

Extenuation of Saline Solutes in Shallow Aquifer of a Small Tropical Island: A Case Study of Manukan Island, North Borneo

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Abstract

Intensive exploitation of groundwater from Manukan Island's aquifer has disturbed the natural equilibrium between fresh and saline water and has resulted in the increase of groundwater salinity and the hydrochemical complexities of freshwater-seawater contact. It was observed that the mixing between freshwater-seawater has created diversity in the geochemical processes of Manukan Island's aquifer and altered the freshwater and seawater mixture away from the theoretical composition line. The results from reactive transport modelling confirmed that the migration of seawater into the fresher parts of the aquifer apparently leads to a calcification of the aquifer despite the seawater being supersaturated for carbonate minerals and shows that the composition of the near coast zone and further landward area may vary and have a significant effect on the processes during the intrusion. It was observed that the effect of freshening aquifer in the landward area near the recharge zone of the study area has incriminated the calcite saturation states of the area. The accumulation of Ca as the interface travels landward up to 100 m from the coast leads to an increasing calcite supersaturation with travelled distance and possibly to the precipitation of calcite.

Keywords: small island; hydrochemistry; PHREEQC; solutes transport

1. Introduction

The presence of freshwater in small islands usually depends on the quantity, its surface and subsurface storage. The interaction between infiltration and groundwater resources is needed in order to sustain the groundwater freshness. This is critical in places where surface water does not exist in an exploitable form, whereas the groundwater resource itself is very limited. Fresh groundwater is the only option which can satisfy small amounts of water demand. Groundwater usage for daily supply in the small island has drastically increased over the last decade due to the rapid increase in visitors to the island (Aris *et al.*, 2009). This has resulted in an enormous leap on groundwater extraction, leading to a contamination of the wells in the island. With this current practice, incursion of seawater into the island's aquifers especially in the low-lying area of the island, is an expected significant consequence. While it is an unseen phenomenon, the influence of seawater intrusion on the ecology of coastal systems especially small islands may be more important than once thought, due to the potential impacts resulting from chemical alterations during the

freshwater-seawater mixing.

Therefore, the understanding of groundwater chemistry is crucial for groundwater management and planning on a small island. Studies on the geochemical constituents in groundwater of small islands may reveal the main processes responsible for the geochemical evolution as shown by previous studies such as Abdullah (2001), Xie *et al.* (2005), Abdullah and Aris (2005), Aris *et al.* (2007), Abdullah *et al.* (2008) and Aris *et al.* (2009). Disturbance of the hydrological balance, for an example due to overextraction of groundwater, could contribute to such a problem. Since the presence of saltwater in fresh groundwater aquifer is manifested by the variation of salinity and major seawater ions (Na, Cl and SO₄), the real overview on its distribution and movement should be forecasted. Thus, as a case study pertaining to that issue, an attempt was made to investigate and describe the dynamics of flow and geochemical processes in a small island aquifer subject to groundwater extraction. This paper presents the results of a preliminary field study of the geochemical processes in a seawater/freshwater mixing within a shallow sandy aquifer in a small tropical island.

2. Materials and Methods

2.1. Study area

Located about 7.5 km from Kota Kinabalu, the capital city of Sabah, Malaysia (Fig. 1), Manukan (5°57'-5°58' N and 115°59'-116°01' E) is one of the islands in the Tunku Abdul Rahman Park. Covering an area of 206,000 m², the island is crescent shaped, one and half kilometres long and three kilometres wide in the middle. Almost 80% of the area of the island is high relief and covered by forest (western side of the island), whereas the remaining 20% has been developed for tourism activities located on the low lying areas of the island (eastern part).

Geologically, Manukan Island was isolated from the mainland about one million years ago (Basir *et al.*, 1991). The island is underlain by interbedded sandstone and shale of the Middle Miocene Crocker Range Formation. The sandstones form cliffs and deep embayments along the coast. The sedimentary rocks of Manukan Island dip at 15-45° to the east-northeast towards the low relief area (Abdullah *et al.*, 1997). The main aquifer of Manukan Island comprises Quaternary carbonates and coarse sandy alluvium, which overlies the older rocks.

The groundwater recharge for Manukan Island aquifer depends entirely on infiltration. Sabah has a

warm and humid climate with annual rainfall between 2000-2700 mm, humidity between 80-90% all year round and temperatures from 21 to 32°C. The climate is affected by the northeast and southwest monsoons, which are dominant during November to March and May to September respectively. The period between the monsoons are marked by heavy rainfall. Presently, Manukan Island depends on a shallow aquifer for its groundwater supply.

2.2. Monitoring well installation and sediment coring

Six monitoring wells with different depths were installed in the study area from south to north. This installation is based on flow measurement that was conducted by Abdullah (2001) using piezometers constructed on the island. The monitoring wells were installed over a distance of 80 meters (Fig. 1) from the coastline and the details of the monitoring wells are specifically indicated in Table 1.

The wells were screened at different elevations between 1 and 4 meters from ground surface level. The sediment cores were taken with a hand auger to a depth of 1.5-4.0 meters during the well construction. After the sediment cores were pulled to the surface, they were immediately sectioned into lengths of 1 m, mixed, sealed and kept in cooler box covered from direct sunlight.

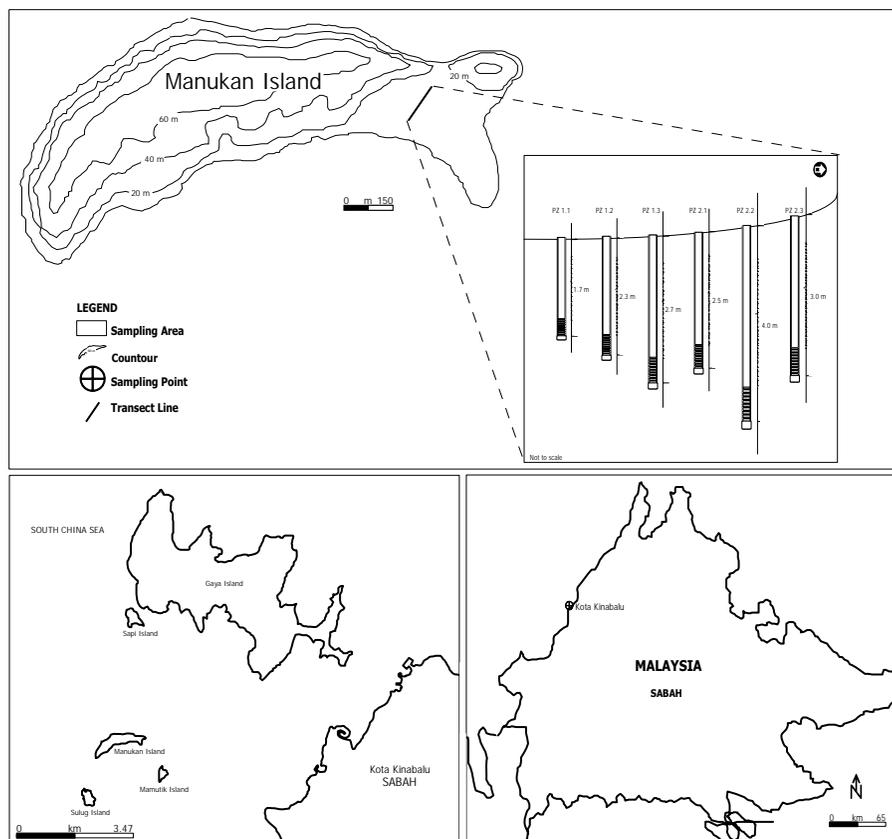


Figure 1. Schematic map showing the geographical locality of Manukan Island

Table 1. Locations coordinate of monitoring wells based on Global Position System (GPS) readings.

| Station's Code | Station's Coordinate | Distance from coastline |
|----------------|------------------------------|-------------------------|
| PZ 1.1 | N 05°58.565' ; E 116°00.350' | 40 m |
| PZ 1.2 | N 05°58.523' ; E 116°00.347' | 42 m |
| PZ 1.3 | N 05°58.525' ; E 116°00.347' | 44 m |
| PZ 2.1 | N 05°58.533' ; E 116°00.353' | 70 m |
| PZ 2.2 | N 05°58.551' ; E 116°00.358' | 74 m |
| PZ 2.3 | N 05°58.546' ; E 116°00.351' | 78 m |

2.3. Laboratory procedures for sediment samples

The content of exchangeable cations (Ca, Mg, Na and K) in sediment samples were determined by ion displacement using separately a 1 M NaCl and a 1 M NH₄Cl solution (Van der Molen, 1958; Andersen *et al.*, 2005; Appelo and Postma, 2005). The cation exchange capacity (CEC) was calculated as the sum of these exchangeable cations.

Prior to the treatment the samples were percolated with ethanol as suggested by Appelo (1996) in Butchner like funnel over 0.45 µm polypropylene micro filter. The purpose of this ethanol percolation is to displace the pore water from the sediment. This is necessary because the contribution of cations from the pore water in sediment with saline or brackish pore water can be more than ten times greater than the exchangeable fraction. About 10 ml of 95% ethanol was carefully poured on the sediment sample and drawn through the sediment by suction. About 30 ml of a 1 M NaCl solution was applied to one set of sub samples and 1 M NH₄Cl solution was added to other set. The centrifuge tubes contains with the sediment pre treated with NaCl and NH₄Cl were shaken end over end for about 10-20 minutes and subsequently centrifuged at 3000 rpm for 30 minutes in order to settle the fines. After the samples were centrifuged, the supernatant was removed with a syringe and filtered through a 0.45 µm filter.

About 15 ml was used for the analysis of Ca, Mg, and K from the NaCl supernatant solution and preserved with 1% 7M HNO₃. The solution from NH₄Cl supernatant was used for determination of Na and also preserved with 1% 7M HNO₃. Samples analysis for Ca, Mg, Na and K were performed using flame (air-acetylene burner) atomic absorption spectrometry (FAAS - Zeeman AAS Z-5000, Hitachi, Japan). The exchangeable cation concentrations are converted from meq/100 g to equivalent fractions (β_T) as suggested by Appelo & Postma, 2005.

2.4. Water analysis procedures

The well water samples were pumped out for some time (5 to 10 minutes) prior to groundwater sampling, to avoid non-representative samples of stagnant or polluted water. The water samples were pumped using a modified dry vacuum pump technique. The analyses of water samples were carried out to assess major cations and anions including sodium (Na), potassium (K), calcium (Ca), magnesium (Mg), bicarbonate (HCO₃), chloride (Cl) and sulphate (SO₄). The water pH and temperature were measured in the field using WTW pH 315i. Water samples were filtered at the time of collection using 25 mm puradisc syringe filtration unit of 0.45 µm pore size Whatman Milipore®, acidified to pH 2 with concentrated HNO₃ acid and analyzed using flame (air-acetylene burner) atomic absorption spectrometry (FAAS - Zeeman AAS Z-5000, Hitachi, Japan). For major cations. SO₄ was measured using HACH (DR/2040 - Loveland, CO, USA) meter, Cl and HCO₃ were analyzed using argentometric and titration methods (APHA, 2005) respectively.

2.5. Reactive transport modelling

The transect (from the coastline to landward area of the Manukan island) is represented by a 100 m horizontal column sectioned into 50 cells of 2 m in length. The incursion of seawater into the sandy aquifer of Manukan island can be justified by the predominantly horizontal intrusion front as suggested by Aris *et al.* (2009).

The model setup only describes the processes in the mixing zone of the aquifer and not in the saline wedge. In the model, the initial groundwater is a mixture of freshwater and seawater end members as a result of incursion of seawater into the aquifer. Details of the model setup are given in Table 2. The seawater chemistry which was set as the intrusion seawater is

Table 2. Parameters for the PHREEQC 1-D reactive transport model.

| Model setup parameters | Value | Comments |
|--------------------------------|--|---|
| Physical parameters | | |
| Model domain | 100 m | Divided into 50 cells of 2 m |
| Boundary conditions flow | Flux | Steady state flow |
| Dispersivity | 0.4 m | Fitted |
| Simulation time | 0.7 yr | Fitted |
| Chemical parameters | | |
| Initial pore water composition | 0-80 m: 15% seawater | Proposed result of seawater intrusion (Aris et al., 2009) |
| | 80-100 m: freshwater | Data from Abdullah et al. (1996) |
| Boundary conditions chem. | Associated flux | Composition is fixed near the recharge area |
| CEC | 0-50 m: 5.7 meq/100 g 50-100 m: 6.9 meq/100 g | Average measured values (this study) Values along flowline were calculated with PHREEQC. Dissolution is when SI < 0 (This study) |
| SI Calcite | 6.3 meq/100 g Seawater : SI ~ 0.81 | |

the average surface seawater presented in Table 3. The transect data was measured just prior to the samples collection and seawater-groundwater chemistry data is used as the initial chemical condition. The variables data were than simulated with PHREEQC (Parkhurst and Appelo, 1999) for the reaction transport modelling in the aquifer.

3. Results and Discussion

3.1. Water chemistry at the seawater/freshwater mixing zone

Table 3 shows the composition of the three main water zones at the study site. Fresh groundwater is of low salinity and dominated by Ca-HCO₃ water type

with insignificant signature of seawater (Abdullah et al., 1996). The seawater sample resembles mean seawater chemistry of the South China Sea with typical values. Water samples from the mixing zone have higher pH compared to the fresh groundwater but lower than the seawater.

3.1.1. Cation exchange and CEC

In the sandy aquifer of Manukan island, the CEC varies between 5.7 and 6.9 meq/100 g with an average value of 6.3 meq/100 g. The CEC shows no distinct trends with distance from the coast but generally shows higher value at the 40 m mark of the monitoring well closest to the coastline and decreased towards 100 m from the coastline as it reaches the recharge area (hilly side). As shown in Fig. 2, the CEC correlates strongly

Table 3. Mean value of hydrochemical parameters for water samples from Manukan island.

| | pH (mg/L) | Ca (mg/L) | Mg (mg/L) | Na (mg/L) | K (mg/L) | Cl (mg/L) | SO ₄ (mg/L) | HCO ₃ (mg/L) |
|--------------------------------------|--------------|-------------------|------------------|----------------------|-------------------|----------------------|---------------------------|----------------------------|
| Fresh groundwater ^a | 6.96 | 60.95 (1.52) | 12.03 (0.50) | 0.92 (0.04) | 4.69 (0.12) | 170.90 (4.82) | 39.87 (0.42) | 355.70 (5.83) |
| Mixing zone groundwater ^b | 7.05 | 932.48 (23.27) | 85.26 (3.51) | 1352.27 (58.86) | 38.21 (0.98) | 2003.55 (56.51) | 271.44 (2.83) | 477.80 (7.83) |
| Seawater | 8.10 | 414.00 (10.33) | 42.51 (17.49) | 11120.20 (483.70) | 416.00 (10.64) | 19750.20 (557.10) | 2600 (27.07) | 125.12 (2.05) |

() bracket indicates mmol/L value

^a From Abdullah et al. (1996)

^b Mean value of hydrochemical parameters for groundwater samples collected at the depth of 1.70 to 4.00 m from g.s.l

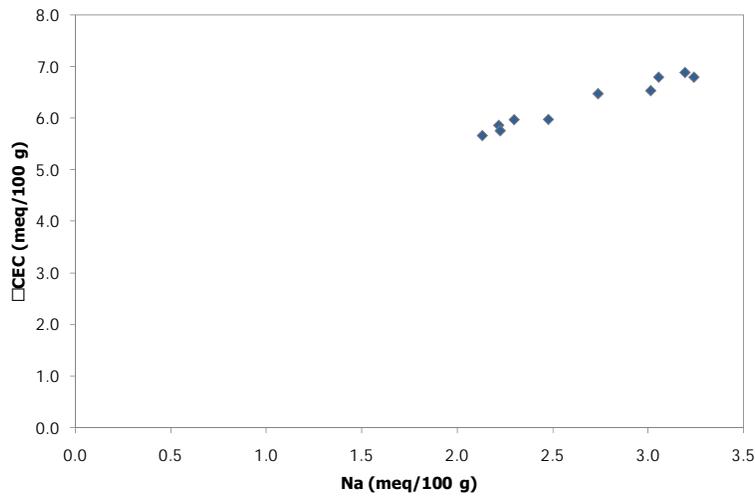


Figure 2. CEC vs Na exchangeable cation content.

($r = 0.962$; $p < 0.01$) with the Na exchangeable cation content of the sediment which is apparently the primary cation exchanger in the Manukan island aquifer which is from the seawater.

Exchangeable cations (Ca, Mg, Na and K) were measured and the results are expressed as equivalent fractions (β_T) of the total CEC (Table 4). Even though the decreasing trend of β_{Na} is not apparent, there were decreases about 0.03 of along the transect. The same observation was observed for β_{Ca} where it increases to around 0.03. β_{Na} and β_{Ca} showed the opposite tendency where the increase of β_{Na} will lead to the decrease in β_{Ca} . There is a strong and significant relationship between β_{Na} and β_{Ca} which clearly indicates that the cation exchange process has occurred between this two ions and significantly controlled the ion compositions in the Manukan island groundwater ($r = -0.995$; $p < 0.01$). Variations in the β_T along the transect especially for the β_{Na} and β_{Ca} are quite prominent because of the higher seawater content in the transect. The increasing trend of β_{Ca} by distance along the transect was associated with the increasing trend of β_{Mg} too. Moving away from the coastline, β_{Mg} increases to around 0.002. β_K is close to 0.004 for most of the

transect. The β_T in the samples taken during the field campaign are internally consistent along the transect although not exactly the same depths were sampled.

The β_T s for Ca and Na calculated with PHREEQC were found to be in equilibrium with the aqueous concentrations as shown in dashed lines in Fig. 3. The calculation was based on the water composition collected from the monitoring wells along the transect and the standard selectivity coefficients in the PHREEQC database. Both the spatial and the temporal shift may cause variability and the difference is particularly important because the density plumes resulting from the inundation were still migrating vertically from the front during the coring. Still, there is a good agreement between the measured composition of the exchange complex and the composition calculated from the water chemistry. In the freshwater-seawater mixing end, the selectivity coefficients were in good agreement with β_{Na} for samples 40 m from the coast but underestimated β_{Na} values for samples 70 m and above from the coast. For Ca, the agreement was more or less similar with the calculated β_{Ca} . As expected, the β_{Na} are higher in the cores collected at up to 50 m from the coast because of the higher seawater content in the transect with Na being the dominating cations. β_{Ca} increases away from the coast from 0.50 to 0.60 indicating that the influence of seawater chemistry to the water composition has decreased significantly over distance from the coast.

In combination, the depletion and enrichment of cations (Ca and Na) in the groundwater and the composition of the exchanger suggest the development of a chromatographic ion exchange pattern in the Manukan island aquifer. Following a transect line as observed from the site exploration, the inferred sequential reactions are:

Table 4. Mean equivalent fractions of Na, K, Ca and Mg on the exchanger measured on core samples based on sampling points.

| | β_{Na} | β_K | β_{Ca} | β_{Mg} |
|---------|--------------|-----------|--------------|--------------|
| PZ 1 | 0.434 | 0.004 | 0.545 | 0.017 |
| PZ 2 | 0.403 | 0.004 | 0.574 | 0.019 |
| Minimum | 0.376 | 0.002 | 0.503 | 0.014 |
| Maximum | 0.477 | 0.005 | 0.601 | 0.028 |
| SD | 0.032 | 0.001 | 0.028 | 0.003 |

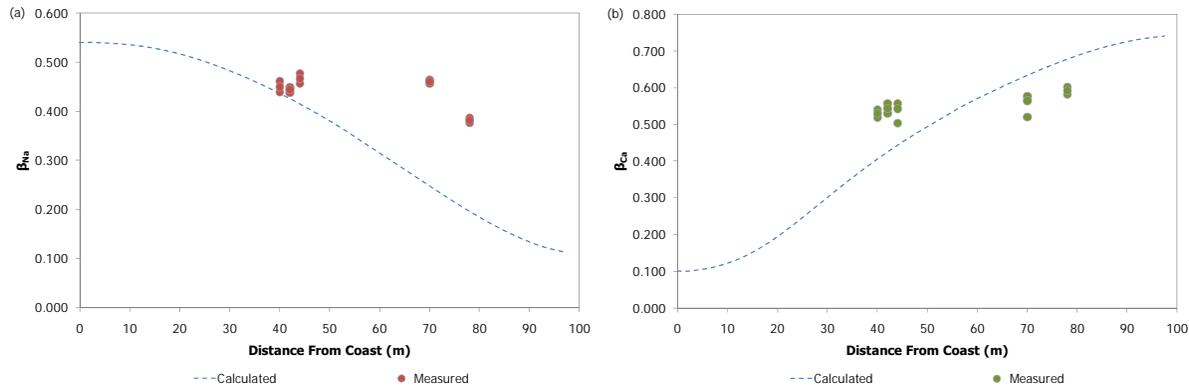
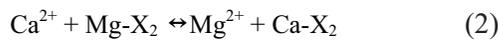
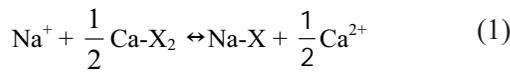


Figure 3. Equivalent fractions of (a) Na and (b) Ca on the exchanger measured on core samples (solid symbols) and calculated using PHREEQC from the groundwater composition (dashed lines).



where X denotes as fixed number of negative charges on the cation exchanger. Equation (1) was normally observed at the aquifer affected with seawater transition while equation (2) which was observed in the monitoring wells at 70 m and above from the coastline normally indicates that the aquifer is experiencing freshening as shown by its Ca-Cl water type. Apart from the observed overall salinization pattern, a minor freshening process of the aquifer can be inferred from the depletion of Na and enrichment of Ca, which was observed at the monitoring wells near to the recharge area (i.e at 70 m and above from coastline). However, a chromatographic sequence of salinization has not been clearly observed in seawater intrusion field-scale experiments, even though the Ca-Cl water type is a ubiquitous indicator of salt water upconing in a coastal area aquifer. The chromatographic sequence is caused by the sequential displacement starting with the cations with the highest affinity for the exchanger. During the seawater intrusion, when seawater mix with freshwater, the cation affinity order is normally $\text{Na} > \text{K} > \text{Ca} > \text{Mg}$ with Ca being displaced from the exchanger in the first order and Na eventually dominating the water and the exchanger. An adverse sequence will be observed in a freshening aquifer and it was found that some of the dissolved Na and K in the displacing seawater may exchange directly with adsorbed Ca on the exchanger further downstream due to dispersion.

3.2. Reactive transport modeling

The model results are shown in Fig. 4. The Manukan aquifer displays a Na-Cl type of water at its up-

low end (PZ 1), changing to groundwater enriched in Ca-Cl further down the flow path. The HCO_3^- increases with the flow path [i.e from Na-Cl; PZ 1 to Ca-Cl; PZ 2). The pattern is typically observed for freshening of an aquifer that originally has been filled with saline water. Representative hydrochemical facies in the aquifer show a marked difference between the headwaters (freshwater from recharge area), where the dominant type is Ca- HCO_3^- , and the coastal strip, where a Na-Cl facies dominates, and this progression clearly indicates freshwater and seawater mixing. In addition to seawater flushing from the aquifer, the Ca in the freshwater will replace Na in the cation exchange on the sediment grains by ion exchange process. Since the amounts of adsorbed Na are much larger than the Ca concentration in the flushing solutions, the ion exchange fronts will move much slower through the aquifer than the chloride front (Appelo & Postma, 2005). In addition, the ion exchange fronts may be coupled to mineral dissolution since binding of Ca ions can cause calcite dissolution, raising the pH and the bicarbonate concentration as seen in PZ 2 monitoring wells. The salt water had been flushed from the aquifer, where the current Cl concentrations at PZ 2 are less than the previous recorded concentrations from the other wells in Manukan island (16 - 37 mmol/L). The modeled Cl concentration [Fig. 4(a)] describes the obtained Cl data quite well with a steep decrease in the seawater influence at 40 to 100 m. Even though the aquifer has been displaced by recharge water, the sediment ion exchanger still contains the saline cations Na, K and Mg and these are at present being displaced by Ca from the recharging water. The concentrations of Na, K and Mg decreased at around 40 m from the coastline following the decreased in Cl (Figs. 4-5) which indicates that a major part of the cation distribution are due to advective transport.

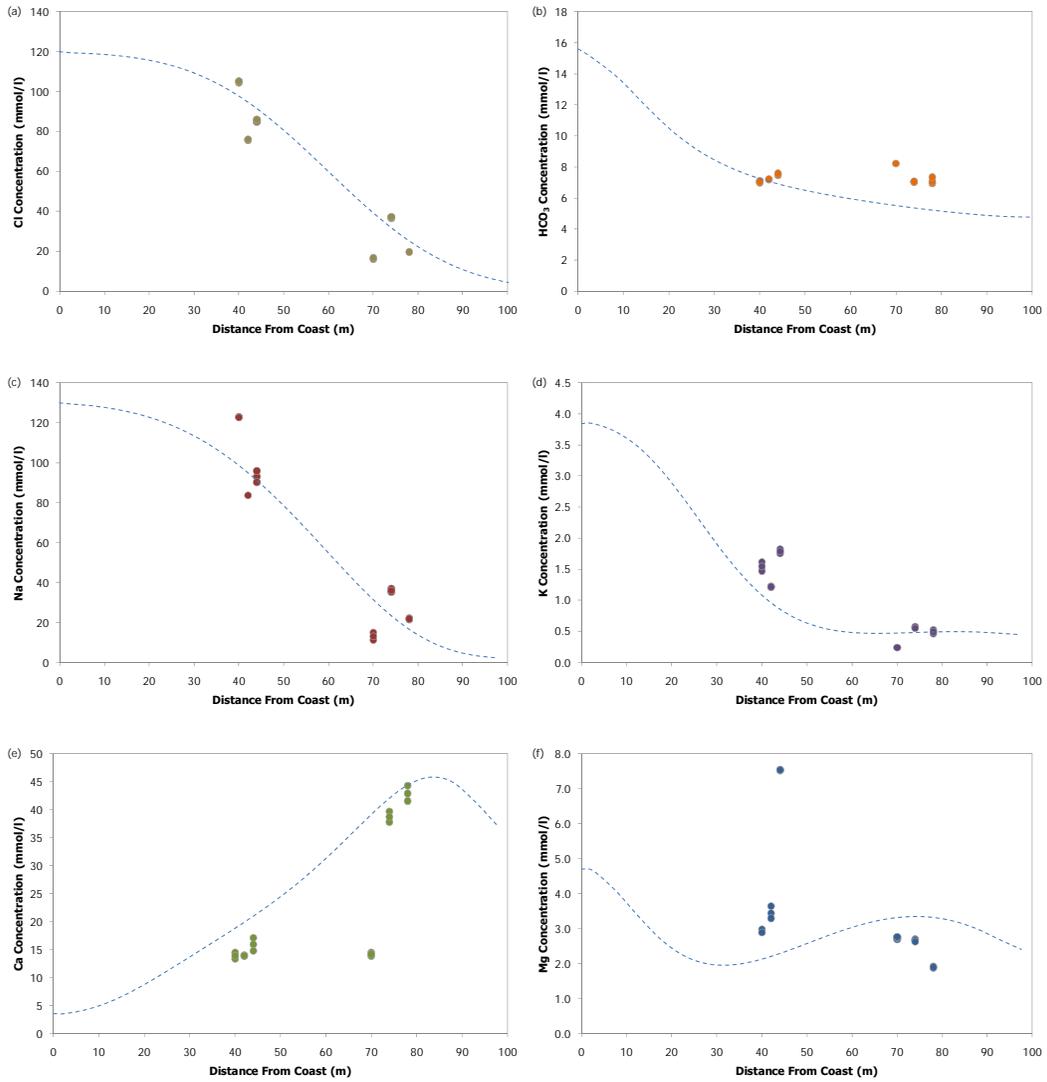


Figure 4. Observed distributions (solid symbols) along flow path from coast of: (a) Cl, (b) HCO_3 , (c) Na, (d) K, (e) Ca and (f) Mg (all in mmol/L). Dashed lines are modeled distributions using PHREEQC with 15 % of diluted seawater mix with fresh groundwater.

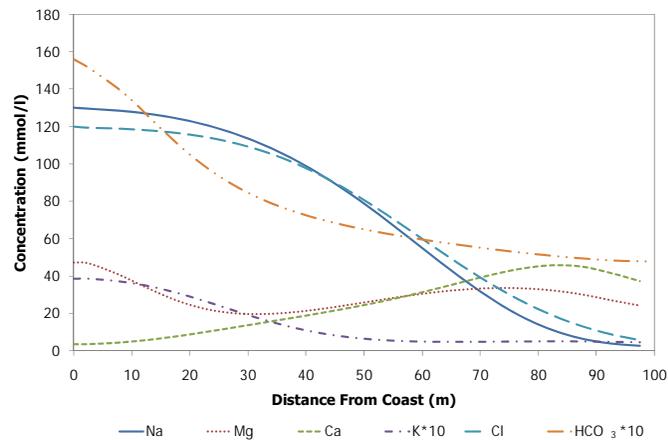


Figure 5. Overall modeled distributions using PHREEQC.

From about 70 to 100 m, Ca gradually displaces the seawater derived cations Mg, Na and K from the exchanger, resulting in Ca concentrations enrichment (Figs. 4-5). The water shifted from Na-Cl to Ca-Cl in all monitoring wells at this distance from the coastline. The shifts clearly indicate the effect of the ion exchange by the decrease in Na and increase in Ca concentrations.

The model overestimates the Ca concentration towards 40 m from the coastline by ~9 - 10 mmol/L, part of which could be due to the dissolving calcite. The calcite species being dissolved are Mg-calcite, rather than the pure calcite used in the model which would also explain the underestimation of Mg at the same distance from the coastline [Fig. 4(f)]. Such elucidation is supported by the fact that the SI values for samples collected at ~40 m from the coastline were lower and near to equilibrium state (SI calcite ~0.21 - 1.05) compared to the samples collected at ~70 m from the coastline (Table 5). The same explanation was made for the underestimates of Mg concentration obtained compared with the modeled data at 70 m from the coastline where SI value for Mg-calcite are supersaturated compared to samples collected at 40 m from the coastline.

Good agreement between the model and the observed values is seen for HCO₃ [Fig. 4(b)]. There is a decreasing trend observed in HCO₃ concentrations over distance even though the concentrations of the observed and modeled are not that apparent at the monitoring wells ~70 m from the coastline. Initially, the calculated HCO₃ in the aquifer receiving recharge water was 5 mmol/L (305 mg/L) (observed at 80 m above from the coastline) and of 7.3 mmol/L (458 mg/L) in the monitoring wells that were significantly affected by seawater (typically at 0 to 40 m). The increase of HCO₃

to 7.5 mmol/L (458 mg/L) in PZ 2 compared to 7.3 mmol/L (445 mg/L) in PZ 1 is partly due to sulfate reduction. The modeled concentration at ~70 m was underestimates compared to actual obtained values and this should be explained by the entrapped SO₄ ion at the aquifer matrix derived from seawater which has undergone the reduction processes that eventually enriched the HCO₃ constituents in the water body as:



The SO₄ reduction effects the carbonate equilibria causing supersaturation with regard to calcite. Since the organic carbon content was not measured in this study, the simulation on the SO₄ transport in the aquifer could not be conducted and can be only justified by the above mentioned processes.

4. Conclusions

The low topography of the study area in Manukan island makes this aquifer prone to frequent inundations of seawater aggravated by continuous groundwater abstraction from its sandy aquifer. These inundations are causing density driven seawater to migrate further into the fresh region of the aquifer (up to 100 m from the coast) and thereby constitute an important additional mechanism for the transport of seawater into the aquifer. The results from reactive transport modeling indicate that the migration of seawater into the fresher parts of the aquifer apparently leads to a calcification of the aquifer despite the seawater being supersaturated for calcite; an indication of seawater chemistry influence to the fresh groundwater aquifer. The main chemical process responsible for an increase of the calcite saturation state is ion exchange process related to the mixing of seawater and fresh groundwater in the carbonate system.

This study also shows that the water composition of the near coast zone and further landward area may vary and have a significant effect on the several processes (i.e redox process and cation exchange process) during the intrusion. It was observed that the effect of freshening aquifer in the landward area near the recharge zone of the study area has incriminated the calcite saturation states of the area. The accumulation of Ca as the interface travels landward up to 100 m from the coast leads to an increasing calcite supersaturation with travelled distance and possibly to the precipitation of calcite.

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Table 5. Average saturation index for carbonate minerals and gypsum calculated with PHREEQC.

| | Aragonite | Calcite | Dolomite |
|------|-----------|---------|----------|
| PZ 1 | | | |
| Mean | 0.49 | 0.63 | 0.95 |
| Min | 0.05 | 0.19 | 0.29 |
| Max | 0.91 | 1.05 | 1.63 |
| SD | 0.36 | 0.36 | 0.55 |
| PZ 2 | | | |
| Mean | 1.03 | 1.17 | 1.46 |
| Min | 0.86 | 1.00 | 2.14 |
| Max | 1.26 | 1.40 | 1.66 |
| SD | 0.17 | 0.17 | 0.17 |

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