

Contamination and Risk Assessment of Heavy Metals from Sadong River, Malaysia

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ABSTRACT

Forty-two surface sediments at different locations around Sadong River, Sarawak, Malaysia were analysed using acid digestion and atomic absorption spectroscopy (AAS). The spatial distributions of heavy metals in surface sediments were determined. An assessment of the possible sources of these heavy metals was inferred. The surface sediments contain variable concentration of Zn (19.29 ± 2.09 to 46.61 ± 1.17 mg/kg), Pb (4.61 ± 0.42 to 69.73 ± 14.80 mg/kg), Cu (3.45 ± 0.28 to 12.30 ± 0.27 mg/kg), Mn (219.59 ± 17.04 to 464.75 ± 17.82 mg/kg) and Fe (1801.85 ± 65.13 to 3447.62 ± 136.63 mg/kg). The relationship between lithogenic and anthropogenic variables were deduced using statistical analysis. The correlation coefficients Zn with Mn ($r = 0.7922$); Fe with Ni (0.6985); Zn with Cu ($r = 0.6328$) and Mn with Cu ($r = 0.6079$) were significantly correlated. The contamination and risk assessment was based on Enrichment factor which showed minor to moderate enrichment of selected heavy metals.

Article History

Received: 19 February 2021
Received in revised form: 30 April 2021
Accepted: 15 May 2021
Published Online: 2 September 2021

Keywords:

Acid digestion, heavy metals, lithogenic, Sadong River, statistical analysis.

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DOI: 10.11113/ajeas.v4.n1.134

1.1 Introduction

The one-way analysis of variance (ANOVA) was used to determine the statistically significant differences between the mean concentration values of heavy metals in surface sediments. Statistical analyses such as correlation analysis, principal component analysis (PCA) and cluster analysis (CA) were determined. The heavy metals data of surface sediments were subjected to statistical analysis to assess the sources of these heavy metals. A number of diverse reference materials such as Fe, Al, Mn and others as well as enrichment calculation methods have been used by several researchers to assess the impact of heavy metal pollution on river sediments (Muller, 1969; Hakanson, 1980; Salomons & Forstner,

1984). The initial crucial step in assessing the impact of sediment pollution and the level of contamination in a given area is to establish a reference background or baseline sample of known metal composition. The main objective was to identify the sources of heavy metals in the river sediments and to understand the geochemical relationships between the heavy metals and major metals.

The Sadong River is one of the major rivers in Sarawak located at the Kota Samarahan District, Sarawak, Malaysia and its length is approximately 40 km (Bryant, 2003; Omorinoye *et al.*, 2019). The composition of sediments in the rivers can be influenced by the deposition of terrestrial materials due to erosion caused by the frequent

rainfall in the study area. River sediments are polluted by heavy metals due to human activities such as transportation (ships and boats), agricultural activities (fishing, oil palm plantation and rice plantation) and sewage discharge into the river. The main pathways of heavy metals partitioning include adsorption, complexation, precipitation and biological uptake (Sany *et al.*, 2013).

Chemical properties of a river are influenced by dissolution of materials in the river, the heavy metals of sediments are related to factors such as bedrock geology, drift cover, land use, climate and topography (Sundararajan and Natesan, 2010; Praveena *et al.*, 2008). Weathering processes can cause the dissolution of particles which are transported as suspended loads in most of the world's major rivers. The products of weathering were carried via the river to the ocean as suspended load as well as bed load representing the coarser fraction (Omorinoye *et al.*, 2019). The organic matter combines with heavy metals to form stable organo-metallic complexes which are very stable and subsequently released into river sediments through several processes such as ion exchange, adsorption, complexation and mineral precipitation (Sundararajan and Srinivasalu, 2010).

Heavy metals originate from both natural processes and anthropogenic activities (Xin *et al.*, 2014). Heavy metals that occur naturally and from industrial activities can increase the concentration of heavy metals in natural rivers. The natural sources are wind-blown dusts, eroded soils and sediments which put huge amounts of solid material into the river (Scott,

1985). Geological sources and living things play an important role in the exchange of metals and ions within the atmosphere and hydrosphere.

River sediments have large amount of fine grained particles such as silt and clay as well as high alkalinity values. These unique features of river sediments directly influence the mobility of heavy metals. The major metals are typically retained and stored in the sediments and are usually retained in the sediments. Sediments serve as traps and can be used to determine the accumulation history of suspended particles and particles bound to metals in different river systems (Tatone *et al.*, 2016). The aim of this study is to describe the statistical analysis of the geochemical data of surface sediments and infer the sources of the heavy metals.

2.1 METHODS

2.2.1 Study area and Sample Collection

Forty-two (42) surface sediment samples were collected between 2016 and 2018 from seven sampling sites (Fig. 1) within the Sadong River, Sarawak, Malaysia. The Sadong River Basin is part of the Borneo Basin of southeast Asia, it is surrounded by plate boundaries, arc systems and marginal ocean basins (Fuller *et al.*, 1999; Nagarajan *et al.*, 2014). The Sadong River Basin is dominated by Cretaceous Eocene age sediments and the sedimentary deposits comprise siliciclastic rocks such as sandstone, shale and siltstone (Hutchinson, 2005; Nagarajan *et al.*, 2014) as shown in Fig.1.

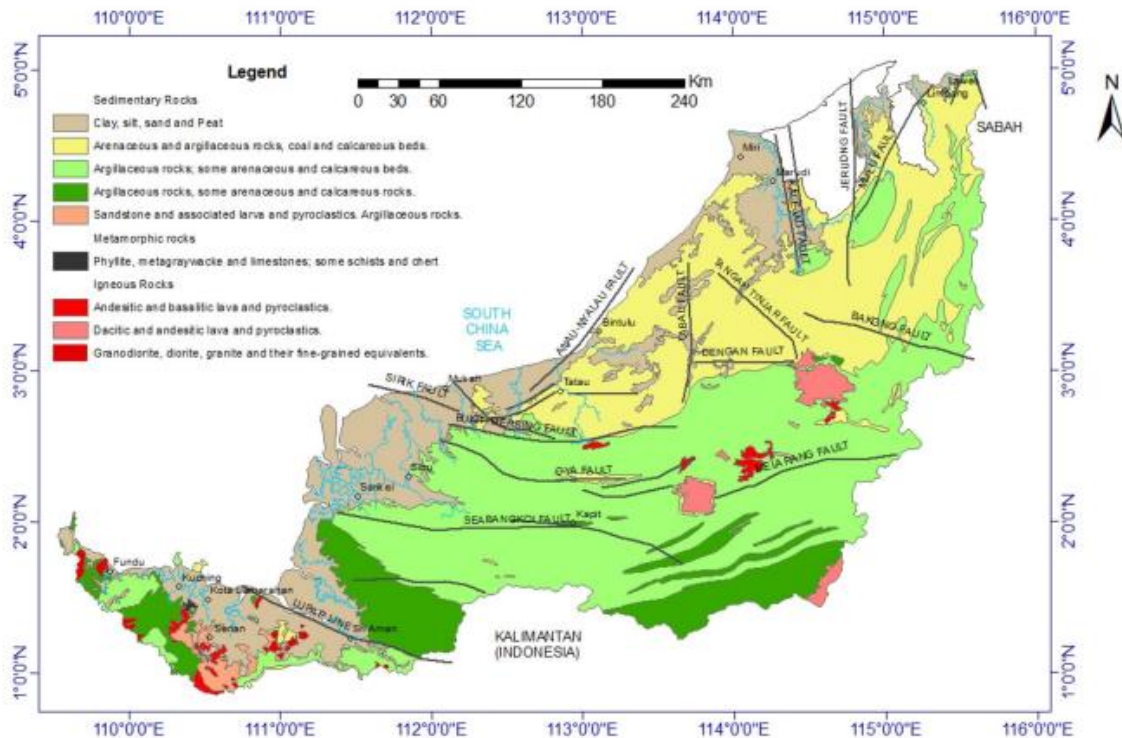


Figure 1: Simplified geological map of the Sarawak Basin (Modified after Mineral and Geoscience Department of Malaysia 2012; Adepehin et al., 2019)

Forty-two representative surface sediment samples with three replicates were taken using a Wildco stainless steel grab sampler at different stations around Sadong River (Table 1). The

coordinates of sampling sites were recorded with Global Positioning System. Figure 2 shows the map of the study area.

Table 1: The sampling location at Sadong River, Sarawak, Malaysia

Stations	Geographical Coordinates	Locality	Major Activities
BS1	N01°33'03.2" E110°45'56.4"	Sebangan	Farming and Fishing
BS2	N01°30'51.0" E110°44'24.6"	Sadong Jaya	Fishing, Small Jetty
BS3	N01°27'19.4" E110°41'08.1"	Sg Buloh	Ferry Service, bridge
BS4	N01°14'49.3" E110°41'06.6"	Simunjan	Fishing and Small Jetty
BS5	N01°14'38.5" E110°40'42.5"	Gedong	Residential area (potential tourist attraction)
BS6	N01°14'42.1" E110°38'09.5"	Sebemban	Farming and Residential area
BS7	N01°09'36.3" E110°33'58.0"	Serian	Residential area

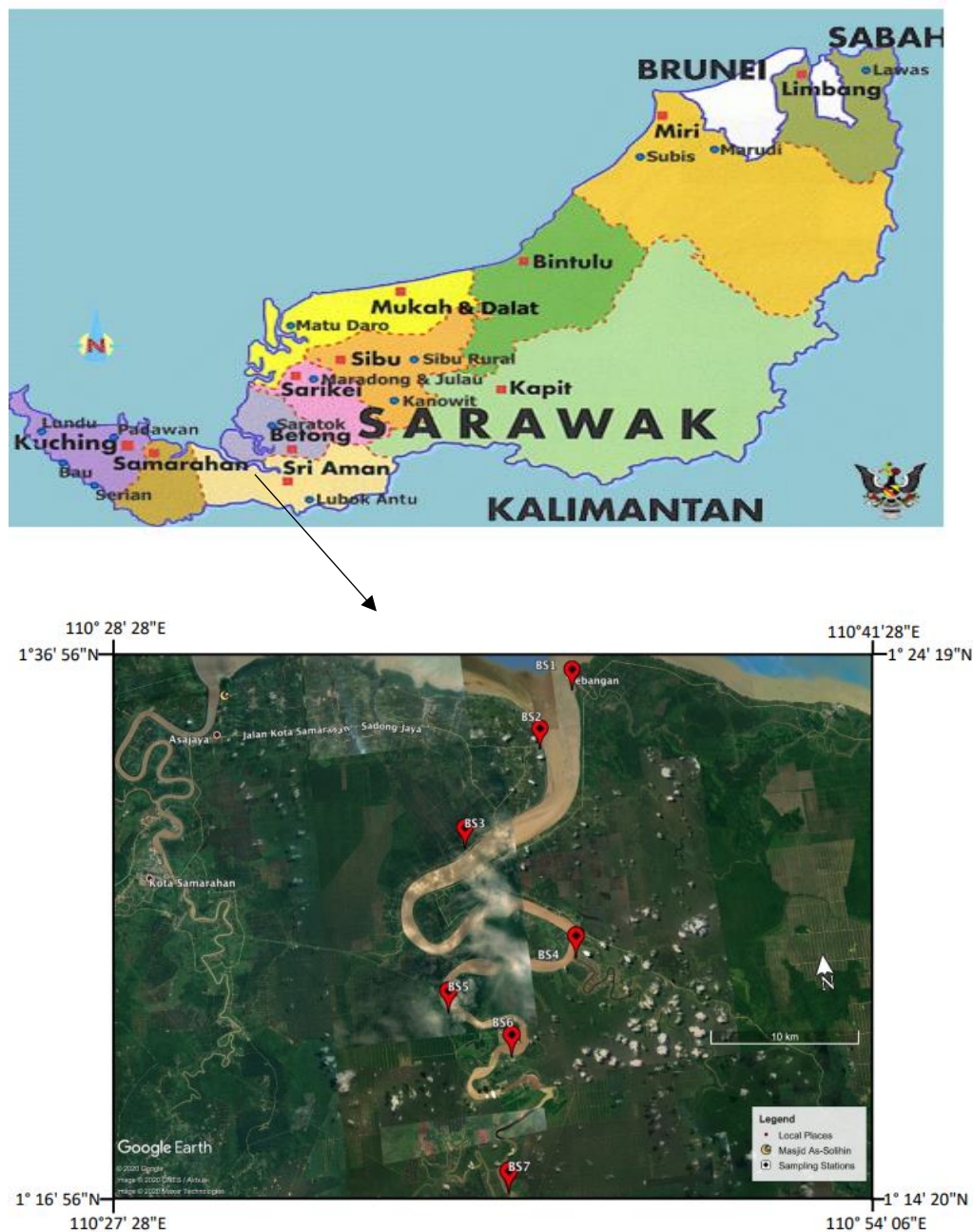


Figure 2: Map showing Sadong River

2.2.2 Sample Preparation and Analytical Technique

Surface samples were collected from the sampling sites at Sadong River and placed in plastic bags. The samples were stored at -40°C until further analysis. The sediments were air dried at room temperature for 72 hours and then pulverised using a mortar and pestle. Approximately 1 g of

each sample was digested with a solution of concentrated HNO_3 : HCl ; 3:1 (v/v). The sediment samples were filtered through $0.45\ \mu\text{m}$ Whattmann filter paper. The filtrate was transferred to a 100 ml volumetric flask and made up to the mark with deionized water. The solution was poured into plastic bottles for heavy

analysis using Atomic Absorption Spectrometry (AAS) laboratory in Faculty of Resource Science and Technology, Universiti Malaysia, Sarawak, Malaysia. All samples were digested in triplicates and subjected to AAS analysis. The metal contents in sediment are reported in mg/kg.

The sample solutions were analysed for heavy metal content using a Thermo Scientific iCE 3000 Atomic Absorption Spectrometry (AAS) Series and the calibration curve method. The heavy metals analyzed for include: Lead (Pb), Zinc (Zn), Iron (Fe), Manganese (Mn), Copper (Cu) and Nickel (Ni), and Cadmium (Cd). The major metals analyzed include Magnesium (Mg), Potassium (K) and Sodium (Na). All statistical analyses were carried out using the software IBM SPSS version 22. The EF method normalises the measured heavy metal content with respect to a sample reference metal such as Fe or Al, these two metals act as a representative for the clay

content (Ravichandran et al., 1995). One of the commonly used methods to distinguish the fraction of metals or enrichment is by normalization with respect to Fe, Mn and Al (Kemp et al., 1976; Loring, 1978; Van Metre & Callender, 1997). However, Fe or Al is widely used as a normalizer because it also compensates for variations in the grain size and composition. Al and Fe represents the quality of aluminosilicates which is the most significant agent for adsorbed metals in the aquatic environments. The variability of the normalized concentrations is expressed as EF, which is the ratio of the concentration of the metal in the sediment sample to the metal concentration corresponding to the pre-industrial period. EF is a useful tool for determining the degree of anthropogenic heavy metal pollution (Bessa *et al.*, 2020). The EF is calculated using the relationship as shown in Equation 1.1:

$$EF = (HM/RE)_{\text{sample}} / (HM/RE)_{\text{Background/upper crust}}$$

$$\text{Equation 1.1}$$

Where HM = heavy metal; RE – Reference element.

3.1 RESULTS AND DISCUSSION

Spatial distribution of the heavy metals and major metals were assessed to understand the variation of concentration along the Sadong River. The surface sediments were collected from the upstream to the downstream of the river. The river banks were visually homogenous and mostly composed of fine-grained sediments (Omorinoye et al., 2019). The surface samples taken at BS1 and BS2 were situated in the downstream of the Sadong River. While BS3, BS4 and BS5 were situated at the midstream of the River and BS7 were the upstream of the Sadong River. Levels of concentration of the heavy metals in surface sediments from all sampling sites were ranged: The surface sediments contain variable concentration of Zn (19.29 ± 2.09 to 46.61 ± 1.17 mg/kg), Pb (4.61 ± 0.42 to 69.73 ± 14.80 mg/kg), Cu (3.45 ± 0.28 to 12.30 ± 0.27 mg/kg),

Mn (219.59 ± 17.04 to 464.75 ± 17.82 mg/kg) and Fe (1801.85 ± 65.13 to 3447.62 ± 136.63 mg/kg).

The concentration of Fe was the highest in all the surface sediments followed by Mn, Zn and Pb while the concentrations of Ni and Cu varied in the surface sediments for the seven stations. Heavy metals in surface sediments from Sandong River include Zn, Pb, Mn, Fe, Ni and Cu. The adsorption of heavy metals ions and complexes in sediments occurred as a result of ion exchange, surface complexation and hydrophobic interaction (Natesan and Seshan, 2010). The fine grained particles play an important role in accumulation, adsorption/desorption and exchange processes of the heavy metal ions.

The concentrations of Fe in surface sediments ranged between 1801.85 ± 65.13 to 3447.62 ± 136.63 mg/kg. This was attributed that Fe is the most common metals found in nature and it is relatively abundant on the earth surface as shown in Figure 2. The concentration of Fe is influenced by the presence of dissolved

rocks and organic matter. The occurrence of Zn and Mn suggested that these heavy metals mainly originated from land based runoff and river discharges.

The surface sediments from BS1 as shown in Fig. 2 contained the highest concentration of Zn, Cu and Mn with concentration 46.61 ± 1.17 , 12.30 ± 0.27 and 464.75 ± 17.82 respectively. This sampling site is characterized by atmospheric deposition, movement of river water as well as tidal and seasonal currents in BS1, BS2 and BS3. This may have caused the variation in mobility, bioavailability and enrichment of heavy metals in surface sediments. Three sampling areas (BS1, BS2 and BS3) are characterized by high concentrations of heavy metals due to the decrease in mobility and dilution. Atmospheric deposition may be the main route for metals to accumulate in the sediments because a large amount of heavy metals can be transported for hundreds of miles from their place of origin. There are high vehicular movement around the Sadong Jaya area (BS2) which can emitted into the sediment and atmosphere. (Manaf *et al.*, 2009). Figure 2 show the distribution of heavy metals in the sediments from seven sampling sites of Sadong River. The BS2, BS3 and BS4 of Sadong River (Table 1) are characterized by boat docking, ferry service whereby some parts of the ship may have been corroded. Several factors such as physicochemical parameters control the rate of adsorption and desorption of these heavy metals (Sany *et al.*, 2013). They occur as particulates, colloids and dissolved phases which are easily absorbed to silica, clay and organic material.

The process of adsorption removes the heavy metal from the river water and stores them in the sediment. Other factors such as the alkalinity and acidity (pH) as well as solubility product of each metal are main parameters that control the concentration of these dissolved metals. The concentrations of Mg and K in surface sediments were recorded at exceptionally high in all sediments compared with Na with concentration ranged for Mg (380.24 ± 29.72 to 1857.16 ± 3.26)

and K ($319.01 \text{ mg/kg} \pm 70.10$ to $1197.35 \pm 32.54 \text{ mg/kg}$), respectively. The concentration of Na is low in the surface sediments of Gedong (BS5), Sebemban (BS6) and Serian (BS7). The concentration of Mg, Na and K detected in surface sediments of Sadong River is lower compared to concentration of Mg ($14,139 \pm 14,653 \text{ mg/kg}$), K ($3,224 \pm 1,426 \text{ mg/kg}$) and Na ($14,287 \pm 8,774 \text{ mg/kg}$) detected in surface sediments from Aliaga region, Turkey (Kara *et al.*, 2015). The concentration of Mg reported in surface sediments from Selangor River Malaysia was 808.46 mg/kg (Othman *et al.*, 2018) while the concentration of Mg and K ranged in Terengganu River in Malaysia between 202.0 to 2769.0 mg/kg and 63.36 to 1730.0 mg/kg respectively (Sukri *et al.*, 2018).

Correlation coefficient and inter elemental association were evaluated using Pearson correlation coefficient (r) and the results are presented in Table 5. The correlation coefficients Zn with Mn ($r = 0.7922$); Fe with Ni (0.6985); Zn with Cu ($r = 0.6328$) and Mn with Cu ($r = 0.6079$) were significantly correlated. Zn shows positive correlation with Mn and Cu which implied that these heavy metals may have originated from the same source. Iron showed a positive correlation with Ni while Mn shows positive correlation with Zn and Cu, which are abundant in the sediment of Sadong River and these heavy metals are usually associated with basement rocks which have been weathered and deposited in the river (Singh *et al.*, 2002). The relationship between Zn and Cu showed they may have originated from the same source, possibly from anthropogenic activities such as untreated waste, municipal effluents and industrial activities. There was no effect on tides and water currents at sites BS5 and BS6, therefore the absorption of Pb by the surface sediments over a long time may be taken into consideration. Copper, Nickel and Lead originated mostly from human activities such as discharge of municipal waste and sewage, burning of fuels as well as application of fertilizers and herbicides on farmland.

Table 2: Pearson correlation coefficient matrix for heavy metals in surface sediments from Sadong River, Samarahan, Sarawak

	Zn	Pb	Mn	Fe	Ni	Cu	As
Zn	1						
Pb	0.3805	1					
Mn	0.7922*	0.1934	1				
Fe	0.3605	0.1388	0.3907	1			
Ni	0.5651	0.1548	0.5755	0.6985	1		
Cu	0.6328	-0.1512	0.6079	-0.2623	0.3057	1	
As	0.943**	0.448	0.870*	0.245	0.424	0.0605	1

The PCA was carried out to predict the possible sources of heavy metals in surface sediments of Sadong River. The PCA result showed three factors were extracted from the PCA using Varimax with Kaiser Normalization rotation method. Principal Component Analysis was carried out based on the concentration of the metals in sediment samples. Principal Component Analysis was applied to quantitatively evaluate the clustering behaviour of the metals. Factor 1 is made up of Cu, Zn, Mn, K, Na and As which are derived from mixture of sources. Factor 2 consists of Ni, Fe and Mg which are believed to be derived from natural sources. These metals were originated from weathering of parent materials (Ke et al., 2017). Factor 3 is made up of Pb which is believed to be derived from anthropogenic sources (Ashraf et al., 2018). Figure 3 depicts the component plot in rotated

space for heavy metals in surface sediments from Sadong River. Factor 1 indicated the occurrence of Cu, Zn, Mn, K, Na and As are from similar pathways in the river system. However, apart from natural weathering of minerals and rocks, direct discharge of domestic waste may be their source (Patel et al., 2018). Lead is often derived from atmospheric deposition due to emission from vehicles (Liu et al., 2014). The presence of only Pb in Factor 3 may be attributed to atmospheric deposition via exhausts from automobiles because of the traffic around some localities around Sadong River. Metals such as Zn, Cd, Cu, and Ni can be principally mobilized through activities of human, while others, such as Fe, Mn, and may be derived from rock weathering (Serrano et al. 2011; Ekoa Bessa et al. 2018; Hossain et al. 2019; Tehna et al. 2019).

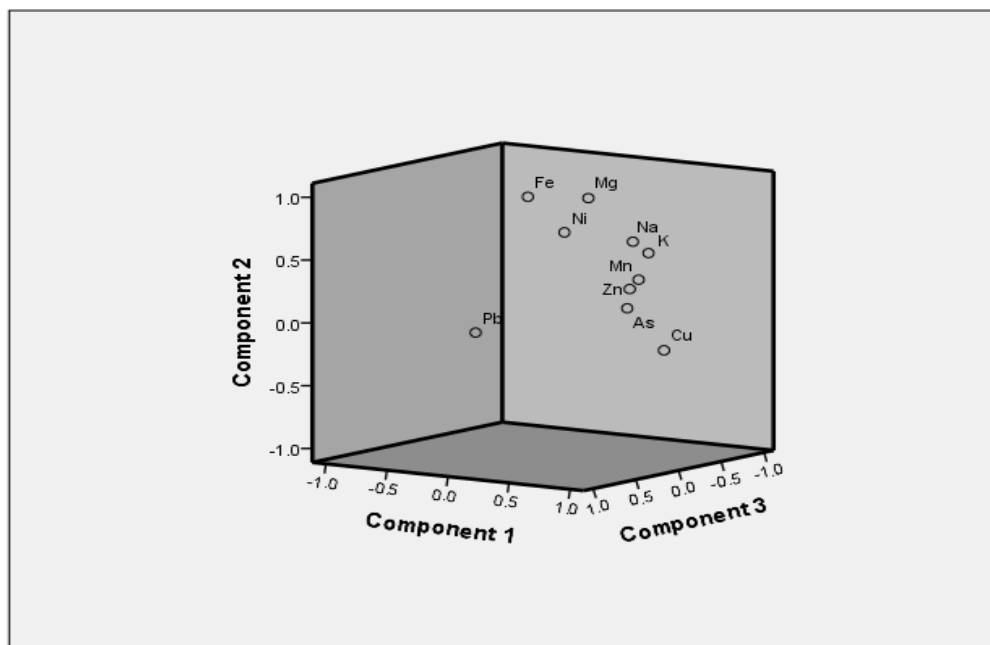


Figure 3: Component plot in rotated space of heavy metals and major metals in surface sediments from Sadong River, Samarahan, Sarawak

Figure 4 shows the EFs of selected heavy metals determined in surface sediment of Sadong River. Value of EF < 3 was observed for Zn in surface sediments of BS2, BS3 and BS5 indicated minor enrichment of Zn occurred. Moderate enrichment of Zn was observed in surface sediments of BS4, BS6 and BS7. Moderately severe enrichment of Zn was observed in surface sediments of BS1. Surface sediments from all the sampling sites of Sadong River have a moderately severe to severe enrichment of Pb in the sediments. An enrichment of Pb in surface sediment from Palk Strait in India has been reported by Kasilingam *et al.*, (2016). Significant enrichment of Pb was observed in surface sediment of BS7, whereas surface sediment of BS4 showed a moderate enrichment of Pb. The EFs of Cu showed minor enrichment in sediments of BS2, Sungai Buloh BS3, BS5 and Serian BS7, whereas a moderate enrichment of Cu was observed in surface sediments of BS4 and moderately severe enrichment of Cu was observed in surface sediments of BS1 and BS6.

These observations showed the river is unpolluted with all heavy metals analysed. The EF of Mn was highest in surface sediments of BS1, showing severe enrichment of Mn and surface sediments of BS3 had a moderate enrichment of Mn. Surface sediments from BS4, BS5, BS6 and BS7 have a moderately severe enrichment of Mn. There was minor enrichment of Fe in surface sediments for all sampling sites with EFs value of 1. The EFs of Ni in the sediments at all sampling sites were less than 1 except for surface sediment of BS2 which indicated a minor enrichment

4.1 CONCLUSION AND RECOMMENDATIONS

The statistical analysis showed that three factors were derived from PCA on distribution of the metals in surface sediments of Sadong River. The factor 1 comprised of Cu, Zn, Mn, K, Na and As which are derived from both natural sources and anthropogenic sources. Factor 2 contained Ni, Fe and Mg are originated from natural sources. Fe and Mg are mainly derived from weathering of siliciclastic rocks and soil in the study area. Factor 3 is made up of Pb which

was derived mainly from anthropogenic sources such agricultural activities and boat docking around the banks of the river. The impact of natural and anthropogenic activities resulted in

the occurrence of heavy metal in the sediments of Sadong River.

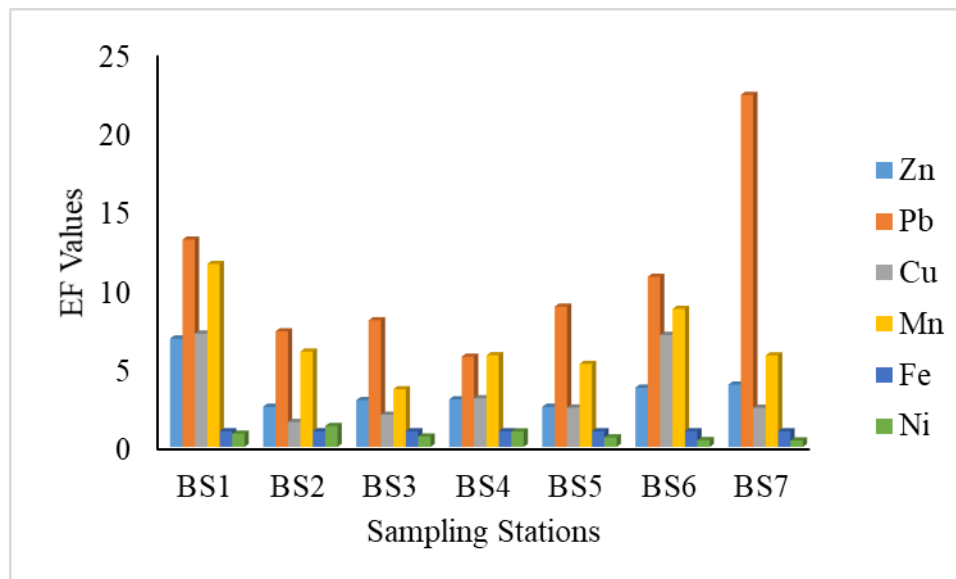


Figure 4: EFs of selected heavy metals in surface sediments from Sadong River, Samarahan, Sarawak

5.1 Conflict of Interest

The authors declare no conflict of interest in this study.

6.1 Acknowledgement

Authors would like to thank the Faculty of Resource Science and Technology, Universiti Malaysia Sarawak, Sarawak, Malaysia.

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