

SPATIAL ANALYSIS OF HEAVY METALS IN MANGROVE ESTUARY AT EAST COAST PENINSULAR MALAYSIA: A PRELIMINARY STUDY

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ABSTRACT

The objectives of this preliminary study are to determine heavy metal concentration in mangrove estuary and to identify spatial patterns in the water quality based on heavy metals concentration. Principal Component Analysis (PCA) and Artificial Neural Networks (ANNs) were selected to analyze the dataset of six heavy metal parameters namely: Ni, Cu, Pb, Cd, As and Zn. PCA results show that the major source of water pollution in mangrove estuary is mostly due to the agriculture activities surface runoff. ANN results show a better prediction performance in discriminating between the regions with an excellent percentage of correct classification.

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This study presents the obligation and expediency of environmetric for the understanding of datasets directing to gain better information about water quality patterns in mangrove estuary based on spatial characterizations at the selected monitoring stations.

Keywords: marine water quality; PCA; ANN; heavy metal; mangrove.

1. INTRODUCTION

Marine water quality testing was compulsory for management of safe and reliable water sources in estuary. Various sources of pollution and destruction of mangrove forest decrease the quality of river water. Poor water quality is harmful to living organism and water ecosystem. The interaction of ground water and surface water was affected by human activities and natural process may cause unfavorable aquatic environment. Water pollution in aquatic ecosystem because of heavy metal is rising at a worrying stage. Based on [1], heavy metal are stored, absorbed or fused in water, sediment and aquatic animals and cannot be degraded. Heavy metal will undergo several processes (precipitation, sorption, complexation and dissolution) once they are released into the water system. These will disturb the aquatic environment quality which will harm the organisms in the ecosystem [2]. Besides, heavy metals have also been involved in forest deterioration because their deposition pattern is correlated with forest forfeiture especially in mangrove ecosystem [3]. Mangrove ecosystem act as a key crucial maker within estuarine systems which serve as natural habitat for fish and crustacean. The mangrove also deliver steadying for coastal landform and mitigation of erosion [4]. Mangrove forest shows decreasing in biodiversity result from mangrove forest ecosystems disturbance [5]. Generally, the mangrove systems (river, estuary and wetland) obtain numerous of pollutants and have become an immense pollution sink in recent decades [6]. Due to the increasing rate of human population, the exploration of mangrove forest through human activities such as logging activities, deforestation, agricultural land and waste from industry gives negative impact to the environment and also to water quality of mangrove forest [5, 7-9]. Scornfully, heavy metals are one of the main anthropogenic toxic compounds reported high in mangrove ascending from several sources including agriculture runoff [6]. Principal Component Analysis (PCA) is one of the chemometric technique which also known

as environmetric techniques have been applied in this study [10-13]. PCA is a powerful multivariate technique for pattern recognition that attempts to explain the large set of interrelated variables and transform them into a smaller set of independent (uncorrelated) variables (principal components) [11, 14]. Besides, PCA provides an understanding of the important patterns and underlying relationship in data, for getting deeper insight into the data and for modelling and visualizing complex data for more insightful analysis [10]. It provides information on the most significant parameters due to the spatial and temporal variations that describes the whole data set by excluding the less significant parameters with minimum loss of its original information [10-13, 15-17]. Artificial Neural Network (ANN) model is one of the application of random or deterministic models which can allow a prediction of the involvedness of the variables [18-21]. Several studies has proven that the ANN model is the best tool to model a non-linear environmental relationship [22]. ANN have the capability to learn non-linear relationships between the variables and complex patterns in datasets that are not well defined by simple mathematical formulae and they can be trained precisely when presented with a new dataset [19, 23]. According to [24], there are still of information on heavy metals in the Malaysian aquatic environment. Most of the studies about heavy metals in mangrove estuary are focused on west coast of peninsular Malaysia but very limited and uncommon studies in east coast of peninsular Malaysia. This is a preliminary study conducted to determine heavy metal concentration in mangrove estuary at East Coast Region, Peninsular Malaysia using PCA. This study also aims to establish an ANN model that can be used for discriminating between heavy metal concentrations in mangrove estuary according to region.

2. EXPERIMENTAL

2.1. Study Area

Monthly field sampling was conducted from June 2016 to September 2016 during southwest monsoon (dry season) at normal tide. Surface water was collected at four sampling stations (S1, S2, M1 and M2) and three stations as control (SM1, SM2 and SM3). Table 1 and Fig. 1 show the coordinate of sampling stations which situated at east coast of peninsular Malaysia and facing South China Sea. Setiu and Merang stations are situated at Setiu district,

Terengganu. Setiu River Basin with catchment area 188 km² approximately and 52km in length and 148,535,660.9m² immensity [25] and Setiu Wetland covers 23,000 ha area and the water column is well mixed and shallow [26]. Merang River Basin with 39,115,835.46m² immensity and the shortest river basin in Setiu district (8km) with catchment area km². Semerak stations is originated from Semerak River Basin which is semi-enclosed lagoon with a total area 1.7 km² with an average depth of 3.12m located in Pasir Puteh, Kelantan. Semerak lagoon contain 5,304,000m² water during high tide [27].

Table 1. Coordinate of sampling station

Station	Latitude	Longitude	Description
SM1	5°86'50.50"N	102°49'40.00"E	Semerak Lagoon
SM2	5°86'60.32"N	102°50'62.95"E	Semerak Lagoon
SM3	5°86'25.74"N	102°51'35.70"E	Semerak Lagoon
S1	5°67'83.80"N	102°71'07.60"E	Setiu Wetland, Kuala Setiu
S2	5°67'80.40"N	102°71'03.70"E	Setiu Wetland, Kuala Setiu
M1	5°53'43.40"N	102°94'62.70"E	Merang Estuary
M2	5°53'34.30"N	102°94'87.00"E	Merang Estuary

2.2. Sample Collection and Preparations

Water sampling were conducted during high and low tides and collected from subsurface water into 1L high density polyethylene (HDPE) bottles prewashed with dilute hydrochloric acid (10%) and rinsed three times with water sample. The samples were stored at 4°C and were transferred to the laboratory for analysis. Acidified water samples (ph < 2 using ultra-pure nitric acid, HNO₃) were carried out using filtered (0.45 µm cellulose membrane filter, Whatman Milipores, Clifton, NJ). In order to safeguard dependable representatives of all analytical results, all reagents used in current study were analytical grade [2]. Additionally, laboratory glassware and apparatuses used in this study were pre-cleaned with 5% HNO₃. 1ml of water sample was transferred into a 10 ml PP volumetric flask and made up to 10ml with 1% Suprapure grade HNO₃ solution, prepared from Milli Q water. Then, diluted samples transferred to the test tube before analyzed using inductively couple plasma mass spectrometry (ICP-MS, ELAN DRC-e, Perkin Elmer, Shelton, CT). Blanks and a series of

standard solutions were prepared. A series of standard solutions were prepared from stock standard solutions (ICP Multi-element standard solution IV, Merck) that were diluted with deionized water.

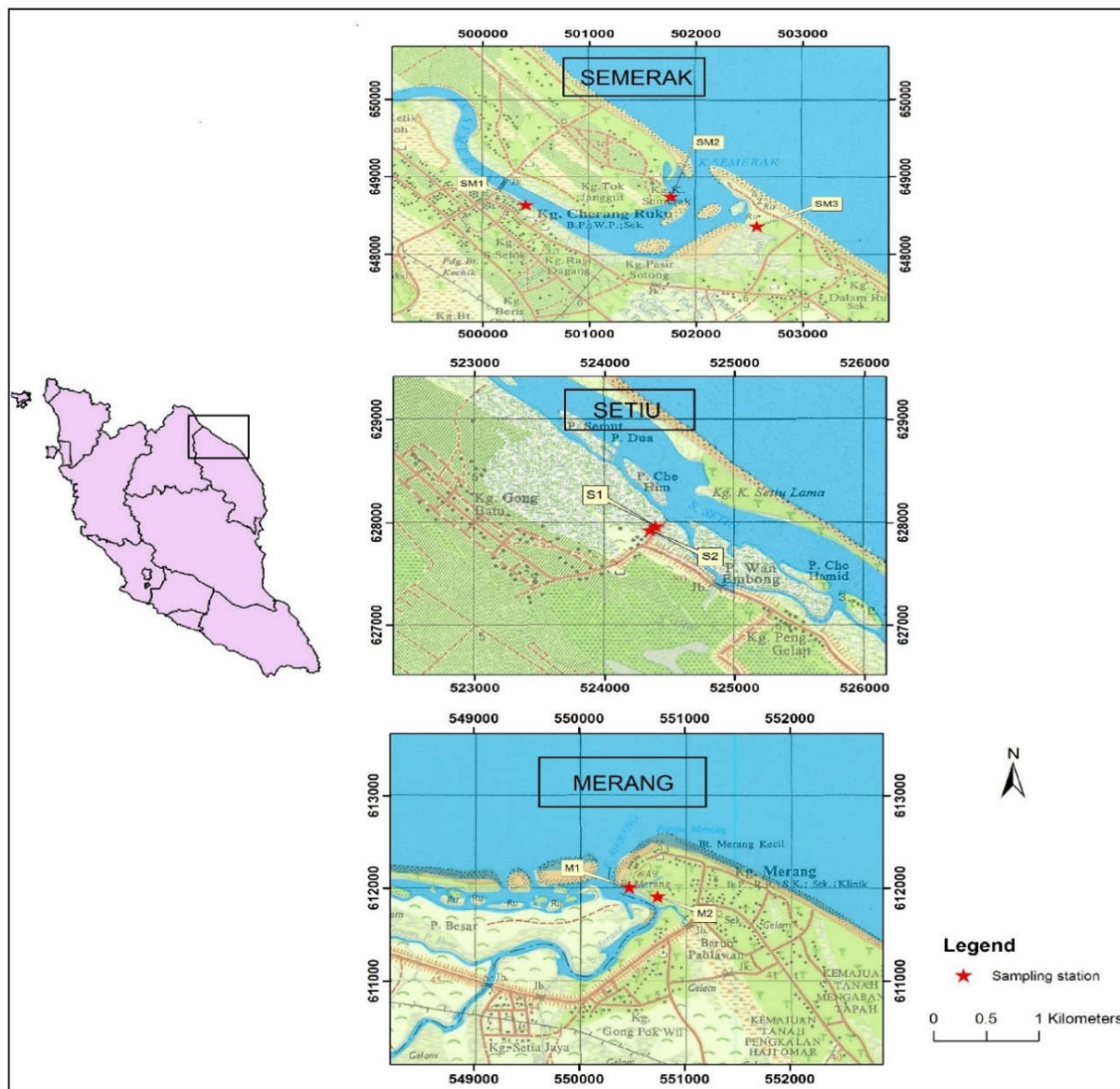


Fig.1. Sampling points at Semerak Lagoon, Setiu Wetland and Merang Estuary

2.3. Statistical Analysis

The Two-way ANOVA without replication was used to reveal the significant value of each water quality parameters in 95% confident level ($\alpha = 0.05$). XLSTAT 2014 software was used to perform statistical analysis. Moreover, the box plot is a visualizing data which represent the descriptive statistics of the data set. The famous ‘stem and leaf diagram’ in box plot are representing data semigraphically [28].

2.4. Principal Component Analysis

The PCs generated by PCA sometimes are not readily interpreted and should be rotated using any of a number of applicable methods such as varimax rotation [10-13]. According to [10], the objective of varimax rotation is to lessen the complexity of the components by making the large loadings larger and the small loadings smaller within each component. The varimax rotation method was applied because this method abridges the factor structure hence makes its explanation easier and more reliable [10, 13]. Based on [29], in the varimax rotation method, only the PCs with eigenvalues greater than 1 (> 1) are used and considered significant in order to obtain the new variables, known as varifactors (VFs) or factor loadings. This approach is known as the Kaiser Criterion. The Kaiser Criterion is used to solve the problem of the number of components to be retained [30-31]. The numbers of VFs gained by varimax rotations are equivalent to the number of variables in accordance with mutual features and can include unobservable, hypothetical, and latent variables [10-11, 31]. The VFs are values that are used to calculate the correlation between variables. VF values which are greater than 0.75 (> 0.75) are considered “strong,” the values ranging from 0.50 to 0.75 ($0.50 \geq \text{factor loading} \geq 0.75$) are evaluated “moderate,” while the values ranging from 0.30 to 0.49 ($0.30 \geq \text{factor loading} \geq 0.49$) are considered “weak” factor loadings [12-13, 32]. In this study, the VFs with complete values larger than 0.75 were set as the selection threshold. Therefore, the results of factor scores after varimax rotation were used for artificial intelligence modelling. The PCA was examined using XLSTAT 2014 software.

2.5. Artificial Neural Network Model

The artificial neural network (ANN) designed to imitate the biological brain system which is able to provide better prediction [18, 33-34]. In this study, ANN was applied and computed using JMP 10 software. ANN have the ability to investigate using six variable parameters (heavy metals) to build supervised classifiers to discriminate between the three different spatial regions (Merang, Setiu and Semerak). Based on Fig. 2, the network architecture of ANN consists of three layers (input, hidden and output). The input layer represents the input variables of heavy metal parameters (Ni, Cu, Pb, Cd, As and Zn) while the outputs characterize the three selected monitoring stations (Merang, Setiu and Semerak). The hidden

layer signifies the interaction among inputs nodes (use as a non-linear transformation function) which is a sigmoid function [19, 35]. According to [19], the nodes' number in the hidden layer was diverse using trial and error procedure until the finest number was attained in order to estimate any non-linear function with any level of accuracy. Thus, this model used to search for the best model for spatial distribution prediction. The correlation of determination (R^2), the root mean square error (RMSE) and the misclassification rate (MR) will determined the networks' performance. By doing this, the best prediction accuracy is based on the lower RMSE and MR value and the higher of R^2 value (near to 1) [19, 36].

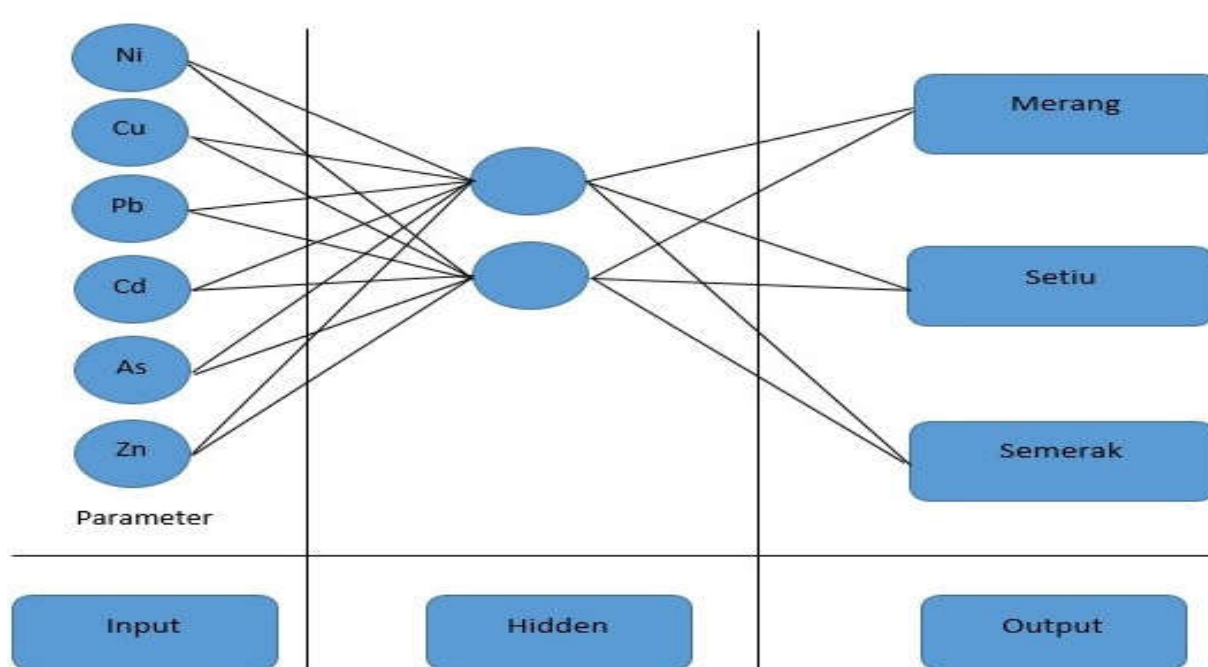


Fig.2. The network structure of the neural network model used in spatial pattern recognition.

3. RESULTS AND DISCUSSION

Analysis of variance (ANOVA) was used to test the significant different of all heavy metals among sampling points. Based on Table 2, since the p-value for rows = $0.5354 > 0.05 = \alpha$ and $F = 0.8571 < 2.3638 F_{crit}$, we cannot reject the null hypothesis. At the 95% level of confidence, we conclude that there is no significant difference in heavy metals distribution by the sampling stations.

Since the p-value for columns = $1.45E-07 < 0.05 = \alpha$ and $F = 12.4940 > 2.3638 F_{crit}$, we reject the null hypothesis, So, at the 95% level of confidence, we conclude there is a

significant difference in heavy metals distribution between the sampling stations.

Table 2. Two way ANOVA

Source of Variation	SS	df	MS	F	P-Value	F crit
Rows	0.05809	6	0.009682	0.857008	0.535404	2.363751
Columns	0.84687	6	0.141145	12.49402	1.45E-07	2.363751
Error	0.406692	36	0.011297			
Total	1.311652	48				

H_0 : there is no significant difference between means of heavy metals at the sampling stations.

H_a : at least one has significant difference in heavy metals distribution between the sampling stations

Descriptive statistic was conducted to describe the main characteristic of mangrove estuary water in the study areas as shown in Table 3. The mean concentrations of heavy metals are in following orders: 1) Setiu Wetland (Zn > Cu > As > Ni > Pb > Cd), 2) Merang Estuary (Zn > Cu > As > Ni > Cd > Pb), 3) Semerak Lagoon (Zn > Pb > As > Ni > Cd). The mean concentrations of heavy metals were compared with Class E (for mangrove estuaries and rivermouth water) Marine Water Quality Index (MWQI) as required to Department of Environment [37]. This study found Zn and Cu concentrations have exceeded the recommended guideline for Setiu Wetland and Merang Estuary. Meanwhile, Semerak Lagoon found only Zn has exceeded the recommended guideline. Summary statistic of heavy metals concentration in Setiu Wetland (S1 and S2), Merang Estuary (M1 and M2) and Semerak Lagoon (SM1, SM2 and SM3) are visualised in box and whisker plots in Fig. 2.

Table 3. Descriptive statistic for heavy metals in study areas

Parameter	Unit	Setiu Wetland				Merang Estuary				Semerak Lagoon				MWQI Class E (2014)
		Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD	
Ni	mg/l	0.0002	0.0107	0.0036	0.0048	0.0002	0.0086	0.0021	0.0037	0.0002	0.0024	0.0019	0.001	NA
Cu	mg/l	0.0002	0.104	0.0351	0.0476	0.001	0.0744	0.0196	0.0311	0.0002	0.0009	0.0007	0.0003	0.0029
Pb	mg/l	0	0.003	0.0005	0.001	0.0002	0.0004	0.0002	0.0001	0.0002	0.003	0.0023	0.0013	0.0085
Cd	mg/l	0	0.0006	0.0002	0.0002	0.0002	0.0012	0.0003	0.0004	0	0.0003	0.0002	0.0001	0.002
As	mg/l	0.0004	0.0687	0.0224	0.0265	0.0079	0.0544	0.0202	0.0164	0.0014	0.0067	0.0022	0.0016	0.02
Zn	mg/l	0	0.205	0.0814	0.095	0.0002	0.191	0.0843	0.0839	0.076	0.174	0.0954	0.0367	0.05

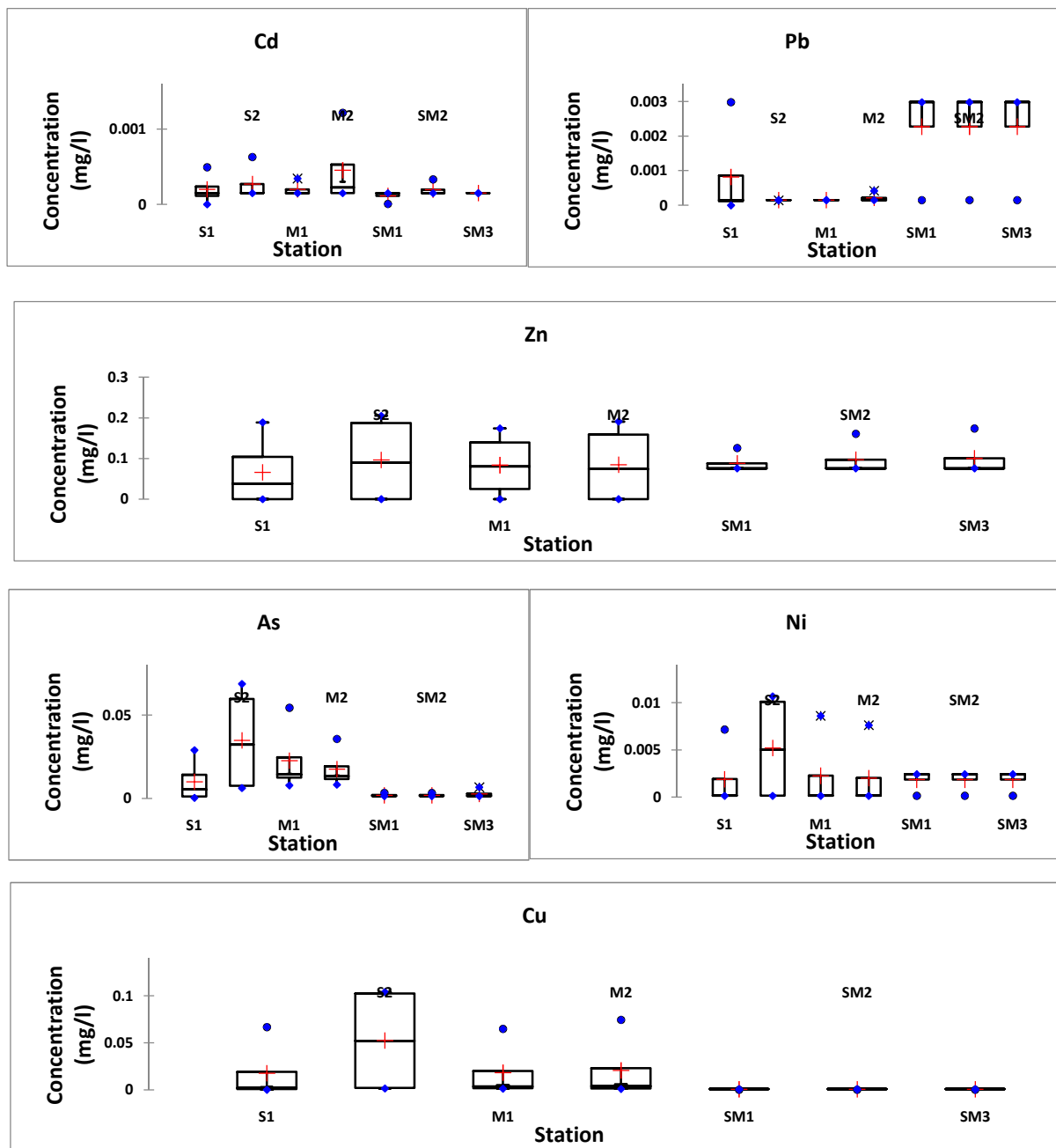


Fig.3. Box plot of heavy metals in mangrove estuary measured

3.1. PCA as Identifying the Source of Variation

The Kaiser-Meyer-Olkin (KMO) result was 0.634 which mean the sampling adequacy was greater than 0.5. This considered that all variables are sufficient and confirming PCA is a useful tool for source apportionment for further analysis [2, 10, 18, 31]. The estimation of the factor loadings was carried out for assessing the correlations between heavy metals variables and the extracted factors. Concordant to eigenvalues, Table 3 shows factor loading after varimax rotation. PCA of the entire data set in Table 4 involved two principal components

with eigenvalues greater than one explaining about 79.374% of the total variance in the heavy metals data set. VF1 shows 56.244% of total variance for Ni, Cu, As and strong negative loading of Zn. This varimax factor represents of industrial waste [38-39]. Based on the result, the value of these variables are very low and below guidelines (except Zn). This VF is consider as surface runoff and the natural geologic sources [2, 40]. Based on [40], topsoil and rocks are drained by rainwater during north-east monsoon season and settled on sediment. Yet, Zn gives strong negatives loading because of this element are higher at Semerak Lagoon which come from river discharge from upstream environment [41-42]. Biplot chart in Fig. 3 represent the heavy metals simultaneously in the new space which shows VF1 linked with S2, M1, M2, SM1, SM2 and SM3 station.

Table 4. Factor loading after varimax rotation

	VF1	VF2
Ni	0.9671	-0.1344
Cu	0.9749	0.1534
Pb	-0.2515	-0.8760
Cd	-0.2753	0.6051
As	0.9213	0.3169
Zn	-0.7026	0.3460
Variability (%)	56.2442	23.1297
Cumulative (%)	56.2442	79.3739

VF2 presents 23.13% total variance of strong positive loadings of Cd and strong negative loading of Pb. This VF is considered as “anthropogenic-toxic” pollution [39]. Shipping waste from fisherman’s boat lead to the significant of Cd in VF2 [2, 11]. Based on [43], anthropogenic inputs through runoff into aquatic environment is concerned for the enrichment of Pb. Though, these two elements have similar mark which the mean value of Pb and Cd are extremely low and followed the MWQI [37]. VF2 linked with S1, SM1, SM2 and SM3 station which represent Pb simultaneously in the new space (Fig. 2). As of Cd is a strong positive loading in VF2, Cd is far from the other elements and significant in M2 station. Thus, Fig. 4 shows that the observations on the factor axes confirm that the heavy metals in

monitoring stations are very well differentiated on the factor axes extracted from the original explanatory variables.

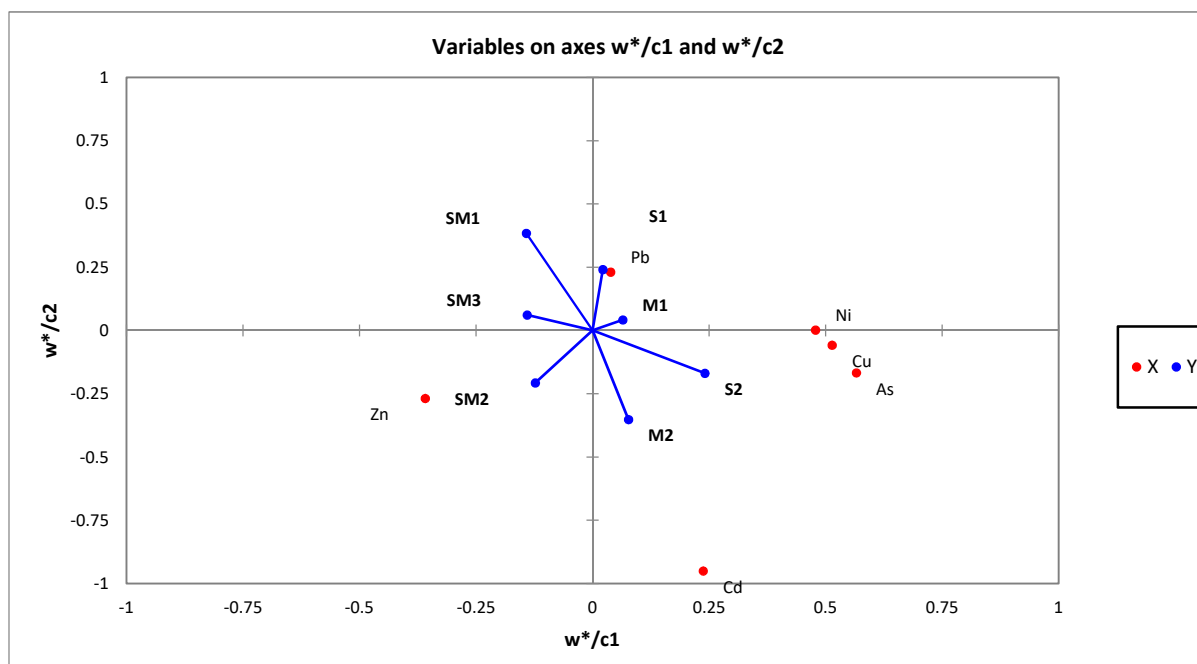


Fig.4. Biplot chart after varimax rotation

*Biplot chart in Fig. 3 shows VF2 linked with S1, S2, SM1, SM2 and SM3 station

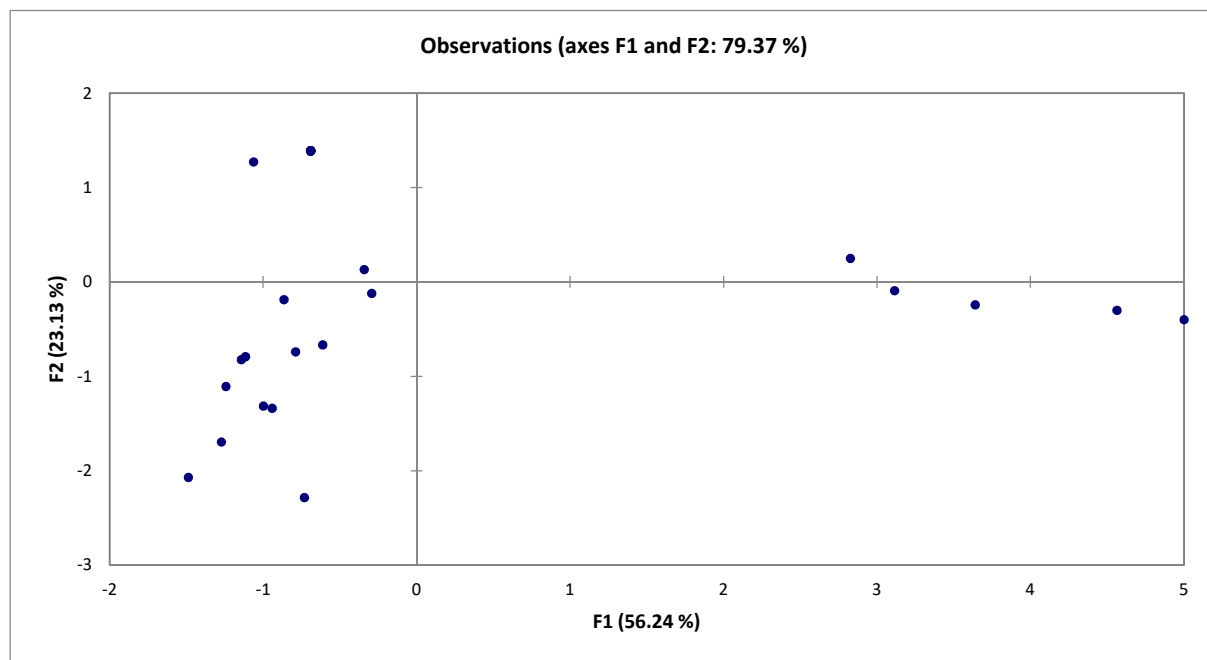


Fig.5. Observation on the factor axes chart

3.2. Predicting the Spatial Distribution of Heavy Metal Parameters Using ANN

A total of two network structures were observed for ANN in order to forecast the spatial distribution of heavy metal samples in mangrove estuary according to regional origin. The

results' summary was obtain in Table 4. ANN model gave acceptable results in discriminating between the heavy metals according the monitoring stations.

Table 5. The prediction performance of spatial pattern recognition (Setiu, Merang and Semerak) using ANN

Model	Network	R ²	RMSE	Misclassification Rate
ANN	[6,0,1,3]	0.6873	0.4568	0.2222
	[6,1,1,3]	0.8	0.399	0.2222
	[6,2,1,3]	0.9614	0.1909	0.1111

*Note: square brackets [w,x,y,z] indicate the ANN structure, w is the number of input nodes, x is the number of hidden nodes, y is the number of hidden layers and z is the number of output nodes

Based on Table 5, two hidden nodes are well-thought-out to be the optimum. After considering two hidden nodes, the model gave the lowest RMSE (0.1909) and MR (0.1111) compared to the other. Table 6 shows the ANN model magnificently discriminated between each monitoring station with an average 85% correct classification. Receiver operating characteristics (ROC) demonstrate the performance of binary classifier system where the discrimination threshold is dissimilar [19]. Thus, Fig. 6 displays the ROC results of the spatial distribution in ANN model were normally distributed by value area respectively; Merang (0.9923), Setiu (0.9231) and Semerak (0.9688). According to the result in Fig. 6, Merang offers more precise information likened Setiu and Semerak which it can be used to achieve higher true positive fraction at the false positive fraction.

Table 6. Classification matrix for ANN of spatial variation in the three regions.

Location	% Correct	Region Assigned by ANN		
		Merang	Semerak	Setiu
Merang	100	5	0	0
Semerak	75	0	6	2
Setiu	80	1	0	4
Total				
Average	85			

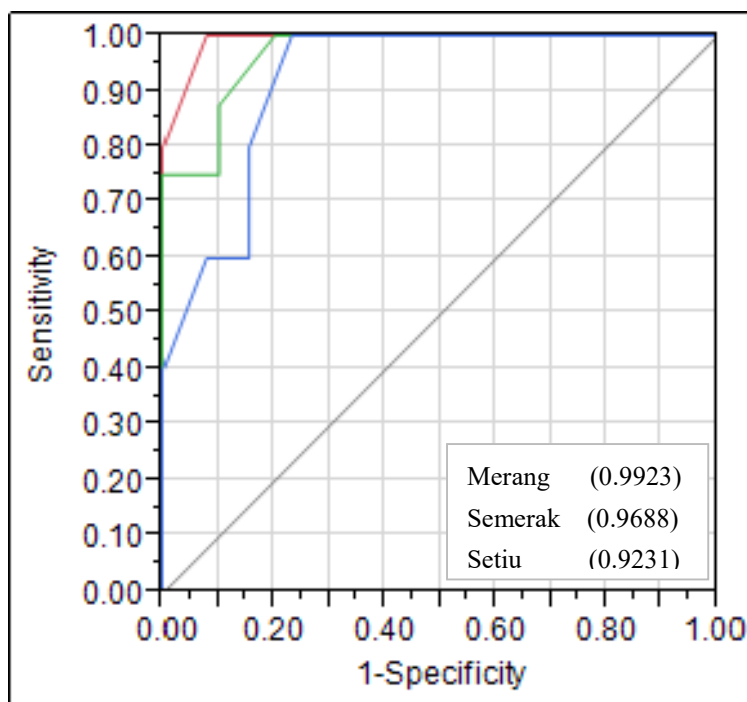


Fig.6. Receiver Operating Characteristics (ROC) for spatial pattern recognition (ANN)

4. CONCLUSION

This is the preliminary study of heavy metal concentration in mangrove estuary at selected area in the east coast of peninsular Malaysia. This study consist of three mangrove estuary areas (Semerak, Setiu and Merang) which were chosen based on lack of studies in term of heavy metal concentrations in mangrove ecosystems which act as a baseline study for the future research. From the finding of PCA, the pollution apportionment from these three areas which affect marine water quality status include industrial waste and ‘anthropogenic-toxic’ pollution. The following conclusions can be drawn in the decreasing trend: 1) Setiu Wetland (Zn > Cu > As > Ni > Pb > Cd), 2) Merang Estuary (Zn > Cu > As > Ni > Cd > Pb, 3) Semerak Lagoon (Zn > Pb > As > Ni > Cd).

The ANN model showed a more reliable prediction performance in discriminating between the monitoring stations with an excellent percentage of correct classification (85%). Thus, the ANN model is proven as a very helpful technique in helping decision makers achieve better heavy metal pollution in mangrove estuary management in order to characterize the spatial

pattern. Hence, long term monitoring for these areas need to be undertaken to ensure the water quality for mangrove estuary will not degrade and that the health of mangrove ecosystem is sustainably conserve. In addition, this paper hopefully could contribute towards more effective heavy metal in mangrove estuary management.

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