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Original Article

Evaluation of Processes Affecting the Variation of Groundwater Quality in Quang Nam, Da Nang, Vietnam

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Abstract: Groundwater quality is vulnerable to various processes. In this study, processes affecting groundwater quality were evaluated in coastal aquifers of Quang Nam - Da Nang (QNDN). A chemical data of 426 groundwater samples from 27 monitoring wells in the period 2011-2018 were analyzed. Principal Components Analysis (PCA) and Base Exchange Indices (BEX_D) were applied for the evaluation. The PCA results suggested the influences of natural processes and anthropogenic activities on the groundwater quality. Seawater influence contributed to the dominant ions in groundwater; mineral weathering and dissolution mainly increased the alkalinity, Ca^{2+}, and Mg^{2+}; SO_4^{2-} reduction explained the low SO_4^{2-} in the groundwater; and reductive dissolution of Fe (hydroxides) caused Fe exceeding WHO's drinking standard. Intensive groundwater abstraction generated up coning of saline groundwater; discharge from agricultural practices, industrialization, and urbanization were considered as sources of high NO_3^- in groundwater. The integration of monitoring data and BEX_D gave a better interpretation of salinization and freshening, which can be masked by the memory effects of seawater transgression and regression in history.

Keywords: Groundwater quality, natural processes, anthropogenic activities, memory effects.

1. Introduction

Groundwater is an essential resource for human life. Nowadays, the intensive groundwater abstraction has been occurred in many coastal aquifers and caused the deterioration of groundwater quality. Groundwater quality is sensitive to various

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geochemical processes and anthropogenic activities. Seawater intrusion is a common process affecting groundwater in the coastal aquifers, which increases Cl$^-$ of groundwater above the World Health Organization (WHO) standard for drinking water (250 mg/L). The weathering of minerals is the process that introduces dominant cations into groundwater such as Ca^{2+} and Mg^{2+} [1, 2]. The natural reduction process is the cause of the occurrences of various contaminants, such as As, Fe, and NH$_4^+$ [3-5]. In addition, human activities, directly and indirectly, affect the groundwater quality. In an urban area, the groundwater contamination has been observed in many places due to the discharge of effluents from industrial [6, 7], domestic waste water, and landfill discharge [8, 9]. In rural areas, agriculture practices are major sources of nitrogen contaminated in groundwater such as NO$_3^-$, and NH$_4^+$ [10, 11]. To identify such factors regulating groundwater quality, the installation of groundwater monitoring network is very important. Based on the monitoring data of groundwater chemistry, various factors regulating groundwater quality can be evaluated and, thus, suitable measures can be given for sustaining groundwater resources.

In the coastal area of QNDN, a monitoring network was installed in 2011 for the purpose of groundwater management. In this area, the groundwater is mainly exploited for supplying the water demand for domestic use, agriculture, and tourism. Due to groundwater abstraction, the deterioration of groundwater quality has been changed. However, the understanding of processes causing the variation of groundwater quality is still limited in this area. Therefore, this study utilizes the monitoring data (from 2011 to 2018) to evaluate processes controlling the variation of groundwater quality in QNDN, Vietnam. The identification of such factors is valuable information for the management and protection of groundwater in coastal aquifer of QNDN.

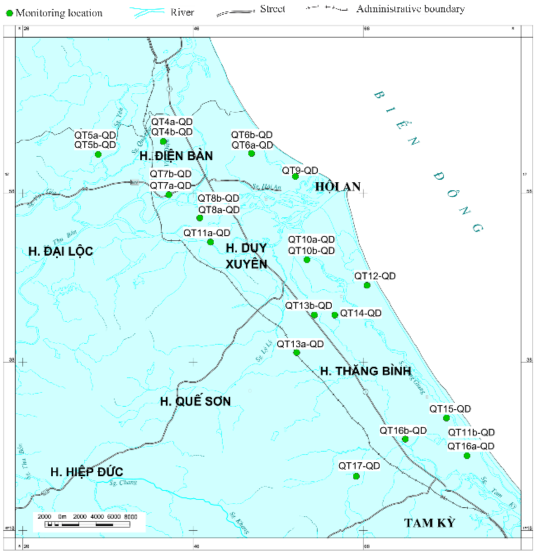

Figure 1. Locations of monitoring wells in the study area.

2. Materials and Methods

2.1. Study Area

The study area locates at 14°54’–16°13’ N and 107°3’–108°45’ E (Figure 1). The total area is 2425.8 km^2. The climate consists of the dry season and the rainy season. The dry season is from February to August and the rain season is from September to January next year. The annual rainfall is high with an average of 2770 mm. The evaporation is also high with an average of 2107 mm/year. The average annual temperature is 25.4 °C. The hydrogeology of the coastal area of QNDN consists of three main aquifers: Holocene (qh), Pleistocene (qp), and Neogene (n). The thickness of Holocene aquifer is from 2 m to 28 m, consisting of sand, silty sand, and gravel. The thickness of Pleistocene aquifer is from 5 m to 50 m. The lithology of Pleistocene aquifer is mostly from gravel sand to silty sand. Neogene aquifer consists of sandstone, siltstone, and conglomerate. The thickness of Neogene aquifer is from 10 m to 30 m.

The groundwater is monitored at depth of 14-50 m, 15-50 m, and 100 m in Holocene, Pleistocene, and Neogene aquifer, respectively.

2.2. Data Source

The data of groundwater chemistry used in this study is provided by Division for Water Resources Planning and Investigation for Central Vietnam. The data is monitored from 2011 to 2018. There are 27 monitoring wells consisting of 16 wells in Holocene aquifer, 7 wells in Pleistocene aquifer, and 4 wells in Neogene aquifer (Figure 1). The groundwater samples were collected twice per year in dry season and rainy season.

The groundwater samples were analyzed for major cations (Ca^{2+}, Mg^{2+}, Na^+, and K^+), major anions (HCO_3^-, Cl^-, and SO_4^{2-}), nitrogen species (NO_3^-, and NH_4^+), and Fe. The analysis of those parameters is followed by the standard of APHA [12]. pH is measured in the field. The ionic balance error of the used data is within ±5%.

2.3. Principal Component Analysis (PCA)

The PCA is applied to initialize the evaluation of the main factors contributing to groundwater composition. The parameters used for the analysis are major cations (Ca^{2+}, Mg^{2+}, Na^+, and K^+), major anions (HCO_3^-, Cl^-, and SO_4^{2-}), NH_4^+, NO_3^-, and Fe. The data is log-transformed and normalized before PCA processing. The principal components with eigenvalues greater than 0.95 are considered for the evaluation of processes controlling groundwater quality in the study area.

2.4. Base Exchange Index (BEX_D)

BEX_D was developed by Stuyfzand [13] to identify whether the state of groundwater is freshening, salinization, or equilibrium in aquifers containing dolomite. The BEX_D background is based on the cation exchange process when seawater intrusion or freshening occurs. The reaction is expressed as follows:

Seawater intrusion:

$Na^+ + 0.5Ca\text{-}X_2 \rightarrow Na\text{-}X + 0.5Ca^{2+}$ (Eqn. 1),

and freshening:

$0.5Ca^{2+} + Na\text{-}X \rightarrow 0.5Ca\text{-}X_2 + Na^+$ (Eqn. 2).

BEX_D calculates the deficit or surplus of (Na + K) from the contribution of seawater as follows:

$BEX_D = Na^+ + K^+ - 0.8768*Cl$ (in meq/L) (Eqn. 3),

Where, the factor 0.8768 is the ratio of (Na + K)/Cl in the mean seawater composition [14].

If:

- BEX_D is negative, groundwater is salinized with the conditions: $BEX_D < -(0.5 + 0.02*Cl)$ and $BEX_D < 1.5*(\sum Cation - \sum Anion)$;

- BEX_D is zero, groundwater is in equilibrium state with the conditions:

$-(0.5 + 0.02*Cl) < BEX_D < 1.5*(\sum Cation - \sum Anion)$; $abs(BEX_D + \{(\sum Cation - \sum Anion)/abs(\sum Cation - \sum Anion)\}* (0.5 + 0.02*Cl)) > 1.5*(\sum Cation - \sum Anion)$;

- BEX_D is positive, groundwater is freshened with the conditions: $BEX_D > (0.5 + 0.02*Cl)$ and $BEX_D > 1.5*(\sum Cation - \sum Anion)$.

3. Results and Discussion

3.1. Groundwater Quality

The groundwater chemical data is summarized in Table 1. In Holocene aquifer, most of groundwater samples (90%) were fresh with Cl^- concentration lower than the limitation of WHO drinking standard (250 mg/L). The groundwater samples with Cl^- exceeding WHO's standard for drinking water were observed in well QT9, which is located close to the coastal line and monitored at the depth of 50 m. The highest Cl^- observed in this well is 3883 mg/L in the dry season of 2013. In addition, the high Cl^- was also found in well QT8a in the dry seasons of 2011, 2012, 2014, and 2016 with the concentrations of 1581, 1205, 1319, and 302 mg/L. pH indicates the neutral groundwater in Holocene aquifer with an average of 7.41. In this aquifer, NO_3^- is below WHO's standard for drinking water (50 mg/L). The highest NO_3^- was observed at well QT8a with the concentration of 43.8 mg/L in the dry season of 2014. The highest NH_4^+ (15.1 mg/L) was also observed at this well at the same time. 75% of monitoring data of Holocene aquifer showed Fe above WHO's standard for drinking water.

Table 1. Descriptive statistics of monitoring data

Aquifer		Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	Cl^-	SO_4^{2-}	pH	NO_3^-	NH_4^+	Fe
Holocene	Min	2.20	0.37	1.80	0.63	0.00	3.55	0.10	6.15	0.01	0.01	0.02
	Max	196	204	3883	167	1940	5762	77.0	8.84	43.8	15.1	12.3
	Mean	23.3	18.1	265	14.6	213	395	8.82	7.41	3.49	0.13	1.61
	Std	20.3	40.2	858	31.5	413	1286	11.9	0.49	5.52	0.98	2.17
Pleistocene	Min	1.60	0.49	2.23	0.41	6.10	3.19	0.15	6.29	0.03	0.00	0.03
	Max	53.9	12.8	56.8	22.4	174	82.3	30.9	8.35	26.4	0.21	12.1
	Mean	9.80	3.54	15.0	5.31	51.9	22.7	5.04	7.34	2.92	0.05	2.19
	Std	8.34	2.37	10.1	5.50	36.2	15.7	6.14	0.43	4.01	0.04	3.11
Neogene	Min	4.41	1.46	8.19	3.12	27.5	8.51	0.10	6.50	0.01	0.01	0.00
	Max	178	124	1241	48.5	659	2340	79.6	8.70	31.3	2.60	9.64
	Mean	37.1	24.1	158	10.2	123	316	14.1	7.42	4.04	0.09	0.95
	Std	37.4	30.1	289	8.05	105	549	21.9	0.43	5.46	0.33	1.63
Unit: mg/L Std: standard deviation												

In Pleistocene aquifer, groundwaters showed a good quality. The highest Cl^- is 82.3 mg/L. NO_3^- was also detected but all samples were lower than 50 mg/L. NH_4^+ is low in this aquifer. However, 77% of the data indicated Fe concentration above WHO's standard for drinking water.

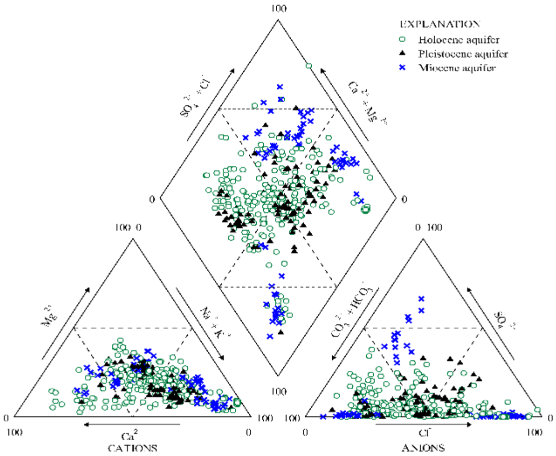

Figure 2. Piper diagram for groundwater types in Holocene, Pleistocene.

In the Neogene aquifer, the saline groundwater was observed in well QT6b with Cl^- concentration up to 2340 mg/L. The other wells are freshwater with Cl^- concentration from 8.51 to 241 mg/L. Although all monitoring wells in Neogene aquifer are at the depth of 100 m, NO_3^- was detected at some sampling periods. The NO_3^- was detected up to 31.3 mg/L at well QT6b in the dry season 2013. Fe concentration is the lowest among aquifers but 66% of data in Neogene aquifer is above WHO's standard for drinking water. In three aquifers, SO_4^{2-} is low during the monitoring period, even in the saline groundwater.

On the Piper diagram (Figure 2), the groundwater shows a variety of groundwater types. In Holocene aquifer, groundwater type varies with Ca-HCO_3, Na-Cl, and Na-HCO_3 types. The groundwater types in Pleistocene aquifer are mainly Ca-HCO_3 and MixNa-Cl. In Neogene aquifer, the groundwater is classified into three main types, which are Ca-HCO_3, MixCa-Cl, and Na-Cl.

3.2. Hydrogeochemical and Anthropogenic Processes

3.2.1. PCA Results

The PCA resulted in four principal components (PCs) with eigen values greater than 0.95 (Table 2) and explained 88% of the total variance. PC1, PC2, PC3, and PC4 explained 52%, 13.9%, 12.1%, and 10.1% of total variance. In PC1, the loading is high in Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, and HCO_3^-. High loading of NH_4^+ and NO_3^- is observed in PC2. PC3 had high loading of SO_4^{2-}, while the loading of Fe is significant in PC4. The chemical parameters with high loading in each PC are used for the evaluation of processes affecting groundwater quality in the study area.

Table 2. The loading of chemical parameters in different principal components

Variable	PC1	PC2	PC3	PC4
Na	**0.984**	-0.005	-0.054	0.017
K	**0.977**	0.019	-0.031	0.014
Ca	**0.614**	0.112	**0.484**	-0.035
Mg	**0.971**	0.033	0.048	0.006
NH4	0.082	**0.869**	-0.175	0.073
Cl	**0.993**	0.015	-0.041	0.009
SO4	-0.078	0.030	**0.915**	0.003
HCO3	**0.978**	-0.052	-0.019	0.017
NO3	-0.068	**0.788**	0.320	-0.080
Fe	0.016	0.004	-0.008	**0.997**
Eigen value	5.21	1.51	1.12	0.98
% of variance explained	52.0	13.9	12.1	10.1
Cumulative %	52.0	66.0	78.1	88.2

3.2.2. Salinization and Freshening Processes

Since Na^+, K^+, Ca^{2+}, Mg^{2+}, and Cl^- are concentrated in seawater, the high loading of those chemical parameters suggests the influence of seawater to the groundwater quality. The plot between ($Na^+ + K^+$) and Cl^- shows that there are samples plotted close to the seawater mixing line (Figure 3a). This means that the groundwater composition was influenced by the seawater mixing. However, there are samples plotted above and below the seawater mixing line, indicating the cation exchange process induced by the salinization and freshening. When the salinization or freshening occurs, Na increases or decreases and, thus, samples indicated the deviation (below or above) from the mixing line. To clarify the salinization and freshening process, BEX_D was applied. The results show that most of monitoring data (83%) is under freshening in Holocene aquifer indicated by positive BEX_D values. The highest extend of freshening is observed in the saline groundwater of well QT9, of which BEX_D is 34.5 meq/L (Table 3). The salinization is observed at well QT8a in dry season of 2012, 2014, and 2016 with BEX_D of -1.42, -0.93, and -1.77 meq/L. In addition, all wells are fresh and monitored at depth less than 40 m, well QT9 is saline and monitored depth of 50 m. These results suggest that the groundwater in Holocene aquifer is vulnerable to the salinization due to the up coning deriving from intensive groundwater abstraction. In Pleistocene aquifer, 82% of groundwaters shows positive BEX_D indicating freshening. The negative BEX_D is low with an average of -0.07 meq/L meaning that these groundwaters are not really in the state of salinization, instead of being close to the equilibrium state. In Neogene aquifer, the salinization is observed in well QT10b and QT6b, while the freshening is in well QT4b and QT7b.

Table 3. Statistical summary of BEX_D in three aquifers

	Holocene	Pleistocene	Neogene
Min	-2.83	-0.43	-6.04
Max	34.5	2.30	5.50
Mean	2.12	0.23	-0.69
Std	6.48	0.32	2.30
Unit: meq/L			
Std: standard deviation			

Since the groundwater in coastal aquifer has been experienced the transgressions and regressions of seawater in the past, the actual freshening or salinization can be masked due to the memory effects [13]. Hence, using BEX_D together with Cl^- of the monitoring data can more precisely identify the freshening and salinization. In well QT9, the saline groundwater showed the decrease of BEX_D and constant or decrease of Cl^- indicating that the saline groundwater is flushing out (Figure 4a). The variation of BEX_D is not significant through time in other wells monitoring of Holocene aquifer. The time series data of wells in Pleistocene aquifer also shows insignificant variation of BEX_D. In Neogene aquifer, the salinization was getting worse in well QT10b. The BEX_D is continuously decrease from -1.10 meq/L to -2.1 meq/L, while Cl^- increases from 117 mg/L to 198 mg/L in the period 2011 – 2018 (Figure 4b). In well QT6b, although individual sample indicate the salinization, the time series data suggests the occurrence of freshening. The BEX_D is significantly increased to less negative (from -5.23 meq/L to -1.86 meq/L in the period 2011 - 2018), and Cl^- drastically decreased from 2340 mg/L to 436 mg/L in the period 2011 - 2018 (Figure 4c).

In summary, the results of BEX_D clarified that most of the groundwaters in the study area are freshened, excepting groundwater in well QT10b. In addition, although the groundwater is on the way of flushing out, its quality can be readily changed due to the impact of saline groundwater intrusion.

3.2.3. Mineral Weathering and Dissolution

In PC1, HCO_3^- also has a high loading of 0.978 suggesting a source from carbonate mineral weathering and dissolution. The plot between (Ca^{2+} +Mg^{2+}) and HCO_3^- shows the influence of carbonate minerals dissolution in groundwater of Holocene and Pleistocene aquifer such as Calcite and Dolomite (Figure 3b). Since weathering and dissolution of carbonate minerals occurred, it would express 1:1 ratio of (Ca^{2+} + Mg^{2+})/HCO_3^- (in meq/L) as follows:

$$CaCO_3 + H^+ \rightarrow Ca^{2+} + HCO_3^- \qquad \text{(Eqn. 4)}$$

$$CaMg(CO_3)_2 + 2H^+ \rightarrow Ca^{2+} + Mg^{2+} + 2HCO_3^- \qquad \text{(Eqn. 5)}$$

In Neogene aquifer, the dissolution of Calcite is more obvious according to samples plotted along the 1:1 ratio line in the plot between Ca^{2+} and HCO_3 (Figure 3c). Samples plotted above and below the 1:1 ratio line indicate the addition or depletion of Ca (Figure 3c), which can be explained as a result of the cation exchange. This is the reason of lower loading of Ca^{2+} extracted from PC1. Furthermore, HCO_3^- also deviates from the 1:1 ratio line suggesting the effects of other processes.

Plot of Ca^{2+} and SO_4^{2-} shows that a part of samples is plotted along 1:1 ratio line (Figure 3d), suggesting the occurrence of the dissolution of sulfate minerals such as Gypsum and Anhydrite. The partly contribution of dissolution of sulfate minerals to the groundwater composition is reflected by the moderate loading of Ca^{2+} (0.484) in PC3.

3.2.4. Sulfate Reduction

The plot between SO_4^{2-} and Cl^- shows that samples are deviated above and below the seawater mixing line (Figure 3e), suggesting processes causing the addition and depletion of SO_4^{2-}. The additional SO_4^{2-} can be derived from the dissolution of sulfate minerals. Pyrite oxidation is also a process increasing SO_4^{2-} in groundwater. When pyrite oxidation occurs, it would be resulted in acidic groundwater [15]. However, the pH of groundwater in the study is from neutral to alkaline and, thus, the occurrence of pyrite oxidation is neglected. In the coastal aquifer, the anoxic condition promotes sulfate reduction, which removes SO_4^{2-} from the groundwater chemistry [3]. Figure 3e shows that the SO_4^{2-} concentration in saline groundwater is not much higher than those of fresh water. This observation means that SO_4^{2-} was strongly reduced in saline groundwater. The simple SO_4^{2-} reduction can be described as the following reaction:

$$SO_4^{2-} + 2CH_2O \rightarrow H_2S + 2HCO_3^- \text{ (Eqn. 6).}$$

According to Eqn. 6, sulfate reduction increases alkalinity in groundwater with 1:1 ratio in meq/L. In saline groundwater, SO_4^{2-} reduced produces a same amount of HCO_3^- explaining how SO_4^{2-} loading in PC1 is low although this PC indicates the influence of seawater, of which composition is high in SO_4^{2-} and low in HCO_3^-.

3.2.5. Reductive Dissolution of Fe (hydr)oxides

The monitoring data shows that 74% of groundwater samples have Fe concentration above WHO's standard for drinking water. PC4 also shows high loading of Fe (0.997) suggesting a role of process introducing Fe into groundwater. In coastal aquifers, the reductive dissolution of Fe (hydr)oxides is frequently observed in strong reducing environment [3]. The Fe concentration is varied in monitoring wells and sampling periods. In overall, the Fe is slightly higher in the dry season. Specifically, the Fe concentrations (on average±standard deviation) observed in the dry season are 2.03±2.51, 2.54±3.22, and 1.19±2.12 mg/L; while, in the rain season, are 1.25± 1.71, 1.81± 2.99, and 0.73± 0.81 in Holocene, Pleistocene, and Neogene aquifers, respectively.

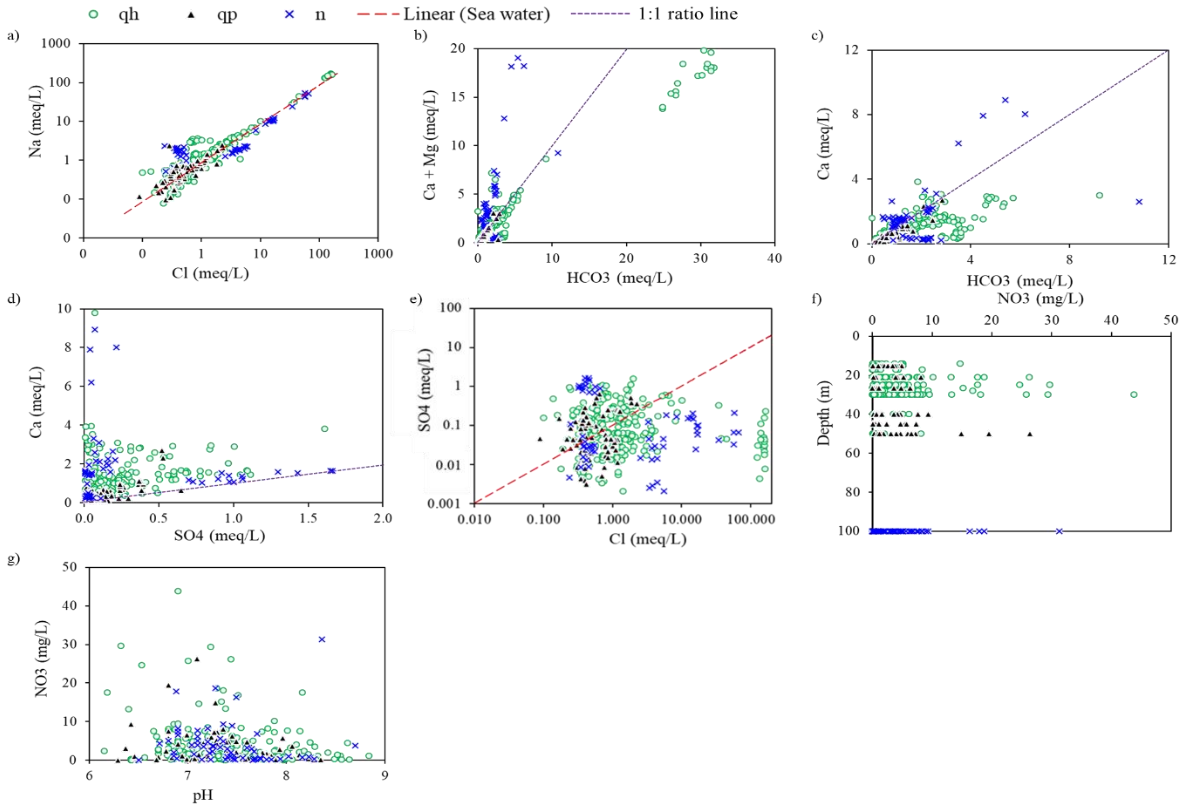

Figure 3. The plots of relation between (a) Na^+ and Cl^-, (b) ($Ca^{2+} + Mg^{2+}$) and Cl^-, (c) Ca^{2+} and HCO_3^-, (d) Ca^{2+} and SO_4^{2-}, (e) SO_4^{2-} and Cl^-, (f) NO_3^- and well depth, and (g) NO_3^- and pH.

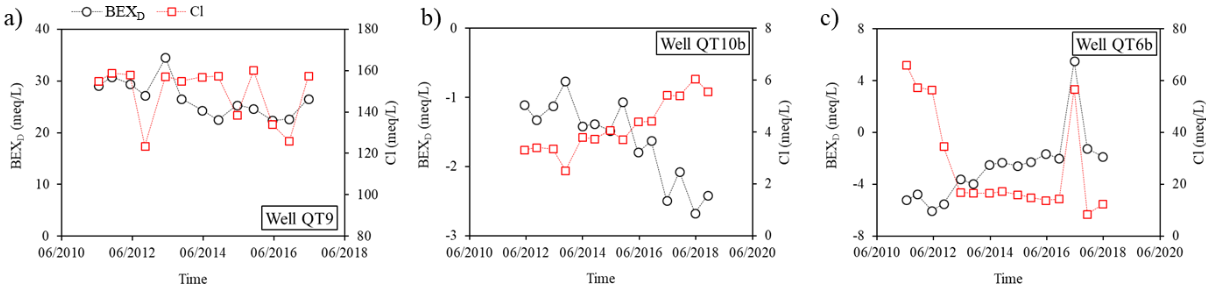

Figure 4. Changes of BEXD and Cl (2011 – 2018) in (a) well QT9, (b) well QT10b, and (c) well QT6b.

3.2.6. Anthropogenic Activities

The high loading of NO_3^- and NH_4^+ in PC2 suggests impacts of anthropogenic activities on the groundwater quality in the study area. Sources of NO_3^- and NH_4^+ are usually from the surface, such as domestic sewage, fertilizers, and industrial effluent. The high NO_3^- is detected in a shallow well with dept less than 30 m and mostly in Holocene aquifer (Figure 3f). This observation suggests the direct effects of human activities on the surface as the Holocene is exposed at some places in the study area. However, NO_3^- is also found in monitoring wells of Neogene aquifer at depth of 100 m with the concentration up to 31.3 mg/L (Figure 3f). The NO_3^- occurs in deep wells may relate to the leakage through the well cases. Hence, more care about nitrate contamination in groundwater is necessary as the study area is on the way of industrialization and urbanization.

In the case of NH_4^+, it is observed together with NO_3^- with low concentration (Table 1). The low NH_4^+ may be associated with the nitrification as simplified reaction as the following:

$$NH_4^+ + 2O_2 \rightarrow NO_3^- + 2H^+ + H_2O \quad \text{(Eqn. 7).}$$

Since the nitrification produces H^+, it decreases pH of groundwater. As a consequence, pH is lowered in groundwater with high NO_3^- concentration (Figure 3g). In addition, the groundwater is highly abstracted in the dry season. This activity has indirectly affected the groundwater quality due to causing the up coning of saline groundwater. The intensive groundwater abstraction also increases the drawdown of water level and, thus, accelerates the infiltration of contaminants from the surface into the groundwater.

4. Conclusion and Recommendation

The study expressed the importance of the groundwater monitoring network on groundwater management and protection. According to the analysis of monitoring data in QNDN area, various factors controlling groundwater quality were identified. The influences of seawater, mineral weathering and dissolution, sulfate reduction, and reductive dissolution of Fe (hydr)oxides are major natural processes controlling groundwater quality in the study area. The natural processes have caused the salinization and high Fe concentration in groundwater quality. On the other hand, anthropogenic activities indirectly generate the up coning of saline groundwater, which resulted in the Cl^- exceeding WHO's standard for drinking water. NO_3^- contamination is also a threat to groundwater quality due to the agricultural practices, industrialization, and urbanization in the study area. Based on the study's results, measures for groundwater management and protection can be efficiently made.

In addition, the utilization of monitoring data eliminated the "memory effect", which is usually masked by the seawater transgression and regression in the history, and, thus, gave the accurate interpretation of freshening and salinization processes.

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