

## Accumulation of Barium, Uranium, Cadmium and Manganese in the Sediment Core from the Pulau Cik Wan Dagang Mangrove Forests, Terengganu, Malaysia

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### ABSTRAK

Akumulasi menegak unsur-unsur Ba, U, Cd dan Mn diukur pada teras sedimen sedalam 150 cm yang diperolehi di hutan paya bakau, Pulau Cik Wan Dagang, Kuala Kemaman, Terengganu, Malaysia. Secara keseluruhannya, kepekatan elemen kajian kecuali Mn menurun mengikut kedalaman dan menunjukkan satu sempadan yang jelas pada kedalaman 70 cm. Bagi menentukan pengaruh pencemaran logam berat, faktor pengkayaan (EF) bagi semua unsur dikira. Dalam kajian ini, nilai EF bagi U dan Cd secara signifikan adalah lebih besar daripada nilai piawai dan dianggap mempunyai sumber yang lebih dominan kepada antropogenik manakala sumber Ba dan Mn adalah dominan secara semula jadi kerana nilai EFnya didapati hampir sama dengan nilai piawai.

### ABSTRACT

The accumulation of elements Ba, U, Cd and Mn with respect to depth was determined in a 150 cm sediment core obtained from the mangrove forests of Pulau Cik Wan Dagang, Kuala Kemaman, Terengganu, Malaysia. Generally, all elements except Mn decrease with depth and their vertical profiles show a clear boundary at about 70 cm deep. In order to assess the influence of heavy metal pollution more precisely, enrichment factors (EF) were calculated. In this study, U and Cd have EF values significantly greater than unity and may therefore be considered to be predominantly anthropogenic in origin whereas Ba and Mn which have EF values about unity are considered to be predominantly terigenous in origin.

**Keywords:** Anthropogenic, barium, cadmium, enrichment factors, manganese, uranium

### INTRODUCTION

Mangroves are woody, seed-bearing, highly specialized plants and are found along sheltered intertidal coastlines of estuaries and lagoons. Their species possess unique adaptations which enable them to thrive in an environment where other plants cannot grow. Recently, reclamation of mangrove ecosystems for industrial, urban and other forms of development has been increasing and it causes irreversible damage in coastal regions throughout the tropics. Along

with the destruction of the mangroves comes the anthropogenic effects associated with the new developments. The sediments from mangrove areas contain a historical record of information on the temporal changes that have been brought about as a result of these actions. This record is altered or smeared by the effects of sediment mixing, which influences the preservation of the physical sedimentary structures (Nittrouer and Sternberg 1979). However, the alteration of the record depends on the intensity, depth and nature of the sediment mixing as well as the sediment accumulation rate. Sediment mixing also alters the biogenic components and pore water concentrations of dissolved chemical species within the sediment mixed layer (Berner 1980; Aller 1982). In addition, Yingst and Rhoads (1980) documented that microbial activities are influenced by sediment mixing rates.

Mangrove ecosystems are important sinks for particulate material supplied by rivers and the atmosphere to the oceans (Yeats and Bowers 1983). In urbanized regions, the sediment is very often influenced by domestic, industrial and mining activities leading to increased trace metal concentrations (Martin and Whitfield 1983; Martin *et al.* 1989). When these sediments are undisturbed, and sedimentation rates are known, they can provide us with a historical pollution record (Goldberg *et al.* 1977; Bertine 1980; Nolting and Helder 1991). Biological, physical, chemical and diagenetic processes can change the sediment properties and those processes can be applied to study the anthropogenic influences on the marine environment. In Malaysia, studies relating to mangroves are not well documented and only little information is available concerning the sedimentation of the mangrove forest (Mohd Lokman *et al.* 1994; Kamaruzzaman 1994). Furthermore, geochemical studies of sediments from the Malaysian mangrove forests have received little attention and only limited studies have been carried out regarding their role in the process of sedimentation (Kamaruzzaman *et al.* 2000). In view of the importance of the mangrove to various aspects of the environment, research on the concentration of heavy metals as well as their distribution pattern in sediment was carried out.

## MATERIALS AND METHOD

### *Sampling Sites*

Pulau Cik Wan Dagang is situated in the district of Kemaman, approximately 170 km south of Kuala Terengganu, Malaysia. The mangroves which are relatively undisturbed have been gazetted as a mangrove reserve forest by the Terengganu Forestry Department. The mangrove area lies on the southern bank of the Kemaman estuary, where both the Kemaman and Chukai rivers flow into. The tide floods the area twice daily as it is semi-diurnal with a mean range of 1.8 m. In this study, a 150 cm sediment core was collected with a D-section core sampler from this area (*Fig. 1*) and was cut into segments of approximately 5 cm intervals, labelled and stored in cleaned acid bottles for analysis. The sediment core was chosen for this study since the area is known to be highly productive (Kamaruzzaman 1994) and with a moderately high

sedimentation rate of  $0.66 \text{ cm yr}^{-1}$  (Kamaruzzaman *et al.* 2000), which is suitable for this kind of study.

#### Analytical Methods

The sediment samples were digested according to the published methods (Noriki *et al.* 1980; Sen Gupta and Bertrand 1995; Kamaruzzaman 1999) with some modifications. An inductively-coupled plasma mass spectrometer (ICP-MS) was used for the quick and precise determinations of Al, Ba, U, Cd and Mn

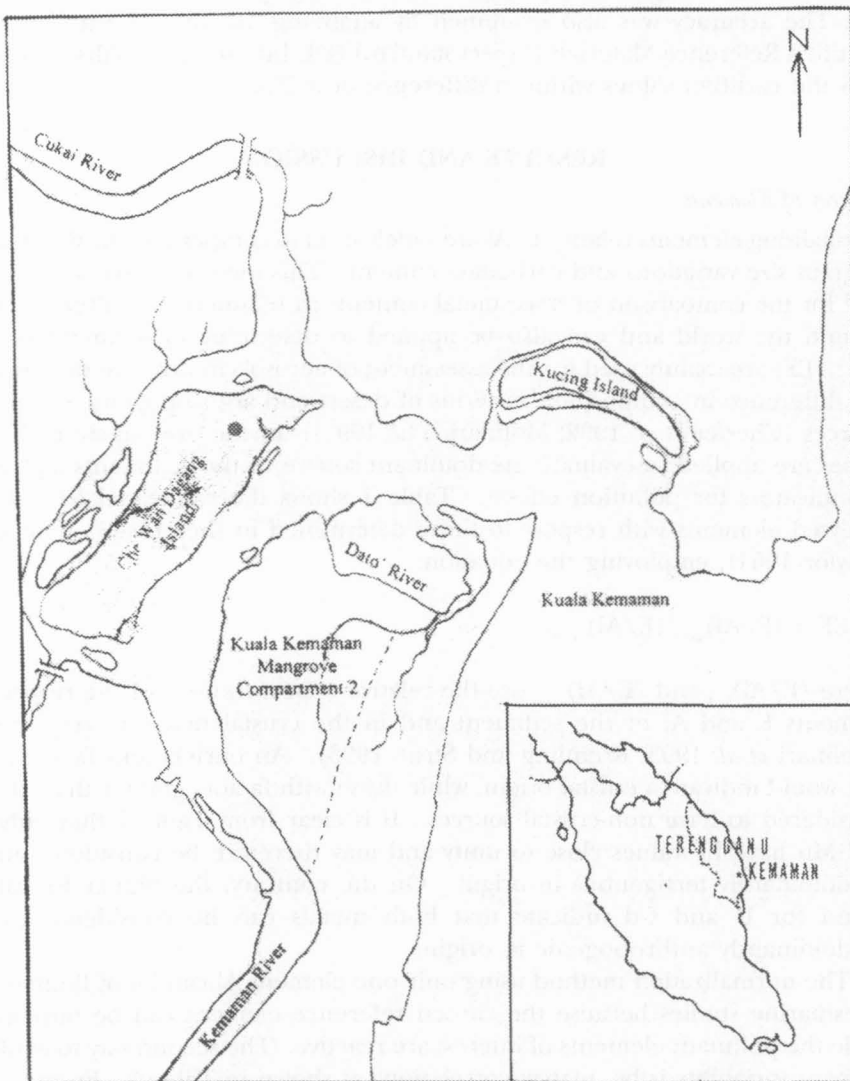


Fig. 1: Location of the core (-) study area in the Pulau Che Wan Dagang, Mangrove Forests, Kemaman, Terengganu, Malaysia

in the digested marine sediment. Briefly, the digestion method involved the heating of 50 mg of a < 63  $\mu\text{m}$  size sample in a sealed teflon vessel with a concentrated acid mixture of HF,  $\text{HNO}_3$  and HCl in the ratio of 2.5 : 3.5 : 3.5. The teflon vessels were kept at 150  $^\circ\text{C}$  for 3 – 5 hours. After cooling, a mixed solution of boric acid and EDTA was added, and the vessel was again heated at 150  $^\circ\text{C}$  for at least 5 hours. After cooling to room temperature, the content of the vessel was transferred into a 10 ml polypropylene test tube and was diluted to 10 ml with deionized water. A clear solution with no residue had to be obtained at this stage. The precision assessed by replicate analyses was within 3%. The accuracy was also examined by analyzing, in duplicate a Canadian Certified Reference Materials Project standard (DL-1a) and the results coincided with the certified values within a difference of  $\pm 3\%$ .

## RESULTS AND DISCUSSION

### *Sources of Elements*

Normalising elements relative to Al are widely used to compensate for differences in grain size variations and carbonate content. This method is also a powerful tool for the comparison of trace metal contents in sediments in different areas around the world and can also be applied to determine enrichment factors (EF). EFs are mainly used for the assessment of aerosols in order to understand the difference in composition in terms of desert and anthropogenic elemental sources (Chester *et al.* 1992; Molinari *et al.* 1993). In the present study the EF values are applied to evaluate the dominant source of the sediments and used as indicators for pollution effects. Table 1 shows the calculated EFs of the analysed elements with respect to those determined in the crustal abundance (Taylor 1964), employing the equation:

$$\text{EF} = (\text{E}/\text{Al})_{\text{sed}} / (\text{E}/\text{Al})_{\text{crust}}$$

where  $(\text{E}/\text{Al})_{\text{sed}}$  and  $(\text{E}/\text{Al})_{\text{crust}}$  are the relative concentrations of the respective elements E and Al in the sediment and in the crustal material, respectively (Molinari *et al.* 1993; Kremling and Strue 1993). An enrichment factor close to 1 would indicate a crustal origin, while those with factors greater than 10 are considered to have non-crustal sources. It is clear from Table 1 that only Ba and Mn have EF values close to unity and may therefore be considered to be predominantly terrigenous in origin. On the contrary, the higher EF values found for U and Cd indicate that both metals can be considered to be predominantly anthropogenic in origin.

The normalization method using only one element Al can be of limited use in estuarine studies because the chosen reference element can be unreactive while the pollutant elements of interest are reactive. The second way to explore element variability is by matrix correlations as shown in Table 2. From Table 2, only Ba, U and Mn showed a fairly moderate correlations coefficient ( $r > 0.5$ ) with Al, indicating the co-occurrence with the lithigenous fraction of the

TABLE 1  
Calculated enrichment factors (EF) of Ba, U, Cd and Mn based on the mean concentrations determined in the whole sub-cores

Depth (cm)	Ba	U	Cd	Mn
5	3.63	24.17	13.14	0.66
10	2.88	14.37	6.09	0.38
15	3.55	18.44	7.67	0.46
20	1.67	20.09	10.28	0.49
25	4.54	25.13	21.48	0.67
30	1.56	18.59	14.06	0.46
35	3.63	17.89	12.49	0.47
40	1.74	24.87	17.21	0.48
45	3.64	18.49	13.37	0.47
50	1.80	19.95	15.27	0.57
55	1.12	20.97	15.49	0.59
60	1.34	18.45	11.35	0.58
65	1.53	15.68	11.24	0.50
70	1.64	18.97	11.37	0.46
75	1.86	23.12	20.27	0.49
80	2.81	21.88	12.78	0.47
85	1.55	20.50	12.87	0.42
90	1.67	19.11	12.00	0.40
95	1.46	17.44	10.97	0.39
100	1.66	18.52	11.82	0.41
105	1.49	18.24	10.98	0.42
110	1.53	18.75	12.32	0.39
115	1.62	19.32	11.96	0.40
120	1.58	19.01	12.98	0.43
125	1.40	17.70	11.41	0.34
130	1.90	24.92	12.97	0.40
135	1.72	19.49	11.07	0.38
140	1.69	15.69	10.74	0.38

TABLE 2  
The correlation coefficient ( $r^2$ ) among 5 analysed elements in the mangrove forests of Pulau Sekeping, Kemaman, Terengganu

Elements analysed	Al	Ba	U	Cd	Mn
Al	1.0000				
Ba	0.5974	1.0000			
U	0.4992	0.0081	1.0000		
Cd	0.0633	0.1920	0.3018	1.0000	
Mn	0.5792	0.2612	0.4756	0.5037	1.0000

sediment. A fairly moderate correlations, ( $r > 0.5$ ) with Mn is showed by U and Cd which indicates their occurrence in fine minerals that make up the bulk of the heavy mineral fraction of the sediment.

#### *Depth Distribution*

Depth profiles of Ba, U, Cd and Mn are shown in *Fig. 2*. Based on the previous studies (Mohd Lokman *et al.* 1994; Kamaruzzaman *et al.* 2000), sediments from the mangrove forests of Kemaman seem to be well mixed, and the sediment supply homogeneous. Although some of these profiles show an increase in concentration toward the surface layer, this is not necessarily an indication for anthropogenic input. It is more likely that early diagenetic processes are responsible for this phenomenon (Ridgway and Price 1987; Macdonald *et al.* 1991). It was noted that the vertical profiles of Ba, U, Cd and Mn show a distinct contrast in the upper and lower parts of the sediments as shown in *Fig. 2*. The concentrations of all elements except for Mn were comparably high and varied widely whereas at greater depth, their vertical profiles showed only slight variation. The mean concentration and relative standard deviations of all four elements for each 5 cm subcore were calculated (Table 3). From the table, we can easily see the existence of a boundary at about 70 cm depth. For the first layer above 70 cm, the average percentage of relative standard deviations for all elements are higher (0.22%) and for greater depths they are relatively smaller (0.05%). Assuming the sedimentation rate obtained from a previous study (Kamaruzzaman *et al.* 2000) can be applied to the boundary, its age turned out to be 91 years old. The average of percentage relative standard deviations of Ba, U, Cd and Mn in the upper layer are 10% or more higher than those in the deeper layer, with Ba showing the greatest contrast of about 25%.

The concentration of Ba ranged from 231 ppm to 760 ppm with an average of 413 ppm. The vertical profile initially decreased steeply with depth (*Fig. 2*). The sharp decrease of Ba in the surface layer may be due to the early diagenetic loss of Ba, being a common phenomenon in the sediments. In other studies at a Pulau Cik Wan Dagang mangrove forest (Leong 2001), Ba has a fairly good correlation with organic matter, suggesting that the sedimentation of Ba is controlled largely by the biogenic matter, although the detrital fraction was dominant in the sediments. Therefore, the low Ba present in the sediments might be due to the low productivity during that period and/or probably due to the dilution of the biogenic matter with very high amounts of terrigenous material transported to the sea.

Uranium was generally constant ranging from 11.5 ppm to 20.1 ppm and averaged at 15.7 ppm (*Fig. 2*). Its mean value was 5 times higher than its average U shale value and calculated EF values suggesting that most of the U might have come from the anthropogenic sources. The slight decrease of U in the surface layer may be due to the early diagenetic loss of U in oxic sediment (Thomson *et al.* 1990), where the bottom water U has diffused downwards into the sediment a few cm deep. The vertical variations in the depth profile might be due probably to temporal changes in biological productivity. However,

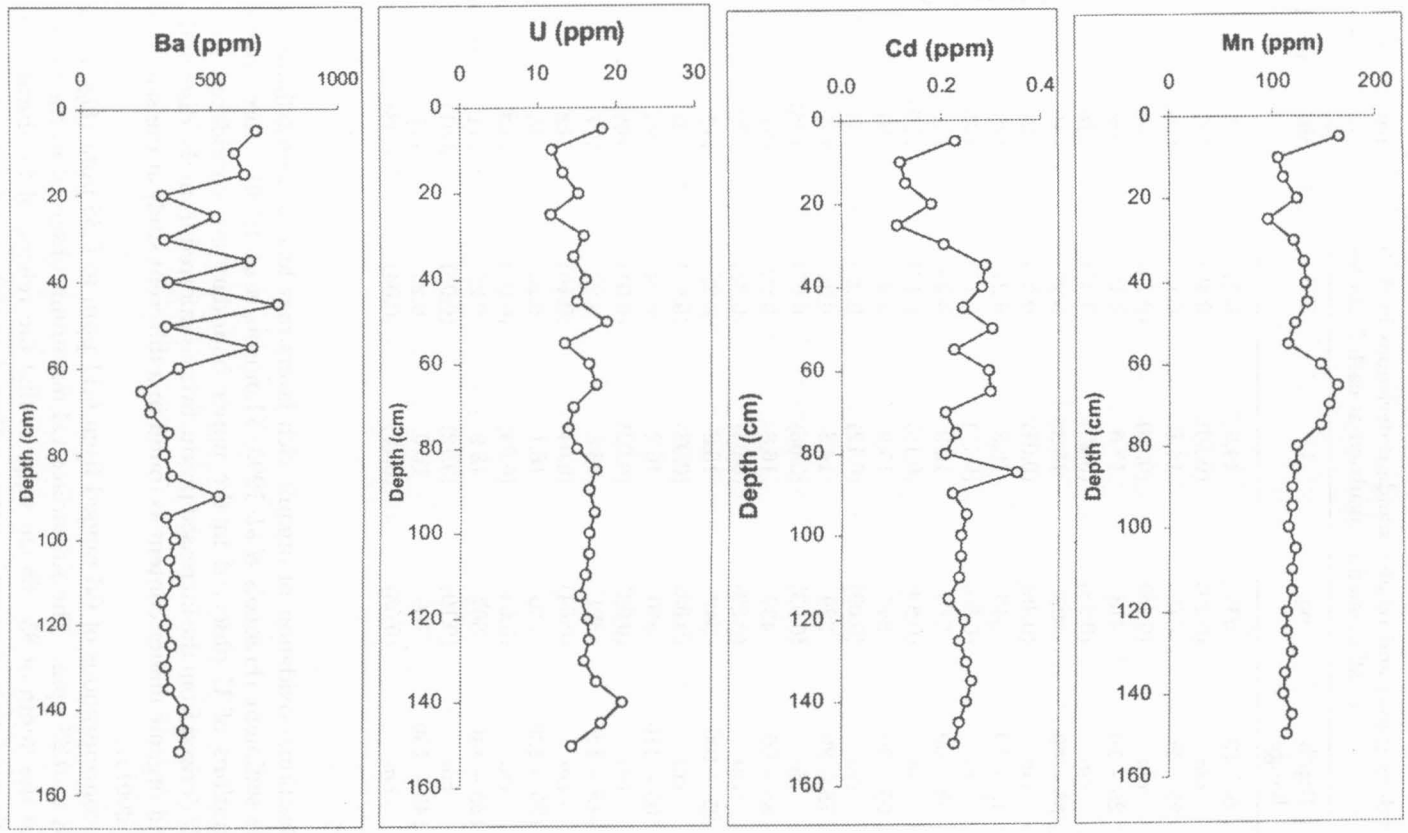


Fig. 2: Vertical profiles of Ba, U, Cd and Mn plotted against depth. The division in the graph is described in the text.



TABLE 3  
Mean values (ppm) and relative standard deviations in % (shown in parentheses)  
of all elements calculated at each 5 cm interval

Depth Range	Ba	U	Cd	Mn
5 – 10 cm	638 (0.10)	14.9 (0.30)	0.17 (0.46)	135 (0.31)
15 – 20 cm	473 (0.48)	14.1 (0.10)	0.15 (0.24)	116 (0.08)
25 – 30 cm	418 (0.33)	13.6 (0.22)	0.16 (0.43)	108 (0.16)
35 – 40 cm	495 (0.46)	15.3 (0.07)	0.29 (0.01)	131 (0.01)
45 – 50 cm	543 (0.56)	16.8 (0.16)	0.27 (0.15)	128 (0.06)
55 – 60 cm	516 (0.39)	14.9 (0.15)	0.26 (0.19)	131 (0.17)
65 – 70 cm	247 (0.09)	15.9 (0.12)	0.25 (0.25)	159 (0.04)
75 – 80 cm	329 (0.03)	14.3 (0.06)	0.22 (0.07)	135 (0.13)
85 – 90 cm	437 (0.29)	16.8 (0.03)	0.29 (0.32)	121 (0.02)
95 – 100 cm	342 (0.08)	16.8 (0.03)	0.25 (0.03)	117 (0.02)
105 – 110 cm	350 (0.03)	16.2 (0.02)	0.24 (0.01)	120 (0.02)
115 – 120 cm	321 (0.04)	15.8 (0.04)	0.23 (0.10)	117 (0.03)
125 – 130 cm	336 (0.04)	16.1 (0.04)	0.24 (0.03)	116 (0.03)
135 – 140 cm	367 (0.10)	18.9 (0.12)	0.25 (0.03)	111 (0.01)
145 – 150 cm	389 (0.03)	16.0 (0.17)	0.23 (0.04)	117 (0.03)

postdepositional oxidation of organic rich layers may lead to redistribution of U in the sediment (Francois *et al.* 1993; Thomson *et al.* 1990). Some higher concentrations of U observed in the upper boundary are considered to be primarily derived from the increased productivity as inferred from the distribution of Ba and organic matter content as found in a different study at the same area (Leong 2001).

The concentrations of Cd ranged from 0.11 ppm to 0.35 ppm (*Fig. 2*) and averaged to 0.23 ppm. The subsurface Cd maximum observed in the vertical profile at the depth of 35 - 65 cm suggests that the release of Cd during early diagenetic oxidation of organic matter rather than diffusion from water at the



bottom is the primary source of dissolved Cd into pore water of suboxic sediments (Gobeil *et al.* 1987; McCorkle and Klinkhammer 1991). Since Cd removal in anoxic, sulfide bearing sediments is related to its strong tendency to form insoluble sulfides in the presence of H<sub>2</sub>S (Elderfield 1981), it is suggested that Cd enrichments in the suboxic sediments in the absence of H<sub>2</sub>S, particularly at the depths of 65 cm and 85 cm, are likely due to CdS precipitation in the presence of trace levels of free sulfide. These small amounts of free sulfide perhaps diffuse upward from underlying anoxic sediments and are sufficient to cause precipitation of CdS.

The oxidizing surface layer containing Mn of about 250 ppm and the existence of the subsurface maxima of Mn are caused by the migration of Mn<sup>2+</sup> with interstitial water and subsequent precipitation as a kind of carbonate on the surface of volcanic pumice (Tsunogai *et al.* 1979). A subsurface maximum in the Mn content within the enriched layer has also been observed in continental shelf sediments (Hartman *et al.* 1979) and is common in pelagic sediments (Wangersky 1962). Since the source of Mn<sup>2+</sup> is not limited at a certain depth, both the dissolution (reduction) of Mn oxide and precipitation as carbonate may occur simultaneously as observed at depths of 20, 50, 60 and 70 cm in this study (Fig. 2). As the sedimentation proceeds, the redox condition of the buried sediment changes to a reducing state and the alkalinity in the interstitial water begins to increase (Hartman *et al.* 1979). The interstitial Mn in the lower part migrates downward through interstitial and forms manganese carbonate precipitate when the carbonate level in the water is sufficiently high. Although it is known that Mn migrates into the reducing environment, depositional migration for other components including Ba and U have not been established. Therefore one should be cautious when interpreting the concentration profiles of metals in a sediment core.

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