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ASSESSMENT OF METAL CONCENTRATION IN DREDGED MARINE SEDIMENTS: CONTAMINATION INDICES

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ABSTRACT

Dredging is a necessary procedure to remove unwanted sediments from the seabed. However, the material was disposed back to the sea. Some of the material was contaminated due to industrial waste, sewage runoff and agricultural discharge. Metals is inorganic pollutant which is undegradable and can be toxic to marine ecosystem. The aim of this study is to determine the metal concentration in the material and evaluate the degree of the contamination. Four dredged marine sediments samples were retrieved from Malaysian waters. X-ray fluoresnce was used to determine the metal concentration and contamination indices were used to evaluate the degree of contaminant of the material. There were six metals detected by x-ray fluorescene; As, Cr, Cu, Pb, Ni and Zn. Based on the contamination degree, all the dredged marine sediments were found to be considerably low degree contamination.

Keywords: dredged marine sediments, dredging, metals, and contamination indices.

INTRODUCTION

Maintenance of waterways requires dredging on a regular basis to prevent flooding, facilitate navigation and allow for use of a given water system (Bert *et al.*, 2012). Dredging works also involve the periodic removal of accumulated bottom sediments from waterways (Pebbles and Thorp, 2001). According to Bortone and Palumbo (2007), the main reason for dredging is maintenance of waterways for shipping and water discharge, capital dredging and remediation of contaminated sites. The removed materials may be treated and reused or disposed under strict environmental controls (International Association Dredging Companies (IADC, 2005).

Many waterways are located in or close to industrial and urban areas. Wastes from industrial, domestic and port enter the waterways by surface runoff (Meegoda and Perera, 2001). Due to human activities during the last decades, the dredged marine sediment have contaminated with various organic and inorganic contaminants. The contaminants of most concern are metal (loids), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and mineral oils (Bert *et al.*, 2012). The type and level of contaminants concentration are different with the dredging location (Millrath, 2002).

Heavy metals are known to be serious components of inorganic pollution in aquatic sediments (Guven and Akinci, 2013). Heavy metal pollutants in the water usually transfer into sediments by physical, chemical and biological processes including ion-exchanging, precipitation, adsorption, hydrolyzation, complexation and flocculation. Heavy metals would be release from sediment to aqueous phase by environmental conditions change like salinity, acidity and redox potential. The sediment become sinks and sources of heavy metals to overlying water columns (Ai-Ping *et al.*, 2006).

In aquatic environment, some metals are knowm to be essential elements and play important roles in biological system at very low concentration (i.e.copper, zinc, iron, manganese and nickel). However, their excessive concentration can be toxic to living organisms and disturb biochemical function in both human and animals (Yielden, 2003 and Sany *et al.*, 2013). Some metals are known to be toxic even at low concentrations, including chromium, lead, cadmium, arsine and mercury (Nguyen *et al.*, 2005). Heavy metals, unlike organic pollutants, cannot be chemically degraded or biodegraded by microorganisms. Thus, their content has steadily increased in water and subsequently accumulated in sediments, plants, fishes, and even in humans (Che *et al.*, 2006).

Heavy metals pollutants after entering into aquatic environment accumulate in tissues and organs of aquatic organisms. Inputs of heavy metals into the soils and water have gradually increased over the past decades as a consequence of anthropogenic activities. Unlike energy which tend to deplete and becomes dispersed at each trophic level, heavy metals become more concentrated with each trophic level in the food chain. Ekpo *et al.*, (2013) reported that concentration of heavy metal become enhanced with progression along the trophic levels in the ecosystem rather than dissipate a process referred as bioaccumulation. The objective of this study is to determine the concentration of metals in dredged marine sediments and to evaluate the degree of contamination by using contamination indices.

MATERIALS AND METHODS

Sample collection and analysis

The disturbed dredged marine sediment (DMS) were collected from Lumut (Perak), Marina Melaka (Melaka), Pasir Gudang (Johor) and Tok Bali (Kelantan)
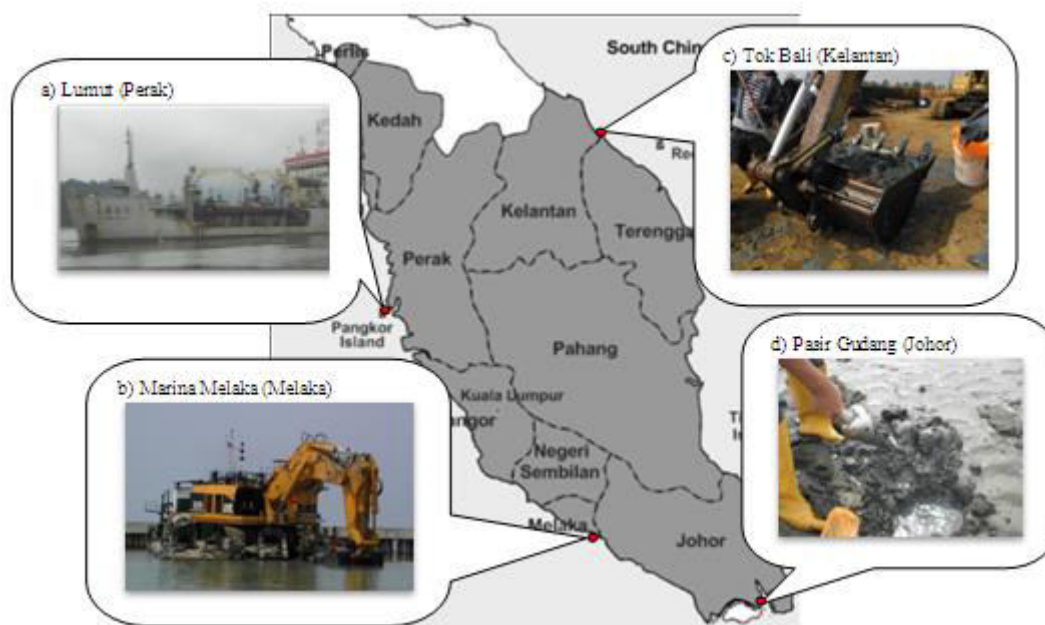
Figure-1. Sampling sites of DMS.

(Figure-1). The soil was retrieved by a trailing suction hopper dredger (TSHD) at a depth of 8-12 m from sea level for the Lumut site. The marine soils from Marina Melaka were dredged by a backhoe dredger (BHD) at a depth of 3.5-6.5 m from sea level. Backhoe dredger was also used at Tok Bali for dredging work and the depth was 3.5 - 5.0 m from sea level. Sample from Pasir Gudang site was taken from 0-10 cm of the sediment surface. Figure-2 shows dredged marine sediment sample.

Figure-2. Dredged marine sediment sample being retrieved from seabed.

The sample for this test was prepared by mixing 9 g fine dried sample with 3 g of wax which based on standard operating procedure of the x-ray fluorescence (XRF) instrument. Then 10 g from the mixture was compressed into a pellet with a diameter 40 mm and 5 mm thickness. The pellet was test by Philips Axis X-ray digital instrument with result in ppm.
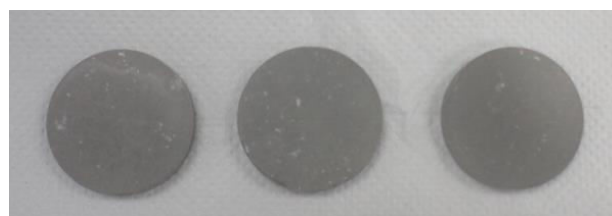

Figure-3. Pellet for XRF analysis.

Contamination assessment

Geoaccumulation index (I_{geo}), contamination factor (C_f), and degree of contamination (C_d) were used to assess metal pollution in dredged marine sediments. Table-1 shows the geoaccumulation index which has seven grades. The geo-accumulation index (I_{geo}) was estimated using the following formula:

$$I_{geo} = \log_2 (C_n/1.5B_n) \qquad (1)$$

where C_n is the measured concentration of heavy metal in sediments and B_n is the geochemical background concentration of the same metal in average shale. The constant 1.5 was introduced to consider the possible variations of the background values due to the lithological variations. The degree of heavy metal contamination in the DMS was determined based on the I_{geo} classes.

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Table-1. Index of geoaccumulation (I_{geo}) of heavy metal in sediment (Muller, 1979).

I_{geo} Class	Sediment accumulation	Pollution Intensity
0	<0	Unpolluted
1	0-1	Unpolluted to moderately polluted
2	1-2	Moderately polluted
3	2-3	Moderately to highly polluted
4	3-4	Highly polluted
5	4-5	Highly to very highly polluted
6	>5	Very highly polluted

The contamination factor (C_f) and the degree of contamination (C_d) were estimated based on the average concentration values of metals following the method of Hakanson (1980). The applied equations are as follows:

$$C_f = M_s/M_b \qquad (2)$$

and

$$C_d = \sum_{i=0}^{n} C_f \qquad (3)$$

where M_s is the metal concentration in the sediment, M_b is the background value of the same metal in average shale and n is number of the investigated heavy metals (in this study n = 6). According to Hakanson (1980), the following terms were used to describe contamination factor (Table-2) and the degree of contamination (Table-3).

Table-2. Contamination factor (C_f) (Hakanson, 1980).

Contamination factor	Description
$1 < C_f$	Low contamination
$1 \leq C_f < 3$	Moderate contamination factor
$3 \leq C_f < 6$	considerable contamination factor
$C_f \geq 6$	Very high contamination factor

Table-3. Degree of contamination (C_d) (Hakanson, 1980).

Degree of contamination	Description
$C_d < 8$	Low degree of contamination
$8 \leq C_d < 16$	Moderate degree of contamination
$16 \leq C_d < 32$	Considerable degree of contamination
$C_d \geq 32$	Very high degree of contamination

RESULTS AND DISCUSSIONS

Concentration of heavy metals

In this study, six elements of heavy metal were detected in all samples. The six elements were arsenic (As), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni) and zinc (Zn). The analyzed metals concentration in Lumut (LT), Melaka (MM), Tok Bali (TB) and Pasir Gudang (PG) dredged marine sediments are shown in Table-4. The highest amount of arsenic (As) was found in MM and PG samples with 18 mg/kg (Figure-4). TB sample contained 16.5 mg/kg and LT sample contained 11.3 mg/kg.

Figure-4b shows the concentration of Cr in dredged marine sedimentss. MM contained highest chromium with 66.7 mg/kg, 59.7 mg/kg for LT, 54.5 mg/kg for PG and 39.5 mg/kg for TB. Meanwhile, the concentrations of Cu for all samples are LT (5 mg/kg), MM (21.7 mg/kg), TB (6.5 mg/kg) and PG (10.5 mg/kg) (Figure-4c).

Table-4. Trace elements of DMS.

Samples	Trace elements (mg/kg)					
	As	Cr	Cu	Pb	Ni	Zn
LT	11.3	59.7	5.0	33.0	18.3	53.7
MM	18.0	66.7	21.7	40.3	22.0	84.3
TB	16.5	39.5	6.5	35.5	12.5	38.0
PG	18.0	54.5	10.5	28.0	9.5	107.0

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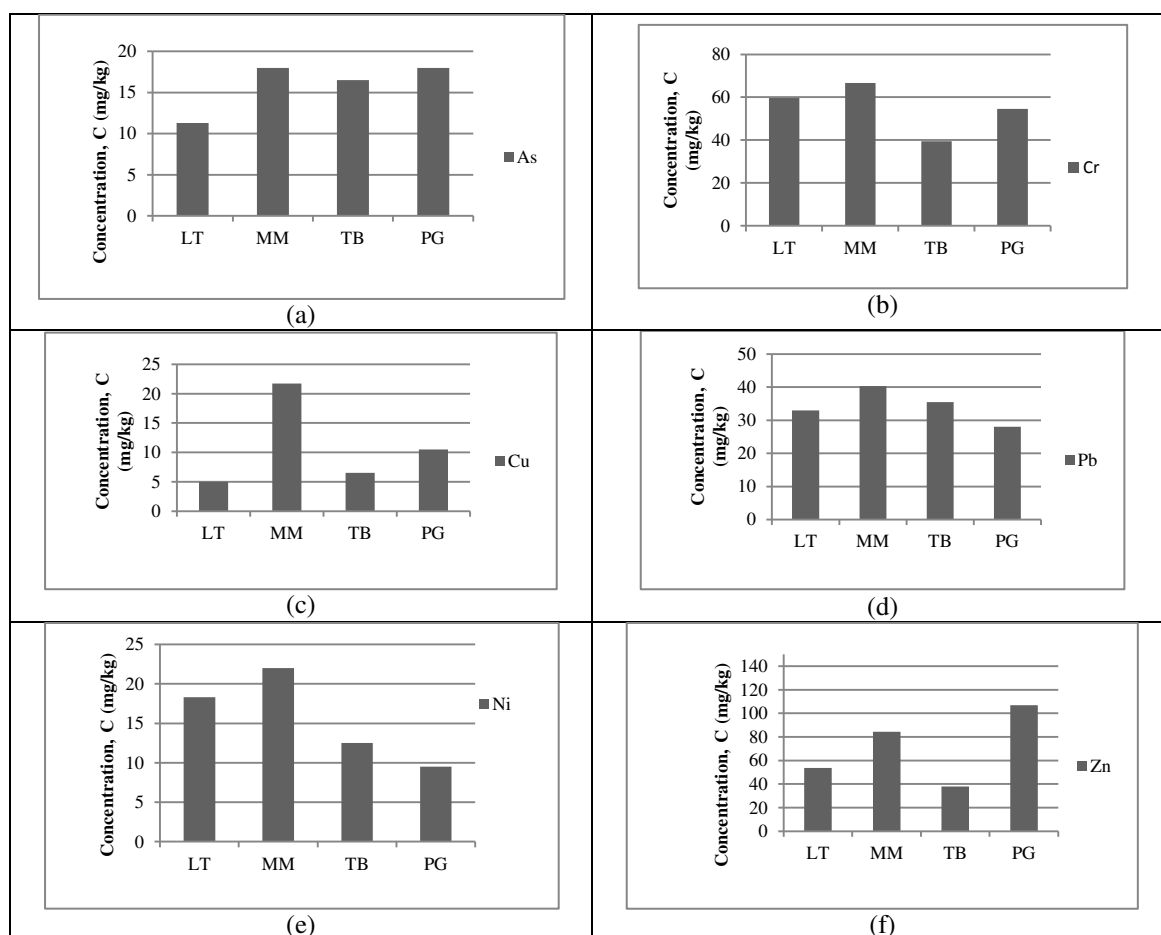
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Figure-4. Trace elements concentrations (As, Cr, Cu, Pb, Ni, Zn) in DMS.

The highest Pb concentration was detected in MM sample (40.3 mg/kg), followed by TB (35.5 mg/kg), LT (33 mg/kg) and PG (28 mg/kg) as shown in Figure-4d. PG contained least amount of Ni compare to the other three samples. The Ni concentration in LT sample was 18.3 mg/kg, MM which indicated the highest concentration 22 mg/kg and TB was 12.5 mg/kg (Figure-3e). Figure-4f shows the concentration of Zn in the dredged marine sedimentss. PG showed highest amount of Zn with 107 mg/kg, followed by MM (84.30 mg/kg), LT (53.70 mg/kg) and TB (38 mg/kg).

Contamination indices

i) Geo-accumulation index (I_{geo})

The spatial distribution of calculated I_{geo} values for each of the studied metals were shown in Figure-5 and Table-5. The I_{geo} values of as for all the sample were greater than 5, indicated that the sampling site were very highly polluted with As. All the sampling sites were unpolluted with Cr, Cu, Ni and Zn. The values of I_{geo} for Pb in LT, MM and TB were between 0 and 1 which showed that these sites were unpolluted to moderately pollute while PG was unpolluted with Pb.
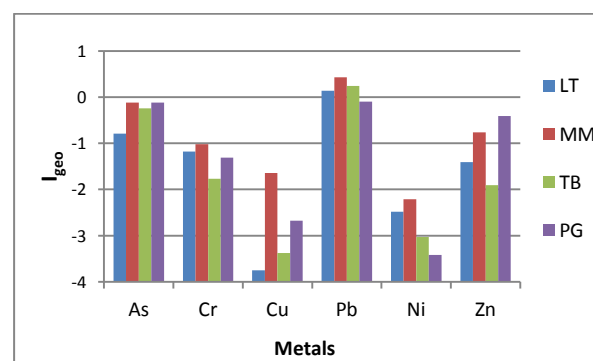
Table-5. Index of geoaccumulation (I_{geo}) of heavy metal in dredged marine sediments.

Location	I_{geo}					
	As	**Cr**	**Cu**	**Pb**	**Ni**	**Zn**
LT	-0.79	-1.18	-3.75	0.14	-2.48	-1.41
MM	-0.12	-1.02	-1.64	0.43	-2.21	-0.76
TB	-0.24	-1.77	-3.38	0.24	-3.03	-1.91
PG	-0.12	-1.31	-2.68	-0.1	-3.42	-0.41

Figure-5. Geoaccumulation index of dredged marine sediments.

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ii) Contamination factor and degree of contamination

All contamination factor and degree of contamination of the pollutants are listed in Table-6. Figure-6 shows the level of contamination factor of the pollutant in the samples. The C_f for arsenic in MM, TB and PG are greater than 1, indicated moderate contamination factor (Figure-6). Leads in all samples also have values greater than 1 and less than 3, which have moderate contamination factor. The C_f value of zinc in PG sample shows moderate contamination factor. The others metals, chromium, copper, nickel were in low contamination. The values of degree of contamination (C_d) for all samples were range 4.13-5.83 (Figure-6), which is below 8. This indicated that all samples have low degree of contamination.

Table-6. Contamination factor and degree of contamination of dredged marine sediments.

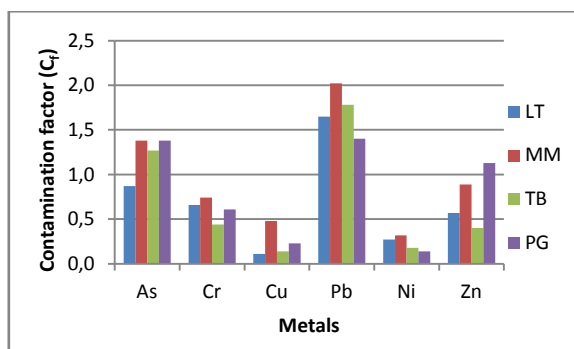| Location	C_f						C_d
	As	Cr	Cu	Pb	Ni	Zn	
LT	0.87	0.66	0.11	1.65	0.27	0.57	4.13
MM	1.38	0.74	0.48	2.02	0.32	0.89	5.83
TB	1.27	0.44	0.14	1.78	0.18	0.40	4.21
PG	1.38	0.61	0.23	1.40	0.14	1.13	4.89

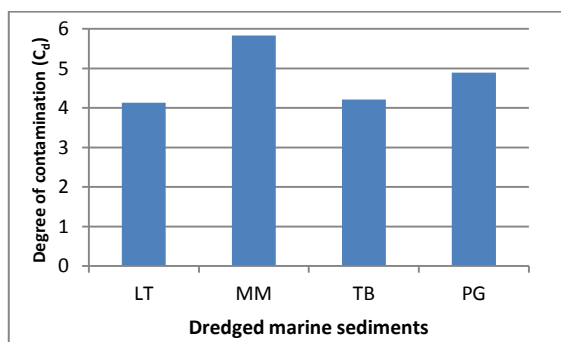**Figure-6.** Contamination factor of dredged marine sediments.

Figure-7. Degree of contamination factor of dredged marine sediments.

CONCLUSIONS

Three contamination indices were used in this study to evaluate the level of metals concentration in DMS. The value of I_{geo} indicated that only Pb in all samples are in class 1 except for PG, class 0. The other 5 metals; As, Cr, Cu, Ni and Zn are categorized in class 0, which is uncontaminated in all samples. The elevated C_f values indicated that the DMS is moderately polluted with As, Pb and Zn. Based on the C_d results, all the DMS are categories as 'low degree contamination.

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