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Distributions of aluminium, cobalt, iron and manganese in the surface sediments of east coast of Peninsular Malaysia

(Taburan aluminium, kobalt, ferum dan mangan dalam sedimen permukaan pantai timur Semenanjung Malaysia)

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ABSTRACT

Surface sediments samples from 30 stations at the east coast of Peninsular Malaysia were collected using the Smith McIntyre grab on June 2008 in order to determine the contents of selected trace metals using the Inductive Coupled Plasma- Mass Spectrometry (ICP-MS). The concentrations of Al, Fe, Co and Mn ranged from 0.96 %-5.74 %, 6.20-8.95 %, 83-155 µg g^{-1} and 404-1350 µg g^{-1} respectively. The variation was then studied by ANOVA, which showed a significant difference for the concentrations of all elements at all sampling stations. The correlation between elements was significant ($p>0.05$) at all stations. Whereas, the geoaccumulation index proved that the surface sediment taken at all stations were unpolluted with Al and Mn (Class 1), moderately polluted with Fe (Class 2) and strongly polluted with Co (Class 4).

Keywords: surface sediments, lithogenic, I_{geo}

ABSTRAK

Sampel sedimen permukaan daripada 30 stesen di pantai timur Semenanjung Malaysia telah dikumpulkan menggunakan cekau Smith McIntyre pada Jun 2008 untuk mengkaji kandungan logam surih terpilih menggunakan Plasma Berpasangan Teraruh-Spektrometri Jisim (ICP-MS). Kepekatan Al, Fe, Co dan Mn masing-masing adalah antara dari 0.96 %-5.74 %, 6.20-8.95 %, 83-155 µg g^{-1} dan 404-1350 µg g^{-1}. Perbezaan itu kemudiannya diuji menggunakan ANOVA, yang menunjukkan perbezaan signifikan bagi kepekatan semua elemen di semua stesen persampelan. Korelasi antara elemen juga signifikan ($p>0.05$) di semua stesen. Manakala, Indeks Geoakumulasi membuktikan bahawa sedimen permukaan yang diambil di semua stesen adalah tidak tercemar dengan Al dan Mn (Kelas 1), sederhana tercemar dengan Fe (Kelas 2) dan kuat tercemar dengan Co (Kelas 4).

Kata kunci: sedimen permukaan, litogenik, I_{geo}

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INTRODUCTION

Marine sediments hold up preserved and age-controlled marine and terrestrial environmental records (Kawahata, 2001); hence contain the history of ancient incidents and processes. These sediments also contain varieties of minerals and heavy metals. Trace metals are known to be contaminants when human activity raises their concentrations in the environment above natural levels (Ming 2001). The high storage capacity for pollutants is one of the characteristics of the sediments. In other words, that in any part of the hydrological cycle for less than 1% of these are actually dissolved in the water and more than 99% are stored in the sediments, which then can produce a remarkable enrichment (Salomons & Stigliani 1995; Naji & Ahmad 2011).

Most of heavy metals are known to be highly toxic. This is because, as ions or in certain compounds, they are very soluble in water and may be absorbed into plants or animal tissues. If the absorption happens, the metals will tend to combine with biomolecules such as proteins and nucleic acids, yet impairing their functions. Heavy metals are also claimed to be pollutant. The word pollutant refers to ‘substances that occur in the environment at least in part of results of man’s activities, and which have deleterious effects on living organisms’, (Gilbert 2004). Pollution in marine area has been a worldwide problem especially when it comes to heavy metals. This is because heavy metals are known to be indestructible and may have some toxic effects on living organisms once they exceed a certain concentration (MacFarlene et al. 2000; Dalman et al. 2006; Chen et al. 2007, Ong & Kamaruzzaman 2009). The introduction of trace metals into the aquatic environment can reach an extend where they can accumulate in sediments through disposal of liquid effluents, chemical leachates and runoff originating from domestic, industrial and agricultural activities as well as atmospheric deposition (Mucha et al. 2003, Naji & Ahmad 2011).

Adjacent estuaries and rivers are known to be one of the sources of trace metal contamination in a marine coastal environment (Ong & Kamaruzzaman 2009). According to Hungspreugs (1988), (Takarina et al. (2004), heavy metals pollution of riverine and coastal environment has been reported throughout the region of Southeast Asia. Industrial development has increased the release of heavy metals to the marine environment. This is an area where the monitoring and enforcement of environmental regulations is difficult due to a lack of laboratory facilities and enforcement mechanism (World Bank, 1994). Environmental concentration is used by comparison with normal or average background levels to provide evidence for contamination (Ming 2001). Usually, trace metals come from the weathering of Earth’s crust, soil erosion, industrial discharge, mining, urban runoff, sewage effluents, air pollution fallout, waste incinerations and other sources. The inorganic chemical manufacturing industries are potential sources for metallic wastes to aquatic ecosystems, so they are regulated minimize inputs (Manahan 2000). This paper aims to measure the concentrations and pollution status of Al, Co, Fe and Mn in surface sediments and to trace the lithogenic transport in the east coast Peninsular Malaysia.

MATERIALS AND METHODS

The sampling was conducted in the Eastern Coast of Peninsular Malaysia, totaling in thirty stations in South China Sea, on a scientific cruise using the Department of Fisheries research vessel, KL Paus (Figure 1; Table 1). The cruise started on 11th June 2008 until 26th June 2008. The water depth ranged from 13.0 to 72.0 meters. Global Positioning System was used to record the exact location of the sampling site. The surface sediment was collected by using Smith McIntyre grab. To avoid any contamination from the grab's wall, the outer part of the sediment sample was removed and only the inner part was further processed. Then, the collected sediment was put in the acid washed plastic bags which were brought back and kept frozen in 4°C until further analysis.

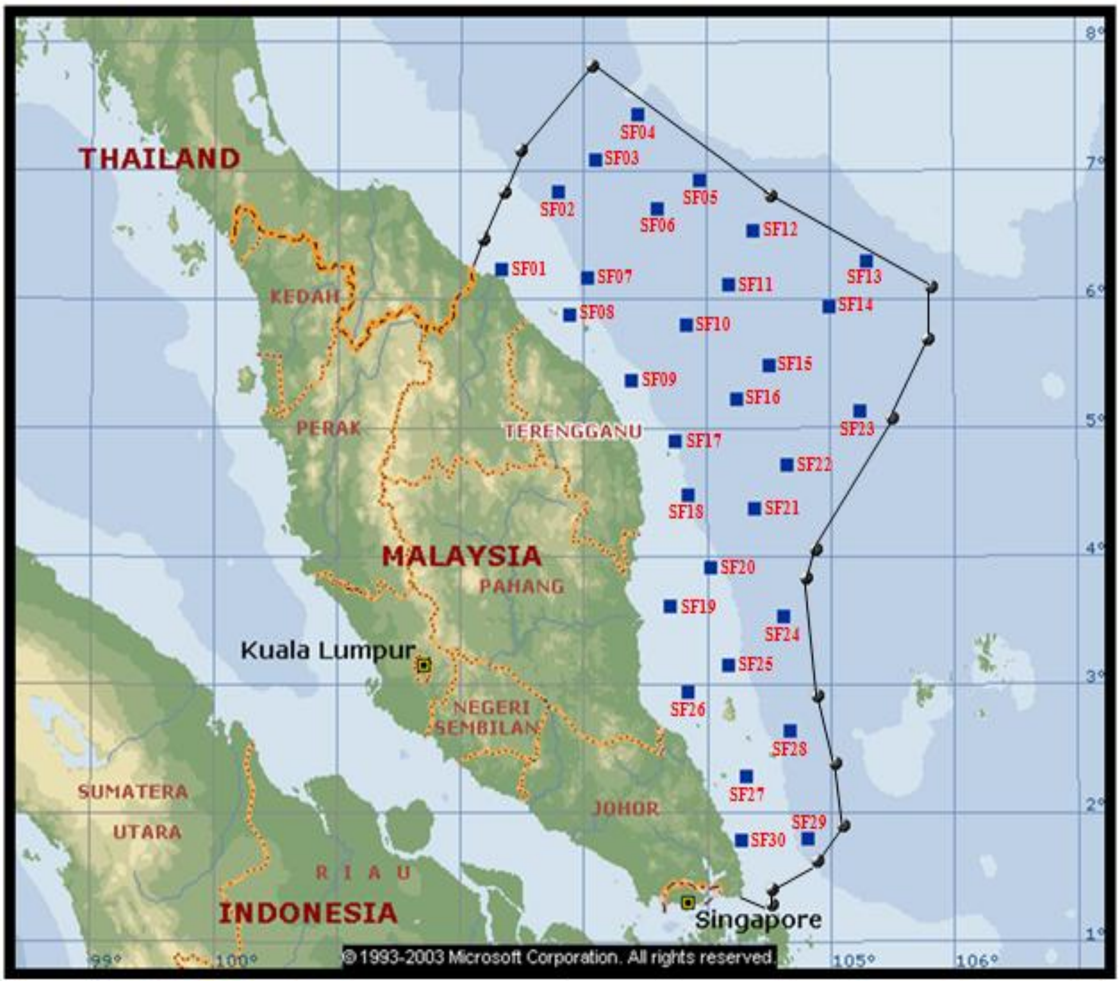

Figure 1. Sampling stations (box in blue)

Table 1: Locations of the sampling stations

Station	Latitude	Longitude	Distance from shoreline (n.m)	Water depth (m)
SF01	06 13.99'	102 19.00'	2.7	13
SF02	06 50.04'	102 47.04'	50	46
SF03	07 05.03'	103 04.99'	73	50
SF04	07 25.98'	103 26.01'	100	61
SF05	06 56.09'	103 56.04'	108	52
SF06	06 42.14'	103 35.17'	80	52
SF07	06 10.00'	103 01.00'	40	45
SF08	05 52.10'	102 51.92'	15	34
SF09	05 22.06'	103 21.97'	14	47
SF10	05 48.20'	103 48.98'	48	55
SF11	06 06.16'	104 09.11'	75	72
SF12	06 32.01'	104 22.11'	101	59
SF13	06 16.98'	105 16.99'	139	55
SF14	05 57.15'	104 58.13'	115	56
SF15	05 29.08'	104 29.02'	80	60
SF16	05 18.50'	104 12.60'	56	60
SF17	04 54.12'	103 42.98'	17	54
SF18	04 28.14'	103 49.98'	20	40
SF19	03 37.07'	103 41.08'	15	23
SF20	03 55.10'	104 00.05'	40	50
SF21	04 22.16'	104 22.07'	52	65
SF22	04 44.19'	104 38.44'	70	66
SF23	05 08.10'	105 12.9'	109	67
SF24	03 32.08'	104 36.00'	70	62
SF25	03 09.14'	104 09.04'	42	41
SF26	02 56.13'	103 49.97'	24	20
SF27	02 16.94'	104 16.97'	19.5	30
SF28	02 39.18'	104 38.91'	47	58
SF29	02 00.55'	104 41.97'	35	46
SF30	01 48.04'	104 15.03'	4.5	14

The sediment sample was dried and grinded. After that, the sample was sieved by using a 63 µm sieve. All the samples were then digested by using the Teflon bomb involving 50 mg of sample with a mixed acid solution (1.5 mL) of concentrated HF, HNO_3 and HCl. The Teflon vessel was kept at 150 °C for 5 hours. After cooling, a mixed solution of boric acid and EDTA (3 mL) was added. The Teflon bomb was heated again for another 5 hours. The digested sampled was then transferred into the test tube and added up with milli-Q water up till 10 mL. Lastly, the samples were detected by using an inductively coupled plasma mass spectrometer (ICP-MS). Quality control test was done by using the Standard Reference Material (SRM 1646a-Estuarine Sediment) published by National Institute of Standard & Technology (NIST) and reached 115 % accuracy for Al, 106 % for Co, 115 % for Fe and 104 % for Mn.

Geoaccumulation index measurement, I_{geo}

The geoaccumulation index was measured by using the Müller formula (1979):

$$I_{geo} = log_2\left(\frac{C_n}{1.5B_n}\right)$$

Where B_n reflects the average shale (Bowen, 1979), Cn reflects the measured elements concentration in the sediment. A constant, 1.5, is used to allow natural fluctuations in the content of a given substances in the environment, with low anthropogenic interruptions (Ong and Kamaruzzaman, 2009).This formula is used along with its classification as in Table 2.

Table 2: Müller Classification to quantify the geoaccumulation index

Value of I_{geo}	I_{geo}	Pollution intensity
< 0	0	unpolluted
0 ~ 1	1	unpolluted to moderately polluted
1 ~ 2	2	moderately polluted
2 ~ 3	3	moderately to strongly polluted
4 ~ 5	4	strongly polluted
5 ~ 6	5	strongly to very strongly polluted
> 5	6	very strongly polluted

RESULTS AND DISCUSSION

Distributions of Selected Metals and Organic Carbon

The concentrations of selected metals in study area are shown in Table 3. Al has a mean concentration of 4.34 ± 1.21 % dry weight with range from 0.96-5.74% dry weight, Fe has a mean of 7.56 ± 0.49 % dry weight with range from 6.20-8.95 % µg g-1 dry weight, Mn has a mean of 664.72 ± 169.13 µg g^{-1} dry weight, ranged from 404-1350 µg g^{-1} dry weight and Co has a mean of 102.27 ± 11.96 µg g^{-1} dry weight, with the range of 83- 155 µg g^{-1} dry weight.

The concentrations of selected metals were not uniform across all stations (Figure 2). However, SF29 showed similar pattern of Co, Fe and Mn distributions. This phenomenon assumed to be caused by the anthropogenic sources from the environment such as eutrophication and land based activities. According to Karbassi et al. (2005), several other studies also showed high concentrations of trace elements in coastal areas caused by input from the environment. Al has a weak correlation with organic carbon (r=0.180, p=0.000), followed by Mn (r=0.077, p=0.000) and Co (r=0.062, p=0.000) while Fe has an intermediate correlation with organic carbon (r=0.433, p=0.000) (Table 4) and this show that iron is associated with organic carbon. Rubio et al. (2000) also stated that there is correlation between organic carbon and iron. In contrast, the correlation of Al, Fe, Co and Mn were found good except for the correlation between Co and Al. This indicates that there is a strong relationship between the selected elements. Plus, this shows that these selected metals come from the same source.

Table 3: Concentrations of selected metals in surface sediments

Station (SF)	Al (%)	Fe (%)	Co ($\mu g\ g^{-1}$)	Mn ($\mu g\ g^{-1}$)
01	5.56	7.84	101.16	651.19
02	5.48	7.23	100.94	645.66
03	5.01	7.36	99.92	730.11
04	7.11	7.86	108.82	863.09
05	4.72	7.22	101.99	660.67
06	5.10	7.74	107.51	807.36
07	5.40	7.31	97.99	684.06
08	4.21	7.18	103.00	650.20
09	4.13	7.51	98.77	673.12
10	3.43	7.15	94.32	558.90
11	5.06	7.30	95.22	628.14
12	4.09	7.32	100.08	621.03
13	4.91	7.48	104.38	656.42
14	4.12	7.60	117.06	785.25
15	3.46	7.32	99.66	633.45
16	3.65	7.21	99.49	519.59
17	4.52	7.90	98.80	689.23
18	3.38	7.05	93.49	520.49
19	0.96	6.20	83.20	404.00
20	4.06	7.56	101.95	604.53
21	3.83	7.39	92.72	536.27
22	4.65	8.04	105.61	744.59
23	5.61	8.05	105.45	823.13
24	4.22	7.92	99.54	681.64
25	3.38	7.71	116.00	680.57
26	1.29	7.21	94.39	383.90
27	4.28	7.94	96.98	560.59
28	4.39	8.19	97.55	639.63
29	4.42	8.95	154.99	1349.84
30	5.74	8.10	97.02	554.82

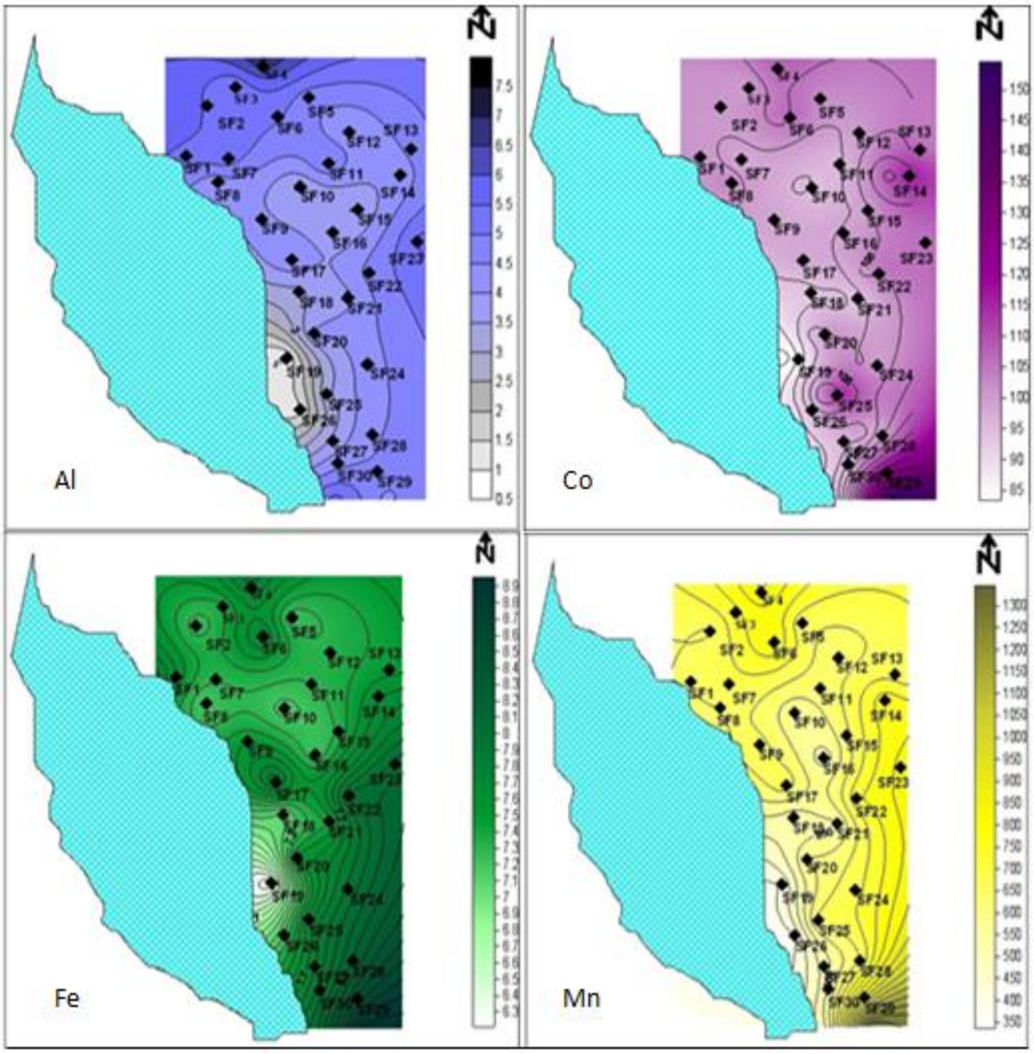

Figure 2. Concentration of selected elements in surface sediments of study area

Table 4. Correlation between element and organic carbon

Element	Al (%)	Co (µg g^{-1})	Fe (%)	Mn (µg g^{-1})
Organic Carbon (%)	0.18*	0.062*	0.433*	0.077*
Al (%)				
Co (µg g^{-1})	0.2601*			
Fe (%)	0.5175*	0.6829*		
Mn (µg g^{-1})	0.7419*	0.5411*	0.553*	

*Correlation is significant at the 0.05 level (2-tailed)

Lithogenic transport

In order to estimate aluminosilicate distribution in study area, a detailed comparison between elements is done by normalizing the concentrations of each element by a reference station (Me/Me_{28}). This reference station should have a concentration that has the intermediate concentrations among all stations. In this study Station SF28 is the reference station. To cancel out dilutions due to biogenic materials, the (Me/Me_{28}) are normalized by the relative Al concentrations (Al/Al_{28}) of each station. Aluminium is used because it has a high natural concentration and low anthropogenic concentration (Masuzawa et al. 1989, Otosaka et al. 2004). In marine environments, stable proportions of elements can be verified. This can be done by normalizing the concentrations of the elements by the Al concentration (Otosaka et al. 2004). In addition, aluminum is an element of the clay mineral structure and is insoluble element in the water column (Thomas et al. 1999). So, Al can be used as a tracer of the lithogenic transport in marine environment (Masuzawa et al. 1989, Otosaka et al. 2004). Assumed that the Al content in aluminosilicates is 8% (crustal average : McLennan 2001), more than 60% of the sediments in the sampling site is composed of aluminosilicates.

Figure 3 shows the concentrations of elements in relative to station SF28, and normalised by Al concentration. Overall, the concentrations of chemical elements were similar for Co, Fe and Mn except with regard to station SF19, SF26 and SF29. In station SF19 and SF26, Co recorded the highest content, followed by Fe and Mn. Whereas, at station SF29, Mn recorded the highest content followed by Co and Fe. The dissolved Fe, Co and Mn are oxidized in the water column, being adsorbed onto particles then settle to the bottom (Santschi et al. 1980, Moffet 1997, Otosaka et al. 2004). Relative to station SF28, depletion of Fe, Co and Mn were only seen in few stations. According to Otosaka et al. (2004), this scenario indicates reduced sediment. As a result of reduced sediment, these Mn, Fe and Co resolves from the sediment and enters the bottom water (Yeats et al. 1979, Sagar 1994, Otosaka et al. 2004. According to Chester (1990), aluminum is an element present in the main rock in the form of aluminosilicate. Aluminosilicate was present with an abundance of clay minerals and a lot of silt (sediment with fine details) (Chester 1990). According to Turekian (1976), aluminum was transported to deeper areas through bottom transport and turbidity currents. Furthermore, according to Chester (1990), high concentration of iron is probably because it is an original element is the natural base (one of the elements most common in the Earth's crust). High iron concentrations in sediment are also caused by natural erosion (Ibrahim 2004).

Next, horizontal transport of elements is also a contributing factor to the concentration of elements, especially manganese because manganese is a trace element that is easy to move through the water (Karbassi & Amirnezhad 2004, Mahmood et al. 2010). According to Barceloux (1999), cobalt can be transported as suspended sediment or in dissolved form by rivers, sea and ocean currents. It can also be precipitated in the adsorbed state with iron oxides and manganese oxides, and with crystalline sediments such as aluminosilicates and goethite. This is because they can retain cobalt. (McLaren et al. 1986, Kim et al. 2006). Overall, this transport of lithogenic material might be caused by ocean current, lateral transport, bottom transport, riverine input or atmospheric input.

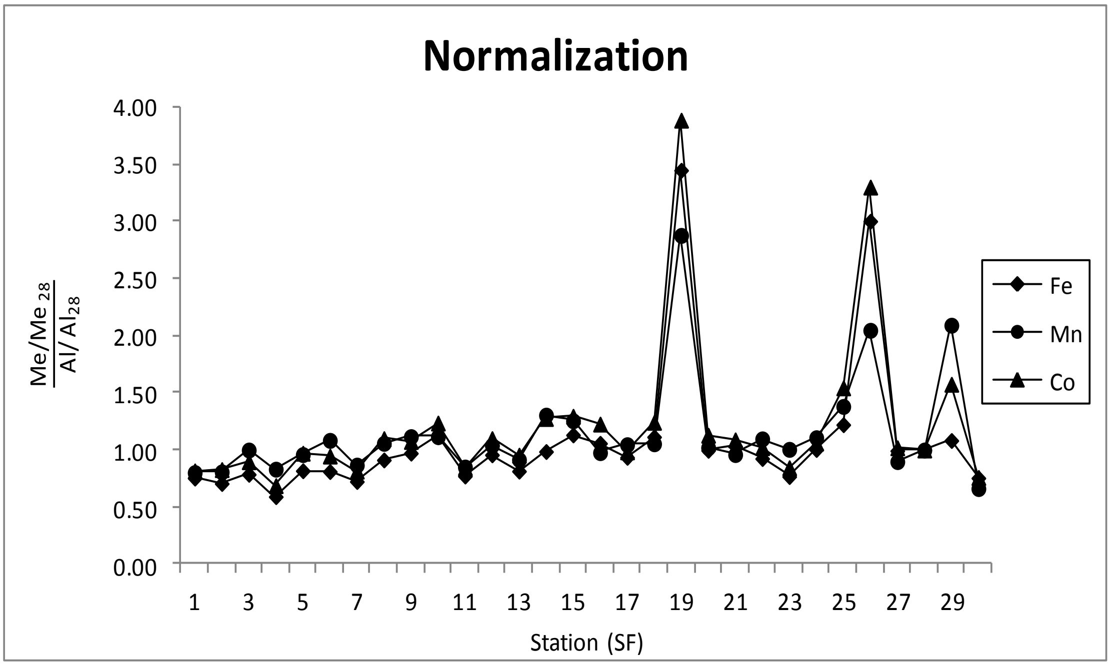

Fig.3: Concentrations of elements in relative to station SF28, and normalised by Al concentration.

Index of geoaccumulation

Marine sediments are composed of terrestrial materials transported by rivers, atmospherically driven dusts, and particles formed in situ biologically or inorganically (Chester 1990). Metal contamination impacts the coastal environment through atmospheric precipitation, fluvial income and direct effluent disposal. Fluvial income is the main carrier of continental metals to coastal zones, especially through suspended particulate materials (Forstner & Wittmann 1983). Particulate materials experience many processes, including interaction with seawater, during transport from their source areas to depositional sites (Calvert & Pedersen 1993). When the rivers reach the marine environment, the main fraction of suspended particulate material is deposited near the coast, acting as geochemical barrier to the flow of metals in the open ocean. Nevertheless, the intense biological activity and the great diversity of environmental conditions, especially in tropical regions, still result in extensive variations in metal content, promoting seasonal removals and regeneration of biological productivity cycles, depositing and removing metals in sediments, and causing chemical exchange between dissolved and particulate phases (Church et al. 1993). Pollution status is used to determine the level of pollution occurs at sampling area. Index of geoaccumulation is used to compare the status of heavy metals concentration with the background values (Ong & Kamaruzzaman 2009).

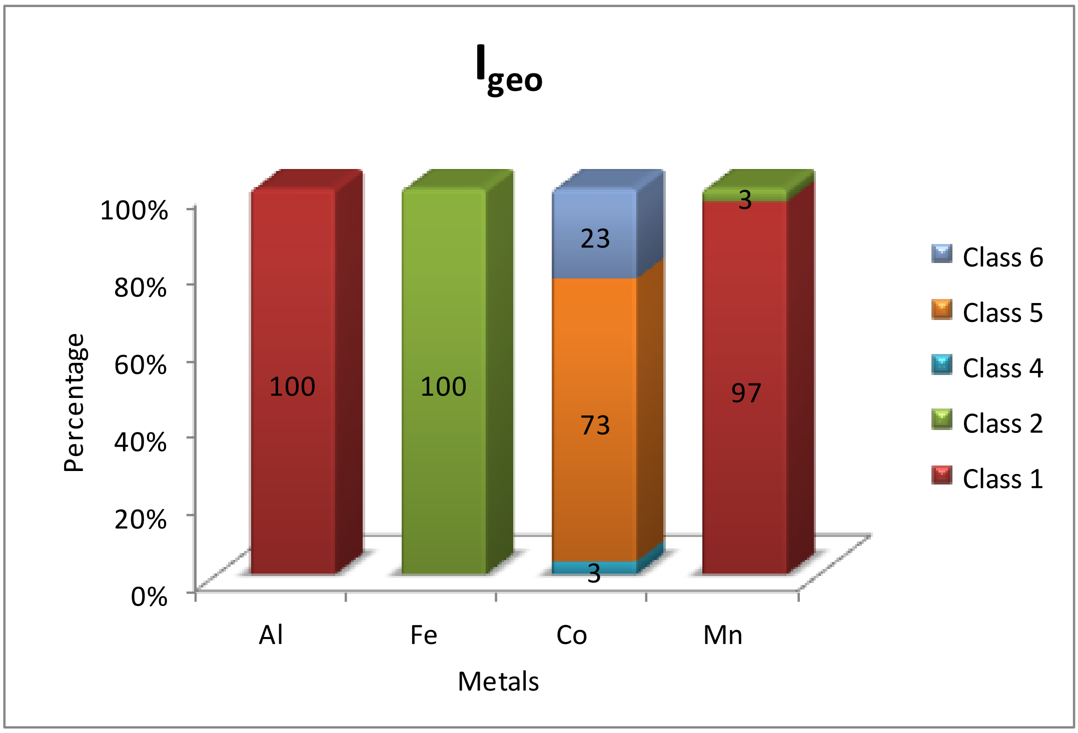

Fig.4: Geoaccumulation index for selected elements

Figure 4 shows the I_{geo} for aluminum, cobalt, iron and manganese in surface sediments of the study area. In this figure, 100% of Al was classified as Class 1 and 100% of Fe was classified as Class 2 respectively. For Co, 3% fell into Class 4, 73% fell into Class 5 and 23% fell into Class 6. While for Mn, 97% fell into Class 1 and the other 3% fell into Class 2. Therefore, it can be said that the studied area is not contaminated with Al and Mn, moderately polluted with Fe and strongly polluted with Co. Iron and manganese has biogeochemical properties that are very fast and active. Iron also has a high affinity for sediments compared to manganese because it was much transported from land to sea (Chambers et al. 2000, Mahmood et al. 2010). Aluminium is said to derive from the immediate vicinity of the naturally occur in the earth's crust (Forstner & Wittmann 1983). Cobalt is naturally occurred in ambient environment. It may enter surface water through run-off, riverine and atmospheric dust. Humans add cobalt by releasing small amounts into the atmosphere from coal combustion and mining, processing of cobalt-containing ores and the production and use of cobalt chemicals. Once it enters the environment, it cannot be destroyed. It may react with other particles or adsorb on sediments. Cobalt will only mobilize under acidic conditions, but ultimately most cobalt will end up in soils and sediments (Chester 1990).

CONCLUSION

In this study, it is found that concentrations of Al, Co, Fe and Mn vary according to the research station. The changes were tested by ANOVA showed significant differences (p = 0.000) for concentrations of all elements at each sampling station. Correlation test revealed that there were strong relationship between elements but not between element and organic carbon in the study area. Normalization of the element to aluminum shows that there is lithogenic transport from the mainland to the study area. Geoaccumulation Index shows that the surface sediment sample taken at all stations was not polluted with Al and Mn, moderately polluted with Fe and strongly polluted with Co.

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