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Vertical Profile of Heavy Metals Concentration in Core Sediments of Sungai Muar, Johor, Malaysia

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Abstract: Two core sediment samples were collected along Sungai Muar, Johor. The down core variation in heavy metals concentrations (As, Cd, Cu, Pb, Zn, Hg) and sediment characteristic were determined by using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) and Particle Size Analyzer, respectively. The result shows the average concentration (ppm) of selected heavy metals in the S4 core followed the order Zn (54.05) >Pb (16.33) > Cu (9.63) >As (3.43) > Cd (0.054) > Hg (0.031). For S5 core sediment, the average concentration for Zn, Pb, Cu, As, Hg and Cd were 59.75, 23.77, 15.04, 3.57, 0.034 and 0.10, respectively. The EF result shows, all the selected heavy metals from S4 and S5 core sediment were came from natural sources (EF<2). For index of geoaccumulation (Igeo), the S4 and S5 core were unpolluted by Zn, Cu, Pb, Cd and Hg, whereas the Aswas in uncontaminated to moderately contaminated status for both core. The average of pollution load values (PLI) was concluded that S5 core was polluted by selected heavy metals, but not in S4. The result of sediment characteristics shows average of mean size value for S4 core sediment was 7.58Ø and was higher than S5 cores, 6.73Ø.

Key words: Particle Size Analyzer · ICP-MS · Enrichment Factors · Index of Geoaccumulation · Sediment characteristics

INTRODUCTION

Heavy metals pollution in aquatic ecosystem has become one of the major critical issues in the world due to their highly toxicity, environmental persistent, non-biodegradable characteristics and ability to be incorporated into food chains [1, 2]. Heavy metal is the type of pollutant that may originate from both natural process and anthropogenic activities [3]. Terrigeneous sources such as weathering of rocks and volcanic eruption arebecome the results for the presence of heavy metals in the aquatic ecosystem [4, 5]. In modern era, industrialization and urbanization have contributed to introduce and increase the level of heavy metals contamination in aquatic ecosystem [6].

River act as the arteries of natural water resources and have made immense contribution to the overall development for country such as provided to power generation, water for domestic and agricultural and industrial consumption. However, the importance role played by rivers for human development has brought with it pollution, degradation and overexploitation. Sediments of the river act as an important geo-marker of heavy metal pollution in the aquatic system and do reflect any environmental changes including the past condition of geochemical and biological condition in an area [7]. The core sediments at Sungai Muar were collected in order tostudy their environmental and geochemical status and determine the strong correlation between human activities to the study areas. In specifically, the purpose

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of this research is to determine the vertical profile of heavy metals in core sediment of Sungai Muar. Sungai Muar is not only used as a route for fisherman activities, but exploited in intensive way by other local surrounding community as a port for shipyard, route for industrial waste and active area for sand mining activities [8]. Furthermore, this study will lead to help for the better aquatic pollution management in the river especially for heavy metals pollution.

MATERIALS AND METHODS

The sampling site of this study was carried out along the Sungai Muar, which located on the southern part of Peninsular Malaysia, Johor (shown in Figure 1). Sungai Muar is one of the major rivers in Malaysia and beginning from a place called "JambuLapan" in Jempol and encompasses an estimated wide of 150 to 300 m [8]. Sungai Muar plays an important role as the natural drainage arteries, meeting the need of an expanding population and supporting agriculture, industrial and socioeconomic development besides being the most important freshwater resource for man. Moreover, this river was flowing through the high polluted area, thus they are highly vulnerable to heavy metal pollution due to urbanization and industrialization. According to the research conducted by Environmental Section (1977), among 42 tributaries in Peninsular Malaysia that was categorized as polluted, Muar River is also undergoing a threatening condition [9].

Sample Collection: There are two vertical core sediment sample were collected by using D-section corer at the downstream and upstream area of the river. The S4 core sample was collected at upstream area of river which is slightly away from the rapid development of Muar town but near with the mangrove area, human housing area and boat jetty. Meanwhile, S5 core sample was obtained at downstream part of the river and near with urban and industrial area. All the samples were preserving at a low temperature and transport to the laboratory for analysis [10,11].

Sediment Samples Preparation: Sediment was cut into segments of approximately 2 cm interval. Any foreign objects such as leaves, snails or shells were discarded. All of the cut sediment samples were transferred to petri disc and dry in an oven at 60°C to a constant weight for one week. Then, the dry samples were grounded into a fine powder by using an agate mortar and pestle and lastly were stored in PE vials until analysis.

Teflon Bomb Digestion: 0.05g sediment sample was transferred into Teflon beaker and 2ml of mixed acid of 3.0 Nitric Acid (HNO3): 3.0 Hydrochloric Acid (HCL): 1.0 Hydrofluoric Acid (HF) was added into the Teflon beaker. The sample was digested within 8 hours at 100 °C in oven. The digested sample was transferred into the 10mL centrifuge tube and Mili-Q water was added to mesh up to 10mL of volume. A clear solution with no residue

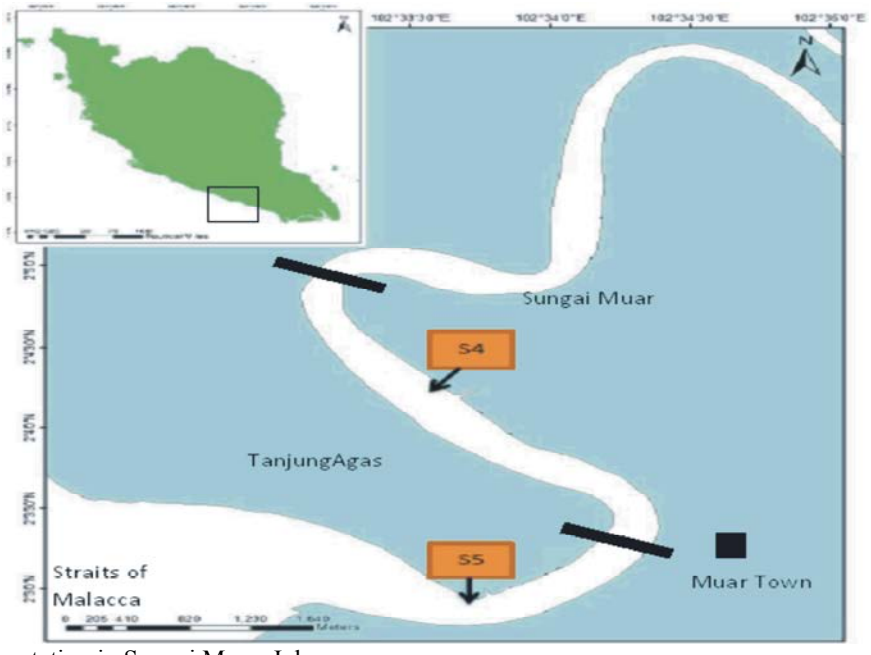

Fig. 1: Map of sampling station in Sungai Muar, Johor

obtained at this stage. Finally, chemical component and metal content was determined and identified by using the instrument of ICP-MS [12,13].

Laser Diffraction Method: The 10 mL of grinded sediment sample was transferred into 50mL centrifuge tube. Then, distilled water was added until reach 40 mL. After that, 10 mL of 20% Calgon solutions (Sodium Hexametaphospahate) was added into centrifuge tube to disperse the sample into single particle. Finally, the samples were analyzed by using Laser Diffractometer Analyser [7].

RESULT AND DISCUSSION

Heavy Metals Distribution: Copper is an essential nutrient and required by the body in very small amount [14]. The calculated average concentration of Cu in S4 (9.63 ppm) and S5 (15.04 ppm) core sediment can be considered as uncontaminated for S4 core contaminated in S4 due to the average level of concentration was far below the background concentration Upper Continental Crust (14.3 ppm) but slightly contaminated in S5core sediment. These indicate that in S4 core, the point sources of this element come from limited sources such as antifouling paint from ships and sewage sludge dumpsites [15]. The relatively excess of Cu concentration in the surface layer of S5 core sediment (29.02 ppm) shown in Figure 2, indicate the presence of recent anthropogenic input to this area including the sewage from roadside and discharge from industrial [16,17]. The lower average Cu concentration in S4 core when compare to S5 core might due to the there are no significant sources of Cu as S4 core located at upstream area that slightly far from main city and industrial area.

Among the selected heavy metals, Zn was show the highest concentration. It might due to its position as 22nd place as abundant element in earth crust [19] The average concentration of Zn (54.05 ppm) in S4 core sediment is slightly higher than upper continental crust (UCC), 52 ppm; suggest that the point sources input including discharges from municipal and sewage effluents to environment were increase the concentration as S4 core was collected near human population area [21]. However, concentration of Zn in S5 core sediment was little bit higher than in S4 core, with value 59.75 ppm. This is probably due the presence of sewage drainage from mainland, agricultural and urban runoff as well as antifouling paint and other human activities that contribute to the enrichment of Zn at S5 core sediment. Other than mentions anthropogenic inputs, the higher concentration of Zn at the upper surface layer in both S4 and S5 core (Figure 2) also contribute by weathering of ultramatic rock [22].

Cd has found widespread used in industrial application mainly in metals plating and chemical industries. The average concentration of Cd in both S4 and S5 core sediment represent the different value which is 0.054 ppm and 0.10 ppm, respectively. The lower average concentration of Cd in S4 suggested that the entrance of Cd come from natural sources such as windblown transport of soil and atmospheric deposition [15,31] in which most of them deposited into bottom sediment. The value of Cd concentration in S5 core was slightly increases when going down from the surface layer and distinct increases was clearly seen start from layer 36cm to 72cm (Figure 2) with the value was 0.11 to 0.20 ppm. The increment concentration value of Cd at the deep layer of core sediment might due to anthropogenic sources of Cd adsorbed by sediment during several decades ago, including discharge of refining waste and untreated sewage effluent [32], agricultural runoff and sand mining activities. The vertical distribution pattern of Cd along the S4 and S5 cores showed fluctuation, this indicate Cd is sensitive to redox changes, where it is known to be soluble in oxygenated conditions and precipitate immediately here post-oxic condition are encountered [33].

Lead (Pb) known as non-essential trace element which toxic to biota when reach certain concentration. Regarding to average concentration, S5 (23.77 ppm) core sediment relatively more polluted with Pb in comparison with S4 core. This condition revealed the atmospheric input that generated from the automobile exhaust emission can be attributed as the most significant sources of Pb to investigated area that situated close to the main road of the cities [23]. The study shows the average value of Pb in S4 core (16.33 ppm) was slightly lower than upper continental crust (UCC) with value 17 ppm [26]. The possible potential sources of Pb in S4 core including leaded petrol stemmed from outboard boat engines from boating activities, as S4 core collected near with boat jetty. On the other hand, Abu-Hilal [27] attributed high Pb concentration to several sources including boat exhaust system, spillage of oil and other petroleum from mechanized boat. The presence of higher concentration of Pb in top layer of S5 core sediment (22.95-37.00 ppm) shown in Figure 2 indicate the vehicle emission that consist of higher proportion of Pb was flow into the river by way of road runoff through the stromwater drain [28].

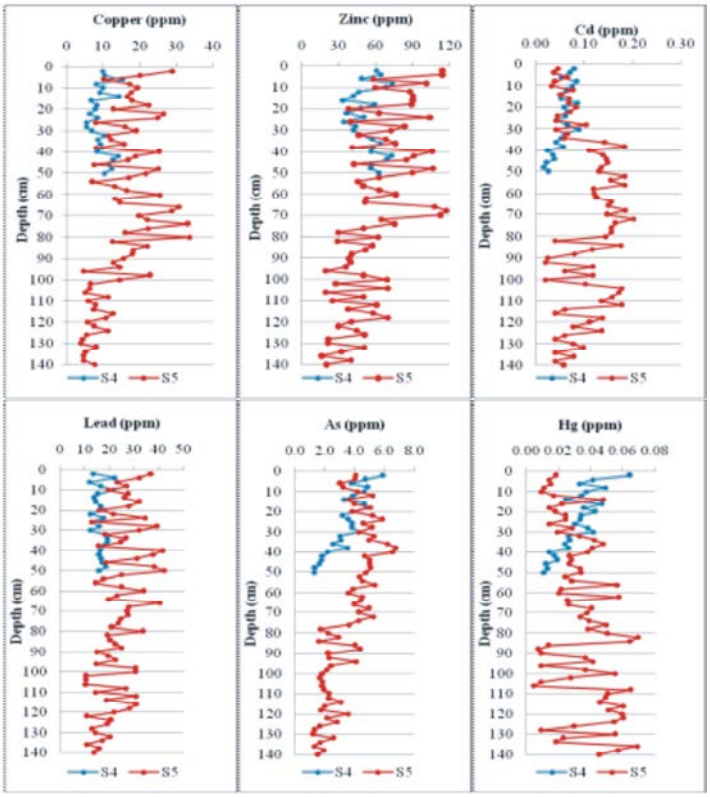

Fig. 2: Concentration of selected heavy metal (ppm) for both S4 and S5 core sediments in Sungai Muar

Moreover, as cited by Segura *et al.* [25] the existence of factories, workshop, industrial activities and disposal activities especially batteries in surrounding area also become the medium that increase the accumulation of Pb in S5 core sediment.

The vertical profile of As for S4 core sediment shows the higher concentration in the upper 10 cm of sediment layer with range of 4.62 to 5.94 ppm (Figure 2). This may be attributed to the atmospheric deposition of waste from fuel combustion activities and human induced activities since S4 located slightly away from urban and industrial area. On the other hand, the higher concentration of As at the surface layer of S4 and S5 core sediment was influenced by early diagenetic processes [24]. In comparison, the average concentration of As in S5 core sediment was higher (3.57 ppm) than S4 core sediment (3.43 ppm) and upper continental crust (2 ppm). Based on Figure 2e, most of the layer of sediment in S5 core were shows higher concentration of As. This indicates the industrial processes including mining, burning of fossil fuels, waste recycling and urbanization discharge has contributed significantly toward the increasing, as S5 was located near the main urban area and Tanjung Agas industrial area. In S4 core, the concentration of Aswas observe in decreasing pattern when going down to the depth layer of sediment (Figure 2e). Mean that, there are no significant sources of As in upstream area of river during several decades ago.

Mercury and its compound play an important part in electrochemistry, used as coolant in certain type of reactor. The average concentration of Hg in S5 core sediment was 0.034 ppm and higher than in S4 core sediment (0.031 ppm). However, the average concentration for both S4 and S5 core was remain below the upper continental crust (0.056 ppm).These suggest there are no significant sources Hg in Sungai Muar. In Figure 2, the maximum concentration of Hg in S4 core sediment can be found at the upper layer of the sediment with value 0.065 ppm, this may attributed to source of Hg might come from atmospheric deposition. Local industrial activities emit Hg into the atmospheric, which can end up in aquatic sediment via wet deposition and sedimentation. As cited by Lamborg *et al.* [29], estimated that 360±60 kg/year of Hg were deposited resulting from wet atmospheric deposition from global sources. Moreover, the higher Hg concentration in the young sediment layer reveal that the emission of Hg may be related to fossil fuel combustion, iron and steel production, sewage sludge and medical waste incinerators [30].

Table 3: Sediment characteristics of S4 and S5 core

Core Sediment	Average and range of Mean Size (Ø)	Average and range of Sand (%)	Average and range of Silt (%)	Average and range of Clay (%)
S4	7.58(7.46-7.71)	0.635(0.00-2.53),	70.68(68.67-72.17),	28.64(26.94-30.93),
S5	6.73(4.88-7.69)	17.48(0.16-58.83),	62.01(32.67-73.24),	20.37(8.47-36.37),

Table 4: The average of Enrichment Factor (EF) value for S4 and S5 core sediment in Sungai Muar

	S4		S5	
Metal	Average of EF value	Heavy Metals Sources	Average of EF value	Heavy Metals Sources
Cu	0.628	Natural	0.679	Natural
Zn	0.973	Natural	0.740	Natural
Pb	0.917	Natural	0.914	Natural
As	1.695	Natural	1.197	Natural
Cd	0.526	Natural	0.713	Natural
Hg	0.547	Natural	0.443	Natural

Sediment Characteristics: River sediment is a mixture of sand, silt and clays which normally come from the sea or land or both. The sediment grain size characteristics and measurement of surface area of sediment are very useful information to sedimentologist, who interest and carried out a research regarding on the soil characterization and trace metals chemistry. Generally, river sediment composed of fine grains size of sediment, such as clays minerals. As a result, heavy metals introduced into an aquatic environment can easily absorbed by sediments due to larger surface area per volume. The result of grain size analysis revealed that Sungai Muar consist of coarse, medium and fine silt. In fact, Table 3 shows all the layer of sediment in both S4 and S5 core show the range value of mean size (Ø) were from 4.48 to 7.71. However, the average of mean size value in both S4 and S5 core sediment were 7.58 Ø and 6.73 Ø, respectively. This indicate that S4 core sediment was classified as fine silt while medium silt for S5 core. High mean value represent fine grain size of sediment and low mean value indicates coarser sediment. The higher average of mean size value in S4 core compare to S5 was probably due to calm condition and weaker current action at the area where S4 core sediment was collected (upstream) allows the deposition of fine and suspended sediment to the bottom.

On the other hand, reduce the hydraulic energy will cause the fine sediment from water column have sufficient time to deposit into bottom [34]. The mean sizes profiles are distributed almost similar along the layer in both S4 and S5 core sediment. Moreover, according to Huang and Lin [35], grain size was influence indirectly by high precipitation during sampling period. The percentage of sand texture in S5 core was higher than S4 core which is the value were 17.48 % and 0.63 %, respectively. This suggest that the wave action, tides cycle and strong water current at S5 core sediment allows the coarse grains to get deposited and washout the fine and suspended sediment [23], since S5 core located near to the mouth of river. For S4 core, the percentage texture of silt and clay were distributed almost similar when going down the depth layer and all of the layers consist of fine sediment. The higher percentage of silt (70.68%) and clay (28.64%) was found in S4 core compare to in S5 core which is the value was 62.01% and 20.37%, respectively. As S4 core was collected at area near with mangrove region, the stronger hydrodynamic energy including tidal action and baroclinic circulation were able to be decrease, which provides times for sinking and re-deposition of silt and clay sediment [34]. In addition, the higher silt and clay percentage observed in S4 core might be due to mangrove tree that able to create their own ecosystem by trapping the fine sediment [36].

Enrichment Factors (EF): The purpose of EF is to determine whether the sediment of study area is affected by anthropogenic sources or naturally effects. Al was chosen as references element in EF calculation Al is the second abundant metals in crust of earth, highly refractory and not influenced by anthropogenic sources [37, 13]. The Table 4 shows the average EF value of the S4 core sediment followed the order As (1.695) > Zn (0.973) >Pb (0.917) > Cu (0.628) > Hg (0.547) > Cd (0.526). For both S5 and S4 core sediment, only As shows the average EF value more than 1 but this metals still categorized as come from natural sources since EF value was less than 2 (EF<2). Moreover, EF value between 0.5 and 1.5 indicate the given metals are entirely derived from crustal materials or natural weathering processes [38]. Based on the average EF value, the contribution from anthropogenic sources was negligible for all selected heavy metals in both S4 and S5 core sediment.

Table 5: Geo-accumulation indices of heavy metals in S4 and S5 core sediment

	S4		S5	
Metals	Average of I-geo value	Pollution Level	Average of I-geo value	Pollution Level
Cu	-1.2092	Uncontaminated	-0.7243	Uncontaminated
Zn	-0.5640	Uncontaminated	-0.5473	Uncontaminated
Pb	-0.6630	Uncontaminated	-0.1947	Uncontaminated
As	0.0826	Uncontaminated to moderately contaminated	0.1129	Uncontaminated to moderately contaminated
Cd	-1.6126	Uncontaminated	-0.7983	Uncontaminated
Hg	-1.5589	Uncontaminated	-1.5339	Uncontaminated

Geoaccumulation Index (I-geo): I-geo is effective and convenient way to determine sediment quality [39]. Table 5 is revealed the quality of core sediment at the Sungai Muar. The I-geo result demonstrate that S4 and S5 core sediment in Sungai Muar were uncontaminated by Cu, Zn, Pb, Cd and Hg indicating that the sediment of the study area are in background value respect to this metals. The average i-geo value for As in both S4 and S5 core sediment were classified as uncontaminated to moderately. This situation suggest the industrial processes including mining, burning of fossil fuels, waste recycling and urbanization discharge has slightly influence the concentration of As both S4 and S5 core sediment.

Pollution Load Index (PLI): PLI is quick tool in order to compare the pollution status of different site of study area [41]. PLI represent the number of times by which the metals content in sediment exceed the background concentration and gives summative indication of the overall level of heavy metals toxicity in a particular sample [42]. In S4 core sediment, the pollution load value varied from 0.548 to 0.969. Table 6 shows the average value of PLI in S4 core was 0.744, this represent the sediment were unpolluted by selected heavy metals (PLI <1). The average of PLI value for S5 core sediment was 1.034 with the range 0.398 to 1.702 along the vertical layer. The average value suggested that the S5 core sediment in Sungai Muar were polluted with respect to the selected heavy.

Correlation Coefficient: Based on Table 6, the negative correlation were observed in S5 core sediment for As, Cd, Pb and sand contents with the correlation coefficient were-0.472,-0.008 and-0.255 respectively. These indicate that these metals are not associated with sand [18]. In S4 core, Cu, Zn and Pb shows negative correlation with average of mean size with the value are-0.302,-0.287 and-0.183, respectively. This negligible relationship indicates that mean size of sediment might not play a significant role in controlling the vertical distribution of these metals. In both S4 core sediment, As, Cd and Hg shows very strong correlation coefficient between them with the value in range of 0.833-0.941. This condition suggesting that these metals are associated with each other and indicate they are might have similar sources in the sediment of Sungai Muar. The higher correlation coefficient between Cu-Zn, Cu-Pb and Zn-Pb can be seen in S5 core (Table 6), indicate these metals have common contamination sources [20].

Table 6: Average and range value of Pollution Load Index Value in S4 and S5 core sediment

Core Sediment	Average of PLI value	Range of PLI value
S4	0.744	0.548-0.969
S5	1.034	0.398-1.702

Table 6: The correlation value between selected heavy metals and sediment characteristics in S4 and S5 core

S4 Core		S5 Core	
Relationship	r-value	Relationship	r-value
As-Cd	0.873	Cu-Zn	0.714
As-Hg	0.941	Cu-Pb	0.714
Cd-Hg	0.833	Zn-Pb	0.806
Cu-Mean size	-0.302	As-Sand	-0.472
Zn-Mean size	-0.287	Cd-Sand	-0.008
Pb-Mean size	-0.183	Pb-Sand	-0.255

CONCLUSION

In conclusion, both S4 and S5 core sediment was contaminated by Asand Zn since their average concentration along the vertical depth represent higher than upper continental crust (UCC). For Pb, the concentration level in S5 core sediment was exceed the UCC value and this indicate there are significant source of Pb enter the river including sewage effluent from urban and vehicle emission.

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