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Chemometric Application on Surface River Water Quality: A Case Study of Linggi River, Malaysia

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A B S T R A C T

Chemometric methods were applied to Linggi River water quality data sets to evaluate spatial - temporal variations and identify sources of pollutants. In 2011, the data sets consist of 11 variables, monthly collected from 26 sampling stations. Three clusters (C1; 6, C2; 10 C3; 10 stations) were generated using a cluster analysis method; four latent factors were identified by principle component analysis with factor analysis method. The scree plot was used to identify the number of principle components and show pronounced change after 4 eigenvalues. Four principle components explained about 80.8 % of the total variance in the water data sets from eigenvalue > 1. Using a discriminant analysis method, the discriminant function weighed with 5 (pH, biochemical oxygen demand, suspended solids, iron and ammonia nitrogen) and 7 (pH, conductivity, biochemical oxygen demand, suspended solids, iron, manganese and ammonia nitrogen) variables capable to distinguish between 84.6 % spatial and 83.3 % temporal variation, respectively. Linggi River water quality status were explained in that pollution mainly originated from agriculture run off or aquaculture residue (nutrients), soil weathering or industrial discharge (metals), and solid waste disposal site or municipal discharge (biochemicals). This study successfully demonstrated that chemometric methods demonstrate useful information for further monitoring strategies.

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INTRODUCTION

Chemometric methods also known as multivariate statistical analysis are useful techniques for obtaining desiredd information about water quality, identifying primary pollution sources and designing monitoring networks for effective water resources management [1, 2]. Its offer superior interpretation of large data sets in verifying temporal and spatial variations, which is influenced by multiple factors [1, 3, 4]. Common methods in chemometric methods are cluster analysis (CA), principle component analysis with factor analysis (PCA/FA), and discriminant analysis (DA).

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The purpose of CA is to assist in partitioning similarities patterns between stations while PCA for pattern recognition of inter correlated variables that explain the variance of a large set. Principle component analysis is being used to identify the ecological aspects of pollutants [5, 6]. An advantage of chemometric application in river studies is interpretation of multi-constituent measurement being easy handled with minimum loss of contribution [7]. Previous studies have been demonstrated that chemometric methods were widely used in Malaysian rivers such as Langat River [5, 8, 9], Juru River [9], Kinta River [10], and Perlis River [11], but not yet in Linggi River.

Linggi River is one of the biggest river basins in Negeri Sembilan, Malaysia. It situated at 2°24'–2 °50' N latitude and 101°53'–102 °12' E longitude heading towards south-western part of the state of Negeri Sembilan. The main stream flows for 83.5 km, with

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more than 21 major tributaries. The water supply resources are demanded by approximately 60% of Seremban population. Major anthropogenic activities nearby this river include palm oil and rubber plantations, aquaculture, paddy cultivations, manufacturing industries and human settlement areas [12, 13].

The aim of this study was to apply the chemometric methods on water quality data set of Linggi River in order to identify sources of pollution and explicit latent factors responsible for temporal and spatial variations.

MATERIALS AND METHODS

Sampling and Laboratory Analysis

The data sets of 26 selected sampling stations, which consist of 11 water quality variables monitored monthly from January to December 2011. The map location and descriptive information of sampling stations are presented in Figure 1 and Table 1, respectively. All surface water samples were collected at a 0.5 m depth at each sampling station taken using 1 L pre-cleaned polyethylene and glass bottles. Samples were then placed in ice filled cool box prior transfer to ALIR, UKM laboratory. The monitored water quality variables namely pH, temperature, salinity, electrical conductivity, dissolved oxygen, biochemical oxygen demand, chemical oxygen demand, suspended solids, ammonia nitrogen, iron and manganese.

Temperature, pH, electrical conductivity, dissolved oxygen and salinity were measured *in situ* using a multiprobe sensor instrument (YSI 550, USA). Other parameters were analyzed in the laboratory using standard methods as following biochemical oxygen demand (5 days incubation), chemical oxygen demand (reactor digestion and colorimetric determination), suspended solids (gravimetric method), ammonia nitrogen (salicylate method), and iron and manganese (flame atomic absorption spectrometry). Metal ions were determined as total dissolved metal concentration after filtering using 0.45 µm. The quality control of water sample was ensured by careful standardization, procedural blank measurements and triplicate samples analyses. Water Quality Index (WQI) was determined by Malaysia Department of Environment formula based on six parameters as given by the following Equation 1 [14];

$$WQI = 0.22{*}S_IDO + 0.19{*}S_IBOD + 0.16{*}S_ICOD + 0.15{*}S_IAN + 0.16{*}S_ISS + 0.12{*}S_IpH \quad (1)$$

where S_I represents sub-index of each parameter, WQI was then used to classify river segment based on National Water Quality Standard Malaysia (NWQS) which categorized water quality into five classes namely class I (WQI > 92.7), class II (WQI 76.6 – 92.7), class III (WQI 51.9 – 76.5), class IV (WQI 31.0 – 51.9) and class V (WQI < 31.0) based on beneficial use of the water [15].

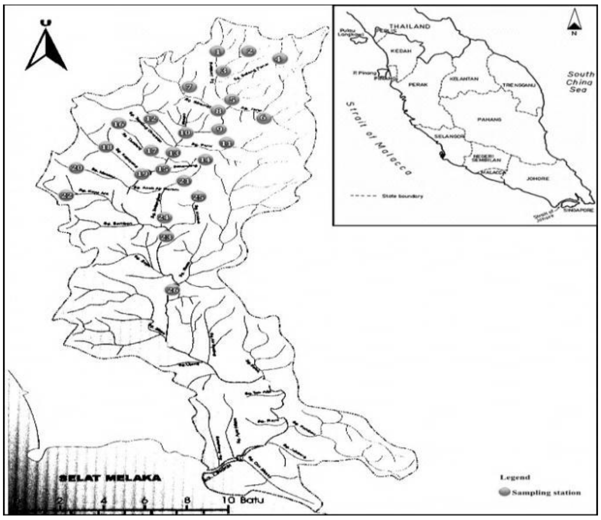


Figure 1. The map location of sampling stations (inside small map of Peninsular Malaysia)

TABLE 1. The descriptive information of sampling stations on the Linggi River

Code Station	Station	Type	WQI	Class
S1	Jerlang River	Tributary	80	II
S2	NgoiNgoi River	Tributary	77	II
S3	NgoiNgoi River Point 2	Tributary	80	II
S4	BatangPenar River	Tributary	86	II
S5	BatangPenar Point 2	Tributary	76	II
S6	Terip River	Tributary	74	III
S7	Sikamat River	Tributary	68	III
S8	Sikamat River Point 2	Tributary	69	III
S9	Shimpa River	Tributary	67	III
S10	Shimpa River Point 2	Tributary	67	III
S11	Paroi River	Tributary	74	III
S12	Temiang Division River	Tributary	75	III
S13	Linggi River Point 1	Main river	66	III
S14	Senawang River	Tributary	63	III
S15	Linggi River Point 2	Main river	62	III
S16	Temiang River	Tributary	61	III
S17	Linggi River Point 3	Main river	60	III
S18	Kepayang River	Tributary	74	III
S19	Linggi River Point 4	Main River	67	III
S20	Mantau River	Tributary	74	III
S21	Anak Air Garam River	Tributary	75	III
S22	KayuAra River	Tributary	66	III
S23	Linggi River Point 5	Main river	62	III
S24	Belangkan River	Tributary	82	II
S25	Nyatoh River	Tributary	69	III
S26	Linggi River Point 6	Main river	72	III

WQI – Water Quality Index

Data treatment: cluster analysis

Cluster system analysis is a repeating process on group similarity until all cluster become one cluster. The final outcome should then exhibit either homogeneity (within the cluster) or heterogeneity (between the clusters) criterion as illustrated in a dendogram [16]. The most common approach in cluster analysis is Hierarchical Agglomerative Cluster Analysis (HACA). It was performed on the normalized data set by Ward's method, using squared Euclidean distance as a measure of similarity. The linkage distance is remarked as D_{link}/D_{max} x 100. The value of D_{link} is the quotient between the linkage distances for a particular case divided by D_{max} as the maximal distance. Then the value was multiply by 100 as a way to standardize the linkage distance [1, 16]. In evaluation of water quality, cluster analysis is usually being group based on geographic location (spatial) and period of study (temporal) before further analysis [1, 3].

Principle component analysis with factor analysis

Principle component analysis is a statistical tool being used for reducing the dimensions of multivariate problems. The extracted of significant principle components will show in scree plot. The first principal component should weighed as much of the variation as possible in the data set. The second principal component is orthogonal to the first and covers as much of the remaining variation as possible till component value can be neglected [17]. To differentiate within principle component value, Kaiser Criterion was also used. Eigenvalue > 1 would consider as significant principle component [18]. The principal component (PC) can be expressed as Equation 2 [16];

$$z_{ij} = a_{i1}x_{1j} + a_{i2}x_{2j} + \ldots + a_{im}x_{mj} \quad (2)$$

where z is the component score, i is the component number, j is the sample number, a is the component loading, x the measured value of variable, and m is the total number of variables.

Factor analysis is used in chemometric methods to provide the most meaningful variables, with minimum loss of original information from PCA. The outcome is attempted to transform inter correlated variables into smaller set of new independent variables, also called as varifactors [1,17]. Varifactors usually are grouped in the studied variables based on communalities features or unobservable, hypothetical, and latent variables. The factor analysis can be expressed per Equation 3 [16]:

$$z_{ij} = a_{f1}f_{1i} + a_{f2}f_{2i} + \ldots + a_{fm}f_{mi} + e_{fi} \quad (3)$$

where z is the measured value of a variable, a the factor loading, f the factor score, i the sample number, j the variables number, m the total number of factors, and e the residual term accounting for errors or other sources of variation.

Discriminant analysis

Discriminant analysis is a step of statistical tool to classify total samples and only performed after knowledge of membership of objects was known to particular groups or cluster. Like cluster analysis, this method will help in grouping samples accordance to sharing under common properties. High yield of samples trueness in classification matrix tables will reflect the effectiveness of data sets [1, 2]. The discriminant function may be expressed as Equation 4:

$$f(G_i) = k_i + \sum_{j=1}^{n} wijpij \quad (4)$$

where i is the number of groups (G), k_i is the constant inherent to each group, n is the number of parameters used to classify a set of data into a given group, wj the weight coefficient, assigned by DA to a given selected parameter (pj) [16].

In this study, water quality data sets were being tested subject to chemometric methods namely cluster analysis (CA), principle component analysis with factor analysis (PCA/FA) and discriminant analysis (DA). Prior to analysis, all data except for discriminant analysis were standardized to experimental data through z-scale transformation. All statistical computations were made by using software Minitab version 17.

RESULTS AND DISCUSSION

Kolmogorov-Smirnov statistics were applied to test the goodness-of-fit of the complex data to log normal distribution. The results test shown all variables are fit well, with 95 % confidence levels on log normal distributed. All the water quality variables results are expressed in mg/l except pH, EC (µS/cm) and temperature (° C). The descriptive statistics of for the 2011 data set on river water quality are summarized in Table 2.

Site similarity

The dendogram of cluster analysis as rendered by the Ward's method is depicted in Figure 2. In this study, all sampling stations were grouped into three clusters namely cluster 1, 2, and 3 at (D_{link}/D_{max}) x 100 < 80. Cluster 1 represent for 6 stations (S1 – S5, S24), cluster 2 (S6 – S8, S10, S13, S15, S17, S19, S20, S23) and cluster 3 (S9, S11, S12, S14, S16, S18, S21, S22, S25, S26) were 10 stations for each corresponds to low level, moderate and high level pollution as matched to water quality indices, respectively. The similarity of cluster 1 corresponds to the upper catchment area, with no anthropogenic activities and far from municipal pollution. Meanwhile, surface waters in cluster 2 are degraded due to aquaculture activities, municipal pollution like dump garbage sites, and untreated wastes discharge from restaurants. Cluster 3 suffered from similarities correspond to major polluting sources from agriculture, wood industry, and settlements.

TABLE 2. The basic descriptive statistic of Linggi River data sets

Variable	Abbreviation	Minimum	Maximum	Mean	[a]SD (±)
pH	pH	6.60	6.97	6.80	0.09
Temperature	Temp	25.11	31.41	27.91	1.36
Salinity	Sal	0.01	0.09	0.04	0.02
Electrical Conductivity	EC	39.33	202.67	100.14	43.73
Dissolved Oxygen	DO	2.83	15.83	6.51	2.26
Biochemical Oxygen Demand	BOD	0.81	3.94	2.76	0.85
Chemical Oxygen Demand	COD	21.44	47.37	32.80	6.93
Total Suspended Solids	TSS	22.30	920.80	301.10	57.01
Ammonia Nitrogen	NH4-N	0.06	3.025	1.01	0.81
Iron	Fe	0.31	1.99	1.49	0.35
Manganese	Mn	0.05	0.55	0.34	0.13

[a]SD – Standard Deviation

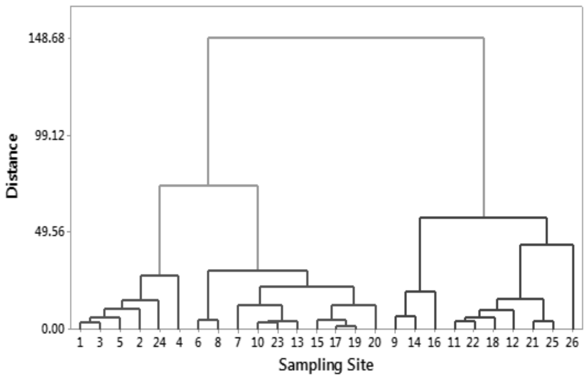


Figure 2. Dendogram of clustering Linggi River sampling stations

Data structure and source identification

The scree plot was used to identify the number of principle components and show pronounced change after 4 eigenvalues (4.76, 1.74, 1.42 and 1.05).Four principle components explained about 80.8 % of the total variance in the water data sets from eigenvalue > 1 (Table 3). In this study, communalities of the variance were high values (0.57 – 0.94) indicate that the extracted factors fit well with the factor solution. The first component with 30.90 % of the total variance had a strong positive loading on ammonia nitrogen, salinity and electrical conductivity. The major source of ammonia nitrogen is likely excreted from agriculture sites in several locations along Linggi river. The second component accounted for 19.80 % of the total variance, with negative loading on pH and temperature which represent physico parameter indicator.

TABLE 3. The varifactors of varimax rotated loading results

Variable	Varifactors (VF)			
	VF1	VF2	VF3	VF4
pH	0.422	**-0.544**	-0.025	0.039
Temperature	0.218	**-0.855**	-0.032	-0.083
Salinity	**0.904**	-0.252	0.075	-0.203
Electrical Conductivity	**0.899**	-0.270	0.084	-0.203
Dissolved Oxygen	-0.127	0.070	0.067	-0.226
Biochemical Oxygen Demand	0.119	-0.462	0.048	**0.839**
Chemical Oxygen Demand	0.415	-0.111	0.085	-0.129
Total Suspended Solids	-0.463	0.272	**0.691**	-0.399
Ammonia Nitrogen	**0.894**	-0.105	0.068	-0.360
Iron	0.463	-0.411	**0.667**	0.078
Manganese	0.139	0.003	**0.907**	0.079
Variance (%)	30.90	19.80	16.20	13.90
Cumulative Variance (%)	30.90	50.70	66.90	80.80

The third component was associated at 16.20 %, weighted on iron, manganese and suspended solids which represent the metal source of variability. The factor loaded with solids explained the run-off phenomenon that brought in high waste disposal [11]. The fourth component had strong positive loading on biochemical oxygen demand presented 13.90 % of the total variance. The factor loadings were marked as strong (VF > 0.75), moderate (0.75 < VF > 0.5) and weak (< 0.50) corresponding to absolute varifactors values, respectively [19].

According to PCA/FA loading results, for Linggi River water quality status, it was presumed that pollution mainly is derived from agriculture run off or aquaculture residue (nutrient), soil weathering or industrial discharge (metals) and solid waste disposal site or municipal discharge (biochemical).

Temporal and spatial variation

A total of 312 observations from this study were grouped into three quarters (1st Quarter January to April, 2nd Quarter May to August, 3rd Quarter September to

December) for temporal variations. The discriminant function indicates a classification matrix of temporal variation with 83.3 % correct assigned using only 7 variables. Thus, the results suggest that variables, namely pH, conductivity, biochemical oxygen demand, suspended solids, iron, manganese and ammonia nitrogen were the most influential variables that can be distinguished between three different quarters or accounting for temporal variations during period of study. Discriminant functions and classification matrix for temporal variations obtained from standard mode are shown in Tables 4 and 5, respectively.

TABLE 4. Discriminant functions of 7 variables for temporal variations

Variables	Coefficient		
	1st Quarter	2nd Quarter	3rd Quarter
pH	-12.520	8.194	-40.979
Conductivity	17.539	-12.573	86.080
Biochemical Oxygen Demand	0.730	-1.085	3.417
Total Suspended Solids	12.036	-2.864	22.899
Iron	-2.001	1.197	-4.867
Manganese	1.412	0.966	1.309
Ammonia Nitrogen	5.348	-0.937	12.385
Constant	-3.201	-1.334	-8.459

TABLE 5. Classification matrix of discriminant analysis for temporal variations

Temporal	Correct (%)	Period assign by Discriminant Analysis		
		Quarter 1	Quarter 2	Quarter 3
Quarter 1	75	78	0	26
Quarter 2	100	26	104	0
Quarter 3	75	0	0	78
Total	83.3	104	104	104

A higher range of suspended solids and ammonia nitrogen would suggest a high load of dissolved organic matter from agriculture runoff and water discharge in clusters 2 and 3 with enhanced atmospheric deposition. However, meteorological data was not obtained to support the assumption. Both metals show different trends in all clusters. It may be presumed that abundance of iron tends to increase due to anthropogenic sources such as the steel factory spotted at station S24. Box and whisker plots of selected variables showing temporal variations are illustrated in Figure 3.

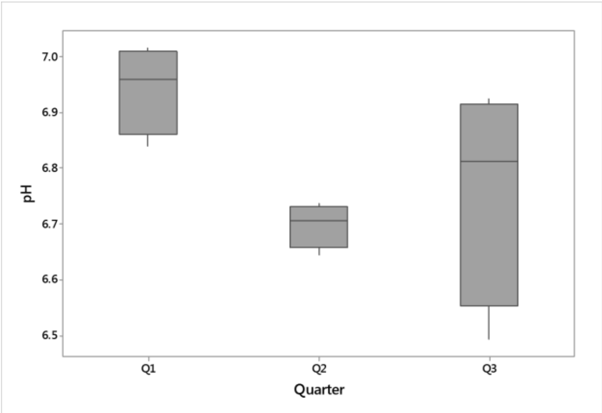
Discriminant functions and classification matrix for spatial variations obtained from standard mode are depicted in Tables 6 and 7, respectively. The discriminant analysis for spatial variation was accounted for after grouping the raw data into three clusters, obtained using cluster analysis. The final result showed that the classification matrix with 84.6 % correct assigned using only 5 discriminant variables. For spatial variations, variables, namely pH, biochemical oxygen demand, total suspended solids, iron, and ammonia nitrogen, are strong enough to distinguish within three clusters. Moreover, cluster 1 showed 100 % of trueness while not all of clusters 2 and 3 made a considerable data reduction for next water monitoring strategies.

TABLE 6. Discriminant functions of 5 variables for spatial variations

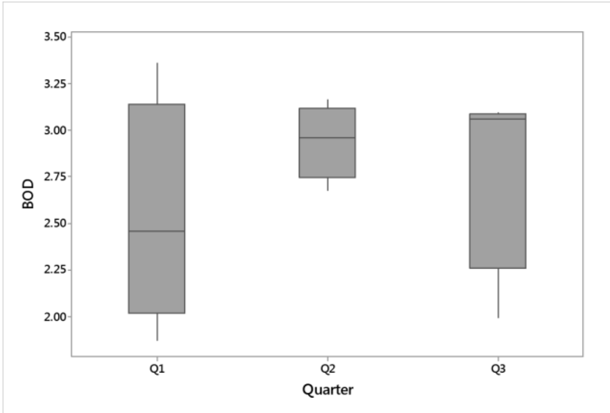
Variables	Coefficient		
	Cluster 1	Cluster 2	Cluster 3
pH	2.129	-1.687	0.219
Biochemical Oxygen Demand	-2.447	0.879	0.615
Total Suspended Solids	-1.636	2.766	-1.370
Iron	-3.252	1.160	0.824
Ammonia Nitrogen	-3.072	0.074	1.614
Constant	-4.657	-2.027	-1.537

TABLE 7. Classification matrix of discriminant analysis for spatial variations

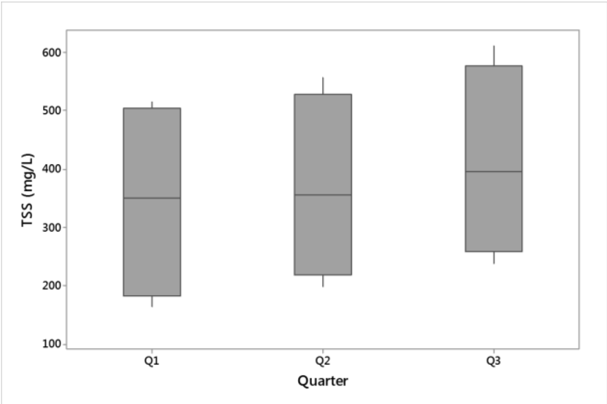
Spatial	Correct (%)	Region assign by Discriminant Analysis		
		Cluster 1	Cluster 2	Cluster 3
Cluster 1	100	72	0	24
Cluster 2	88.9	0	96	12
Cluster 3	72.7	0	12	96
Total	84.6	72	108	132



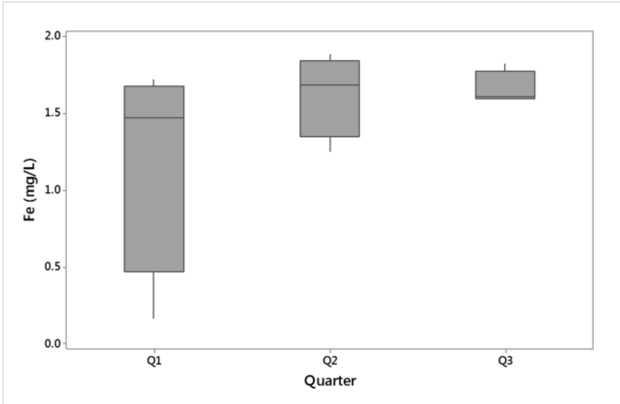
(a) pH



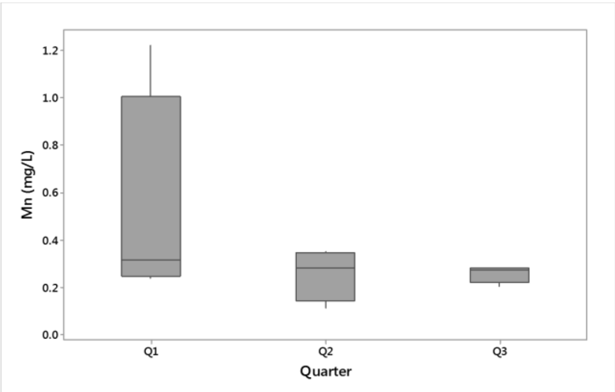
(b) BOD



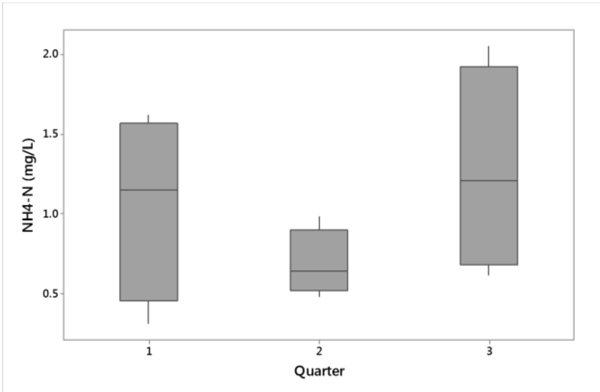
(c) TSS



(d) Fe



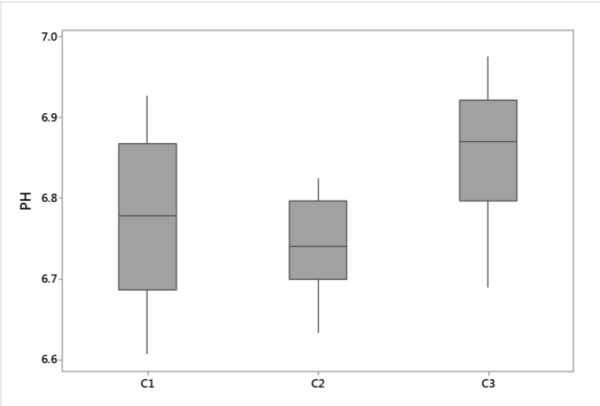
(e) Mn



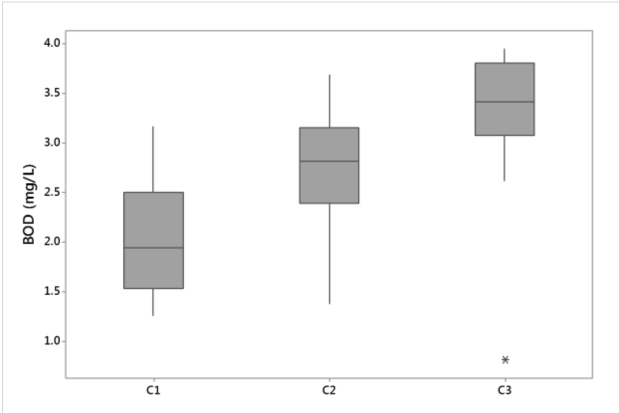
(f) NH_4-N

Figure 3. Box and whisker plots of selected variables showing temporal variations

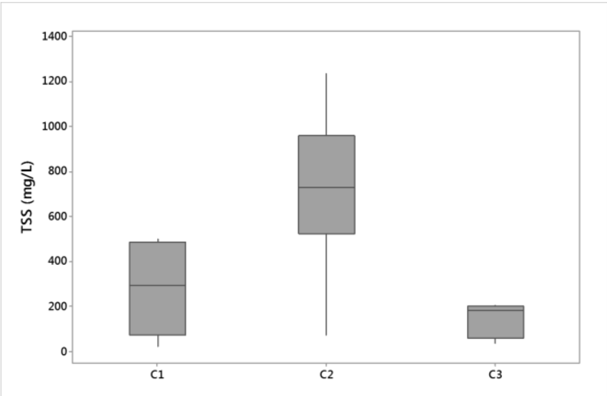
The mean measured from five significant variables (except suspended solids) were highest in cluster 3, as they receive more discharge from agriculture, industry and municipal compared to others. The higher suspended solids mean value suggests a high load of dissolved organic matter from aquaculture, garbage dump sites, or untreated water discharge in cluster 2. This in turn results in the formation of ammonia and organic acids due to anaerobic conditions in the river basin. Hydrolysis of the pollutants would lead to a decrease of pH in this cluster. Box and whisker plots of 5 selected variables showing spatial variations are shown in Figure 4.



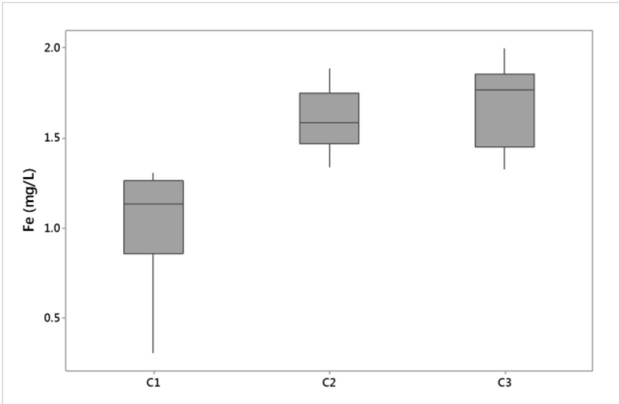
(a) pH



(b) BOD



(c) TSS



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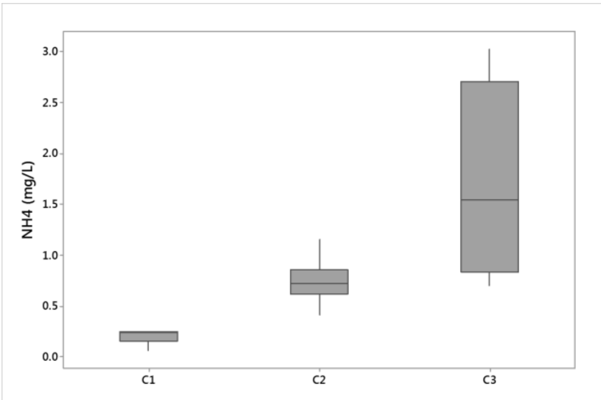

(e) NH_4-N

Figure 4. Box and whisker plots of 5 selected variables showing spatial variations

CONCLUSION

In this study, latent factors in Linggi River water quality in terms of temporal and spatial variations explained the structure of data sets. Cluster analyses successfully examined the existence of three zones (cluster) from 26 sampling stations separately for their differing water chemistry. According to principle component analysis with factor analysis loading results, the four varifactorsextracted indicated that the anthropogenic activities mainly affecting Linggi River include aquaculture, agricultural run-off, industrial discharge, and municipal discharge, especially in clusters 2 and 3. Discriminant analysis confirmed the discriminant function weighed with 5 and 7 variables capable of distinguishing between spatial and temporal variation respectively. The multivariate analysis outcomes may be helpful for further monitoring programs by perhaps limiting the number of sampling stations to those highly representative of the spatial patterns or narrowing the number of variables to the most influence variables in the subject of study.

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Persian Abstract

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چکیده

روش کمومتریک برای داده کیفیت آب رودخانه Linggi استفاده شد که برای ارزیابی تغییرات مکانی، زمانی وشناسایی منابع آلاینده تنظیم شد. در سال ۲۰۱۱، مجموعه داده ها شامل۱۱متغیر، ماهانه از۲۶ ایستگاه نمونه برداری جمع آوری شده است.سه گروه (ایستگاه های C1 ؛ ۶، C2، ۱۰،C3، ۱۰)با استفاده از روش تجزیه و تحلیل گروهی تولید شد. چهار عامل نهفته شده توسط تجزیه و تحلیل مؤلفه های اصلی با استفاده از روش تحلیل عاملی مشخص شدند. نمودار اسکری برای شناسایی تعدادی از اجزای اصلی استفاده شده است و تغییر را پس از ۴ مقادیر ویژه نشان می دهد. چهار جزءاصلی ۸۰/۸٪ از واریانس کل درمجموعه داده آب بامقادیر ویژه >۱ را نشان می دهد. با استفاده ازروش تجزیه و تحلیل مشخص، تابع تفکیک سنجیده شده با ۵ (pH ، اکسیژن مورد نیاز بیولوژیکی، جامدات معلق، آهن و آمونیاک و نیتروژن) و ۷ (pH ، هدایت، اکسیژن مورد نیاز بیولوژیکی، جامدات معلق، آهن، منگنز و نیتروژن آمونیاک) متغیر قادر به تمایز بین ۸۴/۶ ٪ تغییرات مکانی و ۸۳/۳٪ تغییرات زمانی می باشد. وضعیت کیفیت آبرودخانه Linggi به این صورت شرح داده شد که آن آلودگی شد به طور عمده از مواد زائدکشاورزی و یا باقی مانده حاصل از پرورش آبزیان (مواد مغذی)، فرسایش خاک و یاتخلیه صنعتی(فلزات) و دفع زباله جامد یا تخلیه شهری(بیوشیمیایی) سرچشمه گرفته است. این مطالعه با موفقیت نشان داد که روش کمومتریک اطلاعات مفیدی برای راهبردهای نظارت بیشتر نشان می دهد.