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Enrichment and Distribution of Elements Concentration in Sediment of Sungai Balok, Pahang

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ABSTRACT

Sungai Balok, Pahang is one of the rivers that has been impacted by anthropogenic pollution in the surrounding area that contributes to increase the level of river pollution. The purpose of the study was to estimate the pollution levels using sediment indices include pollution load index (PLI), Enrichment Factor (EF) and Index of Geo-accumulation (I_{geo}). The sediment core samples were taken from five sampling points along Sungai Balok from upstream to downstream towards coastal area. The sediment samples were slicedinto 2 cm aggregates from surface down to bottom of the sediment core. Then, the samples were oven dried, pulverized and digested using acid digestion prior to analyze using Inductively Coupled Plasma - Mass Spectrometry (ICP-MS). The average concentration of the elements in sediment core which are shows in the following order: As > Pb > Zn> Cr > Mn. It was found that the elements concentrations distribution varied between the vertical layer. The assessment of the degree of pollutant in sediment were done based on sedimentpollution indices.

1. Introduction

Gebeng is an industrial area in Pahang which was developed since 1970s [1]. Gebeng Industrial Estate (GIE) comprises of industrial, chemical and petrochemical zone. There are several rivers flow through GIE namely Sungai Balok, Sungai Tunggak and Sungai Ular. The most important river is Sungai Balok that flows through the north of the GIE into the final catchment, South China Sea. It was reported that the primary source of pollution of Sungai Balok is from the industrial activities along the river channel [2]. According to DOE, the water quality was reported as class III [3]. The industrial activities and their effluent will be discharged into the water bodies which in the end will cause serious pollution and finally will exert adverse effects and threat to human health [4-6]. The discharged of heavy metals from various industrial, agricultural and municipal sources will result in permanently contaminated water, polluted sediments and the accumulation of chemicals in the

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aquatic-food chain. The study was carried out to assess the pollution level in the Sungai Balok resulted from over the years of industrial activities in Gebeng Industrial Estate (GIE).

2. Literature Review

2.1 Gebeng, Pahang

Malaysia is rapidly growing with few numbers of industrial estates all over its different states. Gebeng is one of main industrial areas in Kuantan, Pahang. Since 1970s the area is established and increasing its industrialization [1]. Gebeng is popularly known with the existence of Gebeng Industrial Estate (GIE). GIE is comprises a world-class industrial, chemical and petrochemical industrial zone. The industrial development that occurs in 8,600 hectares of land is strategically located only 5 kilometers from the Kuantan Port. GIE is also composed of numbers of excellent infrastructure and facilities. Gebeng is also is rapidly expanding to become the leading chemical and petrochemical hub of the region [7]. There are few types of rivers located in the GIE namely Sungai Balok, Sungai Tunggak and Sungai Ular. The origin of Sungai Balok is from the Sungai Batang Panjang. From the Bukit Balok, the Sungai Balok will flow the hill to the northwest of the GIE which serves as its catchment as flows to South China Sea. Recently, the primary source of pollution of Sungai Balok is from the industrial activities along the river channel [2].

2.2 Heavy Metals

Heavy metals are known as the any metallic chemical element that has a relatively which consists high density (superior to 5 g/cm^3) that sometimes can be very toxic to cause health problem [8]. Heavy metals are known to be one of the most common or major worldwide pollutants that can affect the ecosystem which derived from the anthropogenic activities. This contamination is believed to be transported by the metal concentration released by the industrial activity of their waste and process, and as well as from the activity urban zone [9]. The non-essential of heavy metals concentrations can pollute the aquatic environment which can reduce the survival of aquatic organisms including fish and as well as the reduction of the surface sediments quality [10]. Heavy metals can present in a dissolved form in waterways which are transported to streams and rivers by surface runoff or groundwater and thus, leads to the settlement of heavy to sediments. When the heavy metals bound to sediment, they will be affected by processes that affect the sediment directly, such as settlement in slow moving parts of the water body, re-suspension under turbulent conditions and flocculation/settlement in estuaries [11]. The common sources of metals are from the industrial activities, petroleum contamination and sewage disposal. Thus, significant inputs from anthropogenic sources can increase the concentration levels that cause toxicity therefore will be degrading the environment, decreasing the biodiversity and affecting ecosystem and human health [12]. One of the largest problems that associated with the persistence of heavy metals is the potential for bioaccumulation in the food chains [13]. A high bioavailability is linked to high concentrations and bioaccumulation within organisms and its surrounding may lead to negative effects on biodiversity via the inability to secrete physiologically-stored pollutant concentrations, which will cause toxic effects that could progress through certain levels of the food chain and their physiological state [14]. The trace metals play a biochemical role in the life processes of all aquatic plants and animals. Therefore, they are essential in the aquatic environment. However, if it presents in an excessive amount, it will become toxic and harmful [15].

2.3 Types of Sediment

They are few types of sediments by component, namely primary minerals, secondary minerals, organic matter, oxides and hydrous oxides as well as carbonate and sulfate. The primary minerals are formed through the physical weathering process from the parent rock materials. The primary minerals normally found as sand, silt and clay such as quartz, feldspar and micas. As for the secondary minerals, they are normally derived from the physical, chemical and biological weathering. They also normally found in the form of clay soil. Clay is known as secondary minerals that normally containing metals, aluminium, silicon and oxides. This type of sediment also has large specific surface area and charges on the surface which can retain any material or pollutants [26]. The difference properties in size grain particle of sediment are normally due to the sediment density, sediment shape and its chemical composition. The size or grain shape defined as the spherical and roundness of sediment particle. The shape is 15 determined by the type of weathering processes that formed the sediment as well as the transport history of the sediment. As for the density of sediment it is determined by the mineralogy of the sediment itself. Most of the sediment that originated from quartz, settled down at the coastal area because it is abundance in the Earth crust, restrict towards the physical and chemical weathering. On the other hand, the shore and beaches sediment are primarily made of from quartz and feldspar which sometimes will have a portion of heavy minerals in it such as magnetite. The clay sized sediments are also a part of the sediment bed in the estuaries where they settled as bentonite and kaolinite. The size of gravel sediment is ranged from larger than 2 mm which found in the beaches and fluvial area. As for the sand sediment is ranged from 0.063 mm to 2 mm and normally found at the beaches, shelves and estuaries. Next, for the silt-clay size sediment is ranged from 0.063 mm to 0.004 mm which settled down at shelves and estuaries [16].

2.4 Bioaccumulation of Heavy Metals in Sediment

The heavy metals that introduced to the water bodies will normally distribute as water-soluble species, colloids, suspended forms and sedimentary phases [17]. Sediment is also considered as the largest depository of heavy metal. However, the heavy metals contaminants will never stay and accumulate over the sediment forever because under some circumstances, the heavy metals will distribute and transfer to water, aquatic organism and plant by some physical chemical characteristic and condition of the water [18]. Since the sediment is one of the major components in the river ecosystems, the concentration of heavy metals within the sediment can be in contact which they dominate toxicity of heavy metals to the aquatic environment and the end of the day, affect the community’s health through the consumption from the food chain hence, the sediment can be considered as the carrier of heavy metals in aquatic ecosystems [17]. The water and invertebrates are the major component in the bioaccumulation of heavy metals in the sediment. The bioaccumulation by invertebrates or benthos is due to the feeding behavior and ingestion rates [19]. The sediment is considered as the settlement sink in the river, where the marine biota or benthos can accumulate the toxicity within them. Worst, the bioaccumulation can move from the marine biota into the food chain to birds, fish and other animals [20].

3. Method

3.1 Study Area

The samples were collected using a 5 cm diameter hand operated corer capable of collecting 20 - 30 cm long sediment core samples. The location of sampling points is tabulated in Table 1. Figure 1 shows the location of the sampling point. The sediment core samples were cut and sub-sampled by slicing them into 2 cm slices from top to bottom [21]. The sliced sediment core samples were transferred into pre-cleaned plastic container and oven-dried at 60 °C [22]. The sediment samples were pulverized, sieved through 250 um and digested using 10 mL. 65 % HNO_3, 37 % 5 mL HC1 and 1 % H_2O_2 prior to heavy metal analysis using ICP-MS [23].

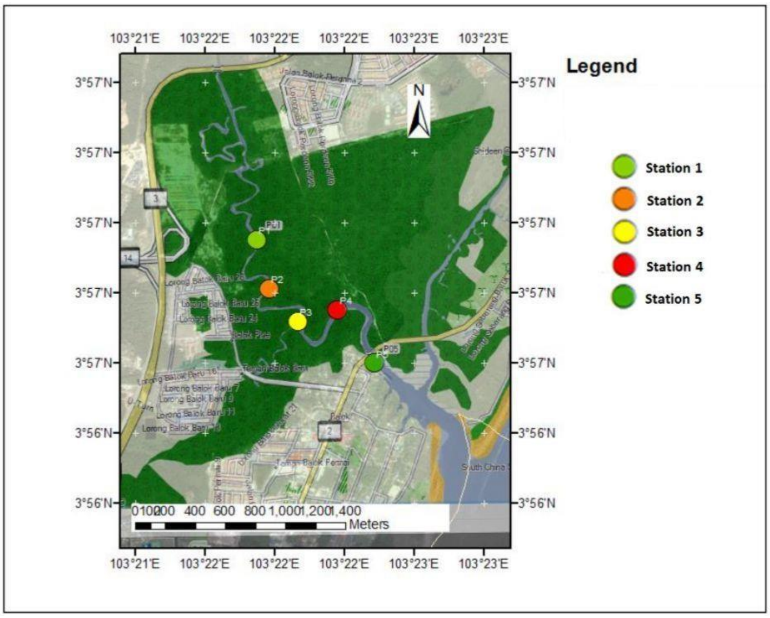

Fig. 1. Sampling points of Sungai Balok

3.2 Samples Preparation for Sediment

For sediment samples, the samples were weighed and oven-dried at 105°C until constant mass. After that, the dried samples were reweighed. Then the samples were pulverized and sieved through 220 µm stainless steel sieves. This step was to break up any aggregates. The surface sediment samples were manually ground in an agate mortar and pestle. After that, the samples were proceeding with acid digestion process. For the microwave acid digestion, a mixture of HNO_3, H_2O_2, and HCl were used for biological and environmental samples. A 0.5000g of each pulverized and sieved surface sediment sample were weighed and mixed with the acids (9ml of 37% HCl and 3ml of 65% HNO_3) in the Teflon Flour Modified (TFM) vessel. After that, the TFM vessel was introduced into HTC safety shield. Then, the combination of acid and samples solution was digested using a milestone ETHOS PLUS with HPR-1000/10s high pressure rotor according to EPA Method 3051.

3.3 Samples Digestion

The accumulation of heavy metals in the sediment may also due to the increasing discharge of municipal and industrial wastewaters, agricultural runoff region in the area [24]. Figure 2 is a flow chart also show that the heavy metal distribution concentration at sampling stations 4 and sampling station 5 are having relatively lower concentration than metal concentration at upstream sampling station (station 1 and station 2); mainly due to downstream sediment having coarser sediment could be resulted from water turbulence of the coastal water. The coarser size had less clay mineral material content which cannot favor the accumulation of heavy metals [25].

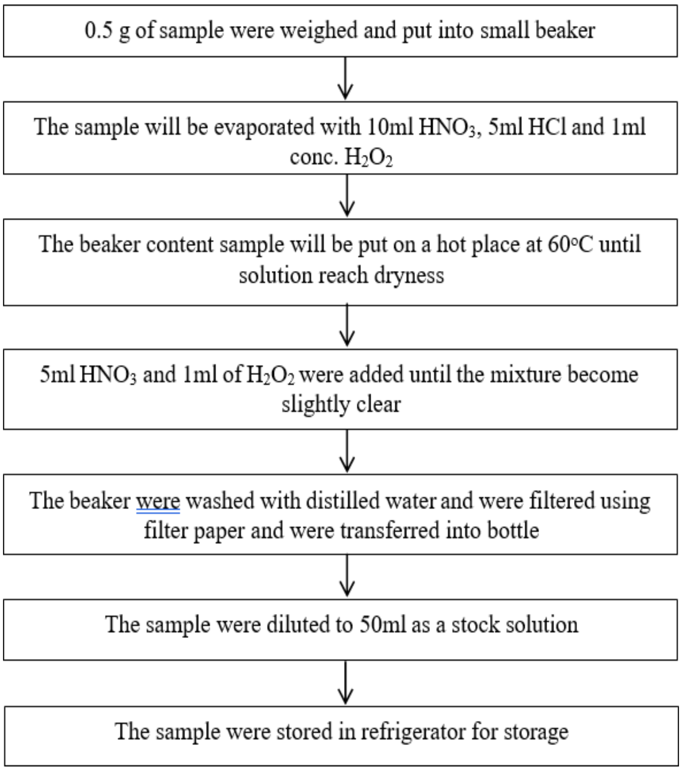

Fig. 2. The flowchart of sediment sample digestion

3.4 Calculation for Enrichment Factor Determination

The determination of enrichment factor will be applying the same procedure as the sample preparation of heavy metals concentration. This method applies the different type of pore-sieves that will be used to separate the sediment fractions to coarse size (250 µm) and fine size (63 µm). Then, after the sample digestion, the samples were analyzed using ICP-OES and the results were further calculated to determine the Enrichment Factor [25].

The Enrichment Factor is a tool that can be used to determine the enrichment and distribution of heavy metals concentration and to evaluate the possible sources of heavy metal either from natural sources or manmade (anthropogenic) in the sample [26]. Eq. (1) was used to determine the Enrichment Factor (EF).

$$EF_{metal} = \frac{(Me/Fe)_{sample}}{(Me/Fe)_{background}} \tag{1}$$

Where,

(Me/Fe) sample = The metal to the Fe ratio in the sample of interest.
(Me/Fe) background = The natural background value of the metal to Fe ratio.

4. Results and Discussion

4.1 Distribution of Heavy Metals Concentration in Sediment of Sungai Balok

Table 1 show the tabulates mean concentration of heavy metals (As, Cr, Fe, Mn, Pb, Zn) in core sediment Sungai Balok.

Table 1
Average of heavy metals concentrations in core sediments in Sungai Balok

Stations	Heavy metals concentration (mg/kg) (n=3)					
	As	Cr	Fe	Mn	Pb	Zn
Station 1	16.86 ± 0.21	85.45 ± 0.01	64859.21 ± 0.01	228.31 ± 0.01	42.07 ± 0.03	287.92 ± 0.01
Station 2	27.10 ± 0.09	66.87 ± 0.01	56845.15 ± 0.01	238.44 ± 0.02	52.82 ± 0.03	256.55 ± 0.01
Station 3	25.41 ± 0.15	26.53 ± 0.01	26706.30 ± 0.01	136.93 ± 0.01	23.82 ± 0.02	116.37 ± 0.01
Station 4	18.41 ± 0.03	36.92 ± 0.04	35476.21 ± 0.01	245.68 ± 0.01	20.00 ± 0.01	107.22 ± 0.01
Station 5	19.70 ± 0.01	26.28 ± 0.01	26944.72 ± 0.02	175.26 ± 0.01	19.95 ± 0.01	99.58 ± 0.03

The concentration of heavy metals in sediment of Sungai Balok varied depending on the sampling location. Most of the heavy metals were detected in high concentration at stations upstream stations of the river away from the shore; indicating possibility of the source of the elements from the upstream of the river closer to the industrial areas. High concentration of Fe was detected in sediment samples may be due to this element is the most abundance in earth's crust. The results concurred with the study reported by Bentum *et al.,* [27].

4.2 The Extent of Pollution

The enrichment factor (EF) is a tool that can be used to determine the enrichment of heavy metals concentration in order to evaluate the possible sources of these metals either from natural or anthropogenic sources [26]. The enrichment factor measured was used Fe as the natural background value [28]. The element mostly used in enrichment factor for sediment normalization is Al because of its characteristics such as high natural concentration, minimal anthropogenic contamination, it is a structural element of clays, and the proportions of metal in the crust are constant [22]. Eq. (1) was used to determine the enrichment factor. In this study, Iron (Fe) was used as a reference element in order to compare the EF in sample with the EF in earth's crust [29].

Enrichment factor (EF) was determined in each sample to indicate how the contaminants migrate into sample. The EF range was shown in Table 2:

Table 2
EF range

Class	Value	Sediment Pollution
0	$EF \leq 1$	No enrichment
1	$1 < EF \leq 3$	Minor enrichment
2	$3 < EF \leq 5$	Moderate enrichment
3	$3 < EF \leq 10$	Moderately severe enrichment
4	$10 < EF \leq 25$	Severe enrichment
5	$25 < EF \leq 50$	Very severe enrichment
6	$50 < EF$	Extremely severe enrichment

Table 3 shows the values of enrichment factor of all elements that obtained in this study. The values of EF for the studied elements were in order As > Zn > Pb > Cr > Mn.

Table 3
Enrichment Factor of heavy metals in core sediment

Location	Enrichment Factor				
	Zn	Mn	Pb	As	Cr
Station 1	5.5	0.3	4.0	12.5	1.1
Station 2	4.9	0.3	5.0	20.1	0.9
Station 3	2.2	0.2	2.3	18.9	0.3
Station 4	2.0	0.3	1.9	13.7	0.5
Station 5	1.9	0.2	1.9	14.6	0.4

The results show that Zn, Pb and As are have the enrichment factor more than 1, indicating possible enriched of these heavy metals especially As due to anthropogenic activities. The sources of As contamination vary may be from the combustion of coal and other fossil fuel for industrial applications to the natural sources [30]. While for Mn and Cr with the enrichment factor <1 are most likely attributed from natural sources.

4.3 Comparison with EPA Region V Standard

The mean concentrations of heavy metals obtained in this study were compared with the sediment criteria proposed by EPA Region V as tabulated in Table 4. The mean concentrations of Pb (32 mg/kg) and Mn (204 mg/kg) were considered as not polluted due to the concentration of these metals not exceeded the limit set by EPA Region V.

Table 4
Comparison of results from this study with EPA Region V

Heavy Metals	Sediment criteria EPA Region V			This study (mg/kg)
	Not Polluted	Slightly Polluted	Severely Polluted	
Pb	< 40	40 – 60	> 60	32
Zn	< 90	90 – 200	> 200	173
Fe	< 17000	17000 – 25000	> 25000	105986
Mn	< 300	-	> 500	204
Cr	< 25	25 – 75	> 75	48
As	< 3	3 - 8	> 8	21

4.4 Distribution of Heavy Metals from the Surface Sediment’s Core Depth Profile

Figure 3(a) to Figure 3(e) shows the heavy metals concentration distribution in sediments core depth profile. Generally, the order of concentration distribution from surface to the bottom sedimentary layers are in the following order Fe> Zn>Mn>Cr>Pb>As. In ideal condition, apart from the time factor, where the surface sediment is the recently incoming sedimentary materials while the bottom most sediment the oldest material, the particle size also play significant role in determining the elemental concentration in the sediments. Fine clay size with high surface area will carry high amount of heavy metals.

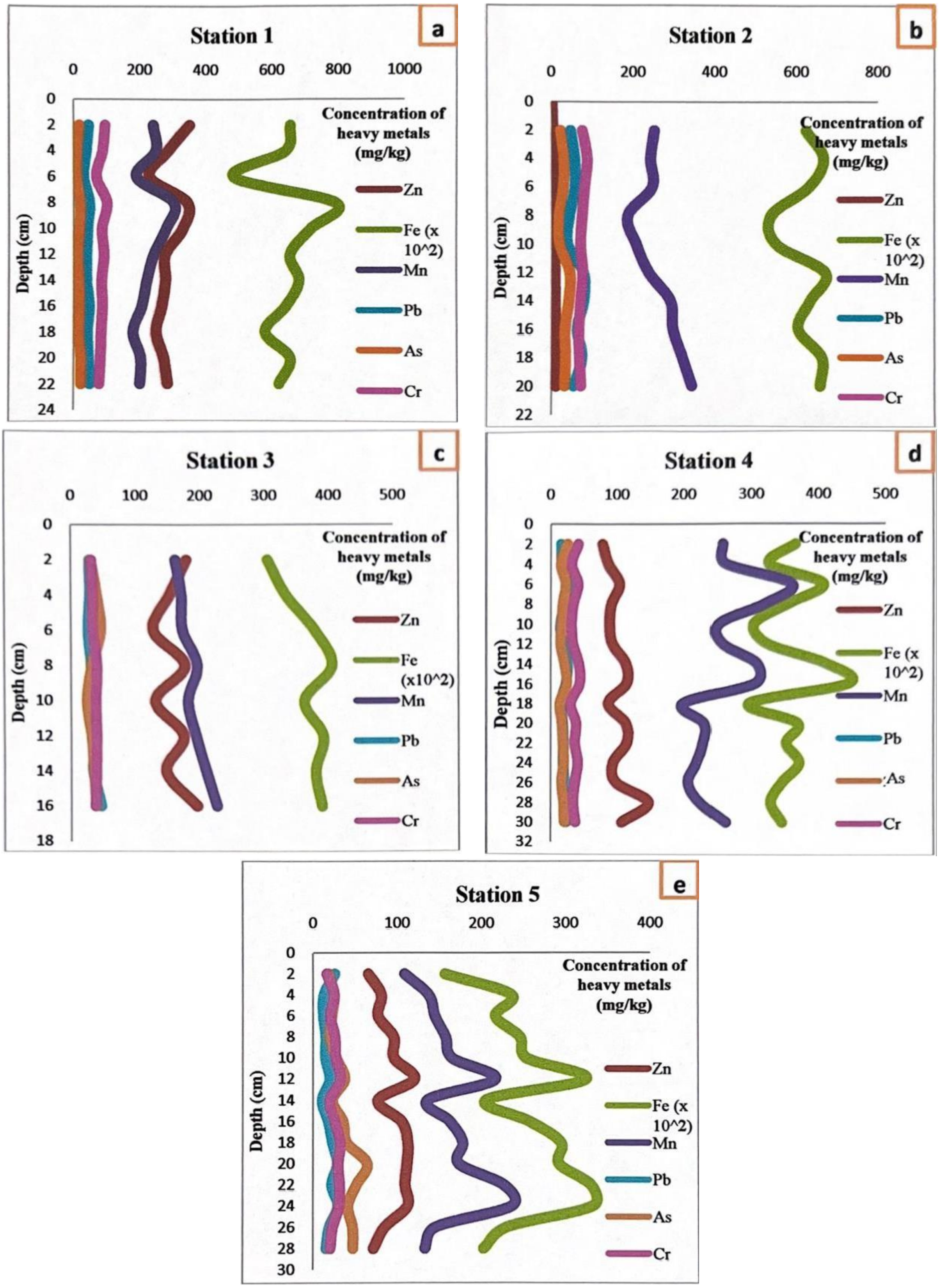

Fig. 3. Graph of heavy metals concentration between layer profiles (a) Station 1, (b) Station 2, (c) Station 3, (d) Station 4 and (e) Station 5

4.4.1 Iron, Fe

The concentration of Fe varies in concentration from the surface to the bottom sedimentary layers. The variation pattern is generally different from one station to the others, for example Fe are the highest in concentration at S7 layer which is located between 12 cm - 14 cm down the sediment core, while in station 2 the Fe is highest in concentration at surface sediment S2 or 0 - 2 cm layers. The variability of the Fe concentration is most likely due to the variation in the sedimentary particle sizes and unlikely due to the time or seasonal deposition factor.

4.4.2 Zinc, Zn

In general, Zn also varies in distribution concentration from surface to the sedimentary bottom layers and show slightly similar trend to Fe distribution patterns but slightly shifted toward higher sedimentary layers at S5 of station 1 located between 10 cm - 12 cm of the sediment core. The variation is mainly due to variability of sedimentary particle sizes. Similar trend was observed for other sediment cores at station 2, station 3, station 4 and station 5. It is important to note that Zn data only available at station 1 for the whole sedimentary cores.

4.4.3 Manganese, Mn

Generally, Mn shows similar distribution patterns relative to Fe and Zn in all sediment cores of all the sampling point (station 1, station 2, station 3, station 4 and station 5) with slightly shifted in peak concentration either down or up of the sedimentary cores. Being an element with normally in close relation to iron the similarity pattern mostly is determined by the particle sizes distribution.

Generally, most of the concentrations of heavy metals were higher at the stations the further in the river which means away from the shore and the concentration is much lower at the stations that were located at the shore. It is high possibility if the source of concentrations were from upstream. Overall, the highest concentration of heavy metal contained in sediment was Fe and the lowest concentration was As. Fe was to be highest because this element is already or readily existed in the Earth's crust which known as the parent rock materoal for sediment [27].

5. Conclusions

The heavy metals and metalloid contents in the Sungai Balok sediments can be arranged in the following order with highest in concentration of Fe > Zn> Mn> Pb> Cr> As. Based on the enrichment factor indicator, if the value of EF less than 1, it indicates the heavy metals is naturally occurring in or exist from the natural activities such as weathering and erosion from the parent rock material. If the value more than 1 it is indicating the enrichment of heavy metals were from the anthropogenic activities. This indicates that the As has very high enrichment. Then, the enriched elements followed by Pb, Zn, Cr and Mn. As for Pb and Zn, they were moderately enriched while as for Cr and Mn, both of these elements were naturally occurring in the sediment of Sungai Balok. The application of enrichment factor and the USEPA sediment criteria Region V signified that As, Pb and Zn are the pollutants that most possibly from the anthropogenic sources. While others most likely from natural sources. From the sediment core results the heavy metal concentration variation mostly due to difference in particle sizes distribution and not by other factors such as temporal or seasonal.

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