



## Assessing major factors affecting shallow groundwater geochemical evolution in a highly urbanized coastal area of Shenzhen City, China



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### ABSTRACT

Groundwater is the most important alternative drinking water source in the coastal urban area of Shenzhen City, China. Understanding the main geochemical factors affecting groundwater chemistry is vital for sustainable water resource management in the long term. Seawater intrusion has been a serious problem, but the situation seems to have recently improved with the establishment of a water supply system and land reclamation. However, total dissolved solids (TDS) concentrations in the groundwater have increased. To determine major factors controlling groundwater geochemical evolution, studies were carried out in the area in Shenzhen most vulnerable to seawater intrusion. Along the groundwater flow path from the bedrock outcrop area to the coastal plain, groundwater evolution processes were studied using multiple hydrogeochemical analyses. Piper plots and hydrochemical facies evolution diagrams show the aquifer vulnerable to seawater intrusion is currently dominated by fresh groundwater of Ca-HCO<sub>3</sub> or Ca-Na-HCO<sub>3</sub>-Cl type; only a few locations close to Shenzhen Bay appear to be Na-Cl type, indicating the threat of seawater intrusion. Anthropogenic pollution is mainly characterized by elevated NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in the form of point sources. Correlation coefficient analysis to evaluate water-rock interactions during granite weathering indicates this process can contribute significant amounts of Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and HCO<sub>3</sub><sup>-</sup> to groundwater. Evaluation of the aggressiveness index (AI), Langelier saturation index (LSI), and Ryznar stability index (RSI) suggest all groundwaters are corrosive and the dissolution of concrete material foundations in urban areas contributes significant amounts of Ca<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> to groundwater. Principal components analysis (PCA) demonstrates water-rock interactions and concrete material dissolution are more important factors than seawater intrusion in terms of affecting shallow groundwater chemistry. This study demonstrates that hydrogeochemical tools are effective in assessing groundwater evolution under the influence of multiple factors in a highly urbanized coastal area.

### 1. Introduction

Higher population densities and living standards in coastal urban areas cause ever increasing demands for fresh water for industrial and domestic use. Because surface water resources are limited, groundwater is playing a more and more important role in water resource supply (Kundzewicz and Doll, 2009; Vandenbohede et al., 2009; Bouwer, 2014). The quality of groundwater can be affected by various geochemical factors in coastal areas, such as seawater intrusion (Giambastiani et al., 2013; Park et al., 2012), land reclamation (Jiao et al., 2006; Chen and Jiao, 2007a, 2007b), wastewater infiltration (Schmidt et al., 2013; Nyenje et al., 2014), sewage exfiltration (Reynolds and Barrett, 2007; Sercu et al., 2011), etc. However, with

complicated influences from multiple anthropogenic and natural processes in some coastal urban areas, identifying each affecting factor can be difficult. Moreover, in some cases only shallow groundwater can be acquired in highly urbanized areas. Effective ways to reveal aquifer status are valuable for achieving integrated and sustainable groundwater resource management.

Shenzhen City is a highly urbanized coastal city located in southern China. The population of 11.37 million consumes water resources of  $2.0 \times 10^9$  m<sup>3</sup> per year. Seawater intrusion has been strongly attacking the local shallow aquifer since 1980 when extensive urbanization began (Fan, 2004; Han et al., 2009; Zhao et al., 2009). In the early 1990s, the status of seawater intrusion began to improve because of the establishment of a public water supply system and large-scale land

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reclamation (Chen and Jiao, 2007a, 2007b; Yu and Feng, 2015). As a result, gradual groundwater freshening has been suggested to be occurring in Shenzhen (Yang et al., 2010; Yin et al., 2011; Lu et al., 2013); however, groundwater salinity data are not consistent with the suggested freshening trend. For example, the recent average total dissolved solids (TDS) concentration in shallow groundwater near the coast of 857.6 mg/L is much greater than the previous average of 71.4 mg/L (Chen and Jiao, 2007b; Yin et al., 2011). The reasons for the increasing groundwater salinity remain unclear as do the processes relevant to the geochemical evolution of this groundwater.

This study aims to assess both natural and anthropogenic factors for a coastal aquifer of Shenzhen under the extensive influence of urbanization. The Futian district, one of the most typical urban areas in Shenzhen and the most vulnerable to seawater intrusion, was chosen for study. Groundwater samples from shallow aquifers (1–6 m deep) were collected and analyzed using hydrogeochemical methods. Hydrochemical parameters were obtained to 1) study the hydrochemical characteristics and types of groundwater, 2) estimate the current influences and understand the evolution of seawater intrusion, and 3) identify other main geochemical processes that affect shallow groundwater composition. The methods adopted and results obtained from this study are helpful for identifying the current contamination status of shallow groundwater and provide a better understanding of the potential threats to local groundwater resources. This study also provides useful reference information for sustainable management of aquifers in other highly urbanized coastal areas.

## 2. Study area

Shenzhen is a mega-city with a total area of 1996.85 km<sup>2</sup>. It is situated in the Pearl River Delta (PRD), with Hong Kong to the south. Influenced by the East Asian Monsoon system, Shenzhen has a subtropical maritime monsoon climate with a mean annual temperature of 22.4 °C and is characterized by two distinct seasons: a wet hot summer from April to September, and a dry mild winter from October to March. The wet season generally contributes nearly 85–90% of the annual 1933 mm of rainfall (Yang and Duan, 2015). The Futian district is located in the southwest of Shenzhen, adjacent to Shenzhen Bay (Fig. 1). The study area can be divided into two parts based on the level of urbanization: a slope area and an urban area (Fig. 1). The slope area is located in the north part, with Meilin slope (elevation 300 m) being the highest feature. Granite from the Jurassic Yanshanian orogeny dominates the slope area. The urban area in the south part is covered with Quaternary materials, including gravels, sands, silts, and clays that range from 2 to 19 m in thickness (Fig. 1). The average infiltration coefficient of the study area is about 0.221, and the total groundwater runoff is reported to be 15,919.20 m<sup>3</sup>/d (Yang and Duan, 2015).

## 3. Methods

Shallow groundwater samples were all taken from monitoring wells with a groundwater sampler from a depth of about 5 m, which is about 1 m below the water table. To investigate seasonal effects on groundwater chemistry, samples were collected in both the wet (June 2015) and dry (January 2016) seasons. Due to management problems, some monitoring wells were inaccessible in the dry season and, therefore, only wet season samples were collected. Groundwater samples from some new boreholes were also collected. In total, 22 samples were taken in the wet season and 24 samples in the dry season. All sampling locations are shown in Fig. 1.

Oxidation-reduction potential (ORP), pH, and temperature were measured by a pH/ORP meter (Hanna HI98121); electrical conductivity (EC) and total dissolved solids (TDS) were measured by portable testing instruments (Hanna HI9034 and HI8733, respectively); and alkalinity as CaCO<sub>3</sub> was titrated using a Hach digital titrator (Model 16900). Hana HI96728 and HI96707 meters were used to measure NO<sub>3</sub><sup>-</sup>-N and

NO<sub>2</sub><sup>-</sup>-N, respectively. Medium range Hana HI96715 (0.00 to 9.99 mg/L) and high range Hana HI96733 (0.0 to 50.0 mg/L) meters were used for NH<sub>4</sub><sup>+</sup>-N measurements. All of the above parameters were measured in the field. Samples for laboratory water chemical analyses were filtered by passing through 0.45-μm filter paper and collected in 125-mL HDPE bottles. Water samples were preserved in a cooler with frozen blue ice in the field and stored in a refrigerator at about 4 °C in the lab. Major ions (e.g., Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>) were determined using an ICS-900 ion chromatograph at the School of Environmental Science and Engineering, Sun Yat-sen University. For quality control, replicate samples and samples with known concentrations were also analyzed.

## 4. Results and discussion

The groundwater table is generally 1.5 to 5.6 m below ground surface, with average depths of 3.9 and 4.0 m in the wet and dry seasons, respectively (Table 1). Shallow groundwater flow in the study area is generally topography driven, flowing from the slope bedrock outcrop area to the urban coastal plain (Fig. 2). Shallow groundwater in the urban area exhibits significantly higher ion concentrations and TDS than that in the slope area, suggesting that the affecting factors could be more complicated in the urban area. Note that additional data from a previous study (Yang and Duan, 2015) were added to our figures to enhance the analysis.

### 4.1. Seawater intrusion

Shenzhen is vulnerable to seawater intrusion, but many studies show that the aquifer beneath Shenzhen is experiencing refreshing as a consequence of recent efforts with respect to public water supply and coastal reclamation (Chen and Jiao, 2007a, 2007b; Yu and Feng, 2015). Shallow groundwater in the study area is enriched in Na<sup>+</sup>, Ca<sup>2+</sup>, HCO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and Cl<sup>-</sup> (Table 1), which are also significant components of typical seawater (Goldberg et al., 1971). Among these major ions, Cl<sup>-</sup> is the most conservative and representative. Cl<sup>-</sup> normally does not take part in any chemical reactions or water-rock interactions and, therefore, is often regarded as a tracer of seawater and used to identify the degree of seawater intrusion in coastal aquifers (Pulido-Leboeuf, 2004; Appelo and Postma, 2009; Park et al., 2012). Fig. 3 shows the relations between Cl<sup>-</sup> and other major ions (Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>) as well as between Cl<sup>-</sup> and TDS. All of the samples from the slope area concentrate around the freshwater end-member and have no obvious relation with the theoretical mixing line. This suggests that the relatively high altitude makes seawater intrusion unlikely to occur in the slope area. In the urban area, results with higher TDS align on or close to the theoretical mixing line between seawater and fresh groundwater, indicating the increasing TDS of those samples is due to the influence of seawater. Most samples with much lower TDS and Cl<sup>-</sup> locate far from the end member of seawater, indicating a limited influence of seawater intrusion.

To understand the status of groundwater influenced by seawater in the shallow groundwater, a Piper plot was created (Fig. 4).

According to Fig. 4, the types of shallow groundwater in the study area do not change significantly with season. Furthermore, most samples are located on or near the mixing line between fresh groundwater and seawater, and more data concentrate around the freshwater-type, indicating the seawater influence is no longer considerable for the shallow aquifer throughout the entire study area. However, some locations still indicate the characteristics of seawater intrusion.

Generally, five groundwater types can be identified: Ca-HCO<sub>3</sub>, Ca-Na-HCO<sub>3</sub>-Cl, Na-Ca-Cl-HCO<sub>3</sub>, Na-HCO<sub>3</sub>-Cl, and Na-Cl. Among them, Ca-HCO<sub>3</sub> is typical fresh groundwater type and Na-Cl is typical seawater type (Appelo and Postma, 2009); Na-HCO<sub>3</sub>-Cl is the typical groundwater type in freshening aquifers (Chen and Jiao, 2007a; Gimenez-Forcada, 2014); and Ca-Na-HCO<sub>3</sub>-Cl and Na-Ca-Cl-HCO<sub>3</sub> groundwater

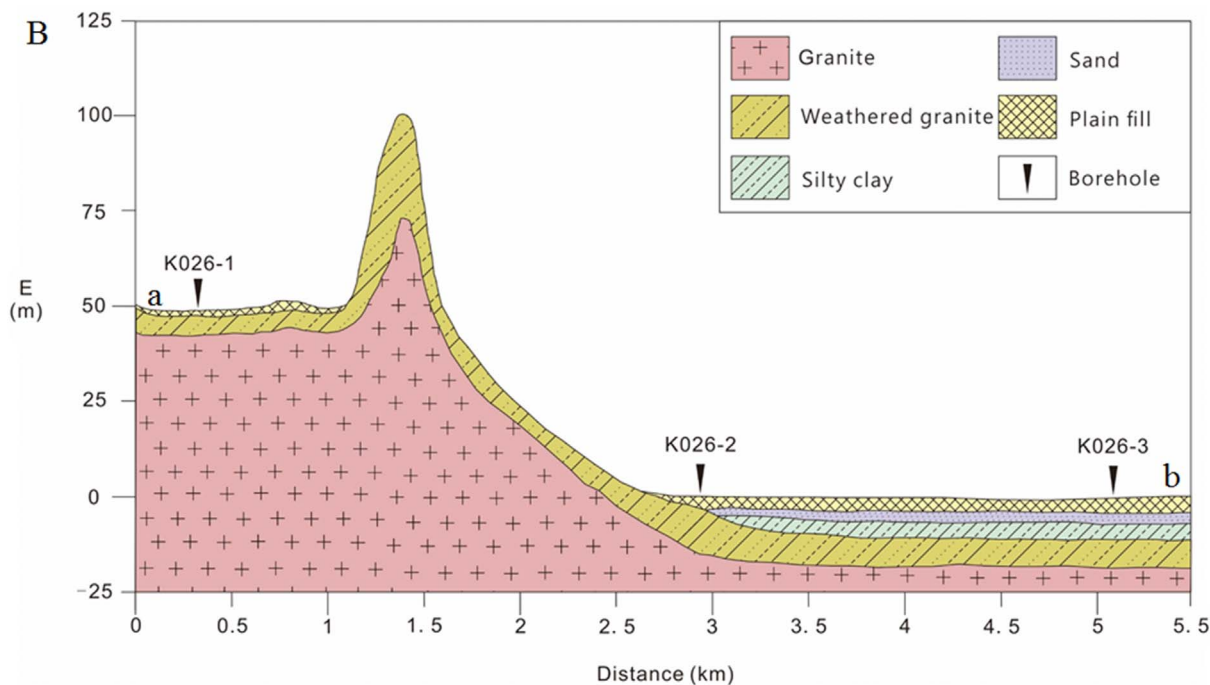
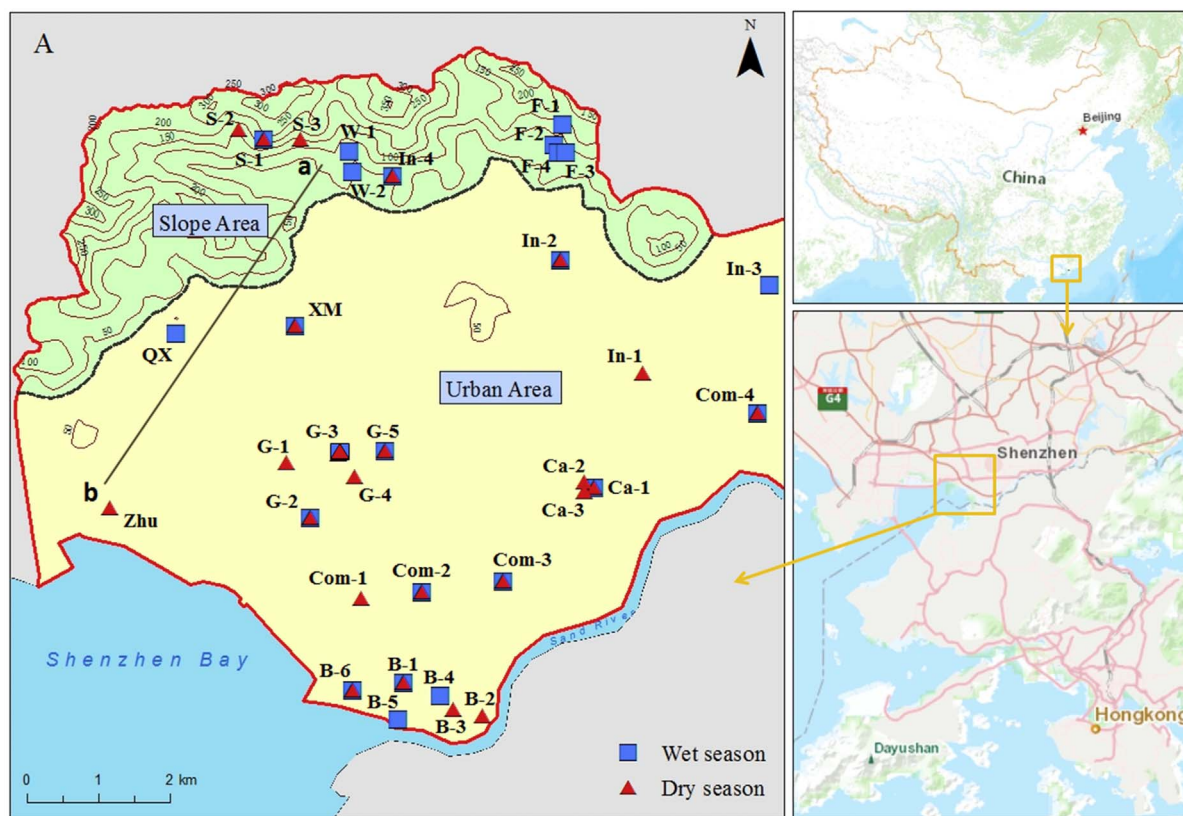


Fig. 1. (A) Map of the study area and sampling locations. Red lines represent the study area of Futian district. Squares represent the sampling locations of wet season and triangles represent the sampling locations of dry season, and the corresponding Sample ID is provided for reference. (B) Cross-section from point a to b in (A). The line marked off the bedrock and sediments of the study area. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

types suggest seawater influences (mixing), with similar groundwater types also found in other aquifers influenced by seawater (Wang and Jiao, 2012; Chandrajith et al., 2016). Samples with the most elevated  $\text{Cl}^-$  ( $> 3200 \text{ mg/L}$  in the wet season) locate very close to Shenzhen Bay are identified as Na-Cl type, indicating the influence of seawater.

To verify the seawater influence analysis from the Piper plot and

achieve a better understanding of the status of the shallow aquifer, a hydrochemical facies evolution diagram (HFE) was also adopted for analysis. HFE diagrams are a new type of analysis diagram that give possible hydrochemical facies and evolution stages considering the percentage of the most significant ions as well as their relationships (Gimenez-Forcada, 2014; Gimenez-Forcada and Román, 2015). Fig. 5



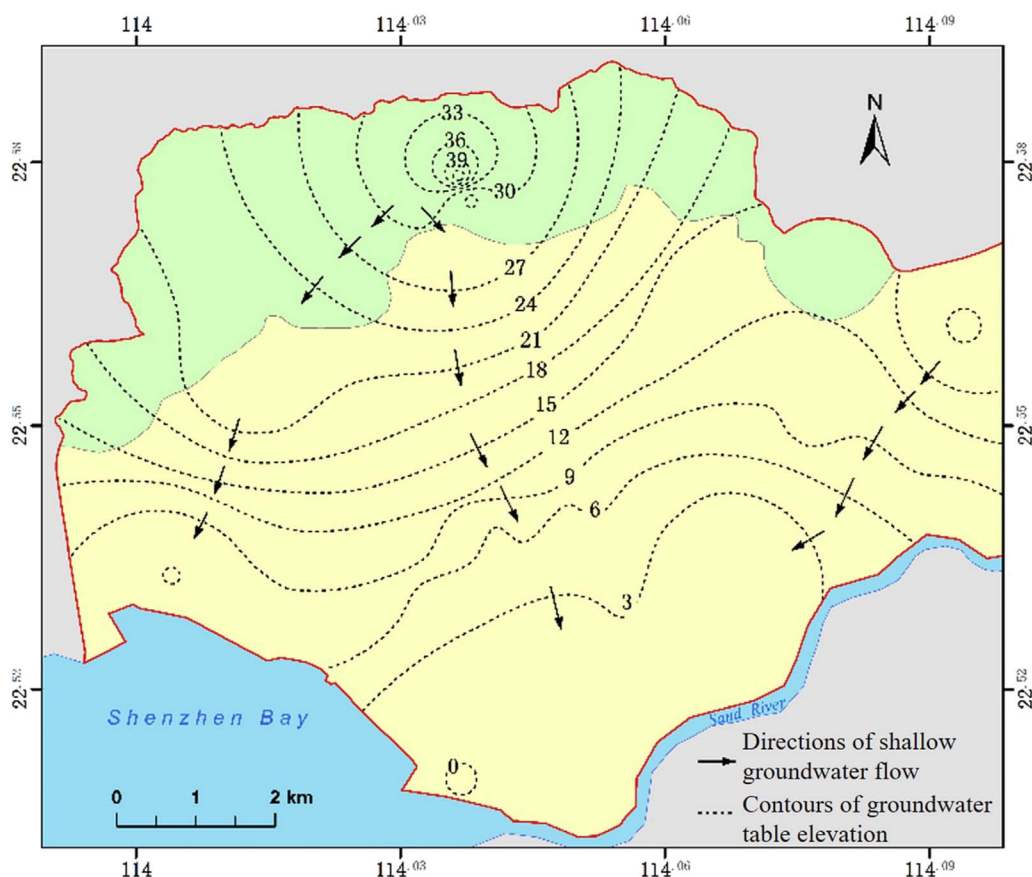


Fig. 2. Contours of groundwater table elevation and directions of shallow groundwater flow. Generally, shallow groundwater flows from the north to the south in the study area.

2009 to 2015, the Cl<sup>-</sup> concentration decreased by 50.6% but TDS increased by 48.8% (Yin et al., 2011). Normally, weakening of seawater intrusion and groundwater refreshing should result in a simultaneous decrease in Cl<sup>-</sup> and TDS. Therefore, their opposing trends indicate that other factors must also significantly affect the local shallow groundwater chemistry.

#### 4.2. Precipitation infiltration

Rainwater is one possible factor influencing the shallow groundwater. Shenzhen suffers from acid rain due to the dissolution of NO<sub>2</sub> and SO<sub>2</sub> emitted from motor vehicles and coal-burning power stations, among other sources. The mean pH and TDS of rainwater from 2005 to 2009 were 4.56 and 6.84 mg/L, respectively (Huang et al., 2010), while the mean rainwater pH and TDS in 2011 were 4.99 and 5.80 mg/L, respectively (Qin et al., 2015). The major source of some rainwater ions, e.g., SO<sub>4</sub><sup>2-</sup>, Ca<sup>2+</sup>, NO<sub>3</sub><sup>-</sup>, and K<sup>+</sup>, is the dissolution of aerosols, particulates, or dust from the atmosphere, while the origin of other ions, e.g., Cl<sup>-</sup>, Na<sup>+</sup>, and Mg<sup>2+</sup>, is marine due to the “near-to-coast” locality of the area (Huang et al., 2010).

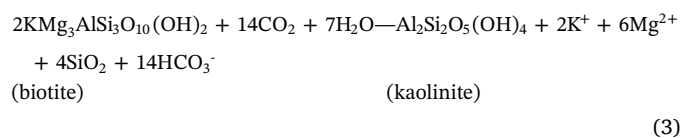
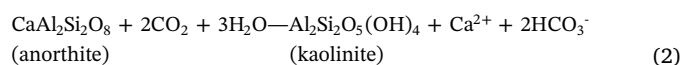
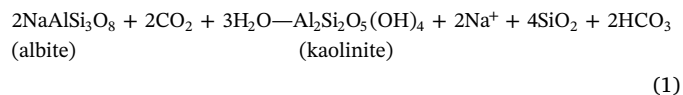
Acid rainwater may influence shallow groundwater by contributing ions. In the study area, the average groundwater pH in the wet season is 7.01 in the slope area and 7.37 in the urban area; in the dry season, the groundwater pH in the slope area increases to 7.38 but in the urban area decreases to 6.94. Groundwater samples collected from the study area generally have a near neutral pH, which suggests limited rainwater influence.

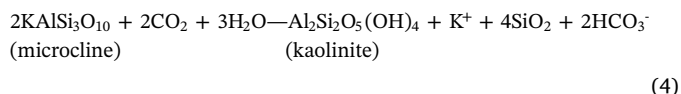
Furthermore, TDS in groundwater is generally higher in the wet season (248.8 mg/L in the slope area and 1136.9 mg/L in the urban area) than in the dry season (39.7 mg/L in the slope area and 334.5 mg/L in the urban area). As noted above, the TDS of rainwater is typically < 10 mg/L, so more infiltration of rainwater into shallow aquifers

in the wet season should dilute the groundwater and result in lower TDS values than in the dry season. However, TDS values in the shallow groundwater are the opposite of what would be expected in the wet and dry season; this suggests that rainwater plays a relatively small role in controlling the composition of groundwater chemistry in the study area compared to other factors.

#### 4.3. Water-rock interactions during granite weathering

Water-rock interactions such as ion exchange processes, mineral dissolution, adsorption, and absorption can progressively modify groundwater chemistry along groundwater flow paths (Leung et al., 2005; Wang and Jiao, 2012). Granite is the most widely distributed bedrock in the study area, and its major minerals (e.g., alkali feldspar, plagioclase, biotite, silica) can contribute specific ions to groundwater due to infiltrating precipitation and weathering (SG, 2009; Cui et al., 2016). Weathering of these minerals occurs as follows:





The above reactions show that granite weathering processes can contribute  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{HCO}_3^-$  to groundwater. Among them,  $\text{HCO}_3^-$  is the common reaction product. Therefore,  $\text{HCO}_3^-$  could be regarded as the representative ion for granite weathering, and has been used as such in a previous study (Leung et al., 2005). To illustrate the relationship between major ion chemistry of groundwater and granite weathering, correlations among  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , and

$\text{HCO}_3^-$  were analyzed using the SPSS 19 software package. Correlation coefficients were not calculated for different seasons but the slope and urban areas were considered separately. This is reasonable because Piper plots show that shallow groundwater types of the study area are similar in both the wet and dry seasons, suggesting major influencing factors do not significantly change with season. All concentration data were normalized and converted to standard Z-scores before statistical analyses (Davis, 2002). Correlation coefficients determined for the slope area and urban area are shown in Tables 2 and 3, respectively.

The results in Table 2 demonstrate that  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ , and  $\text{K}^+$  are all positively correlated with  $\text{HCO}_3^-$ , with correlation coefficients

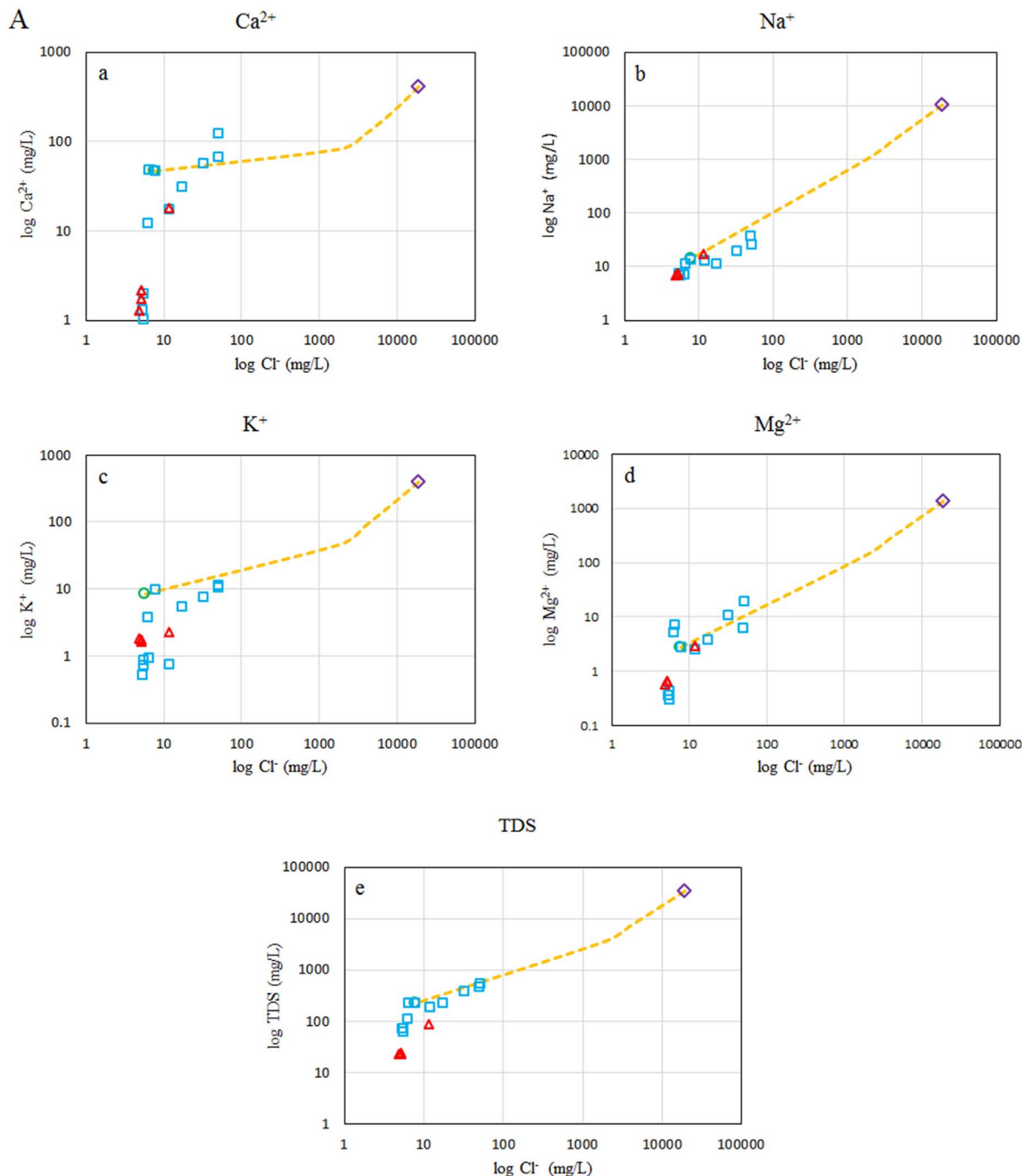


Fig. 3. Relationship between 1)  $\text{Ca}^{2+}$  and  $\text{Cl}^-$ , 2)  $\text{Na}^+$  and  $\text{Cl}^-$ , 3)  $\text{K}^+$  and  $\text{Cl}^-$ , 4)  $\text{Mg}^{2+}$  and  $\text{Cl}^-$ , 5) TDS and  $\text{Cl}^-$  in the urban area. A shows the results of slope area and B shows the results of urban area. Squares represent samples from wet season and triangles represent samples of dry season. The dotted line represents the theoretical mixing line between seawater and fresh groundwater, and the rhombus represents the end member of seawater while the circle represents the end member of freshwater.

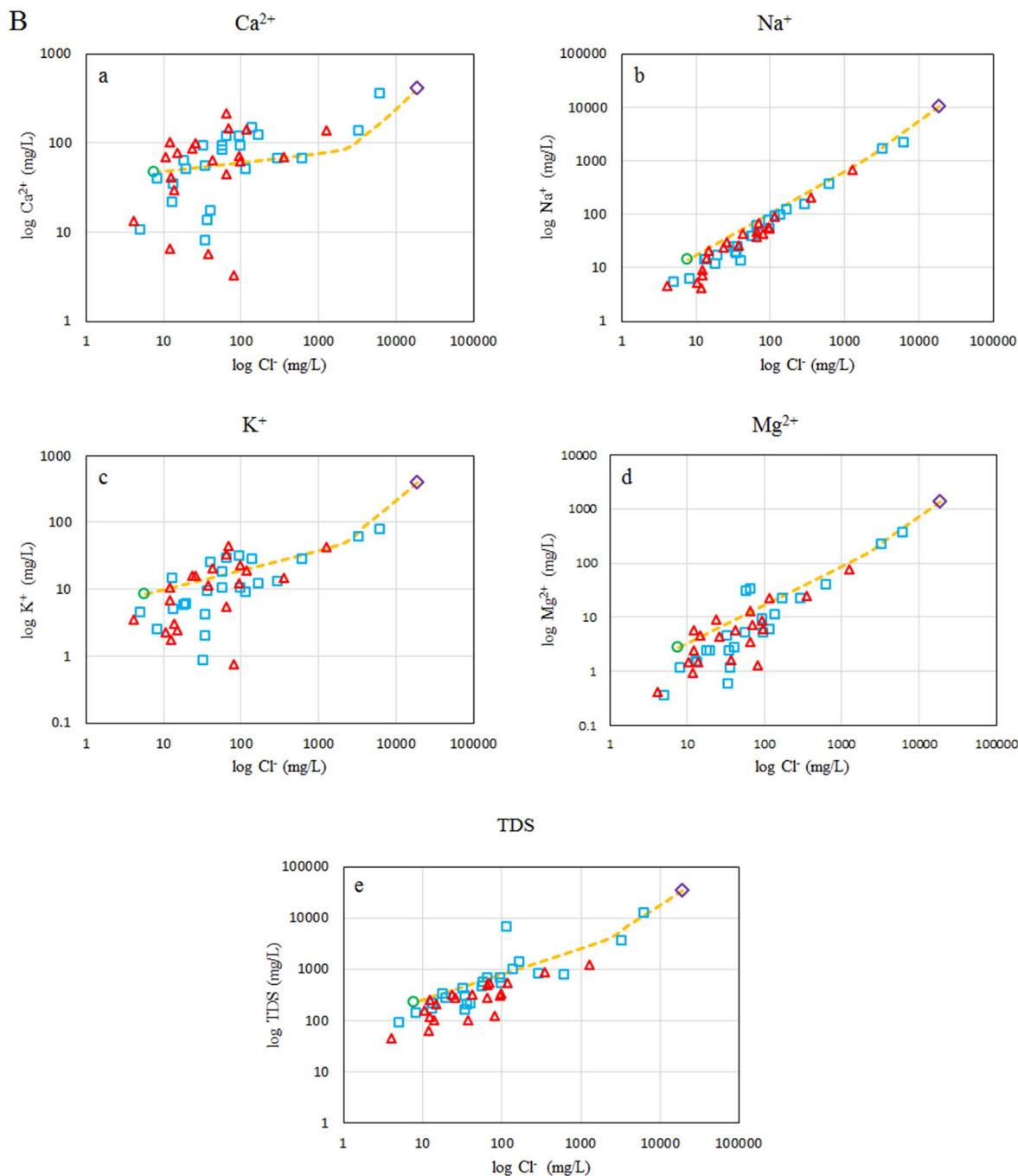


Fig. 3. (continued)

of 0.974, 0.882, 0.749, and 0.803, respectively, in the slope area. However, the results in Table 3 show that corresponding correlation coefficients in the urban area are much lower, at 0.572, 0.300, 0.296, and 0.416, respectively. This indicates that the concentrations of these five major ions are significantly controlled by granite weathering, which is relevant in the slope area but less significant in the urban area. Moreover, these five ions are more enriched in groundwater in the wet season than in the dry season, especially in the slope area (Table 1); this is attributed to the much more abundant precipitation and higher groundwater table during the wet season, which can cause much more intensive granite weathering and water-rock reactions and consequently lead to the dissolution of more minerals.

Furthermore, reaction Eqs. (1)–(4) imply that  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^{+}$ ,  $\text{K}^{+}$ , and  $\text{HCO}_3^{-}$  are largely derived from the weathering of minerals,

including albite, anorthite, biotite, and microcline. Albite and anorthite are usually the first primary minerals to weather significantly, followed by microcline and biotite at later stages, and finally quartz, which is the most stable (Cui et al., 2016). This is consistent with our findings, where the correlation between  $\text{Ca}^{2+}$  and  $\text{HCO}_3^{-}$  ( $r = 0.974$ ) is stronger than that between  $\text{Mg}^{2+}$  and  $\text{HCO}_3^{-}$  ( $r = 0.882$ ) or  $\text{K}^{+}$  and  $\text{HCO}_3^{-}$  ( $r = 0.803$ ) (Table 2). Weathering processes should produce similar concentrations of  $\text{Na}^{+}$  ( $r = 0.749$ ) and  $\text{Ca}^{2+}$ , so the less significant correlation between  $\text{Na}^{+}$  and  $\text{HCO}_3^{-}$  suggests that the concentration of  $\text{Na}^{+}$  is also influenced by other processes.

Granite weathering also occurs in the urban area, as indicated by increasing  $\text{HCO}_3^{-}$  concentrations, from 161.5 to 240.4 mg/L and from 29 to 216.9 mg/L moving from the slope to urban area in the wet and dry seasons, respectively (Table 1), but its influence is masked by other

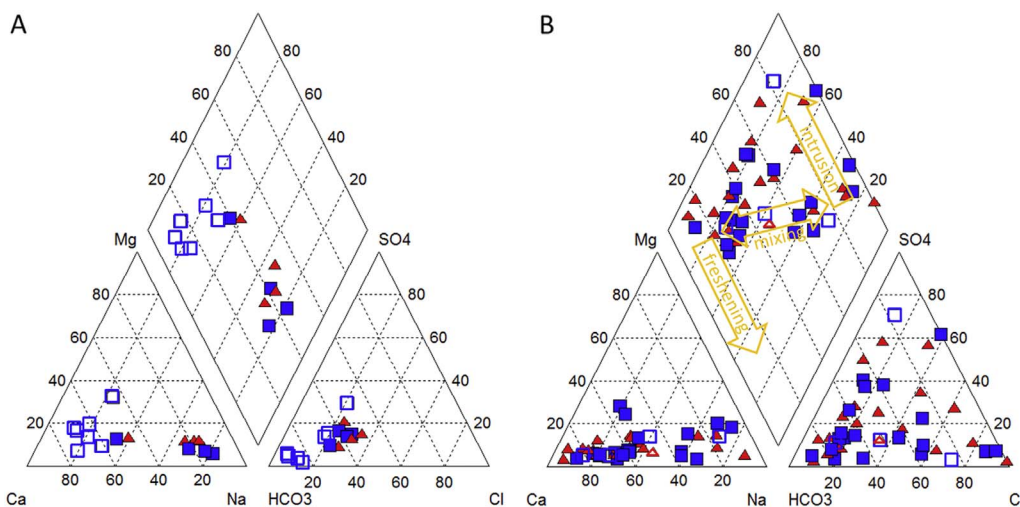


Fig. 4. Piper plots of the groundwater data for slope area (A) and urban area (B) in both wet (squares) and dry (triangles) seasons. Overlapped samples from both wet and dry seasons are filled, and samples from only one season are unfilled.

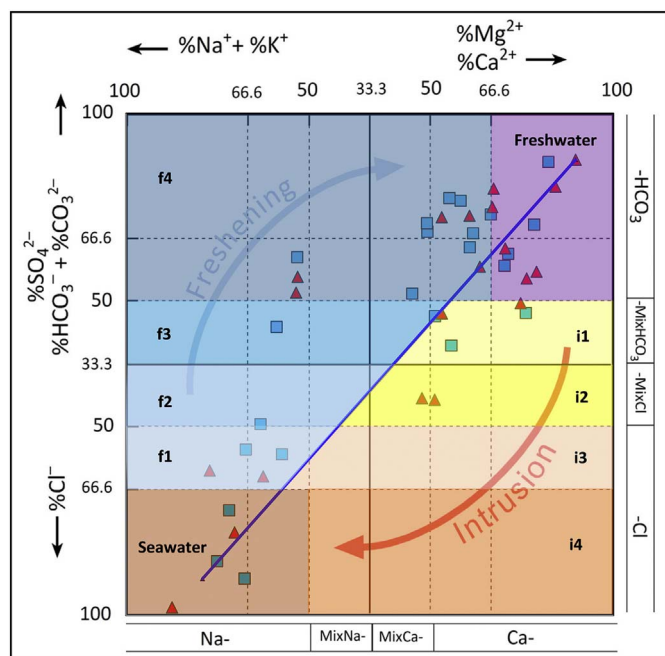


Fig. 5. Hydrochemical facies evolution diagram (HFE). Areas with different colors represent different hydrochemical facies during the mixing of fresh water and seawater. f1–f4 indicate freshening evolution from seawater to fresh water with f1 (Mix Na–Cl), f2 (Mix Na–Mix Cl), f3 (Mix Ca–MixHCO<sub>3</sub> and Ca–MixHCO<sub>3</sub>) and f4 (Ca–HCO<sub>3</sub>). i1–i4 indicate intrusion evolution from fresh water to seawater with i1 (Ca–MixHCO<sub>3</sub>), i2 (Mix Ca–Mix Cl, Ca–Mix Cl), i3 (Mix Na–Cl, Mix Ca–Cl, Ca–Cl) and i4 (Mix Na–Cl). Blue squares represent samples from wet season and red triangles represent the dry season. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

factors that cause the large increase in average TDS (from 248.8 to 1136.9 mg/L in the wet season and from 39.7 to 334.5 mg/L in the dry season). These urban area factors are discussed in a subsequent section.

#### 4.4. Anthropogenic pollution

Hydrological systems of the study area can be influenced by anthropogenic pollution, and so the related effects on shallow groundwater should be taken into consideration. Anthropogenic impacts are generally characterized by high levels of inorganic nitrogen, including

ammonium (NH<sub>4</sub><sup>+</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and nitrite (NO<sub>2</sub><sup>-</sup>) ions (Chan, 2001; Bottrell et al., 2008; Strauch et al., 2008). As shown in Table 1, concentrations of inorganic nitrogen (NH<sub>4</sub><sup>+</sup> or NO<sub>3</sub><sup>-</sup>) in some samples are > 20 mg/L, which suggests anthropogenic pollution is an important factor affecting groundwater chemistry in the form of point sources. The form of inorganic nitrogen can be altered via nitrification and denitrification processes. Nitrification occurs under aerobic conditions and can oxidize NH<sub>4</sub><sup>+</sup> to NO<sub>3</sub><sup>-</sup>; denitrification occurs with the consumption of oxygen and reduces NO<sub>3</sub><sup>-</sup> to N<sub>2</sub>, which could cause inorganic nitrogen loss (as N<sub>2(gas)</sub>) from the groundwater; both processes produce NO<sub>2</sub><sup>-</sup> as the intermediate species (Santoro, 2010; Russak et al., 2015). In the wet season, the average concentration of NO<sub>3</sub><sup>-</sup> is 8.83 mg/L in the slope area and 7.22 mg/L in the urban area, both of which are higher than concentrations of NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub><sup>-</sup> (Table 1) and so suggest that nitrification is the dominant reaction in both areas. In the dry season, the NO<sub>3</sub><sup>-</sup> concentration (5.98 mg/L) is higher than the NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub><sup>-</sup> concentrations in the slope area, but the concentration of NH<sub>4</sub><sup>+</sup> (5.62 mg/L) is far greater than the NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> concentrations in the urban area (Table 1). These inorganic nitrogen concentrations suggest that nitrification is less relevant in the dry season than in the wet season, especially in the urban area. Therefore, the concentration of NH<sub>4</sub><sup>+</sup> is higher in the urban area and almost none of the intermediate species (NO<sub>2</sub><sup>-</sup>) is found in either area in the dry season. The difference between nitrification in the wet and dry seasons is attributed to precipitation. In the wet season, more precipitation brings more oxygen to the groundwater, and therefore the nitrification process predominates; in the dry season, less oxygen is introduced into the groundwater by precipitation, most oxygen is consumed in the slope area, and therefore much less oxygen is available for nitrification in the urban area. Consequently, the dominant form of inorganic nitrogen in groundwater during the wet season in both areas is NO<sub>3</sub><sup>-</sup>, while for dry season is NO<sub>3</sub><sup>-</sup> in the slope area but NH<sub>4</sub><sup>+</sup> in the urban area. As only inorganic nitrogen concentrations (NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>) were identified in this study, it is not possible to confirm the specific sources of the anthropogenic pollution (e.g. septic-tank leachate or domestic sewage) to the groundwater. Stable inorganic nitrogen isotope studies would be carried out in the future to address this issue.

#### 4.5. Dissolution of concrete

Urban areas are crowded with high-rise buildings containing basements and deep foundations (with piers anchored 5 m below the rock head) constructed from concrete. These construction materials are typically situated in situ in Quaternary deposits or rocks and are permanently or periodically below the groundwater table. When



**Table 2**  
Correlation coefficients among major ions in shallow groundwater in slope area (n = 15).

	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	HCO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>
Ca <sup>2+</sup>	1	0.932**	0.774**	0.815**	0.974**	0.866**	0.494	0.120
Mg <sup>2+</sup>		1	0.622*	0.664**	0.882**	0.801**	0.433	0.172
Na <sup>+</sup>			1	0.800**	0.749**	0.919**	0.766**	0.340
K <sup>+</sup>				1	0.803**	0.816**	0.625*	0.105
HCO <sub>3</sub> <sup>-</sup>					1	0.833**	0.361	-0.058
Cl <sup>-</sup>						1	0.711**	0.275
SO <sub>4</sub> <sup>2-</sup>							1	0.579*
NO <sub>3</sub> <sup>-</sup>								1

\*\* Represents correlation is significant at the 0.01 level (2-tailed).

\* Represents correlation is significant at the 0.05 level (2-tailed).

**Table 3**  
Correlation coefficients among major ions in shallow groundwater in urban area (n = 44).

	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	HCO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>
Ca <sup>2+</sup>	1	0.713**	0.665**	0.662**	0.572**	0.693**	0.836**	0.128
Mg <sup>2+</sup>		1	0.987**	0.711**	0.300*	0.993**	0.827**	0.335*
Na <sup>+</sup>			1	0.728**	0.296	0.984**	0.796**	0.301*
K <sup>+</sup>				1	0.416**	0.704**	0.597**	0.211
HCO <sub>3</sub> <sup>-</sup>					1	0.237	0.294	0.06
Cl <sup>-</sup>						1	0.808**	0.323*
SO <sub>4</sub> <sup>2-</sup>							1	0.202
NO <sub>3</sub> <sup>-</sup>								1

\*\* Represents correlation is significant at the 0.01 level (2-tailed).

\* Represents correlation is significant at the 0.05 level (2-tailed).

surrounded and soaked by groundwater, concrete slowly degenerates and leaches compounds including calcium hydroxide. Because Ca(OH)<sub>2</sub> is readily soluble in water to about 1200 mg/L, it can be regarded as an important Ca<sup>2+</sup> source in urban areas (Hawkins, 1997; Leung et al., 2005). Sulphates are also present in cement in the form of gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O), hemihydrate (bassanite, CaSO<sub>4</sub>·0.5H<sub>2</sub>O), and anhydrite (CaSO<sub>4</sub>) (Ylmén et al., 2009), and therefore can also leach out as SO<sub>4</sub><sup>2-</sup>.

The potential aggressiveness of groundwater towards concrete is assessed by three widely used indices: the aggressiveness index (AI), the Langelier saturation index (LSI), and the Ryznar stability index (RSI) (Hawkins, 1997; Leung et al., 2005; Langelier, 1946). The aggressiveness index considers pH, Ca<sup>2+</sup> concentration, and alkalinity (as CaCO<sub>3</sub>). The LSI is an equilibrium model derived from the theoretical concept of saturation and provides an indicator of the degree of saturation of water with respect to calcium carbonate (Langelier, 1946). The RSI attempts to correlate an empirical database of scale thickness observation in water systems to the water chemistry (Roberge, 2012). These indices are calculated as follows:

$$AI = pH + \log_{10}[Ca^{2+}][AlkCaCO_3]$$

$$LSI = pH + pHs$$

$$RSI = 2 \times pHs - pH$$

where

**Table 4**  
Results of different indices for concrete corrosion by groundwater samples from slope area and urban area in both wet and dry seasons in the study area.

Region	Season	AI	LSI	RSI
Natural slope	Wet season	10.34	-1.44	9.89
	Dry season	9.18	-2.38	12.14
Urban area	Wet season	11.33	-0.68	8.71
	Dry season	10.60	-1.47	9.89
Common value of "aggressive" water		< 10	< -0.5 or > 0.5	> 8

$$pHs = (9.3 + a + b) - (c + d),$$

$$a = [\log_{10}(TDS) - 1]/10,$$

$$b = -13.12 \times \log_{10} (^{\circ}C + 273) + 34.55,$$

$$c = \log_{10}(Ca^{2+} \text{ as } CaCO_3, \text{ mg/L}) - 0.4, \text{ and}$$

$$d = \log_{10}(\text{alkalinity as } CaCO_3, \text{ mg/L}).$$

Table 4 compares the different concrete corrosion indices for groundwater samples from the slope and urban areas in both the wet and dry seasons. Groundwater in the study area is clearly corrosive, and more so in the dry season than in the wet season. The corrosive groundwater could accelerate dissolution of concrete materials in the urban area, resulting in the release of both Ca<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup>.

Normally, concentrations of ions such as Mg<sup>2+</sup>, Na<sup>+</sup>, and Cl<sup>-</sup> are much higher in the wet season than in the dry season in the urban area (Table 1), while concentrations of Ca<sup>2+</sup>, K<sup>+</sup>, HCO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> do not change much seasonally. Similar average concentrations are noted for Ca<sup>2+</sup> (72.9 and 74.6 mg/L, respectively) and K<sup>+</sup> (15.5 and 14.5 mg/L, respectively) in the wet and dry seasons. HCO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> concentrations average 230.4 and 86.7 mg/L in the wet season and 216.9 and 71.7 mg/L in the dry season. The unexpectedly high concentrations of Ca<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> in the urban area suggest that dissolution of concrete materials adds significant amounts of these ions to the shallow groundwater. Because groundwater generally moves more slowly in the dry season due to the lower hydraulic gradient, the contact time between groundwater and concrete may increase and result in more Ca<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> leaching from the concrete and accumulating in the groundwater. Ca<sup>2+</sup>, K<sup>+</sup>, and HCO<sub>3</sub><sup>-</sup> could also originate from water-rock interactions. As shown in Table 3, Ca<sup>2+</sup> and K<sup>+</sup> are both significantly related to HCO<sub>3</sub><sup>-</sup>, with correlation coefficients of 0.572 and 0.416, respectively, that are much smaller than those in the slope area. Overall, the data suggest that water-rock interactions also occur in the urban area but their influence is masked by other factors such as concrete dissolution.

Although the data in Table 4 indicate that groundwater in the slope area is normally more corrosive than in the urban area, concrete dissolution mainly occurs in the urban area. The flow of highly corrosive

**Table 5**

Results of varimax rotation of principal component analysis for major ions in shallow groundwater in slope area.

	Factor 1	Factor 2	Factor 3	Community
Ca <sup>2+</sup>	0.90	0.41	0.04	0.98
Mg <sup>2+</sup>	0.95	0.19	0.16	0.96
Na <sup>+</sup>	0.49	0.80	0.20	0.92
K <sup>+</sup>	0.56	0.75	-0.07	0.88
HCO <sub>3</sub> <sup>-</sup>	0.90	0.39	-0.15	0.98
Cl <sup>-</sup>	0.68	0.66	0.18	0.93
SO <sub>4</sub> <sup>2-</sup>	0.16	0.80	0.51	0.92
NO <sub>3</sub> <sup>-</sup>	0.01	0.16	0.97	0.97
% Variance	44.6	33.0	16.6	
% Cumulative	44.6	77.6	94.2	
Source	Granite weathering	Seawater and rainwater	Anthropogenic pollution	

groundwater from the slope area to the urban area can significantly contribute to the observed increase in TDS. This phenomenon would explain the sudden increase of TDS in groundwater (from 248.8 to 1136.9 mg/L and from 39.7 to 334.5 mg/L, in the wet and dry seasons, respectively) (Table 1) over a relatively short flow distance from the slope area to the urban area.

Notably, foundations and other underground structures are more vulnerable to corrosion in the dry season due to the increased contact time. Accordingly, this may negatively affect the structure of high-rise buildings. Furthermore, the groundwater table is generally low in the dry season, so the corrosion of deep foundations could be more significant.

#### 4.6. Estimation of source contribution

Principal components analysis (PCA) was adopted to evaluate and verify the contribution of the above discussed factors to groundwater chemistry. PCA is a useful statistical tool for data analysis and dimensionality reduction and is widely used across various fields of research (Shrestha et al., 2008; Stewart et al., 2014). Considering the biases of covariance structure of compositional data, appropriate transformations of the raw data should be conducted before PCA (Filzmoser et al., 2010). All the raw data was transformed into their Z scores, and then varimax rotation was conducted for PCA (Davis, 2002; Chen et al., 2007; Huang et al., 2010). The results of PCA are consistent with the findings of the process analyses discussed above, and could well explain the situation of shallow groundwater in the study area. Table 5 shows the principal component patterns for the shallow groundwater data from the slope area. Three major factors account for 94.2% of the total variance. The first factor explains 44.6% of the total variance with high loadings of Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and HCO<sub>3</sub><sup>-</sup>, and is attributed to granite weathering. The second factor accounts for 33.0% of the total variance with low loadings of Ca<sup>2+</sup>, Mg<sup>2+</sup>, and

HCO<sub>3</sub><sup>-</sup> and relatively high loadings of Na<sup>+</sup>, K<sup>+</sup>, and SO<sub>4</sub><sup>2-</sup>, and implies the influence of rainwater, which due to the “near-to-coast” effect contains similar ions to seawater (Huang et al., 2010). The third factor accounts for 16.6% of the total variance and has high loadings of NO<sub>3</sub><sup>-</sup> and clearly represents an anthropogenic pollution source, likely a municipal landfill near the slope area. Notably, the sample size for the slope area is relatively small; however, these results help to demonstrate the major influences of granite weathering and water-rock interactions.

The principal component patterns of shallow groundwater samples of the urban area are shown in Table 6. Four major factors are extracted, accounting for 94.7% of the total variance. The first factor explains 35.0% of the total variance and features high loading of Ca<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup>, clearing indicating the influence of concrete material dissolution. The second factor accounts for 29.6% of the total variance with relatively high loadings of Na<sup>+</sup> and Cl<sup>-</sup>, indicating seawater intrusion. The third factor accounts for 16.6% of the total variance and features high loadings of HCO<sub>3</sub><sup>-</sup>, which indicates granite weathering. The final factor contributes 13.5% and features high NO<sub>3</sub><sup>-</sup> loading, suggesting an anthropogenic pollution source such as sewage.

## 5. Summary and conclusions

Shallow groundwater in a coastal urban area of Shenzhen City, China was investigated using hydrogeochemical methods to identify the influence of seawater intrusion and other natural and anthropogenic factors. Season does not affect the types of shallow groundwater found in the study area. The shallow groundwater is mainly dominated by fresh water, and is identified as either Ca-HCO<sub>3</sub> and Ca-Na-HCO<sub>3</sub>-Cl type. These types suggest the influence of seawater (via mixing), but only a few locations close to Shenzhen Bay are Na-Cl type and indicate the active threat of seawater intrusion. However, increasing TDS concentrations suggest that other anthropogenic and natural processes besides seawater intrusion may also significantly influence the shallow groundwater chemistry.

Precipitation infiltration has little direct influence on shallow groundwater chemistry. Anthropogenic pollution is likely to occur in the form of point sources throughout the study area and, as the result of nitrification, the dominant inorganic nitrogen form in groundwater in the wet season is NO<sub>3</sub><sup>-</sup> in both areas while in the dry season is NO<sub>3</sub><sup>-</sup> in the slope area but NH<sub>4</sub><sup>+</sup> in the urban area. In the slope area, granite weathering adds Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and HCO<sub>3</sub><sup>-</sup> to the shallow groundwater. All of the shallow groundwater in the study area is corrosive, and thus concrete materials in the urban area can be dissolved by shallow groundwater and represent another important source of Ca<sup>2+</sup> as well as SO<sub>4</sub><sup>2-</sup>. PCA results suggest four main factors in the urban area account for 94.7% of the shallow groundwater chemistry: concrete material dissolution (35.0%), seawater intrusion (29.6%), granite weathering (16.6%), and anthropogenic pollution (13.5%). All groundwater was corrosive in both the wet and dry seasons, and slightly

**Table 6**

Results of varimax rotation of principal component analysis for major ions in shallow groundwater in urban area.

	Factor 1	Factor 2	Factor 3	Factor 4	Community
Ca <sup>2+</sup>	0.76	0.28	0.52	0.00	0.92
Mg <sup>2+</sup>	0.69	0.67	0.07	0.20	0.97
Na <sup>+</sup>	0.63	0.73	0.06	0.17	0.96
K <sup>+</sup>	0.25	0.84	0.36	0.05	0.89
HCO <sub>3</sub> <sup>-</sup>	0.14	0.14	0.95	0.03	0.94
Cl <sup>-</sup>	0.68	0.69	0.01	0.19	0.97
SO <sub>4</sub> <sup>2-</sup>	0.90	0.32	0.15	0.07	0.94
NO <sub>3</sub> <sup>-</sup>	0.09	0.13	0.02	0.99	0.99
% Variance	35.0	29.6	16.6	13.5	
% Cumulative	35.0	64.5	81.2	94.7	
Source	Concrete material dissolution	Seawater intrusion	Granite weathering	Anthropogenic pollution	

more so in the dry season when the water table is lower. On the basis of the findings of this study, the construction of foundations in the study area should consider the corrosive nature of shallow groundwater and efforts be made to reduce the amount of ions dissolved from building foundations. More work needs to be done to determine what causes the corrosiveness of the shallow groundwater in the study area.

The conclusions of this study give a better understanding of geochemical characteristics of shallow groundwater not only for this region but also for other coastal regions, and therefore provide a good reference for groundwater resource management in coastal urban areas.

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### Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi: <https://doi.org/10.1016/j.gexplo.2017.10.003>. These data include Google map of the most important areas described in this article.

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