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THE SPATIAL DISTRIBUTION OF Al, Fe, Cu, Cd AND Pb IN THE SURFACE SEDIMENT OF BRUNEI BAY, BORNEO DURING THE SOUTHWEST AND NORTHEAST MONSOONS

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Abstract: A geochemical assessment of Al, Fe, Cu, Cd and Pb was carried out for the surface sediment of Brunei Bay. Samples were collected during July 2013 and January 2014 representing the Southwest and Northeast Monsoon respectively. Fe, Pb, Al and Cu were high in July 2013 samples whereas Cd had a similar range of concentration during both sampling periods. Cd showed no particular spatial distribution pattern whereas Fe and Pb were highly distributed at the transect stations compared to the coastal stations due to the river flushed out. In contrast, Al and Cu were uniformly distributed between all stations. The Al, Fe and Pb in Brunei Bay sediment were dominantly bound to the mineral fraction which is also an indicator of natural sources whereas Cu was more dominant in the ion exchangeable fraction which is the most bioavailable fraction. The risk assessment code (RAC) indicated that Al has no risk (RAC<1%) whereas Fe and Pb showed a low risk (RAC<10%) to the benthic organisms. However, Cu presented a very high risk (RAC>50%) in both sampling periods. Even though the RAC showed some risk of these metals on the ecosystem, the geoaccumulation index indicated no serious pollution by these metals. Additionally, the surface sediment of Brunei Bay was dominated by silt and very fine sand. The apparent changes in metals distribution and metals fractionation during January 2014 corresponded to the effects of monsoons. Port activities, aquaculture, fisheries, palm oil and paper mill industries are the potential sources of metals in the Brunei Bay ecosystem.

Keywords: Geochemistry of metals, Northeast and Southeast monsoons, Brunei Bay, Southeast South China Sea.

Introduction

Metals are pollutants that have been discussed due to the rapid growth of urbanization and industrialization. They are classified as inorganic pollutants which are typically persistent and hazardous since they are commonly bioaccumulated and biomagnified in the organisms (Todd *et al.*, 2010; Min *et al.*, 2013). In the marine system, they are commonly distributed amongst the water column, biological factors, and sediment bed. The surface sediment appears to be an ultimate sink, which can be a sensitive indicator for the contaminants monitoring in an aquatic system (Kamaruzzaman *et al.*, 2006).

On the surface sediment, dissolved metals are entrapped in the pore water system due to the sulfide pool and humic acid content will be transported downwards to the anoxic subsurface sediment (Charriau *et al.*, 2011). In the meantime, metals attached to the sediment particles will either accumulate in the sediment, re-suspend into the water column or scavenged by marine organisms (Martino *et al.*, 2002). Metals could transform into numerous species due to dissolution, precipitation, absorption and complexation which are affecting their behavior and bioavailability (Tria *et al.*, 2007). Commonly in the sediment, metal ions form bonds and complexes with carbonates, manganese oxide, iron oxide, organic matter and mineral particles since these compounds could control the mobility, toxicity, binding behavior of metal ions as well as the availability in terms of the biological point of view (Filgueiras *et al.*, 2004; Charriau *et al.*, 2011).

Based on SCOPUS online database since 1968 to 2012 (www.scopus.com), 654 documents were published on the biogeochemistry namely on the metals, rare earth elements and their isotopes. Several scientific articles on the research trend in the bay area were focused on the distribution pattern, pollution status, the source identification as well as contamination of elements in relation to the economic growth and human impact (Meng *et al.*, 2008; Hosono *et al.*, 2010a; Wang *et al.*, 2010a; Hosono *et al.*, 2010b; Wang *et al.*, 2010b; Hosono *et al.*, 2011; Gao & Chen, 2012; Liu *et al.*, 2012; Duo *et al.*, 2013). The database highlighted five articles found in Brunei Bay area were described on proboscis monkey, oil spill issues, dugong distribution and sediment deposition into the Champion Delta (Yeager, 1995; Rahmat & Yusof, 2005; Wing, 2005; Rajamani & Marsh, 2010; Lambiase & Cullen, 2013).

A research activity was carried out in the east region of Brunei Bay by Universiti Malaysia Sabah (Saleh, 2007) highlighted the need of environmental regulation enforcement in order to protect, conserve and improve Brunei Bay ecosystem. In the meantime an article by Bengayawan (2007) in Brunei Times stated that the Asean Regional Centre for Biodiversity Conservation (ARCBC) warned that the rivers and mangroves that drains into Brunei Bay are slowly being destroyed by untreated domestic sewage, sewage effluent, urban waste run-off and worsening of mangrove cutting. Other than that, the ARCBC noted that the indiscriminate dumping of solid waste into the streams and rivers, especially in the Brunei-Muara District, is a problem that requires serious attention.

The present study is purposely to define the current status of metals in the surface sediment of Brunei Bay, Borneo in order to obtain a geochemistry baseline data. The intention of this study is to elucidate the seasonal impact on metals distribution and fractionation during the northeast and southwest monsoon since Brunei Bay is located within the South China Sea area as this area have been experiencing two monsoonal changes.

Materials and Methods

Study Area and Sample Collection

Brunei Bay is located at Northwest of Borneo Island which bordered by Sabah, Sarawak, Brunei and Labuan Island (Figure 1). This coastal water system is characterized by mild-slope bottom and wide mouth facing towards the Southeast of South China Sea. Mangroves, seagrass bed, crocodile, dolphin, turtle, sea-cow, giant fresh prawn and others are commonly found in the area. In recent years the increasing human activities such as palm oil plantation, paper mill, tourism, water villages, ship and water transportation, logging, net fishing and aquaculture are discovered along the coastal region of this bay (Mustafa, 2007). The report says the increasing of demand in the need of living resources for food and business together with the rapid growth of industrialization within and surrounding the Brunei Bay resulting the impact on the bay ecosystem.

Surface sediment samples were collected during July 2013 and January 2014 sampling periods represented the Southwest and Northeast Monsoon respectively. Totaling to 49 stations were selected off Malaysian waters and these stations are divided into 3 sections; coastal, transect and Labuan stations (Figure 1). Surface sediment was collected using the Ponar grab. Few scoops of the sediment (approximately within a surface and 5 cm deep) were sampled using an acid-washed plastic scoop. Samples were then kept in a double polyethylene zip-lock bag under 4 °C and then transported to the laboratory.

Total Digestion

Sediment samples were dried 60 °C under oven heating and mildly crushed using mortar and pestle. Approximately 0.05 g of bulk sediment was digested with a mixture of concentrated Suprapur acids (65% nitric acid, 37% hydrochloric acid and 48% hydrofluoric acid) in a Teflon bomb under oven heating. The digested sample was then diluted to 10 ml with deionized water and kept at 4 °C prior sample analysis

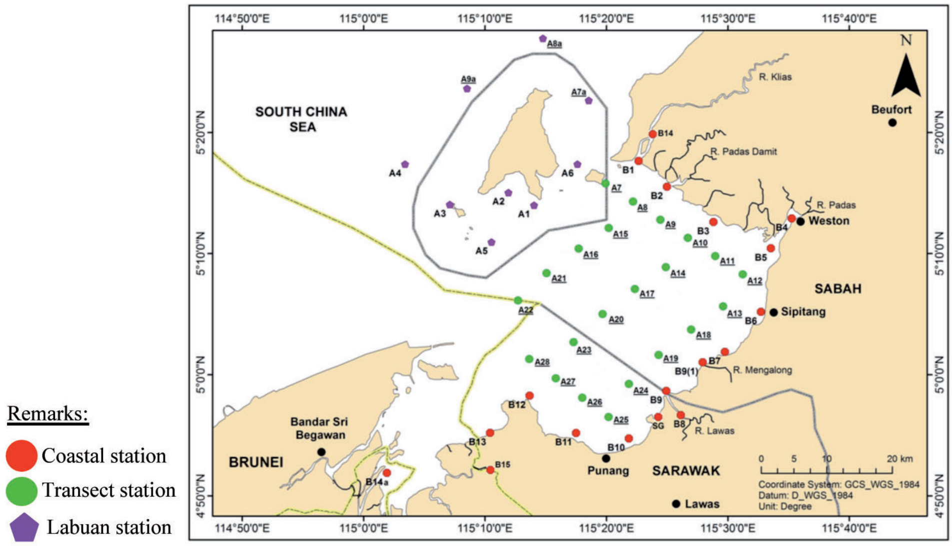


Figure 1: Sampling stations in Brunei Bay, Borneo

using Inductively Coupled Plasma - Mass Spectrometer Perkin Elmer ELAN 9000 (ICP-MS). Blanks were measured to check the metal contamination whereas total digestion method was validated using standard reference material NIST 1646a (Standard Reference Material – Estuarine Sediment from National Institute of Standards and Technology). Concentration of metal in the sediment was calculated using the formula below (Morford *et al.*, 2001; Bidai, 2012; Bidai *et al.*, 2016):

$$\mu g/g = \frac{X \times \text{Stock volume} \times \text{Dilution factor}}{\text{Sample weight (g)}}$$

Remarks:

X: Raw data reading by ICP-MS (µg/L).
Stock volume: Top-up volume after digestion process.

Sediment Sample for Sequential Extraction Analysis

Metals fractionation in the surface sediment of Brunei Bay was carried out according to the procedure by Tessier *et al.* (1979). The method consists of 5 different fractions namely:

(i) Ion-exchangeable fraction: The metal ions which are readily oxidized once they come in contact with air. These species are specifically adsorbed and likely to be released when the ionic composition of water is changed

(ii) Bound to carbonate fraction: Metal fractions that are precipitated or co-precipitated with the carbonates and this phase is also prone to the pH changes.

(iii) Bound to Fe-Mn oxides fraction: The bonds between these oxides with metal ions provides a strong scavenging efficiency for the metals. This metal fraction can be dissolved by changes in the redox potential as these metal species are unstable under reduced condition.

(iv) Bound to organic matter fraction: Refers to the oxidisable phases as the organic material may release the soluble metal ions under oxidizing conditions. Metal ions may be associated through the complexation or bioaccumulation process with various forms of organic material such as living organisms, detritus or coatings on mineral particles.

(v) Residual fraction: Metal ions associated with the mineral matrix.

All collected fractions were then kept at 4 °C prior to analysis using ICP-MS.

Total Organic Carbon in Surface Sediment

The total organic carbon (TOC) analysis was adapted from Walkley and Black (1934). Samples were digested in the water bath heating and titrated. The percentage of TOC in sediment sample was calculated as follows:

$$\% \text{ TOC} = \frac{(V_1 - V_2)}{\text{Sample weight (g)}} \times 0.003 \times 100$$

Remarks:

V_1: Volume of $K_2Cr_2O_7$ used (mL).
V_2: Volume of $FeSO_4$ used (mL).
0.003: The quantity of carbon in 10 mL of potassium dichromate.

Blank was checked for quality control whereas glucose was used to verify the recovery of the procedure. The percentage of TOC in glucose was 36 $\pm$ 5% and percentage recovery of TOC was 38% which is 105.5% of the standard value.

Particle Size Analysis for Surface Sediment

In order to define the size of surface sediment particles, a dry size method by Ellis and Stone (2006) was adopted, analyzed using Particle Size Analyzer Malvern Mastersizer 2000 and computed using the Moment Method. The obtained data was used to classify the sediment particle size into sand, silt or loam by using Wentworth particle size classes (Table 1) as follows:

Table 1: Wentworth classes of particle size

Wentworth size class		Phi (Φ)	Micrometers (μm)
GRAVEL		-12.0	
	Boulder		
		-8.0	
	Cobble		
		-6.0	
	Pebble		
		-2.0	
	Granule		
		-1.0	
SAND	Very coarse sand		
		0.0	
	Coarse sand		
		1.0	500
	Medium sand		
		2.0	250
	Fine sand		
		3.0	125
	Very fine sand		
		4.0	63
SILT	Coarse silt		
		5.0	31
	Medium silt		
		6.0	15.6
	Fine silt		
		7.0	7.8
	Very fine silt		
		8.0	3.9
MUD	Clay		
		14.0	0.06

Results and Discussion

A summary of total metals in the surface sediment of Brunei Bay, Borneo is shown in Table 2.

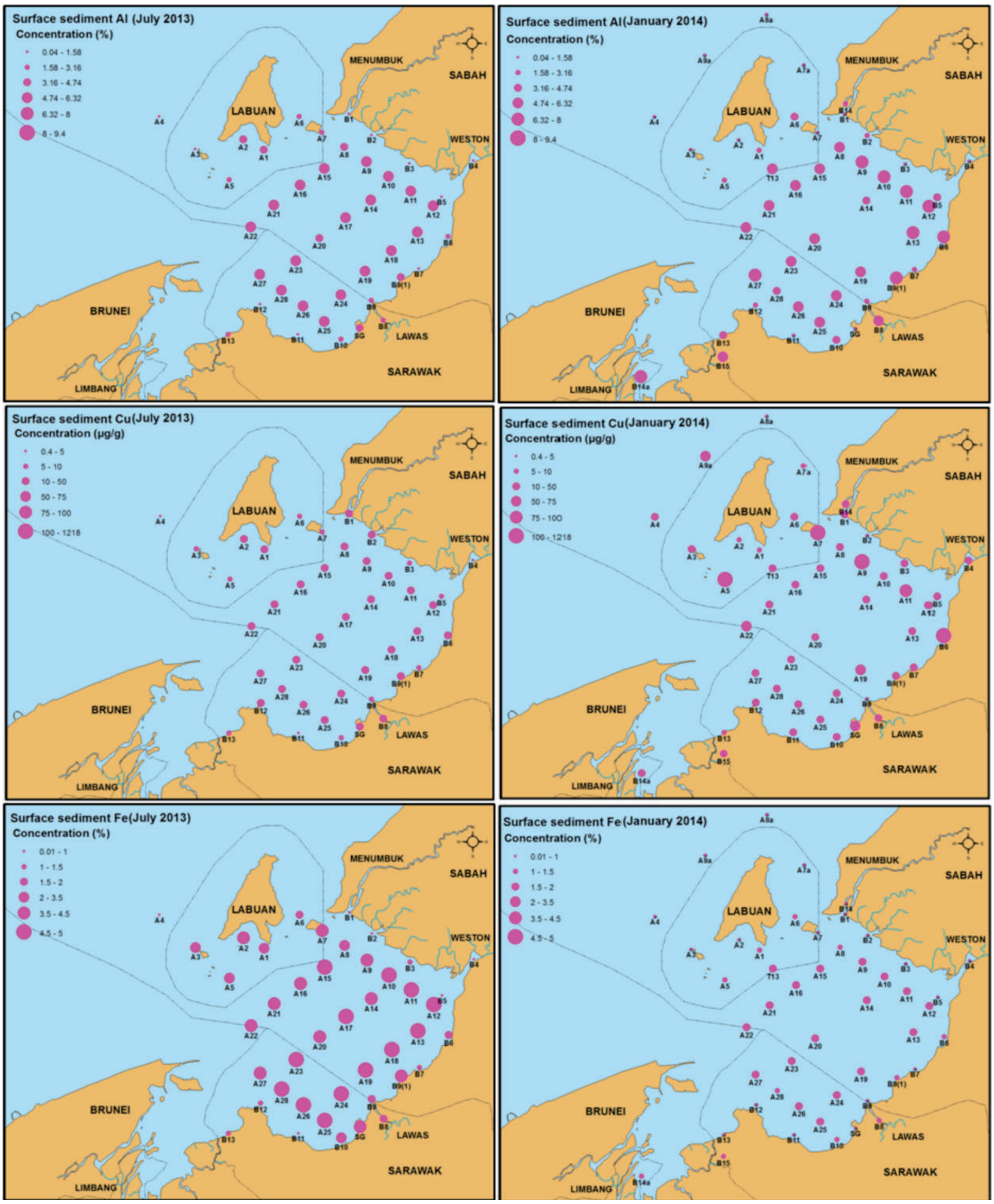
The total concentration of metals (Table 2) and the spatial distribution (Figure 2) showed that Al was high during January 2014 whereas Cd, Fe, Cu and Pb showed higher concentration during July 2013. Al, Fe, Cu and Pb (Figure 2) were proved an accumulation in the bay area as a higher range of concentration are shown in transect stations as compared to coastal and Labuan stations. In the meantime total concentration of Cd showed higher range in coastal stations compared to other stations. In January 2014 sampling period, Brunei Bay have been experiencing northeast monsoon (wet season) as the strong current driven by this monsoon changes the sediment sorting and lead to a lower range of metals in the surface sediment which is in agreement with Zwolsman *et al.* (1993) and Bidai *et al.* (2016). In the meantime during July 2013 (dry season/ southwest monsoon), metals were settled down into the surface sediment and undisturbed due to the weak water current (Owen & Balls, 1997; Adiana *et al.*, 2014).

The bar graphs in Figure 3 show comparison of the average total concentration of metals in this study with the continental crust value adopted from Mason and Moore (1982). Overall, the concentrations of metals found in the surface sediment of Brunei Bay were lower than those found in the continental crust except for Pb. The outlined Pb in the continental crust is 13 ppm but the average Pb in the present study was exceeded approximately 288% and 135% during July 2013 and January 2014 respectively suggesting the aerosol input of Pb from boat and ship traffic, land-based transport and industrial activities surround and within Brunei Bay.

The sequential extraction analysis showed that the sum of all fractions is in the agreement with the total concentrations with satisfactory recoveries (90% - 110%) except for Cd. The recovered sum of the 5 fractions for Cd was lower than 80%. Bar graphs in Figure 4 showed

Table 2: Mean, standard deviation and range (in parentheses) concentration of metals in surface sediment for the different sampling periods

Element (concentration)	Al (%)	Fe (%)	Cu (µg/g)	Cd (ng/g)	Pb (µg/g)
July 2013	3.72 $\pm$ 2.04 (0.44 - 6.08)	3.25 $\pm$ 1.56 (0.64 - 4.96)	15.8 $\pm$ 9.5 (1.80 - 46.2)	128 $\pm$ 91 (46 - 327)	37.4 $\pm$ 18.7 (8.9 - 89)
January 2014	4.01 $\pm$ 2.33 (0.004 - 7.72)	1.11 $\pm$ 0.59 (0.01 - 1.93)	33.0 $\pm$ 38.5 (2.1 - 170)	39 $\pm$ 22 (1 - 142)	17.8 $\pm$ 8.9 (3.7 - 30.9)



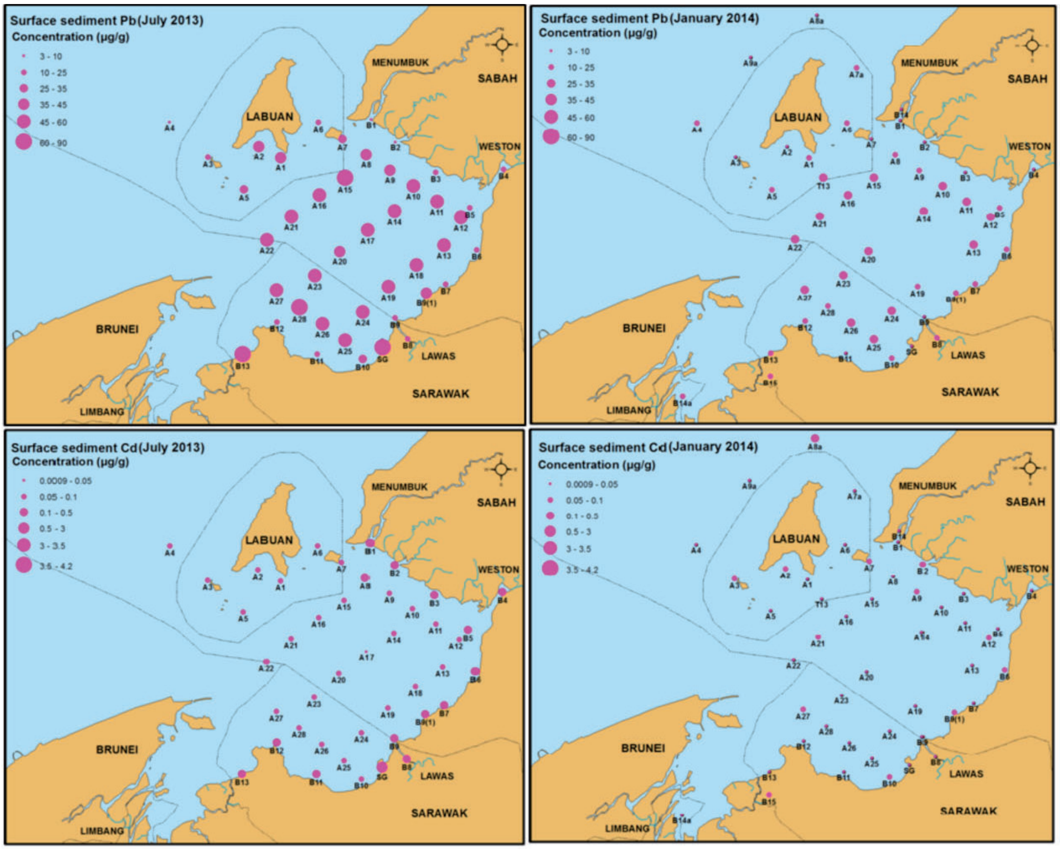


Figure 2: Distribution of metals in the surface sediment of Brunei Bay, Borneo

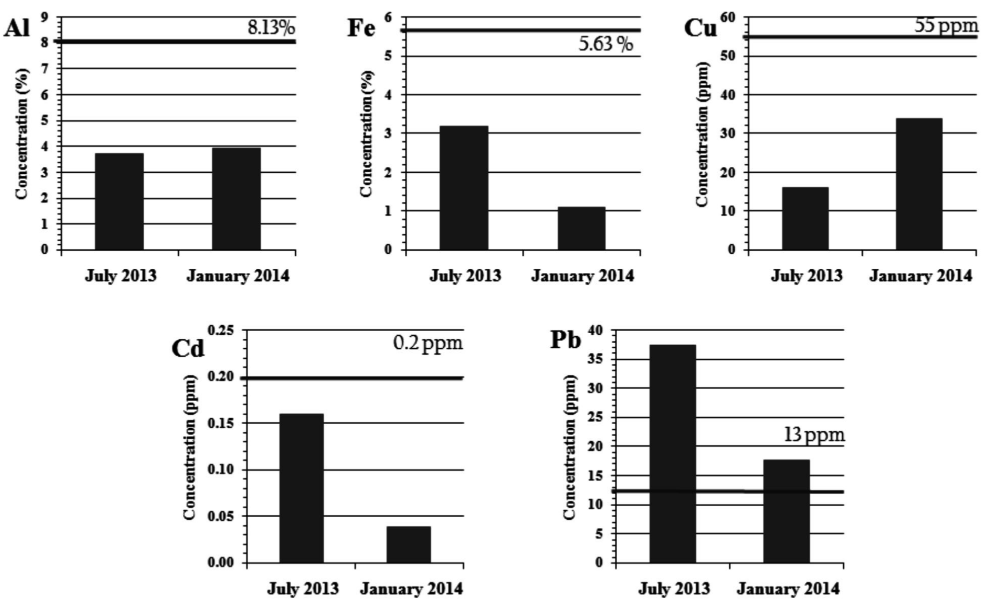


Figure 3: Comparison of metals in the present study with the continental crust value

the percentages of each fraction for Al, Fe, Pb and Cu in surface sediment of Brunei Bay. Al was dominantly bound to the sediment particle sizes as more than 98% of Al was bound with the residual fraction during both sampling periods. The residual fraction is the most stable and least mobile fraction compared to other fractions (Cuong & Obbard, 2006), therefore Al is not bioavailable and bioaccumulated in the Brunei Bay sediment. Fe and Pb were prevalent in the residual fraction but approximately 30.2% to 31.7% of these metals were bound with the organic matter fraction. Fe was high in association to organic matter at stations with low salinity which is in agreement to sediment in Dutch estuaries (Rijkenberg *et al.*, 2006) whereas high correlation of Pb with the organic matter fraction was due to strong attraction of Pb towards organic matter and sulphides (Wang *et al.*, 2010b). While, below than 30% of Fe and Pb were bonded to the Fe-Mn oxides fraction and less than 13% bounded to the carbonate fraction. Apart from that, Fe and Pb bound to minerals, Fe-Mn oxides and carbonate fractions were in agreement to those found by Copaja *et al.* (2014) in Chaopa River basin, Qiao *et al.* (2013) in Shantou Bay and Yuan *et al.* (2004) in the East China Sea. Cu was predominant with ion-exchangeable fraction and less bound with other fractions. According to Copaja *et al.* (2014), the ion-exchangeable fraction is the most labile fraction which is easily uptake by the benthic organisms. Overall, throughout both sampling periods, the percentage of metals bound to ion-exchangeable, carbonate, Fe-Mn oxides and organic matter increased during January 2014 as compared to July 2013 sampling period. The disturbance caused by the strong currents during January 2014 sampling period changes the sediment sorting, redox, salinity and pH of the Brunei Bay surface sediment (Table 3).

The changes in these parameters cause the increased of metals bound to the non-residual fractions of the surface sediment Brunei Bay. According to Qiao *et al.* (2013), Canuto *et al.* (2013) and Gao *et al.* (2010), the strong bonding of metals with the non-residual fractions are due to a strong affinity of metals with humic acids, oxidation and reduction by microbial activities and changes of pH, redox and salinity in the sediment. The percentage of loam during January 2014 increased almost twice of the July 2013 (Table 3). According to Kaufmann and Cleveland (2008), loam sediment type consists of 40% of sand (< 63 μm), 40% of silt (> 2 μm) and 20% of loam (< 2 μm). The increasing of loam caused the increasing of clay content as well as the cation exchange capacity (CEC) in January 2014 samples. Since the high value of CEC showed more capacity to hold the hydrogen ions, it reduces the redox and pH value (de Jonge & de Jonge, 1999). Therefore the sorption rate between metals with the non-residual fraction was increased during January 2014 compared to July 2013 sampling period.

Table 3: Mean and standard deviation of physical parameters in surface sediment for the different sampling periods

Parameter/Sampling	July 2013	January 2014
pH	7.53 ± 0.20	7.20 ± 0.78
Redox (mV)	-172 ± 99	-147 ± 90.5
Salinity (ppt)	32.8 ± 0.85	33.9 ± 0.78
TOC (%)	4.67 ± 0.92	4.75 ± 1.06
Sandy loam (%)	22.58	5.71
Silty loam (%)	64.52	71.43
Loam (%)	12.90	22.56

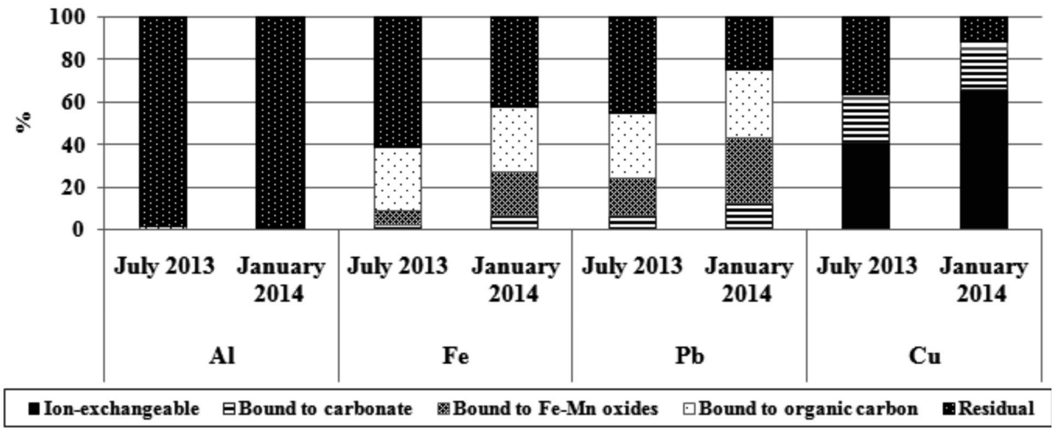


Figure 4: Metals fractionation in surface sediment of Brunei Bay

In order to assess the environmental risks and estimate the possible damage cause by polluted sediments to benthic organisms, a risk assessment code analysis (RAC) was applied based on the ion-exchangeable and carbonate bound fraction, represent the weakly bonded metals and the most bioavailable fractions (Jones *et al.*, 2008; Wang *et al.*, 2010b; Sundaray *et al.*, 2011). The RAC classification is described in Table 4 as below (Wang *et al.*, 2010b):

$$RAC = \frac{\text{sum \% (Exchangeable bound + Carbonate bound)}}{\text{Total fraction}}$$

Table 4: RAC classification

Risk of Assessment	Criteria (%)
No risk	< 1
Low risk	1 – 10
Medium risk	11 – 30
High risk	31 – 50
Very high risk	> 50

The calculated RAC in the surface sediment of Brunei Bay is shown in Figure 5. Based on the bar graph, Al was classed as no risk to the benthic organisms whereas Fe and Pb were proved to have a low to medium risk on the Brunei Bay surface sediment. In the meantime, Cu showed a very high risk during both sampling periods as it is highly bioavailable for benthic organisms in the Brunei Bay. Even though Cu showed very high risk towards the benthic organisms in Brunei Bay, Cu was depleted in comparison with the continental crust value (Figure 3). Cu is counted as one of the nutrient-type metals which is needed in the organisms growth (Lorenzo *et al.*, 2002). On the whole, the RAC value increasing in January 2014 compared to July 2013 sampling period for all metals.

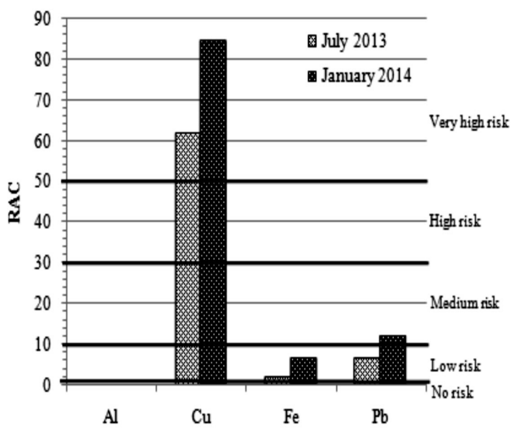


Figure 5: RAC calculated in Brunei Bay surface sediment

Since Pb shows an extremely high concentration compared to the continental crust value (Figure 3) and Cu is observed to have a high mobility and bioavailability behavior (Figure 5), few tests were carried out in order to investigate major contribution by anthropogenic or natural in the Brunei Bay sediment. The first test is normalization; a graph consists of an element versus a normalized element. For the present study, Li was selected as the normalized element since it has the most significant correlation with other elements. The normalization test is a test to compensate for the natural variability of metals in sediments so that any anthropogenic contributions may be detected (Christophoridis *et al.*, 2009; Dung *et al.*, 2013). In the normalization graph, if the plotted element falls within 95% confidence radius, the element is considered to be associated with natural concentration. Whilst of the plotted element falls outside of the 95% confidence level, it is considered as associated with anthropogenic influence. Based on the normalization graphs, data which is fallen outside radius of 95% confident level is proved to be contributed from the anthropogenic sources; for example the circled data in Figure 6 below:

The second test is enrichment factor analysis (EF). Only data proved to be anthropogenic in the normalization test is selected for EF analysis. EF is calculated as below equation and if the calculated EF value is less than 1, it is proved to be input from the anthropogenic sources.

$$EF = \frac{(TMe / TLi) \text{ sample}}{(TMe / TLi) \text{ continental crust}}$$

Remarks:

TMe: Total concentration of an element in the sediment.

TLi: Total concentration of Li in the sediment.

In order to define at what level is an anthropogenic element pollutes the sediment, the final test was carried out namely the geoaccumulation index analysis, I_{geo} (Muller, 1979; Christophoridis *et al.*, 2009). I_{geo} assesses the pollution level based on the below equation and classified the level based on Table 5 class:

$$I_{geo} = \text{Log}_2 (C_n / 1.5B_n)$$

Remarks:

C_n: Measured concentration of the observed metal in the sample of interest.

B_n: Geochemical background concentration of the metal in the continental crust (Mason and Moore, 1982; Wedepohl, 1995).

1.5: Background matrix correction factor due to lithogenic effects.

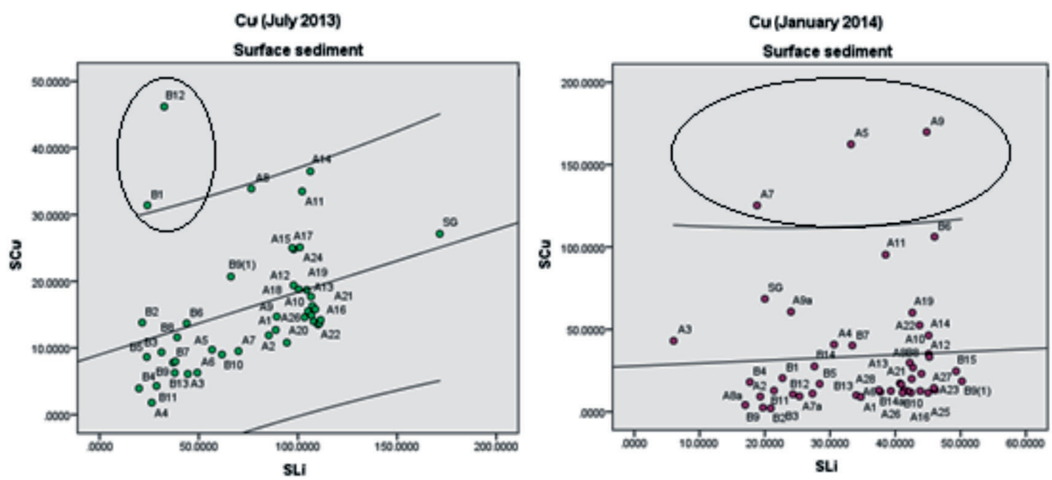


Figure 6: Normalization graph of Cu during both sampling periods

Class	Pollution level
> 5	Very polluted
4 – 5	Polluted to very polluted
3 – 4	Polluted
2 – 3	Moderately polluted to polluted
1 – 2	Moderately polluted
0 – 1	Non polluted to moderately polluted
< 0	Non polluted

Based on the calculated EF and I_{geo} for the present study (Table 6), only Pb at station B13 during July 2013 sampling period is classified as contribution from the anthropogenic. Pb is proved to be moderately polluted the surface sediment at that particular station and sampling period.

Based on the sediment particle size, particle size triangle plots were drawn according to the Moment Method using GRADISTAT *version* 4.0. The triangle plots verified that the surface sediment of Brunei Bay is consisted of silt and silty loam sediment type. In the meantime, low percentages of total organic carbon found were in the range of 3.8% to 5.4%. A Pearson correlation analysis was carried out in order to determine the correlation between metals concentration with sediment particle size and total organic carbon content in the sediment samples. Significant correlation at 0.05 levels (2-tailed) was found between the total concentration of Fe and Pb with the sediment particle size and the total organic carbon fraction (Table 7).

The Pearson correlation (Table 7) showed the increasing of significant correlation value of Fe and Pb with decreasing of sediment particle size as highest value showed correlation with clay fraction during both sampling periods. Even as the lowest significant correlation value was correlated with sand fraction. This finding is in agreement with Ujević *et al.* (2000), Suresh *et al.* (2015) and Bidai *et al.* (2016) as high concentration of metals are strongly correlated with fine sediment particle size. As for the significant correlation of Fe and Pb with total organic carbon strongly suggested the strong attraction of these metals towards the organic materials in the surface sediment of Brunei Bay.

Conclusion

This study provides the first data set on the evaluation of metals concentration in the surface sediment of Brunei Bay, Borneo where such data may has been published but unavailable for access. The total concentration of Al, Fe, Cu and Cd were found to be lower than the continental crust value except for Pb. The fractionation of metals and RAC showed that Al is the least labile whereas Cu is the most labile and bioavailable to the benthic organisms in Brunei Bay. Even though Cu shows some toxicity behavior, the level found during both sampling period did not pollute the surface sediment system due to a low level of concentration compared to the continental crust value. Based on the geochemistry tests to facilitate the anthropogenic input, Pb has moderately polluted

Table 6: EF and I_{geo} value in surface sediment of Brunei Bay

Station		Al		Fe		Cu		Cd		Pb	
		EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}
July 2013	B1					< 1	-				
	B12					< 1	-				
	B13									3.62	2.19
	SG							< 1	-		
January 2014	A1			< 1	-						
	A5					< 1	-				
	A7					< 1	-				
	A9					< 1	-				
	A11	< 1	-							< 1	-

Table 7: Pearson correlation of metals surface sediment of Brunei Bay with total organic carbon and sediment particle sizes

Element		July 2013				January 2014			
		TOC	% sand	% silt	% clay	TOC	% sand	% silt	% clay
Al	Pearson correlation	-.130	-.172	-.221	-.195	-.189	-.181	-.225	-.215
	Sig. (2-tailed)	.405	.271	.155	.211	.198	.217	.124	.142
Cu	Pearson correlation	-.202	-.015	.138	.184	-.111	.119	-.069	-.007
	Sig. (2-tailed)	.195	.922	.378	.237	.451	.419	.643	.965
Fe	Pearson correlation	-.587**	.514**	.708**	.764**	-.503**	.639**	.713**	.803**
	Sig. (2-tailed)	.000	.000	.000	.000	.000	.000	.000	.000
Pb	Pearson correlation	-.491**	.324*	.663*	.752*	-.449**	.622**	.681**	.780**
	Sig. (2-tailed)	.001	.034	.000	.000	.001	.000	.000	.000

the surface sediment of Brunei Bay during July 2013 sampling period by the anthropogenic sources. The decreasing of the residual fraction and the increasing of RAC percentages during January 2014 signified that the Northeast monsoon affected the surface sediment samples. Although it is shown that there is no serious issue of metals pollution in the surface sediment of Brunei Bay but the accumulation behavior found by the high level of concentration in the transects stations as well as the increasing of human activities might cause a serious effect on metals in the future.

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