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Extent and severity of groundwater contamination based on hydrochemistry mechanism of sandy tropical coastal aquifer

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HIGHLIGHTS

- ► The hydrogeological interactions between static and dynamic components determine the groundwater quantity and quality.
- ► Major cations in groundwater were derived from the chemical reaction of the deposited carbonate.
- ► Dissolution of carbonate, weathering and cation exchange processes explain the changes in groundwater compositions.
- ► Cation exchange mechanism is the key factor that modifies and controls the concentration of major cations in groundwater.

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ABSTRACT

Small islands are susceptible to anthropogenic and natural activities, especially in respect of their freshwater supply. The freshwater supply in small islands may be threatened by the encroachment of seawater into freshwater aquifers, usually caused by over pumping. This study focused on the hydrochemistry of the Kapas Island aquifer, which controls the groundwater composition. Groundwater samples were taken from six constructed boreholes for the analysis and measurement of its *in-situ* and major ions. The experimental results show a positive and significant correlation between Na–Cl ($r = 0.907$; $p < 0.01$), which can be defined as the effect of salinization. The mechanisms involved in groundwater chemistry changes were ion exchange and mineralization. These processes can be demonstrated using Piper's diagram in which the water type has shifted into a Na–HCO_3 water type from a Ca–HCO_3 water type. Saturation indices have been calculated in order to determine the saturation condition related to dissolution or the precipitation state of the aquifer bedrock. About 76% of collected data ($n = 108$) were found to be in the dissolution process of carbonate minerals. Moreover, the correlation between total CEC and Ca shows a positive and strong relationship ($r = 0.995$; $p < 0.01$). This indicates that the major mineral component in Kapas Island is Ca ion, which contributes to the groundwater chemical composition. The output of this research explains the chemical mechanism attributed to the groundwater condition of the Kapas Island aquifer.

1. Introduction

Groundwater is of global concern and represents a vital environmental component to island communities' consumption to maintain their lives and is important for drinking water purposes as well as domestic use (Russak and Sivan, 2010). In this case, groundwater storage in small tropical islands is limited and is the only source for supplying water for domestic usage. Usually, the shallow groundwater is recharged by infiltration from precipitation and surface runoff (Amer, 2008; Saxena et al., 2008; Aris et al., 2007). Kapas Island consists of several ephemeral rivers (Abdullah, 1981) that only exist in an exploitable form during heavy rain in the monsoon, which occurs between November and January every year. This has made Kapas Island depend entirely on groundwater resources for its freshwater supply. Kapas Island local communities and tourists use the groundwater for drinking water, agricultural water and domestic use. Such large demands on groundwater in developing tourism areas threaten the quality of groundwater. Since Kapas Island receives considerable attention for ecotourism activities, there is an associated high demand for freshwater. In order to meet the requirement for the freshwater demand, over extraction of groundwater is widely practiced, which further affects the freshness of groundwater (Fleeger, 1999; Aris et al., 2010).

Over abstraction of groundwater may cause the lens of the groundwater aquifer to shrink and enable the vertical and lateral intrusions of seawater (Petalas et al., 2009) into the aquifer and subsequent mixing with freshwater. One of the major phenomena that threaten coastal aquifers is salinization due to seawater intrusion (Petalas et al., 2009; Russak and Sivan, 2010; Fleeger, 1999; Werner et al., 2009; Aris et al., 2009; Praveena and Aris, 2010; Aris et al., 2012).

Groundwater salinization results from the physical and chemical processes that significantly increase the salt concentration in the

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groundwater, as the groundwater occurs in different geological, land-use and climate settings (Salama et al., 1999). Salinization of groundwater is known to have a high concentration of chloride, which is associated with the dissolved solids and conductivity values in the groundwater. Precipitation of minerals through continual evaporation or by evapotranspiration where the infiltration recharge water is taken up by plants leaves solids behind known as salty residue, or, technically, crystalline salt. Hydrolysis is another way for salt accumulation since water is taken in the formation of new minerals in the weathering process or by leakage, which explained the aquifer through confining beds (Salama et al., 1999). High evaporation rates and limited recharge water may be attributed to the groundwater salinization, and, therefore, longer groundwater retention time and more extensive water–rock interaction (Nativ et al., 1997). Salinization is a long-term phenomenon that has become pervasive and renders the fresh groundwater quality unsuitable for human consumption.

The mixing process in the groundwater aquifer might change the constituents and the hydrochemistry of fresh groundwater. The factors that are mostly responsible for the changes of fresh groundwater composition are cation exchange, adsorption of dissolved ions, dissolution of aquifer matrix and the geological formation of the groundwater aquifer. In addition, climate, as represented by precipitation (recharge), has been identified as a critical factor influencing weathering rates that, together, subsequently, determine the carbonate chemistry behavior and hydrochemistry characteristics in the aquifer (Tijani, 1994; Gabet et al., 2010; Russak and Sivan, 2010).

The information on groundwater in small islands, especially concerning groundwater usage or groundwater assessment, is generally lacking or limited (Tijani, 1994). Research on small islands, especially in tropical regions is still in a developing state and the research gaps, particularly those concerning the key issues of small tropical islands, are still big. For example, previous and ongoing research has focused on hydrochemical analysis of groundwater by Aris et al. (2008a) and Abdullah et al. (2008), groundwater management by Praveena et al. (2010a), groundwater quality (Lin et al., 2010), groundwater modeling by Praveena and Aris (2010) and Aris et al. (2010, 2012), and an ionic ratio measurement in groundwater (Isa and Aris, 2012). However, worldwide research on groundwater has been undertaken for more than a century to collect data for different groundwater conditions depending on the lithology and texture of the original aquifer and to understand the physical and chemical behavior of groundwater. For example, groundwater pollution by Oakes et al. (1981), groundwater salinization (Brown et al., 2006; Gaye, 2001; Panda et al., 2007) and up-coning of brines due to over-pumping activities (Rosenthal, 1988). A number of groundwater studies have been conducted in tropical islands; however, they have not been extensively reviewed. Generally, the objectives of those studies conducted mainly involved groundwater resources and seawater intrusion phenomenon. This suggests that groundwater studies in Malaysia are still in their infancy and clearly have a long way to go.

Therefore, to protect the ecological balance of islands, the mixing mechanisms of fresh groundwater and seawater attributed to seawater intrusion as well as its balance need to be identified and investigated so that the responsible bodies of this vital reserve can be well managed. Hence, this study is different from other studies mentioned above as it investigates the hydrogeological and hydrochemical aspects of the groundwater chemistry in Kapas Island, specifically, to identify the salinization or freshening status of groundwater as well as the mechanism controlling its chemistry. This combined approach will provide an overall viewpoint for groundwater studies in respect of small tropical islands since it will give a clear picture of environmental factors affecting the hydrochemistry changes. Concisely, groundwater studies in Malaysia have been limited to academic and research purposes (Praveena et al., 2010b). To provide a direction for future studies involving groundwater of tropical islands in Malaysia, further detailed and site-specific information on soils, aquifer sediment and local hydrogeological conditions is clearly needed. Hence, this study provides and utilizes all information from dynamic components (hydrogeological properties; physical and chemical) and static components (geological information, soil physical and chemical properties). The key issue is to increase the groundwater studies and understanding so that long-term groundwater research and sustainable performance in respect of the environment can be addressed at the outset. This is a crucial step to fill the knowledge gap concerning groundwater studies in Malaysia involving tropical islands.

1.1. Site descriptions

Kapas Island was chosen in this particular study since there is insufficient information concerning the hydrogeochemical condition due to the impact from hectic development, as Kapas Island has become one of the major tourism attractions in Malaysia. Kapas Island is situated approximately 3 km offshore, east of the Marang jetty, Terrengganu, Malaysia. Located between 5° 13.140′ N and 103° 15.894′ E (Fig. 1) with an area of about 2 km^2 (Abdullah, 1981; Shuib, 2003), about 90% of the area is covered by hilly area and the rest represents the coastal area, which has been highly developed for ecotourism activities. The aquifer formation in Kapas Island is made from deposits of carbonate shells that are formed from calcite, aragonite and dolomite minerals.

Kapas Island is underlain by a Permo-Carboniferous metasediment formation and unconformable overlying conglomerate. Kapas Island is also intruded by dolerite dykes, which were further subjected to a final southeast–northwest compression deformation (Shuib, 2003). Metasediment rocks basically represent the sandstone, mudstone, shale and silt while the conglomerate groups were followed up with sandstone and mudstone (Ali et al., 2001). There are several age groups of rocks found in Kapas Island based on the similarity of the metasediments in the Terengganu area, and it can be concluded that the metasediments in Kapas Island could be Permo-Carboniferous (Shuib, 2003). Kapas conglomerate is underlain with other formations based on the previous study by Shuib (2003), and, thus, it is suggested that the Kapas conglomerate formation could be from the Late Permian to Triassic age or still be of Jurassic–Cretaceous age (Shuib, 2003). The location of soil profiling of the study area is shown in Fig. 1 (mark as Control) while the details of the soil profile are shown in Fig. 2 (Abdullah, 1981).

Kapas Island receives an annual rainfall between 1500 and 2800 mm, which is influenced by the monsoon that blows from the middle of November to January. Kapas Island experiences a constant temperature varying from 28 °C to 31 °C and has a warm and humid climate of around 70–80% annually, as it is situated in the tropical region. The groundwater recharge in the Kapas Island aquifer depends entirely on precipitation. Fig. 3 shows the hydrogeological conceptual model for Kapas Island used to understand the hydrogeological concept in the island. The hydrogeological model developed consists of static and dynamic components. The static component symbolizes the aquifer matrix or soil and the dynamic component represents the groundwater as the only water reservoir in the island since no other surface water exists in exploitable form, including evapotranspiration and precipitation. The main input of the conceptual model starts from precipitation, which infiltrates into the groundwater as groundwater recharge, discharge of groundwater from the constructed boreholes, evapotranspiration process from the surface level and to complete the model the water returns as precipitation. These elements/components are shown to be related by exact conceptual, depending on the situation in the present day (Toth, 1970) that controlled the groundwater environment. Another factor that contributes to the process of the conceptual model is climate, comprising the temperature, precipitation and winds while the geology factor is made up of the distribution of different parent rocks and the groundwater. Therefore, the physical and chemical conditions resulting from the combination of hydrogeology and climate may be said to contribute to the hydrogeological groundwater composition. It is possible

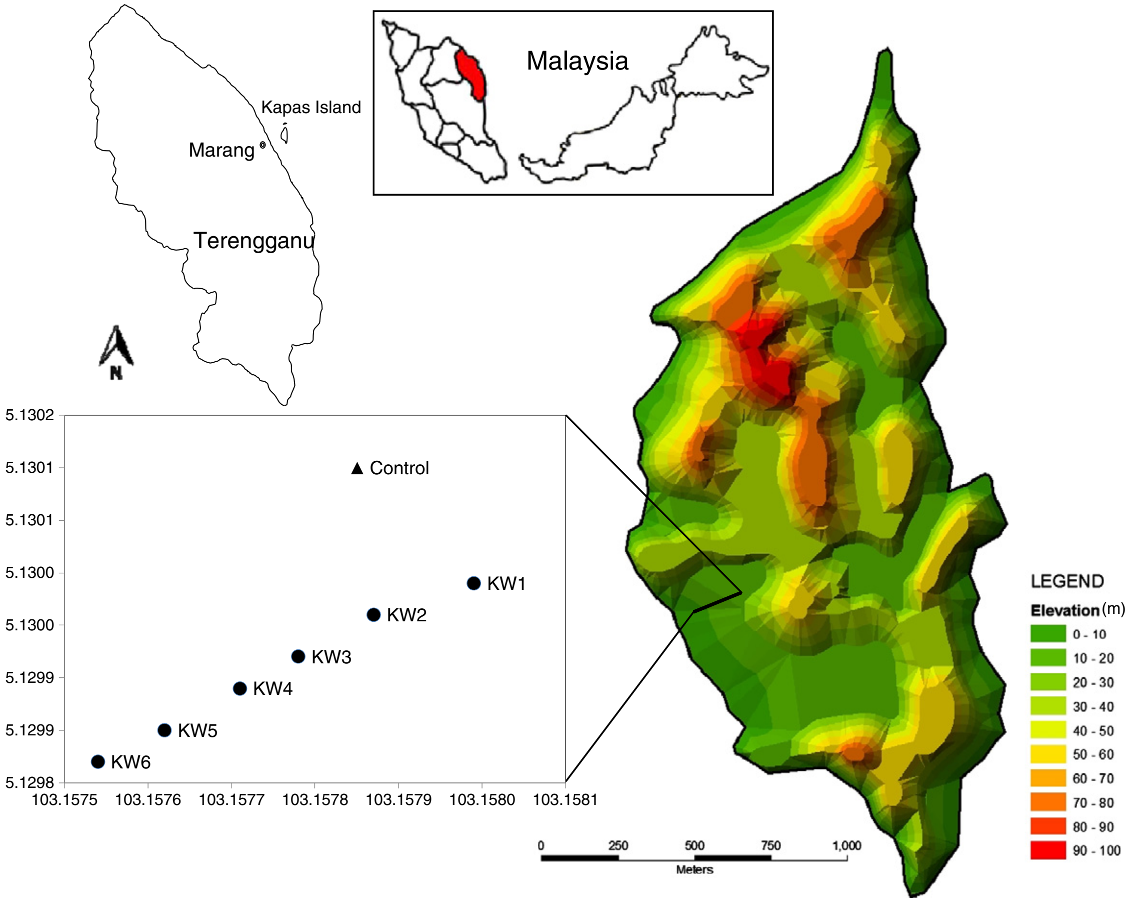

Fig. 1. Schematic map showing the geographical locality with the elevation map of Kapas Island and the location of the boreholes. The control is a soil profiling borehole adapted from Abdullah (1981). The distance from the coastline and the depth for each borehole is tabulated in Table 1.

to describe and analyze the groundwater conditions in the context of the hydrogeological observations of the conceptual model. The hydrogeological conceptual model developed is very important to understand the interaction between the static and dynamic components to identify the mechanisms involved that control the groundwater composition. The use of such a conceptual model for the precise hydrogeological of the sampling area in this study appears to be a prerequisite for groundwater development and management, especially in small tropical islands, as it answers several key issues, which are salinization, mineralization and the chemical processes. The descriptions of the hydrogeological and hydrochemical properties are shown in Table 2.

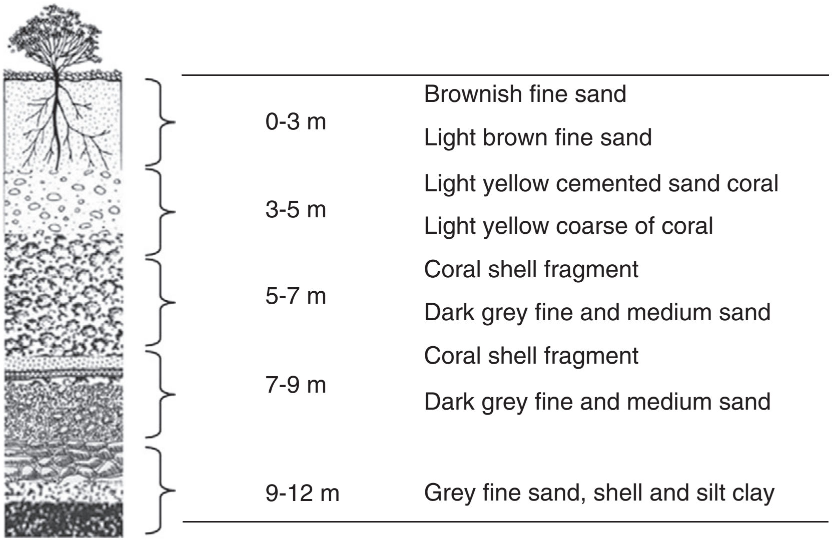

Fig. 2. Soil profile in the study area.
Modified from Abdullah (1981).

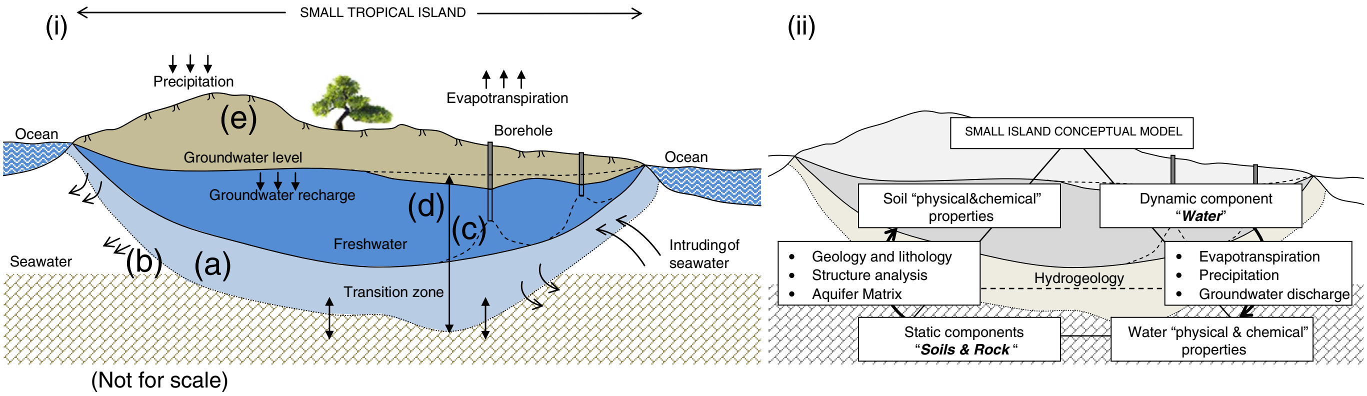

Fig. 3. (i) A simple hydrological conceptual model for Kapas Island aquifer and (ii) the hydrological conceptual model approach used in this study, modified from Winkler et al. (2003). Showing (a) seawater wedge, (b) density-driven circulation-force seawater zone underneath groundwater, (c) seawater upconing due to borehole pumping, (d) average aquifer thickness and (e) ground surface.

2. Methods

From August to October 2010, a total of 108 samples (3 replicates of 36 samples) were collected bimonthly (two sampling campaigns in a month) for physical and chemical analyses. The groundwater and sediment samples were obtained from KW 1, KW 2, KW 3, KW 4, KW 5 and KW 6. The boreholes were constructed (Fig. 1) over a distance of 120 m from the coastline (Table 1). The wells were screened at different elevations between 2.5 and 11.5 m from ground surface level.

The groundwater levels in the Kapas Island aquifer are shown in Table 3 and Fig. 4. The groundwater flows are in the direction from northeast to southwest (toward sea). In general, the water table configuration is a replica of the topography of the area as it increases with increasing elevation. Groundwater flows usually respond to a groundwater recharge event (rainfall). The water enters the rock fractures and flows under the influence of gravity towards low adjacent areas where the groundwater discharges. It is important to identify the groundwater flow-path, which has a significant effect on the groundwater chemistry.

2.1. Sediment samples

The sediment samples in Kapas Island were determined for their exchangeable cations (CEC) using 1 M NaCl and 1 M NH_4Cl extractions (Appelo et al., 1998; Aris et al., 2010). The calculation of CEC is adapted from Radojevic and Bashkin (2006). The sediment samples were divided into two sets: Set A and Set B for determination of exchangeable cations. About 10 g of sediment from each borehole in each set was percolated with 10 ml of 95% ethanol and was allowed to evaporate overnight before further treatment using NaCl and NH_4Cl. The initial percolation with ethanol was to remove solute cations to avoid pseudo cation contribution in the sediments. About 30 ml of 1 M NaCl was added to Set A and 30 ml of 1 M NH_4Cl was added to Set B in 50 ml centrifuge tubes. The centrifuge tubes containing the pre-treated sediments; NaCl (Set A) and NH_4Cl (Set B) were shaken end-over-end for about 20 min and then centrifuged at 3000 rpm for 45 min in order to settle the fines. The supernatant was filtered through a 0.45 μm filter paper (Whatman Milipores, Clifton, NJ, USA) after being centrifuged.

Table 1
Coordinate and features of each sampling station.

Boreholes	Station's coordinate		Distance from coastline (m)	Depth of borehole (m)*
KW 1	05° 12.999 N	103° 15.799 E	119	11.5
KW 2	05° 12.996 N	103° 15.787 E	98	9.1
KW 3	05° 12.992 N	103° 15.778 E	83	3.5
KW 4	05° 12.989 N	103° 15.771 E	68	3.0
KW 5	05° 12.985 N	103° 15.762 E	48	2.9
KW 6	05° 12.982 N	103° 15.754 E	31	2.5

* From ground surface level.

A 15 ml filtered sample from Set A was used for the analyses of Ca, Mg and K (pre treated with NaCl) while Na was analyzed from the Set B samples (pre-treated with NH_4Cl). The Ca, Mg, K and Na in the treated samples were analyzed using a flame atomic absorption spectrophotometer (FAAS, Perkin Elmer, Massachusetts, USA) with triplicate measurements. Low CEC values indicated sandy textured soil while higher CEC values indicated more loam and clay soil characteristics, as shown in Table 4 (Andrews et al., 2004).

2.2. Groundwater samples

Standard procedures for groundwater sample collection were followed. Groundwater samples were pumped out for some time (10–15 min) using a set of purging pump prior to groundwater sampling, to evade non-representative samples of stagnant or polluted water (Petalas et al., 2009; Appelo and Postma, 2005; Aris et al., 2010). It is necessary to empty the borehole or pump out several

Table 2
The description of the hydrogeological and hydrochemical properties in Kapas Island based on the conceptual model.

Hydrology	Mean
Temperature	30.12 °C
Annual precipitation	2274.66 mm/year
Annual evapotranspiration	50% of annual precipitation
Groundwater recharge rate	467 m^3/day
Groundwater discharge rate	1.6 m^3/h
Transmissivity	2.62 m^2/h
Permeability	1.4×10^{-4} m/s
Average aquifer thickness	5 m
Natural groundwater run-off	7.5 m^3/h
Hydraulic gradient	2.9×10^{-3}
Geology	
Highlands	
Metasediment rocks	85%
Lowlands	
Alluvium sands	15%
Water type	
Year 1981	Ca–HCO_3 (Abdullah, 1981)
Year 2010	Ca–HCO_3 and Na–HCO_3

Table 3
Mean water table value for each sampling station in the study area.

Station	Water table (m)						
	Aug 1	Aug 2	Sept 1	Sept 2	Oct 1	Oct 2	Mean
KW 1	2.77	2.97	2.97	2.94	3.03	2.79	2.91
KW 2	2.69	2.88	2.84	2.82	2.89	2.73	2.80
KW 3	2.40	2.89	2.86	2.85	2.88	2.87	2.79
KW 4	2.71	2.85	2.83	2.82	2.82	2.83	2.81
KW 5	2.81	2.97	2.94	2.94	2.88	2.87	2.90
KW 6	2.49	2.59	2.52	2.52	2.42	2.43	2.50

Numbers 1 and 2 represent two sampling campaigns in a month.

Table 4
Interpretation of CEC values to soil texture.

CEC range	Texture	Characteristics
Less than 10 meq/100 g	Sand	Low organic matter and low moisture
10 to 15	Sandy loam	More desirable, higher clay content and well structured
15 to 20	Loam	Higher organic matter content and moisture, well structured
20 to 25	Clay loam	Higher clay content and tight structure
25 above	Clay	Tight soil structure due to high clay content, poor internal drainage.

times to avoid the above problems and collect a representative groundwater sample. The following parameters were measured or analyzed in the field with triplicate measurements in order to acquire representative values of ambient conditions: *in-situ* parameters (measured using Mettler Toledo, Columbus), such as temperature (°C), pH, electrical conductivity (EC; mS/cm), total dissolved solids (TDS; mg/L), oxidation–reduction potential (*Eh*; mV), dissolved oxygen (DO; mg/L) and major anions — bicarbonate (HCO_3), chloride (Cl) and sulfate (SO_4). A pH meter was calibrated to pH 4 and pH 7 using NIST Traceable Buffer prior to measurement, and other instruments were calibrated using specific calibrating solutions. A mean value was calculated for each parameter measured and standard deviation (SD) was used as a precision indicator of each parameter measured for the triplicate samples. Titration methods were performed with 0.02 N HCl for HCO_3 and 0.0141 N $AgNO_3$ for Cl on 50 ml of sample for each parameter measured (APHA, 2005).

SO_4 concentrations in the groundwater were determined using a HACH (DR/2000) meter (HACH, Loveland, CO, USA) on 25 ml of samples (APHA, 2005). Polyethylene bottles soaked in 5% nitric acid overnight and pre-rinsed with deionised water were used to store groundwater samples (APHA, 2005) that needed to be transported to the laboratory for analysis of the major cations (Ca, Mg, Na and K) in an iced cooler box. Groundwater samples were filtered using 0.45 μm filter paper (Whatman Milipores, Clifton, NJ, USA) and acidified with concentrated HNO_3 acid (APHA, 2005; Gabet et al., 2010) to maintain the pH below 2. The acidification process is to avoid bacterial development, block the oxidation effects and prevent the adsorption or precipitation of cations (Appelo and Postma, 2005). The pre-treated groundwater samples were analyzed using a flame atomic absorption spectrophotometer (FAAS, Perkin Elmer, Massachusetts, USA) for major cation constituents. The preservation and data collection of samples were done precisely as suggested worldwide to ensure the quality of data. Careful quality controls and accuracy checks were undertaken to obtain a reliable analytical dataset by checking the blank samples and a five-point calibration curve was constructed for each element. The results of triplicate analyses indicate the precision of the cation measurement. The annual preventive maintenance for the FAAS equipment was done to receive accreditation of the systems and approval of the laboratories by governmental bodies.

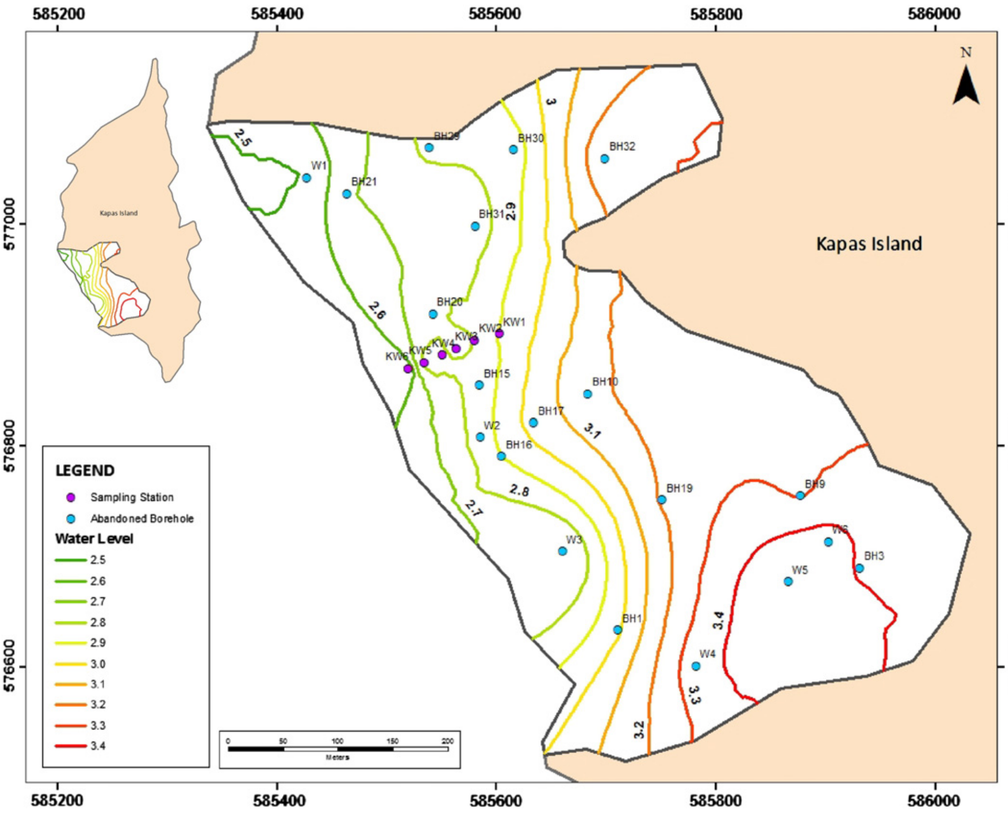

Fig. 4. Water table map for studied area. Stations KW 1 to KW 6 are the studied station. Point "BH" and "W" represent old and abandoned stations from previous report.

Table 5
Descriptive analysis for in-situ parameters and major ions (n = 108).

Boreholes	KW 1	KW 2	KW 3	KW 4	KW 5	KW 6
Temp °C						
Mean	29.1	29.9	30.1	31.0	31.4	31.0
SD	0.7	0.8	0.5	1.2	1.8	1.1
Min	27.8	28.9	29.3	29.9	29.4	29.9
Max	30.2	31.7	31.0	33.7	35.1	32.7
pH						
Mean	7.2	6.9	7.2	7.3	7.2	7.0
SD	0.1	0.1	0.1	0.1	0.0	0.2
Min	7.1	6.8	7.1	7.2	7.1	6.7
Max	7.6	7.1	7.3	7.3	7.2	7.2
Cond (mS/cm)						
Mean	0.56	0.72	0.43	0.42	0.55	0.71
SD	0.02	0.12	0.01	0.01	0.07	0.17
Min	0.54	0.58	0.42	0.41	0.43	0.43
Max	0.62	0.91	0.44	0.44	0.63	0.85
Sal (ppt)						
Mean	0.27	0.35	0.21	0.20	0.27	0.35
SD	0.01	0.06	0.01	0.00	0.03	0.09
Min	0.26	0.28	0.20	0.20	0.20	0.21
Max	0.30	0.45	0.21	0.21	0.30	0.41
DO (mg/L)						
Mean	2.5	2.5	3.2	4.0	3.3	2.4
SD	2.4	2.7	2.3	2.6	0.9	0.8
Min	0.7	0.6	1.4	2.1	2.1	1.4
Max	6.9	7.9	7.1	8.5	4.5	3.8
TDS (mg/L)						
Mean	279.4	358.9	213.3	208.4	276.2	354.9
SD	10.7	59.9	3.7	5.2	33.0	87.0
Min	272.0	292.0	209.0	204.0	212.0	216.0
Max	311.0	455.0	221.0	220.0	314.0	422.0
Eh (mV)						
Mean	1.27	15.34	−3.19	−5.02	1.78	14.01
SD	2.06	7.36	2.38	2.07	4.03	10.60
Min	−4.30	8.50	−6.20	−7.50	−5.60	−3.30
Max	4.30	27.00	0.80	−1.50	7.50	32.30
Ca (mg/L)						
Mean	56.65	53.21	32.22	36.12	50.00	44.09
SD	25.84	31.82	17.02	17.28	31.89	27.01
Min	24.01	10.40	11.40	14.30	18.50	17.80
Max	103.90	118.90	62.20	75.30	145.20	106.70
Mg(mg/L)						
Mean	7.25	7.05	4.08	4.02	3.62	4.72
SD	2.71	3.17	1.46	0.88	1.06	2.11
Min	3.19	2.19	1.62	2.81	1.96	1.98
Max	10.10	10.40	5.70	5.20	5.40	7.60
Na (mg/L)						
Mean	13.77	17.67	9.59	10.47	25.26	37.84
SD	5.06	5.34	4.22	6.53	12.31	20.90
Min	5.80	10.40	0.80	2.96	7.20	7.30
Max	20.20	25.20	13.36	24.48	42.65	58.80
K (mg/L)						
Mean	0.85	2.43	0.62	0.57	0.84	1.45
SD	0.09	0.93	0.16	0.53	0.41	0.85
Min	0.72	1.01	0.38	0.03	0.30	0.65
Max	1.00	3.89	0.88	1.48	1.51	2.95
HCO_3 (mg/L)						
Mean	344.65	427.00	281.28	313.40	467.33	441.50
SD	12.17	77.80	17.67	17.38	205.72	116.56
Min	322.08	323.30	254.98	284.26	241.56	246.44
Max	366.00	578.28	324.52	341.60	893.04	639.28
Cl (mg/L)						
Mean	36.10	41.29	25.21	22.10	48.15	89.30
SD	6.08	6.91	5.03	6.46	15.48	48.71
Min	27.99	32.49	17.99	11.49	19.99	17.99
Max	45.99	55.98	32.99	32.99	78.98	141.96
SO_4 (mg/L)						
Mean	19.33	7.28	11.50	10.33	21.61	16.11
SD	0.84	8.90	1.34	1.19	6.98	4.04
Min	18.00	1.00	9.00	8.00	10.00	9.00
Max	20.00	26.00	13.00	12.00	30.00	20.00

2.3. Data analyses

Descriptive statistics and correlation coefficient analyses were performed for the groundwater samples using Statistical Package for Social Science (SPSS 17.0). The inferential statistics can make judgments of the probability that the difference between groups is a dependable one or independent of each other. The data obtained from this study were graphically presented using a Piper diagram in order to show the mechanisms involved in the groundwater chemistry evolution. The origin and chemical behavior of the groundwater were assessed based on the ionic relationships. These comprise the relationships of Cl/HCO_3 vs. Cl, Na/Cl and (SO_4+HCO_3) vs. $(Ca+Mg)$. The calculation of CEC and the correlations between major cations were studied to clarify which elements contribute to the groundwater composition.

In addition, the saturation states for the samples collected with respect to selected minerals were calculated using PHREEQC software version 2.17. The saturation state of the water samples is defined by the saturation index, (SI) (Appelo and Postma, 2005) and was calculated based on the activities of free ions in solution. Some minerals react fast upon contact with water, mostly minerals, such as gypsum, halite and carbonate, which would meet the equilibrium in a short residence time. The temperature was only considered for certain minerals, typically, silicates; the equilibrium of which is never attained at a low temperature (Appelo and Postma, 2005; Merkel and Planer-Friedrich, 2008). SI values equal to 0 indicate equilibrium between the mineral and the solution while positive SI values show supersaturation and negative SI values reflect subsaturation. SI values can be expressed in numerous ways, such as to describe the dissolution or precipitation of minerals. Other concepts of SI, especially in aqueous chemistry, are widespread and have been widely used to discover the constituents that control the solubility of major elements (Boudot et al., 1996) and to delineate the effect on the freshwater–seawater mixing process in the aquifer (Abdullah et al., 2008).

3. Results and discussion

3.1. Descriptive analysis

During the study period, a large amount of data was collected to provide adequate evidence of the evolution of the hydrochemistry in Kapas Island. Descriptive analyses of the data obtained are shown in Table 5 and correlation data in Table 6.

The pH value of the analyzed samples varies from 6.70 to 7.60. In general, the pH of groundwater samples was slightly acidic to alkaline condition with a mean value of 7.12. This is because the reaction between carbon dioxide and water in a close aquifer system may produce H^+ ion, resulting in an acidic solution, as shown in Eqs. (1) and (2) below.

$$H_2O + CO_2 \rightarrow H_2CO_3 \qquad (1)$$

(Carbonate acid formation)

Table 6
Correlation matrices for all data ($n = 108$).

	Ca	Mg	Na	K	HCO_3	Cl	SO_4
Ca	1	0.000**	0.924	0.972	0.054	0.904	0.812
Mg	0.538	1	0.679	0.274	0.745	0.899	0.796
Na	0.009	0.040	1	0.164	0.008**	0.000**	0.000**
K	−0.003	0.106	0.135	1	0.000**	0.129	0.008**
HCO_3	0.186	0.032	0.253	0.395	1	0.000**	0.653
Cl	−0.012	−0.012	0.907	0.147	0.358	1	0.000**
SO_4	−0.023	−0.025	0.390	−0.254	0.044	0.368	1

Significant value (upper triangle; $p<0.01$**, $p<0.05$*).
Correlation value (lower triangle).

$$H_2CO_3 \rightarrow 2H^+ + CO_3^- \tag{2}$$

While, the reaction between solid species, which is mainly dominated by calcium carbonate in the coastal area, may contribute OH^- ions, which result in alkaline conditions (Eq. (3)), thereby increasing the pH values.

$$H_2O + CaCO_3 \rightarrow Ca^{2+} + HCO_3^- + OH^- \tag{3}$$

Together with pH, the oxidation–reduction process (*Eh*) reactions play a key role in the solubility of various ionic substances (Kresic, 2009). The *Eh* values in this study were in the range of −7.50 to 32.30 mV. The electrical conductivity (EC) value varies from 0.41 to 0.91 mS/cm, while the salinity values vary from 0.20 to 0.45 ppt, respectively. Salinity can include hundreds of different ions; however, relatively few make up most of the dissolved materials in groundwater Cl, Na, Ca, Mg, HCO_3 and SO_4. Salinity is correlated with most of the ions in the groundwater samples with r values (Mg = 0.454, Na = 0.757, K = 0.485, Cl = 0.822 and HCO_3 = 0.440), with $p<0.01$. Salinity also strongly correlated with TDS with r value = 0.999 ($p<0.01$). The Total Dissolved Solid (TDS) values ranged between 204 and 455 mg/L with an average value of 281.9 mg/L. High values of TDS indicate a high concentration of dissolved ions in groundwater samples, which significantly correlated with the high EC values ($r = 0.999$; $p<0.01$). This can be supported by the significant correlation coefficient values of TDS with Mg, Na and K ($r = 0.355$; 0.735; 0.453 and $p<0.01$), respectively.

Dissolved oxygen (DO) varies between 0.60 mg/L and 8.50 mg/L. Groundwater may have low DO value because the groundwater is in a closed system, therefore, a high concentration of DO may be observed during the rainy season as an effect of dilution of the groundwater in which mixing of old groundwater with recent recharge water in a certain sampling campaign. Groundwater in a closed system can be explained with respect to the concentration of gasses and the flows of groundwater recharge and discharge area. Since the groundwater is isolated in contact with the atmosphere, if the remaining gasses that dissolve in the groundwater are consumed, the concentration would decrease while the concentration increases when the gasses are generated during the recharge period. In addition, a low DO value in groundwater probably indicates that the bacteria present have consumed most of the dissolved oxygen for oxidation processes (Rose and Long, 1988). In addition, a high temperature and increased salinity in groundwater may naturally reduce the DO concentration. From this study, the correlation between salinity and DO gives $r = -0.218$ ($p<0.05$), which explains the negative relationship of the concentration of these two elements in the groundwater.

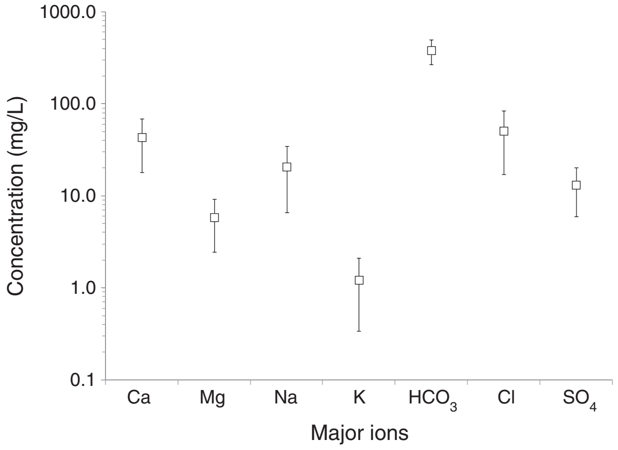

Fig. 5. Total concentration of major ions in groundwater samples for Kapas Island.

From the analysis of major ions, HCO_3, Ca and Cl occurred in high concentrations compared to Na, Mg and SO_4, while the K concentrations in boreholes were low in most cases (Fig. 5). The average concentrations of Ca and Na were 44.67 mg/L and 19.10 mg/L, respectively, while those of Mg and K were 5.12 mg/L and 1.13 mg/L, respectively. These cations are possibly derived from the chemical reaction of the deposited carbonate and other related minerals in the area affecting the groundwater composition, as demonstrated in the saturation indices sub-section. Furthermore, HCO_3 was the most dominant anion, with an average concentration of 379.19 mg/L. The average concentrations of Cl and SO_4 were 43.69 mg/L and 14.36 mg/L, respectively.

Following the detection of sulfide odors in the groundwater samples, a groundwater survey revealed concentrations of hydrogen sulfide in the groundwater at several locations, particularly at station KW 2. Station KW 2 experienced an unpleasant odor of hydrogen sulfide, which is most likely to be a product from the sulfate reduction process as shown in the following equation:

$$SO_4^{2-} + 2HC_2O \leftrightarrow H_2S + 2HCO_3^- \tag{4}$$

The formation of H_2S in groundwater is described as an anaerobic environment, which produces suitable conditions for the growth of sulfate-reducing bacteria (SRB) (Wargin et al., 2007). In most cases, SRB growth in groundwater may result in deterioration of the water quality. According to O'Sullivan et al. (2005), elevated H_2S concentrations might be related to the reclamation practices on the island. Since hydrochemical conditions within the aquifer are generally reduced, H_2S reduction is likely to be the major source for SO_4 to increase.

Other sources of SO_4 concentrations may result from sulfur minerals that constitute a significant percentage of the rock in many carbonate aquifers (Worthington and Ford, 1995). Mann (1958) explained that SO_4 increases in oxic conditions and HCO_3 increases in anoxic conditions. As groundwater mostly comprises more CO_2, which explains the elevation of HCO_3 concentration, the increasing SO_4 concentration can be explained and related to the increasing DO during the period of rain in the sampling campaign.

3.2. Hydrogeochemical facies

From Table 5, the dominant cations are mainly Ca and Na, while the dominant anion is HCO_3. The order of ion constituents (mg/L) is Ca > Na > Mg > K for cation and HCO_3 > Cl > SO_4 for anion and was plotted using a Piper diagram, as shown in Fig. 6.

Generally, Ca and HCO_3 dominate in coastal aquifers due to the wide contact and long retention time in crystallized limestone and marble (Özler, 2010), which cause the carbonate dissolution process. Based on Fig. 6, about 82% of the groundwater samples were classified as Ca–HCO_3 water type while the remaining groundwater samples were classified as Na–HCO_3 water type, which mostly came from KW 6. The water types can be described as Ca–HCO_3 (further inland) to Na–HCO_3 water type as it moves towards the coastal line, which are from station KW 1 (further inland) to KW 6 (Fig. 1). The distribution of groundwater samples was more to area rich in Ca (insert b)

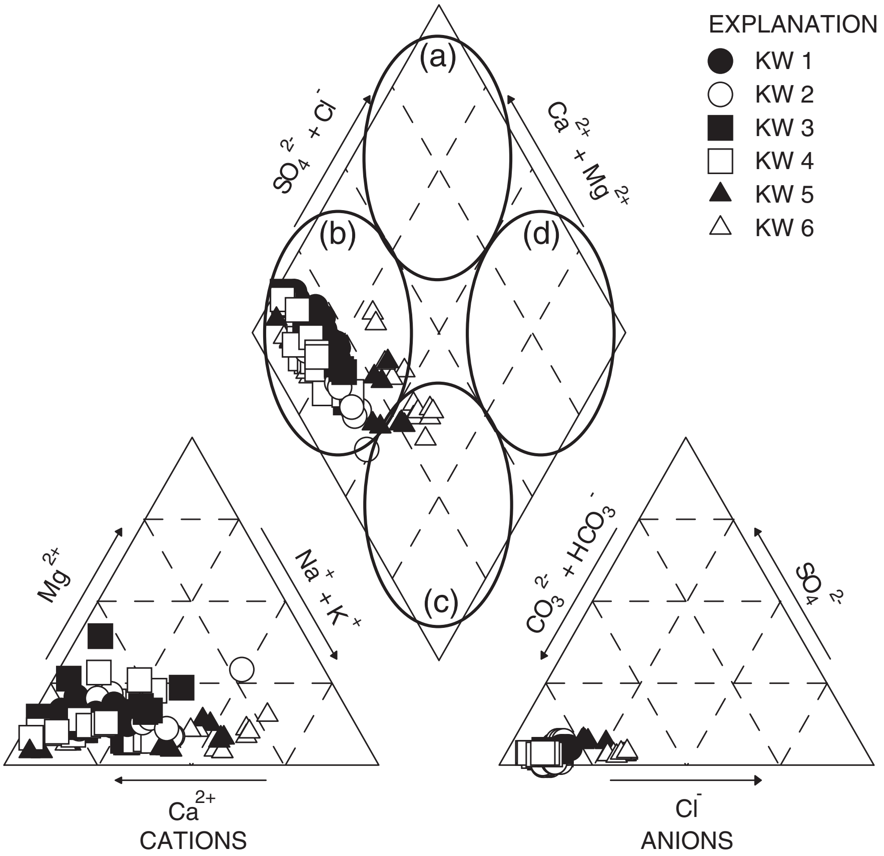

Fig. 6. Piper diagram presentation for groundwater constituents.

that leads to area rich in Na (insert c), as can be seen in Fig. 6. From the statistical analysis, there was a positive correlation between water types and stations ($r = 0.565$, $p < 0.01$). Station KW 5, KW 6 fell in both inserts (b) and (c) as both stations consist of two different types of water, which were Ca–HCO_3 and Na–HCO_3.

3.3. Ionic ratio

Data for the ionic ratio are shown in Table 7. Based on Cl/HCO_3 vs. Cl ionic ratio, it was found that all samples increased with a strong and positive correlation ($r = 0.945$, $p < 0.01$; Fig. 7a). Insert (ii) in Fig. 7a represents KW 6, which was more saline than other (salinity; 0.21–0.41 ppt) sampling stations, resulting in a higher Cl/HCO_3 vs. Cl ionic ratio which is >0.4. The increase of Cl content in Fig. 7a explains the increasing Na content in groundwater ($r = 0.907$; $p < 0.01$; Table 6). Insert (i) in Fig. 7a shows that most of the groundwater samples with low ratio of Cl/HCO_3 vs. Cl are under improved status, which means the freshening of groundwater. The high concentrations of HCO_3 in the groundwater are believed to arise from the dissolution of the aquifer matrix, as elaborated in the saturation indices sub-section. Fig. 7b shows the Na/Cl ratio, in which about 95% of the groundwater sample is <1 which indicates that the mechanism represents the cation exchange process. A Na/Cl ratio that is equal to 1 describes halite dissolution and a ratio of Na/Cl>1 indicates silicate weathering. The scatter plot in Fig. 7c is another example to illustrate the ionic exchange process. About 95% of the samples reflected an ionic exchange leading to SO_4 and HCO_3 accumulation in the groundwater since HCO_3 is the dominant anion in the groundwater. According to Rahman et al. (2010), and Reddy and Kumar (2010), the groundwater samples plotted using a scatter plot of ($SO_4 + HCO_3$) vs. (Ca + Mg) indicate that weathering of minerals is involved in the evolution of the groundwater as the samples fall below the equiline.

The difference in water types – Ca–HCO_3 and Na–HCO_3 – (Fig. 6) is caused by the replacement of cations in the groundwater aquifer. The changes of cation in water type may be derived from the cation exchange process, dissolution of calcite and weathering process. The cation exchange process is the major process that modifies the quality and controls the changes in concentration of major cations in the groundwater of Kapas Island corroborated by the calculation of

Table 7
Ionic ratio for groundwater samples.

Station	Cl/HCO_3 (meq/L)		Ca + Mg (meq/L)		SO_4 + HCO_3 (meq/L)		Na/Cl (meq/L)		Ca/Mg (meq/L)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
KW 1	0.180	0.026	3.423	1.473	6.053	0.206	0.582	0.184	4.761	1.094
KW 2	0.168	0.018	3.021	1.561	7.152	1.156	0.652	0.135	4.411	1.511
KW 3	0.153	0.024	1.943	0.929	4.851	0.300	0.597	0.307	5.087	2.405
KW 4	0.121	0.033	2.133	0.875	5.353	0.288	0.740	0.358	5.627	2.554
KW 5	0.199	0.085	2.793	1.607	8.111	3.351	0.801	0.283	8.809	5.196
KW 6	0.353	0.186	2.588	1.440	7.573	1.933	0.671	0.173	6.178	3.192

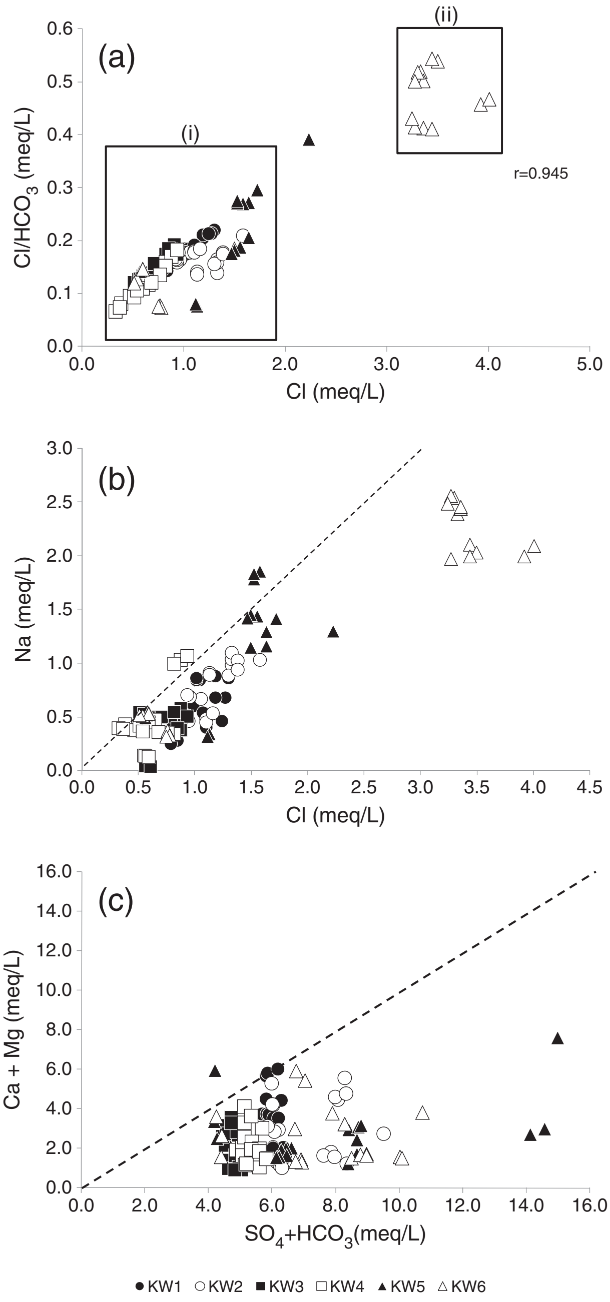

Fig. 7. Distribution of ionic ratio for major ions describing ion exchange processes.

ionic ratio (Fig. 7) and the presentation of ion distribution in Piper diagram (Fig. 6). This was also supported by Russak and Sivan (2010), in that the cation exchange process significantly controls the enrichment or depletion of element concentration in salinization or freshening aquifers.

Table 8
CEC value for every sampling station (n = 18).

Station	CEC (meq/100 g)	SD	Min	Max
KW 1	2.68	0.01	2.67	2.68
KW 2	1.80	0.02	1.77	1.82
KW 3	0.84	0.01	0.83	0.85
KW 4	0.67	0.05	0.62	0.72
KW 5	0.67	0.01	0.66	0.68
KW 6	0.61	0.02	0.59	0.62

3.4. Cation exchange capacity (CEC)

In the sandy aquifer of Kapas Island, the CEC varies between 0.61 and 2.68 meq/100 g with an average value of 1.21 meq/100 g (Table 8). It was found that there is no significantly distinct grouping of these values between stations ($p > 0.05$). According to Appelo and Postma (2005), the area can be classified as a sandy area as represented by low organic matter and low moisture with high permeability (Abdullah, 1981), since the value was below 10 meq/100 g. This is in agreement with the early findings reported by Abdullah (1981) who found the area to be dominated by sandy soils. From the correlation analysis, the CEC values were positively correlated ($r = 0.995$, $p < 0.01$; Fig. 8) with Ca. Hence, $CaCO_3$ is the primary cation exchanger in Kapas Island aquifer, that is, from the aquifer matrix, which is mainly built of sedimentary carbonate (Eq. (3)).

The concentrations of Ca in the groundwater may decrease while the concentration of Na increases during the groundwater mixing, which are the interactions with the aquifer matrix. The Na ions replaced the Ca ions on the solid surface, as demonstrated in equation below.

$$\mathrm{Na{-}X} + \frac{1}{2}\mathrm{Ca}^{+} \rightarrow \mathrm{Na}^{+} + \frac{1}{2}\mathrm{Ca}^{+}. \tag{5}$$

As the Na, which originated from halite minerals (NaCl) that precipitated on aquifer matrix, interacts with the groundwater, the Na ion overtakes the Ca ion and is dominant in the groundwater through the cation exchange process. Thus, the groundwater solution increases with the Na ion.

3.5. Saturation indices (SI)

Data for saturation indices in the study are shown in Table 9. From Fig. 9, the SI of carbonate minerals shows that 76% of the samples were in a condition of under-saturation, which results in the dissolution of carbonate minerals. The carbonate minerals involved in this mechanism were calcite, aragonite and dolomite, which contain Ca and Mg elements. The observation of correlation analysis shows a significant difference between the pH with Ca and pH with Mg ($r = -0.274$ and -0.396, $p < 0.01$), respectively, which plays a role as one of the dissolution factors (Isa and Aris, 2012). The pH values for each station show no distinct group ($p > 0.05$) where the pH observed ranged from 6.70 to 7.60. This indicates that the CO_2 plays an important role as H^+ producer. The groundwater aquifer is in a closed system, which explains the CO_2 concentration, and, hence, may neutralize the OH^- concentrations from the dissolution of $CaCO_3$ (Eq. (3)). Thus, the negative correlations between pH with Ca and pH with Mg were observed. The

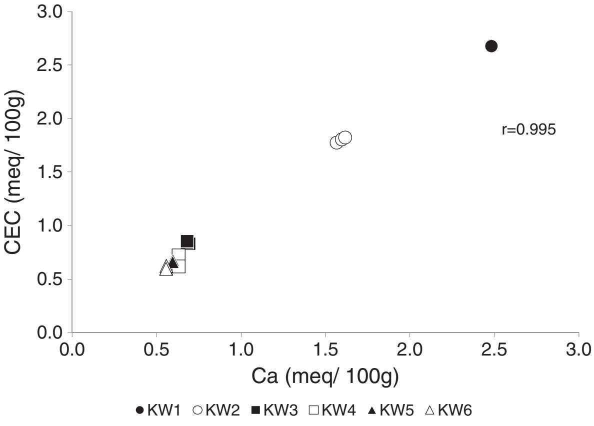

Fig. 8. Correlation of Ca vs. CEC (meq/100 g) (n = 18).

Table 9
Saturation Indices (SI) of calcite, aragonite and dolomite of the study area.

Sampling Station		Calcite	Aragonite	Dolomite
KW 1	Mean	0.10	−0.04	−0.28
	SD	0.18	0.18	0.33
KW 2	Mean	−0.12	−0.26	−0.66
	SD	0.25	0.25	0.40
KW 3	Mean	−0.16	−0.30	−0.77
	SD	0.23	0.23	0.30
KW 4	Mean	0.00	−0.14	−0.50
	SD	0.19	0.19	0.19
KW 5	Mean	0.10	−0.04	−0.46
	SD	0.31	0.31	0.51
KW 6	Mean	−0.16	−0.30	−0.85
	SD	0.19	0.19	0.24

mechanism involved in the aquifer is explained by the following equation; which considers the mechanism in the closed system.

$$CO_2 + H_2O + \underset{\text{(Calcite dissolution)}}{CaCO_3} \rightarrow Ca^{2+} + 2HCO_3^- \quad (6)$$

$$2CO_2 + 2H_2O + \underset{\text{(Dolomite dissolution)}}{CaMg(CO_3)_2} \rightarrow Ca^{2+} + Mg^{2+} + 4HCO_3^- \quad (7)$$

Eqs. (6) and (7) above explain the reaction of carbonate minerals in the closed system of the Kapas Island aquifer. The reaction of CO_2 in the aquifer results in the formation of carbonic acid Eq. (1), which subsequently dissociates to produce H^+ and HCO_3^- ions (Tijani, 1994). Comparative to this study, it was shown that Ca has increased due to the dissolution reaction, as presented in Fig. 9. The Ca was strongly correlated to the saturation state values of selected carbonate minerals, which were calcite ($r = 0.759$, $p < 0.01$), aragonite ($r = 0.759$, $p < 0.01$) and dolomite ($r = 0.662$, $p < 0.01$). In addition, samples between the ratio of 1 to 2 of Ca/Mg ratio reveal the indication of dolomite dissolution, while > 2 is the effect of silicate minerals, which suggests calcite dissolution. About 90% of the samples had a Ca/Mg ratio > 2, which indicates that the groundwater was responsible for the dissolution of minerals mainly from calcite Eq. (6). Groundwater samples that lie between a ratio of 1 and 2 (10%) describe the dissolution of dolomite minerals Eq. (7). Both reactions involved resulted in a concentration of Ca and Mg in the groundwater ($r = 0.538$; $p < 0.01$; Table 6).

The highest cation in terms of the percentage of seawater was nearly 0.8% at station KW 6, located nearest to the coastal area (Table 1). From Fig. 10, the SI values were clustered into two distinct groups, which defined different characteristics and behaviors of the groundwater. The small square in every insert (Fig. 10) shows the assortment of samples from station KW 6 during the six sampling campaigns. Station KW 6 is the nearest to the coastal area, which might have been affected by the movement of seawater laterally into the aquifer system from a single pumping activity as the coastal area is the closest to the transition zone between the groundwater and seawater.

Other than that, Na and Cl are the dominant ions found in saline areas that are carried inland from the ocean by wind, huge waves and rainfall, which later have accumulated in the soil over a long period of time. When the water table increases at certain times during rainy days, salt ions may dilute, which results in the concentration of salt ions in the aquifer (Benyamini et al., 2005). Some salts also occur naturally in the deep sediments and rock strata.

4. Conclusion

Groundwater in small islands has received much attention because it is the only source of freshwater supply to the local community and tourists. The composition of the groundwater is influenced by the hydrology and hydrogeological of the study area, which can be simplified using a complex conceptual model. The conceptual model includes important information gathered from both the previous and present studies. Based on this study, it can be seen that the hydrogeology controls the water chemistry and reveals that the hydrochemistry of Kapas Island is partly due to the chemical inputs from precipitation, chemical weathering process, dissolution of the deposited carbonate shells and the cation exchange processes. The hydrogeological interactions between static and dynamic components determine the groundwater quantity and quality. The groundwater type is largely characterized as a Ca–HCO_3 water type having relatively shifted to a Na–HCO_3 water type. The overwhelming predominance of Na over Ca ions in the groundwater is mostly from the cation exchange process, which can be supported from the ionic ratio calculation (Cl/HCO_3 vs. Cl, Na/Cl and $(SO_4 + HCO_3)$ vs. $(Ca + Mg)$). These observations have major consequences with

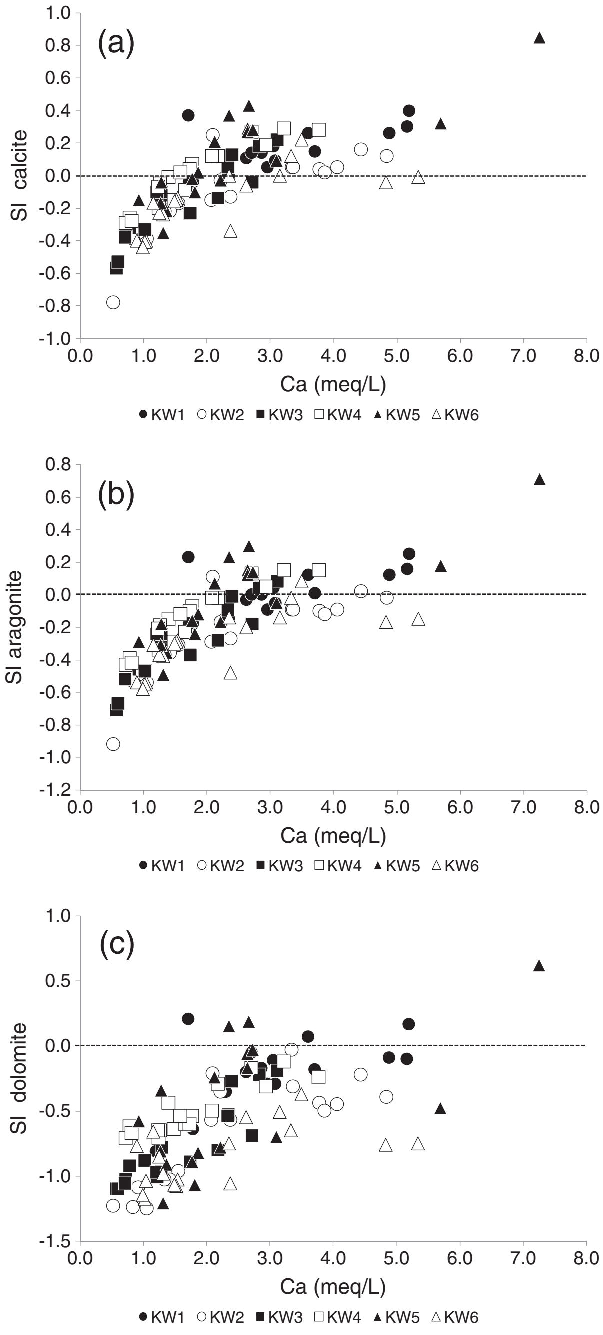

Fig. 9. Distribution of soluble minerals; insert (a) = calcite mineral, insert (b) = aragonite mineral, insert (c) = dolomite mineral and Ca.

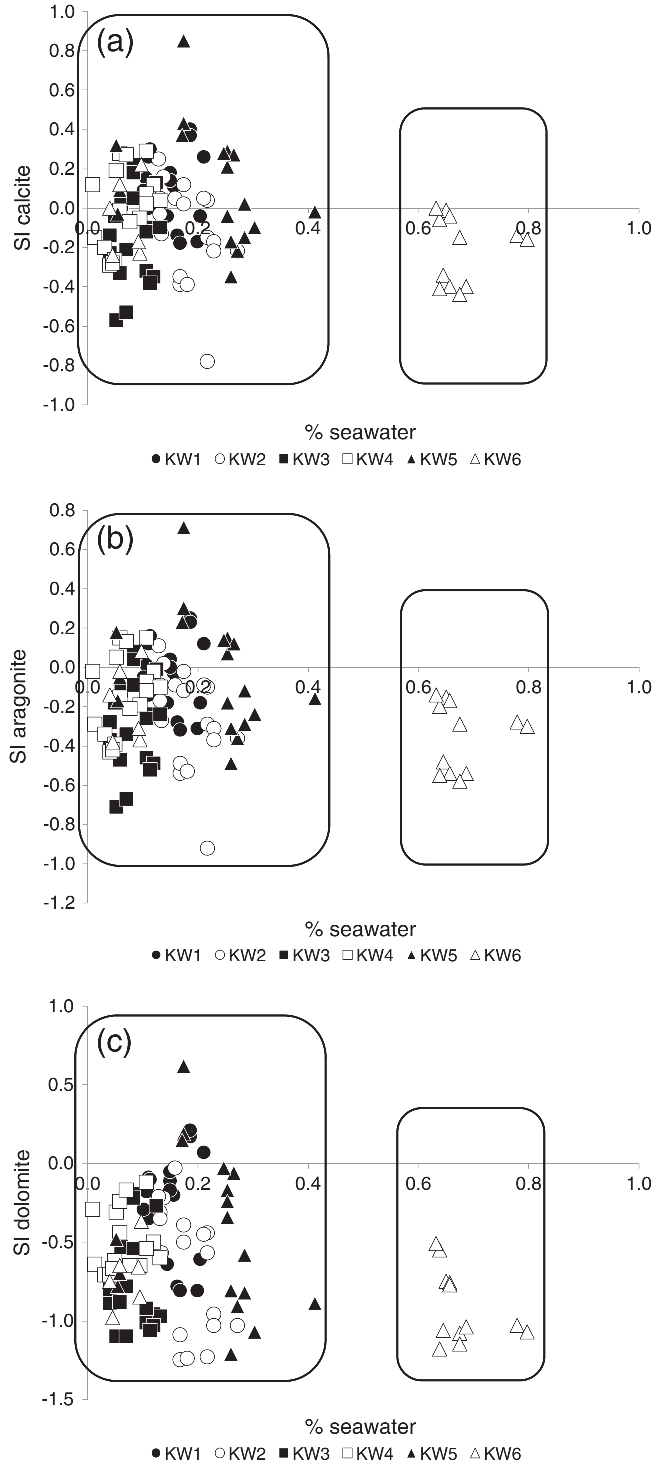

Fig. 10. Distribution of saturation index in Kapas Island; insert (a) = calcite mineral, insert (b) = aragonite mineral, insert (c) = dolomite mineral.

respect to SI determination. Kapas Island mostly experiences the dissolution of carbonate minerals with 76% of the data falling below the state of equilibrium. Although precipitates of carbonate minerals take place (24%) in Kapas Island aquifer, the mechanisms of saline water occurred naturally, in which the percentage of seawater in the aquifer is <1%.

Further investigations are needed to determine the possible alterations to the character of the groundwater, which could also eternize the indigenous environment of the area. The institutional framework must make sure that the planning and development, monitoring activities and management of the water resources take place in the small islands at the national-level. Regulation and enforcement are needed for better protection of groundwater, especially in small islands.

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