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ORIGINAL ARTICLE

Multivariate statistical analysis for identifying water quality and hydrogeochemical evolution of shallow groundwater in Quaternary deposits in the Lower Kelantan River Basin, Malaysian Peninsula

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Abstract A long-term hydrogeochemical data set is used in this study to evaluate the water quality and hydrogeochemical evolution of shallow groundwater in a Quaternary deposit. A multivariate statistical method, hierarchical cluster analysis (HCA), is applied to overcome the problem of a large number of data points in the integration, interpretation and representation of the results. HCA is applied to a subgroup of the hydrogeochemical data set to evaluate their usefulness to classify the groundwater bodies. This subgroup consists of 27 groundwater wells and 15 variables [pH, total dissolved solids, electrical conductivity (EC), Na^+, Ca^{2+}, Mg^{2+}, K, HCO_3^-, Cl^-, SO_4^{2-}, Fe, Mn, NH_4, NO_3^- and SiO_2]. Only 12 chemical variables were used for the analysis. Four clusters have been identified: C1–C4, with two main prevalent facies, Na–HCO_3 and Ca–HCO_3. The hydrogeochemical evolution of shallow groundwater is governed by the processes of precipitation, weathering, dissolution and ion exchange.

Keywords Hydrogeochemistry · Water quality · Groundwater evolution · Hierarchical cluster analysis (HCA) · Quaternary deposits of Lower Kelantan River Basin

Introduction

The study area is located on the northeast coast of the Malaysia Peninsula (Fig. 1) and experiences a tropical rainforest climate, controlled by two monsoons. The southwest monsoon occurring between February and October often brings less rainfall, whereas the northeast monsoon from November to March frequently generates high rainfall intensity over the study area. The mean annual rainfall (1989–2012) recorded from 4 stations is 2649 mm (MGD 2012). Groundwater has been used in Kota Bharu, Kelantan, since 1935 (SMHB 2000). The demand for groundwater for potable usage is estimated to be 163 Ml/d in 2010 and is expected to increase 2.5 % per year (Suratman 2010). Most of the groundwater is abstracted from shallow aquifers except in Tanjung Mas, Pintu Geng and Kg. Puteh where groundwater is drawn from a deep aquifer (PMW 2010). In order to meet the usage demand, the local water authority Air Kelantan Sdn. Bhd (AKSB) has been constructing horizontal collector wells at Wakaf Bharu, Pintu Geng and Kg. Chap (under construction). The major problem in groundwater supply is the high iron concentration that can reach up to 80 mg/L (Hussin 2011). Groundwater supply in Kota Bharu has been treated using conventional treatment and ozonation methods. An alternative iron treatment method using ionic liquid green technology at the laboratory scale was achieved with 95 % of iron removed from the initial concentration. Detailed research, however, is needed before it can be applied for regional- or larger-scale treatment (Hussin et al. 2014). Groundwater resource availability in the Lower Kelantan River Basin depends on thick sequences of Quaternary deposits, with thicknesses of ranging from 25-m inland toward 200 m to the coastal area (Suratman 1997).

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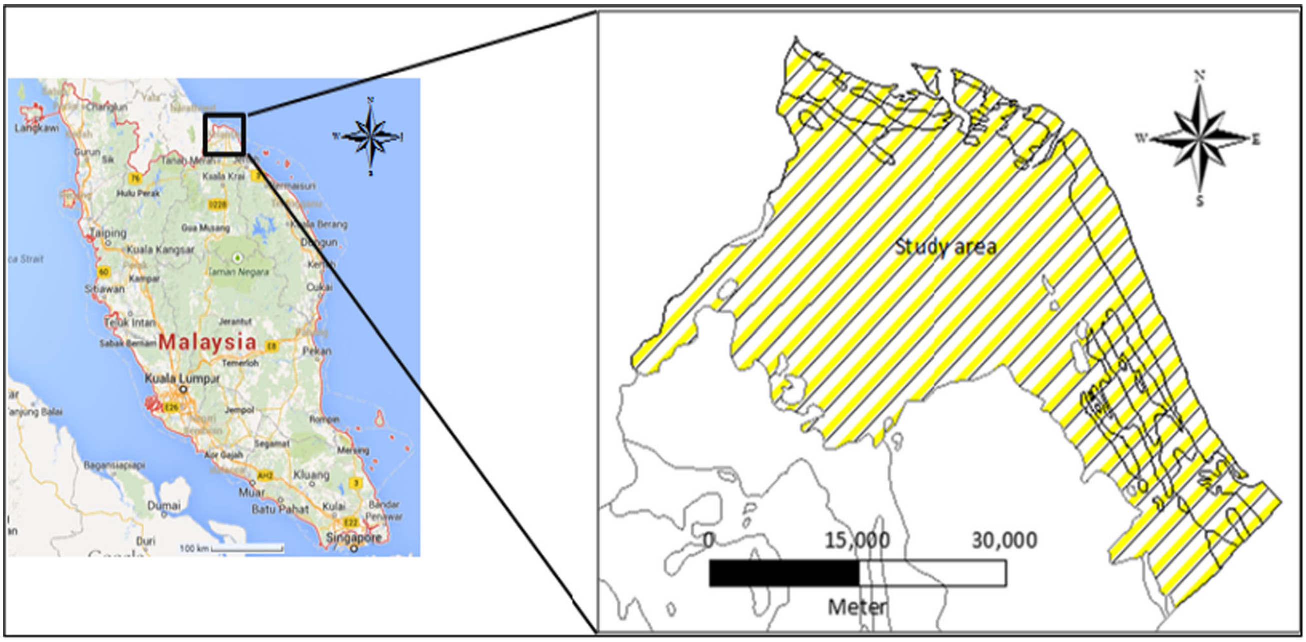


Fig. 1 Location of the Quaternary alluvium aquifer system, Lower Kelantan River Basin

The aims of the study are to identify the groundwater quality and the processes controlling the geochemical evolution of shallow groundwater based on available long-term data recorded from the Minerals and Geoscience Department of Malaysia (MGD) monitoring wells in the Lower Kelantan River Basin. The most well-known multivariate statistical method used in environmental studies, hierarchal clustering analysis (HCA), is used to characterize the hydrogeological and geological context in terms of water quality and hydrogeochemical evolution in the regional basin setting of the Quaternary deposits. HCA therefore is relevant and integral to the aim of the study in comparison with the conventional method of geochemical grouping. This could be beneficial to enhance the management of the basin for future planning and development of the groundwater resources.

Multivariate statistical analysis in hydrogeochemical studies has been proven in numerous related publications such as identifying the groundwater resources, groundwater quality, groundwater flow paths in complex aquifers, aquifer boundaries, geochemical control on composition as well as for separating anomalies such as anthropogenic impacts from background (Steinhorst and Williams 1985; Farnham et al. 2000; Alberto et al. 2001; Lopez-Chicano et al. 2001; Stetzenbach et al. 2001; Locsey and Cox 2003; Pereira et al. 2003; Belkhiri et al. 2010). This type of analysis has been done in combination with classical hydrogeochemical methods such as Piper and Scholler diagrams and Stiff patterns as well as the use of ArcGIS to show the spatial variation mapping of quality characteristics or factors influencing the evolution (Ceron et al. 2000; Güler et al. 2002; Güler and Thyne 2004; Papatheodorou et al. 2007; Helsrup et al. 2007; Andrade et al. 2008; Cloutier et al. 2008; Li and Zhang 2008; El Yaouti et al. 2009; Dassi 2011; Monjerezi et al. 2011).

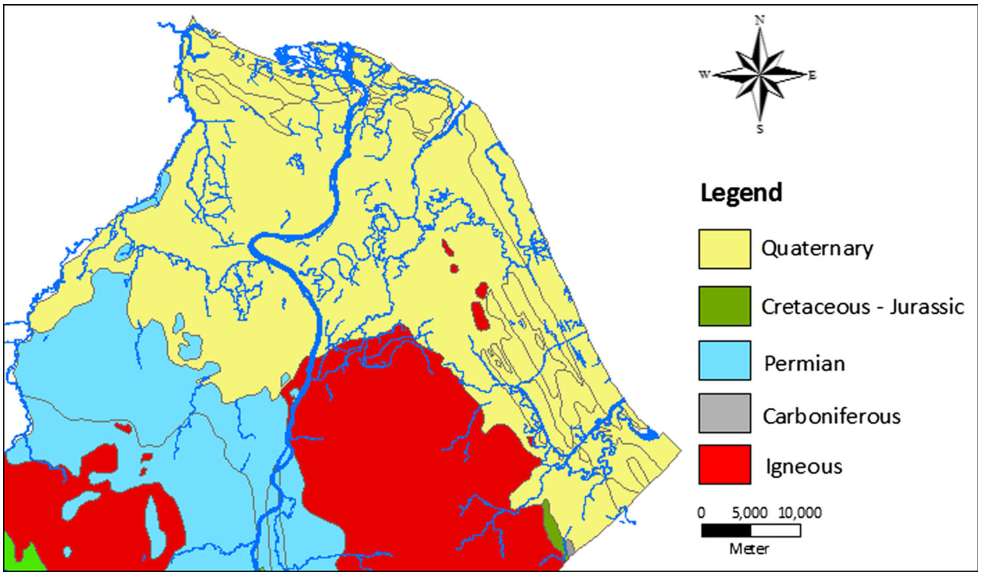
Study area

Geology and hydrogeology

The topography of the study area is generally low lying with a mean elevation <15 m (Hutchison and Tan 2009). The Lower Kelantan River Basin is covered by Quaternary Alluvium deposits (Fig. 2). The depositional environment is shallow marine to estuarine and fluvial (Suntharalingam and Teoh 1985; Loh 1992). This alluvium deposit is lying unconformably on a Permian and Igneous basement. The highest hill is known as Bukit Marak, at 373 m (MacDonald 1967).

This basin is drained by six major rivers; Golok River, Kelantan River, Pengkalan Chepa River, Pengkalan Datu River, Kemasin River and Semarak River. Rivers are considered as potential recharge or discharge areas for groundwater flow (Sofner 1992). A cross section of the Kelantan river bed up to 16 km from the estuary indicated a deep incision into the shallow aquifer and indirectly shows a connection between river and shallow groundwater (Chong and Tan 1986). Regional groundwater recharge is directly from precipitation; meanwhile, the return flow is from irrigation and bank infiltration. Deep aquifer recharge occurs when heads in the shallow aquifer are higher than in the deep aquifer, and the amount depends on the hydraulic

Fig. 2 Geology map of the study area



gradient and permeability along the flow path (Sofner 1992). Estimated value 11–12 % of recharge was used in most numerical modeling conducted for this basin with 5 % recharge used for areas with 1–5 m thicknesses of clay (protective layer) (Tajul Baharuddin 2002; MGD 2012, 2014).

The Quaternary alluvium deposit forms an aquifer system of the Lower Kelantan River Basin, which is characterized by thick sequences of alluvium ranging from 25 m inland up to 200 m at the coastal area (Suratman 1997). The aquifer thickness varies by location. Three layers of aquifer have been identified within the Quaternary alluvium deposit (Ang and Loh 1975; Noor 1979; Chong and Tan 1986; Pfeiffer and Tieddemann 1986; Mohammad 1992; Mohamad and Ang 1996; Hamzah et al. 1997). Layer 1 lies at a depth of approximately 20 m below the ground surface. Layer 2 lies approximately between depths of 20–50 m, while Layer 3 lies at a depth of 50 m. Layer 1 consists of fine light brownish sand with gray clay and shells. Layer 2 consists of light gray, medium to coarse sand. Coarse sand with fine to medium gravel was found in Layer 3. This aquifer is separated by an impervious clay layer with soft, blue-gray clay and shells (Pfeiffer and Tieddemann 1986). Hydraulic continuity between the shallow and deeper aquifer has been demonstrated at Kg. Chap, Bachok, during pumping tests (Noor 1979). The tests carried out at Kota Bharu Waterworks did not reveal any connection between the shallow (Layer 1) and deep (Layer 2 and 3) aquifer (Pfeiffer and Chong 1979). Therefore, a continuity throughout the basin is still to be determined. During the German Mission in Malaysia, Sofner (1992) stated that “the former concept of a three-fold aquifer system which might be true for limited sections has to be considered under regional aspects.” Figure 3 shows the 3D conceptual stratigraphy of the Lower Kelantan River Basin suggested by Sofner (1992). Aquifer Unit 1 is the shallow unconfined aquifer; Unit 2 is the protective clay layer; Units 3a and 3b are the deep aquifer areas consisting of the gravely sand and sandy silty clay, respectively; and Unit 4 is the coarse sand confined aquifer. The contour distribution of piezometric heads for the shallow aquifer is shown in Fig. 4, while Fig. 5 shows the cross-sectional A–A′ and B–B′ of the shallow aquifer (MGD 2012).

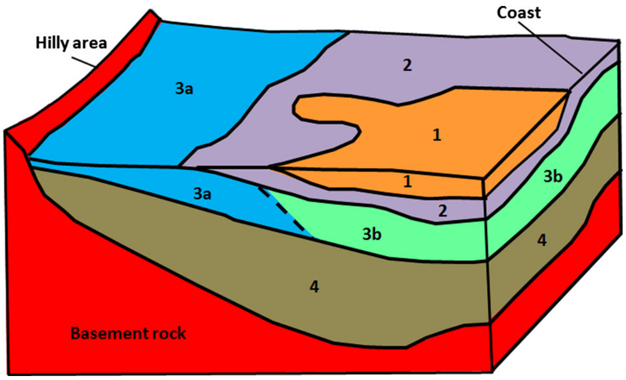


Fig. 3 3D conceptual stratigraphy of the Lower Kelantan River Basin (not to scale). Aquifer Unit 1 is the shallow unconfined aquifer; Unit 2 is the protective clay layer; Units 3a and 3b are the deep aquifer areas consisting of the gravely sand and sandy silty clay, respectively; and Unit 4 is the coarse sand confined aquifer

Methodology

Hydrochemical data

Long-term hydrochemical data recorded from 1989 to 2012 were obtained from the Minerals and Geoscience

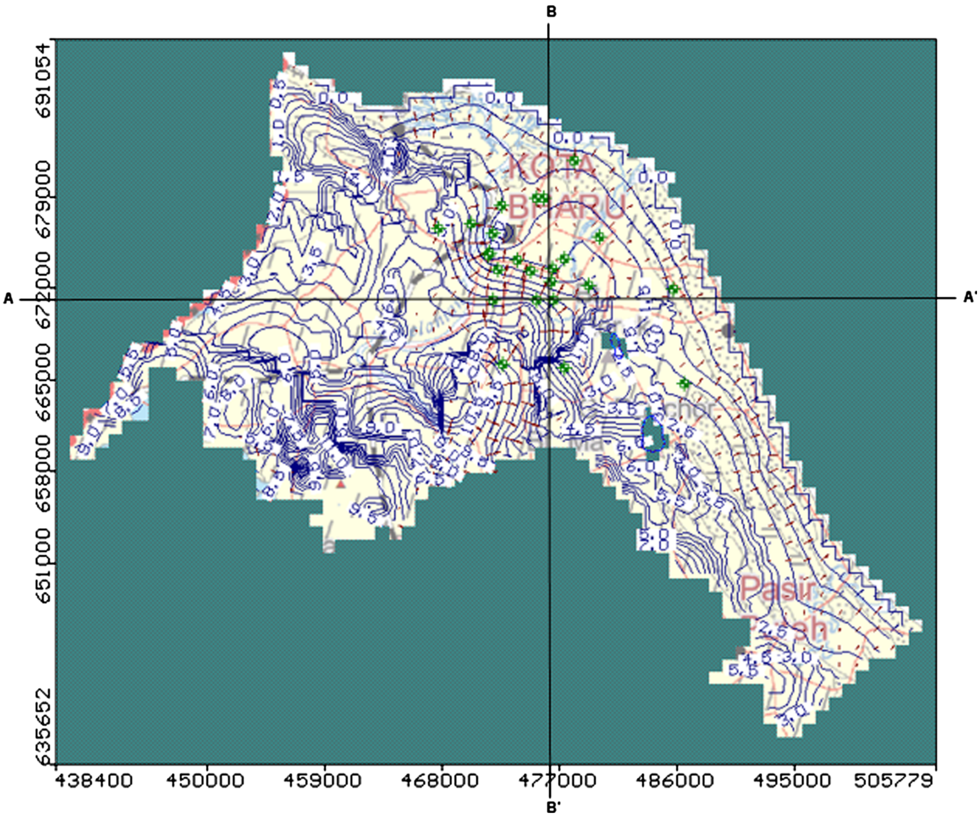
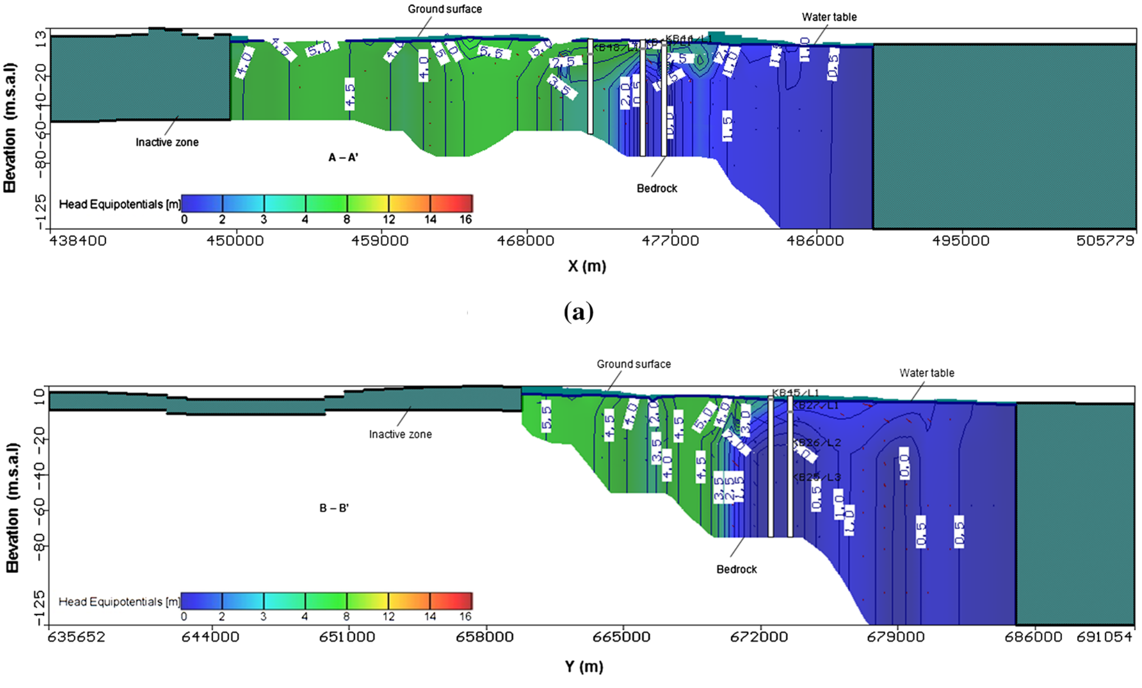


Fig. 4 Contour distribution of the piezometric heads (unit in m.a.s.l and 0.5-m interval) and velocity vectors in the study area (maximum velocity approximately 0.00015 m/s) in the unconfined shallow aquifer. The *green dots* represent the monitoring wells



(b)

Fig. 5 **a** Cross-sectional A–A′: equipotential head distribution (unit in m.a.s.l and 0.5-m interval) and velocity vectors (maximum velocity approximately 3.2 × 10^{-5} m/s); **b** cross-sectional B–B′: equipotential head distribution (unit in m.a.s.l and 0.5-m interval) and velocity vectors (maximum velocity approximately 1.9 × 10^{-5} m/s)

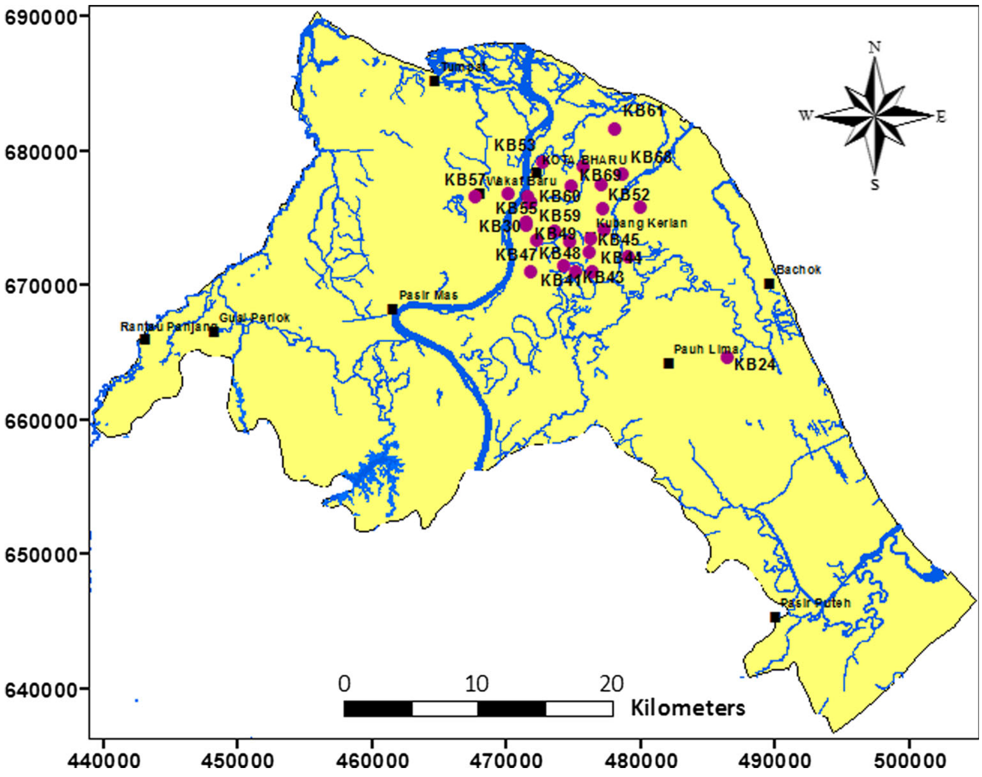


Fig. 6 Distribution of groundwater wells in the Layer 1 aquifer

Department of Malaysia (MGD). The data set focuses on the shallow groundwater, which consists of 27 wells (Fig. 6). These wells have been verified with ion balance errors below ±10 % (Güler et al. 2002; Guggenmos et al. 2011; Moya et al. 2015), which are applied in this multivariate analysis. In common practice, ±5 % of ion balance error is accepted (Freeze and Cherry 1979).

Data preparation for multivariate statistical analysis

Each groundwater well data set is characterized by physical and chemical variables, which presents the regional hydrogeochemical study as a multivariate problem as analysis involves numerous data samples with different hydrochemical problems. In this study, only chemical variables are used for multivariate analysis. Multivariate statistical analysis is a quantitative and independent approach of groundwater classification allowing the grouping and correlations between chemical variables and wells (Cloutier et al. 2008) to identify the most important factor contributing to the similarities between the factors. HCA is applied using SPlus 8.1 (TIBCO Spotfire S + 8.1 2008) software.

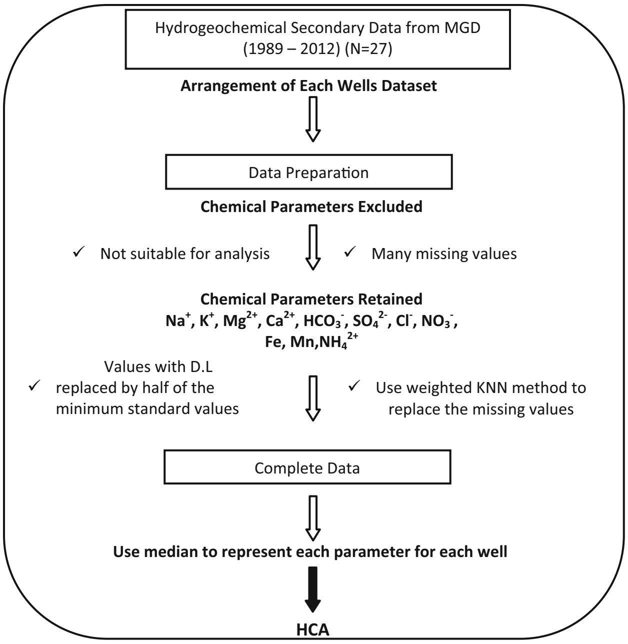


Fig. 7 Methodology flowchart from groundwater wells data analysis

A summary of the methodology used in the preparation of geochemical data for HCA is given in Fig. 7. The original data set consists of 641 samples in total, which involves 27 wells and only 12 chemical variables are considered for HCA. These variables are Na^+, Ca^{2+}, Mg^{2+}, K, HCO_3^-, Cl, SO_4^{2-}, Fe, Mn, NH_4, NO_3 and SiO_2. The data preparation is done in two steps. In the first step, some wells have missing data values; hence, the imputation method was employed. Discarding the data that have missing values may introduce bias or affect the representativeness of the results; this approach also reduces the sample size and results in loss of information. Imputation preserves all the data by replacing missing data with an estimated value based on other available information. Once all missing values have been imputed, the data set can then be analyzed using standard techniques for complete data. There are many different algorithms for imputation; for this study, we applied the weighted k-nearest neighbor (KNN) method (Troyanskaya et al. 2001). The missing value is imputed with the weighted average of the corresponding parameter of the k-nearest neighboring points. The standardized Euclidean distance is used as the metric to estimate the similarity of neighboring points. In the present study, $k = 3$ was used and a complete data set was obtained. Only 22.6 % of samples were involved in the imputation processes. An advantage of the KNN method over other imputation methods is that the replacement values are influenced only by the most similar neighboring points rather than by all points.

In the second step, we need a summary statistic to present a point estimate of each parameter in each well. The median value is chosen as the measure to reduce the effect of outlying observations. Thus, the final data set used for HCA is a data matrix of 27 samples (wells) by 12 chemical variables.

Results and discussion

Hydrogeochemistry/water quality

Table 1 shows the descriptive statistical summary of hydrogeochemical variables analyzed in the study area. All variables are highly dispersed in their distribution. The pH distribution ranging from 5.75 to 7.40 exhibits slightly acidic to slightly alkaline conditions and is within the national drinking water quality standard 5.5–9.5 (MOH 2004). The TDS concentration ranges from 39 to 200 mg/L and is below the 250 mg/L desirable standard (MOH 2004), indicating that the groundwater is suitable for drinking water purposes and is classified as fresh water (Freeze and Cherry 1979) since the concentration is below 1000 mg/L. TDS is influenced by the chemical and biochemical interaction between groundwater and the geologic materials as the water flows through the aquifer layers. Tidal surges from the ocean advance into the nearby rivers (Kelantan, Pengkalan Chepa, Pengkalan Chepa, Pengkalan Datu, Kemasin and Semarak River), especially during the monsoon season where most of the area flooded. The increase in TDS is also a result of human activity at the land surface, including factory waste disposal (Pengkalan Chepa), household rubbish (Kota Bharu), agricultural cultivation sewage effluent and other activities as shown in the land-use map of the area (Fig. 8). Major cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}) and anions (HCO_3^-, Cl^-, SO_4^{2-}) in groundwater show median value ranges (1.60–41.00 mg/L, 1.05–13.00 mg/L, 4.80–26.50 mg/L, 0.90–7.15 mg/L) and (8.00–186.00 mg/L, 3.00–29.50 mg/L, 1.50–34.00 mg/L) below the standard (MOH 2004), respectively. In addition, for the trace elements of Fe^{2+} and Mn^{2+}, the results show median concentrations exceed the desirable limits for drinking water of 0.30 and 0.10 mg/L, respectively. The reason for introducing this limit is to avert taste issues, staining of plumbing fixtures and laundered clothes. Median Fe^{2+}

Table 1 Descriptive statistics of the hydrochemistry of the 27 wells

Parameters	Median	Minimum	Maximum	Standard deviation	Skewness	MOH Standards
pH	6.55	5.75	7.40	0.41	0.02	6.5–9.0
Conductivity	128.00	44.00	335.50	70.17	1.57	NA
TDS	86.00	39.00	200.00	37.95	1.47	1000
Ca^{2+}	5.60	1.60	41.00	7.89	2.93	NA
Mg^{2+}	2.40	1.05	13.00	2.83	2.76	150
Na^{+}	9.60	4.80	26.50	5.72	1.37	200
K^{+}	2.55	0.90	7.15	1.54	1.29	NA
HCO_3^-	34.00	8.00	186.00	42.68	2.80	NA
Cl^-	10.00	3.00	29.50	6.17	1.65	250
SO_4^{2-}	5.00	1.50	34.00	7.04	2.52	250
NO_3^-	2.63	1.50	12.20	2.96	1.38	10
Fe_{total}	2.00	0.10	17.00	4.95	1.92	0
Mn^{2+}	0.10	0.05	0.50	0.11	1.31	0
NH_4^+	0.25	0.06	1.00	0.18	3.06	2
SiO_2	18.29	10.00	43.00	8.36	1.33	NA

Concentration in mg/L, except pH and conductivity (uS/cm), *TDS* total dissolved solids

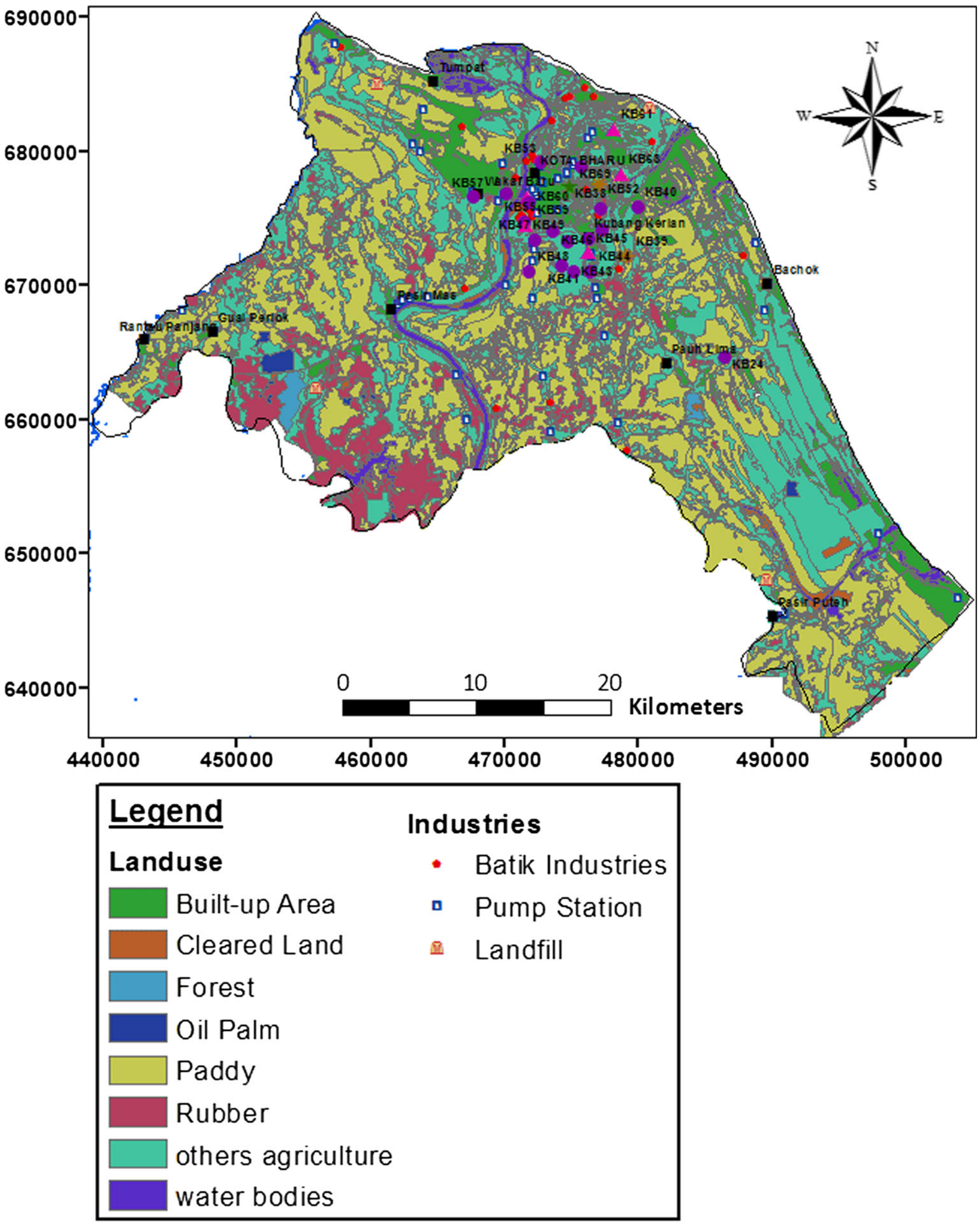
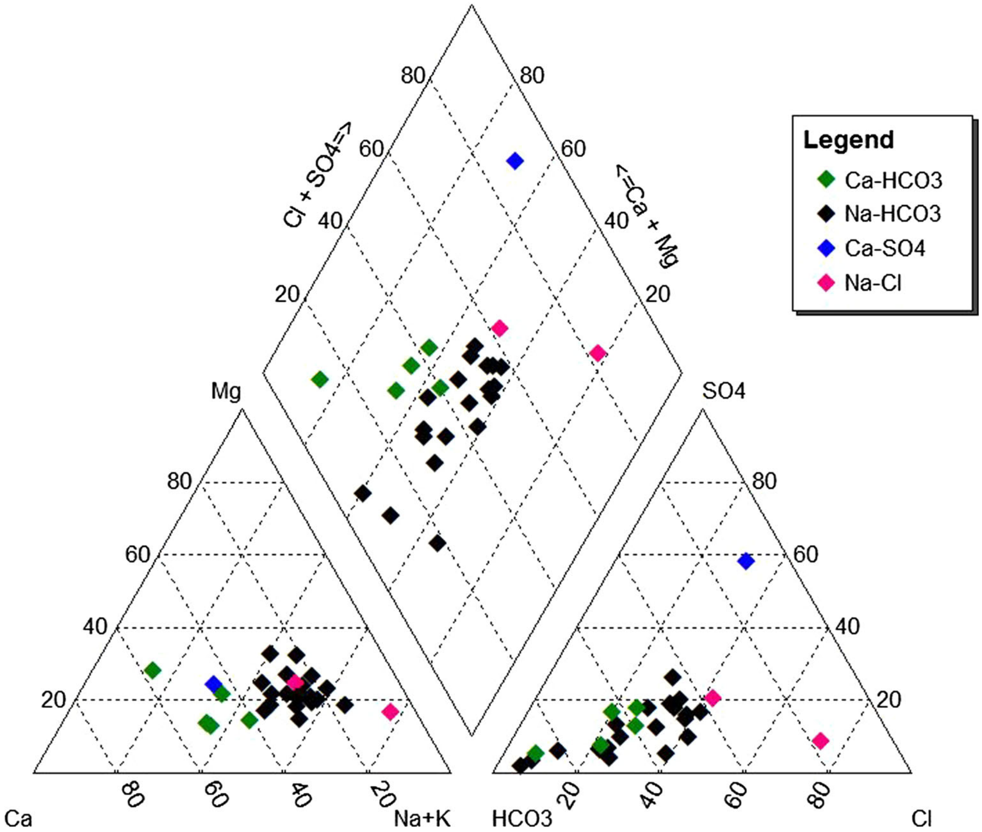


Fig. 8 Land-use map of the Quaternary area in the Lower Kelantan River Basin

ranges from 0.10 to 17.00 mg/L, while Mn^{2+} found in the basin ranges from 0.05 to 0.50 mg/L. Most of the wells have iron concentrations above the MOH limit. This distribution is related to their abundance in the aquifer sediments as well as the geologic condition of the basin. The dissolution of Fe-bearing minerals of pyrite, siderite, magnetite and iron silicate produces mobile ferrous Fe^{2+} in aquifers. Later, Fe^{2+} will oxidize to insoluble ferric iron (Fe^{3+}) and precipitate as ferric hydroxide (reddish/brown color of water) after exposure to the atmosphere (Masoud 2014). The dissolution of Mn minerals (oxides and hydroxides) in sedimentary deposits will oxidize to much less soluble hydrated oxides (Hem 1980) that will form black stains in plumbing fixtures and laundered clothes as well. In groundwater, manganese commonly coexists with iron but in lower concentrations because iron generally has greater crustal abundance (BGS 2003). NH_4^+ and NO_3^- concentrations exceed the desirable limit of drinking water (MOH 2004) at 0.50 and 10.00 mg/L. NH_4^+ and NO_3^- are related to the usage of fertilizer in agriculture and sewage effluent. In general, median concentrations show that groundwater in Quaternary alluvium is suitable for drinking water purposes, but treatment has to be considered for selected variables as discussed above in order to meet the requirement of the national drinking water quality standard (MOH 2004).

Fig. 9 Piper diagram of wells in the Layer 1 aquifer



Groundwater facies (individual wells)

Groundwater facies are interpreted based on major ions in groundwater using Piper diagrams built using Aquachem software version 5.1 (Schlumberger Water Services 2007). The projection of the point from the cation–anion trilinear diagram to the central part of the diamond shape reveals the heterogeneity of the groundwater chemical composition in this study area. Figure 9, the cation triangle, shows clearly that all groundwater is dominated by alkali elements (Na + K) rather than alkali earth element (Ca + Mg). The position of groundwater in the anions triangle indicates that HCO_3 is the dominant ion. Four groups of facies are identified and arranged in a decreasing order of abundance: Na–HCO_3 (74.1 %), Ca–HCO_3 (14.8 %), Na–Cl (7.4 %) and Ca–SO_4 (3.7 %). The general prevalent groundwater flow trend is from Na–HCO_3 to Na–Cl as indicated by the red arrow on the Piper diagram. Figure 10 shows the spatial distribution of the shallow groundwater facies, which is mixed with each other.

Hierarchical cluster analysis (HCA)

Hierarchical clustering is one of the most widely applied classification techniques in earth sciences (Davis 1986) and is often used in the classification of hydrogeochemical data (Steinhorst and Williams 1985; Schot and van der Wal 1992; Riberio and Maacedo 1995; Güler et al. 2002; Cloutier et al. 2008). A dendrogram (Fig. 11) shows the results from the HCA analysis of 27 wells with 12 variables. The Euclidean distance was chosen as the distance measure, or similarity measurement between the well locations. The well locations with the greatest similarities are first grouped. Next, groups of wells are joined with a linkage rule, and the steps are repeated until all wells have been classified. With this hydrochemical data set, Ward's method was more successful to form clusters that are more or less homogeneous and geochemically distinct from other clusters, as compared to other methods such as the weighted pair-group average. Ward's method is distinct from other linkage rules because it uses an analysis of variance approach to evaluate the distances between clusters (StatSoft Inc. 2004). Other studies have also used Ward's method as the linkage rule in their cluster analysis (Adar et al. 1992; Schot and van der Wal 1992; Cloutier et al. 2008). Güler et al. (2002) also found that by using the Euclidean distance as a distance measure and Ward's method as a linkage rule can produce the most distinctive groups.

The classification of the wells into clusters is based on visual observation of the dendrogram. The phenon line was drawn across the dendrogram at a linkage distance of about

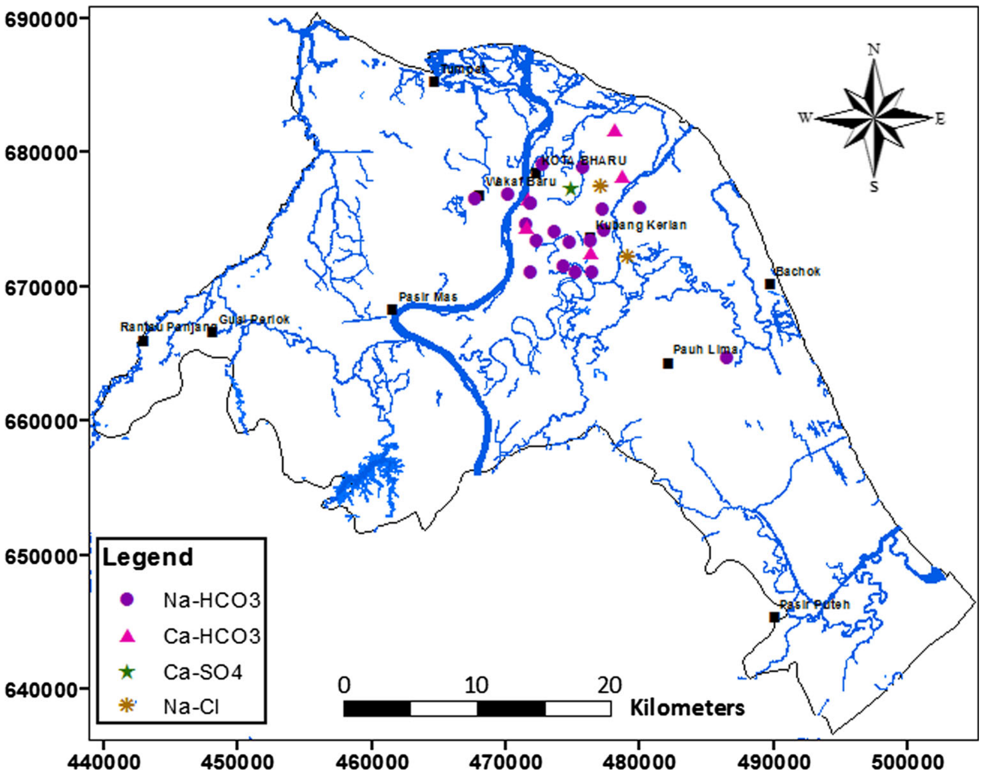


Fig. 10 Spatial distribution of facies in the Layer 1 aquifer

12. Thus, wells with a linkage distance lower than 12 are grouped into the same cluster. Therefore, the phenon line is the benchmark in separating the dendrogram into 4 clusters: C1–C4 (Fig. 11). As mentioned by Güler et al. (2002), fewer or greater numbers of clusters could be defined by moving the position of the phenon line up or down on the dendrogram. This subjective evaluation made HCA a semi-objective method applied for classification. The aims of classification were achieved by having four geochemically distinct clusters in this study area.

Table 2 shows the descriptive statistics of each cluster of groundwater wells using median values of 12 variables and physical data. Later, facies for each cluster are determined using major ions and represented using Stiff diagrams as shown in Fig. 11. An elevated median concentration of minor and trace constituents as indicated in Fig. 11 shows the unique characteristics of the clusters. In general, the four clusters show two main prevalent facies: Na–HCO_3 (C1 and C3) and Ca–HCO_3 (C2 and C4) with slightly distinctive geochemical groups of groundwater wells. C1 groundwater wells are characterized with the lowest concentration of most major ions: Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, HCO_3^{-}, plus with Fe^{2+}, Mn^{2+}, pH, conductivity and TDS, while C3 has elevated concentrations of Na^{+}, Cl^{-}, $SO_4^{2-}$and NO_3^{-} with the lowest concentrations of Mn^{2+}. C2 groundwater wells are indicated by an elevated concentration of Fe^{2+} and NH_4 with lowest Cl^{-}, SO_4^{2-} and NO_3^{-}, while C4 is characterized by elevated concentrations of Ca^{2+}, Mg^{2+}, K^{+}, HCO_3^{-} Mn^{2+} and SiO_2, as well as high pH, TDS and electrical conductivity (EC).

Groundwater clusters and facies group

Figure 12 shows the spatial distribution of groundwater wells according to clusters. The Piper diagram (Fig. 13) presents the groundwater wells labeled according to their respective cluster. An envelope of groundwater wells group facies is drawn on the Piper diagram for comparison. The groundwater wells are scattered among the different facies on the Piper diamond shape and mainly give Na–HCO_3 and Ca–HCO_3 facies, a clear classification of groundwaters in which Na–HCO_3 facies of water quality can be made. Table 3 simplifies the relationship between clusters and facies of groundwater wells. Most of the groundwater wells from C1 to C3 belong to G2 (Na–HCO_3), while C4 represents a mixture of G1 (Ca–HCO_3) and G2 (Na–HCO_3). Therefore, a connection between groundwater well clusters and group facies is apparent as in HCA. Major ions are also considered as the most important ions that control the hydrogeological setting of the basin. The ability of HCA to show the presence of minor and trace elements could help in recognizing the distinct minor element signature related to geological formations.

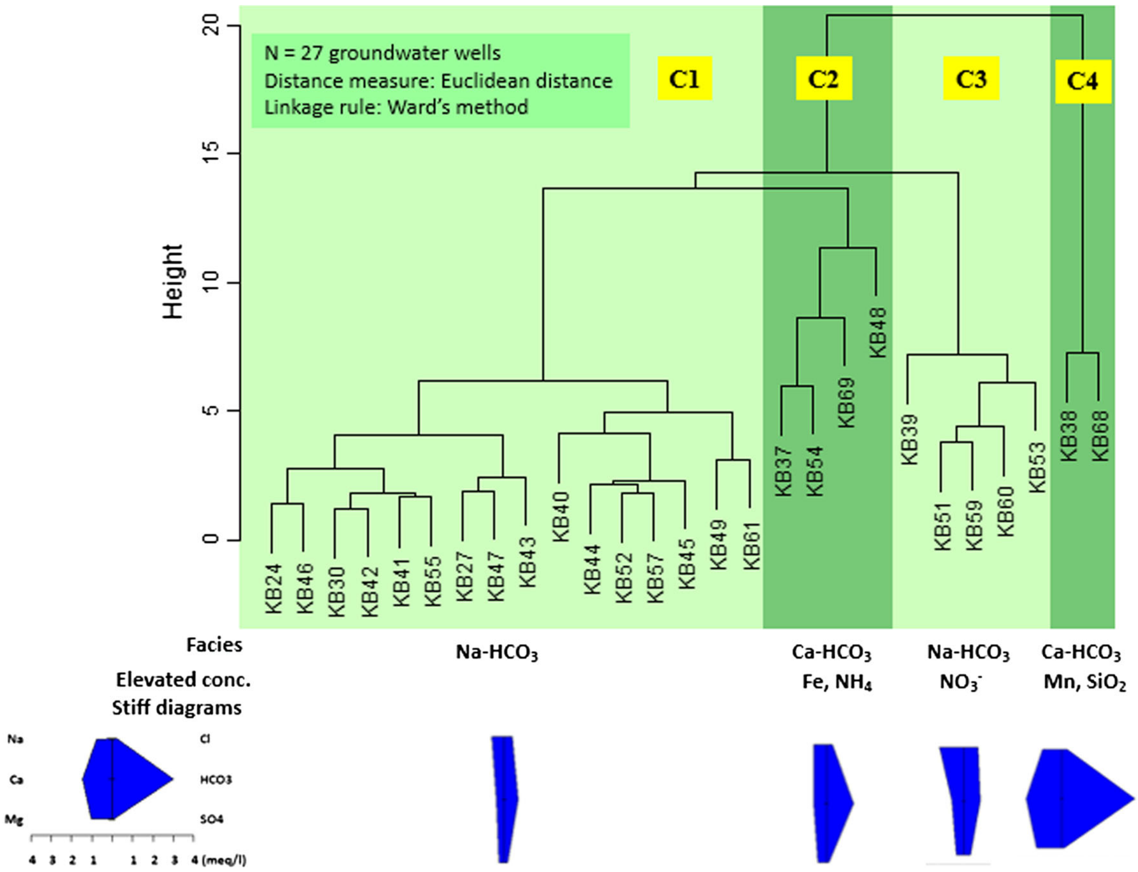


Fig. 11 Dendrogram of the groundwater wells, showing the division into four clusters and the median concentration Stiff diagram of each cluster

Hydrogeochemical evolution

In terms of regional spatial distribution, shallow groundwater is evolving from Ca–HCO_3 to Na–HCO_3 facies. The Ca–HCO_3 facies is characterized as fresh groundwater at or close to recharge areas in the general flow regime. Ca–HCO_3 facies are often associated with carbonate mineral and/or incongruent silicate mineral weathering (Drever 1988; Appelo and Postma 2005). Na–HCO_3 groundwater facies result from the interaction or mixing of groundwaters influenced by two processes of mineral dissolution. Incongruent weathering of albite and related plagioclase feldspars have the potential to result in Na–HCO_3 compositional groundwater facies.

A Gibbs diagram (Gibbs 1970) in Fig. 14 plots the TDS concentration as a function of the weight ratio of Na/(Na + Ca) and Cl/(Cl + HCO_3) to provide information of three major important natural mechanism controlling the hydrogeochemical evolution of groundwater: (1) atmospheric precipitation dominance, (2) rock dominance and (3) evaporation and fractional crystallization dominance. The TDS spreads from low to high with a small variation of concentration. All clusters in Fig. 14 show that rock weathering and precipitation are the dominant mechanisms controlling the hydrogeochemical evolution of shallow groundwater in the study area. Cluster C1 and C3 samples are influenced by the precipitation and freshening, which correspond to recently infiltrated recharge or mixed to intermediate facies (Ghesquiere et al. 2015). Cluster C2 and C4 samples are more influenced by water–rock interaction.

The molar ratio of HCO_3^-/SiO_2 in Table 2 also shows that C1 to C3 clusters have ratios <5, which indicates silicate weathering while C4 is ambiguous. If the ratio is >10 ($HCO_3^- \gg SiO_2$), carbonate weathering is predominant in the area. The saturation indices in Table 2 show that calcite, aragonite and dolomite (SI < 0) are undersaturated, suggesting their absence in the formation and/or not enough time to interact (Wirmvem et al. 2013).

The Na/Cl ratio in Fig. 15a is relatively higher than seawater (0.86) (Hounslow 1995; Millero et al. 2008). The Na/Cl ratio, which is approximately equal to 1, is usually attributed to halite dissolution, whereas >1 is typically interpreted as reflecting Na^+ released from silicate weathering reactions (Fisher and Mullican 1997; Cendon et al. 2010). This ratio suggests that the excess of Na^+ is

Table 2 Geochemical and physical characteristics of each cluster (median concentrations in mg/L)

Parameters	C1	C2	C3	C4
N	16	4	5	2
Ca^{2+}	*4.70*	8.70	9.10	**29.50**
Mg^{2+}	*2.05*	3.35	3.50	**12.50**
Na^{+}	*8.70*	9.85	**21.00**	18.05
K^{+}	*1.80*	2.90	5.05	**5.58**
HCO_3^{-}	*26.50*	55.75	37.00	**183.50**
Cl^{-}	9.00	*7.50*	**18.50**	7.75
SO_4^{2-}	5.00	*3.75*	**11.50**	5.13
NO_3^{-}	2.00	*1.88*	**7.00**	4.50
Fe	*2.00*	**15.25**	0.75	2.53
Mn	*0.10*	0.28	*0.10*	**0.30**
NH_4	0.25	**0.38**	0.25	*0.13*
SiO_2	18.29	23.60	*14.5*	**34.51**
pH	*6.50*	6.63	6.60	**7.35**
Conductivity (µS/cm)	*90.00*	138.75	155.00	**326.25**
Total dissolved solids	*90*	143	136	**219.5**
HCO_3^{-}/SiO_2	*1.37*	1.54	3.02	**5.29**
(Na + K − Cl)/(Na + K − Cl + Ca)	0.37	0.38	**0.40**	*0.30*
Mg/(Ca + Mg)	0.44	*0.38*	**0.44**	**0.44**
CA-I	−0.69	−2.55	*−0.44*	**−3.53**
SI (Anhydrite)	*−4.12*	−4.01	**−3.41**	−3.50
SI (Aragonite)	*−3.05*	−2.43	−2.37	**−0.59**
SI (Calcite)	*−2.90*	−2.28	−2.23	**−0.44**
SI (Chalcedony)	0.10	0.17	*−0.01*	**0.37**
SI (Dolomite)	*−5.74*	−4.55	−4.45	**−0.94**
SI (Goethite)	6.46	7.83	*6.48*	**8.86**
SI (Gypsum)	*−3.89*	−3.77	**−3.17**	−3.26
SI (Halite)	*−8.60*	−8.55	**−7.91**	−8.46
SI (Hematite)	*14.90*	17.65	14.95	**19.70**
SI (Pyrite)	**−86.18**	−87.67	−86.94	*−100.12*
SI (Siderite)	−0.98	0.33	−1.75	**0.40**

N number of samples, Bold: highest values; italics: lowest values

likely from silicate weathering of feldspar or plagioclase and not from the dissolution of halite. Based on the (Na + K+Cl)/(Na + K+Ca + Cl) ratios in Table 2, the source of Na^{+}, K^{+} and Ca^{2+} for all clusters is likely from plagioclase weathering.

The Mg/(Ca + Mg) ratio indicates that sources of Ca^{2+} and Mg^{2+} are mostly from ferromagnesian minerals, with slightly from dolomite and granitic weathering. Table 2 shows that all clusters are supersaturated ($SI > 0$) with respect to hematite and goethite while undersaturated ($SI < 0$) with respect to pyrite and siderite. Under an oxidative environment, the Fe^{2+} is released during dissolution and is precipitated as iron oxide and oxyhydroxides. This indicates that precipitation of iron phases from aquifer layers is thermodynamically favorable. Hematite precipitates more because it is more stable as compared to goethite. Groundwater is undersaturated with respect to major iron phase of pyrite and siderite. Therefore, iron remains dissolved after mobilization. Under an anoxic environment, the pyrite dissolves and is followed by partial oxidation that reflects the increase in Fe^{2+} and SO_4^{2-} in the groundwater.

The relationship between (HCO_3 + SO_4) and (Ca + Mg) in Fig. 15b shows that most of the cluster samples fall along and below the equiline, indicating the influence of silicate weathering. If samples fall above the 1:1 line, they reflect the effect of carbonate and sulfate mineral dissolution (Yidana and Yidana 2010; Yu et al. 2012). Figure 15b indicates that the concentrations of sulfate and bicarbonate are affected by dissolution of silicates minerals.

Thus, the Gibbs diagram as well as the ratios HCO_3/SiO_2, (Na + K+Cl)/(Na + K+Ca + Cl), Mg/(Ca + Mg) and (HCO_3 + SO_4) and (Ca + Mg) strongly indicates incongruent weathering of silicate minerals (water–rock interaction) as the main control on hydrogeochemical

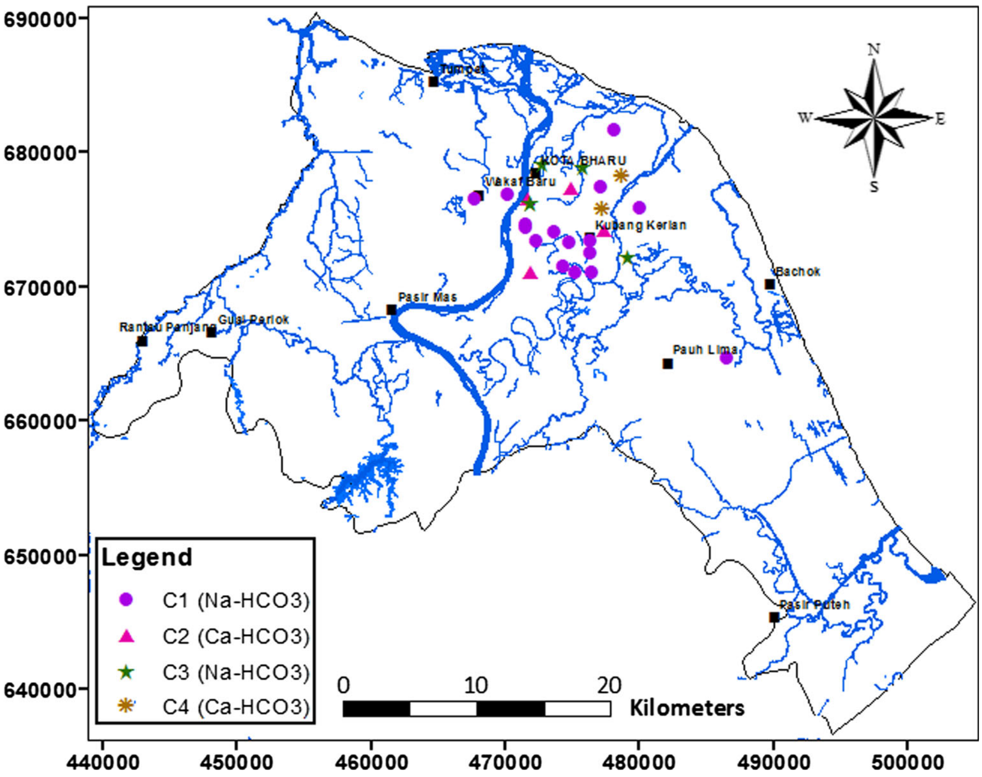


Fig. 12 Spatial distribution of groundwater wells according to clusters

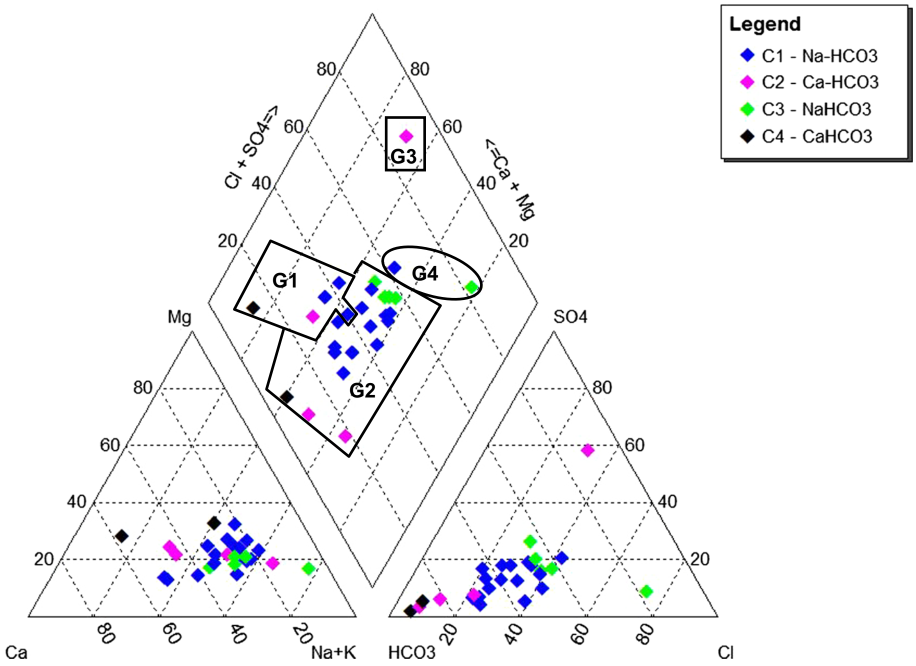


Fig. 13 Piper diagram of wells according to respective clusters in the Layer 1 aquifer. Wells facies are also shown as reference

evolution, in agreement with the geology. Equations 1–5 show the silicate weathering products:

$$\underset{\text{Albite}}{2\mathrm{NaAlSi_3O_8}} + 2\mathrm{CO_2} + 11\mathrm{H_2} \rightarrow \underset{\text{Kaolinite}}{\mathrm{Al_2Si_2O_5(OH)_4}} + 2\mathrm{Na^+} + 2\mathrm{HCO_{3^-}} + 4\mathrm{H_4SiO_4} \quad (1)$$

$$\underset{\text{Albite}}{2\mathrm{NaAlSi_3O_8}} + 2\mathrm{CO_2} + 6\mathrm{H_2O} \rightarrow \underset{\text{Montmorillonite}}{\mathrm{Al_2Si_4O_{10}(OH)_2}} + 2\mathrm{Na^+} + \mathrm{HCO_{3^-}} + 2\mathrm{H_4SiO_4} \quad (2)$$

$$\underset{\text{Anothite}}{\mathrm{CaAl_2Si_2O_8}} + 2\mathrm{CO_2} + 3\mathrm{H_2O} \rightarrow \underset{\text{Kaolinite}}{\mathrm{Al_2Si_2O_5(OH)_4}} + \mathrm{Ca^{2+}} + 2\mathrm{HCO_{3^-}} \quad (3)$$

Table 3 Relationship between well clusters and facies

Clusters	Groups				*N* total
	G1 (Ca–HCO_3)	G2 (Na–HCO_3)	G3 (Ca–SO_4)	G4 (Na–Cl)	
C1	3	12	0	1	16
C2	1	2	1	0	4
C3	0	4	0	1	5
C4	1	1	0	0	2
N total	5	19	1	2	

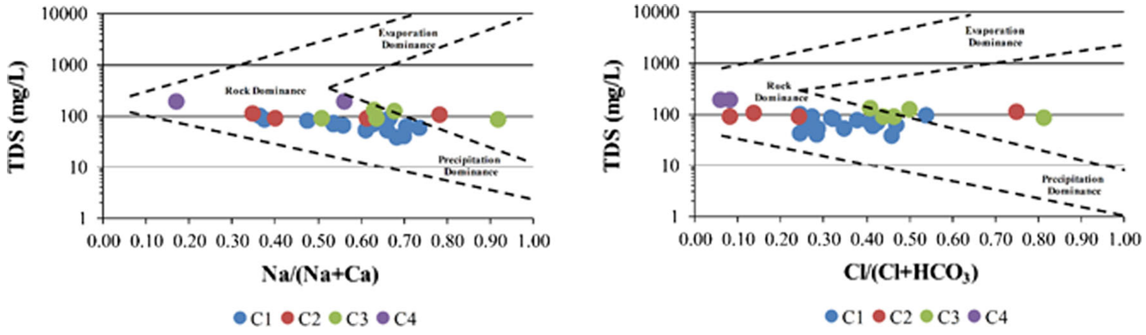
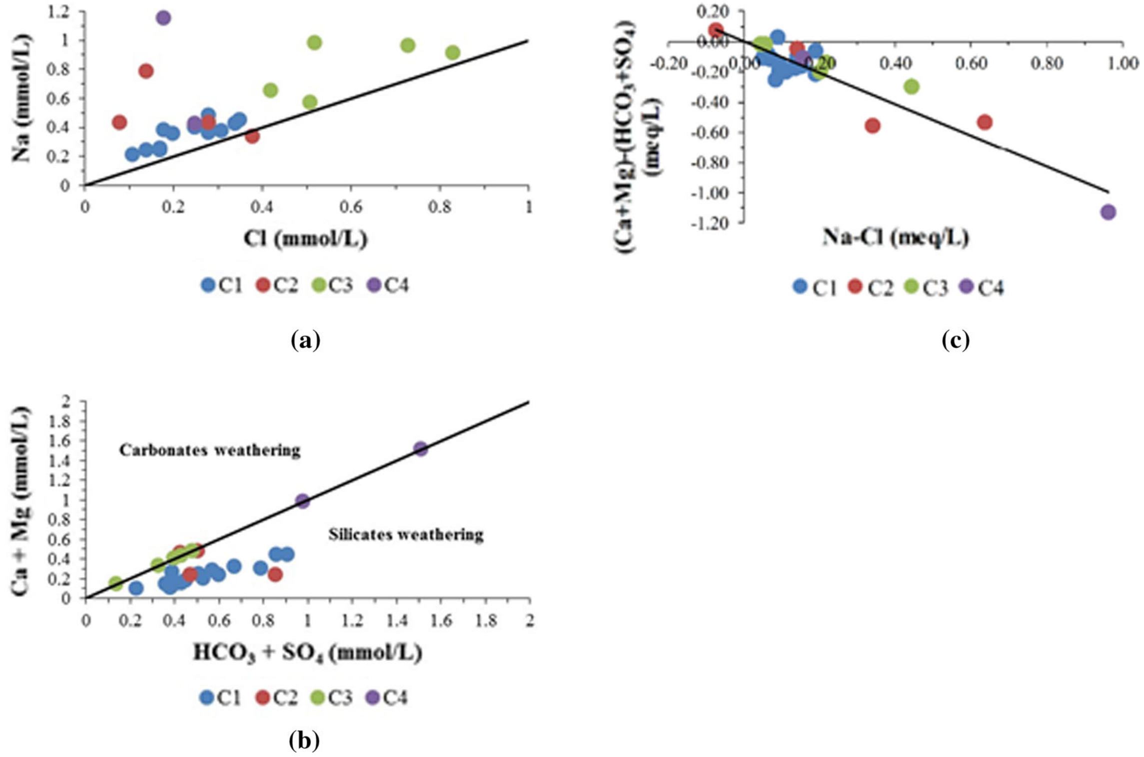


Fig. 14 Plots of TDS versus Na/(Na + Ca) and TDS versus Cl/(Cl + HCO_3)



(a)

(c)

(b)

Fig. 15 Plot of **a** Na versus Cl; **b** Ca + Mg versus HCO_3 + SO_4 and **c** (Ca + Mg)–(HCO_3 + SO_4) versus (Na–Cl)

$$\underset{\text{Biotite}}{2KMg_3AlSi_3O_{10}(OH)_2} + 14CO_2 + 15H_2O \rightarrow \underset{\text{Kaolinite}}{Al_2Si_2O_5(OH)_4} + 2K^+ + 6Mg^{2+} + 14HCO_3^- + 4H_4SiO_4 \tag{4}$$

$$\underset{\text{Biotite}}{2KMg_3AlSi_3O_{10}(OH)_2} + 14CO_2 + 10H_2O \rightarrow \underset{\text{Montmorillonite}}{Al_2Si_4O_{10}(OH)_2} + 2K^+ + 6Mg^{2+} + 14HCO_3^- + 2H_4SiO_4 \tag{5}$$

Ion exchange between the groundwater and its host environment during residence or travel time can be understood by studying the chloro-alkaline indices (Table 2), i.e., CA-I [Cl − Na + K]/Cl, where all ions are expressed in meq/L (Schoeller 1965, 1967; Gupta et al. 2008; Marghade et al. 2012). Na^+ and K^+ ions in water are exchanged with Mg^{2+} and Ca^{2+} ions, if the indices are positive, which indicates a base exchange reaction, whereas negative values indicate chloro-alkaline disequilibrium. The reaction is known as cation exchange. During the process, the host rocks/aquifer materials are the primary sources of dissolved solids in the water. The Schoeller indices of groundwater sample clusters in Table 2 reveal that cation exchange (chloro-alkaline disequilibrium) exists in all clusters. Clay minerals exhibit a preference for ions occupying an exchange site. Kaolinite appears as a dominant clay mineral in the aquifer as studied by Noor (1979). The cation exchange process effectively increases the Na^+ concentrations at the expense of Ca^{2+} and Mg^{2+} as shown in Eqs. 6–8.

$$Ca^{2+} + 2Na - X \rightarrow 2Na^+ + Ca - X \quad (6)$$

$$Ca^{2+} + Mg - X \rightarrow Mg^{2+} + Ca - X \quad (7)$$

Na-X is Na adsorbed onto a clay mineral

$$Mg^{2+} + 2Na - X \rightarrow 2Na^+ + Mg - X \quad (8)$$

The weight ratio of Na/(Na + Ca) in Fig. 14 varies significantly with a small variation of TDS, supporting the conclusion that cation exchange also plays a role by increasing Na and decreasing Ca under the background of rock dominance. During the cation exchange process, the TDS values do not change significantly because 2 mmol/L of Na^+ is released by 1 mmol/L Ca^{2+} exchange, and the weight of 1 mmol/L of Ca^{2+} (40 mg/L) is nearly equal to that of 2 mmol/L of Na^+ (46 mg/L) (Liu et al. 2015).

In addition, a plot of Ca + Mg–HCO_3–SO_4 versus Na–Cl (Fig. 15c) is used to determine the significance of base exchange in enhancing the water chemistry. If cation exchange is the most significant process in the system, the waters should form a line with a slope of −1 (Rajmohan and Elango 2004; Adomako et al. 2011). The diagrams show that all the clusters of groundwater samples give a line with a slope of −1.0287. This confirms that Ca, Mg and Na concentrations are interrelated through cation ion exchange.

Precipitation and river bank infiltration bring recharge into the groundwater system from the inland part of the study area. Infiltration of recharge water into the aquifer layers is very common, especially when there are no impermeable layers of clay overlying the unconfined aquifer. In some parts, the unconfined aquifer may represent a recharge for deeper aquifer systems. As the water recharges, CO_2 rapidly dissolves (Freeze and Cherry 1979) in aquifer layers where CO_2 provides the required acid condition for silicate mineral weathering (Yidana et al. 2012). The dissolution of CO_2 also occurs in soil at partial pressures larger than the atmospheric value, which is primarily caused by root and microbial respiration (Domenico and Schwartz 1990) as shown in Eqs. 9–11. This process will increase the HCO_3^- in groundwater. The main contributor of HCO_3^- is from the hydrolysis of silicate weathering (Eqs. 1–5) as shown in Fig. 15b as an active process in the groundwater flow system.

$$CO_2(g) + H_2O \rightarrow H_2CO_3 \quad (9)$$

$$H_2CO_3 \rightarrow HCO_{3-} + H^+ \quad (10)$$

$$HCO_{3-} \rightarrow CO_3^{2-} + H^+ \quad (11)$$

Conclusions

Analysis of hydrogeochemical data using the HCA multivariate statistical technique can aid in understanding the hydrogeological processes that govern the evolution of shallow groundwater at the regional scale. Based on this study, shallow groundwater is suitable for drinking water purposes with some consideration of treatment needed for selected ions—mostly iron. HCA indicates that the two main prevalent facies that reflect the evolution of groundwater are: Ca–HCO_3 and Na–HCO_3. The main processes that govern the hydrogeochemical evolution are precipitation, weathering, dissolution and ion exchange as discussed above where silicate weathering is the main contributor. In this respect, further studies are proposed in order to enhance the understanding of the process involved in the Lower Kelantan River Basin, which will also be beneficial for the future sustainable development of groundwater resources.

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