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Significance of Hydrogeochemical Analysis in the Management of Groundwater Resources: A Case Study in Northeastern Iran

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1. Introduction

Hydrogeochemistry is a sub-discipline of hydrogeology which is referred to as Chemical hydrogeology in some references (e.g. Domenico and Schwartz, 1990) and Groundwater geochemistry in some others (e.g. Merkel and Planer-Friedrich, 2005). One may also points to "Contaminant hydrogeology" used by Fetter (1998) as another term that carries the same syllabus as Hydrogeochemistry. In all fields of science and engineering, sub-disciplines are developed to accomplish some missions, fulfill some requirements and supplement the base subject. The same is true with hydrogeochemistry. This sub-discipline has been developed to deal with quality, contamination, chemistry, chemical processes and reactions that take place in various groundwater systems. Due to the importance of water quality issues, this sub-discipline has gradually changed into a well established field of research. For instance, Prasanna et al. (2011) clearly demonstrates that the study of quantity of water alone is not sufficient to solve the water management problems because its uses for various purposes depend on its quality. In addition, a number of other researchers show that the hydrogeochemical characteristics of groundwater and groundwater quality in different aquifers over space and time are important parameters in solving the groundwater management issues (Panigrahy et al., 1996; Atwia et al., 1997; Ballukraya and Ravi, 1999; Ramappa and Suresh, 2000). At the start, it should be pointed out that the quality of groundwater depends on the chemical composition of recharge water, the interaction between water and soil, soil-gas interaction, the types of rock with which it comes into contact in the unsaturated zone, the residence time of groundwater in the subsurface environment and the reactions that take place within the aquifer (Freeze and Cherry, 1979; Hem, 1989; Appelo and Postma, 2005). Hydrogeochemical processes such as dissolution, precipitation, absorption and desorption, ion exchange reactions and the residence time along the flow path which controls the chemical composition of groundwater, constitutes the other issues that are dealt with in hydrogeochemistry.

2. Framework of a hydrogeochemical study

There is a routine working flow chart for a hydrogeochemical study. It starts with preliminary desk studies including data collection and collation. Once all the available data-related to the aquifer in question- are assembled and assessed, data gaps are identified and deficiencies are brought into attention. The type of the data that needs to be created and the costs involved are all carefully evaluated. Often, the missing and the necessary data we think about at the first glance are the chemistry data. To gather these, groundwater sampling for chemical analysis are proposed and meticulously planned. Depending on the financial constraints and the level of precision required to solve the problem in hand, the number of samples varies. Sometime, it might be needed to drill new piezometers for this purpose. Before any sampling campaign, necessary equipments such as bottles, additives including acids and reagents, field probes, filter papers, pumps and bailers, various size hoses, isolating box or car fridge and a variety of other stuff- depending on the specific aim of the study- should be prepared. Field vehicle, maps, food and proper shoe and clothing are also needed as is the case with any geological field investigations. Collected groundwater samples are then transported to the relevant laboratory in due time, where they are analyzed using methods specified for different types of chemical elements, anthropogenic constituents, microbes and bacteria. The quality control techniques (duplicated samples, comparing the results which were produced by different laboratories analyzing the same samples, etc) should be undertaken to ensure reliable data. Working with various computer codes to interpret the data and graphically illustrate the results, is the program of work for the next step. At the final stage of the study, hydrogeochemical findings should be compared with the other geological, hydrogeological and field observations results and either revised accordingly or further filed investigation and water sampling be planned.

3. Applications of hydrogeochemical analysis

What we learn from a hydrogeochemical study? What are the objectives of a hydrogeochemical analysis program? Hydrogeochemical investigations are generally carried out to:

1. Identify the cause of anthropogenic contamination and the source of natural salinity
2. Evaluate the fate of the pollutants in the subsurface environment
3. Calculate the time needed to flush out the contaminants and clean the aquifer
4. Pinpoint the type of the geochemical reactions that are taking place within an aquifer system
5. Asses the quality of groundwater for different uses
6. Identify the factors governing the chemical composition of groundwater and the contribution made by each factor
7. Map the spread and the three dimensional picture of the contamination plume
8. Constrain the extent of the saltwater intrusion zone
9. Quantify leakage between surface water and groundwater and locate the hyperhoic zone
10. Compute the natural capacity of the aquifer in attenuating the pollutants

11. Study salinity stratification in groundwater systems and likeliness of variable density flows.

4. A hydrogeochemical case study in Northeastern Iran

Iran is located in a semi-arid area with an annual precipitation rate of 250 mm/year which is less than one third of that of the world average. In the arid and semi-arid environments with limited water resources such as Iran, groundwater constitutes a significant part of the available water resources (Subyani, 2005). Groundwater provides more than half of the total annual water demand in Iran; the uncontrolled groundwater use accompanied by successive draughts in the recent years has adversely affected the quality and quantity of Iran's subsurface waters. This is particularly true in central plateau where high temperature and low precipitation rates prevail. Furthermore, in such environments, groundwater chemistry evolves rapidly and the salinity increases swiftly, leading to restrictions in water utilization and limitations on the development and management of unconfined alluvial aquifers (Subyani, 2005). Several factors lead to rise in the salinity of groundwater. Some of these are local, such as hydrogeological conditions, rate of natural recharge and irrigation, while others are regional in nature. The latter group includes the aridity of the environment and the irregular and unpredictable occurrence of atmospheric precipitation (Eagelson, 1978 and 1979).

5. Description of the study area

Safi Abad watershed in northeastern Iran is located in the south of the Northern Khorasan province, one of the 31 provinces which make up the land of the country (Figure 1). It occupies an area of 2,593 Km², 848 Km² of which is in the form of a plain (Safi Abad plain) and the rest is highlands and mountains. There is no large city in the area and the largest village, Safi Abad, lies in the southern part of the watershed. All together, there are 77 villages in this watershed, the population of which reach 21,152 people. The climate of the Safi Abad watershed is classified as arid and semi-arid with an average annual precipitation of 241 mm. The local temperature rises above 40 °C in hot summer days and it sharply drops to a few degrees below freezing point during the cold winter days. The only local permanent river is Kale Shoor River which flows along east-west direction and is adjoined by two main tributaries. It originates from the mountains in the northern and south eastern parts of the Safi Abad plain and exit from the north western part after draining the area. Due to the scarcity of the surface water resources, water demand for agricultural and potable purposes is supplied by a number of various sizes qanats, springs and water wells mainly in the northern part of the watershed (Note that qanat is a traditional structure, a horizontal gallery which yields groundwater by gravity). Agriculture and animal husbandry are the two main job markets in the region; cotton, a variety of crops, sugar bits, zirah (an Iranian costly product used as flavor in tea and food) and nuts are the main agricultural products. Natural vegetation covers in the highlands consists mainly of short trees and sparsely spaced tall trees.

In terms of regional geology, Safi Abad watershed is a part of the Zone of Central Iran, the oldest rock outcrops of which are Paleozoic in age (Stocklin, 1968). Local geological map of the study area is shown in Figure 2. As it can be seen from this figure, various lithologies

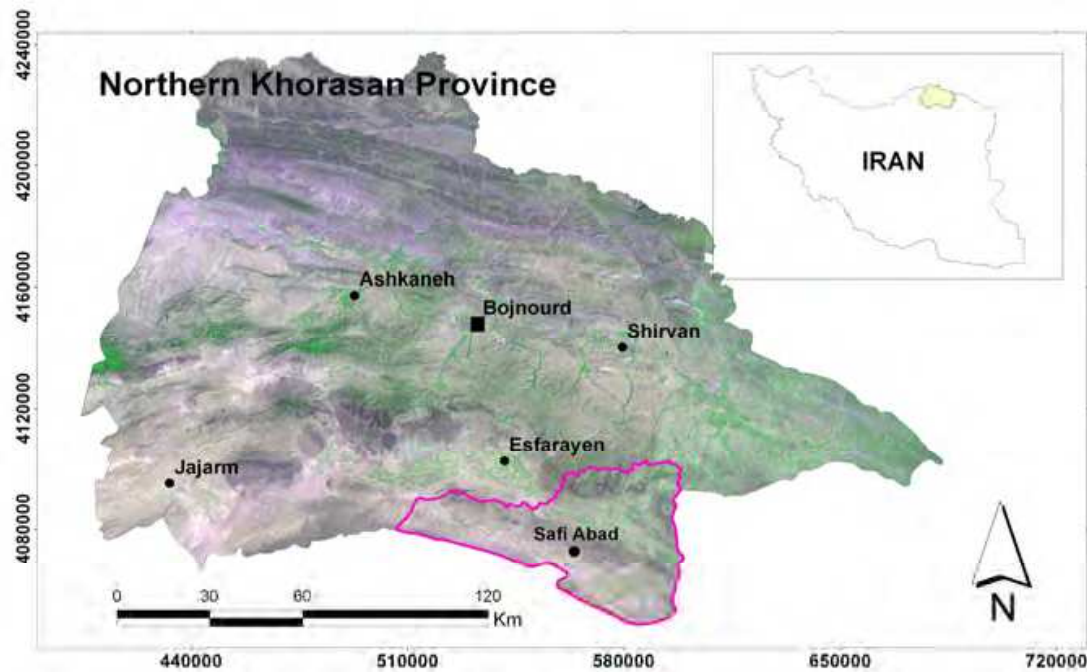


Fig. 1. Geographical position of the study area in the southern part of the Northern Khorasan Province, northeastern Iran.

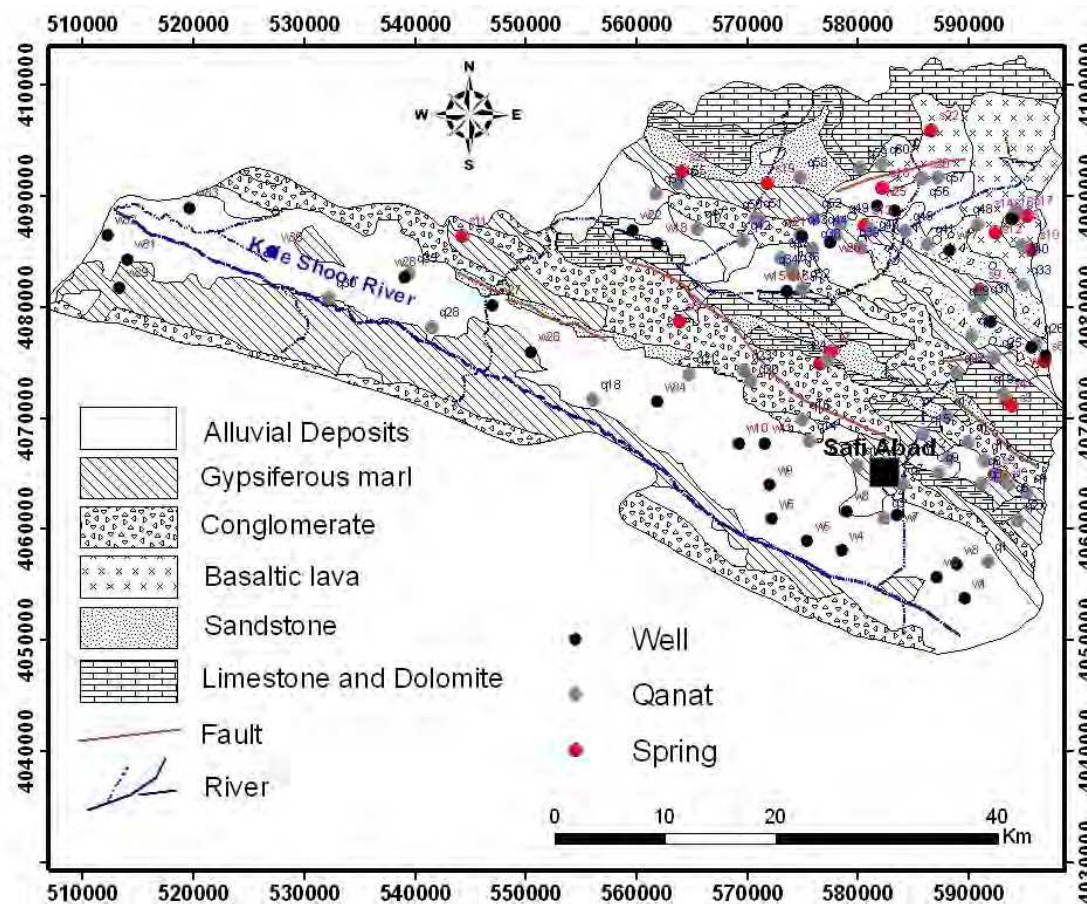


Fig. 2. Geological map of the Safi Abad Watershed (Based on Afshar Harb et al., 1987) and the location of the groundwater samples. Samples from wells, qanats and springs are differentiated.

such as limestone, dolomite, sandstone, shale-sandstone-gypsum sequence and basaltic lava underlie the area. The diverse range of rocks and sediments has led to different erosional pattern in the area. Unconsolidated and easily erodable parts of the plain are covered by marl and conglomerate which make up both the lowlands and the extensive valley floors. In contrast, highlands are formed by massive limestones and Eocene volcanic rocks which are over 2,000 m in height in some points. The minimum elevation in the study area is 1,050 m at the outlet of Kale Shoor river and the maximum elevation is 3,034 m above sea level.

6. Groundwater sampling

In the study area, groundwater utilization takes place through water wells, springs and qanats. There are 133 water wells, 190 qanats and 125 springs in the area which produce 51, 16.4 and 21 MCM (Million Cubic Meters) of groundwater per year, respectively; groundwater production rate is therefore 88.4 MCM per annum in an average year. Wells are mostly located in the plains while the majority of qanats and springs emerge from the highlands and mountains (Figure 2). Like everywhere, more than 95 percents of the extracted groundwater is used in agriculture sector; the proportional contribution of wells, qanats and springs is slightly different, however. A total of 35 water wells, 22 springs and 60 qanats were sampled from different parts of the aquifer underlying Safi Abad watershed in 2010. The sampling sites, altogether 117 sites, are shown in Figure 2. Electrical conductivity (EC) and hydrogen ion concentration (pH) were measured in the field using portable EC and pH meters. Water samples were then analyzed in the laboratory for acidity, electrical conductivity and major ions using standard methods. The summary of the results of the chemical analysis of groundwaters and the calculated charge balance error are presented in Table 1. Low values of charge balance errors demonstrate that the accuracy of the analysis is within the acceptable range.

	EC μS/cm	pH	Anions meq/L				Cations meq/L				% error
			Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	HCO ₃ ⁻	CO ₃ ²⁻	SO ₄ ⁻	Cl ⁻	
Average	3322	7.84	21.4	0.599	6.63	5.83	4.68	0.072	10.83	19.2	-0.75
Minimum	274	6.2	0.2	0	0.2	0.8	1.5	0	0.3	0.3	-6.25
Maximum	35825	8.8	261	14.8	47	77	9.5	0.9	93	285	3.9

Note that percent error values are statistically calculated, otherwise -6.27 is the highest error.

Table 1. Summary of physico-chemical data of the water samples (Number of samples: 117).

7. Results and discussions

7.1 Electrical conductivity and pH

The EC of the samples ranges widely from 274 μS/cm to 35,825 μS/cm, with an average value of 3,322 μS/cm. Forty nine samples have ECs less than 1000, 23 samples less than 2000, 19 samples over 2000 and less than 5000 and the remaining 26 samples have ECs

higher than 5000 $\mu\text{S}/\text{cm}$. This is a clear indication that the aquifer in question has been subjected to salinization processes either naturally or anthropogenically. Saline samples are mostly from the plain and from the wells. Samples pH values are all but one above neutrality which reflects the basic nature of the groundwater. Figure 3 shows that there is a slight negative correlation between EC and pH values, most probably due to high sodium content of these samples as it is demonstrated in the next section.

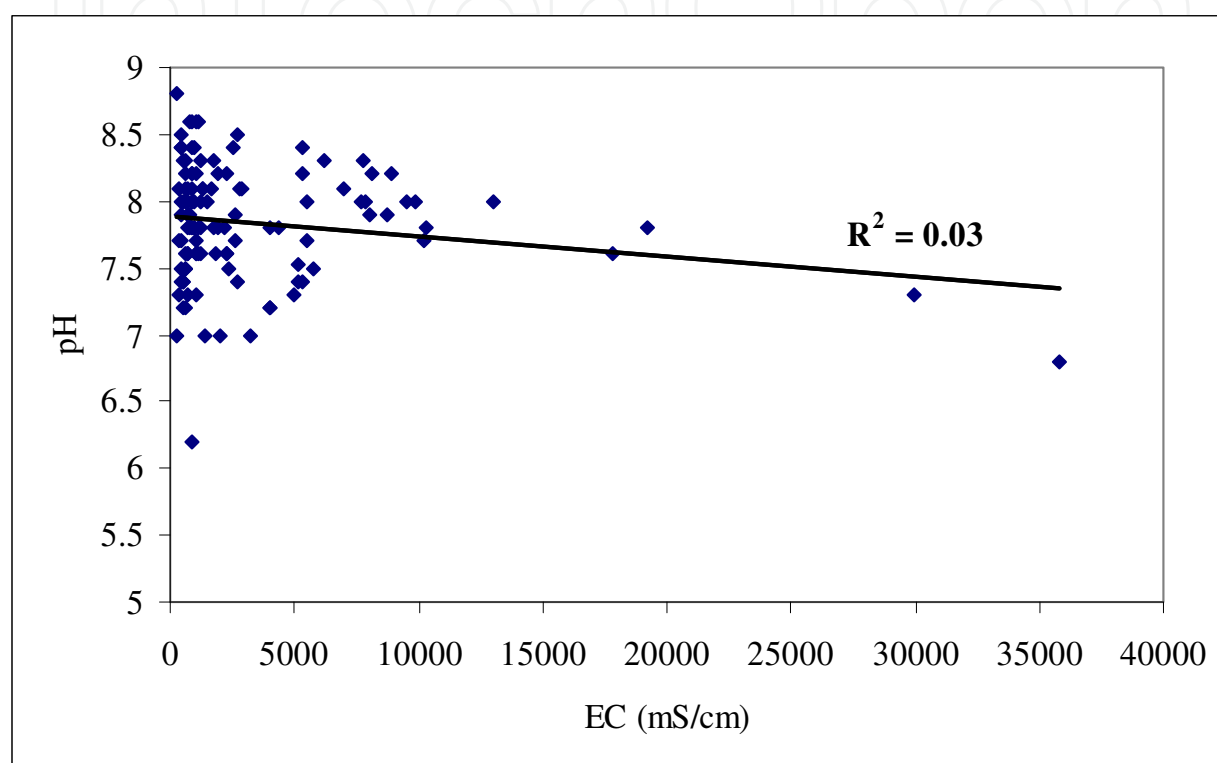


Fig. 3. Relationship between pH and EC values of the groundwater samples

7.2 Hydrogeochemical facies and classification

The chemical composition of groundwater is primarily dependent on the geology as well as on the geochemical processes which take place within the groundwater system. The Piper trilinear diagram (Piper, 1944) has been used for the purpose of characterizing the water types present in the area (Figure 4). Water types are often used in the characterization of waters as a diagnostic tool (Leybourne et al., 1998; Pitkanen et al., 2002). In addition, Piper diagram also permits the cation and anion compositions of many samples to be represented on a single graph in which major groupings or trends in the data can be discerned visually (Freeze and Cherry, 1979). Furthermore, it is used to assess the hydrogeochemical facies. A few conclusions can be inferred from the piper diagram of the collected samples (Figure 4). First, it shows that groundwater in the watershed is of very different types which evolve from bicarbonate to chloride type. Secondly, large percentages of the samples fall within the Na-Cl category followed by Na-HCO₃ type. Thirdly, chloride is the dominant anion found in the groundwater in the study area with the concentration ranging from 0.3 to 285 meq/L.

High concentration of Cl^- may be due to leaching of saline soil residues into the groundwater system, a typical characteristic of arid and semi-arid regions (Zaheeruddin and Khurshid, 2004). Finally, Piper plot shows that sodium with a mean value of 21.4 meq/L dominates the cationic components of the groundwater, although the sum of calcium and magnesium is higher than sodium. Figure 5 illustrates the spatial distribution of different water types throughout the watershed and shows that Na-Cl type waters are mainly concentrated in the plain part of the study area.

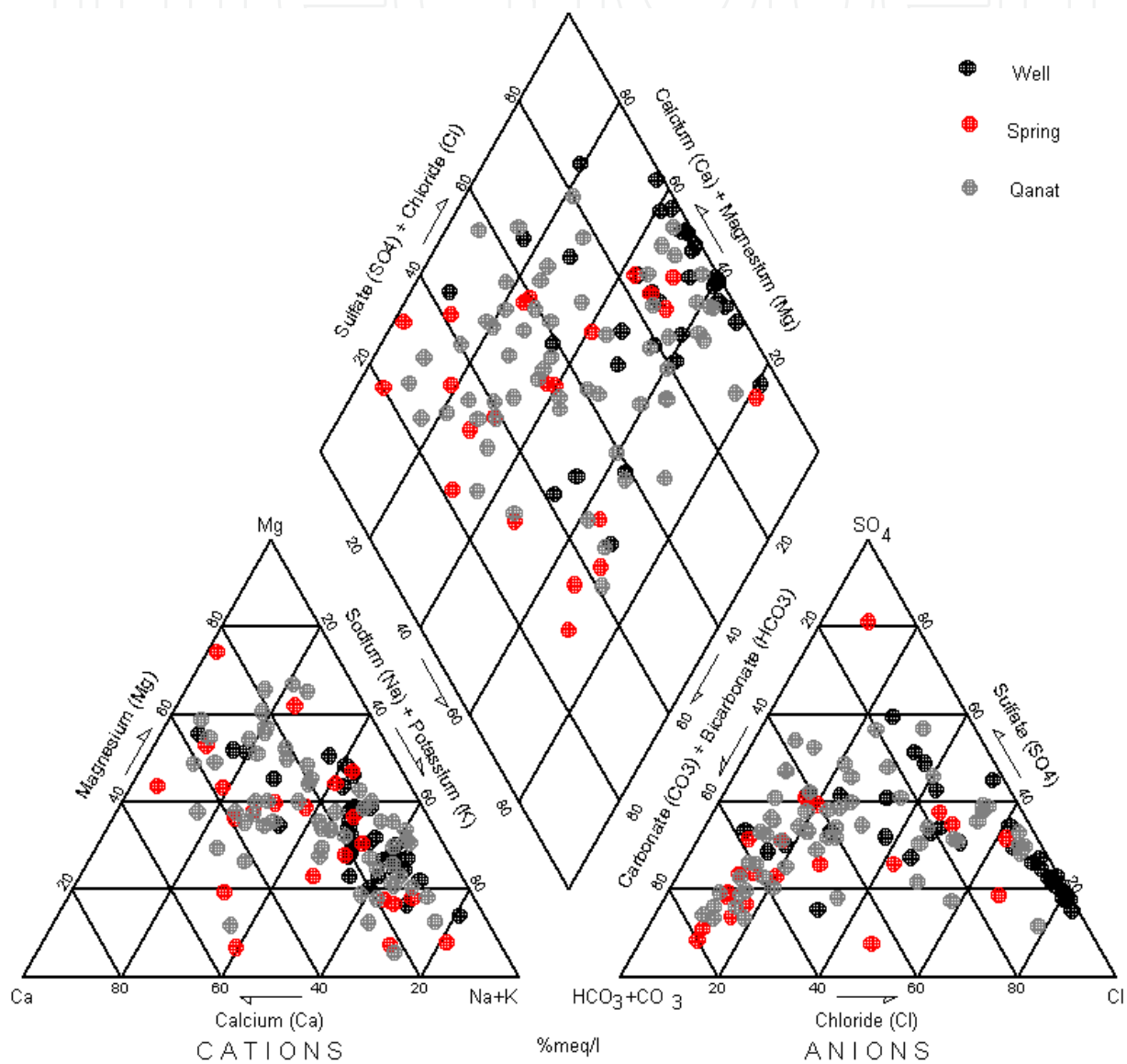


Fig. 4. Piper diagram of the groundwater samples

7.3 Origin of major ions

Concentration of ions dissolved in groundwater is generally controlled by lithology, groundwater flow rate, natural geochemical reactions and human activities (Karanth, 1997;

Bhatt and Salakani, 1996). In fresh groundwaters, both HCO_3^- and CO_3^{2-} originate mainly from the atmosphere but dissolution of sulfates, dolomite, calcite or silicates minerals also contribute to the concentration of these ions. Figure 6a displays the concentration of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus HCO_3^- . It is seen that all samples lie above the 1:1 line; this demonstrates an extra source for both Ca^{2+} and Mg^{2+} ions. The excess concentration of $\text{Ca}^{2+} + \text{Mg}^{2+}$ over HCO_3^- has been balanced by Cl^- and SO_4^{2-} . Excess calcium and magnesium is most likely supplied by the dissolution of various minerals such as dolomite, gypsum, calcite, anhydrite or weathering of silicate minerals such as plagioclase, pyroxene, amphibolites and montmorillonite (Freeze and Cherry, 1979, Boghici and Van Broekhoven, 2001). Plot of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{SO}_4^{2-} + \text{Cl}^-$ shows that all samples lie below theoretical 1:1 line, indicating the equilibrium between $\text{SO}_4^{2-} + \text{Cl}^-$ with alkalis (Figure 6b). Also plot of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus total cations (TC) illustrates that all samples lie far below the theoretical line depicting a contribution of alkalis to the major ions (Figure 6c). Groundwater in the area has a higher average ratio of $\text{Na}^+ + \text{K}^+$ versus total cations (Figure 6d). Plotting $\text{Na}^+ + \text{K}^+$ versus SO_4^{2-} show that all samples lie above 1:1 line (Figure 6e). The excess amount of $\text{Na}^+ + \text{K}^+$ over Cl^- (Figure 6f) at low chloride concentrations reflects input from the weathering of Na and K-rich minerals (Stallard and Edmond, 1983).

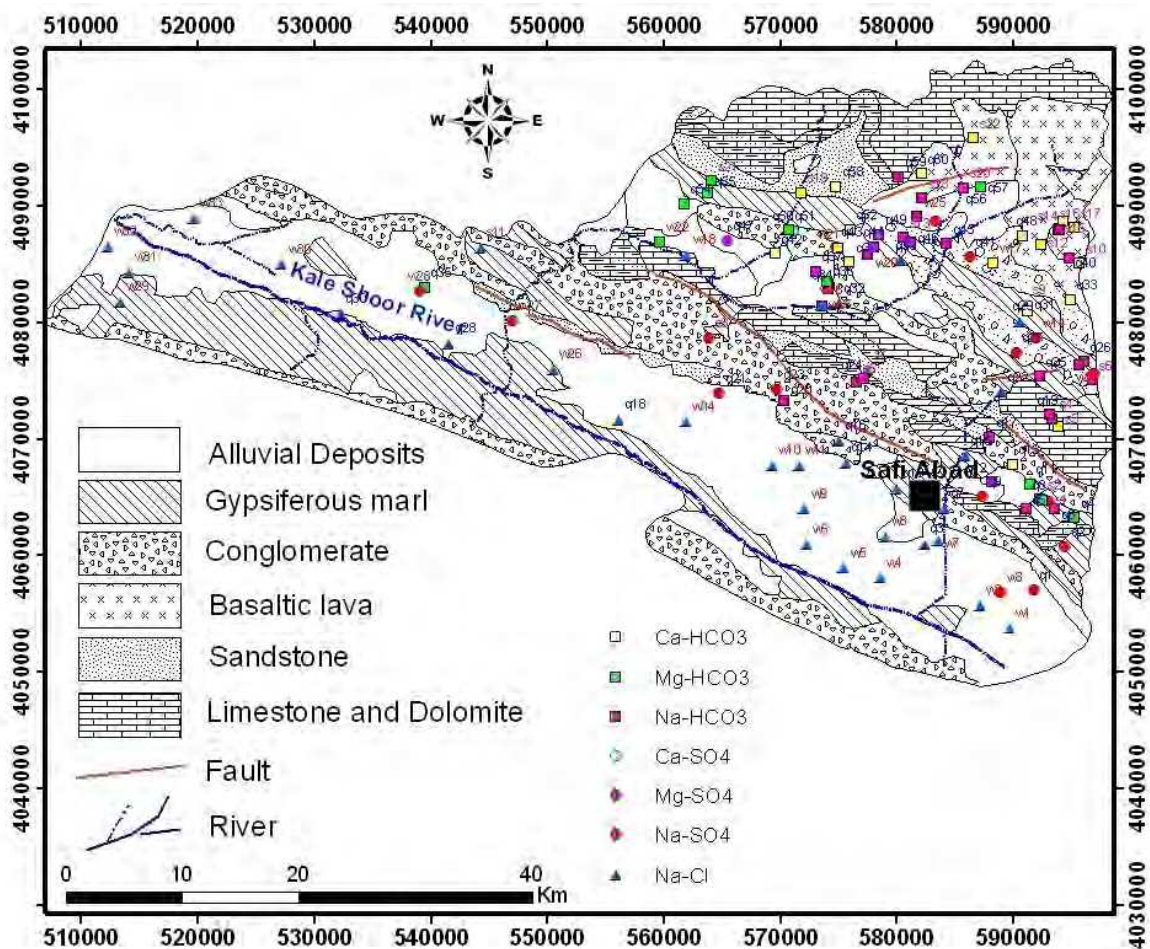
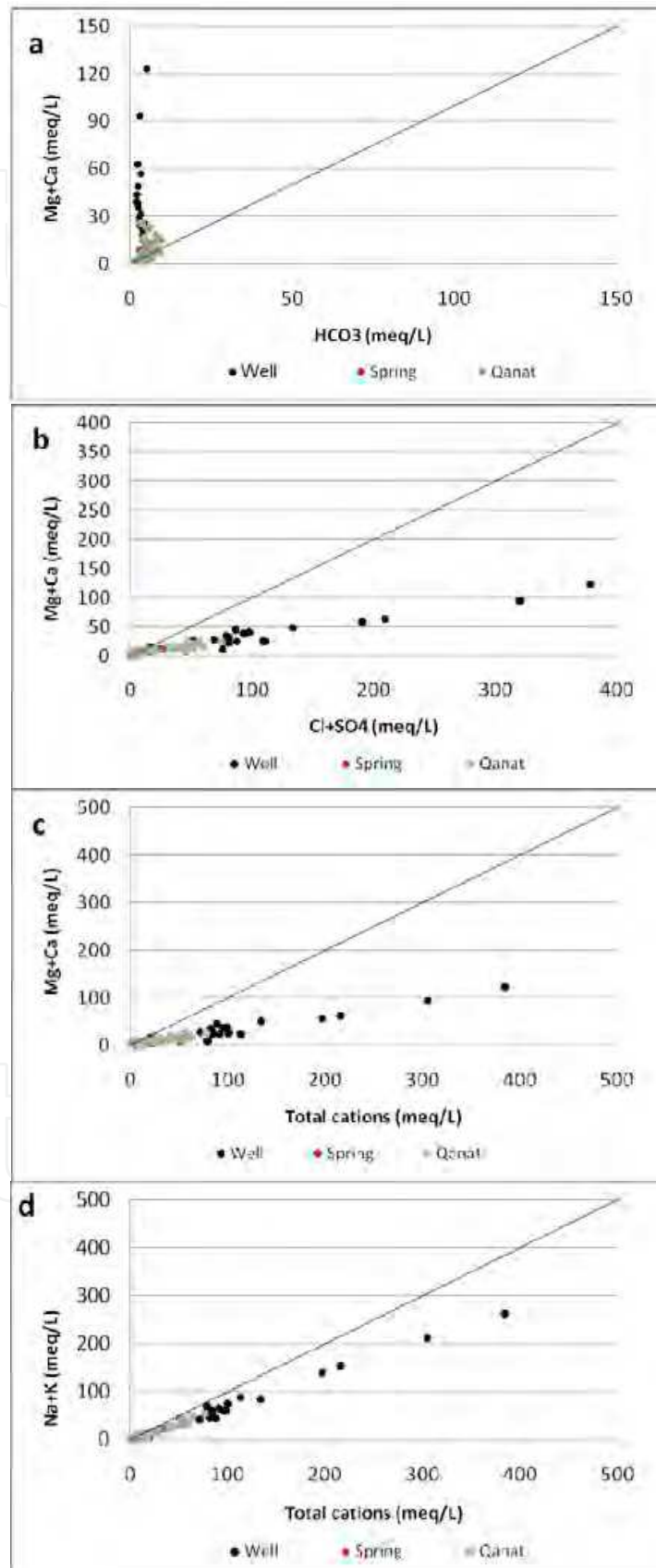


Fig. 5. Spatial distribution of different water types in the study area



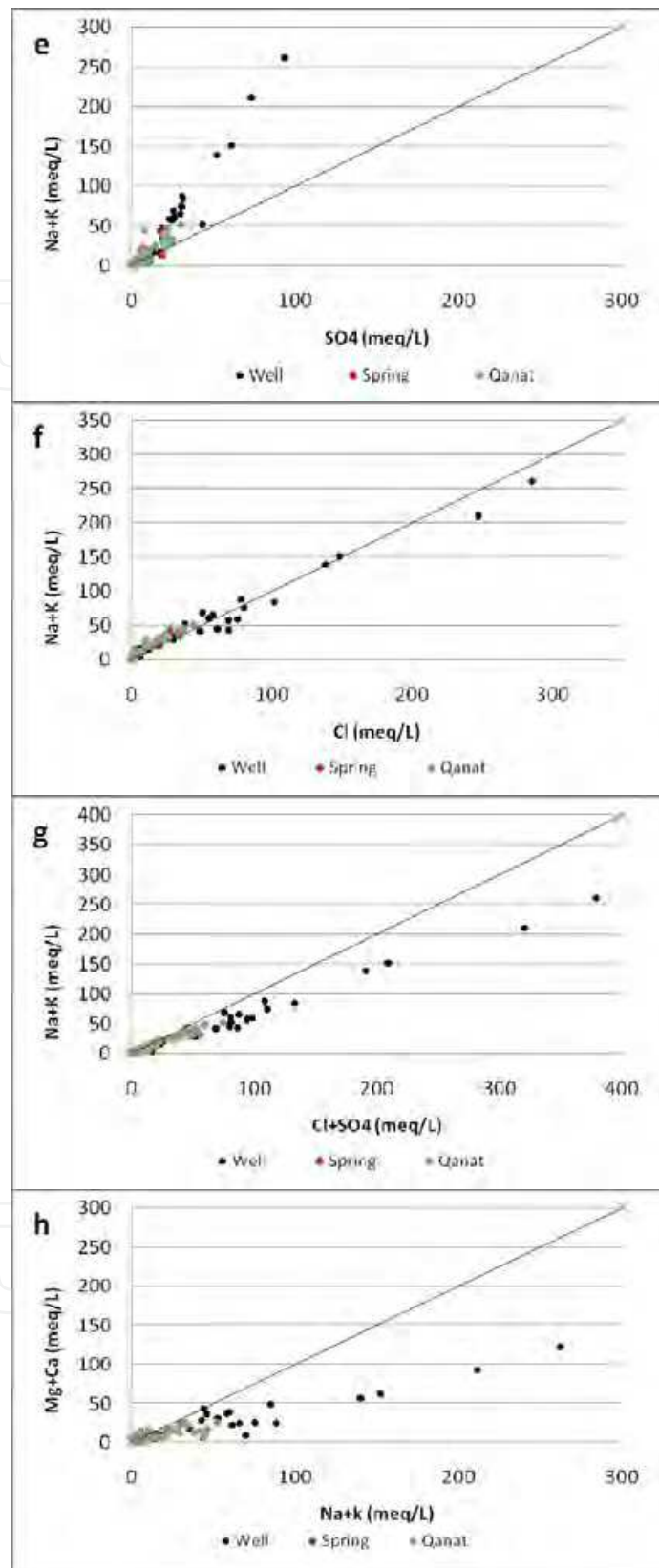


Fig. 6. Scatter plots of a) $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus HCO_3^- , b) $\text{Mg}^{2+} + \text{Ca}^{2+}$ versus $\text{Cl}^- + \text{SO}_4^{2-}$, c) $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus total cations, d) $\text{Na}^+ + \text{K}^+$ versus total cations, e) $\text{Na}^+ + \text{K}^+$ versus SO_4^{2-} f) $\text{Na}^+ + \text{K}^+$ versus Cl^- g) $\text{Na}^+ + \text{K}^+$ versus $\text{Cl}^- + \text{SO}_4^{2-}$ and h) $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{Na}^+ + \text{K}^+$.

Scatter plot of $\text{Na}^+ + \text{K}^+$ versus $\text{SO}_4^{2-} + \text{Cl}^-$ reveals that the increase in alkalis corresponds to a simultaneous increase in $\text{Cl}^- + \text{SO}_4^{2-}$ (Figure 6g), thereby indicating a common source for these ions and also the presence of Na_2SO_4 and K_2SO_4 in the soils (Datta and Tayagi, 1996). In samples with low molar $\text{Na}^+ + \text{K}^+$, $\text{Ca}^{2+} + \text{Mg}^{2+}$ and $\text{Na}^+ + \text{K}^+$ are balanced together and lie at 1:1 line with increase in molar values of $\text{Na}^+ + \text{K}^+$, data move to under the line (Figure 6h).

7.4 Factors governing water chemistry

The mechanism controlling water chemistry and the functional sources of dissolved ions can be assessed by plotting the ratios of Na^+ to $(\text{Na}^+ + \text{Ca}^{2+})$ and Cl^- to $(\text{Cl}^- + \text{HCO}_3^-)$ as functions of TDS (Gibbs, 1970). Gibbs diagram of the water samples (Figure 7) clearly shows that the majority of samples have become saline either via saline water intrusion or evaporative enrichment. As noted by some researchers, evaporation greatly increases the concentration of ions formed by chemical weathering, leading to higher salinity (Jalali, 2007). However, TDS values of over 1000 mg/L are mostly controlled by saline water intrusion. Figure 7 also shows that weathering of rock-forming minerals is only contributing marginally to the salinity of a small number of samples.

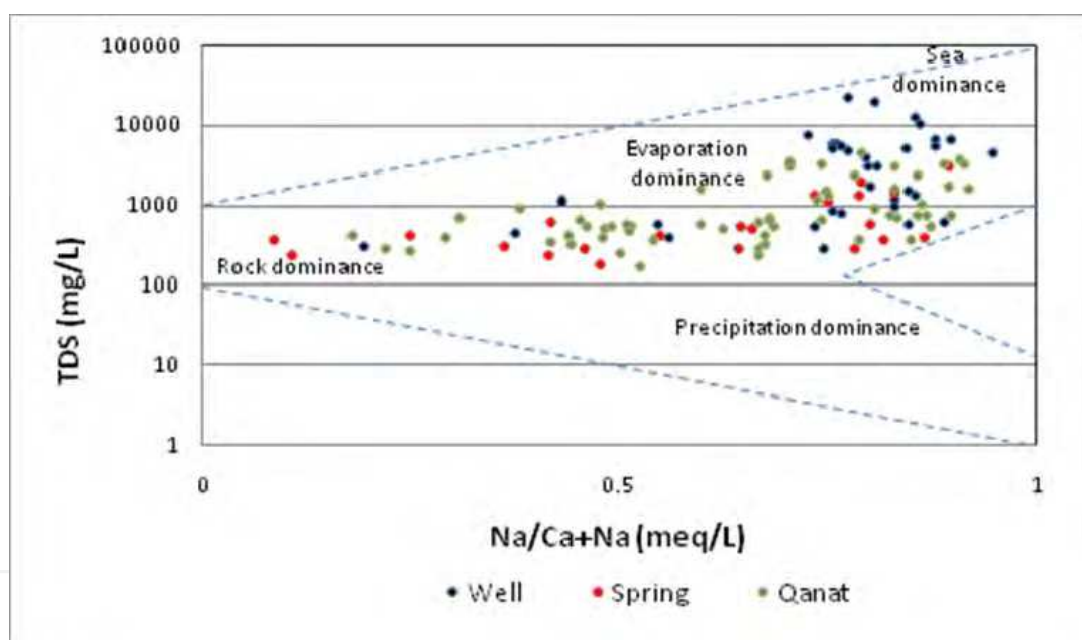


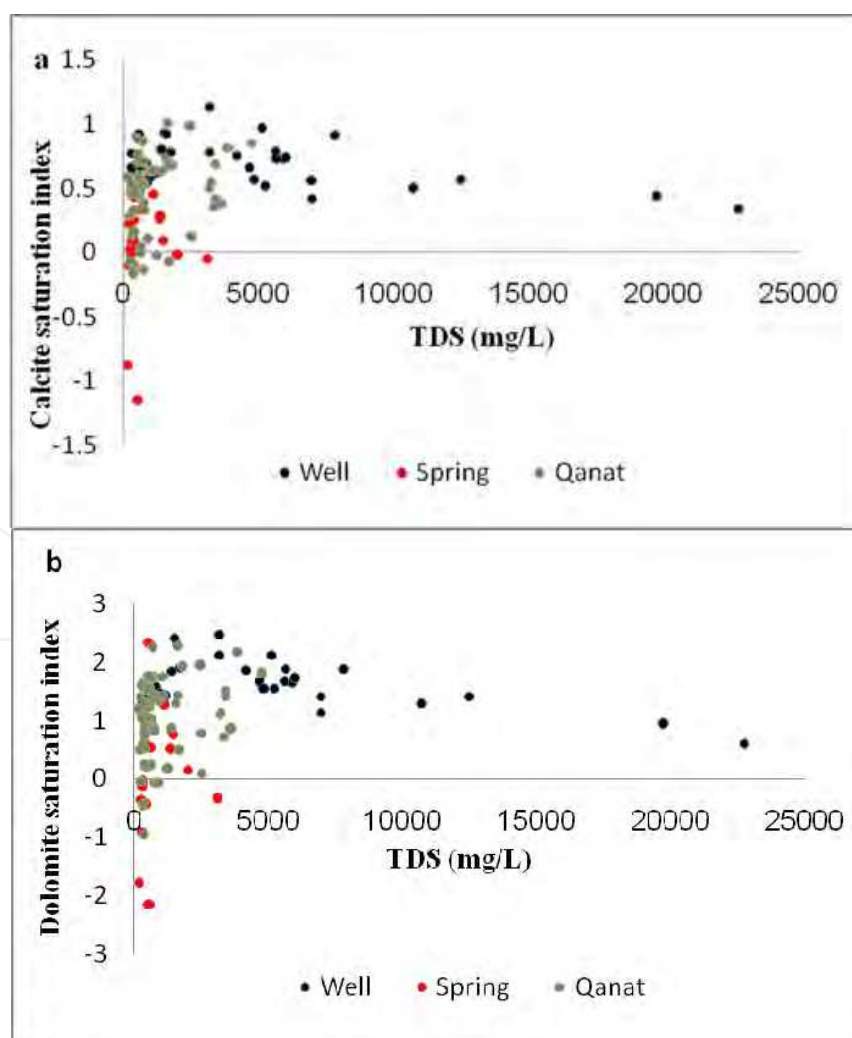
Fig. 7. Gibbs diagram of water samples

7.5 Geochemical modeling

Geochemical interactions often lead to changes in the water chemistry. The potential for a chemical reaction to happen can be determined by calculating the chemical equilibrium of the water with the mineral phases in question. The equilibrium state of the water with respect to a mineral phase can be determined by calculating saturation index (SI) using analytical data. Variation in the groundwater chemistries is mainly a function of the interaction between the groundwater and the mineral composition of the aquifer materials through which it moves (Nwankwoala and Udom, 2011). These processes generally include chemical weathering of minerals, sorption, and desorption, dissolution-precipitation of secondary carbonates and ion exchange between water and clay minerals (Apodaca et al.

2002). During rock weathering, Ca^{2+} , Mg^{2+} , SO_4^{2-} , HCO_3^- and SiO_2 are added to the water. The amount of each depends on the specific mineralogy of the rocks in question (Hounslow, 1995). In this research, the geochemical modeling program PHREEQC (Parkhurst and Appelo, 1999) has been employed to evaluate the water chemistry. The inverse modeling in PHREEQC takes into account uncertainty limits that are constrained to satisfy the mole balance for each element and valance state, as well as the charge balance for each solution within the simulation. In this research, we have employed PHREEQC to calculate saturation indices (SI) of sampled waters, only; it has many other applications.

Figure 8 shows the plot of SI of various minerals against TDS for all samples. With the exception of a few, all samples are super saturated with respect to both calcite and dolomite, suggesting that these carbonate mineral phases are extensively present in the corresponding host rock. Sharif et al., (2008) argue that the common ion effect of gypsum dissolution and calcite precipitation is often accompanied by dolomite dissolution leading to the meaningful increase in the magnesium content of groundwater. This is the case in the study area where higher numbers of samples are super saturated with respect to dolomite than calcite. All samples are under saturated with respect to anhydrite, gypsum and halite, even for very saline samples. Therefore, one may conclude that the interaction between groundwater and aquifer matrix is not significant in controlling chemical characteristics of groundwater in the study area, i.e. the source of the ions is mostly outside of the aquifer matrix.



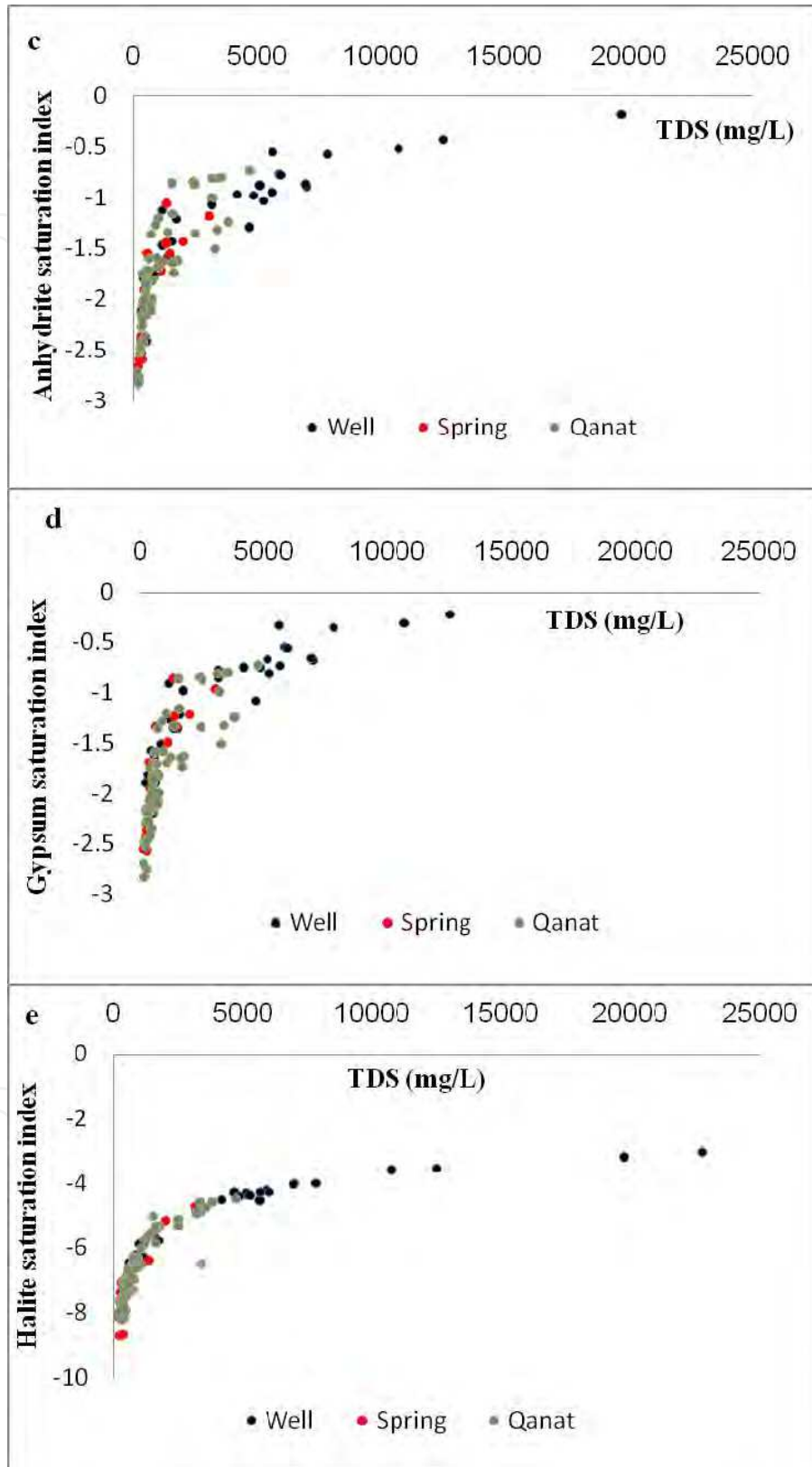
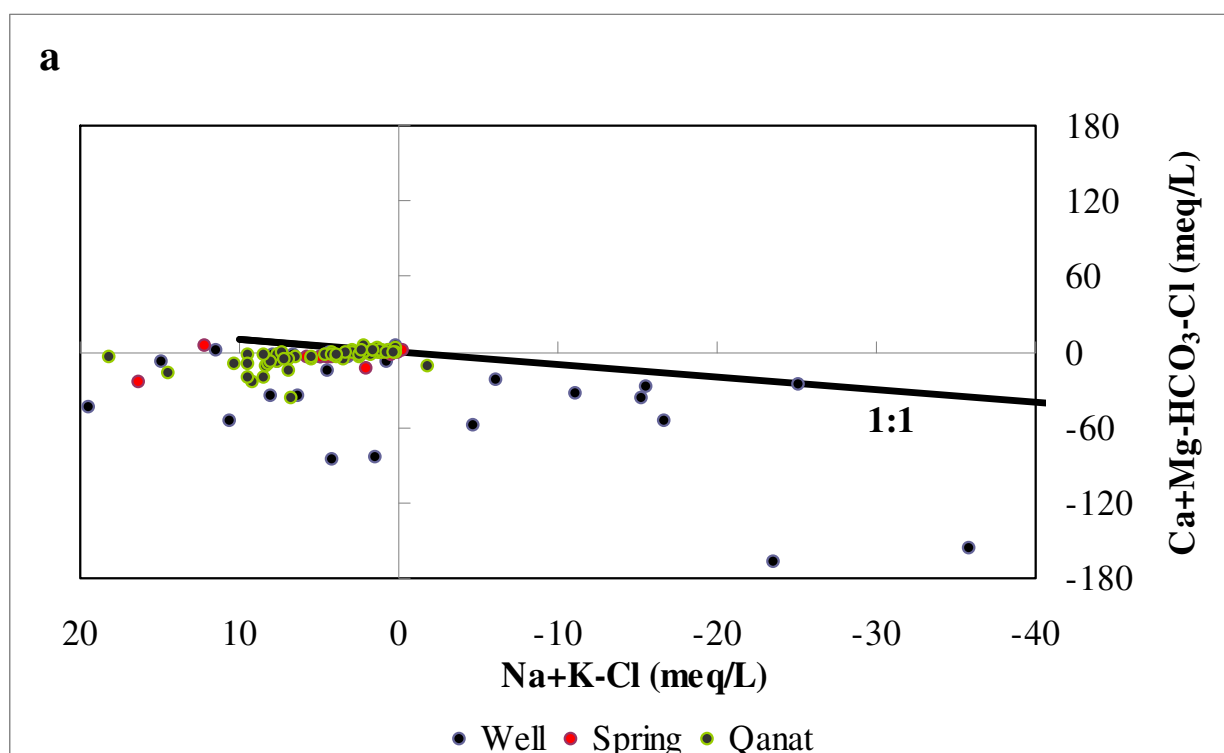


Fig. 8. Plot of the PHREEQC calculated saturation indices (SI) for calcite, dolomite, halite, gypsum and anhydrite versus TDS.

7.6 Ion exchange and reverse ion exchange

Ion exchange involves the replacement of ions adsorbed on the surface of the fine-grained materials of the aquifers by ions that are in the solutions (Todd, 1980). To investigate the importance of ion-exchange processes in groundwater chemistry, we have examined the relationship between the concentration of $(\text{Na}^+ - \text{Cl}^-)$ against $(\text{Ca}^{2+} + \text{Mg}^{2+} - \text{SO}_4^{2-} - \text{HCO}_3^-)$ as suggested by Boghici and Broekhoven (2001) and Jalali (2007). This is shown in Figure 9a. The product of $(\text{Na}^+ - \text{Cl}^-)$ represents excess sodium, that is, sodium coming from the sources other than halite dissolution, assuming that all chloride is derived from halite. Also, the product of $(\text{Ca}^{2+} + \text{Mg}^{2+} - \text{SO}_4^{2-} - \text{HCO}_3^-)$ represents the calcium and/or magnesium coming from sources other than gypsum and carbonate dissolution. In the absence of these reactions, all data should plot close to the origin (McLean et al., 2000). If these processes are significant composition governing processes, the relation between these two parameters should be linear with a slope of -1 (Figure 9a)

The plot of Na/Cl against EC is shown in Figure 9b. This shows that at higher ECs, the ratio of Na/Cl decreases indicating that a) the ratio is approaching seawater ratio of 0.56 and b) reverse ion exchange is far more widespread than ion exchange. Figure 9c shows the amount of $\text{Ca}^{2+} + \text{Mg}^{2+}$ gained or lost relative to that provided by calcite, dolomite and gypsum. When $\text{HCO}_3^- + \text{SO}_4^{2-}$ is low (less than 5 meq/L) and the samples plot on 1:1 line, dissolution of calcite and dolomite is the major process influencing water chemistry but when $\text{HCO}_3^- + \text{SO}_4^{2-}$ is more than 5 meq/L, in addition to calcite and dolomite, dissolution of gypsum is also likely (Kalantary et al. 2007). For all samples of the Safi Abad aquifer, $\text{HCO}_3^- + \text{SO}_4^{2-}$ is >5 meq/L, indicating that gypsum dissolution is likely to occur. Almost all samples lie close to 1:1 line. However, some samples lie above the equiline, pointing to the ion exchange reactions. Also, many samples lie below the equiline indicating reverse ion exchange.



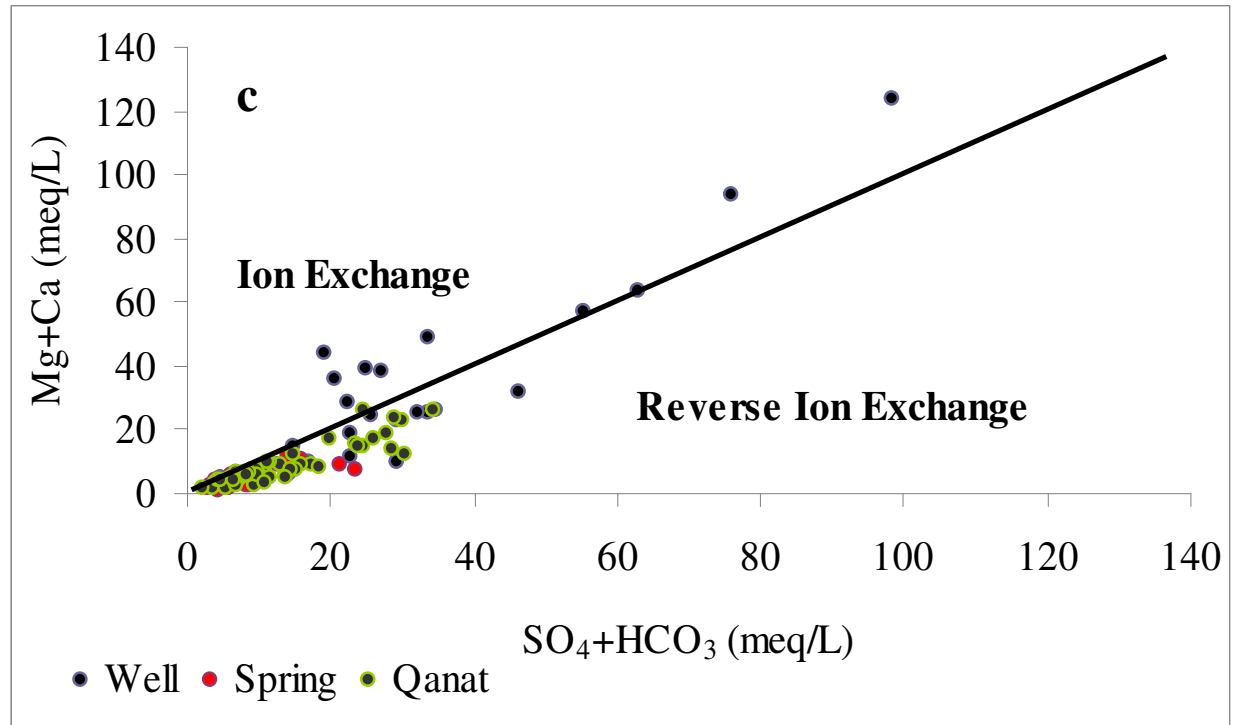
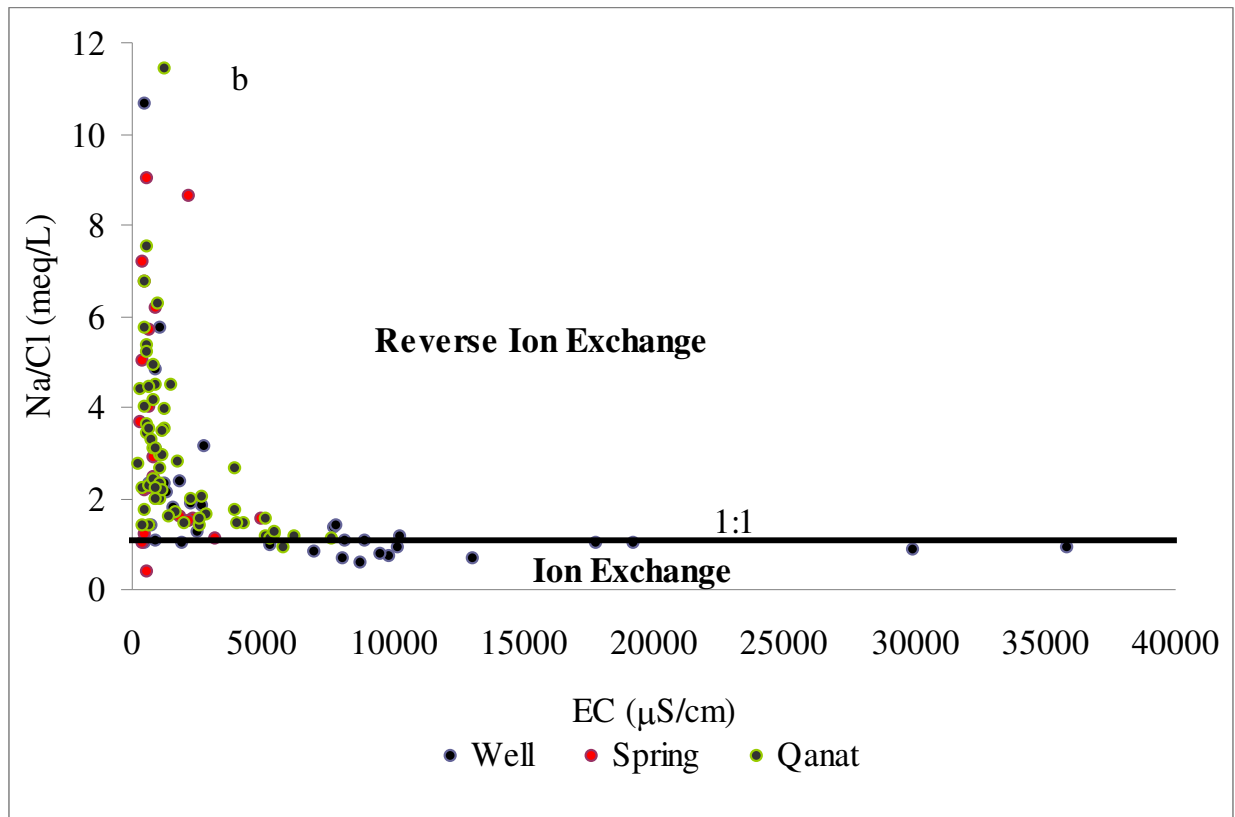


Fig. 9. Plot of a) $\text{K}^+ + \text{Na}^+ - \text{Cl}^-$ versus $\text{Ca}^{2+} + \text{Mg}^{2+} - \text{SO}_4^{2-} - \text{HCO}_3^-$, b) Na/Cl versus EC and c) $\text{HCO}_3^- + \text{SO}_4^{2-}$ versus $\text{Mg}^{2+} + \text{Ca}^{2+}$

8. Implication of hydrogeochemical studies for the management of local groundwater resources

To manage a groundwater resource, we should not only concern about the water quantity and what happens to it, but we should also take care of the quality. Deterioration of groundwater quality may result in serious restrictions on its usages especially when it is used for domestic purposes. The hydrogeochemical approaches implemented in this research shows that a number of groundwater wells and springs have turned into saline most probably due to encroachment of very saline water into these aquifers. Due to aridity, deep saline fossil waters are found in the vicinity of Iranian inland aquifers (e.g. Kazemi, 2011; Kazemi et al., 2001). Therefore, it is well possible that overexploitation of the fresh groundwater resources have impacted the saltwater-freshwater zone, leading to the movement of the fossil waters. If this is the case, this phenomenon will be intensified in the future due to increasing demand for water as well as global warming.

9. Conclusions

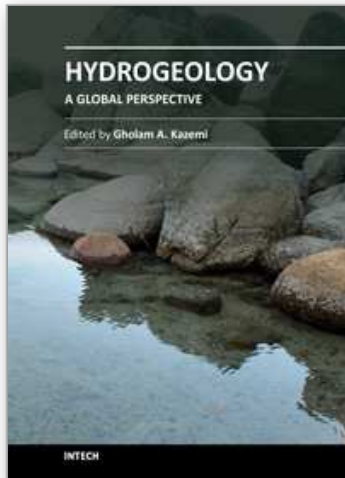
The quality of groundwater resources is as important as their quantity. Hydrogeochemical studies are a useful tool which can help managing quality of these resources. This research shows that rainfall percolating into the mountainous areas of Safi Abad watershed in the northeastern Iran remains mostly fresh while moving in the subsurface environment in the same mountainous area. However, the quality of such groundwaters deteriorates rapidly as it discharges into the adjoining plain. Different hydrogeochemical techniques show that a variety of causes lead to severe salinization of groundwater in the plain including saline water intrusion, leaching of local saline soils into underlying groundwater, intense evaporation and slow rate of groundwater movement. Over exploitation of the groundwater resources during the past 2 decades has probably changed the saltwater-freshwater interface. As a management scenario, the resource managers should first further explore the validity of this hypothesis. Also, it seems that withdrawal of fresh groundwaters in highland areas before letting it to discharge into the aquifer underlying the Safi Abad plain is a possible management scenario to avoid further salinization of this valuable resource.

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The field of groundwater hydrology and the discipline of hydrogeology have attracted a lot of attention during the past few decades. This is mainly because of the increasing need for high quality water, especially groundwater. This book, written by 15 scientists from 6 countries, clearly demonstrates the extensive range of issues that are dealt with in the field of hydrogeology. Karst hydrogeology and deposition processes, hydrogeochemistry, soil hydraulic properties as a factor affecting groundwater recharge processes, relevant conceptual models, and geophysical exploration for groundwater are all discussed in this book, giving the reader a global perspective on what hydrogeologists and co-scientists are currently working on to better manage groundwater resources. Graduate students, as well as practitioners, will find this book a useful resource and valuable guide.

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