

The Chemometric Approach as a Useful Tool in the Identification of Metal Pollution Sources of Riverine-Mangrove Sediment of Kota Marudu, Sabah, Malaysia

Ahmad Zaharin Aris^a, Nadzhratul Husna Ahmad Puad^a, Nur Aliaa Shafie^a, Looi Ley Juen^a, Sarva Mangala Praveena^b, Mohammad Firuz Ramli^a and Mohd Kamil Yusoff^a

^a *Environmental Forensics Research Centre, Faculty of Environmental Studies, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

^b *Department of Environmental and Occupational Health, Faculty of Medicine and Health Sciences, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

Abstract

The chemometric statistical approach was applied to evaluate the level of metals accumulation in sediment and to identify the probable pollution sources in the riverine-mangrove ecosystem of Kota Marudu, East Malaysia. Parameters, such as pH, electrical conductivity, salinity, organic matter, lead (Pb), cadmium (Cd), copper (Cu), chromium (Cr), zinc (Zn), aluminum (Al), nickel (Ni) and iron (Fe) were determined from sediment samples collected from 17 sampling points located throughout the district of Kota Marudu, Sabah. The results from cluster analysis indicate the presence of two prominent clusters that represent sources of pollution that might be induced from natural sources and human activities. Meanwhile, principal analysis from this study has proven that pH, electrical conductivity, salinity, Cd and Cr are responsible for the large spatial variations explaining 31.73% of the total variance, whilst organic matter, Fe and Al explain 24.75% of the total variance. The third factor is followed by Cu and Zn with 15.35% whereas Pb and Ni account for a total variance of 14.44%. The present study reveals the usefulness of the chemometric statistical approach as a remarkable and useful tool to reveal meaningful information concerning the spatial variability of large and complex riverine-mangrove data.

Keywords: chemometric; metals; sediment; HACA; PCA

1. Introduction

Today, the toxicity, persistence and bioaccumulation ability of metals in the natural environment has become one of the most severe environmental problems. Most of the high metal concentrations are derived from anthropogenic sources, especially from industrial activities, discarded automobiles, batteries and wastewater discharge (Bloom and Ayling, 1977; Shriadah, 1999; Marchand *et al.*, 2006; Pekey, 2006). Metals are deposited on the surface of the sediment once it has been transported by the water body (Dwivedi and Padmakumar, 1983), and cannot be degraded either biologically or chemically. They can only be transported over long distances or accumulate in the ecosystem (Marchand *et al.*, 2006). Mangrove ecosystems are very much affected by human activities since they are close to the urban areas (Dwivedi and Padmakumar, 1983) and receive significant pollution loads from the terrestrial environment. They play an important role as a sink for pollutants in the coastal environment (Harbison, 1986). Therefore, the physical and chemical properties of such a complex and dynamic ecosystem

provide useful insights into the probable pollution sources of metals.

Practically, chemometric studies (multivariate statistical approaches) have been widely incorporated with spatial distribution studies in drawing meaningful information from a large amount of data (Davis, 1986; Praveena *et al.*, 2007). Recently, these methods have become important tools in evaluating the complex relationships in environmental sciences (Brown *et al.*, 1994, 1996; Alberto *et al.*, 2001; Juahir *et al.*, 2010; Aris *et al.*, 2012). Generally, these methods are chosen to identify key variables for environmental monitoring purposes and similar contaminant sources (Looi *et al.*, 2013). Principal component analysis (PCA) and hierarchical agglomerative cluster analysis (HACA) are the most common statistical methods used in the interpretation of environmental data (Praveena *et al.*, 2007). These methods are used to categorize variables or data with respect to predetermined criteria (Facchinelli *et al.*, 2001; Praveena *et al.*, 2007; Lim *et al.*, 2012). Since the environment is characterized by inherent variability (natural, anthropogenic, spatial/temporal), a combination of PCA and HACA is used to reveal their

relations, the limits and the hierarchy of the interaction between the components.

The riverine and mangrove ecosystem of Kota Marudu support a number of distinct habitats, and are home to primate species, mammals, birds, insects, fish and reptiles (Malaysian National Oceanography Data Centre (MyNODC, 2010). However, there is no detailed information and preliminary record on the metal study of Kota Marudu mangroves and riverine ecosystems. Therefore, riverine-mangrove sediments from Kota Marudu were chosen for this study to assess the current metals pollution status. The present study aimed to determine the spatial distribution of selected heavy metals and also to identify the controlling factors governing the behavior of the metals in the sediment using PCA and HACA at Kota Marudu riverine-mangroves ecosystems. It is hoped that Kota Marudu could benefit from the initiative and record of the pollution status, especially concerning its sediment and serve as a reliable record for future reference. This research can also provide a platform for better management at Kota Marudu riverine-mangroves area and be of value for specific authorities, such as the Department of Environment Malaysia, Department of Mineral and Geosciences, Ministry of Tourism Malaysia, and the Department of Marine Parks Malaysia in managing environmental pollution.

2. Materials and Methods

2.1. Sampling and analytical procedure

A total of seventeen sediment samples (0-20cm) were collected in triplicate using an Ekman grab sampler

in August 2010 and homogenized into an acid-washed zip locked bag. The exact location for each sampling point is illustrated in Fig. 1.

The locations of the sampling points were determined prior to the sampling and positioned in a way that would reflect the general condition of the study area. The sampling points were selected based on the ecological setting and human activity in the study area. This setting might also provide an opportunity to assess the sampling network and indicate whether, in the future, more or possibly fewer sediment samples would be needed to determine the quality status at various points throughout the ecosystem setting (Aris et al., 2013). The collected samples were stored at 4 °C prior to analysis (Lim et al., 2012). Then the samples were air-dried and ground using an agate pestle and mortar before being sieved through a 2.0 mm (for the determination of the physicochemical parameters) and 63µm sieve (for heavy metal analysis). In order to minimize the mineral contamination, all the apparatus used for the analyses were soaked overnight in 5% HNO₃ and rinsed with distilled water prior to analysis.

Physicochemical parameters, such as pH, electrical conductivity (EC) and salinity were determined by immersed electrodes in a 1:2 w:v ratio for soil to water. For metal analysis, a fine fraction (< 63µm) sample was chosen as it retained higher metal content compared to other larger bulk sediment sizes. About 1.0g of sediment samples were digested using the aqua regia (HCl:HNO₃ = 1:3) method suggested by Radojevic and Bashkin (2007). This method is sufficient for total metals extraction from sediment samples and it is widely used in most soil analyses (Radojevic and Bashkin, 2007). Later, the extracted sample was filtered through

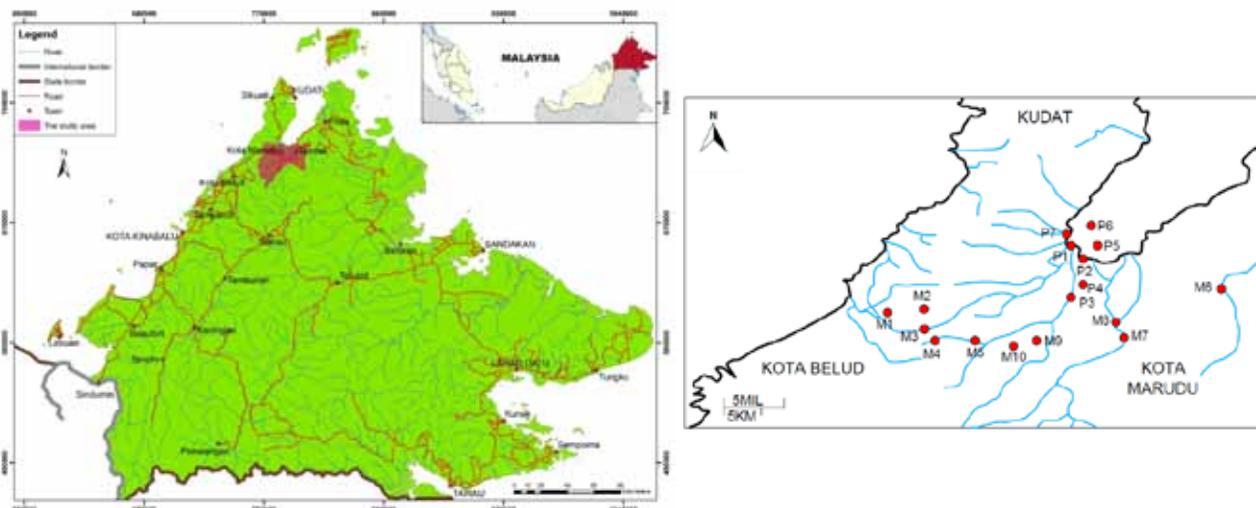


Figure 1. Location of sampling points in Kota Marudu, Sabah

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Whatman filter paper (0.45 µm) and kept in a plastic bottle before it was analyzed by using Inductively Coupled Plasma Mass Spectrophotometry (ICP-MS, Elan DRC-e, Perkin Elmer). The ICP-MS can analyze most of the elements in the periodic table. For each element measured, it is commonly needed to measure just one isotope, since the natural abundance is fixed in nature (PerkinElmerSCIEX Instruments, n.d.). The isotopes analyzed in this study were ^{106}Pb , ^{114}Cd , ^{63}Cu , ^{52}Cr , ^{64}Zn , ^{27}Al , and ^{58}Ni .

Quality control and analysis procedures to obtain good quality and representative results were performed by calibrating the probe meters, analyzing the blank samples and preparing external standards for calibration curves. The calibration curves for each trace element were determined using the 10 mg/L of ICP Multi Element Calibration Standard 3 (Perkin Elmer Pure Plus). In addition, the instrument was operated in accordance with the manufacturer's specifications; the operating conditions are given in Table 1. The performance of the ICP-MS was checked daily to ensure the conditions of the instrument were within the specifications. The extracted sediment samples were analyzed for heavy metal concentrations when all the prerequisite settings of the ICP-MS were in accordance with the accuracy check. Since the detection limits of the ICP-MS were in the range 1-10 µg/L, samples beyond the detection limit were diluted to conform with the acceptable ranges that would allow the detection by ICP-MS. A mean value was calculated for each parameter, with the standard deviation (SD)

being used as an indicator of the precision measured in triplicate.

2.2. Chemometric statistical methods

2.2.1. Principal component analysis

Principal component analysis (PCA) was used in this study, because of its ability to identify and interpret the relationship between variables in the sediment. In addition, it is a useful tool to identify the pattern of correlations within a set of observed variables. PCA is a chemometric statistical method that allows data reduction with minimum loss of original data. Moreover, it is designed to transform the original variables into the new uncorrelated variables called factors, which are linear combinations of the original variables (Alkarkhi *et al.*, 2009). PCA extracts eigenvalues and related loadings from the covariance matrix of the original variables to produce new orthogonal variables through varimax rotation (Mustapha, 2011). Factor loadings are classified as 'strong', 'moderate', and 'weak' with corresponding loading values of >0.75, 0.5-0.75 and 0.30-0.50, respectively (Mustapha, 2011; Liu *et al.*, 2003). Therefore, it is able to identify the most significant factors that explain the whole dataset (Richard and Dean, 2002).

2.2.2. Hierarchical agglomerative cluster analysis

Hierarchical agglomerative cluster analysis (HACA) was used to classify the variables into categories or clusters based on their similarities while

Table 1. Instrument conditions for ICP-MS Model Elan DRC-e

Power	1100 W
Argon plasma gas flow rate	17.00 L/min
Argon auxiliary gas flow rate	1.20 L/min
Argon nebulizer gas flow rate	0.72 L/min
Scan Mode	Peak Hopping
Sweeps per reading	10
Replicates	3
Dwell time	50 ms
Integration time	1500
Estimated Reading time	3.18 s
Estimated replicate time	9.54 s
Estimated sample time	28.62 s
Sample flush	30s 48 rpm
Sample read delay	20s 25 rpm
Sample wash time	30s 48 rpm

Table 2. Concentration of heavy metals in sediment compared with Interim Sediment Quality Guidelines (ISQG)

Parameter	Min	Mean	Max	SD	Interim Sediment Quality Guideline	
					ISQG-low	ISQG-high
Pb ($\mu\text{g}/\text{kg}$)	1.3	53.6	164.6	41.3	50000	220000
Cd ($\mu\text{g}/\text{kg}$)	380.0	18291.8	49638.0	17908.3	1500	10000
Cu ($\mu\text{g}/\text{kg}$)	6.4	247.6	809.0	378.4	65000	270000
Cr ($\mu\text{g}/\text{kg}$)	108.7	423.6	1173.5	322.6	80000	370000
Zn ($\mu\text{g}/\text{kg}$)	145.2	450.5	895.4	200.9	200000	410000
Al ($\mu\text{g}/\text{kg}$)	51660.0	134712.8	259146.0	68170.5	-	-
Ni ($\mu\text{g}/\text{kg}$)	130.0	673.5	1975.0	548.7	21000	520000
Fe ($\mu\text{g}/\text{kg}$)	85912.0	185892.4	319822.6	77900.0	-	-

recognizing the actual groups (Alkarkhi *et al.*, 2009; Richard and Dean, 2002). HACA is often used to find an optimal grouping for which the observations or objects within the cluster are similar and dissimilar between the clusters (Alkarkhi *et al.*, 2009). The variables with most similarity to each other are grouped, and these initial groups are combined according to their similarities (Richard and Dean, 2002). The output of HACA is illustrated by a dendrogram presenting the clusters and their similarity. In this study, HACA was performed on sediment data from Kota Marudu in order to group variables into statistically significant clusters for their similarity and dissimilarity of pollution sources.

3. Results and Discussion

The descriptive statistics of studied heavy metals are shown in Table 2. The highest concentration was detected in Fe. This might be due to the conservative lithogenic element factor. This was followed by the concentrations of Al and Cd. By comparing the data in this study with the Interim Sediment Quality Guidelines (ISQG) in Table 2, the mean concentration of most metals (except Cd and Al) in the sediment samples analyzed were below ISQG-low and ISQG-high ($\mu\text{g}/\text{kg}$) of the Australian and New Zealand Environment and Conservation Council (ANZECC). According to Praveena *et al.* (2008), sediment quality guidelines are used to protect aquatic organisms from the harmful effects related to sediment-bound contamination as well as useful tools for evaluating the potential contaminants within the sediment, which might induce biological effects in an aquatic environment. There are two-sets of values proposed in ISQG: ISQG-low is for a particular contaminant that is not excessive, the element is unlikely to cause any biological effect on organisms inhabiting that sediment and ISQG-high for which the

biological impact is likely to be high. In this study, Cd was found to have a concentration value above the Interim Sediment Quality Guideline (ISQG-high). The discharge of waste from the nearest motor workshops into the river was expected to be one of the contributors to Cd accumulation in sediment. Based on the DOE (1994) report, many motor workshops in urban areas deposit their waste into drains, which eventually flows into the rivers and ends up in the sea, which directly impacts the level of Cd in the sediment.

The metal distribution in sediment follows the order: Fe > Al > Cd > Zn > Ni > Cr > Pb > Cu (Fig. 2). One-way ANOVA analysis showed that the metal concentrations are significantly different between sampling points ($p < 0.05$). The variation in mineralogical composition, rates of mineralization and surrounding land use activities might influence the different amounts of heavy metals accumulated in the samples collected from the study area. Land use within the river basin and coastal area is expanding and is primarily associated with traditional agriculture activities along the lower reaches of the river. Meanwhile, the influence of river flow on sediment transport in the study area also plays a role in the metal distribution and concentration. As the water current increases, the sediment particles are lifted into the water column and transported downstream towards the estuary (Lim *et al.*, 2012). Therefore, large amounts of sediment are transported and may accumulate in the estuary and thus account for a higher level of metal concentration compared to upstream. The ongoing anthropogenic activities in the landward areas cause the accumulation of metals in this estuarine zone.

PCA was carried out on the data set in order to determine the relationship between variables in the sediment. Table 3 displays factor loadings after being rotated with varimax rotation as well as the initial eigenvalues, percentage of variance and cumulative

Table 3. Rotated component matrix

Variable	Factor			
	1	2	3	4
pH	-0.733	-0.197	-0.373	-0.255
Electrical Conductivity	0.910	0.053	-0.091	0.173
Salinity	0.937	0.209	-0.089	-0.063
Cd	0.856	0.273	0.305	-0.132
Cr	0.614	0.298	-0.017	0.580
Fe	0.190	0.918	-0.098	0.063
Al	0.168	0.892	0.253	0.067
Organic Matter	0.272	0.833	-0.040	0.234
Cu	0.110	-0.167	0.943	0.071
Zn	-0.061	0.501	0.790	-0.215
Ni	0.293	0.320	0.005	0.784
Pb	0.463	0.087	0.076	-0.742
Initial Eigenvalue	5.175	1.936	1.916	1.326
Percentage of variance	31.726	24.753	15.352	14.443
Cumulative percentage	31.726	56.480	71.831	86.274

percentage. PCA yielded four factors with eigenvalues greater than 1, explaining 86.27% of the total variance in the data set with more than 56.48% of the variance being explained in the first two principal components (PC). Ideally, an eigenvalue gives a measure of the significance of the factors. For instance, the factor with the highest eigenvalue is the most significant component and responsible for explaining the large variation in the data set (Alkarkhi *et al.*, 2009). The bold values highlighted in Table 3 represent the factor loadings of the variables that are greater than 0.5. The factor loadings are classified as 'strong', 'moderate', and 'weak' with corresponding loading values of >0.75 , $0.5-0.75$ and $0.30-0.50$, respectively (Mustapha, 2011; Liu *et al.*, 2003).

The first factor (F1) accounted for 31.73% of the total variance and a strong positive loading was illustrated by electrical conductivity, salinity, pH, cadmium (Cd), and chromium (Cr). These strong and moderate correlations in the first factor can be clearly proven by the results obtained using Pearson's correlation (Table 4). According to Church (1989) and Praveena *et al.* (2010), 90% of seawater usually contains 3.5% salinity, of which 90% is fully ionized ions, which interacts with conductivity in the sediment during high tide. The association with pH shows that seawater can also influence the pH changes in the buffering process (Hsue and Chen, 2000).

Meanwhile, factor two (F2) signifies organic matter, Al and Fe, as factor loadings with a total

variance of 24.75%. According to Preda and Cox (2000), additional ions can come from the tidal flooding and ion exchange that occur in the system. Moreover, the presence of hydrous aluminum, magnesium or iron silicates in the clay minerals practically induce these elements to be in the same factor.

From the factor loadings for factor three (F3), it was observed that Cu has a moderately significant positive correlation with Zn at $p < 0.05$. The percentage of variance recorded for Cu and Zn was 15.35% at a high loading. This indicates that the precipitation of Cu may be related to the anthropogenic sources and also the complex nature of clay minerals. Cu is among the most abundant metals that can be found in urban areas, industrial waste and sewage, which finally forms particulate and dissolved metals (Kamaruzzaman *et al.*, 2011). On the other hand, zinc (Zn) of natural origin is normally found in the sediment, which is practically derived from the weathering of ultramafic rocks (Kamaruzzaman *et al.*, 2006). In this study, the use of anti-rust paints on the fishing boats may cause the presence of Zn in the sediment. Both elements are correlated with each other, probably because they are mostly abundant in the environment and may also be induced by the natural origin of rocks.

The percentage variance accounted for by factor four (F4) is 14.44% and signifies that Nickel (Ni) and Lead (Pb) have the strongest loading among the others. The adsorption of Ni onto the sediment is believed to be related to the microbial degradation of organic

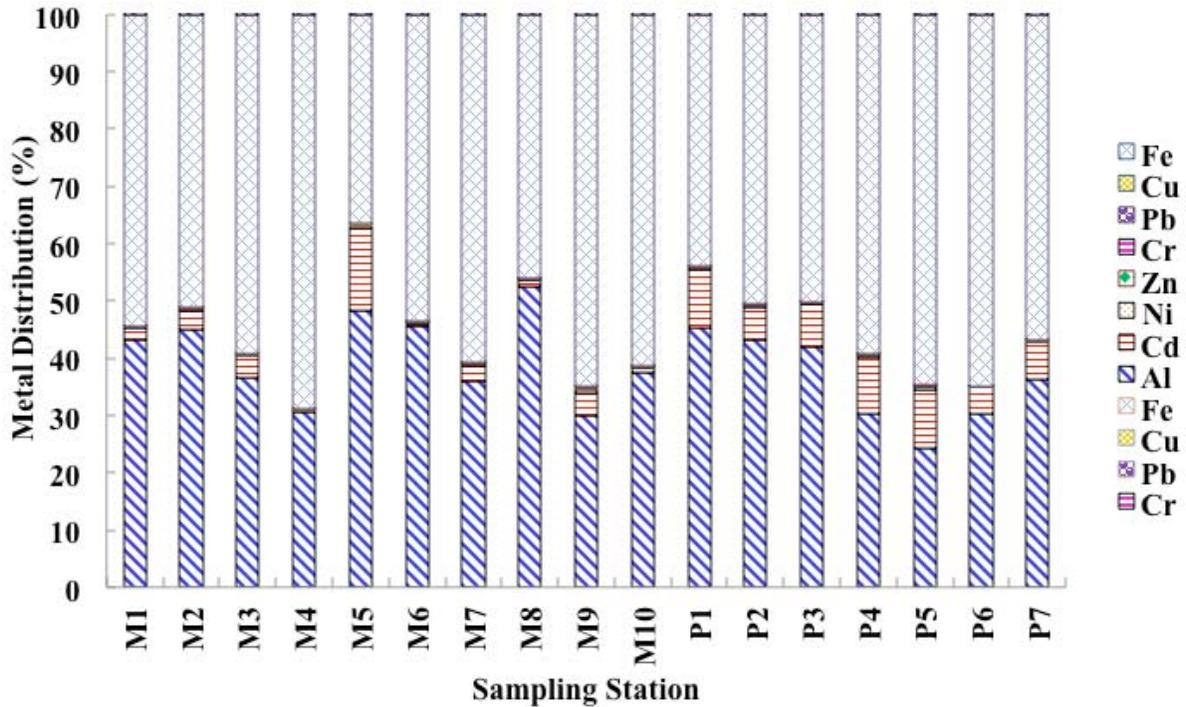


Figure 2. Spatial distribution of selected metals in the sediment samples of Kota Marudu, Sabah, Malaysia

materials, which leads to the rapid removal of the Ni adsorbed on the surface of the clay and detritus particles (Presley *et al.*, 1972). According to EHC 85 (1991), the regular use of nickel as marine anti-fouling paint on the bodies of the boats may contribute to the presence of Ni in the sediment. In this study, station M6 was found to be a boat parking place that might contribute

trace amounts of Ni from the anti-fouling paint used on the boats. However, the correlation of Ni and Pb was not significant to each other. This may be because the elements' pollution in Kota Marudu that poses both of these elements falls in the same factor.

The hierarchical agglomerative cluster analysis was used to confirm and verify the results of factor analysis.

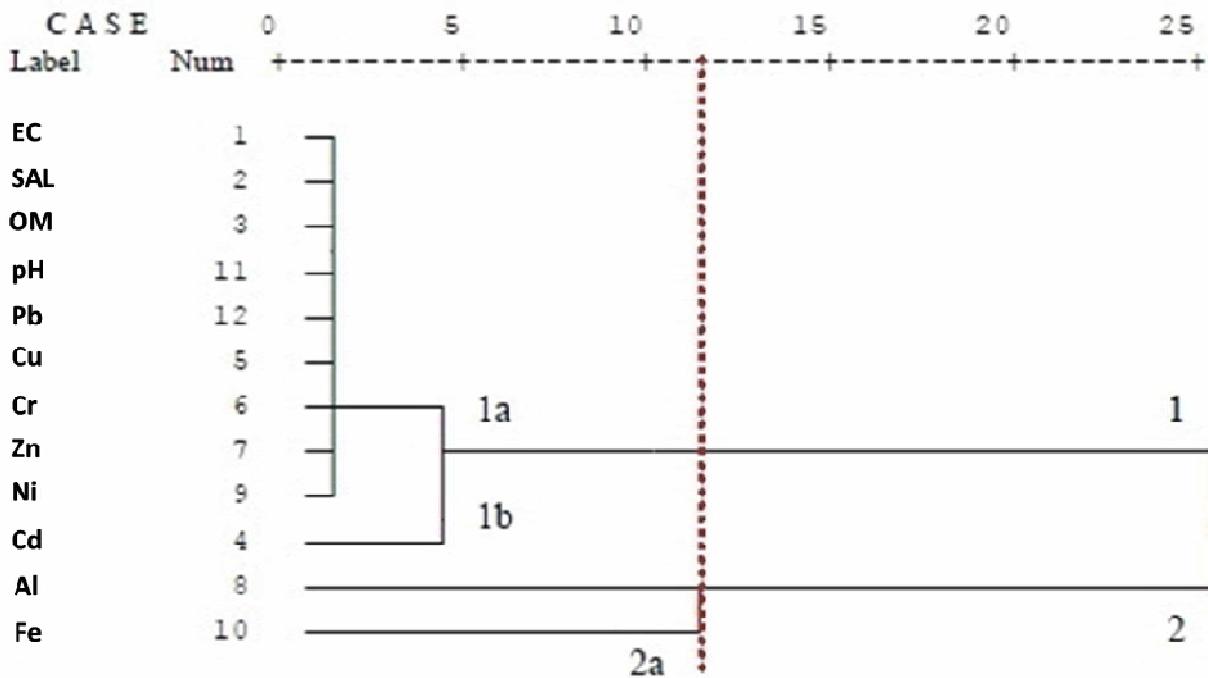


Figure 3. Dendrogram showing hierarchical cluster analysis in the sediment (EC=conductivity; SAL=salinity; OM=organic matter)

The analysis was performed using between groups' linkage (Rescaled Distance Cluster Combine) and portrayed by a dendrogram through hierarchical cluster analysis, as shown in Fig. 3. From the dendrogram, it indicates that there are two distinct groups formed based on the similarity that each variable poses towards each other. In this study, the variability in the metals in the sediment was successfully classified into two distinct factors, namely, the anthropogenic factor and the litogenic factor.

The first cluster identified electrical conductivity, salinity, organic matter, pH, Pb, Cu, Cr, Zn, Ni and Cd, while categories Al and Fe are cluster 2. The variables in the first cluster are similar to the factor loadings in factor one. Strong and significant correlations can be seen from the variables in the first cluster; Cd-Cr ($r = 0.577$; $p < 0.05$), Cu-Zn ($r = 0.604$; $p < 0.05$) and Cr-Ni ($r = 0.778$; $p < 0.01$). The conductivity, salinity, organic matter and pH, which represent the physicochemical parameters, are related to each other. The indirect contact with atmosphere deposition also contributes to the presence of Pb (Liu *et al.*, 2003). Organic matter in the sediment usually has a higher ion exchange capacity compared to sediment colloids, which results in higher cation exchange capacity (Matagi *et al.*, 1998). From the first cluster, it is observed that there are definite sources of pollution, which mainly come from the anthropogenic activities, such as fishing activities, boat parking, harbor, residual oil waste from the nearest workshops, plastic and domestic waste, and agricultural activity. The pollution loads from the terrestrial environment are

transported to the mangrove and coastal areas throughout the riverine system of Kota Marudu. Meanwhile, the elements of Al and Fe in sediments are grouped into cluster 2. Good agreement can be seen from the strong and significant correlation existing between Al and Fe ($r = 0.769$; $p < 0.01$). They are found in abundance in the earth and are often related to each other. Basically, Al and Fe are derived from the natural environment, such as clay-bearing minerals of terrestrial and marine geological deposits (Zhou *et al.*, 2003; Praveena *et al.*, 2007).

4. Conclusion

This study has proven that PCA and HACA are remarkable and useful statistical tools to discover the influential factors involved in the sediment. This study also revealed that the concentration levels of selected metals (except Cd) in the riverine-mangrove sediments of Kota Marudu are still in the safe zone and can be considered clean as they do not exceed the permitted level of National Sediment Quality Guidelines (ISQG). The elevated concentrations of a certain metal can pose a serious threat to the ecosystem in the present and future timeframe. Therefore, monitoring works are crucial for continuous protection of riverine-mangrove ecosystem. The results from the present study can provide a baseline data for the sediment pollution status in Kota Marudu and other riverine-mangrove systems. In addition, the current findings also enhance the understanding concerning the application of the chemometric

Table 4. Pearson's correlation among all the variables.

	pH	Conductivity	Salinity	OM	Pb	Cd	Cu	Cr	Zn	Al	Ni	Fe
pH	1											
Conductivity	-.659**	1										
Salinity	-.658**	.932**	1									
OM	-.364	.462	.484*	1								
Pb	-.162	.160	.416	-.124	1							
Cd	-.713**	.703**	.807**	.406	.549*	1						
Cu	-.353	.036	-.002	-.057	.061	.323	1					
Cr	-.593*	.573*	.514*	.497*	.012	.577*	.027	1				
Zn	-.259	-.106	.011	.336	.200	.360	.604*	-.019	1			
Al	-.477	.211	.319	.786**	.118	.406	.100	.361	.585*	1		
Ni	-.421	.328	.264	.431	-.248	.249	.044	.778**	-.029	.367	1	
Fe	-.305	.178	.351	.765**	.158	.412	-.207	.450	.337	.769**	.454	1

** . Correlation is significant at the 0.01 level (2-tailed).

* . Correlation is significant at the 0.05 level (2-tailed).

statistical approach in the identification of probable pollution sources in the riverine-mangrove ecosystem. It reveals the usefulness of this tool as a remarkable and useful approach to reduce data in order to identify a small number of factors, which enable better information.

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Correspondence to

Associate Professor Dr. Ahmad Zaharin Aris
Environmental Forensics Research Centre,
Faculty of Environmental Studies,
Universiti Putra Malaysia, 43400
UPM Serdang, Selangor,
Malaysia
Tel: +603-8946-7455
Fax: +603-8943-8109
E-Mail: zaharin@upm.edu.my