

Contamination and Pollution Assessment of Heavy Metal in Kuala Perlis Sediment Phase: A Statistical Analysis Approach

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ABSTRACT

A study on heavy metals pollution in sediment of Kuala Perlis was done which four types of heavy metals that are identified in the coastal area. The presence of Cr, Pb, Zn and Cu are resulted from the anthropogenic activities occurs in the land use of Kuala Perlis. Samples were collected with different depth phase (subsurface, 1ft, 2ft and 3ft) at five different points (A,B,C,D and E) based on the different land use activities. The distribution concentrations of Cr, Pb, Zn and Cu in each point were analyzed by using descriptive analysis. Based on the analysis Pb gives the highest in mean concentration compared to others in all points. The relation of metals concentration and the sediment characteristics were analyzed by Pearson correlation analysis. The correlation analysis shows the positive correlation among the metals with percentage of silt and clay and negative correlation among the metals with the sediment pH. The sediment pollution assessments were investigated by using contamination indices such as Enrichment Factor (EF), Geo-accumulation Index (Igeo), Contamination Factor (CF) and Pollution Load Index (PLI). Among the four metals tested Pb gives the highest value of EF, Igeo and CF in sediments collected from each sampling points meanwhile point A gives the highest PLI value. Based on the terminologies, since the PLI value <1 so, the sediment in Kuala Perlis coastal area are considered as unpolluted sediment.

Keywords: Heavy Metals, Sediment, Descriptive Analysis, Pearson Correlation, Contamination Indices

1. INTRODUCTION

The coastal area in the marine environment is being continuously polluted with chemical pollutants such as heavy metals resulted from the lithogenic and anthropogenic activities [1]. Lithogenic refers to the natural process of rocks weathering and volcanic activities that enriching the water reservoirs and waterways with heavy metals [2]. Meanwhile, anthropogenic activities refers to the human activities rises from the industries, agricultures, and urban development constructions that transported the pollutants to the marine water by the rivers and waterways [2]. These chemical pollutants, leads to the bad implications towards the marine environment and human populations [1].

The increasing of anthropogenic activities in Kuala Perlis, such as industries and shipping activities that operates daily, more toxic contaminants were release to the marine waterways [3]. These toxic contaminants such as heavy metals finally deposited in the marine sediment and undergo the process of bioaccumulations over a certain time exposure [4]. Within this period, the metals could

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leach its toxicity and this will give the dangerous effect towards the marine ecosystems. Since the heavy metals pollution cannot be seen by naked eyes, so a pollution study must be conducted to assess the quality of the sediment in Kuala Perlis. This study is important to give the understanding and basic knowledge to the community regarding the pollution degree in the area as well as provide a supervision for the future environmental control.

The purpose of this study was to identify the type and distribution of heavy metals that polluting the sediment of Kuala Perlis Coastal as well as to assess the degree of sediment pollution based on different depth and land use activities. Descriptive analysis was used to describe the distribution of the metals, meanwhile Pearson correlation analysis was used to examine the relationship between heavy metals and the sediment characteristics. Moreover, by applied the mathematical equations such as the contamination indices to the sediment phase as the contamination indicator in this study will enable us to obtain a good evaluation of the degree of pollution in Kuala Perlis coastal area [5].

2. MATERIALS AND METHODS

2.1 Research Area and Sampling Location

20 sediment samples were collected at five different points pointed as A (Jetty port and Sg. Perlis estuary), B (Seafood restaurant), C (Roadside area), D (Power plant) and E (Sg. Bahru estuary and residential area) along Kuala Perlis coastal area as shown in Table 1. The samples on each point were taken at different depth which were the subsurface (0-15cm), 1ft, 2ft and 3ft by using had mud auger. The sediment samples were taken to laboratory for further analysis.

Table 1 Location of research area

Point	Location	Coordinate	Sea level
A	Jetty port	N06°23'56.6" E100°07'46.1"	± 7m
B	Seaside seafood restaurant	N 06°23'41.3" E100°07'41.8"	± 6m
C	Main road, village area	N 06°21'57.0" E100°08'34.8"	± 8m
D	Power plant	N 06°20'21.5" E100°09'08.4"	± 11m
E	Fisherman floating village, residential area	N 06°19'52.1" E100°09'23.7"	± 6m

2.2 Experimental Methods

The sediment samples were oven dried at 110°C for 24 hours. The characteristics of sediment such as moisture content were measured and the sediment pH was examined by pH meter. The dried samples were sieved according to ASTM D422-63 to obtain the particle size distribution of the sediment. The dried samples also were sieved into 60 µm to identify the types of heavy metal by using XRF (X-Ray Fluorescence) analyser [3]. Then, 2g of dried samples was digested with H₂O₂, HNO₃, HClO₄, and H₂SO₄ [6] [7]. The digested samples were filtered and analysed by AAS (Atomic Absorption Spectrometer) to obtain the metal's concentration [6].

2.3 Statistical Analysis

Descriptive analysis such as mean, median, maximum and minimum value of the data were used to calculate the statistical concentration of metals to evaluate data distribution [8]. Followed by the simplest form, boxplot present the visual display of the metal concentration in five sample statistics which includes minimum value, lower quartile, median, upper quartile and maximum value [9]. Pearson correlation analysis was done to investigate the relationship between metals concentration (Cr, Cu, Pb and Zn) and the sediment characteristics (pH, moisture content, particle size distribution and Fe concentration). All statistical analysis were conducted and processed with IBM SPSS statistics V23.0.

2.4 Contamination Indices

Four types of contamination indices were used in this study to assess the sediment pollution. Equation 1, 2 3 and 4 were used to calculate the value of Enrichment Factor (EF), Geo-accumulation Index (Igeo), Contamination Factor (CF) and Pollution Load Index (PLI) respectively. The average shale concentration given by Turekian and Wedepold [1] were used as the background concentration for the tested element while Fe is chosen as a reference element[10] since it is the normalizer and major element in earth crust [11]. The indices values were analysed according to the terminologies of contamination indices as shown in Table 2.

$$EF = \frac{\left(\frac{C_n}{C_{ref}}\right)_{sample}}{\left(\frac{B_n}{B_{ref}}\right)} \quad (1)$$

where C_n (sample) is concentration of the examined element in examined environment, C_{ref} (sample) is the concentration of the examined element in reference environment, B_n is the concentration of the reference element in examined environment and B_{ref} is the concentration of the reference element in reference environment [11].

$$I_{geo} = \text{Log}_2 \left(\frac{C_n}{1.5B_n} \right) \quad (2)$$

where C_n is the concentration of metal in examined environment, and B_n is the concentration of metal in reference/background environment [12].

$$CF = (C_{sample}/C_{background}) \quad (3)$$

Where C_{sample} is the metal concentration of the sample and $C_{background}$ is the background concentration of the metals

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{\frac{1}{N}} \quad (4)$$

where CF is the contamination factor of the element while N is the number of metals[13].

Table 2 Terminologies and categories of contamination indices assessment

EF		I_{geo}			CF		PLI	
Value	Enrichment condition	Value	Class	I_{geo} condition	Value	Condition	Value	Condition
EF<1	No enrichment	≥ 0	0	Unpolluted	CF<1	Low CF		
EF=1-3	Minor enrichment	0-1	1	Unpolluted-moderately polluted	$1 \leq CF < 3$	Moderate CF		
EF=3-5	Moderate enrichment	1-2	2	Moderately polluted	$3 \leq CF < 6$	Considerable CF	>1	Polluted
EF=5-10	Moderately severe enrichment	2-3	3	Moderately-strongly polluted				
EF=10-25	Severe enrichment	3-4	4	Strongly polluted				
EF=25-50	Very severe enrichment	4-5	5	Strongly-extremely polluted	CF ≥ 6	Very high CF	<1	Unpolluted
EF>50	Extremely severe enrichment	>5	6	Extremely polluted				

3. RESULTS AND DISCUSSIONS

3.1 Descriptive Analysis

The descriptive statistics and boxplot of Cr, Cu, Pb and Zn for 5 different sediment points known as point A, B, C, D and E are shown in Figure 1. Based on the ascending mean of the metals in point A, B and C show a similar pattern on the element concentration with (Pb>Zn>Cr>Cu) that Pb gives the highest while Cu is the lowest concentration respectively. However, a result of point D and E shows that the highest concentration is recorded by Pb while Cr gives the lowest mean concentration. Based on the maximum value, Pb shows the highest value in point E, Cr shows the highest in point A, Cu and Zn highest in point E. Whereas the minimum concentration of Pb, Cr, Cu and Zn is recorded at point B, E, D and E accordingly.

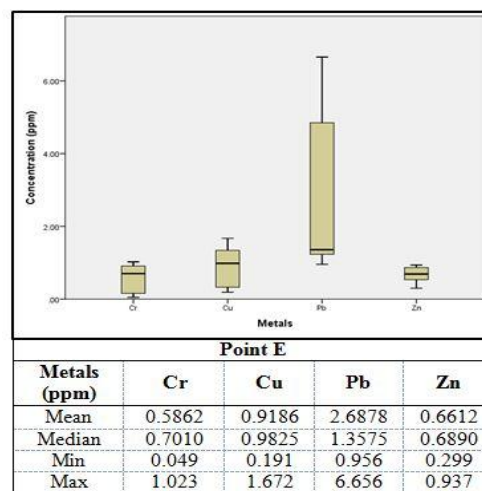
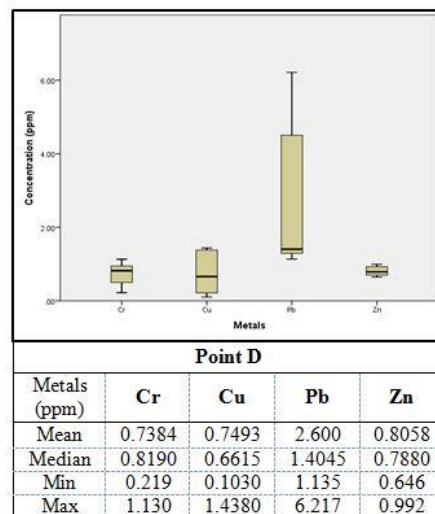
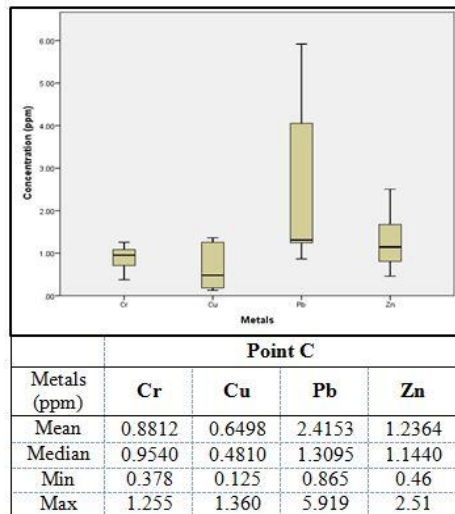
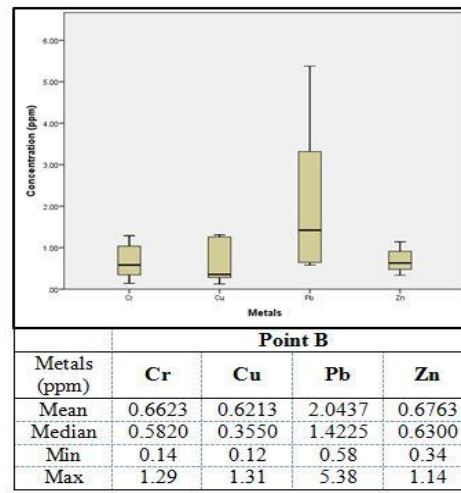
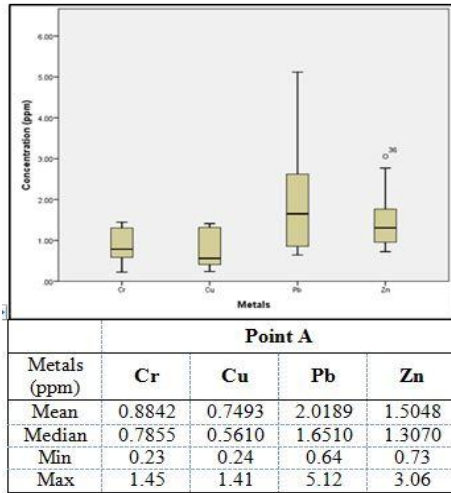


Figure 1. Boxplot and descriptive analysis of metals concentration in each study point.

The boxplot of Pb shows the positive skewness distribution since the median are close to the lower quartile and the data average of Pb concentration are skewed to the right and it indicates a distribution with an asymmetric tail extending toward more positive value in all of the points. Meanwhile, Zn and Cu in point E shows the negative skewness distribution since the median are close to the upper quartile whereas in point A, B, C and D both of these metals shows positive skewness distribution. As for Cr, point A and B gives the positive skewness distribution while negative skewness distribution in point C, D and E. Based on these analysis, it concludes that, Pb is the main contribution of the heavy metals concentration in Kuala Perlis.

The anthropogenic activities such as manufacturing industries, smelting and refining of metals, fossils or gasoline fuels burning, sewage sludge and domestic water waste contribute to the release of high Pb concentration in Kuala Perlis[14][5]. Besides, chromium is the source from electroplating, steel manufacturing leather tanning and textile industries that produce dyeing wastewater and sludge release to the marine waterways[15][5]. As for the main contributors of Cu are the improper disposal of waste such as the lubricants, automobile waste containing copper wires, electrodes and alloy that carried by the wastewater flowing to the coastal waterways[3]. The existence of Zn in the sediment are due to mining and coal combustion, steel processing in industries ,fertilizers and manufacturing that containing metals [16].

3.2 Pearson Correlation

The Pearson correlation analysis tabulated in Table 3 was used to analyse the correlation between the metals concentration and sediment characteristics. Cr and Zn significantly have the strong positive correlation while Pb gives strong negative correlation with the sediment moisture content (MC). Moisture content in the sediment plays the important roles of metal mobilization between the interphase of sediment and sea water exchange. The interchange process decreases in low water content and this resulted the high in precipitation and sedimentation of metal from the water to the sediment thus increases the level of the metal concentration [17]. During the high in moisture content, the increase in metal resuspension and desorption in sediment phase, which increase the concentration of metals in water flows thus increasing the chances of the interaction between the over layers water and the sediment surface [17].

The pH correlated negatively with the metals concentration which Cr gives the strong negative correlation among the four metals. The decrease in pH value resulted in increase the metals concentration in the sediment phase due to the low pH sediment could facilitate the migration, mobilization and availability of heavy metals in sediment [8]. Cr is significantly correlate with the particle size distribution which shows highly correlation in sand and negatively correlation in silt and clay. Sediment grain size plays the significant factors in the concentration and distribution of heavy metals in estuarine sediment [1]. The metal capacity absorption is in the order of clay>silt>sand since the fine fraction has the high capacity to retain heavy metals compared to the other fraction[1] [18]. In addition, there are some of the metals that were positively correlated to each other. Fe shows high positively correlated towards Cr and Zn, Cr shows highly correlated with Zn as well as Pb and Cu. The high positively correlation of these metals is due to the metals have the similar anthropogenic or lithogenic origins and chemical behaviour which probably presented partly in oxide form and partly in hydroxide form [8]. So, by comparing the parameters that have analysed, moisture content affect most of the metals distribution and concentration in the sediment

Table 3 Correlation analysis of metals and sediment characteristics

	MC	pH	Sand	Silt & Clay	Fe	Cr	Pb	Zn	Cu
MC	1								
pH	-0.327	1							
Sand	-0.106	-0.101	1						
Silt & Clay	0.106	0.108	1.000**	1					
Fe	0.397	-0.032	0.285	-0.283	1				
Cr	0.499*	-0.532*	-0.208	0.218	0.580**	1			
Pb	-0.467*	-0.008	-0.036	0.035	-0.163	-0.196	1		
Zn	0.476*	-0.402	-0.186	0.179	0.469*	0.656**	-0.431	1	
Cu	-0.159	0.260	-0.589**	0.596**	-0.409	-0.435	0.452*	-0.208	1

Contamination Indices

Enrichment Factor (EF)

Table 4 represent the minimum, maximum and mean value of Enrichment Factor (EF) of Cr, Pb, Zn and Cu respectively at four different depths in the study area. By referring to the terminologies tabulated in Table 2, the mean of each metal was analysed and assessed the environmental condition.

Table 4 Enrichment factor of metals in sediment phase

		Enrichment Factor (EF)											
Metal conc (ppm)		Cr			Pb			Zn			Cu		
Point/Depth (ft)	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	
A	0ft	0.82	1.85	1.46	4.29	33.31	16.30	1.11	2.15	1.75	0.69	3.68	1
	1ft	1.09	2.36	1.62	6.76	16.29	12.23	1.45	3.04	2.10	0.86	4.45	2.29
	2ft	0.60	3.15	1.90	7.67	36.10	20.59	1.82	6.96	4.06	2.46	7.42	4.32
	3ft	0.58	2.46	1.43	6.22	32.90	17.15	1.61	5.06	2.92	2.18	4.94	2.79
B	0ft	0.92	3.22	1.96	6.55	34.79	19.49	1.33	2.70	1.89	1.52	6.56	3.22
	1ft	0.81	3.10	1.78	7.69	44.73	23.59	0.97	2.64	1.71	1.44	7.22	3.51
	2ft	0.68	2.18	1.37	6.30	41.26	20.25	0.96	1.89	1.41	0.99	5.46	2.68
	3ft	0.44	3.28	1.84	8.89	75.75	34.75	1.00	2.63	1.68	0.77	7.75	3.68
C	0ft	0.72	1.20	1.03	4.78	20.56	10.86	0.60	2.91	1.80	0.41	3.04	1.51
	1ft	0.54	1.64	1.12	6.99	28.19	14.46	0.62	2.36	1.60	0.36	3.63	1.77
	2ft	1.49	2.19	1.82	9.51	40.67	20.43	1.14	2.86	1.95	0.70	4.74	2.39
	3ft	0.80	2.40	1.70	11.56	52.80	25.35	1.73	2.20	1.95	0.66	5.27	2.70
D	0ft	0.70	1.50	1.22	8.63	32.06	17.05	1.05	1.59	1.32	0.90	4.64	2.53
	1ft	0.47	1.42	1.02	7.41	27.04	14.25	0.92	1.15	1.06	0.41	3.54	1.86
	2ft	0.90	1.47	1.18	7.86	37.66	18.46	1.04	1.16	1.10	0.40	4.25	2.24
	3ft	0.45	2.06	1.33	11.78	57.82	27.45	1.26	1.79	1.46	0.43	5.94	3.20
E	0ft	0.11	1.90	1.03	10.80	38.86	20.36	1.01	1.56	1.19	1.22	5.73	3.52

1ft	0.24	1.82	1.12	9.67	44.10	21.64	0.92	1.57	1.23	1.26	5.69	3.51
2ft	0.89	4.90	3.27	26.61	125.43	61.32	1.63	4.25	3.35	2.15	16.00	9.43
3ft	0.22	3.79	2.38	19.47	135.53	61.05	1.28	2.99	2.41	1.73	14.94	9.03

The assessment reveals that, Cr shows the minor enrichment for point A, B, C and D in all of the phases. Point E gives the similar condition in 0-1ft but slightly different in 2-3ft which in moderate enrichment. The severe enrichment of Pb shows in all phases in point A. Meanwhile, point B and point C shows the severe Pb enrichment in 0-1ft and very severe enrichment for 2ft-3ft. The phase 0-2ft of point D reveals that Pb is in severe enrichment while 3ft in very severe enrichment. Point E shows that 0-1ft are in severe enrichment condition and 2-3ft in extremely severe enrichment of Pb in the sediment. Besides, point A and E gives the similar result of Zn enrichment which stated that the minor enrichment in 0ft,1ft and 3ft while 2ft gives the moderate enrichment. The minor enrichment of Zn in all the phases also shown in point B,C and D. The minor enrichment of Cu for 0ft,1ft and 3ft and moderate enrichment for 2ft shows in point A while point B reveals the opposite results which moderate enrichment in 0ft,1ft and 3ft and minor enrichment in 2ft. Point C and D gives the similar enrichment which is minor enrichment in all phases. Moderate enrichment of Cu in 0-1ft and moderately severe enrichment in 2-3ft of point E. This concludes that, Pb shows the high in enrichment in the sediment of all of the points.

In addition, Figure 2 shows the mean of EF value in different depth of the sediment sample collected from the study point. Based on the graph Pb gives the highest EF mean in all of the point. The enrichment increases as the depth of the sampling point increases. The condition was due to the accumulation of metals as the bottom of sediment over a period of time [19]. The accumulation causes the metals were unable to migrate away thus increase the potential of sediment contamination due to the leaching of metals toxicity process from the bottom of marine sediment [19] [20].

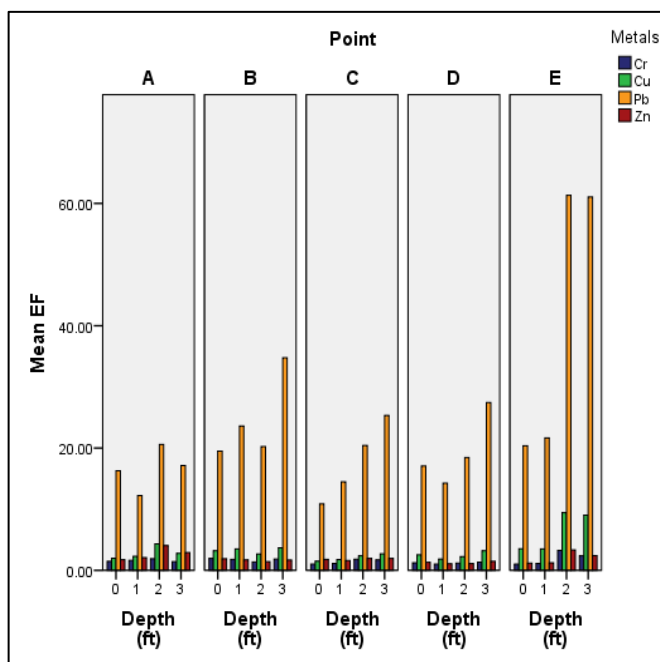
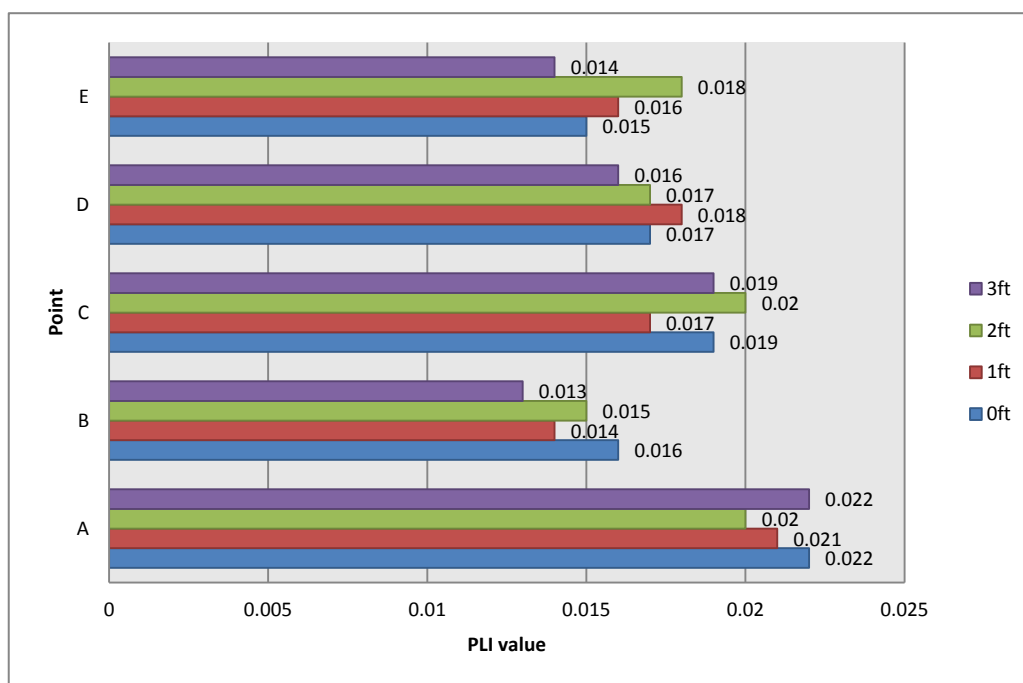
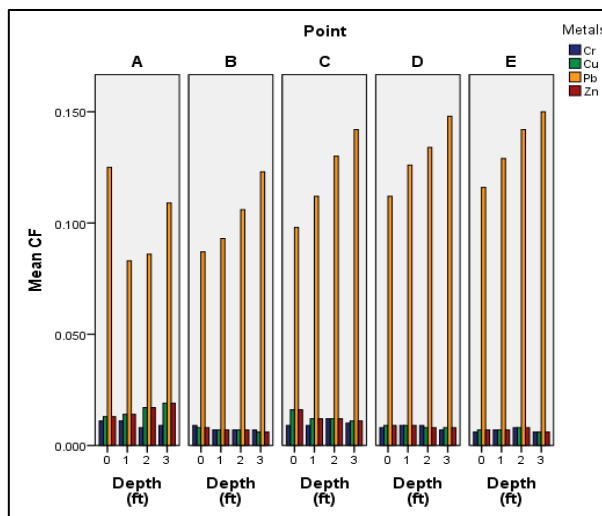
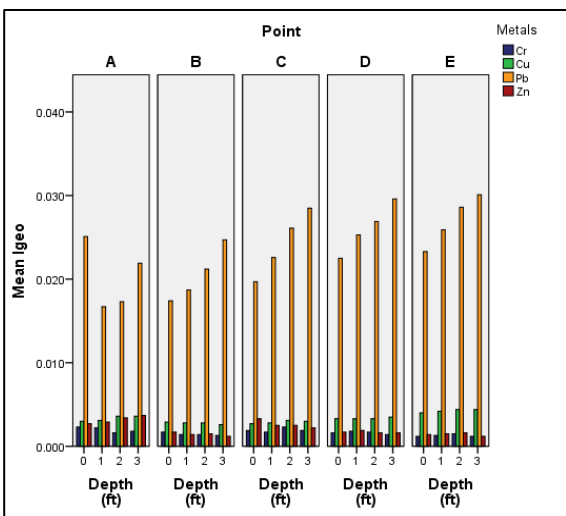


Figure 2. Enrichment factor in point A, B, C, D and E with different depth. *Geo-accumulation Index (Igeo), Contamination Factor (CF) and Pollution Load Index (PLI)*

Table 5 shows the mean of Igeo, CF and PLI for the Cr, Pb, Zn and Cu in point A,B,C,D and E. Based on the terminologies in Table 1, all of the point reveals the similar assessment shows that the sediment in the class 1 which in the unpolluted-moderately polluted conditions since the Igeo mean in the range of 0-1. The CF mean in all of the point shows the value <1 so, all of the metal in the point were in low contamination factor. Meanwhile, the mean of PLI in all of the sediment point were also <1 so the sediment in Kuala Perlis considered as unpolluted sediment condition. In addition, Figure 3 and 4 shows the pattern of Igeo and CF with different depth in point A, B, C, D and E respectively. As for point A, The Igeo and CF of Pb decrease in 1ft and increase back up to 3ft while point B, C, D and E increase in depth. CF value of Cu and Zn in point A increase with depth while Cr increase in depth as well as the Cr, Cu and Zn value in point B,C,D and E. Figure 5 shows the PLI value of the sediment in Kuala Perlis. Although the sediment considered as unpolluted but the pollutant still exists the sediment which gives the point A has the highest PLI value. The heavy anthropogenic activities such as shipping and ferries operates daily at the Jetty Port contribute to the existence of pollutant in that area[21]. Besides, the municipal waste carried by the waterways from the Perlis River estuary is one of the factors contribute to the sediment pollution.

Table 5 Geo-accumulation index, contamination factor and pollution load index in sediment phase

Metal conc (ppm)	Geo-accumulation Index (Igeo)				Contamination Factor (CF)				Pollution Load Index (PLI) Mean	
	Mean				Mean					
Point/Depth (ft)	Cr	Pb	Zn	Cu	Cr	Pb	Zn	Cu		
A	0ft	0.0023	0.0251	0.0027	0.0030	0.011	0.125	0.013	0.015	0.022
	1ft	0.0022	0.0167	0.0029	0.0031	0.011	0.083	0.014	0.016	0.021
	2ft	0.0016	0.0173	0.0034	0.0036	0.008	0.086	0.017	0.018	0.020
	3ft	0.0018	0.0219	0.0037	0.0036	0.009	0.109	0.019	0.018	0.022
B	0ft	0.0017	0.0174	0.0017	0.0029	0.009	0.087	0.008	0.014	0.016
	1ft	0.0014	0.0187	0.0014	0.0028	0.007	0.093	0.007	0.014	0.014
	2ft	0.0014	0.0212	0.0015	0.0028	0.007	0.106	0.007	0.014	0.015
	3ft	0.0013	0.0247	0.0012	0.0026	0.007	0.123	0.006	0.013	0.013
C	0ft	0.0019	0.0197	0.0033	0.0027	0.009	0.098	0.016	0.014	0.019
	1ft	0.0017	0.0226	0.0025	0.0028	0.009	0.112	0.012	0.014	0.017
	2ft	0.0023	0.0261	0.0025	0.0031	0.012	0.130	0.012	0.015	0.020
	3ft	0.0019	0.0285	0.0022	0.0030	0.010	0.142	0.011	0.015	0.019
D	0ft	0.0016	0.0225	0.0017	0.0033	0.008	0.112	0.009	0.017	0.017
	1ft	0.0018	0.0253	0.0019	0.0033	0.009	0.126	0.009	0.016	0.018
	2ft	0.0017	0.0269	0.0016	0.0033	0.009	0.134	0.008	0.016	0.017
	3ft	0.0014	0.0296	0.0016	0.0035	0.007	0.148	0.008	0.017	0.016
E	0ft	0.0012	0.0233	0.0014	0.0040	0.006	0.116	0.007	0.020	0.015
	1ft	0.0013	0.0259	0.0015	0.0042	0.007	0.129	0.007	0.021	0.016
	2ft	0.0015	0.0286	0.0016	0.0044	0.008	0.142	0.008	0.022	0.018
	3ft	0.0012	0.0301	0.0012	0.0044	0.006	0.150	0.006	0.022	0.014



4. CONCLUSION

The heavy metal pollution assessment in sediment is important to give the basic information regarding the status of marine pollution in Kuala Perlis. Besides, this study also provides useful statistical methods such as descriptive analysis, boxplot, and correlation analysis and contamination indices for the processing and analysing the heavy metals sediment contamination in Kuala Perlis. This study reveals that Kuala Perlis were currently polluted by four types of heavy metals Cr, Pb, Cu and Zn which indicates Pb is the major contribution to the sediment pollution. These metals were highly negative and positive correlated with the sediment characteristics. As the

overall assessment, PLI value stated that Kuala Perlis sediment was currently in unpolluted condition.

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