Oceanological and Hydrobiological Studies

International Journal of Oceanography and Hydrobiology

Volume 45, Issue 3, September 2016
pages (353-367)

ISSN 1730-413X
eISSN 1897-3191

Temporal and spatial variability of heavy metals in Marudu Bay, Malaysia

by

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DOI: **10.1515/ohs-2016-0032**
Category: **Original research paper**
Received: **January 19, 2016**
Accepted: **February 29, 2016**

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Abstract

The current study was conducted to estimate the baseline concentration of heavy metals in the surface sediment of Marudu Bay. Environmental parameters were measured at the seafloor and samples of the surface sediment were collected at monthly intervals for the period of 12 months. The organic content, total N, total P and concentration of 16 trace metals in the surface sediment were analyzed. The baseline concentration of metals was estimated by geochemical normalization. Anthropogenic inputs of metals were then estimated by calculating the enrichment factor for each element. The result demonstrated that the C/N ratio of sediment at Marudu Bay varies from 15 to 342, which indicates the dominance of terrestrial organic matter. The baseline concentration of V, Fe, Mn, Zn, Ti, Rb and Sr were 26.74 mg kg^{-1}, 1.04%, 205.31 mg kg^{-1}, 34.09 mg kg^{-1}, 507.61 mg kg^{-1}, 93.25 mg kg^{-1}, 37.56 mg kg^{-1}, respectively. The concentration of most metals was comparable to the baseline, except Mn and Zn which showed higher concentrations in most parts of Marudu Bay. In conclusion, the metal concentration in Marudu Bay is still within the permissible levels and should not cause any threats to public health.

Key words: marine sediments, heavy metals, nutrients, enrichment factor

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Introduction

Marudu Bay is located close to one of the largest marine protected areas in Southeast Asia, the proposed Tun Mustapha Marine Park and the coral triangle. The intensive land use in recent years for agricultural and industrial development in the neighboring areas has become a cause for concern (Tan & Ransangan 2015b; 2016) due to possible threats to the environment in the adjacent marine protected areas. Heavy metals discharged into estuarine and coastal waters are rapidly bound to particulate matter and settle on the sea bottom (Inengite et al. 2010; Zaaboub et al. 2014). The accumulation of high concentrations of heavy metals in the marine environment may cause adverse effects on marine organisms and cause acute heavy metal poisoning in humans through bioaccumulation of metals from the food chain (Wang et al. 2002; Bradl 2005; Kamaruzzaman et al. 2011).

Heavy metals can be defined as trace elements that are toxic even in low concentrations, and have a relatively high density with relative atomic mass ranging between 63.5 and 200.6 (Wang et al. 2002; Alluri et al. 2007; Lakherwal 2014). Marine, surface sediments are useful to illustrate the most recent deposition of heavy metals in a given area (Zaaboub et al. 2014). However, the concentration of heavy metals in marine sediment is a result of combined accumulation of natural and anthropogenic inputs (Sany et al. 2014). Therefore, the distinction between these two metal proportions is required in order to quantify the anthropogenic loads of heavy metals in an area. Normalization procedures are commonly used to compensate the natural variability of metals in the sediment, which would then enable the detection and quantification of any anthropogenic metal contribution to the system (Loring 1991; Siddique et al. 2009).

Generally, the normalization approaches consist of granulometric and geochemical methods. Granulometric normalization approaches use a certain fraction of sediments, mainly silt and clay, whereas geochemical normalization approaches employ certain conservative elements like Al, Fe, Li or Rb to minimize the influence of particle size composition on the concentration of heavy metals (Hanson et al. 1993; Daskalakis & O'Connor 1995; Zhang 1995; Covelli & Fontolan 1997; Rae 1997; Schiff & Weissberg 1999; Rubio et al. 2000; Siddique et al. 2009). The enrichment factor (anthropogenic metal contribution) of metals is usually estimated based on the ratio between a sample and background value, which gives a value range of 1 $\pm 2\sigma$ (Middleton & Grant 1990).

To date, studies of heavy metals in the Malaysian marine environment are very limited. Only a few reports are available on heavy metal concentration in bivalves (Yap et al. 2004; 2006; 2007; Yap & Al-Barwani 2012), sediment of coastal areas in Peninsular Malaysia (Ismail et al. 1993; Shazili et al. 2006) and the west coast of Sabah (Mokhtar et al. 1994; Ali et al. 2014). Although the north coast of Sabah is a gazetted marine protected area, the information on the baseline levels of heavy metals is still lacking. A detailed and comprehensive study of heavy metal contamination in Sabah coastlines is necessary to establish the baseline information on the level of heavy metals in the sediments. Therefore, the main objective of this study was to determine the possible anthropogenic sources of heavy metals in Marudu Bay.

Materials and Methods

Sampling sites

Sampling was conducted in Marudu Bay (6° 35′ to 7° N and 116° 45′ to 117° E), north of Sabah (Figure 1). The bay has an equatorial climate with uniform temperature, high humidity and heavy rainfall due to its proximity to the equator (Malaysian Meteorological Department, 2014). In general, the Asian tropical monsoon may be divided into three monsoons, the Southwest Monsoon (SWM) from May to September, the Northeast Monsoon (NEM) from November to March and the Inter-seasonal Monsoon (IM) in April and October.

The potential anthropogenic activities possibly contributing to the accumulation of heavy metals in Marudu Bay include: (1) runoff of pesticides from a palm tree plantations at the southwest coast, (2) discharge of untreated domestic waste from areas of high population density at a bay pocket, and (3) effluent from a palm oil processing factory and liquid wastes from auto service workshops on the east coast of Marudu Bay (Tan et al. 2016). Ten sampling sites were located in Marudu Bay. Stations 1, 2 and 3 were located at the bay pocket, stations 4 and 5 were located on the west coast, stations 6, 7 and 8 on the north coast and stations 9 and 10 on the east coast of Marudu Bay. The water depth at stations 1, 2, 3 and 10 was below 5 m, at stations 4, 5, 7 and 9 ranged from 5 to 10 m, and at stations 6 and 8 – over 10 m.

Environmental variables

Environmental variables, including water temperature (°C), salinity (PSU), pH, and dissolved oxygen (%) at 0.5 m above the seafloor, were measured using

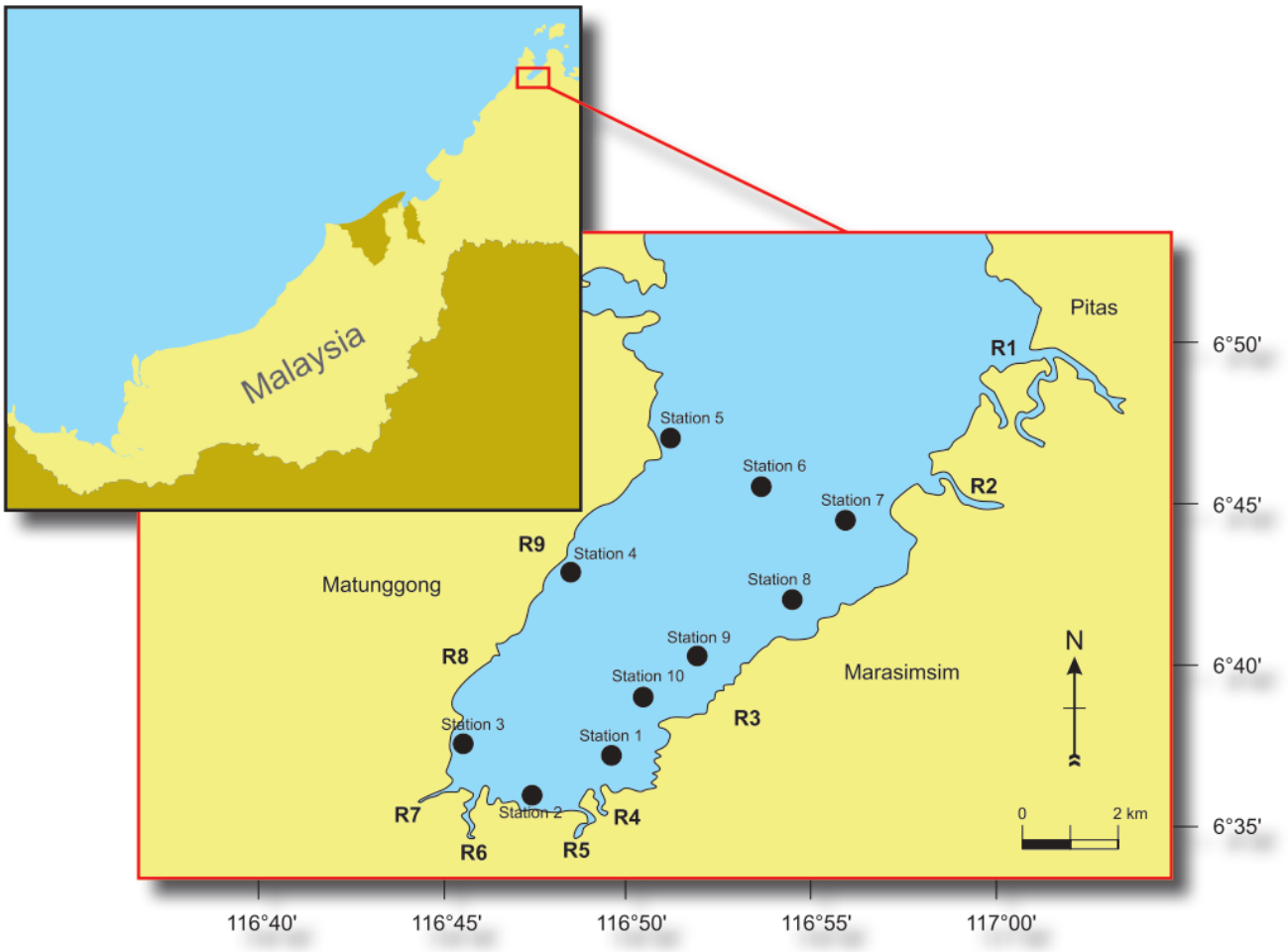

Figure 1

Location of the ten sampling stations in Marudu Bay. Note: R1= Pitas River; R2= Bangkoka River ; R3= Marasimsim River ; R4= Taritipan River ; R5= Raku River ; R6= Sumbilingan River ; R7= Marudu River ; R8= Karangawan River ; R9=Matunggong River

a multifunctional environmental sensor (YSI; Loveland, Co, the USA). Water depth (m) was measured by a depth sounder, whereas water current speed was measured using a current meter (Stanley, USA) deployed at 10 min intervals at 0.5 m above the sea bottom. Sampling was conducted for 12 months from February 2014 to January 2015 during the full moon (spring tide), at approximately the same time (from around 8.00 to 8.30 am). Secondary data on the monthly amounts of rainfall and wind speed were obtained from the Meteorological Department, Kota Kinabalu.

Organic content and sediment composition

The upper sediment layers were sampled using a Peterson stainless steel grab with an area of 0.04 m^2 and a penetration of about 5 cm. Samples were then kept in airtight metal-free polyethylene bags and stored at 4°C in dark conditions. A small sediment subsample of 5 g was oven-dried in a laboratory, at 105°C until constant weight was obtained and then heated in a muffle furnace at 550°C for 6 h. The organic carbon was determined according to the equation presented below:

$$Organic\ carbon = \left(\frac{DW - DW_{550}}{DW} \right) \times 100$$

where:

DW = Dry weight after oven-drying at 105°C

DW_{550} = Dry weight after combustion at 550°C for 6 h

A 100 g sediment subsample was air-dried at room temperature on plastic trays, ground and mixed

thoroughly. The sediment clay-silt percentage was measured by a laser diffraction particle size analyzer (Sequola, Canada) according to Agrawal and Pottsmith (2000). The total nitrogen and total phosphorus in the air-dried sediment were determined using the Kjeldahl method and the molybdenum-antimony spectrophotometric method (Bao 2005), respectively.

Heavy metals

All the equipment and glassware were fist acid washed in 10% nitric acid solution to avoid metal contamination. A subsample (0.5 g) of bulk sediment was acid digested (6:1:2; nitric acid: perchloric acid: hydrochloric acid) at 180°C for 10 h according to Matthai et al. (1998). The extract was collected in 0.5 M HNO_3 and filtered through a GF/C filter paper (47 mm, 0.45 µm, Whatman) and stored in scintillation low potassium Glass bottles at 4°C until analysis.

The concentration of various trace metals, including Al, Li, Cd, V, Fe, Mn, Zn, As, Ti, Rb, Sr, Cr, Cu, Ag, Ni and Pb, was measured by a Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). A Perkin-Elmer Sciex ELAN 5000 ICP mass spectrometer was used. A standard torch for this instrument was used with an outer argon gas flow rate of 15 l min^{-1} and an intermediate gas flow of 0.9 l min^{-1} (Date & Gray 1988). The procedural blanks and quality control samples were made from the standard solution for each metal and analyzed once in every ten samples to check for sample accuracy (Yap et al. 2006). For accuracy, both within and between-day relative percentage difference (RSD) values were determined by analyzing two in-house laboratory control samples. Within-day precision was determined by analyzing the laboratory control samples in triplicate on each day, and between day precision was measured by using the mean results of the triplicate samples analyzed each day on 5 different days.

Statistical analysis

The environmental parameters, nutrients in sediment and heavy metal concentrations were analyzed for their significance using the SPSS Windows Statistical Package (version 21). Tests were considered to be significant at $p<0.05$. Prior to the analyses, all variables were tested for normality and homogeneity of variance. One-way ANOVA was performed, followed by Turkey multiple comparison tests (Turkey HSD) to make specific contrast in spatial and temporal variations of environmental parameters, silt-clay percentage, organic content, nutrients and heavy metal concentrations. Eight environmental variables were reduced to three principal components. To help the interpretation of the principal components, varimax rotation was performed (Wang et al. 2006). Correlation coefficients were calculated between the main environmental variables, principal components and the concentration of heavy metals.

Normalization was performed by the geochemical normalization method as described by Newman and Watling (2007). Al, Fe and Li were selected as alternative reference elements. Linear regressions were first performed for each heavy metal on the alternative reference elements. The appropriate reference element for each metal was selected based on the highest coefficient value of the regression (Newman & Watling 2007). Subsequently, data sets in a scatter plot were tested for linearity of distribution under the condition of $p<0.05$ and $r^2>0.9$. Metals with concentrations higher than the upper limit of the 95% confidence interval were eliminated as outliers. The procedure was repeated until the above two criteria were satisfied (Loring 1991).

The baseline concentration of heavy metals was calculated using the linear regression equation between a heavy metal element and a reference element (Colizza et al. 1996; Newman & Watling 2007).

$$C_M = aC_N + b$$

where:

C_M = Baseline concentration

C_N = Concentration of a reference element

a and b = Regression constants of the equation

The enrichment factor was calculated according to Middleton and Grant (1990).

$$EF = \frac{X(s) / Al(s)}{X(b) / Al(b)}$$

where:

X = Element concentration

s = Sample

b = Background value

Results

Environmental variables

Temporal and spatial variations of environmental variables are illustrated in Figure 2 and Figure 3, respectively. In general, water temperature at the sea bottom of Marudu Bay was significantly higher ($p<0.05$) in SWM than in NEM. Water salinity was significantly higher ($p<0.05$) from April to June than in other months. Water pH was significantly higher ($p<0.05$) from May to July than in December and from February to March. Current speed was significantly higher ($p<0.05$) from April to September than in other months. DO was significantly higher ($p<0.05$) from April to August than in the other months.

In respect of the spatial variation, no significant differences ($p>0.05$) were observed in temperature and pH among the stations. However, water salinity in the mouth of the bay (stations 4, 5 and 6) was significantly higher ($p<0.05$) compared to station 3. DO at stations 2, 3, 5, 7 and 10 was significantly higher ($p<0.05$)

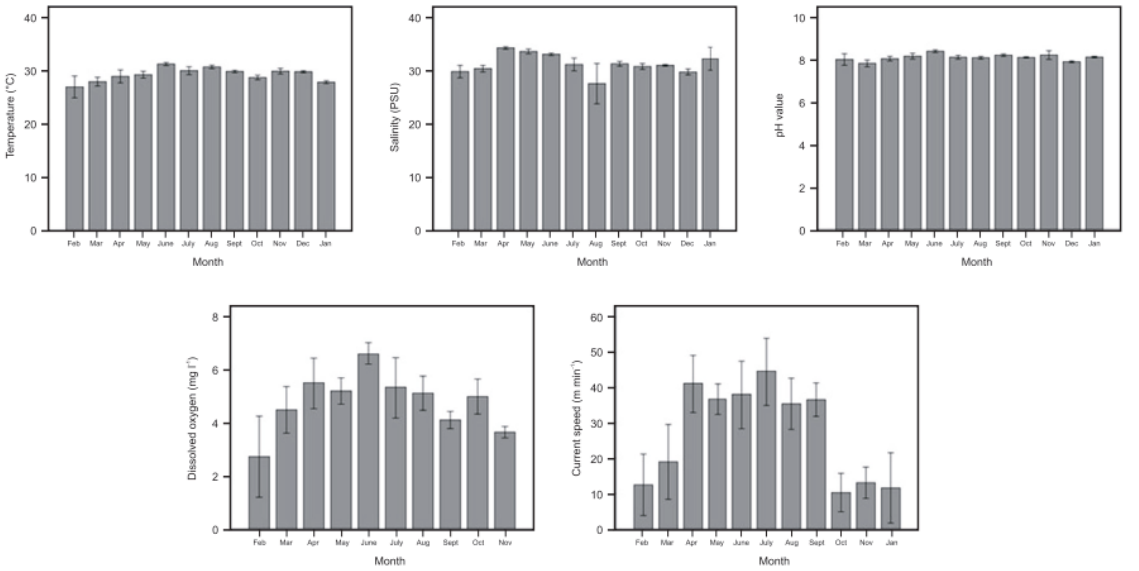

Figure 2

Temporal variations of environmental parameters (mean ±SD) in Marudu Bay from February 2014 to April 2015

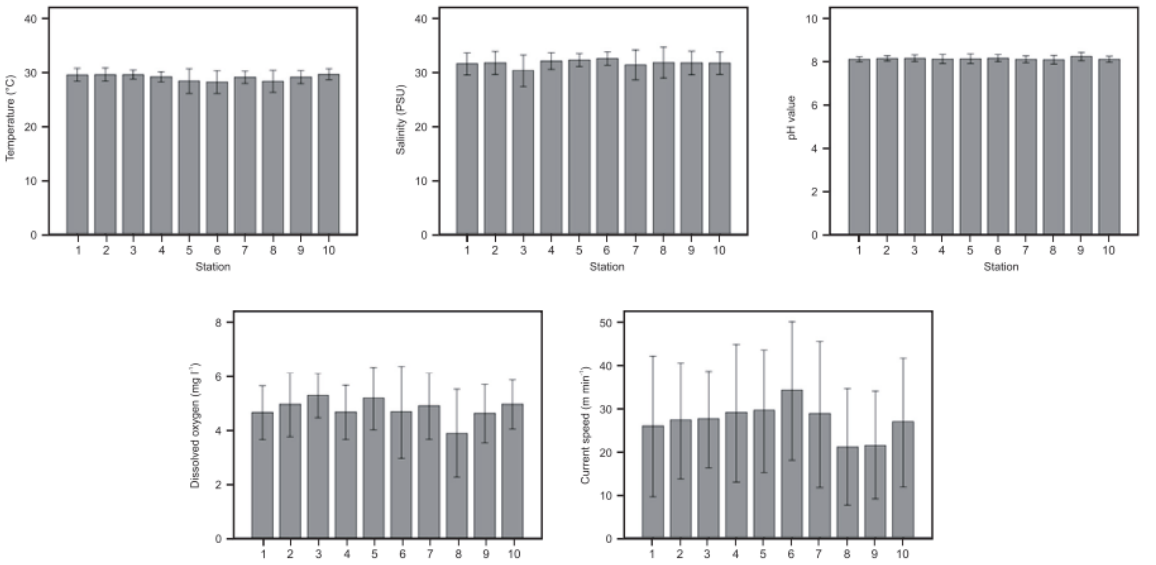

Figure 3

Spatial variations of environmental parameters (mean ±SD) in Marudu Bay from February 2014 to April 2015

compared to station 8. Current speed at station 6 was significantly higher ($p<0.05$) compared to stations 1, 8 and 9.

Marudu Bay received heavy rainfall in October 2014, December 2014 (maximum: 502.1 mm) and January 2015, while the lowest rainfall of 1.1 mm occurred in March 2015 (Figure 4). The average wind speed recorded in January to March (2.8 to 3.1 m s^{-1}), July (2.8 m s^{-1}) and October 2014 (2.7 m s^{-1}) was significantly higher compared to other months (1.9 to 2.5 m s^{-1}).

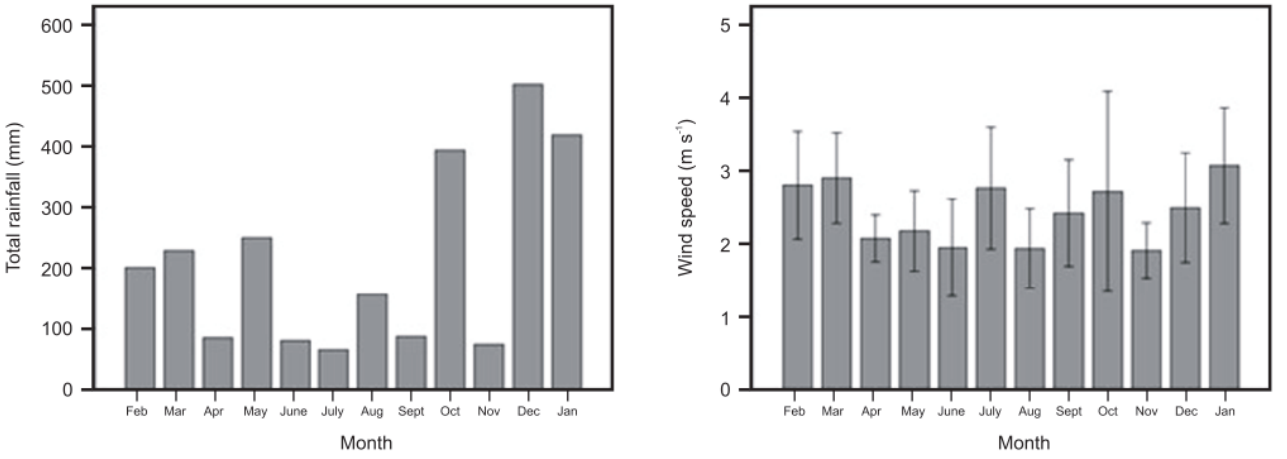

Figure 4

Total rainfall and mean wind speed (mean± SD) in Marudu Bay from April 2014 to April 2015

Nutrients and granulometric characteristics of the sediment

Granulometric characteristics of the sediments from each station are summarized in Table 1. In general, the clay composition at all stations was similar ($p>0.05$). However, the silt composition was significantly ($p<0.05$) higher in the bay pocket (stations 1, 2, 9 and 10) than in the other areas. The sand composition was significantly higher at stations 3, 4, 5 and 6 compared to other stations. The organic content, total phosphorus, total nitrogen, the C/N ratio and clay-silt composition in sediment ranged from 1.47 to 16.84%, 0.001 to 0.21%, 0.01 to 0.21%, 15.19 to 342.26 and 13 to 85%, respectively.

Temporal variations of nutrients and clay-silt composition in sediment are illustrated in Figure 5. Organic content in sediment was significantly lower ($p<0.05$) in February and from April to May compared to other months. Total phosphorus was significantly higher ($p<0.05$) from February to March and from September to November 2014 than in other months. Total nitrogen was significantly higher ($p<0.05$) from May to August 2014, but significantly lower ($p<0.05$) from March to April and from October to December 2014. The C/N ratio was significantly higher ($p<0.05$) from February to April and October to December, but significantly lower ($p<0.05$) in February and from May to September. Clay-silt composition in sediment was significantly higher ($p<0.05$) from December to January than in other months.

In respect of the spatial variation, the organic content in sediment was significantly higher ($p<0.05$) at stations 6 and 8, but significantly lower ($p<0.05$) at stations 3, 5 and 7 (Figure 6). Total phosphorus in sediment was significantly higher ($p<0.05$) at stations 1, 6 and 8 compared to other stations. The total nitrogen in sediment was significantly higher ($p<0.05$) at stations 1, 6, 8 and 10, but significantly lower ($p<0.05$) at stations 3, 4, 7 and 9. The C/N ratio in

Table 1

Granulometric characteristics of the sediments from each station in Marudu Bay

	Clay (% ±SD)	Silt (% ±SD)	Sand (% ±SD)
S1	2.8 ± 0.7	57.1 ± 8.7	40.5 ± 8.6
S2	2.6 ± 0.7	53.7 ± 8.9	44.0 ± 8.8
S3	2.2 ± 0.6	40.3 ± 7.9	66.1 ± 13.8
S4	2.6 ± 0.7	44.5 ± 14.5	61.9 ± 19.0
S5	2.4 ± 0.8	45.3 ± 13.1	61.1 ± 18.3
S6	2.2 ± 0.9	39.0 ± 17.8	67.6 ± 20.4
S7	2.5 ± 0.8	50.3 ± 10.6	47.5 ± 10.6
S8	2.3 ± 0.7	47.9 ± 8.4	52.5 ± 7.8
S9	2.6 ± 0.7	58.4 ± 11.0	39.3 ± 10.7
S10	3.0 ± 0.9	56.5 ± 16.3	40.9 ± 15.9

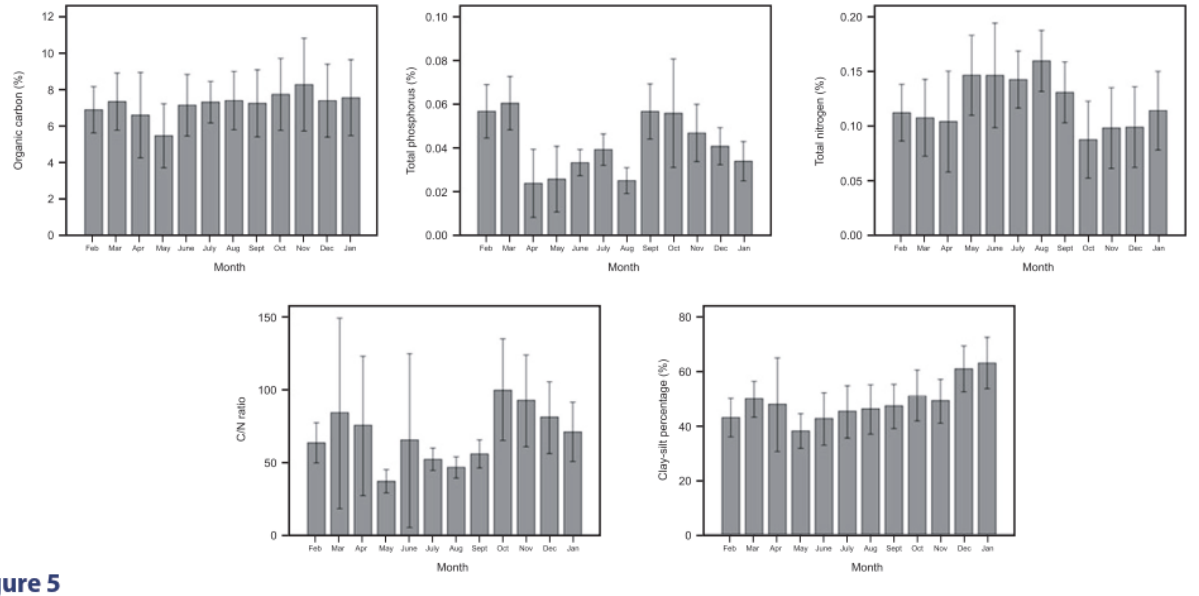

Figure 5

Temporal variations of nutrients and clay-silt percentage (mean± SD) of sediment in Marudu Bay from February 2014 to April 2015

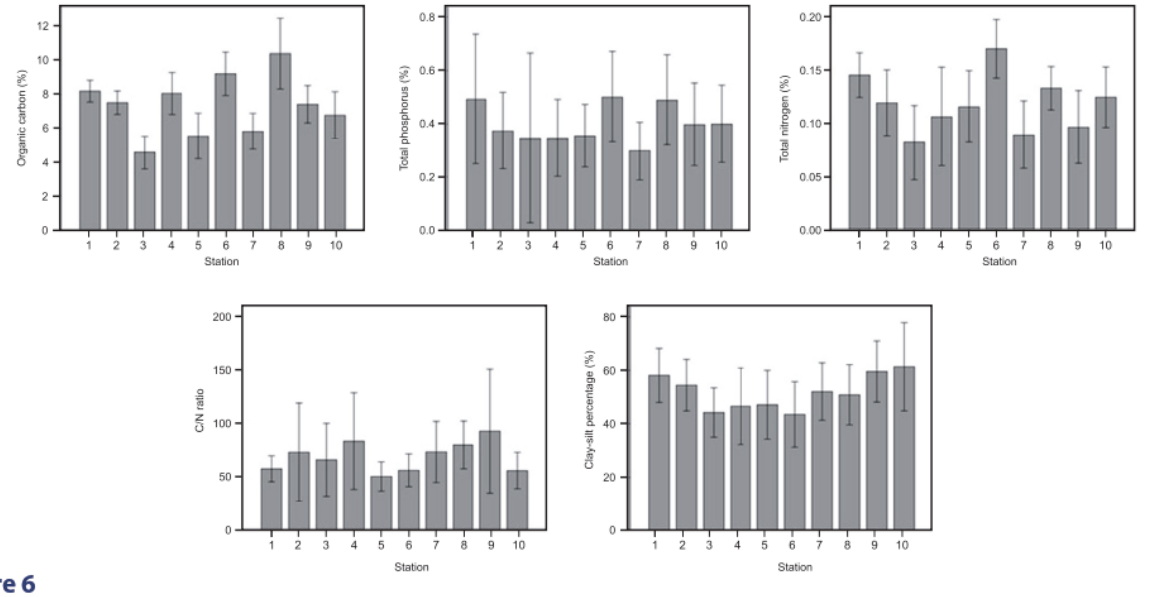

Figure 6

Spatial variations of nutrients and grain size (mean ±SD) of sediment in Marudu Bay from February 2014 to April 2015

sediment was significantly higher ($p<0.05$) at stations 4, 7, 8 and 9, but significantly lower ($p<0.05$) at stations 1, 5, 6 and 10. Clay-silt composition at stations 1, 2, 9 and 10 was significantly higher ($p<0.05$) compared to other stations.

Concentration of metals

Concentrations of various metals in coastal sediment of Marudu Bay are summarized in Table 2. Both within-day and day-to-day precision values were <4% RSD over 5 independent days (data not shown). The concentration of Ag and Cd in sediment of Marudu Bay was below the detection limit in most data points. No significant differences were observed for Al, Fe, Zn, Ti, Cd, V, Rb, Sr, Cu, Ag, Ni and Pb throughout the stations. However, significant differences were recorded in Sr, Mn, As, Li, Cr and Ni. The concentration of Sr was significantly higher ($p<0.05$) at stations 6 and 9 than at the other stations. Mn was significantly higher ($p<0.05$) at stations 6 and 10 than at stations 3, 5

Table 2

Concentrations of metals in the sediment of Marudu Bay

	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10
Al (%)	3.02 ± 1.29	3.08 ± 1..11	2.14 ± 0.92	2.54 ± 1.03	2.15 ± 1.73	3.81 ± 1.18	2.43 ± 1.02	2.98 ± 1.18	2.92 ± 1.11	2.86 ± 1.20
Fe (%)	1.22 ± 0.67	1.24 ± 0.60	0.90 ± 0.42	0.90 ± 0.44	0.88 ± 0.54	1.18 ± 0.53	0.93 ± 0.52	1.40 ± 0.86	1.33 ± 0.77	1.32 ± 0.54
Mn (mg kg^{-1})	275.60 ± 204.90	231.80 ± 134.80	119.40 ± 90.20	224.78 ± 126.64	188.71 ± 232.11	341.65 ± 176.60	93.71 ± 75.86	318.22 ± 222.28	291.50 ± 218.56	277.50 ± 167.94
Zn (mg kg^{-1})	89.30 ± 91.60	57.30 ± 38.80	62.20 ± 60.50	42.77 ± 21.74	51.33 ± 30.63	69.20 ± 50.50	51.81 ± 35.60	49.18 ± 20.80	50.57 ± 19.41	47.86 ± 19.02
As (mg kg^{-1})	2.19 ± 1.96	2.57 ± 2.01	2.93 ± 2.04	2.98 ± 2.01	2.77 ± 2.21	2.62 ± 1.72	3.76 ± 2.19	0.66 ± 1.42	0.00 ± 1.43	2.62 ± 2.19
Ti (mg kg^{-1})	364.1 ± 450.20	347.0 ± 415.1	246.8 ± 246.3	236.18 ± 266.87	251.63 ± 288.83	316.76 ± 337.46	271.31 ± 324.69	468.66 ± 598.90	388.01 ± 575.39	300.51 ± 385.30
Cd (mg kg^{-1})	0.00 ± 0.43	0.00 ± 0.38	0.00 ± 0.36	0.27 ± 1.62	0.00 ± 0.61	0.00 ± 0.36	0.00 ± 0.38	0.00 ± 0.40	0.00 ± 0.66	0.00 ± 0.39
V (mg kg^{-1})	31.38 ± 16.17	32.20 ± 16.88	23.31 ± 12.65	27.38 ± 13.56	22.26 ± 16.38	36.31 ± 17.02	26.26 ± 14.58	31.59 ± 22.97	30.92 ± 17.63	29.90 ± 16.74
Rb (mg kg^{-1})	78.90 ± 19.30	78.49 ± 18.53	61.43 ± 23.46	82.96 ± 29.64	68.13 ± 34.11	110.45 ± 21.83	71.29 ± 26.54	72.33 ± 24.48	67.98 ± 27.04	94.00 ± 27.71
Sr (mg kg^{-1})	31.20 ± 8.40	26.60 ± 7.50	25.82 ± 10.20	35.95 ± 11.68	33.98 ± 14.88	52.67 ± 15.06	26.35 ± 8.08	40.20 ± 15.05	33.27 ± 9.96	37.10 ± 11.89
Li (mg kg^{-1})	47.60 ± 23.30	44.60 ± 24.30	23.46 ± 24.86	33.12 ± 31.69	47.84 ± 65.71	31.28 ± 28.94	29.95 ± 32.34	70.03 ± 45.07	98.02 ± 56.97	43.48 ± 39.54
Cr (mg kg^{-1})	0.00 ± 0.43	0.00 ± 0.38	0.00 ± 0.36	0.27 ± 1.62	0.00 ± 0.61	0.00 ± 0.36	0.00 ± 0.38	0.00 ± 0.40	0.00 ± 0.66	0.00 ± 0.39
Cu (mg kg^{-1})	57.40 ± 58.00	115.80 ± 169.30	59.56 ± 52.48	61.60 ± 68.39	61.49 ± 76.73	53.69 ± 57.96	57.92 ± 71.63	95.83 ± 143.35	79.80 ± 76.99	56.30 ± 64.58
Ag (mg kg^{-1})	0.00 ± 0.95	0.00 ± 0.90	0.00 ± 0.60	0.00 ± 0.66	0.00 ± 0.80	0.00 ± 0.68	0.00 ± 0.70	0.00 ± 1.03	0.00 ± 0.88	0.00 ± 0.65
Ni (mg kg^{-1})	71.90 ± 24.70	65.30 ± 29.40	24.47 ± 23.33	37.29 ± 40.31	36.48 ± 31.15	41.47 ± 22.94	30.46 ± 17.75	109.21 ± 54.56	135.09 ± 54.62	66.00 ± 27.97
Pb (mg kg^{-1})	4.7 ± 5.0	3.20 ± 6.24	1.78 ± 6.72	4.87 ± 6.53	4.98 ± 5.17	2.78 ± 5.89	2.90 ± 7.02	3.29 ± 4.19	3.13 ± 3.61	6.16 ± 2.44

and 7. Concentration of As at stations 8 and 9 was significantly lower ($p<0.05$) compared to stations 3, 4, 5, 7 and 10. Cr at stations 8 and 9 was significantly higher ($p<0.05$) than at stations 3, 4, 6 and 7. Ni at stations 8 and 9 was significantly higher ($p<0.05$) compared to other stations. Li at station 6 was significantly higher ($p<0.05$) than at any other stations.

In respect of temporal variations, no significant differences were recorded in Zn, Ti, Cd, Rb, Ag, Ni, Pb, Sr, Cr and Ni throughout the year. However, the concentration of Fe, Mn, V, Li, As and Cu in sediment of Marudu Bay has a similar pattern, with a significant increase in May and September, the inter-seasonal monsoon (Figure 7).

Relationship between environmental parameters and metals concentration

Three principal components (PC) were extracted from the principal component analysis of seven environmental variables (Table 3). The PCs explained 28.5%, 27% and 13.6% of the total variance, respectively. The total initial eigenvalues were 3.067, 2.366 and 1.005 for PC1, PC2 and PC3, respectively. Temperature, pH, current speed and dissolved oxygen showed a significant positive loading on PC1. Clay-silt composition had a strong positive loading on PC2. The organic content showed a strong positive loading on PC3, while dissolved oxygen produced a significant reverse effect. In general, it appears that PC1 represents the southwest monsoon where temperature, current speed and dissolved oxygen are higher. PC2 is associated with the particle size and the composition of clay-silt in sediment, whereas PC3 represents a high microbial activity, which involves high organic content and low dissolved oxygen.

Pearson's correlation coefficients between logarithmically transformed environmental parameters and the concentration of trace metals are presented in Table 4. In general, the concentration of most

Table 3

Principal component analysis: Varimax rotated component matrix

	PC1	PC2	PC3
Temperature	**0.85**	0.07	0.05
pH	**0.70**	-0.20	0.20
Salinity	0.37	-0.24	-0.01
Current	**0.84**	-0.01	-0.14
Dissolved oxygen	**0.70**	0.18	**-0.37**
Particle size	0.09	**-0.94**	0.03
Clay-silt composition	-0.02	**0.93**	0.02
Organic content	-0.04	0.15	**0.92**
Total variation explained (%)	**28.5**	**27.0**	**13.6**

Significant loading are boldfaced.

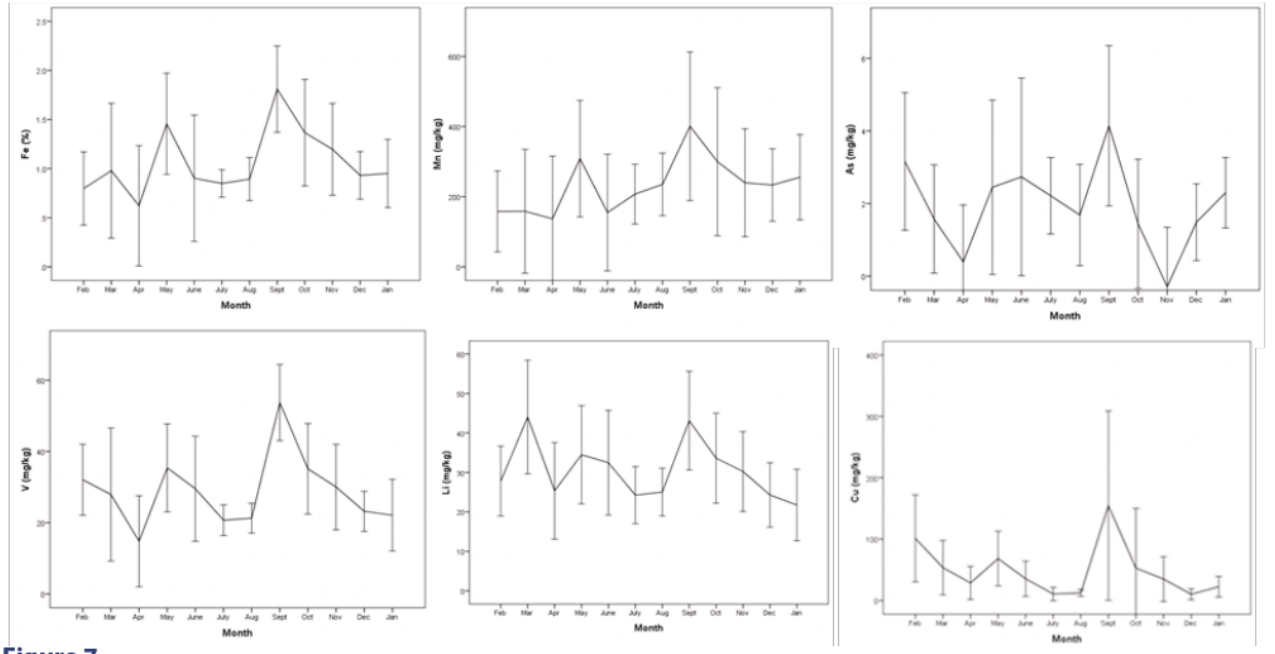

Figure 7

Temporal variations of heavy metal concentration in Marudu Bay

trace metals (V, Mn, Zn, Rb, Sr, Cr, Ni, Li, Fe and Al) was positively correlated with organic content in sediments, except for As which was negatively correlated. In addition, the concentration of Zn, Rb, Cr, Ni, Pb, Li was positively correlated and the concentration of As and Cd was negatively correlated with PC2 and clay-silt composition. The concentrations of most metals were positively correlated with PC3 and negatively correlated with DO and current speed.

Metal enrichment

The resultant regression line with 95% confidence limits between metals and reference elements are illustrated in Figure 8. Metals V, Fe and Mn are very strongly positively correlated with Al, and the r^2 values are 0.895, 0.847 and 0.740, respectively. Whereas, Ti showed a strong positive correlation with Fe (r^2=0.855).

On the other hand, Rb and Sr were positively correlated with Li at r^2 of 0.695 and 0.501, while Zn was correlated with Al at r^2 of 0.365. Other metals, i.e. Cr, As, Cu, Ag, Ni and Pb, did not correlate with any reference elements.

Almost all data points were retained for V and Al regression plot. For other metals, however, many data points were reduced prior to the regression analysis by removing data with the metal concentrations much higher than the upper limit of the 95% confidence interval (Table 5). The reduced data indicated that the baseline concentrations of V, Fe, Mn, Zn, Ti, Rb

Table 4

Pearson's correlation coefficients between environmental parameters after logarithmic transformation and the concentration of trace metals

	V	Mn	Zn	As	Ti	Rb	Sr	Cr	Cu	Cd	Ag	Ni	Pb	Li	Fe	Al
PC1	-	.12*	-.30**	-	-	-	-.15*	-	-.14*	-	-	-	-.32**	-	.12*	-
PC2	-	-	.25**	-.21**	-	.40**	-	.16**	-	-.77**	-	.34**	.21**	.21**	-	-
PC3	.25**	.31*	.12*	-.13*	.20**	.376**	.46**	.26**	-	-	-	.42**	-	.43**	.23**	.25*
Clay-silt	-	-	.13*	-.16*	-	.23**	-.17**	.17**	-	-.742*	-	.32**	-	-	-	-
DO	-.20**	-.14**	-.21**	-	-.26**	-	-.18**	-.18**	-.31**	-	-	-.22**	-	-	-	-
Current	-	-	-.29**	.14*	-	-.19	-.18**	-.26**	-.13**	-	-	-.22**	-.23**	-	-	-
Sal.	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
pH	.13*	.15**	-.18*	.23**	-	-	-	-	-	-.68*	.54*	-	-.36**	-	.13*	.22*
Temp.	-	.18**	-.29**	-	-	-	-.23**	-	-.18**	-	.52*	-	-	-	-	-
Organic	.18**	.23**	.16**	-.26**	-	.38**	.44**	.20**	-	-	-	.41**	-	.46**	.19**	.22*

The significant coefficients:**, p<0.01; *, p<0.05

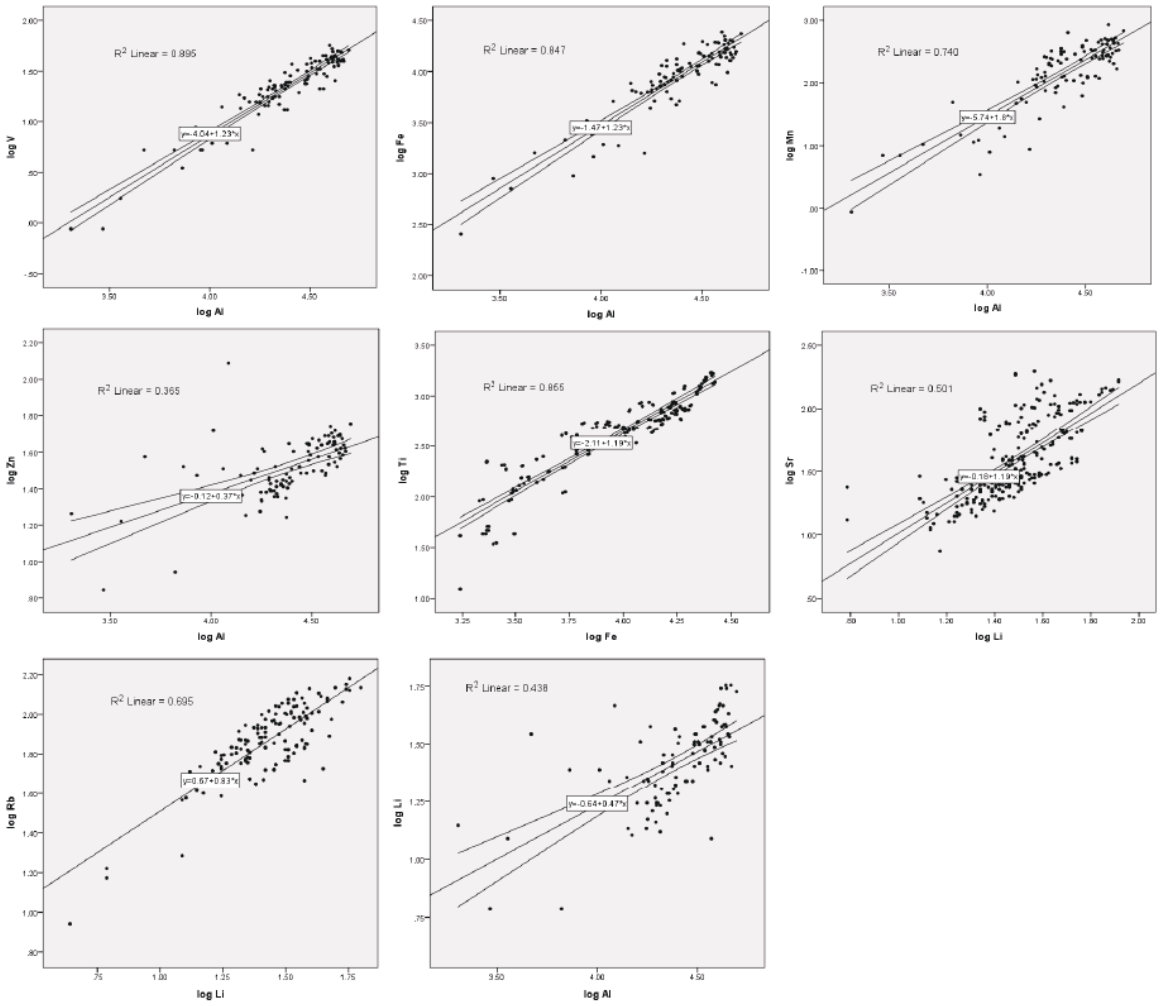

Figure 8

Scatter plots of metals with a suitable reference element in the coastal sediment of Marudu Bay. The solid line represents the regression line with $p<0.05$, while dashed lines represent the 95% confidential intervals.

Table 5

Linear regression equations for various trace metals and the reference elements and the corresponding calculated baseline concentration in the sediment

Element	N	n	Sample removed	Formula	Background
V	330	329	1	$\log_{10}V = -3.99 + 1.22*\log_{10}Al$	26.74 mg kg^{-1}
Mn	326	300	26	$\log_{10}Mn = -5.68 + 1.80*\log_{10}Al$	205.31 mg kg^{-1}
Zn	326	284	42	$\log_{10}Zn = -1.62 + 0.71*\log_{10}Al$	34.09 mg kg^{-1}
Fe	326	319	7	$\log_{10}Fe = -1.40 + 1.20*\log_{10}Al$	1.04 mg kg^{-1}
Ti	188	174	14	$\log_{10}Ti = -1.83 + 1.12*\log_{10}Fe$	507.61mg kg^{-1}
Rb	138	94	44	$\log_{10}Rb = 0.59 + 0.9*\log_{10}Li$	93.25 mg kg^{-1}
Sr	266	190	76	$\log_{10}Sr = -0.74 + 1.51*\log_{10}Li$	37.56 mg kg^{-1}
Cr*					
Cu*					
Cd*					
Ag*					
Ni*					
As*					
Li*					
Pb*					

* The metal does not correlate with any reference elements.

and Sr were 26.74 mg kg^{-1}, 1.04%, 205.3 mg kg^{-1}, 34.1 mg kg^{-1}, 507.6 mg kg^{-1}, 93.3 mg kg^{-1} and 37.6 mg kg^{-1}, respectively.

The degree of metal enrichment estimated based on the geochemical normalization is summarized in Table 6. In general, the content of V at all stations was similar to the baseline value with very low variations of about 10%. Slight enrichment with Mn relative to the baseline concentration was observed throughout the bay (except stations 3 and 7), whereas high enrichment with Zn occurred at the bay pocket. On the other hand, depletion of Ti relative to the baseline concentration was observed at all stations. Moreover, depletion of Rb and Sr relative to the baseline concentration was recorded at most stations and their accumulation at station 6.

Table 6

Metals enrichment

	V	Mn	Zn	Ti	Rb	Sr
S1	1.07	1.22	2.39	0.67	0.61	0.60
S2	1.08	1.01	1.50	0.67	0.64	0.54
S3	1.12	0.75	2.35	0.66	0.96	1.00
S4	1.11	1.19	1.36	0.63	0.92	0.99
S5	1.07	1.18	1.93	0.69	0.52	0.65
S6	0.98	1.20	1.47	0.64	1.29	1.53
S7	1.11	0.52	1.72	0.70	0.87	0.80
S8	1.09	1.43	1.33	0.80	0.38	0.52
S9	1.09	1.34	1.40	0.70	0.25	0.31
S10	1.08	1.30	1.35	0.55	0.79	0.77

Discussion

Environmental parameters

During the northeast monsoon (November to March), the wind carries the moist air from the South China Sea, thus bringing heavy rainfall to the northern coast of Sabah (Malaysian Meteorology Department, 2014). The heavy rainfall resulted in the lower water temperature and salinity during the northeast monsoon compared to the southwest monsoon. On the other hand, Marudu Bay also experienced a relatively stronger current speed and higher DO during the southwest monsoon compared to the northeast monsoon. This is because Marudu Bay is sheltered by Banggi Island from the strong water current during the northeast monsoon, whereas no island blocks the current from the southeast during the southeast monsoon (Azanza et al. 2008).

With regard to the spatial variation, station 3 was a shallow area (3 m) located right in front of the Marudu River, while the mouth of the bay was located in deeper water (>20 m) and far from the source of fresh water. Therefore, the effect of dilution by fresh water results in a stronger effect at station 3 compared to the mouth of the bay. Since the bottom water is in direct contact with the sediment, some of the other parameters (particularly pH and DO) are highly affected by microbial activity and chemical composition of the sediment (Sany et al. 2014).

Nutrients and granulometric characteristics of sediment

Based on the granulometric characteristics of sediments in Marudu Bay, it can be concluded that the mouth of the bay was exposed to a strong current. The finer sediment particles are known to be transported from high to low energy areas (Molinaroli et al. 2009). Therefore, this could explain the accumulation of silt in the bay pocket of Marudu Bay.

In general, the C/N ratios in the surface sediment of Marudu Bay were higher than 20, which indicates that the accumulated organic matter came mainly from terrestrial sources (Tyson 1995; Zaaboub et al. 2014). The high terrestrial organic input is expected due to the location of Marudu Bay close to a mangrove swamp, a palm oil plantation, and areas of high population density.

With regard to the temporal variation, total phosphorus in the surface sediment was significantly higher during the high rainfall season, the northeast monsoon. Soil and rock minerals are rich in phosphorus (Sany et al. 2014). Higher levels of phosphorus content in the surface sediment during the higher mean rainfall season could be contributed by river runoff of soil and weathering rocks (Anderson 2009; Tan & Ransangan 2015a). On the other hand, the total nitrogen in the surface sediment was high during the southwest monsoon. In addition, the C/N ratio was much lower during the southwest monsoon compared to the northeast monsoon. Organic matter of higher plants is known to have a lower nitrogen content, while phytoplankton is rich in nitrogen and deficient in C/N ratios (Carpenter & Capone 1983). Therefore, the significantly higher total nitrogen content and lower C/N ratios in sediment during the southwest monsoon suggest that the organic content was contributed by sedimentation of dead algae cells after blooming events. The results of the current study also showed that the clay-silt composition in the surface sediment was generally higher in months with a weaker current speed. This is because a low current speed induces the deposition of large quantities of low-density particles (Storlazzi & Field 2000; Spagnoli et al. 2008; Yang et al. 2008).

With regard to the spatial variation, a higher level of phosphorus and nitrogen was recorded at stations 1, 6 and 8 as compared to other stations. Station 1 was adjacent to a rocky shore and areas of high population density. The continuous supply of P and N from the rock weathering by a strong wave and domestic effluents could explain the higher levels of P and N at station 1 (Anderson 2009; Tan & Ransangan 2015a). On the other hand, the water at stations 6 and 8 (26 m and 15 m, respectively) was much deeper than at the other stations (2 to 8 m). At the stations with a water depth below than 10 m and with the mean wind speed of 15.8 m s^{-1}, the Ekman layer (the wind-affected depth) extends to the bottom (Adam et al. 2011). This creates a well-mixed homogeneous water column which brings nutrients from the sediments (Anderson et al. 2002). On the other hand, the Ekman layer does not reach the bottom of the deeper water at stations 6 and 8. Therefore, the accumulation of nutrients occurred and resulted in a higher level of organic content, P and N. On the other hand, higher C/N ratios were observed at stations 4, 7, 8 and 9, which indicates a higher level of terrestrial organic inputs compared to other stations (Zaaboub et al. 2014). Station 4 was located near the mangrove swamp, while the east coast of Marudu Bay (stations 7, 8 and 9) was located near a palm oil processing factory. Therefore, the proximity of these stations to the source of terrestrial organic matter might explain the high C/N ratios in the current study. The clay-silt composition in the surface sediment was higher on the southeast coast of Marudu Bay (stations 1, 2, 9 and 10). The higher silt-clay composition in the sediment of the mouth of the bay might result from a longer water resident time in the areas (Molinaroli et al. 2009).

Concentration of metals

Sediments represent an accurate time-integrated record of environmental contamination and therefore can be used as an economic monitoring tool to detect the anthropogenic impact over time (Daskalakis & O'Connor 1995; Sany et al. 2014). In addition, sediments also play a major role in the transport and storage of heavy metals. They are also frequently used to identify the source of contaminants and to determine the dispersion pathways in the marine ecosystem (Murray 1996; Birch et al. 2001). Therefore, the current study used marine sediment as a monitoring tool to evaluate the organic and heavy metal contamination status in the coastal area of Marudu Bay.

The concentrations of most elements (Table 1) in the current study were lower than the recommended value of metals in the sediment as suggested by FAO/WHO and USEPA guidelines and the Pollution Load Index (PLI). However, the concentrations of Al, Mn, Zn, Rb and Ni were slightly higher, whereas the Cu concentration in sediment of Marudu Bay was much higher than the recommended value, but still within the permissible limit of 108 mg kg^{-1} (Sulochanan et al. 2007). Compared to other parts of Malaysia, most of the metals concentrations in Marudu Bay (except Cu) were lower than those recorded in the coastal areas of Peninsular Malaysia, i.e. 2.5 to 5% of Fe, 3000 to 5000 mg kg^{-1} of Mn, 600 to 900 mg kg^{-1} of Zn and 10 to 30 mg kg^{-1} of Pb (Ismail et al. 1993; Yap et al. 2002; Shazili et al. 2006). Cu comes mainly from shipping activities and the use of pesticide and insecticides (Alluri et al. 2007; Kamaruzzaman et al. 2011). The organic matter in the sediment of Marudu Bay is characterized by high C/N ratios, which indicates terrestrial deposition (Tyson 1995; Zaaboub et al. 2014). Therefore, the large oil palm plantation in the surrounding coastal area could explain the high Cu concentration in the sediment of Marudu Bay.

In terms of spatial comparison, significantly higher Cr and Ni concentrations were recorded at stations 8 and 9. Ni and Cr are the wastes from various industries, including steel, textile, electronic and zinc-based casting industries (Kamaruzzaman et al. 2011). Stations 8 and 9 were located near the palm oil processing factory, which combined with the presence of a relatively large number of small workshops or electronic service and repair shops in the neighborhood resulted in the higher Ni and Cr concentrations. On the other hand, station 6 is the only station with the water depth (>20 m) beyond Ekman's depth of about 15 m. The accumulation of metals that strongly bind to organic detritus is expected (Xu et al. 2010). In addition, higher Mn and As concentrations were recorded at station 3 – the main artisanal fishing ground, and at stations 5 and 7 – the mouth of the bay. The intensive boating and shipping activities in the areas might explain the higher levels of As and Mn (Alluri et al. 2007).

In terms of temporal comparison, the concentration of Fe, Mn, As, V, Li and Cu significantly increased during the inter-seasonal monsoon. Interestingly, the increase in the metal accumulations corresponded to the increase in the C/N ratio and organic content in the sediment. The direction of the wind begins to change during the inter-seasonal monsoon (Malaysian Meteorology department, 2014), hence the poor water circulation during this period might account for the accumulation of terrestrial organic matter in Marudu Bay (Azanza et al. 2008).

Relationship between environmental parameters and metal concentration

The results of the current study demonstrated that most of the metal elements in the sediment responded positively to finer sediments and organic content. Similar results have been reported in other studies where finer sediments are enriched with heavy metals (Weiguo et al. 2001). Fine-grained sediments are known to have a higher surface-to-volume ratio, while organic matter has higher chemical affinity for trace elements (Weiguo et al. 2001; Sany et al. 2014). Therefore, trace metals are accumulated in fine particles with high organic content.

On the other hand, the negative correlation between As and fine particles could result from the fact that the As concentration in the current study was too low to be detected (the result might not reflect the actual conditions). It has been suggested that solubility of metals is affected by physical parameters (Ideriah et al. 2012). This is reflected in the current study where various metals responded to temperature and pH. Lower pH has been suggested to favor metal accumulation (Ideriah et al. 2012). This theory was confirmed in the current study, where V, Mn, As, Ag, Fe and Al positively correlated with pH. However, a few elements (Pb, Cd and Zn) showed the opposite trend. This could be related to the origin of heavy metals, which resulted in different degrees of physical and chemical bonds with the sediments in different environmental conditions (Pempkowiak et al. 1999).

Metal enrichment

The current study adopted the geochemical normalization method to measure the degree of metal pollution in Marudu Bay. The geochemical normalization has proved to be superior compared to the granulometric method, because it compensates for the mineralogical as well as natural granular variability of the metal concentration in the sediment (Loring 1991). In addition, the geochemical normalization can also minimize the contamination due to minimal manipulation of a sample (Din 1992; Shazili et al. 2006).

Al has been frequently used as a reference element in the study of marine sediment, due to its natural origin with minimal human impact (Din 1992; Trimble et al. 1999; Weisberg et al. 2000). However, Fe and Li acted as reference elements for various heavy metals in Marudu Bay, whereas Al rarely did, which suggests that heavy metals in Marudu Bay had been influenced by anthropogenic sources (Weiguo et al. 2001).

In the V/Al regression plot, the high regression coefficient of r^2=0.895 was obtained without removing any data points. In addition, the enrichment study also suggests that minimum or no enrichment with V occurred in Marudu Bay. This clearly indicates that V in Marudu Bay is currently not affected by anthropogenic activities. For other elements, various data points were suspected of being enriched, and therefore were removed until r^2>0.9 was achieved for the purpose of accurate baseline concentration determination. Mn and Zn enrichment was observed at the stations located near areas of high human population density. The anthropogenic sources of Mn and Zn are mainly contributed by paint, metal components and sewage effluents (Alluri et al. 2007; Ideriah et al. 2012). Therefore, the increase in the Zn and Mn enrichment factor could be contributed by river runoff of industrial and domestic effluents.

On the other hand, Rb and Sr seem to be transported from other stations and selectively accumulated in the mouth of the bay (station 6), because Rb and Sr are strongly bound to the organic detritus matter (Xu et al. 2010). Transporting of organic matter from the shallow water (Ekman layer) to deep water (station 6, depth >20 m) indirectly carried also Rb and Sr. Some metals, including Cr, Cu, Ag, Ni, Ca and Pb, were not correlated with any reference elements. Therefore, the enrichment factor cannot be calculated. Since the concentration of these metals was too low and did not coincide with any reference elements, the combination of geochemical and granulometric normalization should be considered in future studies to minimize the grain-size effect on metal concentrations.

Conclusion

Organic matter in the sediment of Marudu Bay comes mainly from terrestrial sources. The heavy metal concentrations in the sediment of Marudu Bay are currently within the allowable level and pose no threat to public health. In addition, the levels of heavy metals in the sediment of Marudu Bay are similar or lower compared to other regions. However, localized anthropogenic enrichment with heavy metals, particularly Cu, might require attention. Spatial and temporal monitoring strategies are critical to combat and manage heavy metal pollution in Marudu Bay.

Acknowledgments

This work was financially supported by the Niche Research Grant Scheme (NRGS0003) from the Ministry of Higher Education Malaysia.

References

Adam, A., Mohammad-Noor, N., Anton, A., Saleh, E., Saad, S. et al. (2011). Temporal and spatial distribution of Harmful Algal Bloom (HAB) species in coastal waters of Kota Kinabalu, Sabah, Malaysia. *Harmful Algae* 10: 495-502.

Agrawal, Y.C. & Pottsmith, H.C. (2000). Instruments for particle size and settling velocity observations in sediment transport. *Marine Geology* 168: 89-114.

Ali, S.A.M., Tan, W.H., Tair, R., Naser, A.A. and Sualin, F. (2014). Surface sediment analysis on heavy metals in coastal area of UMS- Tuaran, Sabah. *Borneo Science* 34: 6-10.

Alluri, H.K., Ronda, S.R., Settalluri, V.S., Suryanarayana, V. & Venkateshwar, P. (2007). Biosorption: An eco-friendly alternative for heavy metal removal. *African Journal of Biotechnology* 6(11): 2924-2931.

Anderson, D.M., Glibert, P.M. & Burkholder, J.M. (2002). Harmful algal blooms and eutrophication: Nutrient sources, composition, and consequences. *Estuaries* 25: 704-726.

Anderson, D.M. (2009). Approaches to monitoring, control and management of harmful algal blooms (HABs). *Ocean and Coastal Management* 52: 342.

Azanza, R.V., David, L.T., Borja, R.T., Baula, I.U. & Fukuyo, Y. (2008). An extensive *Cochlodinium* bloom along the western coast of the Palawan, Philippines. *Harmful Algae* 7: 324-330.

Bao, S.D. (2005). Agricultural soil analysis, third ed. China Agriculture press, Beijing.

Birch, G.F., Taylor, S.E. & Matthai, C. (2001). Small-scale spatial and temporal variance in the concentration of heavy metals in aquatic sediments: a review and some new concepts. *Environmental Pollution* 113: 357-372.

Bradl, H. (2005). Heavy metals in the environment: Origin, interaction and remediation. Elsevier Academic Press, London.

Carpenter, E.J. & Capone, D.J. (1983). Nitrogen in the marine environment. Stony Book, Marine Science Research Center. 900p.

Colizza, E., Fontolan, G. & Brambati, A. (1996). Impact of a coastal disposal site for inert wastes on the physical marine environment, Barcola-Bovedo, Trieste, Italy. *Environmental Geology* 27(4): 270-285.

Covelli, S. & Fontolan, G. (1997). Application of a normalization procedure in determining regional geochemical baseline. *Environmental Geology* 30: 34-45.

Dasklakis, K.D. & O'Connor, T.P. (1995). Normalization and elemental sediment contamination in the coastal United States. *Environmental Science and Technology* 29: 470-477.

Date, A.R. & Gray, A.L. (1988). Applications of inductivity coupled plasma source mass spectrometry. Blackie Publishers, Glassgow.

Din, Z. (1992). Use of aluminium to normalize heavy-metal data from estuarine and coastal sediment of Strait of Melaka. *Marine Pollution Bulletin* 24: 484-491.

Hanson, P., Evans, D.W. & Colby, D.R. (1993). Assessment of elemental contamination in estuarine and coastal environments based on geochemical and statistical modelling of sediments. *Marine Environmental Research* 36: 237-266.

Ideriah, T.J.K., David-Omiema, S. & Ogbonna, D.N. (2012). Distribution of heavy metals in water and sediment along Abonnema Shoreline, Nigeria. *Resources and Environment* 2(1): 33-40.

Inengite, A.K., Oforka, N.C. & Leo, C.O. (2010). Survey of heavy metals in sediments of Kolo creek in the Niger Delta, Nigeria. *Africa Journal of Environmental Science and Technology* 4(9): 558-566.

Ismail, A., Badri, M.A. & Ramlan, M.N. (1993). The background levels of heavy metal concentration in sediments of the west coast of Peninsular Malaysia. *The Science of the Total Environment Supplement* 134: 315-323.

Kamaruzzaman, B.Y., Mohd Zahir, M.S., Akbar John, B., Jalal, K.C.A., Shahbudin, S. et al. (2011). Bioaccumulation of some heavy metal by green mussel *Perna viridis* (Linnaues 1758) from Pekan, Pahang, Malaysia. *International Journal of Biological Chemistry* 5(1): 54-60.

Lakherwal, D. (2014). Adsorption of heavy metals: A review. *International Journal of Environmental Research and Development*. 4(1): 41-48.

Loring, D.H. (1990). Lithium- a new approach for granulometric normalization of trace metal data. *Marine Chemistry* 29: 155-168.

Loring, D.H. (1991). Normalization of heavy metal data from estuarine and coastal sediments. ICES *Journal of Marine Science* 48: 101-115.

Malaysian Meteorological Department. Annual Report (2014). Putrajaya, Malaysia: Malaysian Meteorological Department.

Matthai, C., Birch, G.F., Szymezak, R. (1998). Suboxic, early diagenetic process in surficial sediments near a deepwater ocean outfall, Sydney, Australia. *Journal of Geochemical Exploration* 64: 1-17.

Middleton, R. & Grant, A. (1990). Heavy metals in the Humber estuary: Scrobicularia clay as a preindustrial datum. *Proceeding of the Yorkshire Geological Society* 48: 75-80.

Mokhtar, M., Awaluddin, A. & Low, Y.G. (1994). Water quality of Inanam river estuary and the Ko-Nelayan tiger prawn aquaculture ponds in Sabah, Malaysia. *Hydrobiologia* 285(1): 227-235.

Molinaroli, E., Guerzoni, S., Saretta, A., Mosiol, M. & Pistolato, M. (2009). Thirty-year changes (1970 to 2000) in bathymetry and sediment texture recorded in the Lagoon of Venice sub-basins, Italy. *Marine Geology* 258: 115-125. DOI: 10.1016/j.margeo.2008.12.001.

Murray, K.S. (1996). Statistical comparisons of heavy metal concentration in river sediments. *Environmental Geology* 27: 54-58.

Newman, B.K. & Watling, R.J. (2007). Definition of baseline metal concentrations for assessing metal enrichment of sediment from the south-eastern Cape coastline of South

Africa. *Water SA* 33(5): 675-691.

Pempkowiak, J. (1992). Enrichment factors of heavy metals in the Southern Baltic surface sediments dated with Pb-210 and Cs-134. *Environment International* 17: 421-428.

Rae, J.E. (1997). Trace metals in deposited intertidal sediment, in biogeochemistry of intertidal sediments. In T.D. Jickells, J.E. Rae (Eds.), (pp. 16-31). Cambridge University Press.

Rubio, B., Nombela, M. & Vilas, F. (2000). Geochemistry of major and trace elements in sediments of the Ria de Vigo (NW Spain): an assessment of metal pollution. *Marine Pollution Bulletin* 40(11): 968-980.

Sany, S.B.T., Hashim, R., Rezayi, M., Salleh, A. & Safari, O. (2014). A review of strategies to monitor water and sediment quality for a sustainability assessment of marine environment. *Environment Science Pollution Research* 21: 813-833.

Schiff, K.C. & Weisberg, S.B. (1999). Iron as a reference element for determining trace metal enrichment in Southern California coastal shelf sediments. *Marine Environmental Research* 48: 161-176.

Shazili, N.A.M., Yunus, K., Ahmad, A.S., Abdullah, N. & Abd Rashid, M.K. (2006). Heavy metal pollution status in the Malaysian aquatic environment. *Aquatic Ecosystem Health and Management* 9(2): 137-145.

Siddique, A., Mumtaz, M., Zaigham, N.A., Mallick, K.A., Saied, S. et al. (2009). Heavy metal toxicity levels in coastal sediments of Arabian Sea along the urban Karachi (Pakistan) region. *Marine Pollution Bulletin* 58(9): 1406-1414.

Spagnoli, F., Bartholini, G., Dinelli, E. & Giordano, P. (2008). Geochemistry and particle size of surface sediments of Gulf of Manfredonia (Southern Adriatic Sea). *Estuarine Coastal and Shelf Science* 80: 21-30.

Storlazzi, C.D. & Field, M.E. (2000). Sediment distribution and transport along a rocky, embayed coast: Monterey Peninsula and Carmel Bay, California. *Marine Geology* 170: 289-316.

Sulochanan, B., Krishnakumar, P.K., Prema, D., Kaladharan, P., Valsala, K.K. et al. (2007). Trace metal contamination of the marine environment in Palk Bay and Gulf of Mannar. *Journal of Marine Biology Association India* 49(1): 12- 18.

Tan, K.S. & Ransangan, J. (2015a). Factors influencing the toxicity, detoxification and biotransformation of paralytic shellfish toxins. In D.M. Whitacre (Ed.), *Reviews of environmental contamination and toxicology Volume 235* (pp. 1-25). Springer International Publishing Switzerland.

Tan, K.S. & Ransangan, J. (2015b). Factor influence the larvae distribution and spat settlement of Perna viridis in Marudu Bay, Sabah, Malaysia. *Advance in Environmental Biology* 9(17): 18-23.

Tan, K.S. & Ransangan, J. (2016). Feeding behaviour of green mussels, *Perna viridis* farmed in Marudu bay, Malaysia. *Aquaculture Research* 1-16. DOI: 10.1111/are.12963.

Tan, K.S., Denil, D.J. & Ransangan, J. (2016). High mortality and poor growth of green mussels, *Perna viridis* in high chlorophyll-a environment. *Ocean Science Journal* In press

Trimble, C.A., Hoenstine, R.W., Highley, A.B., Donoghue, J.F. & Ragland, P.C. (1999). Baseline sediment trace metals investigations: Steinhatchee river estuary, Florida, northeast Gulf of Mexico. *Marine Georesources and Geotechnology* 17(2-3): 187-197.

Tyson, R.V. (1995). Sedimentary organic matter. Organic facies and palynofacies. London: Chapman and Hall. 615p.

Wang, W.X., Yan, Q.L., Fan, W. & Xu, Y. (2002). Bioavailability of sedimentary metals from a contaminated bay. *Marine Ecological Progress Series* 240: 27-38.

Weiguo, Z., Lizhong, Y. & Hutchinson, S. (2001). Magnetic normalization of particle size effects in a heavy metal pollution study of intertidal sediments from the Yangzte estuary. *Science in China* 44: 185-189.

Weisberg, S.B., Wilson, H.T., Heimbunch, D.G., Windom, H.L. & Summers, J.K. (2000). Comparison of sediment metal: Aluminium relationships between the eastern and gulf coasts of the United States. *Environmental Monitoring and Assessment* 61(3): 373-385.

Xu, H., Liu, B. and Wu, F. (2010). Spatial and temporal variations of Rb/Sr ratios of the bulk surface sediments in Lake Qinghai. *Geochemical Transactions* 11(3).

Yang, S.L., Li, H., Ysebaert, T., Bouma, T.J., Zhang, W.X. et al. (2008). Spatial and temporal variations in sediment grain size in tidal wetlands, Yangtze Delta: On the role of physical and biotic controls. *Estuarine Coastal and Shelf Science* 77: 657-671.

Yap, C.K., Ismail, A. & Tan, S.G. (2004). Heavy metal (Cd, Cu, Pb and Zn) concentrations in the green-lipped mussel *Perna viridis* (Linnaeus) collected from some wild and aquaculture sites in the west coast of Peninsular Malaysia. *Food Chemistry* 84: 569-575.

Yap, C.K., Ismail, A., Tan, S.G. & Rahim Ismail. A. (2006). Is gender a factor contributing to the natural variations in the accumulation of heavy metals (Cd, Cu, Pb and Zn) by the green-lipped mussel *Perna viridis*? *Indian Journal of Marine Science* 35: 29-35.

Yap, C.K., Edward, F.B. & Tan, S.G. (2007). Determination of heavy metal distribution in the green mussel *Perna viridis* as bioindicators of heavy metal contamination in the Johore Straits and Senggarang Peninsular Malaysia. *Trends in Applied Science Research* 2(4): 284-294.

Yap, C.K. & Al-Barwani, S.M. (2012). A comparative study of condition indices and heavy metals in *Perna viridis* population at Sebatu and Muar, Peninsular Malaysia. *Sains Malaysiana* 41(9): 1063-1069.

Zaaboub, N., Oueslati, W., Helali, M.A., Abdeljaouad, S.A., Huertas, F.J. et al. (2014). Trace elements in different marine sediment fractions of the Gulf of Tunis (Central Mediterranean Sea). *Chemical Speciation and Bioavailability* 26(1): 1-12.

Zhang, J. (1995). Geochemistry of trace metals from Chinese River/ Estuary systems: an overview. *Estuarine Coastal and Shelf Science* 41: 631-658.