DETERMINATION OF SELECTED METALS IN SEDIMENT FROM BESUT AND DUNGUN RIVER

By

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DECLARATION

I declare that this thesis entitled "Determination of selected metals in sediment from Besut and Dungun River" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : ______________________________

Name : ______________________________

Date : ______________________________

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LIST OF ABBREVIATIONS

EF	Enrichment Factor
Igeo	Geoaccumulation Index
MPI	Metal Pollution Indices
PCB	Polychlorinated Biphenyl
PLI	Pollution Load Index
SB	Besut River
SD	Dungun River
SPSS	Statistical Package Social Science
USEPA	United State Environmental Protection Agency
WHO	World Health Organization

LIST OF SYMBOLS

%	Percentage
<	Less than
>	More than
°C	Degree Celsius
Al	Aluminium
As	Arsenic
Ca	Calcium
Cd	Cadmium
Cr	Chromium
Cu	Copper
Fe	Iron
Hg	Mercury
K	Potassium
Mn	Manganese
Ni	Nickel
Pb	Lead
Si	Silicon
Ti	Titanium
ppm	Part per million
Ti	Titanium
Zn	Zinc
Zr	Zirconium

Determination of Selected Metals in Sediment from Besut and Dungun River

ABSTRACT

Metals are the elements that occur naturally to environment in form of rock, soil or water. If high concentration of metals was detected in the environment it will cause bad effects to the surrounding. In this study, metals concentration in sediment from Besut and Dungun River were investigated. The objectives of this study are to determine the concentration of metals in sediment and to evaluate sources and level of metal pollution using pollution assessment such as Enrichment Factor (EF) and Geoaccumulation Index (Igeo) in Besut and Dungun River. Seven sediment samples from Besut River and three from Dungun River were dried and sieved by using 75 µm sieves. Sediment samples were analysed by X-ray fluorescence. Based on the mean values of the metal concentrations, the metals follow in the following order, from the highest to the lowest; Si > Fe > Al > K > Ti > Zr >Ca> Mn. When the EF value is calculated, the results showed that Fe, Mn, K, Ti, and Zr were come from anthropogenic sources and Al, Ca, and Si were come from natural sources. The results was supported by Igeo when it was found that both rivers were moderately polluted to extremely polluted by Fe, Mn, K, Ti, and Zr. Most of the pollution shown that the sources were come from point sources such as activities nearby to a river bank, water gate, road bridge and aquaculture at the T junction of a river.

Penentuan Logam terpilih dalam Sedimen Dari Sungai Besut dan Sungai Dungun

ABSTRAK

Logam adalah unsur yang wujud secara semula jadi dalam bentuk batu, tanah atau air di persekitaran. Jika kepekatan logam di persekitaran tinggi ia akan menyebabkan kesan buruk. Kepekatan logam di dalam sedimen dari Sungai Besut dan Dungun telah dikaji. Objektif kajian ini adalah untuk menentukan kepekatan logam dalam sedimen dan menilai sumber dan tahap pencemaran logam menggunakan penilaian pencemaran seperti Penilaian Faktor Pengkayaan dan Indeks Geoakumulasi. Tujuh sampel sedimen dari Sungai Besut dan tiga dari Sungai Dungun telah dikeringkan dan disaring dengan menggunakan penapis bersaiz 75 mikrometer. Sampel sedimen telah dianalisis menggunakan X-ray pendarfluor. Berdasarkan nilai purata kepekatan logam, turutan logam dari tertinggi ke terendah adalah; Si> Fe> Al> K> Ti> Zr> Ca> Mn. Keputusan menunjukkan bahawa Fe, Mn, K, Ti, dan Zr berpunca daripada sumber antropogenik dan Al, Ca, dan Si daripada sumber semula jadi berdasarkan nilai Faktor Pengkayaan. Keputusan ini disokong oleh Indeks Geoakumulasi apabila didapati bahawa kedua-dua sungai adalah pada tahap sederhana tercemar hingga sangat tercemar disebabkan oleh Fe, Mn, K, Ti, dan Zr. Kebanyakan sumber pencemaran adalah berpunca daripada aktiviti berdekatan dengan tebing sungai, pintu air, jambatan dan akuakultur di persimpangan sungai.

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Metals are the elements that occur naturally to environment in form of rock, soil or water. If human beings or organisms received too little or too much metals concentration, defect symptoms can arise and give bad effects. A higher concentration of a metal also can cause problem such as health risk to human and environment. This issue became a concern to the international researcher in twentieth century because it is considered a risk to the environment and human health (Elekes, 2014).

One type of metals that are potentially cause risk to environment is heavy metals which consist of Cu, Pb and Cr and soon. Heavy metals are widely used in the industrial activity. For example, the source of cadmium comes from industrial discharge and the application of fertilizer and sewage sludge on farmland (Järup, 2003). Trace metals is also one type of metals that are causing pollution in the environment. For examples, Mn can be found in the environment by the anthropogenic activities such as emission of iron and steel production, discharge from municipal wastewater and sewage sludge (Howe *et al.*, 2004).

In industrialization era, without practice the proper waste management would threaten the aquatic life by accumulation of metals in rivers. The increase of metals in water has change the natural concentration in sediment. As we know, anthropogenic activity nearby the river can cause heavy metal contamination. Examples of heavy metal contaminant from anthropogenic activities such as industrial effluents, aquaculture, solid waste disposal, agriculture runoff, mining and metal processing (Zulkifli *et al*., 2015). These sources contain a huge amount of concentration of heavy metal and directly contaminated the surrounding areas.

Metal contamination also can affect both human health and aquatic life such as decrease the water quality and able to accumulate in fish body. If human ate those contaminated fish it were cause to health problem and also death (Adeosin *et al.*, 2015). It was found that the *Oreochromis niloticus* species has exceeded the amount of heavy metals in body part which posed health hazards for consumer (Saeed and Shaker, 2008).

Evidently, trace metals also gives bad effects to the aquatic life. For example, study done by Noor and Zutshi (2016) had investigated the bioaccumulation of metals in Rohu fish between two lakes; Vengaiah and Yellamallappa Chetty Lake. It was found that the trace metals such as Al, Fe and Zn had accumulated in the gills fish than its muscle which causing the system in their body cannot function properly.

Water resources like river are commercially used to supply clean water for for the food sources, bathing, washing and industrial usage. When anthropogenic activities increase, its can cause the heavy metal contamination in both river and sediment (Shanbehzadeh *et al.*, 2014). Due to this problem, we are not suggested to get water supply from the contaminated river.

The significance of this study is to have useful and updated data of the metals concentration in both rivers which contributed water and soil pollution in Besut and Dungun River. At the same time, the local authority and non-government agencies aware about current situation happened in Besut and Dungun River. For example, frequently do enforcement to the active industrial companies by paying penalties of illegal pollutant released from their company.

1.2 Problem Statement

The accumulation of heavy metal in sediment was dangerous threat to the aquatic life, human health and the environment. Trace of metal contamination in sediment could affect the water quality and bioaccumulation of metal in aquatic life (Yisa *et al*., 2012).

Besut and Dungun River were used as water sources for aquaculture, municipal water supply, and wastewater dilution and it also were exposed to anthropogenic activities such as oil spill from boats, agriculture runoff and residential area (Suratman *et al.*, 2009; Suratman, 2013).

The discharged from the anthropogenic activities such as spills from boats were introduced the metals and other substances such as hydrocarbon that could affect the aquatic life and the environment. For example Saeed and Shaker (2008) had studied heavy metals pollution in water and sediments and their effect on *Oreochromis niloticus* in the northern delta lakes, Egypt. Study was found that the lakes are heavily polluted and their reading is beyond the standard guidelines.

Despite for all the anthropogenic activities identified in Besut and Dungun River, there are no strict regulation that have been used by the government to reduce emission and discharge that comes from these activities. Hence, this study is to provide the government a database of levels of metals in both rivers.

1.3 Objectives

i. To determine the concentration of heavy and trace metals in sediment from Besut and Dungun River
ii. To evaluate sources and level of metal pollution using pollution assessment such as Enrichment Factor (EF) in Besut and Dungun River

CHAPTER 2

LITERATURE REVIEW

2.1 Metals

Metals are commonly derived from the minerals that are naturally found in the environment. It can be obtained from natural and anthropogenic sources such as weathering of soil and industrial activity respectively. Metals also can be classified into light, heavy, semimetal (i.e. metalloids), toxic, and trace and it's depending on its density, weight, atomic number, and degree of toxicity (Roberts *et al.*, 2005).

There are certain types of metals that can be harmful to the environment such as heavy metals (Singh *et al.*, 2011). The potential element of heavy metal that present in the sediment or soil are Cadmium (Cd), Arsenic (As), Chromium (Cr), Lead (Pb), Nickel (Ni), and Mercury (Hg). These element is commonly come from agriculture runoff, industrial sewage sludge and exhaust gases petrol engines (Sherene, 2010).

Previous study done by Saeed and Shaker (2008), had studied heavy metals pollution in water and sediments and their effect on *Oreochromis niloticus* in the northern delta lakes, Egypt. Study was found that the lakes are heavily polluted and their reading is beyond the standard guidelines. The edible part of *Oreochromis niloticus* that are from Lake Edku and Manzala also concentrated with Cd and Cr.

The presence of metal contamination in the environment occurred by different pathways such as diffusion into the bloodstream through fish gill and skins, soil, water and air. In the same study, the accumulation in the fish species are higher in the fish organs such as gills and liver (Saeed and Shaker, 2008). Moreover, aquatic organisms have the ability to accumulate and biomagnified contaminants like heavy metals, polycyclic aromatic hydrocarbons and PCB in the environment (Davies, *et al*., 2006).

Certain metals are essential to the environment and for animal and human health. For examples, Fe, Cu, Cl, Mn, and Zn are the essential element of metals that acts as micronutrient and is essential to their nutrition and growth (Johnston, 2005). The source of these metals may come from natural and anthropogenic origin.

For example, the study of trace metals (V, Al, Sn, As, and Se) in seawater, sediments and some fish species from Marsa Matrouh Beaches in north-western Mediterranean coast, Egypt done by Ghani (2015). The results showed that Sn in sediment sample was come from anthropogenic source while the other metals were come from natural source.

In the same study done by Ghani (2015), the researcher was using the Metal pollution indices (MPI) to determine the amount of metals in the fish. The recorded values were lower than 1 except in demersal fish species S. *undosquamis* (1.430) indicating that it is safe for human consumption.

2.2 Besut and Dungun River

Dungun district consists of 35% lowland area, 20% swamp and other water bodies, and 45% forest reserves. Dungun River is the river that includes in the water bodies in Terengganu, approximately 80km long before reaching South China Sea (Department of Irrigation and Drainage, 2011). The lengths of Besut River is approximately 69km and have main streams such the La, Keruak, Angga, Tadau and Jertih Rivers (Department of Irrigation and Drainage, 2011). There are no major industrial activities in both rivers. However, both river basins were used as water sources for aquaculture, municipal water supply, and wastewater dilution (Suratman *et al.*, 2009; Suratman, 2013).

From the previous study, Suratman *et al.* (2009) had focused on the distribution of trace metal in Besut River Basin. It was found that the selected metal, Cu, Fe, and Pb still not higher than other study area. However, continuous inputs from anthropogenic activities may increase the concentration of trace metal in that basin time to time. Continuous study needed in order to monitor the Besut River.

Another study done by Ghazali *et al.* (2011) had investigated the distribution of Cd, Mn and Pb in the south China sea off the south Terengganu Coast, Malaysia during post-monsoon and pre-monsoon. It was found that the concentration of Lead was slightly higher in Dungun river estuary compared to other sampling points. The results also showed that Dungun River area has the highest concentration of trace metal.

The study of effects of Northeast Monsoon on trace metal distribution in the South China Sea off Peninsular Malaysia by Ghazali *et al.* (2014) showed that the concentration of trace metals Cd and Mn was highest in the area of Dungun river estuary. The result also showed that the factor of metal distribution was the anthropogenic activities and the climatic change between Northeast and Southwest monsoon.

2.3 Sediment

Sediments are combination of several components of mineral species such as organic debris and its act as the accumulation of metals in the environment. It also used to assess the impact of anthropogenic activities to the aquatic environment (Zulkifli *et al*., 2015). The sediment pollution occurred when sediment are additional influenced with chemical adsorption between the metals, grain size, and organic matter (Aprile and Bouvy, 2008).

Besides, sediment was categorizing as nonpoint source pollutants because it comes from various sources and being introduced to the river through the surface runoff. When anthropogenic activities occurred near the river and produced potential source of heavy metal such as agricultural runoff and oil spill from the ships and boats, the soil particles that are transported by surface water movement can cause sediment pollution (Asplund, 2000).

2.4 Potential source of metals in sediment

Aquatic environment has been introduced with metals through natural conditions such as weathering of minerals and anthropogenic sources such as industrial waste, agriculture runoff, burning of fossil fuels, vehicular emission, mining and metallurgical processes and their waste disposal (Likuku *et al.*, 2013).

Metals are being released into the environment are mainly comes from the anthropogenic activity as they increase more concentration to the naturally amount of metal that should be in the environment. For example, it was found that there are major changes of water quality in Terengganu river basin is due to the several anthropogenic activities that are found in that river (Suratman *et al*., 2015).

2.4.1 Natural

The natural source of heavy metal comes from the earth and it’s become a concern when higher concentration of the metal is occurred in the environment. Natural sources commonly comes from the weathering of soil and rock, erosion, wildlife fire, and volcanic eruptions activities and being released naturally into the rivers (Ren *et al*., 2015).

From the study done by Ren *et al*. (2015), they had analysed the heavy metal pollution in Yellow River sediment. The results showed that metals Cu, Fe and Zn were come from natural sources such as erosion and weathering.

The highest heavy metals contamination in some commercial fish at Ogun River, Opeji, Ogun State, Nigeria was Zinc (Zn) and its comes from agriculture activities and biological breakdown of rocks in the river (weathering). The results show that the fish in the river are still safe to be consumed (Adeosin *et al.*, 2015).

Study done by Kljakovic *et al.* (2008) that determined the content of trace metals (Cd, Pb, Cu, Zn and Ni) in sediment of Soline Bay, Rogoznica, Croatia showed that metals of Cd and Ni comes from natural sources. These results were supported by the statistical analysis of correlation coefficient and PCA analysis.

2.4.2 Anthropogenic

Anthropogenic source comes from the human activities such as mining, industrial effluent, agricultural runoff, residential waste and vehicle emission (Rajeswari and Sailaja, 2014). As for the source that cause heavy metal adsorption in the sediment is from the vehicle emission such as boat and ship, agriculture activity near the river, and the residential effluent from unmanaged drainage.

The highest heavy metals contaminations were Cd, Cu, Fe, and V and comes from the anthropogenic activities such as industrial activities, mining and exploitation of oil reservoirs (Roozbahani *et al.*, 2015).

Mokhtar *et al.* (2015) have studied that the major metals contamination in Langat Estuary River are comes from anthropogenic activities that occurred at Port

Klang and Langat River through water pathways. Highest concentration of Ni and Zn was found from the study.

Results from the study done by Aprile and Bouvy (2008) showed that metals such as Zn, Cu, Pb and Ni in Tapacurá River Basin were accumulated in the sediment. It also showed that the metals distribution came from anthropogenic source such as fertilizers and herbicides, municipal sewage, and industrial effluents.

Another study done by Kamaruzzaman *et al.* (2006) that investigated the concentration of Cu, Mn, Pb, Zn, and Th in the sediment of Paka estuary, Terengganu showed that higher concentration of metals were at the surface of sediment. According to the EF value, results also showed that all the metals except for Mn come from anthropogenic activity.

2.5 Potential effects of metals

The presence of metals can affected the system of the environment if it is contained a high concentration of the metals. Some of the metals can be taken up by organism and some of them are toxic.

Exposure of Al can lead human to be associated Alzheimer diseases which can affect human brain by causing forgetfulness and loss of mental skills. It also causes aluminosis condition by breathing Al (Newton and Edgar, 2010).

Concentration of Fe in the river affects water quality and bioavailability. In ponds with acidic sediment, iron deposited on the exoskeletons and gills of shrimp which will cause physical damage to the gills and unsightly blotches on shrimp at harvest (Claude, 2008). Next, metal such as Mn can give both negative and positive impact to human body. However, the need to take up Mn only in minimum amount and if excess, it can cause health problem such as sleepiness, emotional disturbance and the worst is can be paralyse (Newton and Edgar, 2010).

While other metals such as Ca, K and S were the metals that are essential to both human and plants. Plants and animals must take up these metals in other to ensure their growth and to keep functions in their system work properly. Besides, there were metals such as Si, Ti, and Zr that are occur naturally in the earth crust. These metals can cause some health problem such as irritation in eyes and skins, respiratory problem (Newton and Edgar, 2010).

2.6 Summary from previous studies

Based on the Table 2.1, it shown total concentration of heavy metal from different river from Malaysia and other countries studies. From previous study, there were no studies about the concentration of metals Si, Ca, and Ti and K and Zr in Malaysia. However, there were studies of metals Al, Mn and Fe.

A low concentration of metals such as Al, Mn, Fe were found in the study done by Shaari *et al.* (2015) that determined the distribution of metals in sediment of EEZ of the east coast of Peninsular Malaysia especially nearshore of Terengganu and Pahang. However, Fe concentration was slightly higher due to the presence of anthropogenic activity that is iron ore mining.

The high concentration of Al and Fe were found in the study of distribution of metals in coastal sediment of Port Klang except for Mn. The results shown that Igeo accumulation for both metals were moderately pollute and comes from anthropogenic source (Sany *et al*., 2013).

Meanwhile, the study done in Kelantan River showed that the concentration of Fe and Mn was lower compared to the background level of earth crust. The results shown that the metals come from natural origin (Ahmad *et al.*, 2009).

Results obtained by researchers to study the level of heavy metal contamination at Balok River in Pahang found that the highest concentration of metals was Fe. The results also indicate that Fe and Mn sources come from both natural and agricultural source. It was supported by the Igeo and EF assessment that there is no pollution that cause by the industrial discharge (Abdullah *et al.*, 2015).

The study of Mn and Fe contamination in mangrove sediment, Sarawak showed that the levels of concentration in the sediment are higher than the standard guideline such as USEPA and WHO Sediment Quality Guidelines in that study (Billah *et al.*, 2014). The results also compared to worldwide studies which showed that the concentration of the metals were within the range.

Another study in Malaysia had investigated the heavy metals distribution coastal sediment and the relation with grain size in Johor Port. It was found that high concentration of Fe and Mn in the fine sediment. However, the pollution assessment, EF value of Mn was indicates that it comes from natural origin (Abdullah *et al.*, 2015).

Metals that are studied from other countries are Al, Fe, Mn, Ca, K, Si, and Ti and Zr. The study of metals distribution along the coastal of Tamilnadu showed that the concentration of metals mentioned above except Zr are not pollute the sediment (Harikrishnan *et al.*, 2016).

Furthermore, other study that had been done at Southwest Nigeria River were showed that the metal contamination in urban stream sediment were cause by

Mn. Major metals such as Fe, Ca, K were below the average of shale (Odukoya and Akande, 2015).

Study done by El-sorogy and Attiah (2015) that investigated metal contamination in coastal sediments, seawaters and bivalves of the Mediterranean Sea coast, Egypt only showed the results of the concentration of Fe, Mn, K and Zr. According to their EF value, the results indicate that the source is natural origin. However, based on the Igeo Accumulation index, Al, K, and Mn were unpolluted and Fe was moderately polluted the sediments.

Lastly, the study of trace metal distribution and environmental risk in sediments from the Lake Kalimanci showed that the concentration of Al, Fe, Mn, Ca, K, Si, Ti except Zr were comes from natural origin. The source of these metals comes from mineral weathering (Vrhovnik *et al.*, 2013).

2.7 Assessment of metals pollution

2.7.1 Enrichment Factor (EF)

Enrichment factor (EF) is used to know the origin of the elements found in river sediment (Qingjie *et al.*, 2008). The calculated EF value from the analysed heavy metals is also to evaluate whether the source of heavy metals was from natural or anthropogenic source (Parizanganeh *et al.*, 2012). Al was chosen as normalization in determined the EF due to the content of Al that is mostly made up from natural resources as used in study done by (Vrhovnik *et al*., 2013).

Table 2.1: Previous studies from Malaysia and International countries in ppm

	Malaysia studies						**International studies**			
Element	EEZ, Peninsular Malaysia	Port Klang, Selangor	Kelantan River	Balok River, Pahang	Mangrove sediment, Sarawak	Johor Port	East coast Tamilnadu	Southwest Nigeria River	Mediterranean Sea Coast, Egypt	Lake Kalimanci
Al	423000.00	14724.00	-	-	-	-	23691.00	26000.00	-	79000.00
Fe	756000.00	6547.00	3860.00	47028.00	14895.00	47000.00	19302.00	75000.00	198100.00	66000.00
Mn	2700.00	231.43	394.00	155.90	91.60	1886.00	367.65	1261000.00	5700.00	6000.00
Ca	-	-	-	-	-	-	12076.00	2000.00	-	38000.00
K	-	-	-	-	-	-	6179.00	300.00	640.00	23000.00
Si	-	-	-	-	-	-	174699.00	-	-	235000.00
Ti	-	-	-	-	-	-	8460.00	100.00	-	4000.00
Zr	-	-	-	-	-	-	-	41000.00	3700.00	-
References	Shaari *et al.* (2015)	Sany *et al.* (2013)	Ahmad *et al.* (2009)	Abdullah *et al.* (2015)	Billah *et al.* (2014)	Abdullah *et al.* (2015)	Harikrishnan *et al.* (2016)	Odukoya and Akande (2015)	El-sorogy and Attiah (2015)	Vrhovnik *et al.* (2013)

Olubunmi and Olorunsola (2010) have investigated the status of heavy metal pollution of sediment of Agbabu Bitumen Deposit Area, Nigeria. The EF results show that the ranges of heavy metal enrichment are from no enrichment to moderate enrichment. It also showed the heavy metal pollution higher in the dry season than in the wet season. The low EF values obtained also indicate that the moderate heavy metal pollution observed in the soil comes from the natural processes.

The results obtained by researcher from the study of heavy metal concentration in surface sediment at Tanjung Lumpur, Pahang shows that the concentration of heavy metals in downstream the of river was high but still within their background values. They used EF indices to determine whether the source of heavy metals comes from anthropogenic or natural origin. The result shows that the sediment was polluted by lead which is coming from anthropogenic origin (Yunus *et al.*, 2011).

Furthermore, there were also studies that investigated the EF of the trace metals. For examples, the trace metal deposition in sediment from Tanjung Pelepas Harbour were investigated by using EF to determine the origin of the studied metals such as Al, Fe, Ti, Pb, Cu and Zn. The results showed that Al, Fe and Ti were comes from natural source, Cu and Zn comes from anthropogenic source, while Pb comes from both source (Yusoff *et al.*, 2015).

Another study on metals pollution on coastal sediments of Sindh Pakistan using Geochemical Approach such as EF was done by Zubair *et al.* (2014). The results recommended that trace metals Mn and Zn in the coastal sediment comes from anthropogenic activities and this due to the the industrial discharge nearby the sampling points.

2.7.2 Geoaccumulation (Igeo)

Geoaccumulation (Igeo) is used to determine the degree of contamination in the sediment by comparing the background value of every individual element with the analysed concentration element (Wang *et al.,* 2010).

The pollution of heavy metals in Tigris River was determined by using Pollution Load Index (PLI) and Geoaccumulation Index (Igeo). The Igeo index showed that all the heavy metals found in grade 0 and 1 and this suggest that this river has natural value of background concentrations for Mn, Cu, and Ni. However, Pb and Cd have exceeded the normal value in the soil (Rabee *et al.,* 2011).

Apart from that, the study of heavy metal contamination levels of Balok River, Pahang was found that the concentrations of heavy metal are slightly higher than background values. However, the results show that there are no excessive metals were released in the river (Abdullah, *et. al.,* 2015). The researcher used geoaccumulation and EF index to evaluate the levels of contamination in that river.

The metal pollution assessment in sediments of Dikrong River, NE India also was using geoaccumulation index to show the degree of contamination of metals such as Al, Fe, Ti, Mn, Zn, Cu, Cr, Ni and Pb. The results reveal that Al, Fe, Ti, Mn, Zn, Cr, Ni was not contaminated the sediment. However, Cu and Pb showed that they were comes from anthropogenic source (Chakravarty and Patgiri, 2009).

CHAPTER 3

MATERIAL AND METHODS

3.1 Sampling Locations

Study site was located on the East Coast of Peninsular Malaysia, in the state of Terengganu. In this study there were 2 selected rivers with spotted anthropogenic activities within the site. The sampling location was shown in Figure 3.1 while the geographical position and description of the sampling location was shown in Table 3.1. Samplings of the sediments were done at selected riversides of Besut and Dungun Rivers as shown in Figure 3.1b and 3.1c. The anthropogenic activities around the study area were mainly effluent from river tributaries of agriculture activities, boating and transportation activities, residential area, tourism and fishing activities as referred in Table 3.1.

3.2 Sediment and analytic method

In this study, 10 points for both rivers have been identified as sampling points of the sediment. The points have been selected based on the presence of anthropogenic activities along the river that are connected to the South China Sea as shown in Table 3.1. A flow chart of sampling analysis is shown in Figure 3.2.

3.2.1 Sediment sampling

Total of 10 samples of sediment were collected with an Ekman grab which was pushed with pressure through the water to obtain sediment layer at a depth of approximately 4 cm at each sampling point. There are 7 sediment samples were collected in 7 sampling point from Besut River. However, there were only 3 sediment samples were collected at Dungun River due to the bad weather condition.

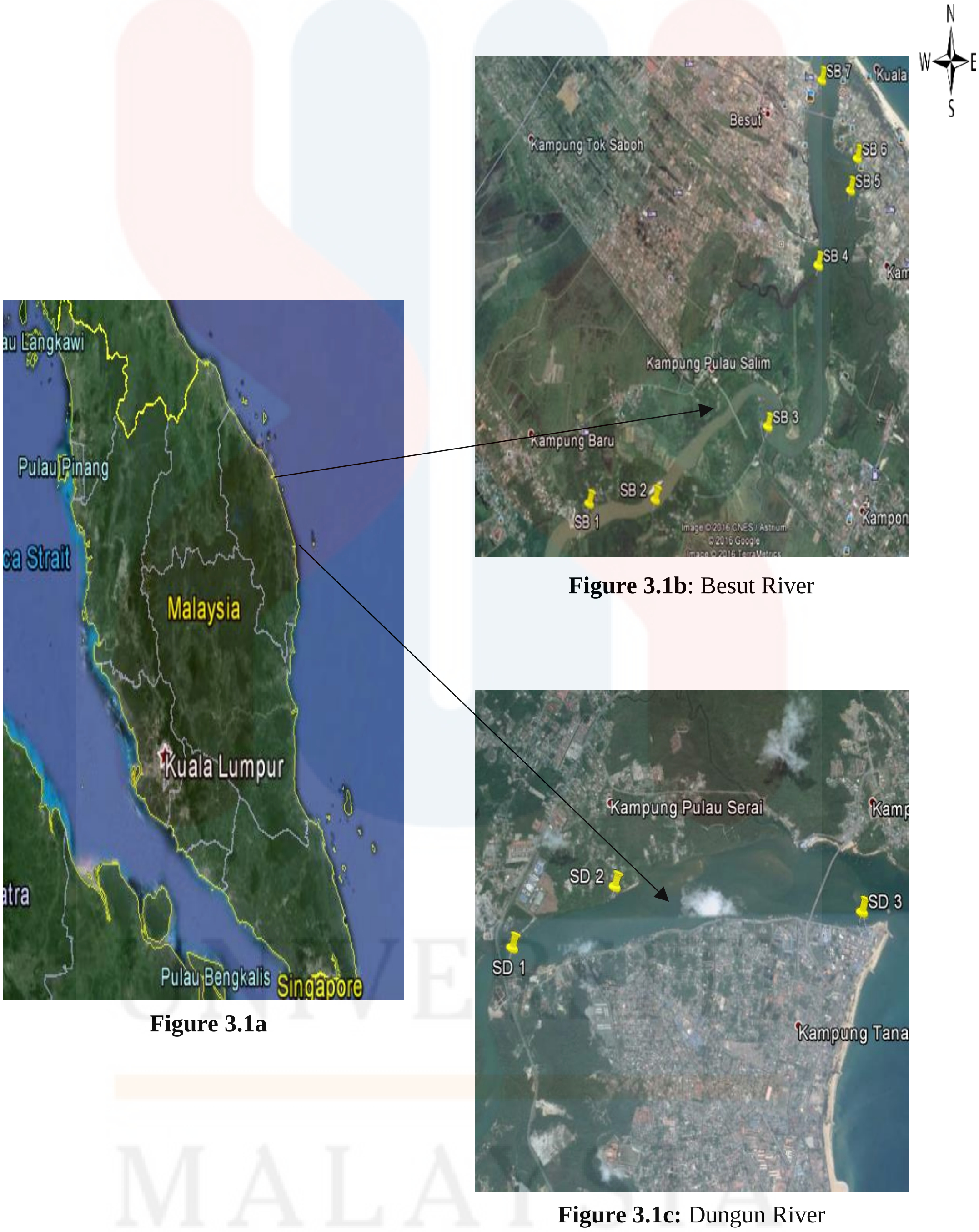

Figure 3.1a

Figure 3.1b: Besut River

Figure 3.1c: Dungun River

Figure 3.1a, b, c: The map of sampling locations in Terengganu, Malaysia

Source: Google Earth

Table 3.1: The geographical position and description of sampling locations.

Sample ID	Location	Geographical Position		Description
		Latitude (North)	Longitude (East)	
SB 1	Besut River	N 05^{O} 47'40.5"	E 102^{O} 31'49.4"	Watergate
SB 2	Besut River	N 05^{O} 47'42.0"	E 102^{O} 32'18.5"	Nearby to river bank
SB 3	Besut River	N 05^{O} 48'04.1"	E 102^{O} 33'08.1"	Agriculture runoff/ Teluk Mak Koor
SB 4	Besut River	N 05^{O} 48'53.4"	E 102^{O} 33'31.1"	T junction point of river and Aquaculture
SB 5	Besut River	N 05^{O} 49'17.1"	E 102^{O} 33'46.9"	Kupang harvesting area
SB 6	Besut River	N 05^{O} 49'27.3"	E 102^{O} 33'49.3"	Kg. Seberang Kastam Boat Parking
SB 7	Besut River	N 05^{O} 49'52.4"	E 102^{O} 33'33.0"	Jetty to Perhentian Island
SD 1	Dungun River	N 04^{O} 46'34.7"	E 103^{O} 23'37.8"	Nearby to road bridge
SD 2	Dungun River	N 04^{O} 46'50.5"	E 103^{O} 24'10.0"	Boat Parking
SD 3	Dungun River	N 04^{O} 46'44.9"	E 103^{O} 25' 29.0"	Jetty

Note: SB= Besut River SD= Dungun River

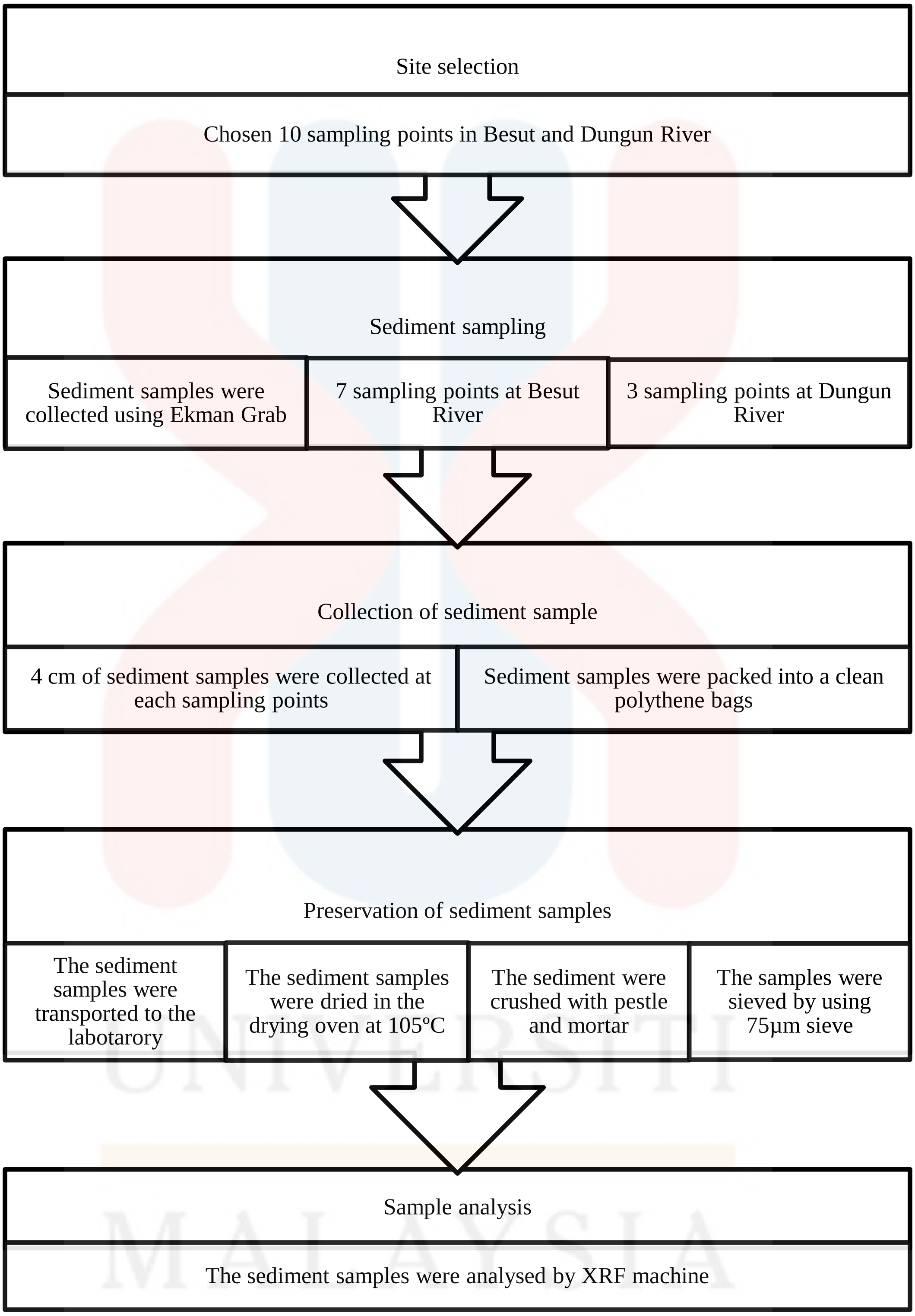

Figure 3.2: Flow chart of sampling analysis

3.2.2 Sediment preservation and preparation

The sediment samples were packed in clean polythene bags and transported to the laboratory until the further analysis. They were dried in drying oven at 105 ºC until there was no further change in weight. Dried sediments for each sampling station were crushed with a pestle and mortar. The samples were sieved by using 75µm and packed in clean polythene bags prior for XRF sediment samples that must be prepared in a powder form. Then, the prepared samples will be analysed by the XRF instrument.

3.3 Instrumentation

The metals of Al, Fe, Mn, Ca, K, Si, Ti and Zr were determined by X-ray Flouresence(XRF) Instrument. It were used for the determination of the metals that contain in the powdered sediment sample. Values of the elements Al, Cr and Fe are provided by XRF in weight percentage (wt%) in oxidized forms. The XRF results for the oxidized forms were multipled by 10,000 for wt% to ppm values.

3.4 Statistical analysis

Statistical analysis for Kruskal-Wallis test was performed using SPSS 21. Kruskal Wallis test is the non-parametric test that analysed the variance. It is to test whether there are differences in the median values of three or more independent samples (Nahm, 2016). The formula to Kruskal Wallis test is shown in equation 3.1 and 3.2 below:

$$H = \frac{12}{N(N+1)} \sum_{j=1}^{k} \left(\frac{R_J^2}{n_J}\right) - 3(N+1) \qquad (3.1)$$

(Rj = rank sum of each sample)

$$N = \sum_{j=1}^{k} n_j \qquad (3.2)$$

(n_j = number of samples for each group, k = number of groups)

3.5 Assessment of Heavy metals pollution in different rivers sediment

In order to assess the level of pollution in river sediment, Enrichment Factor (EF) and Geoaccumulation (Igeo) were calculated to observe the current status of metal contamination whether it comes from natural or anthropogenic sources.

3.5.1 Enrichment Factor (EF)

Enrichment factor (EF) is used to know the origin of the elements found in river sediment (Qingjie *et al*., 2008). In this study, the reference element is referred to the content of samples is almost the same as the Earth Crust content. According to Wedepohl (1995), the concentration of Al, Fe, Mn, Ca, K, Si, Ti and Zr in the Earth Crust were shown in Table 3.2.

Table 3.2: Concentration of metals in Earth's Crust

Metals	Concentration in Earth's Crust (ppm)
Aluminium(Al)	77440
Iron(Fe)	30890
Manganese(Mn)	527
Calcium(Ca)	29450
Potassium(K)	28650
Silicon(Si)	303480
Titanium(Ti)	3117
Zirconium(Zr)	237

(Source: Wedepohl 1995)

The formula to calculate EF is shown in equation 3.3 below:

$$EF(X) = \frac{(X/N)_{sample}}{(X/N)_{control}} \tag{3.3}$$

Where,

EF(X) = the enrichment factor for the metal X

(X/N_{sample}) = the ratio of the concentration of metal X to major metal N (Fe or Al) in a reference material

$(X/N_{control})$ = the ratio of the concentration of the metal x to major metal N (Fe or Al) in a reference material such as the control sample

Enrichment categories were listed in the Table 3.3.

Table 3.3: Enrichment factor categories

Enrichment factor (EF)	Enrichment factor (EF) Categories	Metals sources
EF < 2	no enrichment	Natural weathering process
2 ≤ EF < 5	Moderate enrichment	Anthropogenic sources
5 ≤ EF < 20	Significant enrichment	Anthropogenic sources
20 ≤ EF < 40	Very high enrichment	Anthropogenic sources
EF ≥ 40	Extremely high enrichment	Anthropogenic sources

(Source: Yuan *et al.* 2013)

3.5.2 Geoaccumulation (Igeo)

Geoaccumulation (Igeo) is used to determine the degree of contamination in the sediment by comparing the background value of every individual element with the analysed concentration element (Wang *et al.*, 2010). Igeo will be calculated using below equation. It expressed by Muller's equation as shown in equation 3.4:

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5B_n}\right] \tag{3.4}$$

Where,

C_n = element concentration in the analysed samples

B_n = the background value of the element in Earth's Crust

which is then compared to the 6 classes of Muller's classification as shown in Table 3.4.

Table 3.4: Muller's classification for Geoaccumulation index

Igeo Value	Class	Qualification of Sediment
≤ 0	0	Unpolluted
0–1	1	From unpolluted to moderately polluted
1–2	2	Moderately polluted
2–3	3	From moderately polluted to strongly polluted
3–4	4	Strongly polluted
4–5	5	From strongly polluted to extremely polluted
>5	6	Extremely polluted

(Source: Wang *et al.*, 2010)

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Concentration of metals in sediment of Besut and Dungun River

Table 4.1 presents the mean concentrations of the selected metals measured in the sediments from Besut and Dungun River. Based on the mean values of the metal concentrations, the metals follow in the following order, from the highest to the lowest; Si > Fe > Al > K > Ti > Zr >Ca> Mn for both river, respectively.

The data shows clearly that Si is the element that is highest in the samples. Meanwhile, Mn is the metal that was observed in the lowest mean concentration. For Si, the highest mean value is come from SB1 which is 386290 ppm. Meanwhile, Mn concentration was 2250 ppm. Mean concentration and concentration of metals in sediment from Besut and Dungun River is presented in Figure 4.1 and 4.2.

The concentrations of Al in both rivers were presented in Table 4.1 and Figure 4.3. As shown in figure 4.3, it was observed that the highest concentration of Al was in point SD1 (201000 ppm) and the lowest concentration was SB2 (1800 ppm) in Besut River. The mean concentration of Al in all sampling points was 143280 ppm as shown in Figure 4.1.

The source of Al concentration may be come from the activities that are found in SD1 (nearby road bridge) in which the source of Al may come from the discharge that is pass through the bridge. It also may due to the availability of Al in the sediment.

Table 4.1: Mean of metals concentration with standard deviation in sediment of all samples in Besut and Dungun River (ppm)

Sample ID	Al	Fe	Mn	Ca	K	Si	Ti	Zr
SB1	149000.00	154000.00	3400.00	9100.00	127000.00	479000.00	22200.00	19200.00
SB2	1800.00	5000.00	3100.00	8900.00	241000.00	401900.00	11100.00	3900.00
SB3	145000.00	312000.00	2100.00	8700.00	77800.00	344000.00	18400.00	3400.00
SB4	143000.00	249000.00	2000.00	19100.00	95000.00	351000.00	20200.00	3700.00
SB5	149000.00	237000.00	1500.00	8800.00	103000.00	375000.00	22000.00	5300.00
SB6	152000.00	258000.00	1800.00	8900.00	101000.00	378000.00	22900.00	7100.00
SB7	161000.00	283000.00	3000.00	18300.00	92200.00	353000.00	21900.00	2000.00
SD1	201000.00	701000.00	2900.00	9100.00	173000.00	462000.00	33500.00	20000.00
SD2	180000.00	200000.00	1100.00	6800.00	132000.00	406000.00	14900.00	2300.00
SD3	151000.00	296000.00	1600.00	14800.00	105000.00	313000.00	20400.00	5600.00
Mean	143280.00	269500.00	2250.00	11250.00	124700.00	386290.00	20750.00	7250.00
SD	52930.41	175949.01	790.57	4428.76	48848.72	52354.53	5825.85	6688.51

Note: SB= Besut River, SD= Dungun River

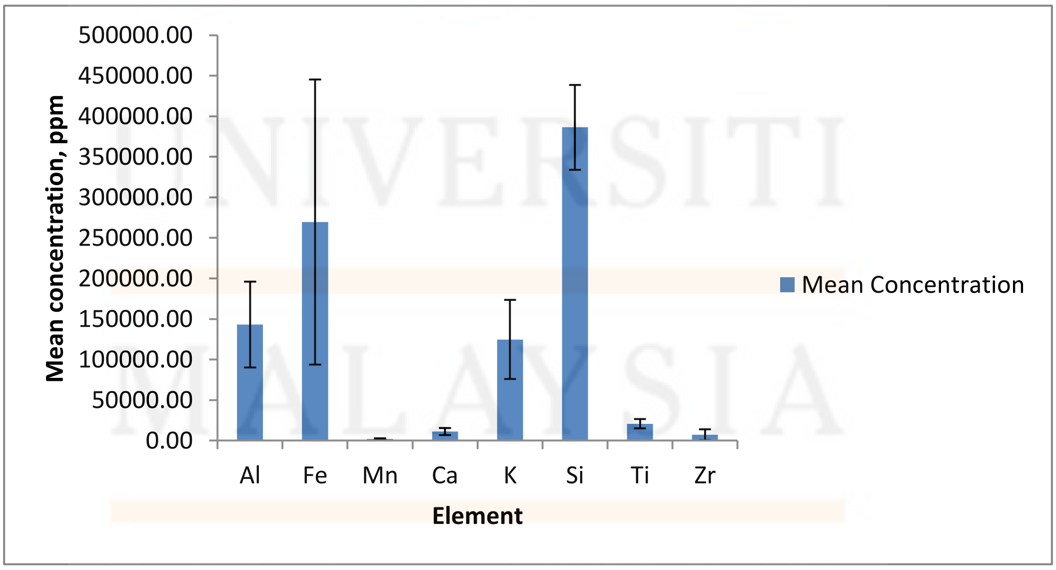

Figure 4.1: Mean concentration of metals in all sediment samples from Besut and Dungun River.

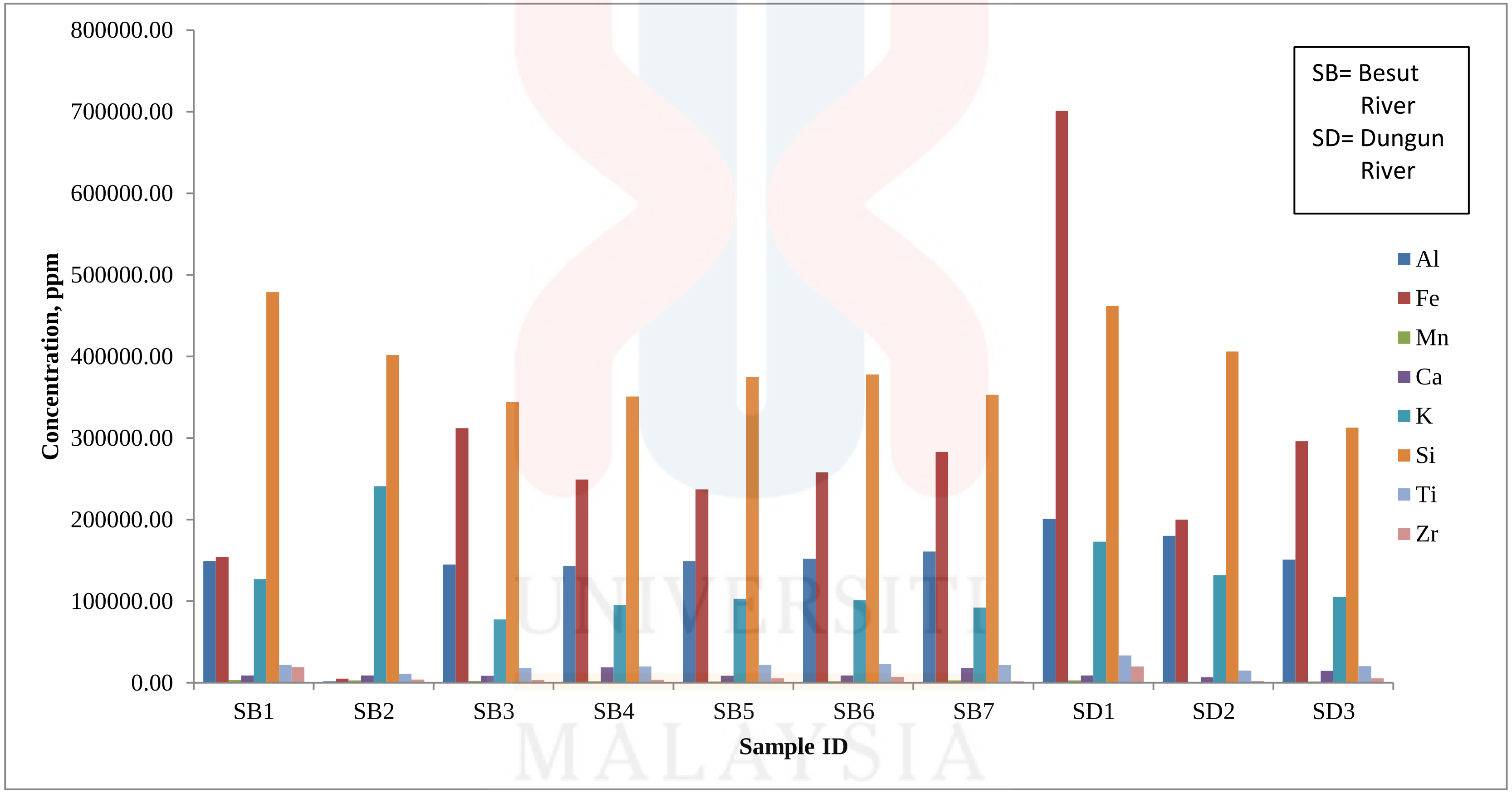

Figure 4.2: Concentration of each metal in all sediment samples from Besut and Dungun River.

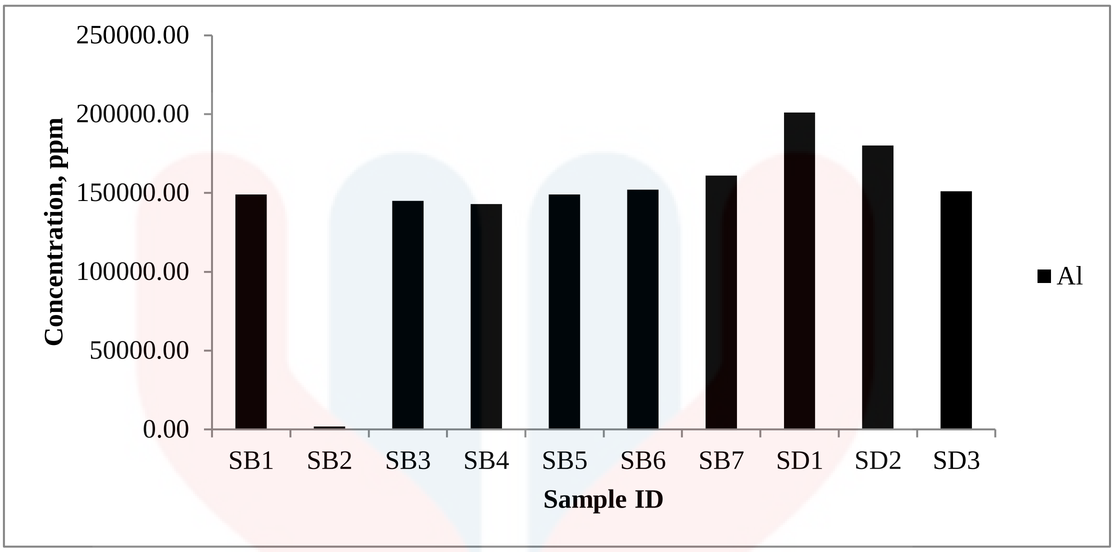

Figure 4.3: Concentration of Al in sediment from Besut and Dungun rivers.

The concentrations of Fe in both rivers were presented in Table 4.1 and Figure 4.4. As shown in Figure 4.4, the highest concentration of Fe were found in SD1 (701000 ppm) and the lowest concentration was in point SB2 (5000 ppm).

The mean concentration of Fe in all sampling points was 269500 ppm as shown in Figure 4.1. SD1 is located nearby with road bridge. High concentration of Fe may be because the sampling point was nearby the road bridge. The discharge of improper waste disposal that comes from the bridge may increase the availability of Fe in the sediment.

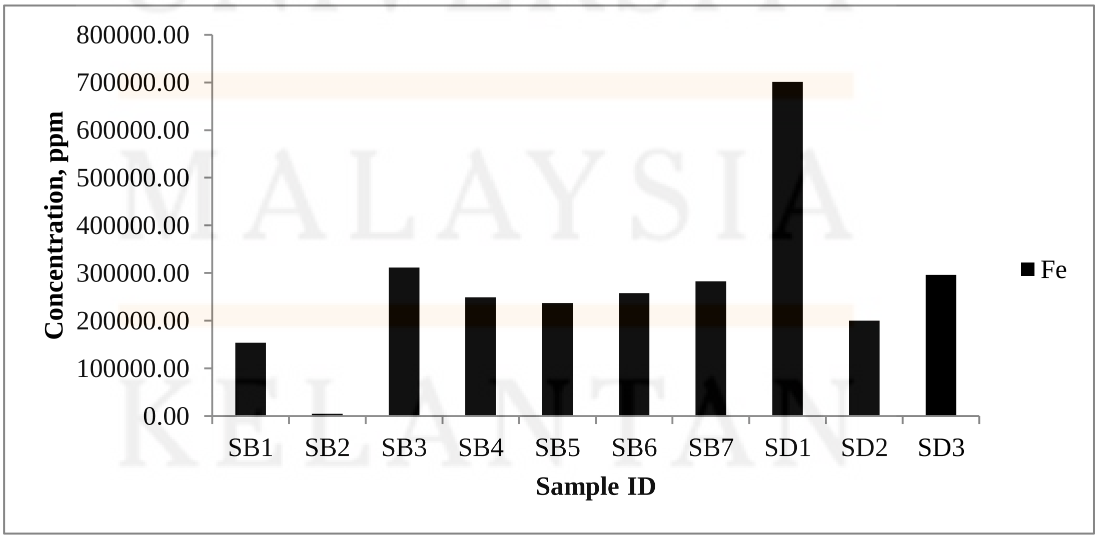

Figure 4.4: Concentration of Fe in sediment from Besut and Dungun rivers.

The concentrations of Mn in both rivers were presented in Table 4.1 and Figure 4.5. The highest concentration of Mn was found in SB1 (3400 ppm) for both rivers which the points was located at the watergate as shown in the Figure 4.5. Meanwhile, the lowest concentration was at point and SD2 (1100 ppm) and were located at boat parking area. The mean concentration of Mn in all sampling points was 2250 ppm as shown in Figure 4.1.

The occurrence of Mn was never a pure element in the nature. The contribution may come from the anthropogenic such as industrial activity and other waste discharge. The source of Mn concentration may be come from the activities that are found in watergate such as industrial effluent and discharge from nearby village. Besides, the discharge from the boat activity can be the source of Mn concentration in the sediment.

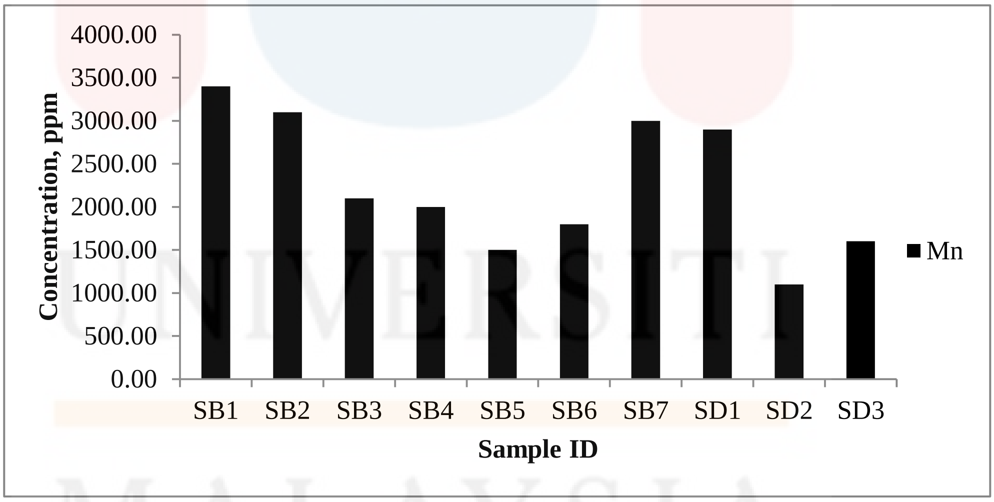

Figure 4.5: Concentration of Mn in sediment from Besut and Dungun rivers.

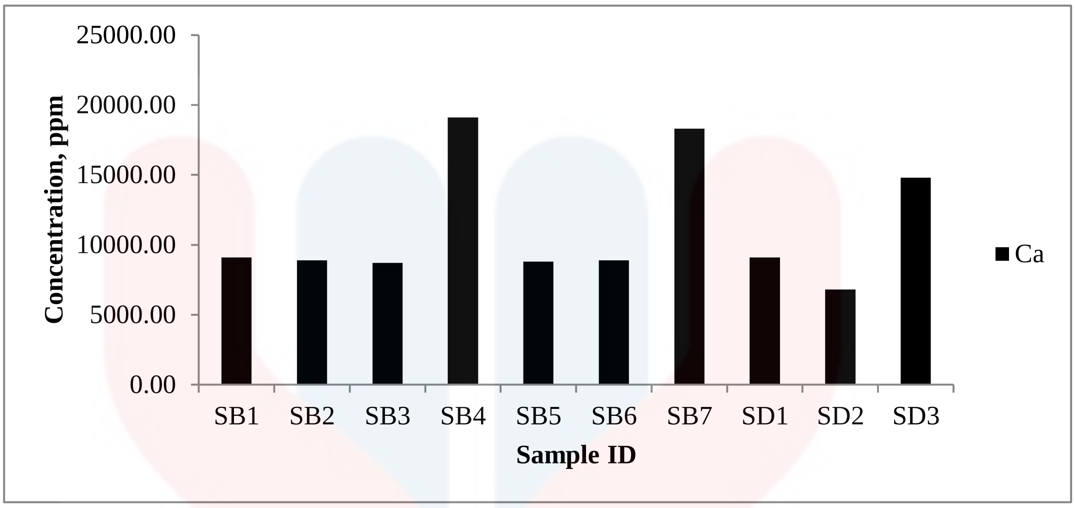

Figure 4.6: Concentration of Ca in sediment from Besut and Dungun rivers.

The concentrations of Ca in both rivers were presented in Table 4.1 and Figure 4.6. The mean concentration of Ca in all sampling points was 11250 ppm as shown in Figure 4.1. High concentration of Ca was in point SB4 (19100 ppm) and the lowest in SD2 (6800 ppm). The contribution of Ca might be due to the location of the point which is at the junction of the river. The source might come from the village in the riverside area and also the availability of the metals itself in the sediment of the river.

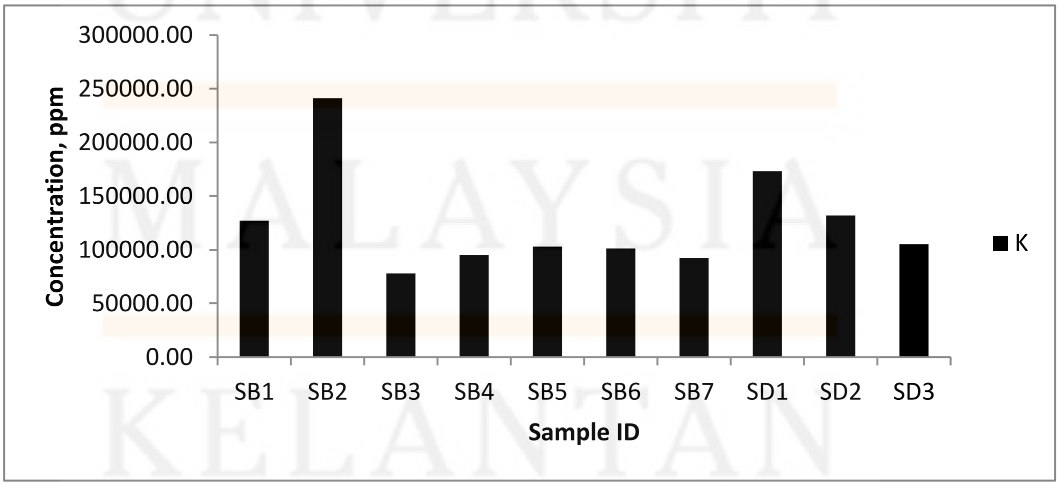

Figure 4.7: Concentration of K in Sediment from Besut and Dungun rivers.

The concentrations of K in both rivers were presented in Table 4.1 and Figure 4.7. As shown in figure 4.7, it was observed that highest concentration of K was at point SB2 (241000 ppm) and the lowest was at point SB3 which is 77800 ppm. The mean concentration of K in all sampling points was 124700 ppm as shown in Figure 4.1. SB2 point is located nearby the riverbank. The source may come from the activities that involved with cleaning activities. This is because K compound is commercially used by human in the form of soap and detergent and also agriculture use (Newton and Edgar, 2010).

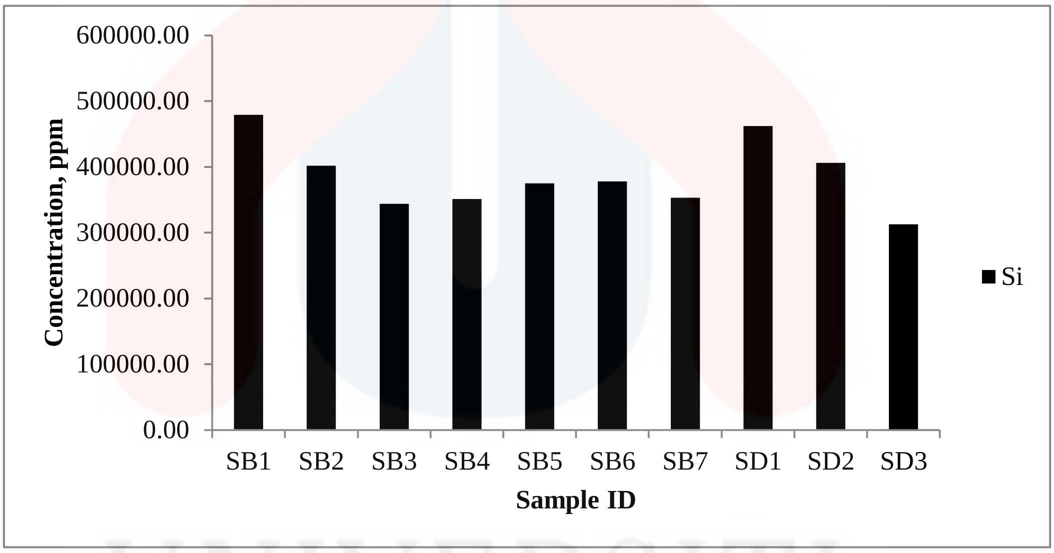

Figure 4.8: Concentration of Si in Sediment from Besut and Dungun rivers.

The concentrations of Si in both rivers were presented in Table 4.1 and Figure 4.8. The mean concentration of Si in all sampling points was 386290 ppm as shown in Figure 4.1. The highest concentration of Si was shown in point SB1 (479000 ppm) which is located at Watergate of the river while, the lowest Si concentration was at point SD3 (313000 ppm) which is located at a Jetty.

All points at Besut River shows that the concentration of Si found were in uniform data. Even though Si concentration is the highest among all points; Si metal was occurring naturally in the sediment of the river. This is due to the abundance amount of Si in the nature.

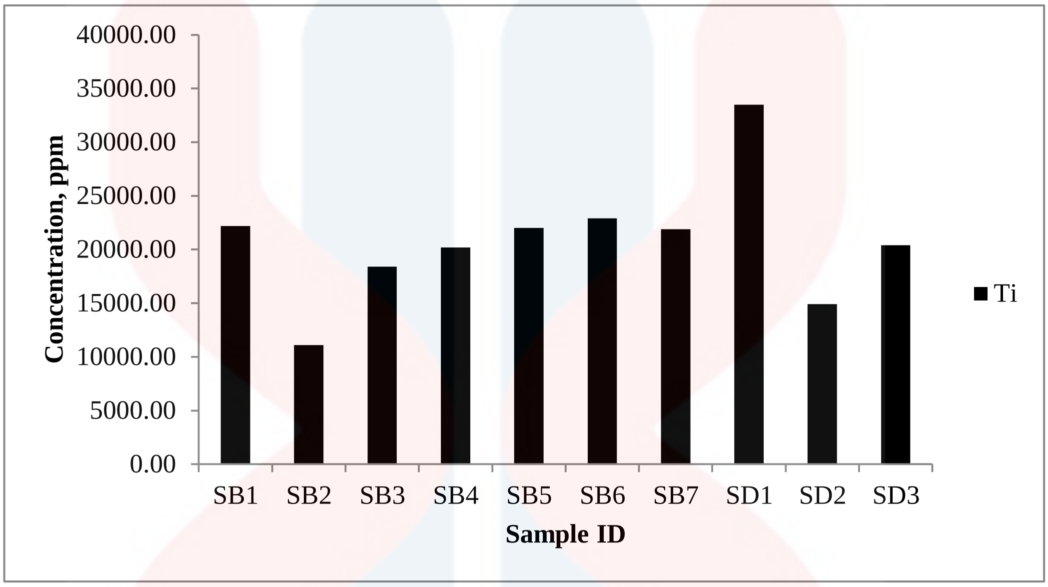

Figure 4.9: Bar graph of Ti in Sediment from all point in Besut and Dungun rivers.

The concentrations of Ti in both rivers were presented in Table 4.1 and Figure 4.9.As we can see in Figure 4.9, it was shown that the highest concentration of Ti was at point SD1 (33500 ppm) and the lowest at point SB2 (11100 ppm). The mean concentration of Ti in both rivers was 20750 ppm.

Point SD1 was located nearby road bridge and while SB2 at the riverbank. High concentration of Ti in this location might due to the samples were collected nearby with roadbridge. The source also may due to the discharge of waste from upstream and the metal may be accumulated in the sampling point which is nearby river bank.

The concentrations of Zr in both rivers were presented in Table 4.1 and Figure 4.10. The mean concentrations of Zr in both rivers were 7250 ppm. The highest concentration in Besut River was SD1 (20000 ppm) and the lowest was in SB7 (2000 ppm). The source of Zr in the sediment might be due to the activities nearby the road bridge such as disposal of domestic waste from the residential area since Zr examples in daily use were flash bulb and lamp filament (Newton and Edgar, 2010).

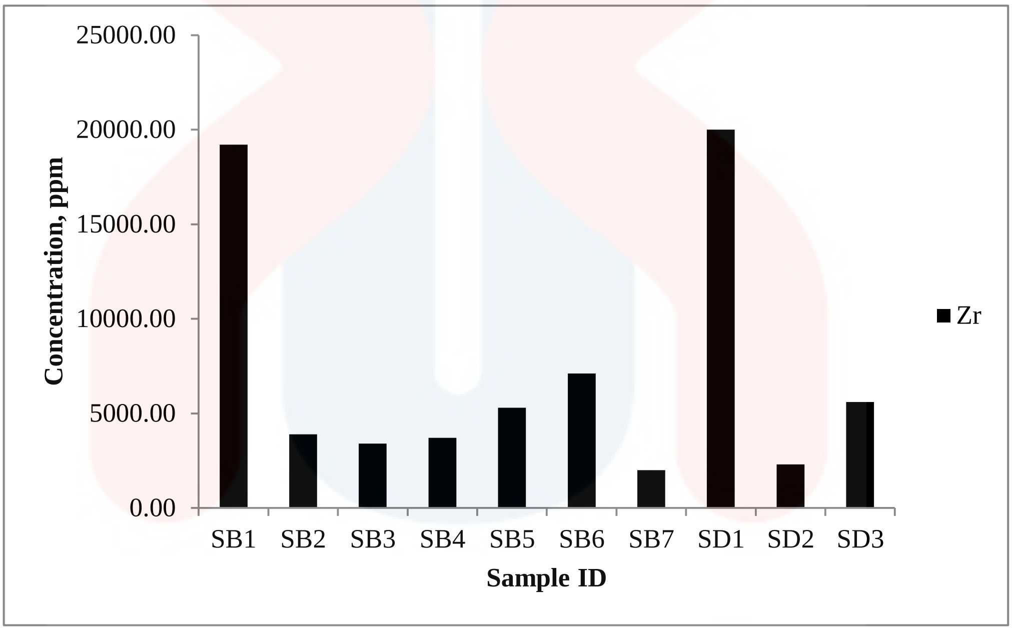

Figure 4.10: Concentration of Zr in sediment from Besut and Dungun rivers.

4.2 Comparison with other study

In this section, the results of the metals concentration in Besut and Dungun River were compared to the Malaysia and international studies to see whether the present study were comparable or not with other studies. However, the metals that from previous study was just only Al, Mn and Fe for the Malaysian studies whereas others will be compare to the international studies. The results were mentioned in Table 4.2.

Firstly, highest concentration of Al in Besut (161000 ppm) and Dungun (201000 ppm) were compared to several studies in Malaysia as shown in Table 4.2. Concentration of Al in EEZ of the East Coast of Peninsular Malaysia reported by Shaari *et al.* (2015) was lower than the concentration of both rivers. Also, both river were higher when compared with study done by (Sany *et al.*, 2013) at Port Klang. As for international studies, both rivers were higher than in Lake Kalimanci reported by Vrhovnik *et al.* (2013), East coast Tamilnadu as reported by Harikrishnan *et al.* (2016) and Southwest Nigeria River reported by Odukoya and Akande (2015).

The highest concentration Fe in Besut (312000 ppm) and Dungun (701000 ppm) were compared to several studies in Malaysia as shown in Table 4.2. In Malaysia studies, the concentration of Fe in Besut River is lower compared to EEZ of the east coast of Peninsular Malaysia as reported by Shaari *et al.* (2015). However, the concentrations of Fe in both rivers were higher than in Balok River, Pahang studies of Abdullah *et al.* (2015) and Johor Port: East Coast Peninsular Malaysia as stated by Abdullah *et al.* (2015). Both rivers also have higher concentration than in Mangrove sediment, Sarawak studies of Billah *et al.* (2014), in Port Klang, Selangor as reported in Sany *et al.* (2013) and Kelantan River as reported by Ahmad *et al.* (2009).

In international studies, Besut and Dungun River was considerably higher than in the study done by El-sorogy and Attiah, (2015) at Mediterranean Sea Coastal, Egypt, Southwest Nigeria River reported by Odukoya and Akande (2015), Lake Kalimanci reported by Vrhovnik *et al.* (2013), and East coast Tamilnadu as reported by Harikrishnan *et al.* (2016), respectively.

Highest concentration Mn in Besut (3400 ppm) and Dungun (2900 ppm) were compared to several studies in Malaysia as shown in Table 4.2. It was found that the concentration of Mn in Besut and Dungun River was higher than in EEZ of the east coast of Peninsular Malaysia, Johor Port: East Coast Peninsular Malaysia, Kelantan River, Port Klang, Selangor, Balok River, Pahang, and Mangrove sediment, Sarawak that was reported by Shaari *et al*. (2015), Abdullah *et al*. (2015), Ahmad *et al.* (2009), Sany *et al.* (2013), Abdullah *et al.* (2015) and Billah *et al.* (2014), respectively.

When compared to international studies, the concentration of Mn in East coast Tamilnadu as reported by Harikrishnan *et al.* (2016) was lower than in Besut and Dungun River. However, the concentration of Mn in Lake Kalimanci and Mediterranean Sea Coastal, Egypt were slightly higher than in Besut and Dungun River as reported by Vrhovnik *et al.* (2013) and El-sorogy and Attiah (2015). Moreover, the results were lower compared to southwest Nigeria River study done by Odukoya and Akande (2015).

Highest concentration Ca in Besut (1900 ppm) and Dungun (14800 ppm) were compared to several studies in Malaysia as shown in Table 4.2. In Malaysia, there is no studies of Ca concentration was found. When compared to international studies, the concentration of Ca in the concentration of Ca in Lake Kalimanci was higher than Besut and Dungun River as reported in Vrhovnik *et al.* (2013). However, concentration of Ca in East coast Tamilnadu and Southwest Nigeria River was lower than Besut and Dungun Rivers as reported by Harikrishnan *et al.* (2016) and Odukoya and Akande (2015).

Highest concentration K in Besut (241000 ppm) and Dungun (173000 ppm) were compared to several studies in Malaysia as shown in Table 4.2. In Malaysia, there is no studies of K concentration was found. In international studies, the concentration of K in Besut and Dungun River are higher than Lake Kalimanci, East coast Tamilnadu, Mediterranean Sea Coast, Egypt and Southwest Nigeria River as reported by Vrhovnik *et al.* (2013), Harikrishnan *et al.* (2016) El-sorogy and Attiah (2015) and Odukoya and Akande (2015).

Highest Si concentration in both rivers was 479000 ppm in Besut and 462000 ppm in Dungun. In Malaysia, there is no studies of Si concentration was found. In other countries studies, the concentration of Si in Besut and Dungun River are higher than Lake Kalimanci and East coast Tamilnadu as reported by Vrhovnik *et al.* (2013) and Harikrishnan *et al.* (2016).

As for Ti, there is no studies of Ti concentration was found in Malaysia. However, in the international studies, the highest concentration of Ti in Besut and Dungun Rivers are 22900 ppm and 33500 ppm respectively which is higher than East coast Tamilnadu, Lake Kalimanci and Southwest Nigeria River as stated by Harikrishnan *et al.* (2016), Vrhovnik *et al.* (2013) and Odukoya and Akande (2015).

Lastly, highest Zr concentration in both rivers area was 19200 ppm in Besut and 20000 ppm in Dungun. As for Zr, there is no studies of Zr concentration was found in Malaysia. In the international studies, the results showed that Zr concentration in Southwest Nigeria River was lower than in both rivers as mentioned in Odukoya and Akande (2015). However, Zr concentrations of both rivers were higher than in Mediterranean Sea Coastal, Egypt study done by El-sorogy and Attiah (2015).

Table 4.2: Comparison between the present study and previous studies in ppm

Location/ Element	Al	Fe	Mn	Ca	K	Si	Ti	Zr	References
Present Study									
Besut River	161000.00	312000.00	3400.00	19100.00	241000.00	479000.00	22900.00	19200.00	Present Study
Dungun River	201000.00	701000.00	2900.00	14800.00	173000.00	462000.00	33500.00	20000.00	
Malaysian studies									
EEZ of the east coast of Peninsular Malaysia	42300.00	756000.00	2700.00	-	-	-	-	-	Shaari *et al.* (2015)
Port Klang, Selangor	14724.00	6547.00	231.43	-	-	-	-	-	Sany *et al.* (2013)
Kelantan River	-	3860.00	394.00	-	-	-	-	-	Ahmad *et al.* (2009)
Balok River, Pahang	-	47028.00	155.90	-	-	-	-	-	Abdullah *et al.* (2015)
Mangrove sediment, Sarawak	-	14895.00	91.60	-	-	-	-	-	Billah *et al.* (2014)
Johor Port: East Coast Peninsular Malaysia	-	47000.00	1886.00	-	-	-	-	-	Abdullah *et al.* (2015)
International studies									
East coast Tamilnadu	23691.00	19302.00	367.65	12076.00	6179.00	174699.00	8460.00	-	Harikrishnan *et al.* (2016)
Southwest Nigeria River	26000.00	75000.00	1261000.00	2000.00	300.00	-	100.00	41000.00	Odukoya and Akande (2015)
Mediterranean Sea Coast, Egypt	-	198100.00	5700.00	-	640.00	-	-	3700.00	El-sorogy and Attiah (2015)
Lake Kalimanci	79000.00	66000.00	6000.00	38000.00	23000.00	235000.00	4000.00	-	Vrhovnik *et al.* (2013)

4.3 Enrichment factor (EF)

The contribution of anthropogenic sources can be described using assessment of enrichment factor (EF) to assess the level of contamination. The EF value of all metals was presented in Table 4.3.Enrichment Factor of Fe, Mn, Ca, K, Si, Ti, Zr in sediments from Besut and Dungun River were shown in the Figure 4.11.

Based on EF value of Fe, all points in Besut and Dungun River found Fe contamination having EF value higher than 2. The EF value that had been recorded in point SB1, SB4, SB5, SB6, SB7, SD2 and SD3 had been categorized into ($2 \leq EF < 5$) EF classes. This is due to that point had undertaken moderate enrichment of Fe contamination. However, EF value for Fe in points SB2, SB3, and SD1 was detected and categorized into ($5 \leq EF < 20$) EF classes. These classes were shown that the degree of Fe contamination were significant enrichment.

Based on EF value of Mn, only point SB2 was extremely high enrichment and had been classified into ($EF \geq 40$). The EF value that had been covered in point SB1, SB3, SB4, SB7 and SD1 had been classified into ($2 \leq EF < 5$) EF classes. This shown that points had moderate of Mn contamination. Other than that, SB5, SB6, SD2 and SD3 were categorized into ($EF < 2$) EF classes. The levels of Mn contamination in all points were considered from moderate enrichment to the extremely high enrichment.

Based on EF value of K, only point SB2 was extremely high enrichment and had been classified into ($EF \geq 40$). The EF value that had been covered in point SD1 and SB1 for Fe and had been classified into ($2 \leq EF < 5$) EF classes. This suggested that points had moderate enrichment of Fe contamination. Other than that, SB2, SB3, SB4, SB5, SB6, SB7, SD2 and SD3 were categorized into ($EF < 2$) EF classes. The levels of Fe contamination in all points were considered from moderate enrichment to the extremely high enrichment.

Based on EF value of Ti, all points in both river found is contaminated and having EF value higher than 2. The EF value that had been covered in point SB2 was classified into (EF $\geq$ 40) classes. This suggested that points had extremely high enrichment of Ti contamination. Other than that, all other points were categorized into (2 $\leq$ EF < 5) EF classes. The levels of Ti contamination in all points were considered from moderate enrichment to the extremely high enrichment.

Based on EF value of Zr, the EF value that had been covered in points SB1 and SB2 had been classified into (EF $\geq$ 40) EF classes. This suggested that point had extremely high enrichment of Zr contamination. Meanwhile, SB7 and SD2 had been classified in (2 $\leq$ EF < 5) EF classes and SB3, SB4, SB5, SB6, SB6, and SD3 in (5 $\leq$ EF < 20). The levels of Zr contamination in SB1 and were considered moderate enrichment to the significant enrichment. Other than that, only Zr in point SD1 having a very high enrichment of Zr and was categorized into (5 $\leq$ EF < 20) EF classes. The levels of Zr contamination in both rivers were considered from moderate enrichment to the extremely high enrichment.

The highest EF value of Si was found in SB2 and was classified into (EF $\geq$ 40) which indicate that extremely high enrichment of Si contamination. Besides Si, metal Ca was also found only in one point which is in SB2. The EF value of Ca was classified into (5 $\leq$ EF < 20) EF classes which indicate that significant enrichment of Ca contamination in SB2. However, all points beside SB2 found were no enrichment of Si and Ca. This result shows that the metals were comes from natural source in all points except for SB2.

According to Yuan *et al.* (2013), elements that having EF value more than 2 was come from anthropogenic activity. Therefore, the result showed that the contamination of K, Fe, Mn, Ti, and Zr which occurred in all points were mostly contributed by anthropogenic sources in or nearby each point such as run off from agricultural activities, boating activities and also domestic waste that goes into the river before being accumulated in the sampling points.

Thus, the origin of Si and Ca that had happened in all points for both rivers can be classified as naturally occurring process. The sources come from the natural occurrence and bedrock of the sediment.

Table 4.3: EF value in all points of Dungun and Besut River

Element	EF value									
	SB1	SB2	SB3	SB4	SB5	SB6	SB7	SD1	SD2	SD3
Fe	2.59	6.96	5.39	4.37	3.99	4.26	4.41	8.74	2.79	4.91
Mn	3.35	253.07	2.13	2.06	1.48	1.74	2.74	2.12	0.90	1.56
Ca	0.16	13.00	0.16	0.35	0.16	0.15	0.30	0.12	0.10	0.26
K	2.30	361.90	1.45	1.80	1.87	1.80	1.55	2.33	1.98	1.88
Si	0.82	56.97	0.61	0.63	0.64	0.63	0.56	0.59	0.58	0.53
Ti	3.70	153.21	3.15	3.51	3.67	3.74	3.38	4.14	2.06	3.36
Zr	42.10	707.96	7.66	8.45	11.62	15.26	4.06	32.51	4.18	12.12

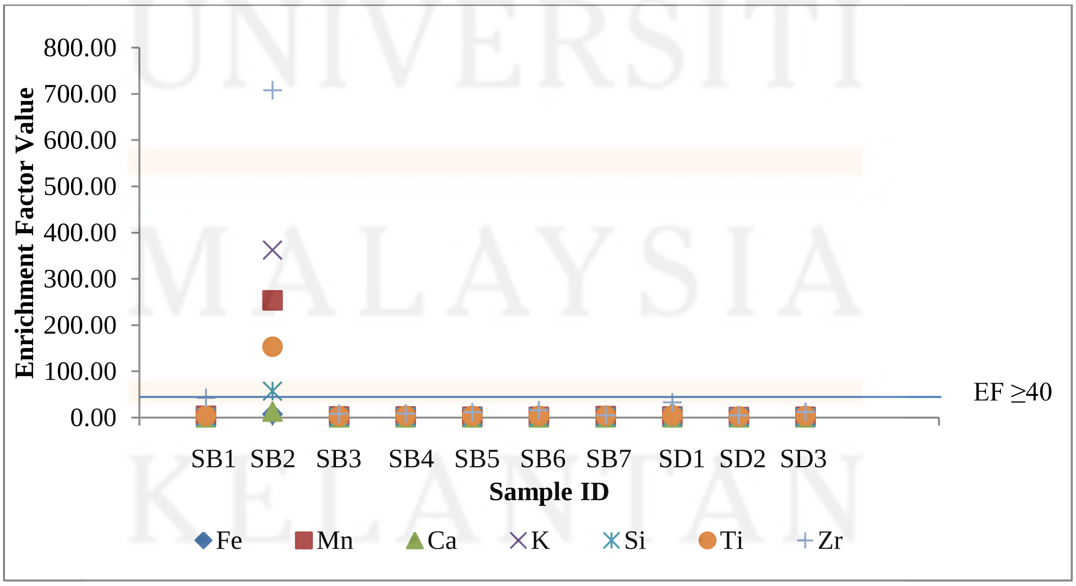

Figure 4.11: Enrichment Factor of Fe, Mn, Ca, K, Si, Ti, Zr in sediments from Besut and Dungun River

4.4 Geoaccumulation index (Igeo)

In order to evaluate the level of metals in sediment, Geoaccumulation index (Igeo) was used to compare the present study Igeo value with the Igeo classes so that sediment qualification can be determined. The calculated results of Igeo values were shown in Table 4.4 for both rivers. Igeo value of Al, Fe, Mn, Ca, K, Si, Ti, Zr in sediments from Besut and Dungun River were shown in Figure 4.12.

Aluminium showed unpolluted to moderately polluted in all points from both rivers and possess the Igeo class 1 whereas Igeo of Al in SB1, SB4, SB5, SB6, SB7, SD1, SD2, and SD3 were unpolluted to moderately polluted which possessed Igeo Class 1.

Next, the Igeo value of Fe showed strongly polluted possess the Igeo class 4 only for point SD1, whereas Igeo of Fe in SB3, SB4, SB5, SB6, SB7, SD2, and SD3 were moderately to strongly polluted (Igeo Class 3) and Fe in SB1 were moderately polluted (Igeo Class 2). Lastly, Fe in point SB2 was categorized as unpolluted and has the Igeo Class 0.

The Igeo value of Mn showed moderately to strongly polluted possess the Igeo class 3 for point SB1 whereas Igeo value of Mn in SB2, SB3, SB4, SB6, SB7, and SD3 were categorized as moderately polluted (Igeo Class 2). Meanwhile, Mn in SB5 and SD2 was from unpolluted to moderately pollute in Besut and Dungun River which have the Igeo class 0.

Potassium showed moderately polluted to strongly polluted possess the Igeo class 3 for points SB2 and SD1, whereas Igeo of K in SB1, SB4, SB5, SB6, SB7, SD2, and SD3 were moderately polluted (Igeo Class 2) and K in SB3 were unpolluted to moderately polluted (Igeo Class 1).

For Ti, point SB2, SB3 and SD2 were found as moderately polluted (Igeo class 2). The Igeo value of Ti was categorized as moderately to strongly polluted (Igeo class 3) for point SB1, SB4, SB5, SB6, SB7, SD1 and SD3.

Next, for Zr, SB1 and SD1 was qualified as extremely polluted and possess in class 6. The Igeo value of Zr in points SB2, SB3, SB4, SB5 and SD3 were showed strongly polluted possess the Igeo class 4 whereas in SB6, and were categorized as strongly polluted to extremely polluted (Igeo class 5). Other than that, Igeo value of Zr in SB7 and SD2 were classified as moderate to strongly polluted and were possess in class 3. Hence, Ca and Si consistently qualified as unpolluted (Igeo Class 0) in all points.

As we can see in the table 4.4, the highest Igeo value (class 6) was Zr which qualified as extremely polluted in SB1 and SD1. Through observation in these points, the source might come from the construction of small dam in point SB1. Meanwhile, in SD1, the point was nearby the road bridge. Since, transportation emission is one of examples of anthropogenic activity. The source of metals might come from it.

Table 4.4: Igeo value in all points of Besut and Dungun River

Sample ID/ Element	Measured Igeo value								Igeo class value							
	Al	Fe	Mn	Ca	K	Si	Ti	Zr	Al	Fe	Mn	Ca	K	Si	Ti	Zr
SB1	0	2	2	0	2	0	2	6	1	2	3	0	2	0	3	6
SB2	0	0	2	0	2	0	1	3	0	0	2	0	3	0	2	4
SB3	0	3	1	0	1	0	2	3	1	3	2	0	1	0	2	4
SB4	0	2	1	0	1	0	2	3	1	3	2	0	2	0	3	4
SB5	0	2	1	0	1	0	2	4	1	3	1	0	2	0	3	4
SB6	0	2	1	0	1	0	2	4	1	3	2	0	2	0	3	5
SB7	0	3	2	0	1	0	2	2	1	3	2	0	2	0	3	3
SD1	1	4	2	0	2	0	3	6	1	4	2	0	3	0	3	6
SD2	1	2	0	0	2	0	2	3	1	3	1	0	2	0	2	3
SD3	0	3	1	0	1	0	2	4	1	3	2	0	2	0	3	4

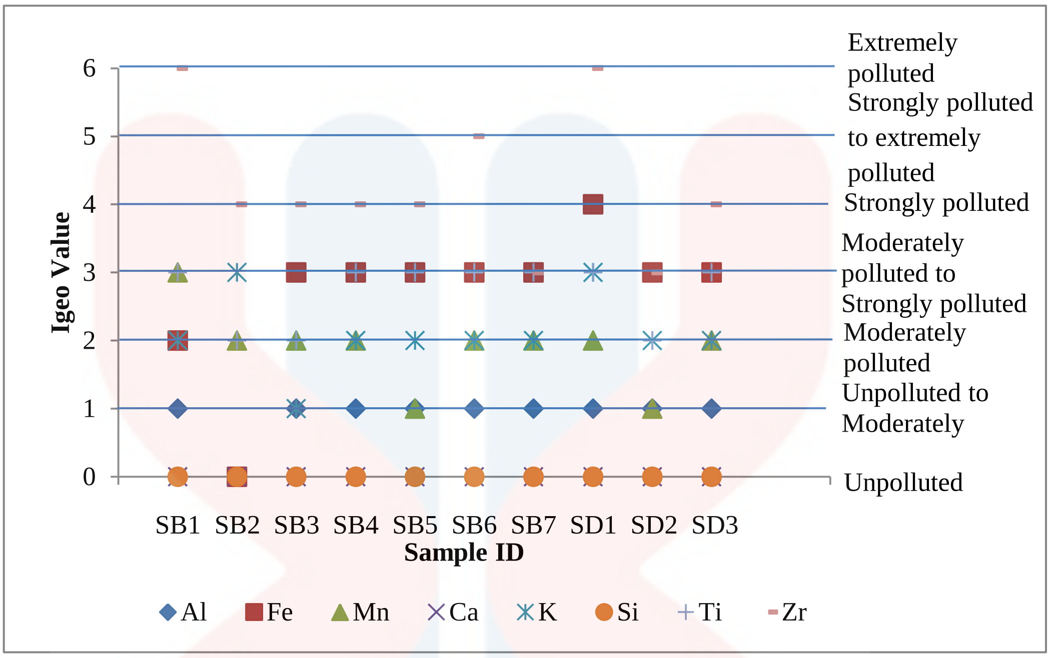

Figure 4.12: Igeo value of Al, Fe, Mn, Ca, K, Si, Ti, Zr in sediments from Besut and Dungun River

4.5 Statistical Analysis (Kruskal-wallis test)

Kruskalwallis test was performed to evaluate the difference of metal concentration in each river, Besut and Dungun River. The results showed that both rivers have significant different in all metals concentration found. In this study, the hypothesis was;

H_{null} = all metals concentration are the same

H_1 = all the metal concentration is different

For Besut River, the Kruskalwallis H test was 44.708, and the p value was 0.05. Since the p value for each metals concentration (0.03) was lower than the Kruskalwallis p value, the H_{null} was rejected and it was concluded that all the metals concentration was different.

As for Dungun River, the Kruskalwallis H test was 21.587, and the p value was 0.05. Since the p value for each metals concentration (0.03) in Dungun River was lower than the Kruskalwallis p value, the H_{null} was rejected and it was concluded that all the metals concentration was different.

The results showed that the metals concentration in each river were influenced and might come from different type of sources. This also supported the results from the enrichment factor (EF) and Geoaccumulation Index mentioned in the section 4.3 and 4.4.

Table 4.5: Kruskal Wallis test for Besut and Dungun River

Rivers	Kruskal Wallis Test	P value
Besut River	44.708	0.000
Dungun River	21.587	0.003

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

The mean metals concentration in Besut and Dungun River were ranged from 2250 to 386290 ppm. The factors that contributed to the variability of metals in sediment are due to their source distribution and transportation mechanism. Different distribution pattern shown in every point might have attributed the concentration of metals in sediment. The source such as non-point and point sources may cause the distribution pattern of metals in Besut and River.

Most of the pollution was suggested to come from point sources such as activities nearby to a river bank, water gate and road bridge that can be contributed to the accumulation of metals in the mentioned sampling point. Moreover, T junction of river and aquaculture activities also may become a source of metals pollution in both rivers.

When the EF value is calculated, the results showed that Fe, Mn, K, Ti, and Zr were come from anthropogenic sources and Al, Ca, and Si were come from natural sources. The results was supported by Geoaccumulation Index when it was found that both rivers were moderately polluted to extremely polluted by Fe, Mn, K, Ti, and Zr.

In conclusion, this study is used to update information on the level of metals concentration in both rivers which contributed water pollution. Authorities such as government and non-government agencies should be aware about these issues so that the rivers can be managed well. For example, enforcement of law to the industrial companies nears the rivers by paying penalties. Further study can be done time to time to see the changes in metal concentration in sediment for both rivers.

5.2 Recommendation

Based on the results and the conclusion from this study, it was clear that both rivers were polluted by certain metals that can harm aquatic life and human being.

To suggest, metals monitoring and research should be done from time to time so that the levels of metals in both rivers always in low level of concentration. For example, further study can be done by adding more parameter such as observing the water quality of the rivers and the level of metals in the aquatic life.

Another suggestion is the metals must be analysed using advanced instrument such as Atomic Absorption Spectroscopy (AAS). This is due to the accurate and precise results can be obtained from AAS. It is also easy to set up and run.

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APPENDIX A

Table A.1: Kruskal wallis test at Besut River

	Element	N	Mean Rank
Concentration	Al	7	34.36
	Fe	7	41.29
	Mn	7	5.50
	Si	7	53.00
	Ti	7	26.29
	K	7	35.14
	Ca	7	19.43
	Zr	7	13.00
	Total	56	

Test Statistics[a,b]

	concentration
Chi-Square	44.708
df	7
Asymp. Sig.	.000

a. Kruskal Wallis Test

b. Grouping Variable: element

Table A.2: Kruskal wallis test at Dungun River

	Element	N	Mean Rank
concentration	Al	3	17.00
	Fe	3	20.67
	Mn	3	2.33
	Si	3	22.00
	Ti	3	10.67
	K	3	14.33
	Ca	3	7.00
	Zr	3	6.00
	Total	24	

Test Statistics[a,b]

	concentration
Chi-Square	21.587
df	7
Asymp. Sig.	.003

a. Kruskal Wallis Test

b. Grouping Variable: element

APPENDIX B

Figure B.1: 4cm of sediment was collected by using Ekman grab

Figure B.2: The sediment samples is kept into clean polythene bag until the further analysis

Figure B.3: Sediment samples were categorized before was put in Drying Oven for 105°C (24 hours).

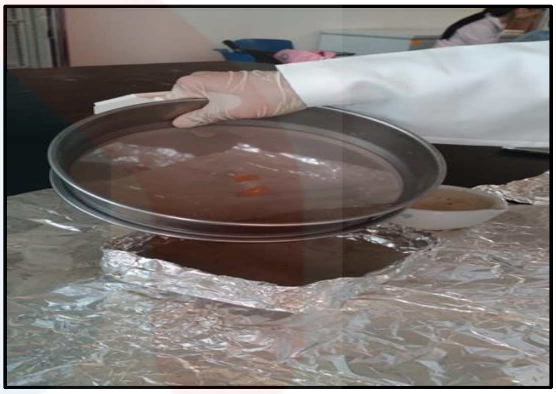

Figure B.4: The samples were sieved by using 75µm sieve

Figure B.5: The sediment samples were analysed by XRF instrument