

EVALUATION OF HEAVY METAL CONTAMINATION LEVELS OF BALOK RIVER SEDIMENTS IN PAHANG, MALAYSIA BASED ON GEOACCUMULATION INDEX AND SUPPORTED WITH ENRICHMENT FACTOR

(Penilaian Paras Pencemaran Logam Berat Terhadap Sedimen Sungai Balok di Pahang,
Malaysia Berdasarkan Kepada Indeks Geo-akumulasi dan Di Sokong oleh Faktor Pengkayaan)

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Abstract

This study was conducted in order to assess the level of metal pollution in river sediment samples, which indirectly representing the general quality of the Balok River. The samples of river sediment have been collected at nine sampling stations from the particular river. The total content of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn were measured using inductively coupled plasma optical emission spectrometer (ICP-OES) after an acid extraction. The mean concentrations (mg/kg) were found at 0.49, 16.16, 42.93, 24.31, 2734.69, 155.61, 16.04, 29.31, 50.71 and 159.40 for the respective metals. The assessment of the river quality was done based on the information showed by the enrichment factor (EF) and geoaccumulation index (I_{geo}). Analysis of the I_{geo} indexes clearly indicate that most of the sediment samples analyzed in this study contained the selected heavy metals in the levels of unpolluted to moderately pollute. Based on the low EF values recorded in this study, it can be concluded that the Balok River sediments is not significantly contaminated with the studied heavy metals.

Keywords: sediment, acid extraction, enrichment factor (EF), Geo-Accumulation Index (I_{geo}), heavy metals

Abstrak

Kajian ini dijalankan untuk menilai tahap pencemaran logam di dalam sampel sedimen sungai, yang secara tak langsung secara umum akan menggambarkan kualiti Sungai Balok. Sampel-sampel sedimen sungai telah diambil di sembilan tapak persampelan daripada sungai tersebut. Jumlah kandungan Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V dan Zn telah diukur menggunakan plasma gandingan teraruh – spectrometer emisi optik (ICP-OES). Nilai kepekatan purata (mg/kg) bagi setiap logam yang dikaji adalah 0.49, 16.16, 42.93, 24.31, 2734.69, 155.61, 16.04, 29.31, 50.71 dan 159.40 bagi masing-masing logam. Penilaian kualiti sungai telah dilakukan berdasarkan kepada maklumat yang diberikan oleh faktor pengkayaan (EF) dan indeks geo-pengumpulan (I_{geo}). Analisis terhadap indeks I_{geo} jelas menunjukkan bahawa kebanyakan sampel sedimen yang dianalisis mengandungi kandungan logam berat pada paras tidak tercemar sehingga sederhana tercemar. Berdasarkan kepada nilai-nilai EF yang rendah direkodkan dalam kajian ini, maka boleh disimpulkan bahawa sedimen Sungai Balok tidak dicemari secara ketara dengan logam dikaji.

Kata kunci : sedimen, pencernaan berasid, faktor pengkayaan (EF), Indeks Geo-akumulasi (I_{geo}), logam berat

Introduction

River sediments are a major carrier of heavy metals in an aquatic environment. Sediments are mixture of several components of mineral species and represent the ultimate sink of heavy metals discharged into environment [1]. Chemical leaching of bedrocks, water drainage basins and runoff from river banks are the primary sources of heavy metals in river sediment [2]. Disposal of industrial wastes and applications of biocides for pest are the other anthropogenic sources of metals for river sediment [3]. Heavy metals are categorized as a serious pollutant for an aquatic life because of their toxicity and persistence in the environment [4]. Some of the metal pollutants have cancer-causing properties and poisonous than the latter can cause environmental and health problems to human and animals [5]. In general, the metal pollutants considered as non-biodegradable materials and usually it is difficult to remove these pollutants from the water system [6]. When enter the environment, metals will distributed between the aqueous phase and the suspended sediments during their transport. Hence, water sediment is regarded as the potential reservoir for metals and plays an important role in adsorption of dissolved heavy metals [7]. Polluted sediments also can act as the important sources of heavy metals, imparting them into the water and reduced water quality [8]. Metal concentration in river sediment can be regarded as a good indicator of the river contamination. A Gebeng industrial area is considered as one of the most developed industrial sites in Kuantan, Pahang. Since 1970s the area has increased its industrial activities. The area is actively involved in various types of activities including petrochemical, multifarious industries and the latest rare-earth processing plant. The wastes produced from this industrial area are believed indirectly discharged into the adjacent river. Balok River is one of the several rivers in Kuantan that adjacent to the Gebeng industrial estate. Balok River originates as Batang Panjang River from the hill to the northwest of Gebeng Industrial area and generally serves the western catchment of this industrial area. The industrial site is within the Balok River catchment and thus all discharges from the industrial site will enter this river system. Balok River flows along the western boundary of Gebeng industrial area to the south before its confluence into the coastal waters of the South China Sea. The primary source of pollution into this river is the drainage discharge from the Gebeng Industrial area along the river channel. This study was conducted in order to measure the concentration levels of the selected heavy metals in Balok River sediment and to evaluate the pollution status of the river sediment based on geoaccumulation index and enrichment factor.

Materials and Methods

Sampling Location and Sample Collection

The sediment samples were collected at nine sampling stations along the Balok River which located close to the commercial beach area in Kuantan, eastern part of peninsular Malaysia. Figure 1 shows the nine selected sampling stations along the Balok River. The river flows pass through the wide industrial facilities which include steel industries, palm oil mills and pipe coating and others. The Global positioning System (GPS) was utilized to determine the actual coordinate of the sampling stations and to reconfirm the location of stations during the subsequent sampling periods. The Ponar type grad sampler was used in this study to collect river sediment at about 20 cm deep core. The sediment samples were then kept in a cleaned polyvinyl plastic container to avoid any contamination and transported to the laboratory for further treatment and analysis. The sediment samples were taken along the river within a 5 km long where the distance between sampling sites is 500 m. The sediment sample has also been collected from the remote area of Janda Baik in Bentong as a control. The control sample has been treated and analyzed in the same manner as the real sediment samples.

Samples Treatment and Analysis

The sediment samples were oven-dried at 100°C, ground with a mortar and sieved. All the laboratory apparatuses were soaked in 10% HNO₃ (v/v) overnight and rinsed with de-ionized water before being used. A 0.5g sample of the sieved sediment was digested with HNO₃, HCl and H₂O₂ in the sealed PTFE vessels using the Milestone Ethos 1 microwave oven. Ten heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn) were quantified by inductively coupled plasma optical emission spectrometer (Perkin Elmer Optima 5300DV ICP-OES). All the sediment samples were analyzed in triplicate including the blank sample to reduce the uncertainty reading of the concentration measurement. The sample of the certified reference material (European Reference Material ERM[®]-CC141) was analyzed to confirm the accuracy of the analytical method used in this study. The percent recovery obtained for all studied metals were as follows: Cd (80%), Cu (104.80%), Mn (100.80%), Ni (94.90%), Pb (118.01%) and Zn (121.60%).

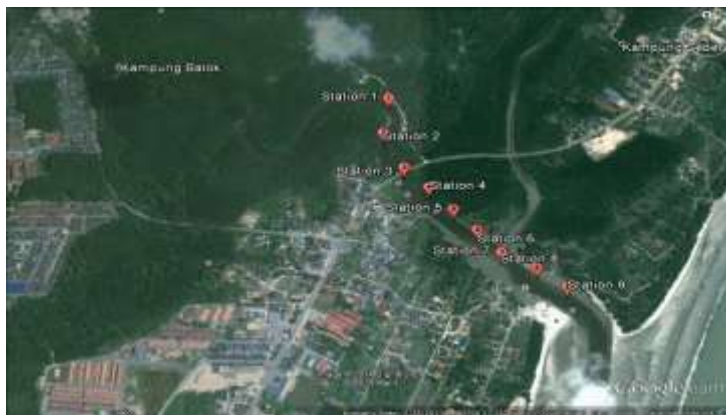


Figure 1. The study area showing all the nine sampling locations along the Balok River

Results and Discussion

Heavy Metal Concentrations

Table 1 represents the total concentrations of the selected metals measured in the studied sediments. Based on the mean values of the metal concentrations, the metals follows in the following order, from the highest to the lowest; $\text{Fe} > \text{Zn} > \text{Mn} > \text{V} > \text{Cr} > \text{Pb} > \text{Cu} > \text{Ni} > \text{Co} > \text{Cd}$. The data clearly indicate that Fe and Zn are the two elements that dominated in the surface sediment samples of the Balok River. The existence of these two metals in river sediment could be related to the natural origin [9]. Anthropogenic related elements, V, Cr, Pb and Ni showed a lower concentration than the lithogenic related elements, Fe and Zn. Meanwhile, Co and Cd are the two metals that were observed in the lowest mean concentration.

In general, the metal contents of the sediment samples were significantly higher compared to the background concentration. This indirectly indicates the influence of the contribution of anthropogenic activities to the elevation of the metal contents to this particular river. The concentrations of all elements were measured in low percent of standard deviation except for Co and Pb which were measured with 78.7 and 54.5 % deviation respectively. The high value of standard deviation would indicate a strong skewness of the concentration of these two elements among the nine sampling stations. The reason for the strong skewness could be explained by a high concentration of the two metals that was recorded at St9 with 48.4 and 70.8 mg/kg for Co and Pb respectively.

Inter-element Relationships

Inter-element relationships data are useful to obtain the interesting information on element sources and its pathways [10]. The inter-relationship of the metals obtained in this study is shown in Table 2. The inter-relationship data clearly indicate that Fe was significantly correlated with the metals of Zn, Co, Pb and Cu. Fe is more readily deposited element and its silicate, carbonate and hydroxide are less soluble compared to the other metals [11]. Fe is considered as among the major components of the earth's crust and accumulation in sediment is the result of long-term exposure. This strongly suggests that the existences of the five elements in the river sediment were controlled by the "natural factor" of the lithogenic process during weathering progress of natural parent materials such as rocks and soils.

Table 1. Concentration of heavy metals measured at nine sampling stations throughout the river

Sampling Point	Element (mg/kg dry weight)									
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
St1	0.8	6.0	33.8	10.6	32244	72.0	12.8	18.0	40.4	67.0
St2	0.4	8.4	46.4	26.4	41940	102.0	15.4	18.4	45.8	139.8
St3	0.4	12.8	59.2	27.2	53736	184.4	22.2	31.2	70.2	197.2
St4	0.4	11.8	54.8	28.4	50424	115.8	18.6	22.4	56.2	186.8
St5	0.4	13.4	44.8	24.2	47964	144.0	16.6	23.8	54.0	176.0
St6	0.6	13.4	49.8	23.2	53400	185.8	18.2	26.8	64.4	184.8
St7	0.4	20.4	45.6	26.2	38448	243.4	18.0	34.2	64.0	172.2
St8	0.4	10.8	28.6	11.0	37992	184.2	12.4	18.2	36.8	140.2
St9	0.6	48.4	23.4	41.6	67104	171.6	10.2	70.8	24.6	170.6
Mean	0.5	16.2	42.9	24.3	47028	155.9	16.0	29.3	50.7	159.4
% STD	29.7	78.7	27.9	38.4	23	33.7	33.5	54.5	36.2	34.1
Control	0.2	4.8	14.0	9.8	29388	57.0	6.2	5.0	38.8	36.0

Table 2. Pearson's correlation matrix for the heavy metal concentration in Black River sediments

	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
Cd	1.000									
Co	0.124	1.000								
Cr	-0.445	-0.498	1.000							
Cu	-0.207	0.774	0.088	1.000						
Fe	-0.051	0.733	0.026	0.844	1.000					
Mn	-0.400	0.379	0.054	0.251	0.237	1.000				
Ni	-0.469	-0.426	0.966	0.072	0.045	0.264	1.000			
Pb	0.170	0.985	-0.410	0.796	0.768	0.376	-0.329	1.000		
V	-0.380	-0.463	0.911	-0.042	-0.075	0.349	0.965	-0.376	1.000	
Zn	-0.640	0.308	0.528	0.635	0.674	0.603	0.596	0.327	0.521	1.000

Meanwhile, the low inter-relationships of Fe with the metals of Cd, Cr, V, Pb and Ni indirectly suggest that the existence of Cd, Cr, V, Pb and Ni in the sediment samples is dominated by the anthropogenic factors. The high correlations were observed for the pairs of Pb/Co, Ni/Cr, V/Cr and Ni/V for their existence in the sediment samples. This would strongly suggest that all of the metals were possibly originated from the same sources. On the other hand, Cd did not show a strong correlation with all of the others metals as well as Mn, indicating that their existences in the sediment samples were not contributed by the same contributor.

Principal component analysis (PCA)

Multivariate statistical analysis was applied to identify the possible pollution sources, as well as to reveal the elemental composition of such possible sources. In this study, the calculation of PCA was carried out based on the concentration data of the 10 metals in sediment samples. Principal component analysis, PCA was applied to quantitatively evaluate the clustering behavior of the metal concentration data. This particular statistical analysis was used in order to draw out a small number of latent factors for analyzing the relationship between the heavy metals, hence, reducing the original data [12]. The outcomes of the PCA statistical analysis are used to characterize

the possible different pollutant sources that may contribute some amount of heavy metals to the river. Table 3 shows the summarized information obtained after the data of the metal concentration were treated with the PCA analysis.

The treatment of PCA analysis has successfully extracted two component factors, PC1 and PC2 from the 10 variables. The PCA analysis was done based on the criterion of an eigenvalue of one. The cumulative variance of the two factors explained 80.54% of the examined data. The factor 1 (PC1), extracted 40.85% of the total variance of the studied data set has a large factor loading for metals, Pb, Co, Cu, and Fe in the group. All metals in PC1 showed almost equally high factor loading except for Mn. The average concentrations of these elements all exceeded their background values. These metals, especially Cu and Mn are usually related to the excess application of fungicides or pesticides [13,14]. But with the present of Pb and Fe in the same PC group, the existence of metals in the sediment could be related to the anthropogenic origin. Based on the surrounding environment of the study area, it is possible to assume that the existences of Pb and Fe in sediment were strongly associated to be “agrochemical” inputs and the local fisheries activities.

Table 3. actor loading of the selected metals in sediment

Elements	Component		Communalities
	Factor 1, PC1	Factor 2, PC2	
Pb	0.922	-0.314	0.948
Co	0.915	-0.375	0.978
Cu	0.905	0.124	0.834
Fe	0.891	-0.149	0.800
Mn	0.499	0.379	0.393
Ni	0.126	0.965	0.937
V	-0.153	0.932	0.892
Cr	-0.150	0.928	0.884
Zn	0.652	0.715	0.936
Cd	-0.149	-0.654	0.450
Eigenvalue	4.085	3.969	
% of variance explained	40.85	39.69	
% cumulative	40.85	80.54	

Factor two (PC2), which accounts 39.69 % of the total variance, has a high positive loading for Ni, V, Cr, and Zn. Meanwhile, Cd has a negative loading in the group. Ni, V and Cr which recorded nearly the equal factor loading in the group with 0.965, 0.932 and 0.928 respectively, strongly suggest the influenced of the anthropogenic factors. The elements of Ni and V normally associated with the power plants and any combustion-related activities. Some of the elements were also released from the plant facilities in the liquid form and flow out to the adjoining water system with factory waste water. The high concentration of these elements within the study area could be reflected in the fishing activities and the occurrence of the oil spill from the nearby boat jetty.

Enrichment Factor

The total concentrations of the studied pollutants do not represent the real level of pollution emanating from natural or anthropogenic sources [15]. The application of the statistical analysis, Enrichment Factor (EF) could provide a rough idea of the anthropogenic impact on river sediment pollution [16]. The EF calculation enables to reduce the metal variability associated with variations in mud/sand ratios. The EF statistical analysis normalizes the measured heavy metals concentration with respect to a sample reference metal such as Fe and Al [17]. In this study, iron (Fe) has been selected as the element of reference in the calculation the enrichment factor index. The assumption was

made that the association or influence of anthropogenic sources to the atmosphere is insignificant or unimportant. The EF value was calculated as equation 1 below[18]:

$$EF = [M_{\text{sample}} / Fe_{\text{sample}}] / [M_{\text{baseline}} / Fe_{\text{baseline}}] \quad (1)$$

M_{sample} indicates the concentration of heavy metal in the sediment sample. Fe_{sample} is the concentration of the reference metal in the sample, M_{baseline} is the concentration of heavy metal in a control sample and Fe_{baseline} is the concentration of the reference metal in a control sample. In this study, the control sample was collected in the remote area which considerably out of any possible industrial contaminants influences.

Table 4. Enrichment factor value of sediment sample in Balok River

Element	Sampling Station								
	St1	St2	St3	St4	St5	St6	St7	St8	St9
Cd	4	1	1	1	1	2	2	2	1
Co	1	1	2	1	2	2	3	2	4
Cr	2	2	2	2	2	2	3	2	1
Cu	1	2	2	2	2	1	2	1	2
Mn	1	1	1	1	2	2	3	3	1
Ni	2	2	2	2	2	2	2	2	1
Pb	3	3	3	3	3	3	5	3	6
V	1	1	1	1	1	1	1	1	0
Zn	2	3	3	3	3	3	4	3	2

An indication of the EF value; < 2: Deficiency to minimal enrichment, 2-5: Moderate enrichment, 5-20: significant enrichment, 20-40: very high enrichment and > 40: Extremely high enrichment.

Table 4 shows the EF values of the analyzed elements obtained in this study. The EF values indicate that most of the metal contents in Balok river sediments are not so different in their concentrations compared to the average soil content. The average of the EF's values for most of the studied metals were observed below than 3, which indicates the occurrences of minimal to moderate enrichment of the metals in the sediment samples. As shown in Table 4, the EF values of Pb and Zn for almost all of the sampling stations were higher than 2 except for St1 and St9 for the Zn metal. For Pb, the significant enrichment was observed in two sampling stations, St7 and St9 with the EF values recorded as 5 and 7 respectively. This would suggest that a substantial portion of Pb and Zn concentration in sediment samples were contributed by anthropogenic sources, primarily related to the fisheries and boating activities by the local residents. St7 located at the meeting point of Balok River and Tunggak River while St9 located next to the Balok beach side which is a famous beach resort in the state. Based on its geographical position, the increases of metals concentration in river sediments in this study can be linked with various tourism and hospitality industry, which grew rapidly in the surrounding area.

Geo-Accumulation Index

In order to assess the contamination of the examined sediment with organic and inorganic substances, the Geo-accumulation index is utilized by comparing the control level with the current concentration of the metals. The geo-accumulation index (I_{geo}) in this study was calculated by using the following equation 2 [19].

$$I_{\text{geo}} = \log_2 [C_n / (1.5 (B_n))] \quad (2)$$

The C_n representing the concentration of the metal n in the studied sediment while the B_n representing the background value of the metal n .

Table 5: The classification of the sample sediment quality based on the I_{geo} indexes

Element	Sampling Station								
	1	2	3	4	5	6	7	8	9
Cd	2	1	1	1	1	1	1	1	1
Co	0	1	1	1	1	1	2	1	3
Cr	1	2	2	2	2	2	2	1	1
Cu	0	1	1	1	1	1	1	0	2
Fe	0	0	0	1	1	1	0	0	1
Mn	0	1	2	1	1	2	2	2	2
Ni	1	1	2	1	1	1	1	1	1
Pb	2	2	3	2	2	2	3	2	4
V	0	0	1	0	0	1	1	0	0
Zn	1	2	2	2	2	2	2	2	0

Class 0: Unpolluted, 1: From unpolluted to moderately polluted, 2: Moderately polluted, 3: From moderately polluted to strongly polluted, 4: Strongly polluted, 5: From strongly polluted to extremely polluted, 6: Extremely polluted.

The classification of the sample sediments can be made based on the geo-accumulation indexes where it can be explained by seven different indications that covered unpolluted to extremely polluted levels. Table 5 presents the I_{geo} values of the studied metals. In general, the I_{geo} values for ten studied metals at all sampling stations were below 1 which clearly indicates that the studied sediment samples were not polluted with the studied metals. Most of the studied sediment was equally polluted with Pb, Cr and Zn at the moderate class by having the I_{geo} value equal to 2 and above. It is suggested that nearly all the analyzed sediment samples were not being contaminated with the studied metals except with Pb, Cr and Zn.

Conclusion

The average concentrations of the studied metals in the Balok River sediments are significantly higher compared to the background levels. The highest concentration of the element was recorded for Fe which ranging from 3224 to 67104 mg/kg. The concentration of metals in sediment samples can be arranged in descending order as Fe > Zn > Mn > V > Cr > Pb > Cu > Ni > Co > Cd. The results of PCA generally indicate that the studied metals contained in the sediment samples were originated from two different sources, anthropogenic and natural origin. The group of Fe, Mn, Co, Pb and Cu are possibly related to the natural and agricultural origin, while the group of Ni, V, Cd, Cr and Zn were contributed by industrial activities. The information obtained from the analyses of Enrichment factor, EF and supported by the Geo-Accumulation Index, I_{geo} clearly indicates that no extremely excessive metals were discharged into the Balok River by the adjacent industrial activities.

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