476	.296	.489	.009	1.000			
Cu	.004	-.172	.221	.597*	.711**	.000	1.000		
Pb	.564*	-.697**	.312	.279	.743**	.161	.434	1.000	
Zn	.320	-.714**	.595*	.715**	.703**	.474	.562*	.693**	1.000

* Correlation is significant at $p < 0.05$ ** Correlation is significant at $p < 0.01$ (n = 11)

The concentration of Cd in the sediment was significantly and positively correlated ($p < 0.01$) with OM level in samples collected during the dry season. The concentrations of Cu and Zn were also positively correlated ($p < 0.05$ and $p < 0.01$) with the OM level in sediment collected during the wet season. The OM in the soil sediment is very important in controlling metal mobility. The OM in the soil sediment is mainly supplied from fresh litter leaching and also from decomposition of OM on the soil surface [20,21,22]. The capacity of the soil sediment to dissolve metals by forming organic complexes (i.e., complexation capacity and stability of the complexes) has also been measured [23,24,25,26]. Until now, OM already present in the soil was regarded as the source of important metal carriers.

The concentration of heavy metals in the sediment collected during the wet season was significantly and positively correlated ($p < 0.01$) with CEC. It seems that the complexation of heavy metals is governed by the CEC. During the dry season, only the concentration of Zn was positively correlated with CEC in the sediment. The lack of significant correlation between Cd and CEC may indicate that the complexation of Cd is not controlled by the CEC. The concentrations of Cd and Zn in the sediment were significantly and positively correlated (Cd, $p < 0.01$, Zn, $p < 0.05$) with the clay content. The total concentration of heavy metals reflects variations in sediment texture.

Examination of heavy metals concentration in fractions of mangrove sediments with different grain sizes revealed high concentrations of Cd and Zn in the clay texture compared with other textures. This may indicate that finer particles retain some metals better than coarse particles and that metal concentrations in the sediment are inversely related to particle size [27]. In fact, metals are most often associated with fine grains [28]. Traditionally, the fine-grained fraction of the sediment has been used to examine metal contamination in sediments[29]. The clay fraction is known to be the most important substrate for metal attachment, and metal concentrations tend to increase from sand to silt [30]. Haque and Subramanian [31] reported that metal adsorption capacity increased with decreased grain size (sand < silt < clay) due to increased surface area.

Conclusion

The study site at Tha Chin Estuary is covered by mangrove forest. All wastes wherever they come from agriculture, farming, industrial etc., are flowed to Tha Chin River. Eventually, all those waste are

piled up at this estuary where screens all wastes before they are flowed to the sea. Moreover, heavy metals are kept in soil sediment. This usefulness of this mangrove forest can prevent heavy metal contaminated in soil which will cause problem to fish shores. In this study, the factors responsible for accumulation of heavy metals were (i) OM content, (ii) CEC and (iii) sediment texture. Concentrations of four heavy metals were higher at the landward stations and decreased gradually toward the sea. The abundance of the heavy metals had the following order: Zn > Pb > Cu > Cd. Future research and analysis of mangrove ecology should include cambium activity analysis, which could indicate how distributions of heavy metals and the chemical and physical properties of sediment may influence mangrove plant growth.

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