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Heavy Metals Contamination in Sediments along the Eastern Coast of the Gulf of Thailand

Waewtaa Thongra-ar [a], Chaluay Musika [a], Wanchai Wongsudawan [a] and Arvut Munhapol [a]

[a] *Institute of Marine Science, Burapha University, Chonburi 20131, Thailand*

Abstract

Levels of Hg, Cd, Pb, Zn, Cu, Ni, Fe and Mn in surface sediments were investigated in 52 stations along the Eastern Coast of the Gulf of Thailand. The sediment samples were collected in March 2004. Concentrations of the heavy metals ranged from 0.005 - 0.121 μg g^{-1} for Hg, <0.006 - 0.19 μg g^{-1} for Cd, 1.69 - 66.3 μg g^{-1} for Pb, 7.48 - 131 μg g^{-1} for Zn, 14.4 - 103 μg g^{-1} for Cu, <0.64 - 79.9 μg g^{-1} for Ni, 1.17 - 92.8 mg g^{-1} for Fe and 0.03 - 1.71 mg g^{-1} for Mn. Only Pb and Cu in the sediments exceeded the effects range low of the proposed marine and coastal sediment quality guidelines for Thailand. The calculated geoaccumulation index (I_{geo}) showed that the sediments were moderately polluted with Pb in some locations, particularly at Map Ta Phut Industrial Estate, and were slightly polluted with Cu, Zn and Mn at some sampling stations. All metals (except Cu) were associated with each other in the sediments. Organic matter, clay and silt were the major sediment components responsible for most metals sorbed in the study area.

Keywords: heavy metal; sediment; pollution; geoaccumulation index; the Gulf of Thailand

1. Introduction

Heavy metal contamination is an environmental problem in both developing and developed countries throughout the world. Human activities increase the release of heavy metals to natural environment because most metals are widely used in a variety of industrial and agricultural applications. High concentrations of heavy metals could have toxic effects on living organisms, accumulate in marine food chain and affect human health through consumption of contaminated seafood. After being discharged into aquatic environments, heavy metals are subsequently deposited into bottom sediments. However, under some conditions, they can be released back to the water column as a result of physical, chemical or biological processes (Bakan and Balkas, 1999). Thus, sediments serve as a potential risk source as well as ultimate sink of heavy metals in aquatic environments and are considered to be a good environmental indicator of metal pollution (Soares *et al.*, 1999).

The Eastern Coast of the Gulf of Thailand has a total coastline of approximately 500 kilometers, covering 5 Provinces: Chachoengsao, Chon Buri, Rayong, Chanthaburi and Trat. This regional coast has various kinds of anthropogenic activities including agriculture, fisheries, tourism, industrial and urban communities. There are two industrial estates located on the coast as a result of the Eastern Seaboard (ESB) Development Programme which was first introduced during the 5th National Economic and Social Development Plan (1981-1984). Map Ta Phut Industrial Estate, Rayong Province, for example, has been developed as the heavy industry zone with a gas separation plant, petroleum refining plants, petrochemical industries, chemical fertilizer, iron & steel, and power plants. Laem Chabang Industrial Estate was established in Chon Buri Province for light industries and an international deep sea port. Since the east coast has been developing with a high expansion rate of industrialization and urbanization, these activities will substantially increase the potential risk of heavy metal pollution along the coast which will subsequently degrade natural resources and aquatic environments. Lately, this area has faced the problem of mercury discharged by industries especially around Map Ta Phut Industrial Estate (Chongprasith and Wilairatanadilok, 1999).

Therefore, the present study was undertaken 1) to investigate current metal concentrations in sediments of the Eastern Coast of the Gulf of Thailand and the relationship between these contaminants and some sediment characteristics, and 2) to evaluate the potential for heavy metal pollution in the sediments from anthropogenic inputs.

2. Materials and Methods

2.1. Sediment Sampling

Surface sediments were collected from 52 stations along the Eastern Coast of the Gulf of Thailand. The sampling stations were divided into 4 zones: A, C, E and G , emphasizing on industrial and aquacultural areas (Fig. 1). At each station, composite sediments of 3 subsamples were collected in March 2004 using a modified Peterson grab. The samples were freeze-dried, manually sieved through a 2-mm mesh size nylon sieve and then homogenized using agate mortar and pestle. Duplicate subsamples of each composite sediment were taken for metal analysis and sediment characterization.

2.2. Heavy Metal Analysis

Heavy metals determined included Hg, Cd, Pb, Zn, Cu, Ni, Fe and Mn. The sediment samples were microwave digested using a mixture of aqua regia (HNO_3 : HCl, 1:3 v/v) and hydrofluoric acid (HF) (Loring and Rantala, 1992), followed by instrumental analysis by flame atomic absorption spectrophotometer Model AAnalyst 100, Perkin-Elmer (Zn, Cu, Ni, Fe, Mn) or graphite furnace atomic absorption spectrophotometer Model 4110 ZL, Perkin-Elmer (Cd, Pb). For Hg analysis, the samples were digested with a mixture of 9:1 (v/v) HNO_3/HCl (Randlesome and Aston, 1980) and determined by a flow injection mercury hydride system (FI-MHS, Model FIAS 100) combined with 4110 ZL atomic absorption spectrophotometer (Perkin-Elmer).

The precision and accuracy of the analytical technique were assessed by analysis of marine sediment reference material (MESS-3) from the National Research Council of Canada and found to be very close to the certified values (Table 1).

2.3. Sediment Characterization

Selected sediment characteristics were determined as follows: pH in water at a sediment to solution ratio of 1:2.5, calcium carbonate by the gravimetric method for loss of carbon dioxide (Goh *et al.*, 1993), organic matter (OM) by acid-dichromate oxidation method (Nelson and Sommers, 1982), cation exchange capacity (CEC) by the ammonium acetate saturation method (Chapman, 1965), Fe and Mn oxides by dithionite-citrate method (Ross and Wang, 1993) and particle size distribution by the hydrometer method (English *et al.*, 1994).

3. Results and Discussion

3.1. Metal Concentrations

Concentrations of the heavy metals in surface sediments of the Eastern Coast of the Gulf of Thailand are presented in Table 2. The highest average concentrations of Hg, Zn and Mn in the sediments were observed in Bangpakong to Angsila area (zone A) with relatively high concentrations found at Chon Buri Bay. This was possibly due to industrial and domestic wastes discharged from Bangpakong estuary and municipal wastewater discharges from Chon Buri municipality, a densely-populated area of the city. The highest average concentrations of Cd and Pb were found in Map Ta Phut area (zone

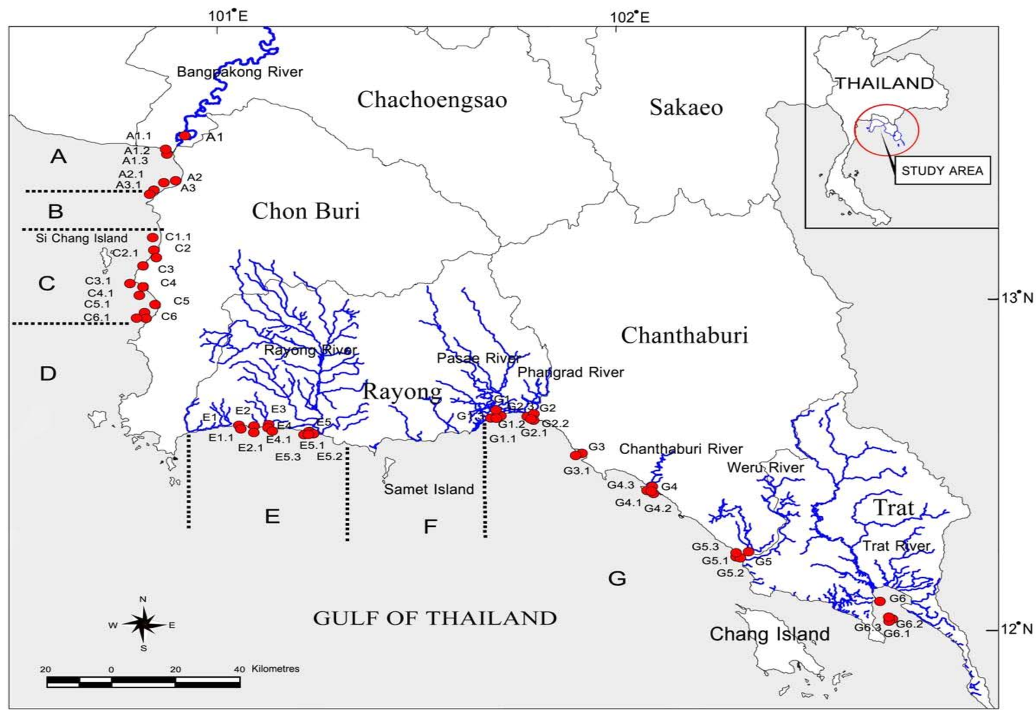

Figure 1. Map of study area and sampling stations. Details of sampling sites are given in text.

Table 1. Results of analysis of marine sediment reference material (MESS-3)

Heavy Metal	Certified Value (µg g^{-1} dry weight)	Measured Value (µg g^{-1} dry weight)	% Recovery	% RSD	Method Detection Limit (µg g^{-1} dry weight)
Hg	0.091 ± 0.009	0.098 ± 0.009 (n=5)	98.9 - 124.2	9.4	0.004
Cd	0.24 ± 0.01	0.26 ± 0.02 (n=5)	94.5 - 118.2	7.7	0.006
Pb	21.1 ± 0.7	18.6 ± 0.7 (n=3)	84.0 - 90.4	3.8	0.62
Zn	159 ± 8	150.0 ± 1.3 (n=5)	93.6 - 95.3	0.9	1.28
Cu	33.9 ± 1.6	38.1 ± 3.2 (n=5)	105.2 - 128.0	8.4	1.92
Ni	46.9 ± 2.2	39.5 ± 1.8 (n=4)	78.9 - 87.9	4.5	0.64
Fe	43,400 ± 1100	44,182 ± 285 (n=5)	100.8 - 102.5	0.7	0
Mn	323 ± 12	265.2 ± 27.6 (n=5)	72.7 - 88.6	10.4	19.0

E), while those of Cu, Ni and Fe were observed in Chanthaburi to Trat area (zone G), especially in the estuaries. We also found that Pb, Zn and Cu in sediments of Map Ta Phut were slightly higher than those in Laem Chabang , whereas the other metal concentrations were not much different between the two areas.

Comparing the metal concentrations in this study with those of previous studies (Terai *et al*., 1995), it was found that most metals (Pb, Zn and Cu) in the sediments had increased during the past 10 years due to the industrialization and economic development. However, the present metal concentrations in the Eastern Coast sediments were significantly lower than those in other industrialized/urban ports in the Asian countries such as Port of Singapore, Manila Bay (Philippines), Jakarta Bay (Indonesia), Mason Bay (Korea), Western Xiamen Bay (China) and Hong Kong coast (Table 3).

With respect to the proposed marine and coastal sediment quality guidelines for Thailand (PCD, 2006), only concentrations of Pb and Cu in the sediments exceeded the effects range low (ERL), but did not exceed the effects range median (ERM) of the guidelines. The high Pb concentrations were found at Map Ta Phut Industrial Estate and Rayong estuary, whereas high Cu was observed at several stations in the estuaries in Chanthaburi Province and Kung Kraben Bay (Figure 2). Only sediment Ni exceeded the ERM of the guidelines which was found at Trat estuary. There are no guideline values for Fe and Mn. Although Hg is a pollutant of

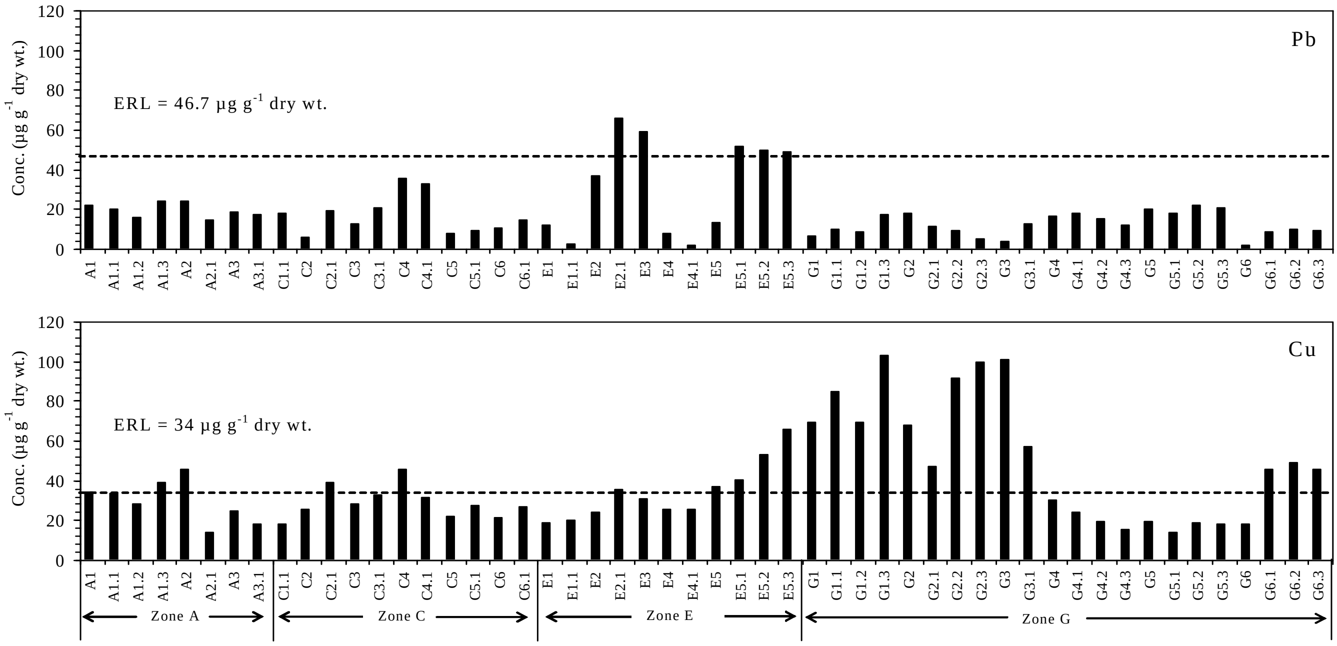

Figure 2. Pb and Cu concentrations in sediments of the Eastern Coast of the Gulf of Thailand. Station codes are the same as shown in Figure 1.

Table 2. Sediment heavy metals and selected sediment characteristics of the Eastern Coast of the Gulf of Thailand in March 2004

Zone*	Location		Hg	Cd	Pb	Zn	Cu	Ni	Fe	Mn	pH	$CaCO_3$	OM	CEC	Total Oxides (mg g^{-1})		Particle Size Distribution		
			(µg g^{-1} dry wt.)						(mg g^{-1} dry wt.)			(%)	(%)	(cmol kg^{-1})	Fe oxides	Mn oxides	Sand (%)	Silt (%)	Clay (%)
A	Bangpakong River - Ang Sila, Chon Buri Province (8 stations)	Average	0.055	0.056	19.8	74.5	29.9	24.7	27.3	0.99	7.8	2.1	2.8	22.5	2.17	1.05	41.0	39.6	19.4
		Min	0.018	0.024	14.9	21.3	14.4	5.19	8.21	0.28	7.3	0.41	0.8	5.4	0.94	0.35	7.1	16.6	15.0
		Max	0.121	0.19	24.4	131	46.0	45.6	40.5	1.71	8.3	4.7	5.0	36.2	6.62	2.04	68.4	75.3	28.2
		GeoMean	0.045	0.044	19.5	60.1	28.0	17.5	22.0	0.83	7.8	1.3	2.5	17.6	1.80	0.86	33.8	34.7	18.9
C	Laem Chabang, Chon Buri Province (Bang Pra - Na Klua) (11 stations)	Average	0.036	0.047	17.3	30.7	29.1	10.9	11.9	0.26	8.4	7.2	2.2	9.9	0.97	0.17	59.6	29.2	11.2
		Min	0.010	0.008	5.80	7.48	17.9	<0.64	1.85	0.06	8.1	2.6	0.5	1.8	0.34	0.04	22.1	1.6	6.4
		Max	0.117	0.15	35.5	64.5	46.0	22.3	22.3	0.56	8.9	12.4	4.1	21.2	1.77	0.34	91.4	61.3	17.9
		GeoMean	0.028	0.032	15.0	25.1	28.1	7.3	9.25	0.21	8.4	6.5	1.7	6.9	0.86	0.14	52.5	16.2	10.4
E	Map Ta Phut, Rayong Province (Nong Fab - Rayong River) (11 stations)	Average	0.049	0.059	31.9	45.1	34.4	14.6	12.6	0.15	8.2	3.5	2.2	10.1	1.70	0.13	60.0	29.0	15.2
		Min	0.007	<0.006	1.69	7.49	18.8	4.15	1.17	0.03	7.3	0.48	0.2	1.4	0.19	0.02	18.1	0	6.0
		Max	0.116	0.17	66.3	107	66.3	32.3	24.5	0.41	8.9	8.8	4.3	26.0	5.82	0.32	93.4	65.0	37.8
		GeoMean	0.031	0.033	18.6	32.1	32.0	11.1	7.58	0.10	8.2	2.6	1.5	6.4	0.96	0.08	50.1	11.4	12.5
G	Chanthaburi - Trat (Prasae River - Trat River) (22 stations)	Average	0.034	0.032	12.6	38.1	50.5	28.2	32.3	0.51	7.8	2.3	2.5	12.5	4.42	0.31	45.3	40.2	14.5
		Min	0.005	<0.006	2.21	10.5	14.4	9.85	7.18	0.11	7.0	0.13	0.5	2.2	0.46	0.05	16.1	1.0	6.3
		Max	0.081	0.059	22.2	101	103	79.9	92.8	1.16	8.9	6.1	6.2	33.4	9.03	0.60	91.7	70.6	22.2
		GeoMean	0.024	0.027	11.1	32.7	40.8	23.1	26.7	0.43	7.8	1.5	1.9	9.7	3.67	0.25	36.9	28.6	13.5
Sediments of the Eastern Coast of the Gulf of Thailand (52 stations)		Average	0.041	0.044	18.8	43.6	39.4	21.1	23.0	0.45	8.0	3.5	2.4	13.0	2.77	0.36	50.8	50.8	14.7
		Min	0.005	<0.006	1.69	7.48	14.4	<0.64	1.17	0.03	7.0	0.13	0.2	1.4	0.19	0.02	7.1	7.1	6.0
		Max	0121	0.19	66.3	131	103	79.9	92.8	1.71	8.9	12.4	6.2	36.2	9.03	2.04	93.4	93.4	37.8
		GeoMean	0.029	0.031	14.4	33.8	33.8	14.9	15.9	0.30	8.0	2.2	1.8	9.1	1.82	0.21	41.8	21.5	13.2
Proposed Sediment Quality Guidelines for Thailand (PCD, 2006)		ERL	0.15	1.2	46.7	150	34	20.9	-	-	-	-	-	-	-	-	-	-	-
		ERM	0.71	9.6	218	410	270	51.6											
Crustal Average (Taylor, 1964)			0.08	0.2	12.5	70	55	75	56.3	0.95	-	-	-	-	-	-	-	-	-
Sediment Accumulation Index (I_{geo})			<0	<0	<0 - 1.26	<0 - 0.22	<0 - 0.22	<0	<0 - 0.09	<0 - 0.18	-	-	-	-	-	-	-	-	-

* Main activity in the area of zone A and G is aquaculture whereas that of zone C and E is industry.
ERL = effects range low
ERM = effects range median
GeoMean = geometric mean

Table 3. Comparison of sediment metal concentrations in the study area with those in other Asian countries (all concentrations in μg g^{-1} except Fe and Mn in mg g^{-1})

Study Area	Year of Study	Hg	Cd	Pb	Zn	Cu	Ni	Fe	Mn	References
Eastern Coast of the Gulf of Thailand	2004	0.005 - 0.121 (0.041)	<0.006 - 0.19 (0.044)	1.69 - 66.3 (18.8)	7.48 - 131 (43.6)	14.4 - 103 (39.4)	<0.64 - 80 (21.1)	1.17 - 92.8 (23.0)	0.03 - 1.71 (0.45)	This study
Eastern Coast of the Gulf of Thailand	1993	-	ND - 0.47 (0.14)	ND - 12.9 (3.0)	0.7 - 112 (23.3)	0.2 - 37.7 (8.6)	-	-	-	Terai *et al.* (1995)
Laem Chabang Industrial Estate	1998	<0.005 - 0.032	<0.1	<2 - 22	4 - 41	<2 - 12				PCD (1999)
Map Ta Phut Industrial Estate	1998	<0.005 - 0.172	<0.1 - 0.2	2 - 55	<2 - 179	<2 - 19	-	-	-	PCD (1999)
Ha Long Bay, Vietnam	No data	0.137 - 0.193 (0.178)	0.48 - 1.22 (0.75)	10.0 - 15.4 (12.4)	75.9 - 97.7 (88.0)	15.7 - 22.0 (18.8)	-	-	-	Cu (1999)
Port of Singapore	1994 - 1996	-	-	36 - 151 (87)	204 - 758 (497)	90 - 447 (257)	9 - 31 (20)	29 - 43 (34)	-	Orlic *et al.* (1997)
Manila Bay, Philippines	1994 - 1995	-	0.1- 0.43	8 - 80	79 - 283	46 - 346	45 - 86	20.7 - 30.5	1.07 - 2.06	Narcise and Jacinto (1997)
Hong Kong Coast	1998 - 2004	0.05 - 8 (0.19)	0.1 - 5.3 (0.33)	9 - 260 (53.56)	17 - 790 (147.73)	1 - 4,000 (118.68)	5 - 220 (24.72)	3.4 - 79 (28.62)	0.047 - 1.6 (0.52)	Zhou *et al.* (2007)
Western Xiamen Bay, China	2004 - 2005	-	0.11 - 1.01 (0.33)	45 - 60 (54)	65 - 223 (139)	19 - 97 (44)	25 - 65 (37.4)	30.8 - 48.1 (39.4)	-	Zhang *et al.* (2007)
Masan Bay, Korea	2004 - 2005	-	0.10 - 7.47 (1.24)	13.03 - 82.2 (43.97)	80 - 378.7 (206.3)	13.5 - 90.7 (43.4)	10.2 - 40.4 (28.8)	-	-	Hyun *et al.* (2007)
Jakarta Bay, Indonesia	1994	-	2.85 - 3.93	45 - 136	94 - 222	20 - 93	27.8 - 38.9	-	0.39 - 0.54	Hutagalung (1995)

The numbers in parenthesis are average values.
ND = non detectable

Table 4. Correlation matrix of heavy metals and selected sediment characteristics (n = 52)

	Hg	Cd	Pb	Zn	Cu	Ni	Fe	Mn	pH	$CaCO_3$	OM	CEC	Fe Oxides	Mn Oxides	Clay	Silt	Sand
Hg	1.000																
Cd	.715**	1.000															
Pb	.559**	.612**	1.000														
Zn	.811**	.628**	.474**	1.000													
Cu	-.128	.008	-.062	.003	1.000												
Ni	.587**	.368**	.129	.818**	.020	1.000											
Fe	.315*	.054	.071	.492**	.167	.509**	1.000										
Mn	.334*	.075	.036	.594**	.057	.515**	.697**	1.000									
pH	-.492**	-.219	-.255	-.619**	-.043	-.503**	-.697**	-.567**	1.000								
$CaCO_3$	-.279*	-.062	-.066	-.415**	-.251	-.256	-.363**	-.286*	.499**	1.000							
OM	.678**	.394**	.348*	.558**	-.206	.443**	.345*	.352*	-.549**	-.035	1.000						
CEC	.689**	.413**	.408**	.791**	-.125	.627**	.576**	.677**	-.661**	-.288*	.719**	1.000					
Fe Oxides	.402**	.055	.074	.390**	.022	.408**	.575**	.366**	-.679**	-.421**	.553**	.480**	1.000				
Mn Oxides	.349*	.108	.092	.610**	-.097	.470**	.440**	.803**	-.492**	-.277*	.401**	.721**	.175	1.000			
Clay	.551**	.418**	.602**	.667**	-.213	.504**	.336*	.383**	-.559**	-.331*	.515**	.633**	.431**	.395**	1.000		
Silt	.595**	.413**	.481**	.584**	-.070	.425**	.424**	.439**	-.643**	-.213	.671**	.756**	.554**	.434**	.638**	1.000	
Sand	-.629**	-.440**	-.448**	-.640**	.104	-.478**	-.439**	-.485**	.671**	.252	-.687**	-.789**	-.593**	-.477**	-.668**	-.982**	1.000

*, ** : Significant at $p = 0.05$ and 0.01, respectively

concern in Thailand, the concentrations found were still within the guidelines. The highest Hg concentrations (0.121 μg g^{-1}) were observed at Chon Buri Bay (zone A), followed by the concentrations of 0.117 and 0.116 μg g^{-1} found at Ao Udom (zone C) and Map Ta Phut Industrial Estate (zone E), respectively.

3.2. Sediment Characteristics and their Relationship with Metal Concentrations

Selected sediment characteristics are shown in Table 2. The sediments were slightly alkaline with pH ranging from 7.0-8.9. Sediment organic matter ranged from 0.2-6.2 % with high values (>4%) found in estuarine sediments, especially at Bangpakong, Weru and Trat estuaries where the sediments were also high in CEC (>20 cmol kg^{-1}). $CaCO_3$ content in the sediments was between 0.13-12.4 % with high values found in Laem Chabang area from Bang Pra - Na Klua (zone C). Higher Fe oxides contents were also observed in the sediments than Mn oxides. The high Fe oxides were mainly found at estuaries in Trat and Chanthaburi Provinces, whereas the high Mn oxides were mainly found at Bangpakong estuary.

All metals (except Cu) showed significantly positive correlation with Hg, indicating their common source and association with each other (Table 4). More importantly, Hg was more associated with Zn (r = 0.811) and Cd (r = 0.715) than the other metals. Most metal concentrations showed negative correlation with sediment pH, calcium carbonate and sand, whereas these showed positive correlation with CEC, organic matter, Fe and Mn oxides, silt and clay. The above correlations suggest that organic matter, silt and clay were the major metal sorbents of the sediments, whereas calcium carbonate and sand acted as diluant materials of the heavy metals in the sediments. Fe and Mn oxides were also sorbents for most metals, but not for Pb, Cd and Cu. However, organic matter content was considered to be the most important controlling factor in the abundance of Hg in the sediments (r = 0.678), while clay was the most important sediment component responsible for the abundance of the other metals. This is because Hg exhibits stronger affinity for organic matter in soils and sediments (Schuster, 1991) than do soil inorganic components (Yin *et al*., 1996). Copper showed no correlation with other metals and sediment characteristics. This suggests that there may be significant anthropogenic sources of Cu to the coastal area.

In addition, the organic rich sediments showed the high CEC values (r = 0.719) and also showed good correlations with clay (r = 0.515) and silt (r = 0.671). This is because the CEC of soils or sediments is attributable to organic matter and clay fractions, and organic matter can also occur with clay minerals (Sparks 1995). Therefore, the sediments containing high organic matter, clay and silt had the potential to retain heavy metals and high CEC of the sediments could result in high metal concentrations.

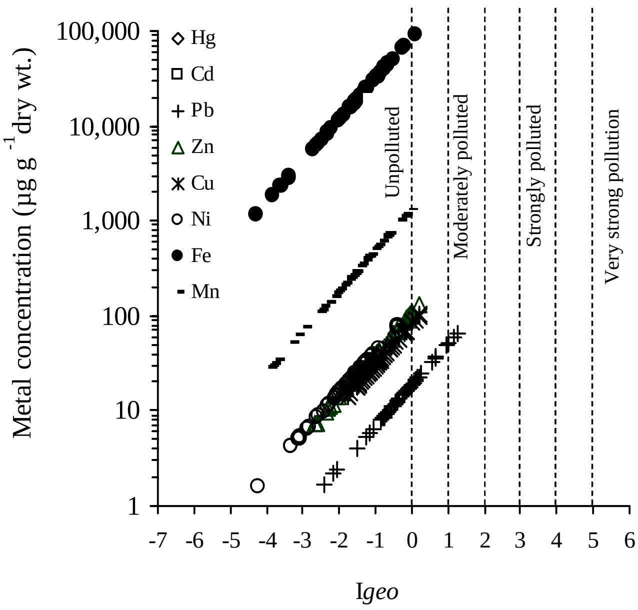

Figure 3. Plots of heavy metal concentrations and I_{geo} index.

3.3. Assessment of Heavy Metal Pollution

The geoaccumulation index (I_{geo}) was used to define the degree of anthropogenic pollution in the sediments. The index is the enrichment on geological substrates and can be calculated using the following equation (Förstner *et al.*, 1993):

$$I_{geo} = \log_2 C_n/1.5 \times B_n$$

where C_n is the measured concentration of the examined metal in sediments; B_n is the geochemical background concentration of the metal and 1.5 is the correction factor for variation in background values due to lithogenic effects.

The I_{geo} consists of seven grades, whereby the

Table 5. Geoaccumulation index classification (Förstner *et al.*, 1993)

Sediment Accumulation Index (I_{geo})	I_{geo} Class	Pollution Intensity
>5	6	Very strong pollution
>4-5	5	Strong to very strong
>3-4	4	Strongly polluted
>2-3	3	Moderately to strongly
>1-2	2	Moderately polluted
>0-1	1	Unpolluted to moderate
<0	0	Practically unpolluted

highest grade (6) reflects 100-fold enrichment above background values. Förstner *et al.* (1993) listed geoaccumulation classes and the corresponding pollution intensity for different indices as presented in Table 5. The index has been applied by many researchers (e.g., Singh and Hasnain, 1999; Bakan and Balkas, 1999; Zhang *et al*., 2007) to distinguish heavy metal levels in anthropogenically enriched sediments from preindustrial or natural background levels in equivalent sediments.

However, this index depends on the choice of an appropriate natural background value. Since there are no metal background values for this study area, the crustal average values (Taylor 1964) were used to calculate this index. Results of I_{geo} values of the sediment heavy metals are given in Table 2 and Figure 3. According to the I_{geo} classification, the study area may be considered moderately polluted with Pb (I_{geo} > 1) in some locations with the highest contamination observed at Map Ta Phut Industrial Estate. This may be due to the input of various industries around and automobile aerosol lead. In addition, a lower degree of pollution was found in the sediments by Cu (at Prasae estuary and Kung Kraben Bay), Zn (at Chon Buri Bay), and Mn (at Bangpakong estuary). There was no pollution from Hg, Cd and Ni in the sediments and Fe could be neglected due to very low I_{geo} value obtained (I_{geo} = <0-0.09).

In conclusion, the examined heavy metals in sediments of the Eastern Coast of the Gulf of Thailand showed wide ranges of concentrations due to spatial variations of metal distribution. The differences could be attributed to the sediment characteristics and land-based point and non-point inputs, especially from industrial activities. The two approaches in evaluating heavy metal pollution, comparing with Thailand's sediment quality guidelines and use of the geoaccumulation index gave quite similar information that the sediments were polluted with Pb and Cu, but no categorization of the degree of pollution based on the former approach. In addition, the present study showed that anthropogenic activities for heavy metals contribution in the area were marked at minimal.

Acknowledgements

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Correspondence to
Dr. Waewtaa Thongra-ar
Institute of Marine Science,
Burapha University,Bangsaen,
Chon Buri 20131 Thailand
Email : waewtaa@buu.ac.th