Spatial variability in surface water quality of lakes and ex-mining ponds in Malacca, Malaysia: the geochemical influence

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ABSTRACT

A surface water quality assessment was carried out to explore the spatial variations between lakes and ex-mining ponds in Malacca with the aid of chemometrics tools. The physio-chemical data included pH, dissolved oxygen, total dissolved solids, electrical conductivity, biological oxygen demand, and ammoniacal nitrogen, which were measured in situ. Meanwhile, the elemental concentrations were determined by inductively coupled plasma mass spectrometry. Hierarchical cluster analysis of the water quality dataset revealed a clustering pattern that was strongly associated with their underlying geochemical variations. A similar trend was also demonstrated in the principal component analysis which suggested that the variability in the water quality depended mainly on the nature of ponds/lakes, whereby the dominance of Ca, Mg, electrical conductivity, and total dissolved solid could plausibly be linked with the dissolution of rocks; and part of the variation was originated from their current uses. Based on the linear discriminant analysis model, the ex-mining ponds were characterized with As, Ca, Mg, and Mn concentrations whereas Na, Fe, pH, and ammoniacal nitrogen levels were associated with the lakes. Despite the inherent variability characteristics between both water sources, the corresponding metal index revealed no significant As a threat. But, in most cases, Fe exceeded the reference limit, which was attributed to its natural abundance. Therefore, these surface water sources could be considered as potential reservoirs for potable supply after conventional treatment.

Keywords: Chemometrics; Drinking water; Geochemistry; Metal; Pattern recognition

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1. Introduction

The need for adequate and safe drinking water is increasing globally due to urban expansion and an increase in human population [1]. The quality of water is a major factor of consideration to maintaining a healthy ecosystem due to the reported effects of water pollutants to humans, animals, and plants [2,3]. Many countries depend on surface water such as lakes and rivers, canals, and underground sources for daily human consumption and other domestic needs [4–6]. Rainfall is also used to support drinking water supplies and agricultural activities; this expanded the idea and need for rainwater harvesting [7,8]. Many other sources of water can be explored for daily water supply, and to meet up with the public demand.

Malacca experiences seasonal water shortages, and thus this causes limited water availability and potential issues of water quality. Therefore, there is an urgent need to search for other supporting sources. Malacca experienced the worst water crisis in 1991 due to a very low water level from its major reservoirs; the Durian Tunggal Lake (LDT) and Jus Dam (JD). These water sources are not enough to cater for the three districts of Jasin, Alor Gajah, and Malacca Tengah, and thus larger portion of the daily water supply in Malacca is imported from Muar River in Johor [9]. Furthermore, the authorities in Malacca have identified additional potable water sources to complement the public needs [10].

Surface water quality is greatly influenced by a myriad of factors among which is mineral exploration such as mining [11]. In most developing countries, mining is associated with lack of proper environmental laws and monitoring [12,13], thereby resulting in surface and underground water pollution [14-16]. The pollutants of concern are toxic metals which are not easily removed from the environment, and increase in organic matter usually associated with crushing and other mining operational processes [17,18]. The acidic medium generated is due to oxidation of metal ores which increases the metal levels in ex-mining water by liberating more metals into the solution [19-21]. However, a basic medium is observed when the basement or host rocks neutralize the acidity produced due to buffering processes, resulting in low metal levels [22,23]. Therefore, a detailed understanding of the geology and geochemical characteristics is pertinent in evaluating the levels of hazardous metals in the catchments [15].

In this study, chemometric techniques which included hierarchical cluster analysis (HCA), principal component analysis (PCA), and linear discriminant analysis (LDA) were applied to evaluate the water quality dataset collected from selected ex-mining ponds and lakes in Malacca, in addition to the conventional metal index approach. The multivariate models were aimed to unveil the underlying variations in metal concentrations and physico-chemical parameters with respect to the geological influences and to characterize the water samples from both sources.

2. Materials and methods

2.1. Study area

Malacca is a state situated in the southwestern part of Malaysia which borders Negeri Sembilan to the north and

west, and Johor to the south. The estimated population of Malacca is 913,210, with major land use as agricultural activities. Its small size and annual volume of rainfall result in limited water resources as compared to other Malaysian states [24]. Mining operation started in the 1820 s and expanded widely into the 1870 s until now. Tin mining is dominantly practiced with the resulting tailing which contains sand (80%), slime (20%), and sandy slime [12]. The geology of Malacca is mostly influenced by phyllite. However, there is sufficient quantity of limestone in Jasin district with substantial alluvium and schist sand extended up to Johor state [25]. Similar earlier findings by Schwartz and Askury [26] reported a sequence of limestone and pelitic deposits in Malacca with granite abundance, which is less evolved in terms of chemical composition. The largest surface of ex-mining land (356 ha) is found in Jasin District, where most ex-mining ponds in this study are located (Fig. 1). The ex-mining ponds in Malacca are mostly utilized for recreational activities, such as picnics, surfing, and fishing; while the lakes are used for agricultural purposes and water supply as listed in Table 1. Agricultural activities around the studied sites are mostly rubber and palm oil cultivations with the largest rubber plantation industry in Malaysia, covering over 1,200 ha [27,28].

2.2. Materials

All reagents used were of analytical grade or better. Deionized water was obtained by using ELGA[®] PURELAB[®] UHQ II system (UK); HNO₃ Suprapur[®] grade from Merck (Germany); multi-elemental calibration standard solution was from Agilent Technologies (Newcastle); certified reference material for trace elements in freshwater (SRM1643f) from National Institute of Science and Technology (USA). The plasticware involved were soaked in 15% HNO₃ (v/v) and rinsed at least twice with UPW.

2.3. Sampling

By using Wildco water sampler, nine sub-samples were collected per site from a depth of 25 cm in October 2016 which usually records low rainfall [17,31]. The samples drawn were immediately preserved to determine the total metals by the addition of concentrated HNO₃ (Suprapur[®], Merck) to pH < 2 before being transported to the laboratory in acid pre-clean polyethylene bottles conserved at 4°C [32].

2.4. On-site measurement

The physico-chemical parameters, that is, pH, electrical conductivity (EC), dissolved oxygen (DO), ammoniacal nitrogen (AN), and total dissolved solids (TDS) were analyzed by using YSI Pro multi-parameter portable water quality meter (professional series); and the biological oxygen demand (BOD) was determined via portable water meter (s/no 005) to minimize temporal changes due to bacterial concentration.

2.5. Elemental analysis

The preserved water samples obtained from the study sites were filtered via 0.45 μ m polytetrafluoroethylene



Fig. 1. Map of sampling sites.

Table 1 Details of sampling sites

Туре	Coordinate	Sites	Code	Uses	Estimated size (Km ²)	Reference
Ex-mining	2.5168°N 101.9633°E	Lake Rantau	LR	Agriculture	1.98	[17]
	2.2705°N 102.4881°E	Lake Biru Chinchin 1	LBC1	Recreation	1.96	[29]
	2.2711°N 102.4939°E	Lake Biru Chinchin 2	LBC2	Recreation	0.89	[29]
	2.2743°N 102.4931°E	Lake Biru Chinchin 3	LBC3	Recreation	1.26	[29]
Lake	2.4440°N 102.4000°E	Jus Dam	JD	Reservoir	5.5	[24]
	2.3532°N 102.2989°E	Lake Durian Tunggal	LDT	Recreational and Reservoir	0.91	[30]
	2.2772°N 102.4850°E	Lake Tebat	LT	Agriculture	0.68	[30]
	2.2752°N 102.3022°E	Lake Bukit Katil	LBK	Agriculture	0.59	[30]

(PTFE) membrane filters prior to the analysis of metals and metalloids by using an Agilent 7500ce (USA) inductively coupled plasma-mass spectrometer (ICP-MS). The concentrations of dissolved As, Ca, Cd, Fe, Mg, Mn, Na, and Pb were determined under the operating conditions as listed in Table 2, which was verified via SRM1643f [15]. For quality assurance, the validity of the process was inspected for every 10 samples by using quality control samples prepared from appropriate dilution of the reference solution.

2.6. Metal index

Metal index (MI) conveys the overall water quality by taking in to account the potential health effects related to the presence of metals and metalloids [33]. The basis of evaluation entails comparison between the measured concentrations against reference values, which can be expressed as below [34]:

$$MI = \sum \frac{C_i}{(MAC)_i}$$
(1)

where C_i and $(MAC)_i$ are, respectively, the mean concentration for *i*th element and its maximum permissible level derived from the Malaysian Interim National Water Quality Standards (INWQS) of 50, 100, and 1,000 µg/L for As, Mn, and Fe, respectively [35]. The higher the ratio of C_i with respect to $(MAC)_{i'}$ the more deteriorated the quality of the water is. Therefore, MI > 1 is a threshold of warning which signifies a potential threat [33,36,37].

Table 2 ICP-MS operating conditions

Parameter	Setting
Plasma RF powers, W	1,600
Reflected powers, W	<15
Plasma gas flow, L/min	12
Carrier gas flow, L/min	0.9
Makeup gas flow, L/min	0.25
Collision gas	Helium
Collision gas flow, L/min	3–5

2.7. Multivariate analyses

The water quality dataset was preprocessed with Microsoft[®] Excel 2010, and the respective method detection limits were adopted for those non-detected cases. Pattern recognition techniques, that is, HCA, PCA, and LDA, were applied by using SAS[®] JMP Pro 12.

2.8. Hierarchical cluster analysis

HCA was performed on the standardized dataset to illustrate the spatial relation between the water samples by a dendrogram. The hierarchical linkages were constructed according to Ward's method, whereby dissimilarities were expressed in terms of Euclidean distance [16,38]. The hierarchical sequence of the selected water quality variables was further expanded via two-way clustering [39].

2.9. Principal component analysis

PCA was applied to the correlation matrix to explore the inherent pattern in the water quality dataset based on orthogonal transformation. This was achieved by summarizing the original variation through a manageable number of uncorrelated principal components [40]. The score, $z_{ij'}$ corresponding to *i*th PC of *j*th sample is given by linear combinations of *m* number of original water quality variables, $X_{mi'}$ which be written as:

$$z_{ij} = a_{i1}X_{1j} + a_{i2}X_{2j} + a_{i3}X_{3j} + \dots + a_{im}X_{mj}$$
⁽²⁾

where a_{im} reflects the loadings where the number of extracted PCs was depended on those with eigenvalue > 1.

2.10. Linear discriminant analysis

LDA was applied to infer the spatial classification of water samples (i.e., ex-mining pond and lakes) based on the relative similarities of the between-group and within-group [15]. The canonical score, $z_{jk'}$ corresponding to *j*th discriminant function for *k*th water sample is expressed as:

$$z_{jk} = w_0 + w_1 X_{1k} + w_2 X_{2k} + w_3 X_{3k} + \dots + w_n X_{nk}$$
(3)

where w_0 is an intercept, w is the discriminant weight, and X is the water quality variable.

3. Results and discussion

3.1. Quality assessment and quality control

The accuracy of ICP-MS measurement was checked as shown in Table 3 with mean recoveries that ranged between 90% and 107% (whereby coefficients of determinant $R^2 > 0.999$, in all cases; which demonstrated the fitness in the determination of those elements in freshwater with a certain degree of confidence [41,42]. The detection limits of the metals under consideration are 0.0002, 0.0001, and 0.6 mg/L, respectively, for As, Mn, and Fe.

3.2. Surface water quality

3.2.1. Physico-chemical characteristics

The physical-chemical measurements of the water catchments (Table 4) were found to be within the Drinking Water Quality Standard for raw water and Malaysia's INWQS [35,43]. The pH of all sampling sites ranged between 6.5 and 8.5, except LR (10.3 \pm 0.1) which was located in Negeri Sembilan state near Malacca with a more basic rock basement [44]. Similarly, LR that surrounded by much greenly vegetation has recorded the highest DO of about 9.16 mg/L, whereas less than 5 mg/L (the recommended level) were observed in both LT and LBK. The low dissolved oxygen levels might be associated with the low water level at LT, whereby the imbalance between oxygen supply from the surface waters and removal of oxygen from bottom water, as well as the organic matter corresponding to residential discharges into LBK [45,46].

3.3. Metal index

The water quality status of the ex-mining ponds and lakes are listed in Table 5, which revealed a notable variation. According to literature reports, high metal concentrations including As are commonly related to ex-mining ponds [16,17,47–50]. For instance, elevated metal levels were reported in ex-mining ponds located in Pahang, Malaysia. However, such a trend was not apparent, whereby As was the only toxic element that was detected in all sampling sites [20]. The low metal concentrations observed could plausibly be explained by the buffering of waterbody due to dissolution of limestone host rocks. The acidic solution produced due to oxidation of metal ores liberated more metals in the solution, but the acidity is neutralized by the influence of limestone basement in the study area [51], thereby lowering the metal levels [21,52–54].

Among the metals only As, Mn, and Fe are considered in the MI computation so as to minimize the possible additive effects from other essential elements [34,55,56]. Moreover, the reference value for Ca is not even specified under the Malaysian drinking water standards INWQS [35,43]. As shown in Table 5, LT is a popular fishing lake with an overall MI of 2.71. Such value gives a negative impression on the corresponding water quality as it exceeded the reference limit of 1, which suggests potential health risks associated with its use [57]. The noticeable contributions could be linked with the arsenical pesticides used to preserve wooden boats [58,59], in addition to the natural

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Table 3	
Performance of ICP-MS on SRM1643f	

Element	Certified value (µg/L)	Measured value (µg/L)	Recovery (%)
As	57.4 ± 0.4	57 ± 2	98 ± 5
Ca	$(29.4 \pm 0.3) \times 10^3$	$(27.8 \pm 0.2) \times 10^3$	95 ± 3
Cd	5.9 ± 0.1	6.0 ± 0.6	102 ± 9
Fe	93.4 ± 0.8	101 ± 1	107 ± 2
Mg	$(7.45 \pm 0.06) \times 10^3$	$(6.77 \pm 0.08) \times 10^3$	91 ± 11
Mn	37.1 ± 0.6	39.2 ± 0.3	105 ± 1
Pb	18.48 ± 0.08	17 ± 2	90 ± 9

Table 4

Physico-chemical characteristics of water samples from lakes and ex-mining ponds

Type Ex-mining				Lake				
Site	LBC1	LBC2	LBC3	LR	JD	LDT	LT	LBK
DO	5.1 ± 0.2	6.1 ± 0.1	5.0 ± 0.2	9.2 ± 0.3	4.9 ± 0.1	6.1 ± 0.5	4.6 ± 0.3	3.7 ± 0.1
BOD	0.90 ± 0.01	1.15 ± 0.02	1.16 ± 0.02	1.96 ± 0.01	1.25 ± 0.03	1.09 ± 0.01	2.12 ± 0.01	1.50 ± 0.01
pН	7.2 ± 0.1	7.3 ± 0.1	7.0 ± 0.1	10.3 ± 0.1	8 ± 1	7.2 ± 0.1	7.1 ± 0.1	6.9 ± 0.1
TDS	47.5 ± 0.1	59.0 ± 0.3	55.9 ± 0.1	77.0 ± 0.9	44.9 ± 0.1	61 ± 1	26.7 ± 0.1	48.0 ± 0.3
AN	0.02 ± 0.01	0.07 ± 0.01	0.07 ± 0.01	0.22 ± 0.02	0.28 ± 0.04	0.17 ± 0.01	0.03 ± 0.01	0.09 ± 0.01
EC	82.3 ± 0.1	102.3 ± 0.1	97.5 ± 0.1	136.0 ± 0.5	77.4 ± 0.1	107.6 ± 0.1	46.0 ± 0.1	80.0 ± 0.4

Electrical conductivity in µs/cm, pH (no unit), other physico-chemical parameters in mg/L.

Table 5 Metal index corresponding to water samples from lakes and ex-mining ponds

Туре	rpe Ex-mining				Lake			
Site	LBC1	LBC2	LBC3	LR	JD	LDT	LT	LBK
As	0.19	0.13	0.14	0.05	0.01	0.03	0.21	0.09
Mn	0.23	0.51	1.12*	1.32*	1.28*	0.20	1.07*	1.70*
Fe	0.01	0.07	0.31	0.34	0.19	0.11	1.44*	0.86
MI	0.43	0.71	1.57*	1.71*	1.49*	0.33	2.71*	2.65*

The concentration of Cd and Pb were less than the limit of detection, and metal indices were <0.03 and <0.002, respectively. *Metal and overall MI > 1.

abundance of Fe and Mn in the soils and earth's crust [60]. In this context, both Fe and Mn have considerably lower toxicity [61,62]; they can be significantly removed during the aeration stage of water treatment process [63–65]. Therefore, the impacts of Fe and Mn in LBC3, LR, JD, LT, and LB (MI > 1) should not be of health concern.

The MAC of the metals under consideration especially As (50 μ g/L) is similar to that in Georgia, but much higher than European union and World Health Organization values of 10 μ g/L [66]. Similarly, MAC for Mn is 400 μ g/L [56], which is higher than INWQS of Malaysia. The differences in MACs result in variations in MI values, whereby larger MAC accommodates higher metal concentrations [57]. The MAC for Fe is 1,000 μ g/L in all reference standards. Unless introduced in extreme quantities, iron does not cause any human health or other ecotoxicological risks [43,55,56].

The study findings on low metal concentrations in ex-mining ponds agreed with literature reports on the

geological analysis of Malacca ex-mining lands which showed little or no influence of metallic elements [29]. The variations in metal concentrations could be due to the natural percentage abundance and geological background. Therefore, it is imperative to analyze and monitor the presence and levels of metals in water so as to identify the possible risk associated with its usage [67].

3.4. Hierarchical cluster analysis

The clustering patterns of the sampling sites with respect to surface water quality were examined by using HCA. The sites could reasonably be grouped into two main clusters, reflecting the variability in water quality characteristics as outlined in the two-way dendrogram (Fig. 2). Due to the similarities in underlying geological features and water chemistry, the catchments located in Jasin District formed a major cluster that partitioned from LR sited at the neighboring state [15,22,68]. Under similar circumstances, the Jasin cluster could be further subdivided based upon the background and/or uses of the catchment sites where the ex-mining ponds (LBC1, LBC2, and LBC3) were clustered and distanced from other lakes. Despite the division due to background differences, the attributions from similarities in the land use helped the agglomeration, for instance, those ex-mining ponds that were reclaimed for recreational activities were linked with a recreational lake (LDT). Such clustering results suggested that the variability in surface water quality was strongly associated with the spatial pattern of catchment characteristics. In this regard, the variations of As, Mn, Fe, and BOD levels in catchments provided a reflection of the land use, as shown in Fig. 2.

3.5. Principal component analysis

The natural variations in the water characteristic between the samples from ex-mining ponds and lakes were explored with PCA, whereby the first two PCs described 46.2% and 23.6% of the total variance which mainly originated from the geological difference and land-use. As shown in Fig. 3, the PC1 loading with major components, that is, EC, DO, TDS, and Ca was indicative of the dissolution of rocks that corresponded to weathering/leaching [15]. In a similar context, the strong loadings of Mn, Fe, and BOD on PC2 could be attributed to the natural background concentrations and/or imbalance caused by the low water level of the sites. Low water levels could result in elevated concentrations of metals, especially with natural abundance. This is because, at low water level, there is a higher rate of metal exchange between the water column and bottom sediments. The extent of metal migration is influenced by its occurrence in the sediment, pore solutions in the bottom sediments, and the physico-chemical properties or states which emerge at the water/sediment boundary [69–71]. Similarly, low DO and high BOD are associated with the bottom water in the aquatic ecosystem due to the imbalance between oxygen supply from surface waters and the removal of oxygen from bottom water [45,46].

Period of low water levels was also reported to be characterized by high EC [72]. Ex-mining pond LR was clearly separated from all other studied sites along PC1 with higher component loading, which could be due to differences in land use and natural geological background. JD that is used as a reservoir is located around the origin; however, ex-mining ponds LBC and LDT which are utilized for recreational activities, showed deviation from the origin and were loaded with As, Mg, and Na. Similarly, LT and LBK utilized for agricultural purposes deviated from the origin along PC2 and loaded with Fe, Mn, and BOD.

3.6. Linear discriminant analysis

LDA was successfully applied to examine the discriminating features between the lakes and ex-mining ponds. Under such circumstances, ex-mining ponds are commonly associated with high metal levels as compared to lakes due to the impact of acidic pH [16]. The catchments of ex-mining ponds are commonly found in acidic nature



Fig. 2. Two-way clustering of lakes and ex-mining ponds in Malacca.



Fig. 3. PCA biplot of metals and physico-chemical parameters in lakes and ex-mining ponds in Malacca.



Fig. 4. Canonical plot for the discrimination of water samples from lakes and ex-mining ponds.

as the oxidation consequence of the parent ore. However, geological variations may lead to a basic pH to a certain extent. In a similar context, high EC is associated with an increased level of dissolved metals from crushed rocks by the ex-mining activities [16,73,74]. Therefore, As, Mn, Ca, Mg, and EC are the parameters associated with ex-mining ponds, whereas pH and Na are associated with lakes (Fig. 4). Such outcomes support that weathering and dissolution of rocks are the main sources of metals in the ex-mining ponds. For instance, weathering of host rocks and mine tailing were found associated with high metal levels in mines and adjacent rivers [75,76].

4. Conclusion

This work revealed the spatial variations in quality parameters between lakes and ex-mining ponds in Malacca. The results demonstrated that the quality of surface water could not be judged solely based on the historical identity of the sites because the impacts of geochemistry and current land-use are more crucial. The outcomes of multivariate analyses suggested the major variations in the water quality were derived from the dissolution of underlying rocks and current anthropogenic inputs, such as agricultural and recreational activities instead of ex-mining. Since the contribution of As was relatively low, the overall MI suggested that those sources could be considered as the potential reservoirs for potable supply after conventional treatment processes.

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