ORIGINAL ARTICLE

Classifying Sources of Nitrate Contamination in an Alluvial Deposit Aquifer System Using Hydrogeochemical Properties and Multivariate Statistical Techniques

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ABSTRACT

Introduction: This study determined nitrate concentration and identify the classifying sources of nitrate pollution in the alluvial deposit aquifer system in Bachok, Kelantan. **Materials and Methods:** A total of 300 groundwater samples were collected in two different areas; agricultural area (150 samples) and non-agricultural area (150 samples). The samples were analyzed for nitrate and other parameters such as pH, EC, NH4+, TDS, turbidity and salinity. The multivariate analyses were used to identify factors that govern the groundwater quality and potential nitrate sources in the study area. **Results:** Samples in the agricultural area were slightly acidic (5.89 ± 0.67), contained high nitrate (15.10 ± 15.90 mg/L NO3-N), NH4+ (0.82 ± 1.24 mg/L) and turbidity (3.25 ± 2.78 NTU). The principal component analysis (PCA) have identified the groundwater quality in the study area was influenced by the natural processes and anthropogenic activities. Based on the hierarchal cluster analysis (HCA), Cluster II in the agricultural area was identified to be most heavily nitrate contamination, while Cluster III in the non-agricultural area was identified to be strongly affected by seawater intrusion. **Conclusion:** The findings of this study are useful for developing protection alternatives of private well waters to prevent further deterioration of groundwater quality by nitrate such as control of nitrogen fertilizer use, manure applications and other agricultural practices in the agricultural area. In order to reduce the health risk of nitrate, private well water users in this area should be advised to treat their water or find alternative sources for drinking.

Keywords: Groundwater, Nitrate, Principal Component Analysis, Hierarchical Cluster Analysis

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INTRODUCTION

Less than 10% of the present water use in Malaysia is from groundwater resources (1). The groundwater is being utilized for public water supplies (65%), industries (30%) and irrigation (5%). The use of groundwater for domestic purposes is mainly confined to rural and remote areas, where there is no piped water supply (2-4). The public water supplies in Kelantan state in the eastern part of Peninsular Malaysia, mainly rely on groundwater (70%) (2, 3). In 2010, about 165 million liters of groundwater per day was withdrawn and it was expected to increase by 2.5% per year (5, 6). The demand for groundwater increases significantly in Kelantan as they are facing with water quality problems of it surface water resources (3, 4). However, the local residents of north Kelantan, especially those living in villages normally get their water supply from their private well; dug well or borehole types (6, 7).

The private well water users tend to be exposed more frequently to higher levels of nitrate because they used it without any treatment and monitoring, shallower and closer to nitrate sources compared to public supply wells (8-11). The choice of Bachok district for this study is based on the fact that the aquifer in this area is covered by alluvial deposit, made up of gravel, sand, clay and silt, which is fragile and easily depleted due to over exploitation of groundwater and anthropogenic activities at the surface (6, 12). Being shallow and relatively unprotected, the aquifer in this area is exposed to higher risk of nitrate contamination.

Futhermore, land use of Bachok district which is covered primarily by the agricultural area also poses a significant potential for groundwater contamination by nitrate. Extensive research has indicated that agricultural activities may cause groundwater nitrate contamination and exceed the maximum acceptable level of nitrate for drinking water (10 mg/L NO3-N) (3, 13, 14). According to Nemčić-Jurec and Jazbec. (2017), the agricultural activities contribute to an increased nitrate concentration in groundwater either directly by nitrate leaching from agrochemicals or by agrochemicals that affect the processes in the soil and increase nitrate leaching from the soil (15). Other than agricultural activities, the population growth in the Bachok district also increases the risk of nitrate contamination to the groundwater in the non-agricultural area. Dense populations and discharges from point sources like septic systems or broken sewer systems contribute significantly to groundwater contamination by nitrate (11, 15, 16).

Investigations from previous studies have discovered the effects of nitrate on human health including methemoglobinemia, multiple sclerosis, Non-Hodgkin lymphoma, diabetes and thyroid gland hypertrophy and gastric cancer (17-21). Moreover, the discharge of high nitrate concentration to the surface water also has a negative impact on the environment, with the development of anoxic zones and eutrophication (20, 22, 23). World Health Organization (WHO) and Ministry of Health Malaysia (MOH) have recommended a nitrate concentration in drinking water less than 10 mg/L NO3-N (24, 25).

This study aims to measure nitrate concentration in the groundwater and classifying the source of nitrate pollution in the alluvial deposits aquifer system in the study area using the hydrogeochemical properties and multivariate statistical methods.

MATERIALS AND METHODS

Study area

This study was conducted in Bachok district, Kelantan, which located at 6^0 4′ 0″ North and 102^0 24′ 0″ East. The area is approximately 264.64 km3 with the mean of annual rainfall and temperature is 2700 mm/year and 27 0C, respectively. This is governed by the northeast and southwest monsoon which responsible for two distinct seasons; wet season and dry season. The northeast monsoon (wet season) occurs between November to March regularly generates high rainfall intensity and causes flood events (6). Meanwhile, the southwest monsoon (dry season) occurs between February and October often generates a low intensity of rainfall (6, 26). The mean of relative humidity is ranging from 80 – 87% with the seasonal variation conforms to the rainfall distribution pattern in the area (26).

The study area has a gentle-to-flat topography with mean elevation less than 15 m (26). The abundance of groundwater resources is related to the geologic condition of the study area, which is covered by Quaternary alluvium (2, 26, 27). The Quaternary alluvium deposits mainly consist of unconsolidated to semiconsolidated gravel, sand, clay and silts that occupy in the study area and along the river valley (12, 27). The thicknesses of alluvial deposits are ranging from 25 m inland to > 200 m near the coastal area (12).

Sample size Calculation

Due to effects of nitrate on human health, the sample size based on the calculation formula for estimating the size for group comparison (two-groups) as follows;

$$\text{Formula, } n = \frac{\{Z_{1-\alpha/2}\sqrt{2\bar{P}(1-\bar{P})} + Z_{1-\beta}\sqrt{P_1(1-P_1)+P_2(1-P_2)}\}^2}{(P_1-P_2)^2} \quad \textbf{(Eq. 1)}$$

where,

$\bar{P}$ = $(P_1 + P_2) / 2$

P_1 = Estimated proportion, larger (33.8%) (Volkmer et al., 2005)

P_2 = Estimated proportion, smaller (17.1%) (Volkmer et al., 2005)

$Z_{1-\alpha/2}$ = Standardized value for confidence interval, 95% CI = 1.96

$Z_{1-\beta}$ = Standardized value for power, 80% of power = 0.842

The proportions (prevalences) from Volkmer et al. (2005) were used to calculate the sample size in the present study (28). A study by Volkmer et al. (2005) evaluated the effect of nitrate level in the drinking water on the incidence of urological malignancies in a German community (28). They found that the incidence per 100 000 inhabitants/year of urinary tract tumours was 33.8% in group A and only 17.1% in group B. The group A had a drinking water supply with nitrate level 60 mg/L and 10 mg/L in group B.

Example of sample size calculation in this present study as follow;

$$\bar{P} = (0.338 + 0.171)/2 = 0.255$$

$$n = \frac{\{1.96\sqrt{2(0.255)(0.745)} + 0.842\sqrt{(0.338)(0.662) + (0.171)(0.829)}\}^2}{(0.338 - 0.171)^2}$$

$$n = \frac{\{(1.96)(0.616)+(0.842)(0.605)\}^2}{0.028}$$

$$n = 105.26 \approx 105 \text{ for each group}$$

Based on the calculation, it is estimated that a minimum number of 105 well waters are required to each group to examine the significant differences in the health risk of nitrate exposure experienced by the respondents in the agricultural and non-agricultural areas at 95% level of confidence and power of 80%. This study was able to gather a total of 300 well water samples which include 150 well water samples from the agricultural area and 150 well water samples from the non-agricultural area (Fig. 1).

Sampling and data collection

This study compared the concentration of nitrate in groundwater samples between the agricultural and non-agricultural areas. The agricultural area refers to the

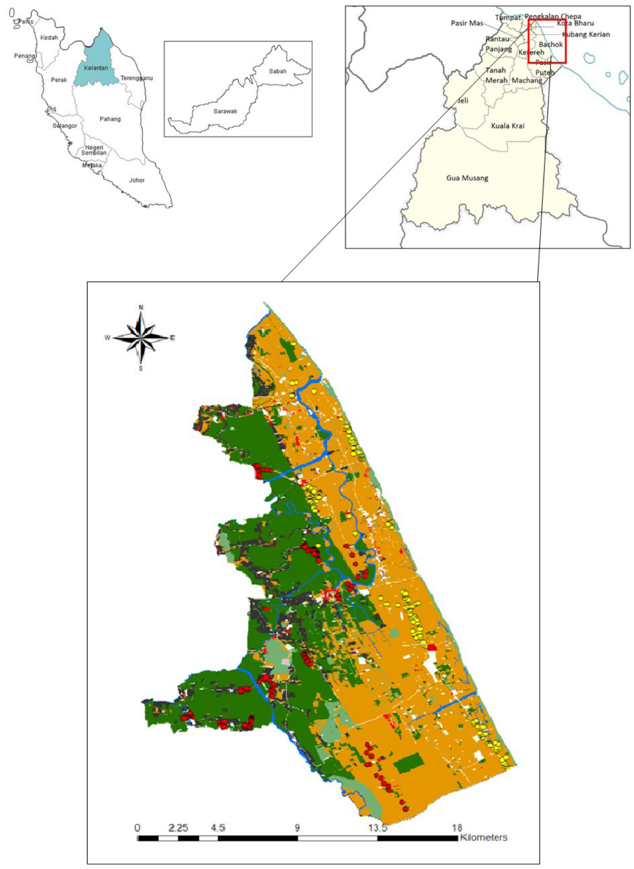

Figure 1: Location of the study area and sampling points (n = 300)

paddy farming area and non-agricultural area refers to the residential and industrial area. All the private wells chosen for groundwater sampling were used for drinking. A Global Positioning System (GPS) was used to record the spatial location (i.e., longitude and latitude) of each sampling well. Nitrate and physicochemical parameters were measured immediately during each field work using the YSI Professional Plus handled multi-parameter meter. This is to acquire representative values of water quality and also to avoid any biochemical changes in the samples (29).

Statistical analysis

Before performing multivariate statistical analysis, standardization of data using Z-score has been done to avoid misclassification of results or being influenced by great magnitude variables (30, 31). The Z-scores was used to standardize the data using the equation below (30);

$$Z_i = \frac{x_i - \text{mean}}{s} \qquad \textbf{(Eq. 2)}$$

where;

Z_i = Standardized Z scores

X_i = Value of each variable

mean = Mean value of the normal distribution

s = Standard deviation of the distribution

Principal Component Analysis (PCA) was used in this study to transform a complex data of groundwater to a smaller set of uncorrelated comprehensive components and arrange them in decreasing order of importance for easy data interpretation without losing significant information (30, 32, 33). The strength of associations are classified as loading values > 0.75 as 'strong', loading values 0.75 – 0.5 as 'moderate'; and loading values 0.5 – 0.3 as 'weak' (34). Only components with eigenvalues equal or greater than one were selected and rotated by the varimax method to enhance and strengthen the differences between the components (31, 33, 35).

Hierarchical Cluster Analysis (HCA) was performed to group sampling groundwater into clusters on the basis of similarities within a class and dissimilarities between different classes which respect to the pre-determined characteristics (32, 33, 35, 36). In the present study, Ward's method was used as linkage rule and Euclidean distance for the classification scheme. The results were illustrated by a dendrogram. The statistical analyses were carried out using IBM SPSS version 21.0.

RESULTS

Nitrate concentration and hydrogeochemical parameters of the study area

Groundwater samples from the agricultural area were slightly acidic (5.89 ± 0.67), contained high nitrate (15.10 ± 15.90 mg/L NO3-N), ammonium (NH4+) (0.82 ± 1.24 mg/L) and turbidity (3.25 ± 2.78 NTU). In contrast, samples in the non-agricultural area were neutral water pH (6.44 ± 0.77), have high electrical conductivity (EC) (230.46 ± 22.88 μS/cm) and total dissolved solids level (TDS) (176.13 ± 178.15 mg/L) and low level of nitrate (5.81 ± 5.08 mg/L NO3-N).

Sixty-nine (69, 46%) groundwater samples from the agriculture area and 36 (24%) samples from the non-agricultural area exceeded MOH and WHO drinking water quality standards for nitrate (10 mg/L NO3-N) (24, 25). NH4+ levels in 23 (15.33%) samples in the agricultural area and five (3.33%) samples in the non-agricultural area were more than 1.5 mg/L, the maximum acceptable limit in drinking water recommended by MOH (24). Turbidity level in 33 of the samples in the agricultural area exceeded the WHO and MOH-recommended value (5 NTU) (24, 25). Meanwhile, seven (4.67%) samples in the non-agricultural area exceeded allowable limit of EC in drinking water by the WHO (500 μS/cm) (25). Only three (2.0%) samples in the non-agricultural have exceeded allowable limit of TDS in drinking water recommended by MOH (1000 mg/L) (24).

Principal Component Analysis (PCA)

The Kaiser-Meyer-Olkin values for the PCA model in the agricultural and non-agricultural areas were 0.762 and

0.771 respectively, which supporting the factorability of the correlation matrix (Table I). Seven factors were yielded for both areas (PC1-7), however, only factors with eigenvalues greater than or equal to 1 were accepted as possible sources of variance in the data. Hence, first three and two factors in the agricultural and non-agricultural areas were selected, respectively. The contribution of the first three factors (PC1, PC2 and PC3) in the agricultural area were 57.08%, 16.08% and 14.83%, respectively, accounting for approximately 87.99% of the total variance. Meanwhile, in the non-agricultural area, two factors (PC1 and PC2), were affecting the quality of groundwater were identified, which explains 65.47% of the total variance. PC1 and PC2 account for 48.44% and 17.04% of the total variance, respectively.

The varimax rotated component matrix loadings for the three components for the agricultural area are presented in Fig. 2 where evidently showed strong-to-moderate positive association with EC (0.988), TDS (0.984), salinity (0.979), nitrate (0.833) and pH (0.600) in PC1. PC2 had a strong – to – weak positive association with turbidity (0.905) and pH (0.456) while PC3 had a strong positive association with NH4+ (0.952) and weak negative association with pH (- 0.341).

In the non-agricultural area, PC1 had strong-to-moderate positive association of EC (0.953), TDS (0.951), salinity (0.903), pH (0.636) and NH4+ (0.573) while PC2 had a strong positive association with nitrate (0.747) and high negative association with turbidity (- 0.774) (Fig. 3).

Hierarchical Cluster Analysis (HCA)

The HCA analysis resulted in three clusters for both agricultural and non-agricultural areas (Fig. 4 and 5). The mean + SD and range values of parameters are presented in Table II to quantify the variation of parameters between clusters.

Most of the samples in the agricultural area were grouped in Cluster I (n = 109) followed by 23 samples in Cluster II and 18 samples in Cluster III. Nitrate concentration in Cluster II (40.45 ± 8.61 mg/L NO3-N) and III (28.80 ± 20.84 mg/L NO3-N) were beyond the maximum contaminant level value (10 mg/L NO3-N). The groundwater samples in Cluster II was slightly acidic while in Cluster III was alkaline. Nitrate and NH4+ concentrations were the highest in Cluster II, with the mean values of 40.45 mg/L NO3-N and 1.25 mg/L, respectively while EC and TDS were the highest in Cluster III.

As for samples in the non-agricultural area, most of the samples grouped in Cluster I (n = 110) followed by 33 samples in Cluster II and seven samples in Cluster III. Cluster I have pH, NH4+, EC, TDS and salinity at the lowest level among the three groups. Nitrate concentrations were the lowest in Cluster II. Meanwhile, all parameters showed the highest concentrations in Cluster III.

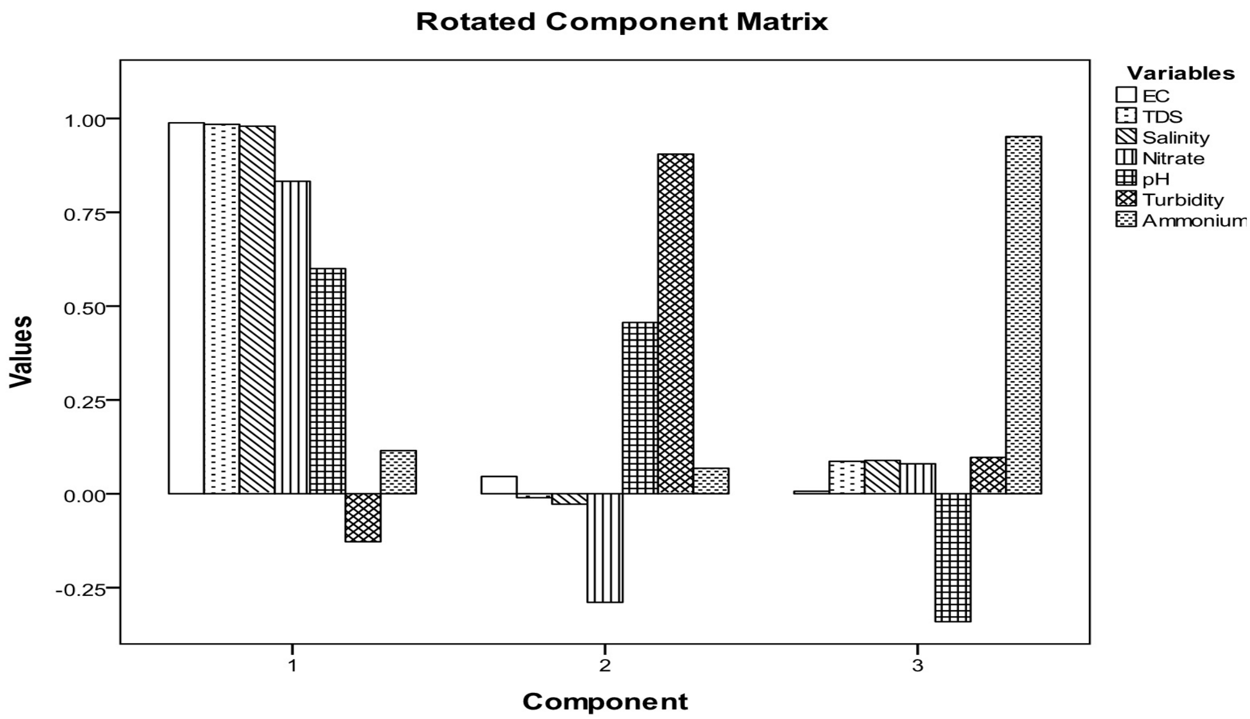

Figure 2: Loadings for rotated component matrix in the agricultural area (rotation method: Varimax with Kaiser normalization) of PCA (n = 150)

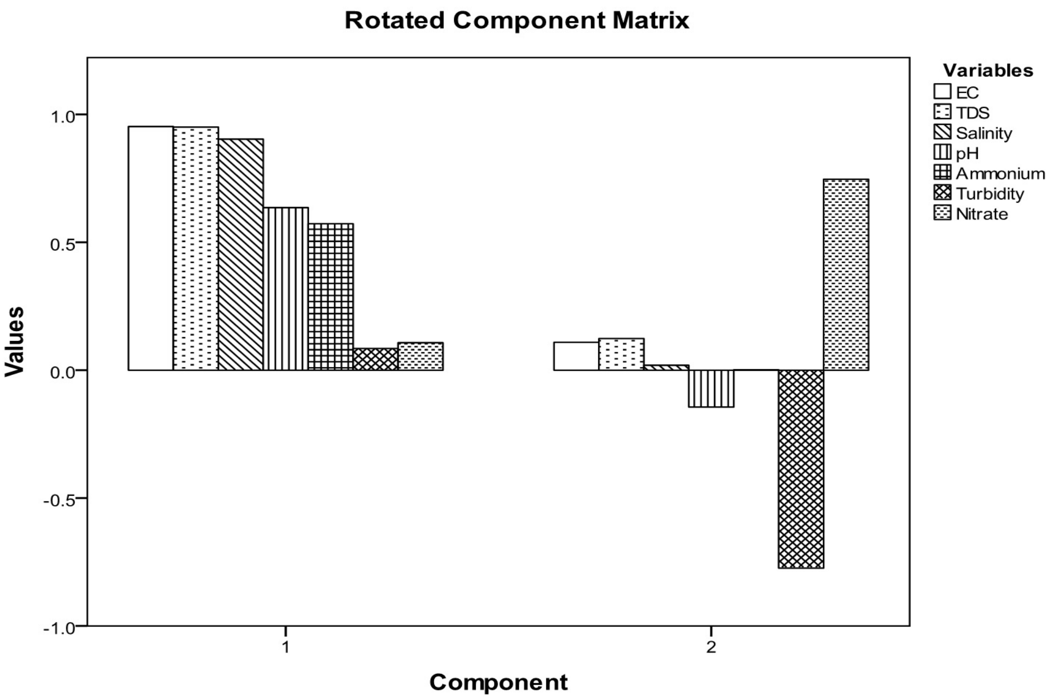

Figure 3: Loadings for rotated component matrix in the non-agricultural area (rotation method: Varimax with Kaiser normalization) of PCA (n = 150)

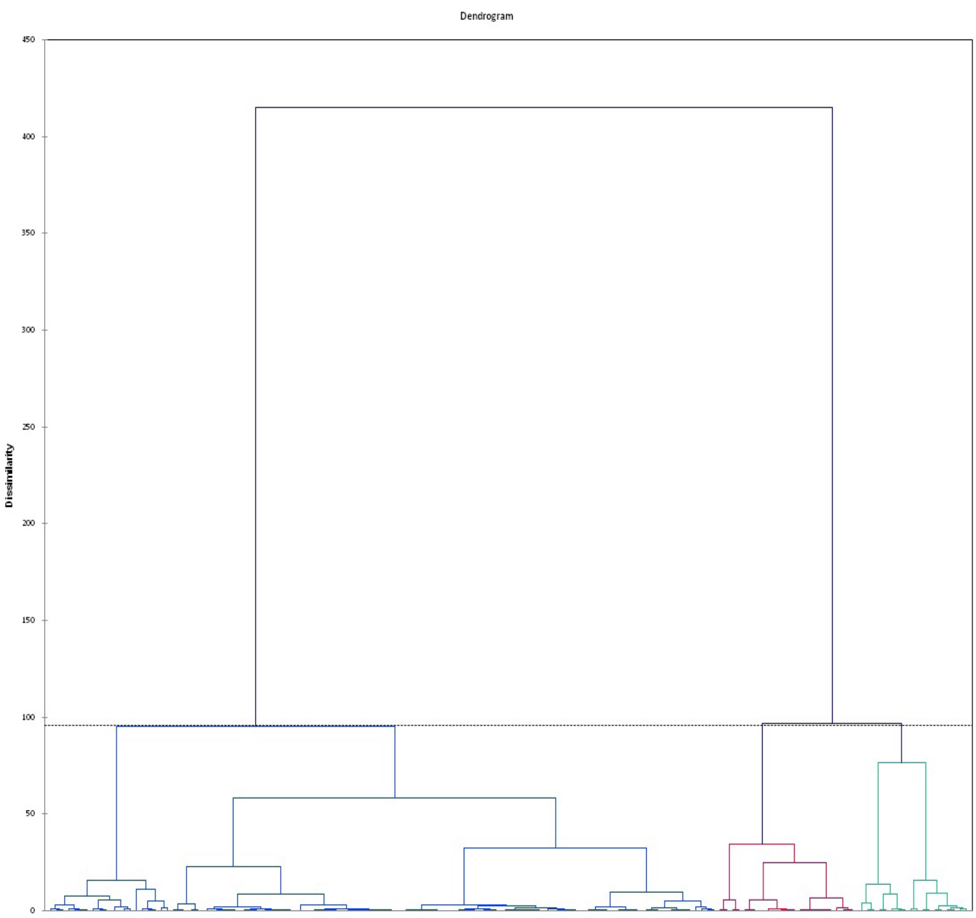

Figure 4: Dendrogram for the well water grouping with respect to their physicochemical parameters in the agricultural area (n = 150)

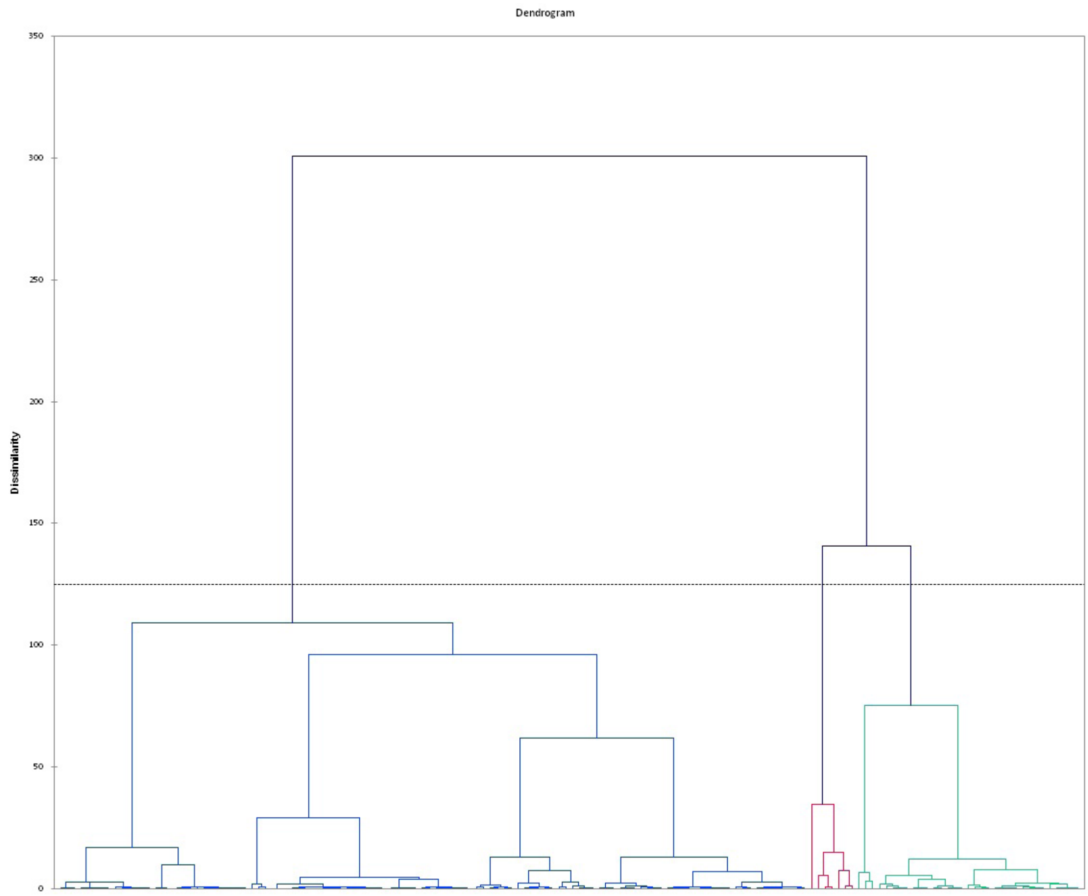

Figure 5: Dendrogram for the well water grouping with respect to their physicochemical parameters in the non-agricultural area (n = 150)

DISCUSSION

Factors affecting groundwater chemistry

The results form PCA analysis provided some information on factors affecting the chemistry of groundwater in the study area. In the agricultural area, high factor loadings on EC, TDS and salinity of PC1 indicates the occurrence of high dissolved ions in the groundwater such as inorganic salt and organic matter (30). The occurrence of dissolved ions in the groundwater was influenced by many factors such as evaporation, mineralogy, type of aquifers, as well as from CO2 which dissolves in precipitation as it falls to the ground (37). According to Shamsuddin et al. (2015), the presence of contaminant from the agricultural activities and groundwater contact with the rock and weathered rock also contributed to the high loadings of these parameters (38). Meanwhile, high factor loading on pH of PCI is likely due to the hydrolysis of bicarbonate (OH-) and the dissociation of H2CO3 (H+) in the groundwater (32).

High factor loading on nitrate of this factor was attributed to the agricultural activities such as the application of nitrogen fertilizers, animal waste and mineralization of soil organic nitrate in this area (32, 34). According to Mahasim et al. (2005), nitrate from nitrogen fertilizers used in the plantation and fields can be transported into the wells either vertically or horizontally with the rainwater during the rainy season (39). Hussain et al. (2013) concluded that soil physical properties contributed the most variations of nitrate leaching in a paddy field at Kelantan (40). However, in this study, soil physical properties of the study area were not evaluated.

PC2 had a strong positive loading on turbidity. Turbidity is related to suspended matter such as clay, silt and organic matter (10, 41). The study area is underlain by Quaternary alluvium deposits which mainly consist of unconsolidated to semiconsolidated gravel, sand, clay and silts (6). Therefore, this correlation indicates the influences of geology on the turbidity of groundwater samples in the agricultural area. A study by Kura et al. (2013) in Kapas Island's groundwater also found microbial activity and organic matter are associated with an increased turbidity (42). High positive loading on NH4+ of PC3 was related to the organic matter contents of the sediment (30). Usman et al. (2014) also reported a similar result in their study and indicated the high amount of NH4+ in groundwater in the agricultural area resulted from the manure application (43).

In the non-agricultural area, the PC1 had high positive loadings on EC, TDS and salinity, indicate the water-rock interaction and seawater intrusion (34). The location of the non-agricultural area in the coastal area confirms the suggested hydrogeochemical explanation (10, 34, 37). This finding is consistent with Gasim et al. (2017), which stated that these parameters are related to saline water contamination into the coastal aquifer (44). The high loading on NH4+ in the non-agricultural area is likely due to the result of organic matter degradation in the aquifer and the influenced by other sources such as sewage infiltration, livestock operations and industrial processes that located nearby the sampling sites (30, 44). This finding is consistent with Mohd Kamal et al. (2015), which found high NH4+ in the urban area. Septic system also results in locally elevated recharge rates of NH4+ (45). According to Mohd Kamal et al. (2015), NH4+ in the aquifer can cause degradation of groundwater quality and usability, it can substantial effects on water-rock interactions, and it can be a substantial source of nitrogen in surface waters receiving groundwater discharge (45).

High positive loading on nitrate of PC2 in the non-agricultural area could be due to the anthropogenic activities such as livestock activities and septic systems (46). Narany et al. (2017) conducted a study in the northern Kelantan clearly indicated the significant role of anthropogenic sources such as livestock areas, discharge from septic tanks, and leaking sewers in the residential area to the groundwater nitrate contamination (14). Shamsuddin et al., (2015) also reported the same result, showed the seepage of the liquids due to the occurrence of sewage and septic tanks contributed to high nitrate concentration in groundwater (38). However, the natural sources of nitrate such as local mineral deposits (e.g., potassium nitrate), decomposition of biomass, atmospheric precipitation (as NH3), nitrogen-fixing bacteria activities and nitrogen-bearing rock units also contribute to high groundwater nitrate concentrations in the non-agricultural area (47, 48).

The strong negative loading on the turbidity of this factor is likely due to the location of the non-agricultural area which situated in the coastal area. The sandy soil of the coastal area which has low suspended solids contributes to the negative association of turbidity of this factor. A study conducted by Gasim et al. (2017) in East Coast of Terengganu stated a possible explanation to this occurrence probably due to the presence of the coloured dissolved organic matters that absorb UV and blue wavelength in water presence it may reduce the turbidity readings (44).

Spatial similarity and site grouping

In the agricultural area, samples in the Cluster II were identified to be most heavily nitrate contamination. The mean of nitrate was at least four times as high as maximum nitrate levels in the drinking water quality standard. The application of nitrogen fertilizers in this area may lead to nitrification process. The ammonia compounds in the fertilizer are oxidized to nitrate by ammonia-oxidizing bacteria under aerobic conditions (31). This process produces hydrogen ions, leading to decrease in pH of groundwater. Therefore, high nitrate in the groundwater is predicted to have low levels of pH. Hamzah et al. (2014) also found a similar result in Kelantan and they stated that the lower pH is probably due to the acidic type of rock of the well location, in which most of the sampling sites are located on granite rocks which consist of acid intrusive rocks (49). Besides, it may also be attributed to the interaction of surface water and the carbon dioxide in the atmosphere and gain entry into the well. When the groundwater from an open dug well is exposed to the air, the carbon dioxide that presents in the air gets dissolved in the water and results in decreasing of pH (49).

In the non-agricultural area, mean values of nitrate in all cluster did not exceed the drinking water quality standard, however were slightly higher than the natural concentrations of nitrate in groundwater (3 mg/L NO3-N). Based on the physicochemical parameters of Cluster III, it was observed that seawater intrusion strongly affected this cluster. These samples were characterized by high EC, TDS and salinity values compared to the other clusters. The location of the non-agricultural area in the coastal area, shallow groundwater levels and flat topography of the study area may cause seawater moves inland during high tides and be a factor to high salinity of groundwater (46). Samsudin et al. (2008) also reported the high salinity of the aquifer in the coastal area of northern Kelantan was initially believed to be associated with the influence of seawater, which had intruded into the aquifer (50). According to Mohammed and Ghazali. (2009), high pumping rates and continuous pumping from the wells near the coastal areas will lead to saltwater intrusion (27).

CONCLUSION

The multivariate statistical methods were applied in this study to analyze complex data of groundwater samples. The PCA results indicate factors that influence groundwater quality in this study were related to the natural processes and anthropogenic activities. Based on HCA results, Cluster II in the agricultural area was identified to be most heavily nitrate contamination, while Cluster III in the non-agricultural area was identified to be strongly affected by seawater intrusion. The results from this study can be used for future groundwater management plans in the study area. For example, the information about the levels and sources of nitrate in the groundwater are useful to the responsible agencies such as Department of Environment (DOE), Department of Mineral and Geoscience (DMG), Ministry of Health (MOH) and Department of Agriculture (DOA) for developing protection alternatives such as control of nitrogen fertilizer use, manure applications and other agricultural practices in the agricultural area which contributing to the groundwater nitrate contamination.

Many private well water users lack the knowledge and resources needed to routinely monitor and maintain their well water. So, they can be educated through outreach programmes that explain how activities at the surface ultimately impact the groundwater quality.

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