

Hydrochemistry and quality assessment of groundwater in the Ardabil area, Iran

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Abstract In the study area, groundwater is the main water resource for various purposes such as drinking, agriculture and industrial. To evaluate the hydrochemical characteristics of groundwater and suitability for drinking, irrigation and industrial purposes, seventy-seven samples were collected and analyzed for various ions. Results show that, groundwater in the study area is mainly hard to very hard, and slightly alkaline-fresh to brackish in nature. According to the hydrochemistry diagrams, the main groundwater types are Ca, Mg-HCO₃, Na-HCO₃ and Na-Cl. Calculation of mineral saturation index indicate that the groundwater samples are saturated with respect to carbonate minerals and under-saturated with respect to sulfate minerals such as gypsum and anhydrite. The mineral weathering, mixing, ion exchange and anthropogenic activity are the dominant hydrogeochemical natural processes. Results of investigating the quality of heavy metals and calculating the heavy metal index indicated that the groundwater of study area is not contaminated with heavy metals. In this research, the various indices were used to determine the quality of groundwater for various uses. Calculate the indices and comparison results with the WHO standards to determine the quality of groundwater for various uses indicated that the most of the groundwater in study area is chemically suitable for drinking, industrial and agricultural uses.

Keywords Ardabil · Groundwater quality · Heavy metals · Hydrochemistry · Hydrogeochemical processes

Introduction

Groundwater is the major source of water for domestic, agricultural and industrial purposes in many countries. The rapid increase in the population of the country has led to large scale groundwater developments in some areas. Intensive cultivating and urban development has caused a high demand on groundwater resources in arid and semi-arid regions of the world and Iran, while putting these resources at greater risk to contamination (Asghari Moghaddam and Najib 2006; Khazaei et al. 2006; Jalali 2007; Giridharan et al. 2008; Aghazadeh and Mogaddam 2010; Esmaeili et al. 2015; Li 2016). The evaluation and management of groundwater resources require an understanding of hydrogeological and hydrogeochemical properties of the aquifer (Umar et al. 2001). Hydrogeochemical processes that are responsible for altering the chemical composition of groundwater vary with respect to space and time.

Chemical characteristics of groundwater play an important role in assessing and classifying the quality of water. Groundwater quality depends on a number of factors such as geology, degree of chemical weathering of the various rock types, quality of recharge water, and water–rock interaction (Domenico and Schwartz 1990; Guler and Thyne 2004; Ayenew et al. 2008; Giridharan et al. 2008; Aly 2015).

During the past decade, various geochemical methods have been successfully used to assess groundwater quality in world and Iran (e.g., Pazand and Javanshir 2014; Barzegar et al. 2016; Fijani et al. 2016; Kavurmac and

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Ustun 2016). Additionally, numerous recent studies have focused on analyzing the natural concentrations of several ions and metals in groundwater, to isolate anthropogenic and natural sources that affect groundwater quality, and establish interactions that take place within the aquifer (e.g., Aghazadeh and Mogaddam 2010; Jacintha et al. 2016; Ehya and Marbouti 2016; Sethy et al. 2016). A large number of groundwater studies also have focused specifically on the assessments of its suitability for drinking and irrigation purposes (Subramani et al. 2005; Kord et al. 2013; Sarikhani et al. 2015; Cao et al. 2016; Golchin and Azhdary Moghaddam 2016).

Groundwater is an important water resource for drinking, agriculture and industrial uses in Ardabil, a city in northwest of Iran. In study area, agriculture is the most important economic activity affecting the changes in groundwater quality by anthropogenic activity. However, in this area, quality assessment of groundwater has received little attention and efforts to use hydrochemical data to solve particular problems are less or nonexistent. Therefore, hydrochemical analysis of the groundwater has become a high priority concern.

Thus main objectives of the present study is (1) to identify the major hydrogeochemical processes (2) to determine the groundwater quality and (3) to delineate regions where groundwater is suitable or unsuitable for drinking, irrigation and industrial purpose.

Description of the study area

The study area is part of Ardabil province in northwest of Iran and located between the latitudes of $38^{\circ} 2'$ to $38^{\circ} 27' N$ and longitudes of $48^{\circ} 6'$, to $48^{\circ} 39' E$ (Fig. 1). Ardabil plain covers an area of 893 km^2 with average altitude of 1400 m.a.s.l. The climate of study area is cold and semi arid. The air temperature is highest in August (25.4°C) and lowest in February (-6.9°C) with an annual average of 9.5°C . According to the data between 1990 and 2010 recorded at the Ardabil meteorological gauging station, the average annual precipitation is 286.3 mm. About 65% of the annual precipitation falls between December and May, including a snowy period from November to March.

The main surface drainages are the Gharehso River and Balikhlichai River, which extends across the entire length of the Ardabil Plain. The average discharge of the Gharehso River is $7.9 \text{ m}^3/\text{s}$, with a maximum discharge of $15.6 \text{ m}^3/\text{s}$ (Ardabil Regional Water Authority (ARWA) 2014). The land use and land cover analysis show that agriculture land is the dominant land use category and comprises 80% of the total land (Kord et al. 2013). The most important economic activity of the area is agriculture and the chief crops are wheat, potatoes, barley, corn, sugar beet, and beans.

Base geological structural deviations of Iran, the investigated area is located in the Alborz-Azerbaijan zone of Iran and it is covered by Jurassic to Quaternary sediments (Nabavi 1976). The exposed lithological units of the Ardabil plain range in age from Jurassic to Quaternary (Fig. 2). Jurassic formations in the study area chiefly comprise alternation of shale, sandstone, marl and siltstone with intercalation of limestone. Crystallized thick bedded limestone with alternation of tuffaceous sandstone is cretaceous formation in study area. Trachyandesite, lapillus, tuff, volcanic breccias and granodiorite igneous rocks of Tertiary age are outcropped in many mountainous areas. Quaternary volcanic rocks in the study area were related to Sabalan volcanic activity and which mainly consists of lava andesitic and basalt. As a consequence, the Quaternary deposits are mainly characterized by clay, silted gravel with medium grained.

The result obtained from well logs, indicates that in Ardabil plain aquifer is unconfined and the thickness of the alluvium aquifer in average is 65 m (Ardabil Regional Water Authority (ARWA) 2014). The maximum thickness is about 220 m, which lies in the east and central part of the plain. The general groundwater flow direction in the aquifer is from S, SE to NW, and its depth to water table varies from 0.5 to 56 m below the ground level. Abstraction rate from the aquifer during the water year 2009–2010 is about 219.3 million cubic meters. Eighty-five percent of exploited groundwater is used for agriculture, 14% for drinking and 1% is used by the industry. In study area, the maximum and minimum water level is in May and September, respectively (Ardabil Regional Water Authority (ARWA) 2014). The average fall of groundwater level in the last 30 years in the Ardabil aquifer is about 0.32 m (Ardabil Regional Water Authority (ARWA) 2014), that the reason is intense over-pumping and rapid development of agriculture and industry.

Materials and methods

In this research to evaluate the quality of groundwater, 77 groundwater samples were collected from shallow and deep wells and springs of the area during September 2010. Groundwater samples were analyzed for various chemical parameters as described by the American Public Health Association (APHA 1995). These parameters include hydrogen ion concentration (pH), electrical conductivity (EC), total hardness (TH), total alkalinity (TA), total dissolved solids (TDS), and important cations such as calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}), potassium (K^{+}) and iron (Fe^{2+}) as well as anions such as carbonates (CO_3^{2-}), bicarbonates (HCO_3^{-}), chlorides (Cl^{-}), nitrates (NO_3^{-}), Phosphate (PO_4^{3-}), sulfates (SO_4^{2-}) and heavy

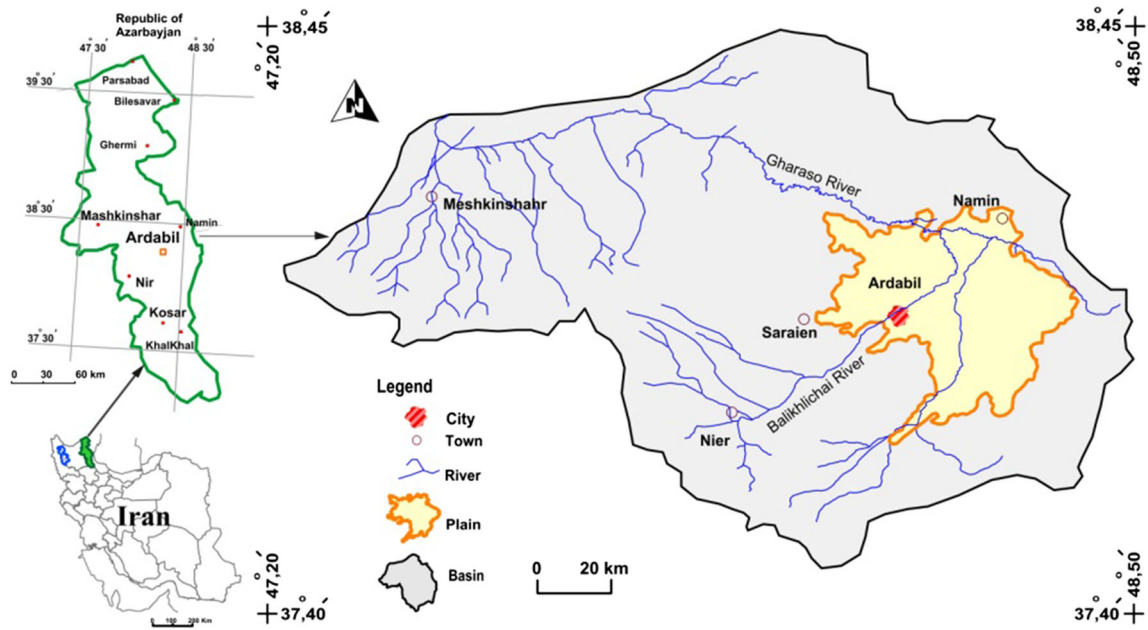


Fig. 1 Location map of study area and drainage pattern

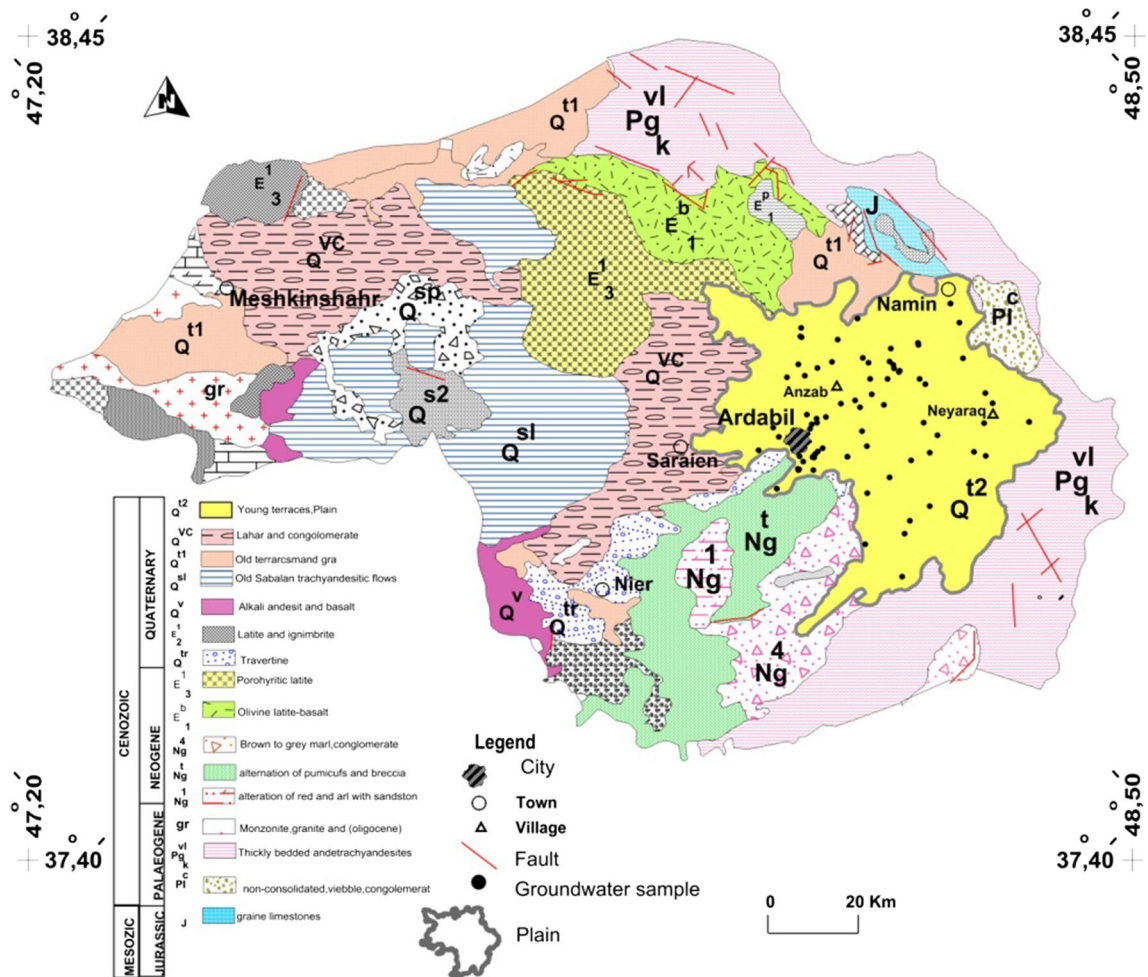


Fig. 2 Geology map of study area and groundwater sample points

metals (As, Cu, Cr, Ni, Zn, Mn, Hg). The pH and electrical conductivity (EC) were measured in field using portable conductivity meters. Na^+ and K^+ were determined by flame photometer. Fe^{2+} , PO_4^{3-} , NO_3^- and SO_4^{2-} were analyzed by spectrophotometer. TH and TA as CaCO_3 , Ca^{2+} , CO_3^{2-} , HCO_3^- and Cl^- were analyzed by volumetric method. Mg^{2+} was calculated from TH and Ca^{2+} contents. Heavy metals were analyzed by ICP-Mass methods. Based on the physico-chemical analyses, some important parameters and indices were calculated. Iso-concentration maps were constructed using the Arc GIS software. Saturation indices for carbonate and sulfate minerals and chemical facies were also computed through the computer programmers, PHREEQC and AUCHEM. The correlation of the analytical data has been attempted by plotting different graphical representation for the classification of water and to study the suitability of groundwater for utilitarian purposes by ascertaining various factors on which the chemical characteristics of water depend. The suitability of the groundwater for drinking, industrial, domestic, and irrigation purposes was evaluated by comparing the values of different water quality parameters with those of the World Health Organization (WHO 2004) guideline values for drinking water.

Results and discussion

Chemical characteristics of groundwater

The pH in groundwater samples varied from 6.37 to 8.14 that indicating slightly acidic to slightly basic in nature. In the study area, the electrical conductivity (EC) of groundwater varies widely between 366 and 4981 $\mu\text{S}/\text{cm}$. In 52% of the groundwater samples the enrichments of salts are low ($\text{EC} < 1500 \mu\text{S}/\text{cm}$), 43% of samples are medium ($\text{EC}: 1500 \text{ and } 3000 \mu\text{S}/\text{cm}$) and 5% of samples are high ($\text{EC} > 3000 \mu\text{S}/\text{cm}$). Higher value of EC can be due to the dissolution of minerals and the influence of anthropogenic contamination, causing increases in ionic concentration. Total dissolved solids (TDS) in the study area vary from 92 to 4508 mg/l L and groundwater is generally fresh to brackish in nature.

Large variations in EC and TDS values are primarily attributed to geochemical process like ion exchange, evaporation, sediment dissolution, and rainwater infiltration (Ehya and Marbouti 2016) and anthropogenic sources (such as domestic sewage, septic tanks, and agricultural activities) (Hosseini-fard and Mirzaei Aminiyan 2015).

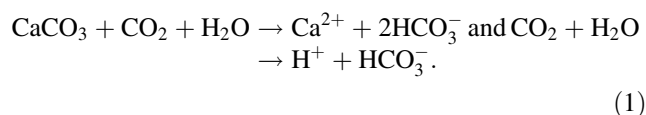
The value of total alkalinity (TA) is observed from 78 to 774 mg/L (Table 1) that, it is caused by HCO_3^- ion only, as the pH is between 6.37 and 8.14. This alkalinity also suggests that dissolution has been occurred due to

interaction between soil and rainwater, giving groundwater alkaline character (Singh et al. 2012).

The TH (as CaCO_3) in the groundwater of study area is between 49 and 1519 mg/L. According to the classification of TH, approximately 2% of the groundwater samples come under the fresh, 10% are moderately hard category, 23% of the groundwater samples come under the hard category and the remaining 66% of the groundwater samples fall in the very hard category. Cation concentrations and ratios can trace water-rock interaction processes, such as mineral weathering and cation exchange (Han et al. 2009). The concentrations of calcium range from 12.5 to 404 mg/l (Fig. 3a; Table 1), which is derived from calcium rich minerals like feldspars, pyroxenes and amphiboles. The major source of magnesium (Mg^{2+}) in the groundwater is due to ion exchange of minerals in rocks and soils by water. The concentrations of Mg^{2+} found in the groundwater samples vary in the range 1.4–121.7 mg/l (Fig. 3b; Table 1).

In the study area, the Na^+ and K^+ concentrations in groundwater range from 6.2 to 399 and 0.4 to 138 mg/l, respectively (Fig. 3c; Table 1). High concentrations of Na^+ in the groundwater are attributed to cation exchange among minerals, anthropogenic activities (spatial agricultural activities) and poor drainage conditions.

The carbonate and bicarbonate concentration in groundwater is derived from carbonate weathering, as well as dissolution of carbonic acid in the aquifers (Kumar et al. 2009) (Eq. 1).



The value of HCO_3^- is observed from 96 to 945 mg/L, which is the dominant ion in some of the groundwater (Fig. 3d; Table 1). The higher concentration of HCO_3^- in the water infers a dominance of mineral dissolution. The carbonates available in carbonate rocks could have been dissolved during irrigation, rainfall infiltration and groundwater movement, and added to the groundwater system with recharging water (Singh et al. 2012). The concentration of chloride ranges from 9 to 667 mg/l and increases from the recharge to discharge area and is dominant anion in some of the samples (Fig. 3e; Table 1). Sulfate varies from 4.2 to 1286 mg/l (Fig. 3f; Table 1). The abundance of the major ions in groundwater is in following order Ca^{2+} , $\text{Na}^+ > \text{Mg}^{2+} > \text{K}^+$ and HCO_3^- $\text{SO}_4^{2-} > \text{Cl}^- > \text{CO}_3^{2-}$. Minimum, maximum and average values of physical and chemical parameters of groundwater samples are presented in Table 1.

The concentration of nitrate (NO_3^-) in the study area varies from 2.26 to 130 mg/l with an average value of

Table 1 Summary statistics of the analytical data in study area

Parameters	Units	Minimum	Maximum	Mean	Standard deviation	Most desirable limits (WHO 2004)	Maximum allowable limits (WHO 2004)
pH	–	6.37	8.14	7.37	0.36	5	8.5
EC	μmoh/cm	366	4981	1424	830	1400	–
TDS	mg/l	92	4508	918.6	705	500	1500
Na	mg/l	6.2	399.2	131.5	89.9	–	200
K	mg/l	0.4	138.5	23.76	26	–	20
Ca	mg/l	12.5	404.6	112.6	88.6	75	200
Mg	mg/l	1.4	121.7	42.3	24.36	50	150
Cl	mg/l	8.9	667.4	161.3	134.5	200	600
HCO ₃	mg/l	95.6	945.3	381.23	190.3	–	300
SO ₄	mg/l	4.2	1286	273.4	224	299	400
PO ₄	mg/l	0	1.72	0.35	0.4	–	250
NO ₃	mg/l	0	130	26.5	24.7	–	50
F	mg/l	0.15	1.18	0.51	0.23	–	1.5
Fe	g/lμ	0.95	181	29.7	29.3	–	300
Mn	g/lμ	0	16.43	5.5	4	–	500
Cr	g/lμ	0	12.8	0.53	1.5	–	50
As	g/lμ	0	9.42	0.43	1.3	–	10
Cu	g/lμ	0	11.4	2.2	2.3	–	2000
Hg	g/lμ	0	3.12	0.3	0.44	–	6
TH	mg/l	48.8	1518.58	457.95	292.77	100	500
TA	mg/l	78	774	312	156	–	–
SAR	–	0.22	6.1	2.7	1.5	–	–
%Na	%	9	97.3	40.8	17.32	–	–
RSC	meq/l	–18.59	5.86	–2.9	4.53	–	–
PI	%	30.87	93	59	18.98	–	–
SI _{calcite}	–	–2.01	1.12	–0.39	0.69	–	–
SI _{dolomite}	–	–5.62	1.29	–1.59	1.36	–	–
SI _{gypsum}	–	–2.48	–0.44	–1.11	0.42	–	–
SI _{anhydrate}	–	–2.73	–0.7	–1.36	0.42	–	–
SI _{aragonite}	–	–2.17	0.96	–0.54	0.68	–	–

EC electrical conductivity, TDS total dissolved solids, TH total hardness, SAR sodium adsorption ratio, RSC residual sodium carbonate, %Na sodium percent, PI permeability index, SI saturation index

26.4 mg/l (Table 1). Groundwater can be contaminated by fertilizer application, human and livestock sewage, deposition of plants and other wastes rich in nitrates (Chukwura et al. 2015; Vincy et al. 2015).

The nitrate concentration was relatively high in some of the groundwater samples. Spatial distribution of the nitrate concentration in the groundwater is illustrated in Fig. 3g. The concentration of nitrate was relatively high in around the Ardabil city, south and south-eastern parts of the study area. However, in most of the samples (88%), the nitrate concentration was below the permissible drinking limits set by the WHO (50 mg/l). In the study area, agricultural activities are mainly dependent on the groundwater resources. A very high nitrate concentration is observed along the Balekhli River, which may be due to the seepage

of irrigation waters from agricultural fields where chemical fertilizers are being used in discriminately (Kord et al. 2013). The high nitrate values in around the Ardabil city are attributed to the overflowing sewage water. In addition to this, phosphate concentrations were found to vary from 0.01 to 1.72 mg/L (Fig. 3h; Table 1).

Fluoride (F[–]) is one of the main trace elements in groundwater, which generally occurs as a natural constituent. The concentration of fluoride in groundwater in study area varied from 0.15 to 1.18 mg/l (Table 1), and the mean value was 0.5 mg/l. Concentration of fluoride was low in the major part of the study area, which indicates limited lithogenic input of fluoride ion in groundwater samples. The spatial distribution of fluoride ion concentration in groundwater is illustrated in the Fig. 3i. The iron

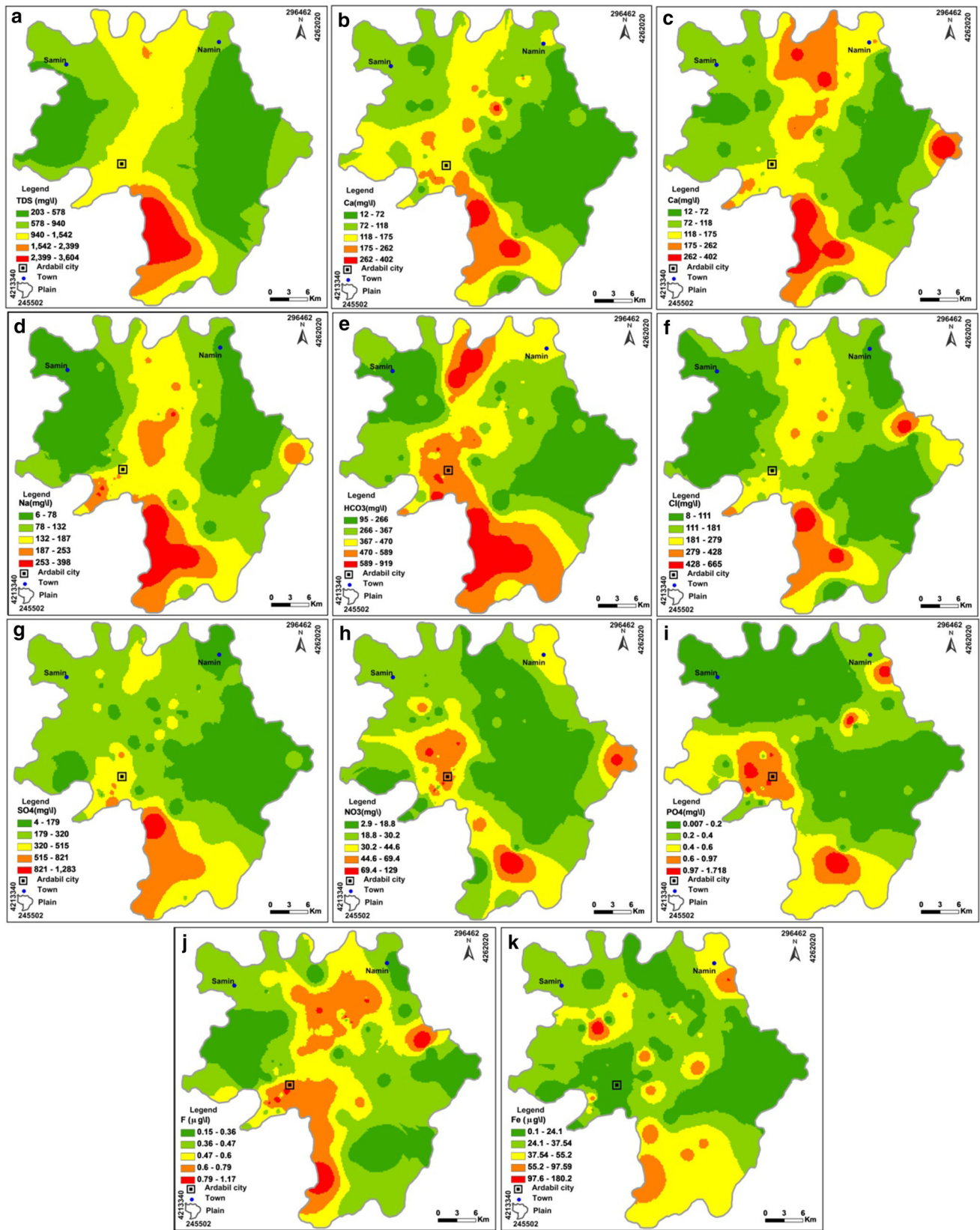


Fig. 3 Spatial Distribution of TDS, Ca, Mg, Na, HCO₃, Cl, SO₄, NO₃, PO₄, F and Fe in the Ardabil area

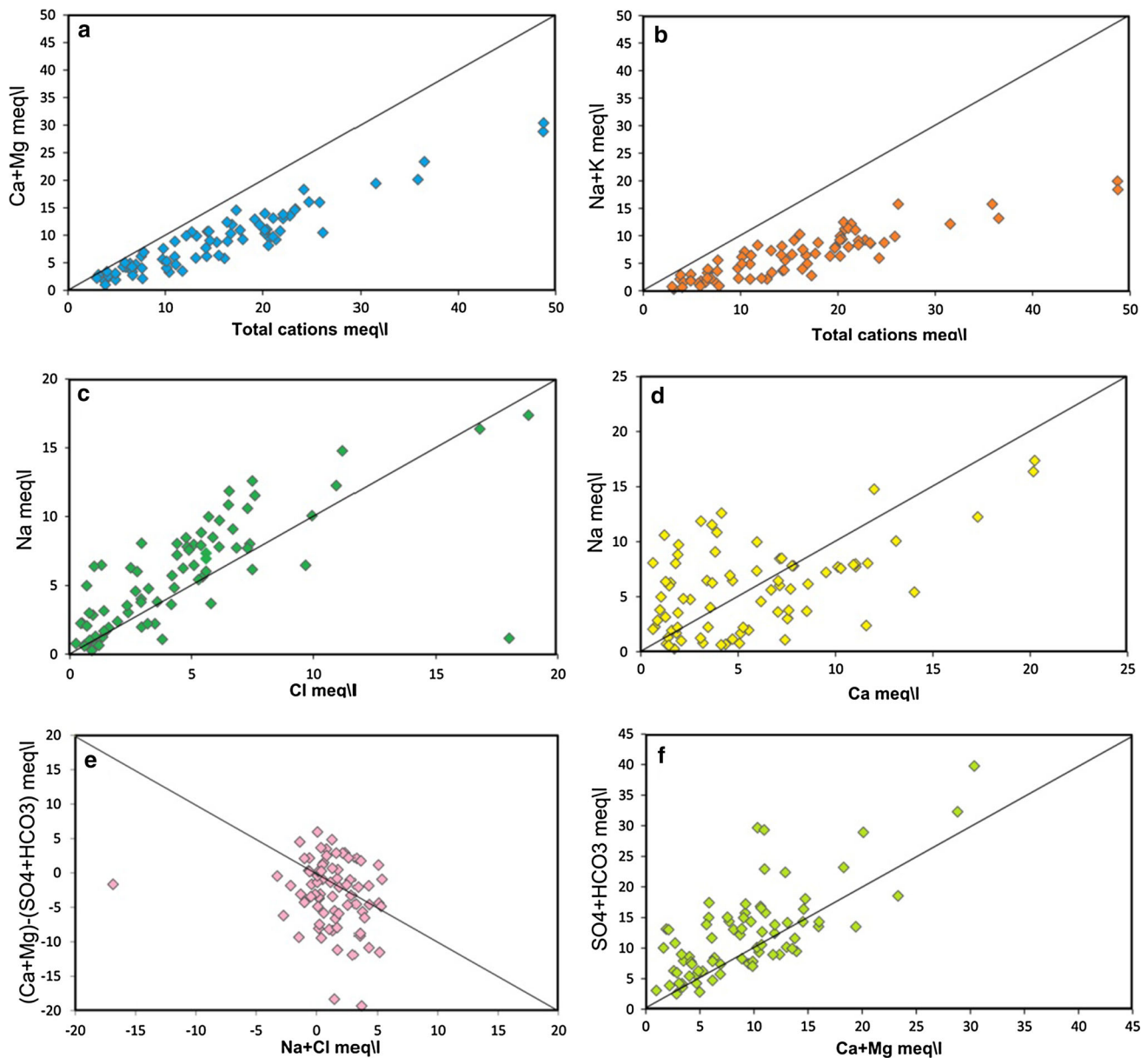


Fig. 4 Graphs of different parameters in groundwater of study area (solid line denotes 1:1)

concentration generally varies from 0.001 to 0.18 mg/l with an average concentration of 0.003 mg/l (Table 1). The high iron concentration is observed mainly in the northwest and also in the east parts of the study area. A very high concentration of iron in groundwater is found in northeast side villages of Barogh (0.18 mg/l). All the samples from study were within the prescribed standard desirable limit set by the WHO.

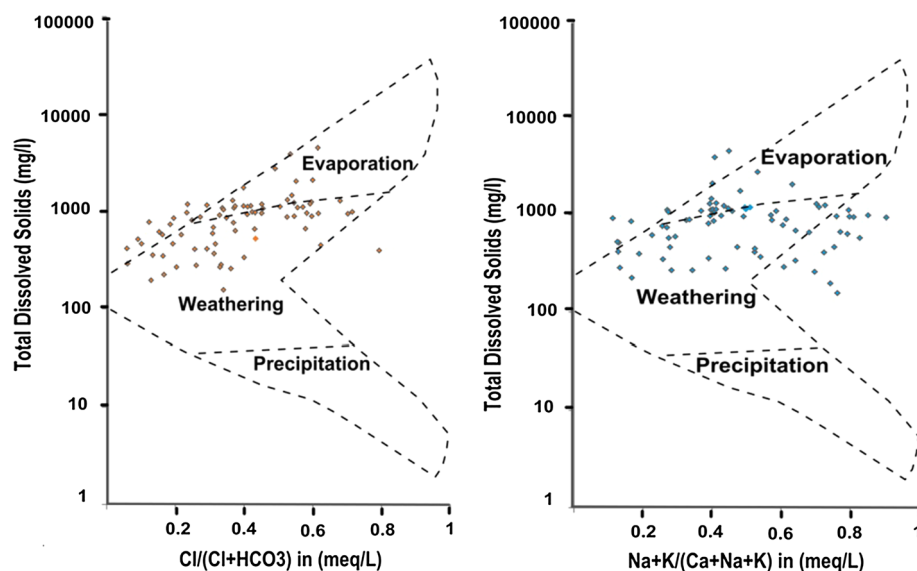
Hydrochemical evaluation

The major ion chemistry of groundwater is a powerful tool for determining solute sources and for describing groundwater evolution as a result of water–rock interaction

leading to the dissolution of carbonate minerals, silicate weathering and ion exchange processes (Kumar et al. 2009). Results from the chemical analyses were used to identify the geochemical processes and mechanisms in the groundwater aquifer system. The chemical data of the groundwater samples is plotted for $\text{Ca}^{2+} + \text{Mg}^{2+}$ and $\text{Na}^{+} + \text{K}^{+}$ vs. TC (total cation) (Fig. 4a, b). The graphs show that the most of the samples far below the theoretical line (1:1), indicating supply of cations via ion exchange, silicate weathering and/or soil salts is more significant.

High concentration of Na with respect to Cl or depletion of Na with respect to Cl is the evident of cation exchange reactions (Sethy et al. 2016). Result shows that, most of the samples have a Na/Cl ratio around or above 1, indicating

Fig. 5 Mechanisms governing groundwater chemistry in Gibbs diagram



that an ion exchange process is prevalent in the study area (Fig. 4c). Also, Fig. 4d shows the ion exchange reactions, where Na^+ is plotted against Ca^{2+} , in which Ca^{2+} levels are observed between 0.6 and 20 meq/l, while Na^+ levels are found between 0.3 and 17.4 meq/l. If the ion exchange is the only controlling process of groundwater composition, the relation between $(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{SO}_4^{2-} + \text{HCO}_3^-)$ and $\text{Na}^+ - \text{Cl}^-$ should show negative linear trend with a slope of unity, considering the participation of cations in the ion exchange reaction (Fisher and Mullican 1997). In Fig. 4e, the samples show a trend of $(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{SO}_4^{2-} + \text{HCO}_3^-)$ versus $\text{Na}^+ - \text{Cl}^-$ with a negative slope of less than unity, but they spread above and below the linear trend. This suggests that the controlling of groundwater quality depends not only on the involvement of ion exchange process, but also on the involvement of other processes. Otherwise, the spreading of sample points above and below the linear trend should not be expected. The graph of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{SO}_4^{2-} + \text{HCO}_3^-$ will feature a nearly 1:1 line if dissolutions of calcite, dolomite and gypsum are the dominant reactions in the system (Srivastava and Ramanathan 2008; Fijani et al. 2016). Ion exchange tends to shift the points right because of the excess of $\text{SO}_4^{2-} + \text{HCO}_3^-$ ions, which may be due to anthropogenic input in the groundwater system (Fisher and Mullican 1997; Barzegar et al. 2016). The graph of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{SO}_4^{2-} + \text{HCO}_3^-$ (Fig. 4f) shows that most of samples fall above the 1:1 ratio line and show a deficiency of $\text{Ca}^{2+} + \text{Mg}^{2+}$ relative to $\text{SO}_4^{2-} + \text{HCO}_3^-$. Higher concentration of Na^+ in the groundwater is an index of ion exchange process. Also, the scatter diagram of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{SO}_4^{2-} + \text{HCO}_3^-$ shows that silicate weathering was the primary process involved in the evolution of groundwater. If bicarbonate and sulfate are

dominating than calcium and magnesium, it reflects that silicate weathering was dominating and, therefore, was responsible for the increase in the concentration of HCO_3^- in groundwater. The study of Ca/Mg ratio also revealed that the dissolution of silicate minerals was one of the prime processes involved in attaining the present chemical makeup of the groundwater. The $\text{Ca}^{2+}/\text{Mg}^{2+}$ ratio of 1 indicated dissolution of dolomite and of >2 reflected an effect of silicate minerals on the groundwater chemistry; it also suggested dolomite dissolution for Ca^{2+} , Mg^{2+} concentration in groundwater (Barzegar et al. 2016). Majority of the samples have $\text{Ca}^{2+}/\text{Mg}^{2+}$ ratio between 0.6 and 3.3, indicating effect of silicate minerals on the groundwater chemistry.

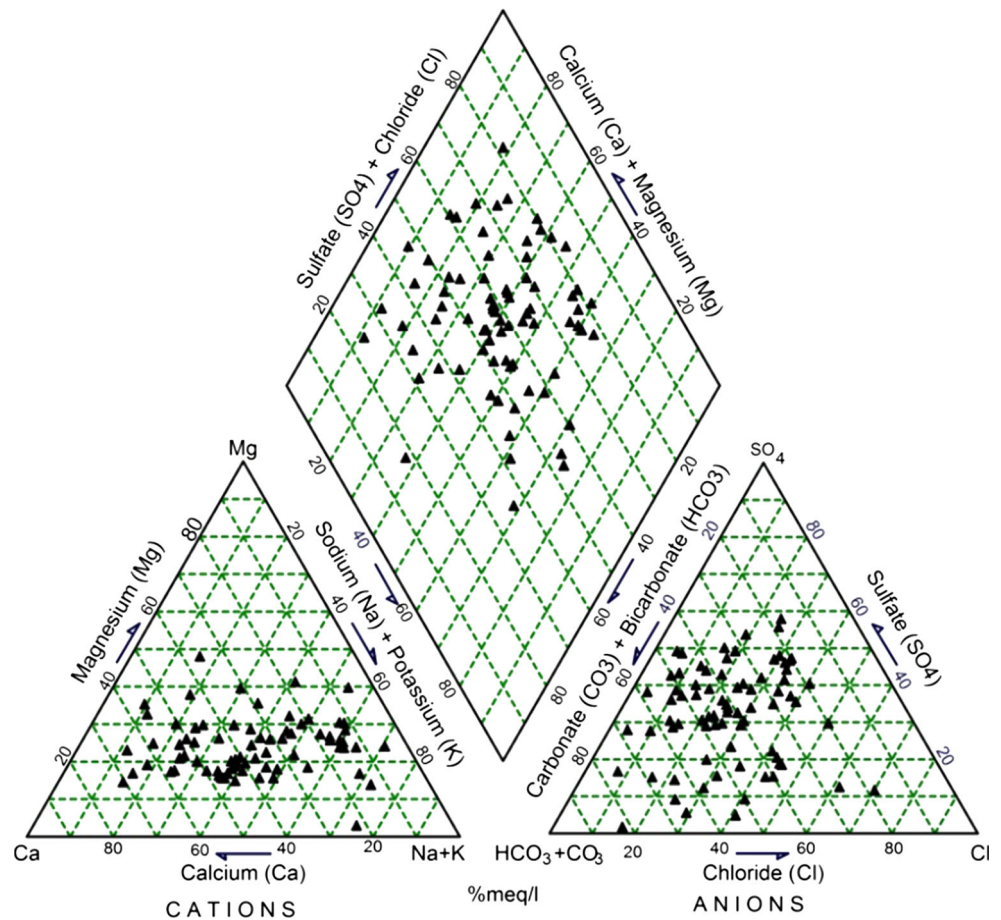
The chemical data of groundwater sample were plotted in Gibbs's diagrams (Gibbs 1970) (Fig. 5). The clustering of the data points in Gibbs diagram indicates that most of samples the chemical weathering of rock-forming minerals is influencing the groundwater quality and also in some of samples evaporation is influencing the groundwater quality.

Hydrochemical facies and water type

The hydrochemical facies reflect the effect of chemical processes in the lithological environment and the contained groundwater flow patterns (Back 1966; Freeze and Cherry 1979).

In this study, we used to Piper and Durov diagrams to determine the hydrochemical facies of groundwater. The values obtained from the groundwater sample analyzing, and their plot on the Piper's diagrams (1944) (Fig. 6) reveal that in the most samples no cation–anion exceed 50%. However the major cations are Ca^{2+} , Na^+ and the anion is HCO_3^- , Cl^- . In the study area, 22% samples

Fig. 6 Chemical facieses of groundwater in Piper diagram



belong to the Ca, Mg-HCO₃, 53% of the samples belong to the Na-HCO₃, 18% of the samples belong to the Na-Cl and 7% of the samples belong to the mixed water types. The sodium-chloride water type in study area is due to the low velocity of groundwater, ion exchange, long time contacts of water and the type of the rocks.

To determine the domain hydrochemical processes and type of ion exchange chemical data of groundwater samples have been plotted on the Durov diagram (Durov 1948; Lloyd and Heathcode 1985). According to Durov diagram (Fig. 7) the majority of samples fell in mixed zone, indicating in the most samples none dominate cation–anion and the major hydrochemical processes are mixing and normal ion exchange.

Saturation indices and water mineral equilibrium

The equilibrium state of the water with respect to a mineral phase can be determined by calculating a saturation index (SI) using analytical data (Garrels and Mackenzie 1967). Changes in saturation state are useful to distinguish different stages of hydrochemical evolution and help identify which geochemical reactions are important in controlling

water chemistry (Drever 1997; Langmuir 1997; Coetsiers and Walraevens 2006). The saturation index of a mineral is obtained from Eq. 2 (Garrels and Mackenzie 1967).

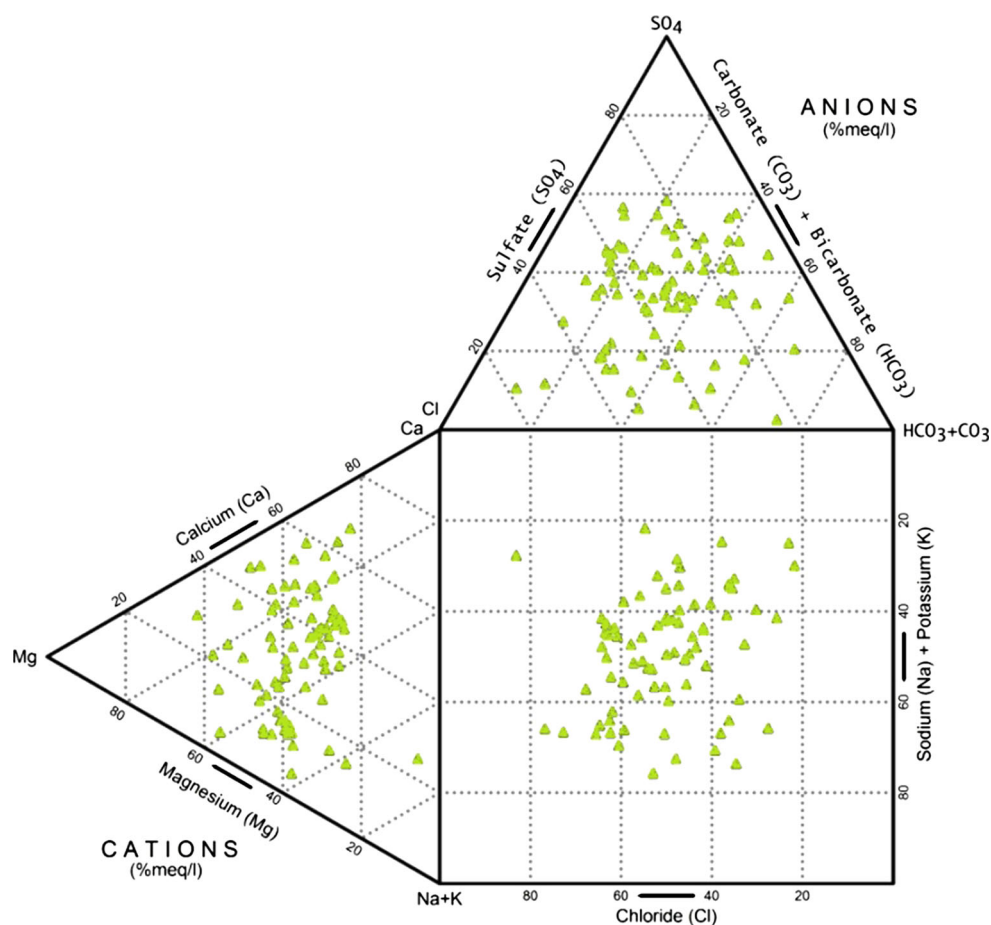
$$SI = \log \frac{IAP}{KT}, \quad (2)$$

where IAP is the ion activity product of the dissociated chemical species in solution, Kt is the equilibrium solubility product for the chemical involved at the sample temperature.

The saturation indexes were determined using the hydrogeochemical equilibrium model, PHREEQC for Windows (Parkhurst and Appelo 1999).

An index (SI), less than zero, indicate that the groundwater is under-saturated with respect to that particular mineral. An index (SI), greater than zero, specifies that the groundwater being supersaturated with respect to the particular mineral phase and therefore incapable of dissolving more of the mineral. Such an index value reflects groundwater discharging from an aquifer containing ample amount of the mineral with sufficient resident time to reach equilibrium (Appelo and Postma 1996; Langmuir 1997). In Table 1 the SI for calcite, dolomite, anhydrate and gypsum are shown. The plot of saturation indices of calcite (SIC)

Fig. 7 Chemical facieses of groundwater in Durov diagram



versus dolomite (SID) demonstrates that the 91% of the waters are under-saturated with respect to dolomite and 68% are under-saturated with respect to calcite (Fig. 8). The SID and SIC values are 1.29 to -5.62 and 1.12 to -2.01 , respectively. Figure 9 shows the plots of SI against TDS for all the investigated water. All of groundwater samples were under-saturated with respect to gypsum and anhydrite because the major lithological units of the around Ardabil plain are volcanic rocks and there are not insufficient evaporate minerals for solution.

Heavy metals concentrations and classification of groundwater base heavy metal index

The most common heavy metals of concern in water are chromium (Cr), iron (Fe), nickel (Ni), and zinc (Zn), copper (Cu), lead (Pb), arsenic (As), mercury (Hg) and cadmium (Cd). Excess levels of these heavy metals can damage human health and ecosystems (Ehya and Marbouti 2016). The concentrations of heavy metals in groundwater samples from different location are given in Table 4, and compared with WHO standard (2004). Results of investigating the quality of heavy metals show that the abundance of the heavy metals in groundwater

are in following order: $Fe > Mn > Zn > Cu > Cr > Ni > As > Hg > Ce$ (Table 1).

The Fe and Mn in the groundwater of study area ranged from 0.95 to 181 $\mu\text{g/l}$ with an average value of 29.7 $\mu\text{g/l}$ and from 0 to 16.4 $\mu\text{g/l}$ with an average value of 5.5 $\mu\text{g/l}$, respectively. Chrome levels in the groundwater of study area ranged from 0 to 12.8 $\mu\text{g/l}$, with an average of 0.53 $\mu\text{g/l}$ (Table 1). The average Arsenic concentration in the groundwater was 0.4 $\mu\text{g/l}$, with a range of 0–1.88 $\mu\text{g/l}$ (Table 1). The Cu and Hg in the groundwater of study area ranged from 0 to 11.4 $\mu\text{g/l}$ with an average value of 2.21 $\mu\text{g/l}$ and from 0 to 3.12 $\mu\text{g/l}$ with an average value of 0.29 $\mu\text{g/l}$, respectively. The heavy metal concentrations sampled from the groundwater of study area are also compared with WHO Standard (2004) (Table 1). The result shows that almost in the all of samples concentrations of heavy metals are lower than the corresponding WHO values.

In this study, two heavy metal index (Heavy metal Pollution Index (HPI) developed by Prasad and Bose (2001) and the Heavy metal Evaluation Index (HEI) proposed by Edet and Offiong (2002) used to classify the groundwater. To calculate the HPI and HEI index the groundwater of study area, the concentration value of seven

Fig. 8 Plot of saturation indices (SI) for calcite (SIC) versus dolomite (SID)

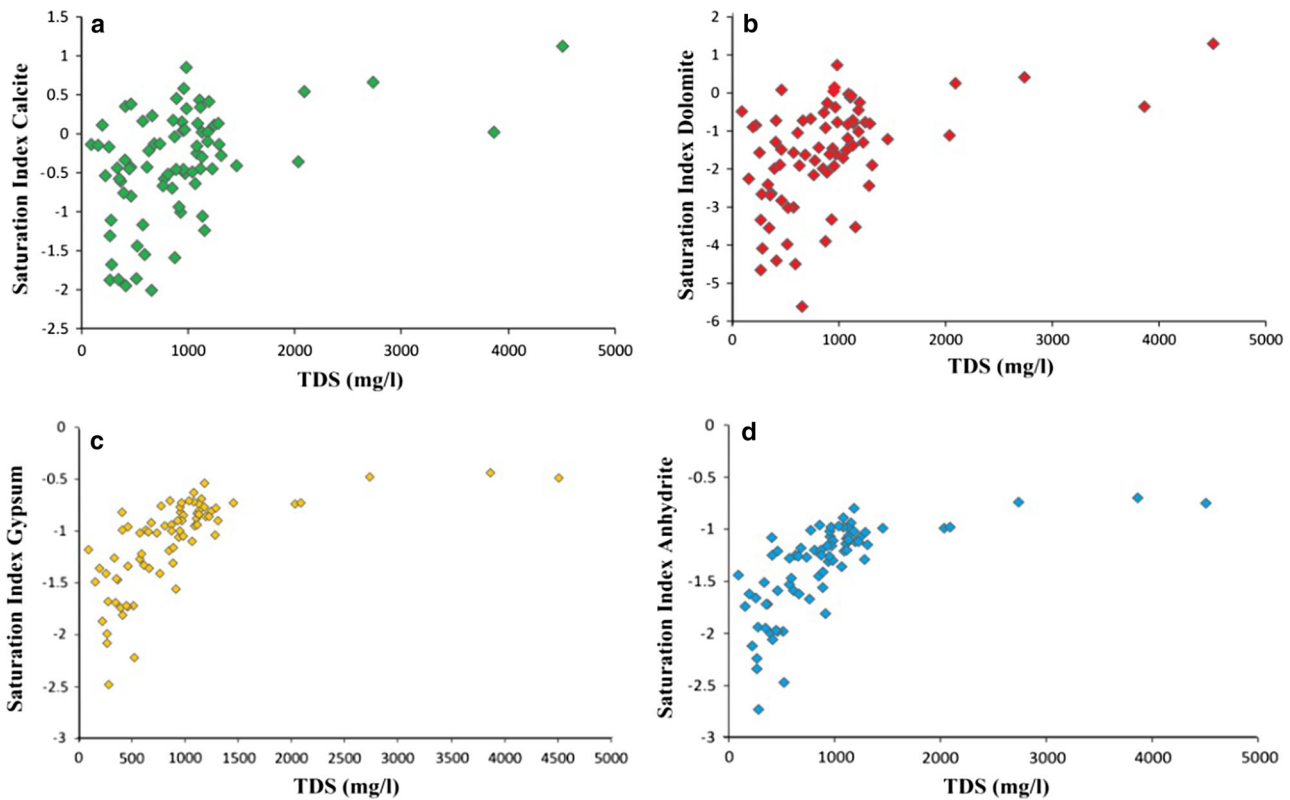
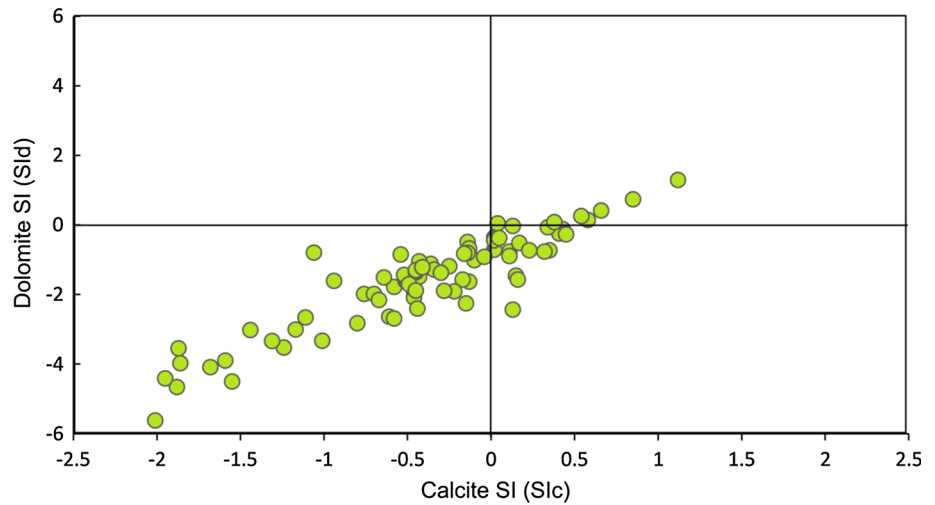


Fig. 9 Plot of saturation indices (SI) versus total dissolved solid (TDS)

heavy metals (Cr, Cu, Zn, Ni, As, Mn, and Hg) have been taken into account.

The HPI index represents the composite influence of heavy metals on the overall quality of water. The HPI is a method of assigning a rating or weighting (W_i) for each chosen parameter and selecting the pollution parameter on which the index is to be based. The rating is a value between zero and one, reflecting the relative importance of individual quality considerations and defined as inversely proposal to the recommended standard for each

heavy metal. Water quality and its suitability for drinking can be examined by determining its quality index (Mohan et al. 1996; Prasad and Kumari 2008; Ehya and Marbouti 2016). For this study, the concentration limits (i.e., maximum admissible concentration for drinking water (MAC), highest permissible value for drinking water (S_i) and the maximum desirable value (I_i) for each heavy metal) were taken from the international WHO standard. The HPI index can be calculated as Eq. 3 (Mohan et al. 1996):

$$\text{HPI} = \frac{\sum_{i=1}^n W_i Q_i}{\sum_{i=1}^n W_i}, \quad (3)$$

where Q_i is the sub-index of the i the parameter, W_i is the unit weighting of the i the parameter and n is the number of parameters considered. The sub-index (Q_i) of the parameter is calculated by Eq. 4:

$$Q_i = \sum_{i=1}^n \frac{|M_i - I_i|}{S_i - I_i} \times 100, \quad (4)$$

where M_i is the monitored value of the i th parameter, I_i is the ideal value of i th parameter, and S_i the standard value for the i th parameter.

In the study area, the median of HPI was 18.03 (Table 2), which is below the critical value of 100. The maximum and minimum value of HPI are 41.5, 3.1, respectively. The results show that groundwater of study area are not contaminated with heavy metals.

The HEI index gives an overall quality of the water with respect to heavy metal content. The HEI is computed as Eq. 5:

$$\text{HEI} = \sum_{i=1}^n \frac{H_c}{H_{\text{mac}}}, \quad (5)$$

where H_c is the monitored value of the i th parameter and H_{mac} the maximum admissible concentration of the i th parameter (Edet and Offiong 2002).

The median of HEI was 0.0013 (Table 2), which is below the critical value so that groundwater of study area is not contaminated with heavy metals.

Drinking and irrigation water quality

The analytical results have been evaluated to ascertain the suitability of groundwater of the study area for drinking and agricultural uses. The drinking water quality is evaluated by comparing with the specifications of TH and TDS set by the WHO (2004). According to WHO (2004) specification TDS up to 500 mg/l is the highest desirable and up

to 1500 mg/l is maximum permissible (Table 3). Based on this classification, 92% of samples belongs to maximum permissible category and remaining samples are exceeding the maximum allowable limits. Maximum allowable limit of TH for drinking is 500 mg/l and the most desirable limit is 100 mg/l as per the WHO international standard. Based on this classification, it indicates that 40% of the samples exceed the maximum allowable limits; such water cannot be used for domestic purposes, because it coagulates soap lather.

The development and maintenance of successful irrigation projects involve not only the supplying of irrigation water to the land but also the control of salt and alkali in the soil (Haritash et al. 2008; Li et al. 2016). Salinity and indexes such as, sodium absorption ratio (SAR), sodium percentage (%Na), residual sodium carbonate (RSC), and permeability index (PI) are important parameters for determining the suitability of groundwater for agricultural uses (Srinivasa 2005; Raju 2007; Kord et al. 2013).

Electrical conductivity is a good measure of salinity hazard to crops as it reflects the TDS in groundwater. The Wilcox (1955) classified ground waters on the basis of electrical conductivity (Table 3). Based on this classification, %22 of samples belongs to the good category; %70 of samples belongs to the permissible category and %8 doubtful categories. Stuyfzand (1989) classified water on the basis of Cl^- ion concentration into eight divisions as shown in Table 3. Based on this classification, %13 of groundwater samples were very fresh, %38 fresh, %40 fresh-brackish and %9 were brackish on the basis of Cl^- concentration.

Sodium adsorption ratio (SAR) is an important parameter for determining the suitability of groundwater for irrigation because it is a measure of alkali/sodium hazard to crops (Subramani et al. 2005). SAR is defined by Karanth (1987) as Eq. 6

$$\text{SAR} = \frac{\text{Na}^+}{\sqrt{\frac{\text{Ca}^{2+} + \text{Mg}^{2+}}{2}}}, \quad (6)$$

Table 2 HPI calculation for median groundwater concentration (in $\mu\text{g/l}$)

Heavy metals	M_i	S_i	I_i	MAC	Sub. index (Q_i)	Unit weightage (W_i)
As	0.15	50	10	50	24.63	0.02
Cr	0	50	10	50	25	0.02
Cu	1.48	1500	50	1000	3.346	0.001
Hg	0.17	6	1	1	16.6	1
Mn	4.95	300	100	300	47.52	0.003
Ni	0.21	70	20	20	39.58	0.05
Zn	1.54	15000	5000	5000	49.99	0.0002

$\sum W_i = 1.0945$; $\sum W_i Q_i = 19.74$; $\text{HPI} = 18.04$, M median concentration value, $n = 77$ S highest permissible value, I maximum desirable value, MAC maximum admissible concentration

Table 3 Classification of groundwater based on total hardness (TH), electrical conductivity (EC), chloride concentration, Sodium adsorption ratio (SAR), sodium percent (%Na) and residual sodium carbonate (RSC)

Classification scheme	Categories	Ranges	Percent of samples
TH (Sawyer and Mccarty 1967)	Soft	<75	2
	Moderately hard	75–150	10
	Hard	150–300	23
	Very hard	>300	65
EC (Wilcox 1955)	Excellent	<250	–
	Good	250–750	22
	Permissible	750–2250	70
	Doubtful	2250–5000	8
	Unsuitable	>5000	–
Cl ⁻ classification (Stuyfzand 1989)	Extremely fresh	<0.14	–
	Very fresh	0.14–0.85	13
	Fresh	0.85–4.23	38
	Fresh brackish	4.23–8.46	40
	Brackish	8.46–28.21	9
	Brackish-salt	28.21–282.06	–
	Salt	282.06–564.13	–
	Hypersaline	>564.13	–
SAR (Richards 1954)	Excellent	<10	100
	Good	10–18	–
	Doubtful	18–26	–
	Unsuitable	>26	–
Na% (Wilcox 1955)	Excellent	0–20	13
	Good	20–40	35
	Permissible	40–60	34
	Doubtful	60–80	17
	Unsuitable	>80	1
RSC (Richards 1954)	Good	<1.25	88
	Medium	1.25–2.5	3
	Bad	>2.5	9

where all ionic concentrations are expressed in meq/l. The SAR values range from 0.22 to 6.1. According to the Richards (1954) classification based on SAR values (Table 3), all of samples belong to the excellent category. Sodium replacing adsorbed calcium and magnesium is a hazard as it causes damage to the soil structure and becomes compact and impervious (Raju 2007; Hosseinifard and Mirzaei Aminiyan 2015). The sodium percent (%Na) is obtained by the Eq. 7

$$\text{Na\%} = \frac{(\text{Na}^+ + \text{K}^+) \times 100}{(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+)}, \quad (7)$$

where all ionic concentrations are expressed in meq/l, according to the Wilcox (1955) classification based on %Na values (Table 3), %13 of samples belong to the excellent category, %35 of samples good category, %34 permissible category, %17 doubtful category and the remaining samples belong to the unsuitable category. High

concentrations of Na are undesirable in water due to adsorption of Na onto the soil cation exchange sites, dispersion of soil aggregates, and reduction of soil permeability (Pazand and Javanshir 2014; Sathy et al. 2016; Golchin and Azhdary Moghaddam 2016). The excess sum of carbonate and bicarbonate amounts in groundwater over the sum of calcium and magnesium amounts also influences the unsuitability for irrigation (Aghazadeh and Mogaddam 2010; Hosseinifard and Mirzaei Aminiyan 2015; Sarikhani et al. 2015).

Residual sodium carbonate (RSC) has been calculated by the Eq. 8

$$\text{RSC} = (\text{HCO}_3^- + \text{CO}_3^{2-}) - (\text{Ca}^{2+} + \text{Mg}^{2+}), \quad (8)$$

where all ionic concentrations are expressed in meq/l (Eaton 1950). The classification of irrigation water according to the RSC values in waters containing more than 2.5 meq/l of RSC are not suitable for irrigation, while

those having 1.25–2.5 meq/l are doubtful and those with less than 1.25 meq/l are good for irrigation-n (Richards 1954) (Table 3). Based on this classification, %88 samples belong to the good category, %3 samples belong to the doubtful category and %9 belongs to unsuitable category.

The permeability index (PI) values are also used to determine for irrigation water quality. It is defined as Eq. 9

$$PI = \frac{Na + \sqrt{HCO_3}}{Ca + Mg + Na} \times 100, \quad (9)$$

where all the ions are expressed in meq/l (Ragunath 1987). WHO (1989) uses a criterion for assessing the suitability of water for irrigation based on permeability index. According to PI values, the groundwater of the study area can be designated as class II (25–75%) indicate that the 88% of groundwater is suitable for irrigation excepting the 12% samples, which is classified as class I (>75%).

Industrial water quality

The quality requirements for industrial water supplies range widely and almost every industrial unit has its own standards. Industries frequently suffer from the common undesirable effects of incrustation and corrosion, which the chemical reactions are caused by inferior water quality. In this study, we used Langelier saturation index (LSI), Ryznar stability index (RSI) Larson–Skold index (L-S index) and Puckorius scaling index (PSI) for determines the corrosive and scaling ability of water samples.

Langelier saturation index (Langelier 1936) is a system for estimating or predicting the amount or degree of problems with lime scale, caused in a particular water supply as it determines the corrosive or incrusting ability of a water sample.

The positive value of LSI indicates that the water is over or super saturated, depositing $CaCO_3$ on the surface of metal and corrosion rates will be negligible. A negative index indicates that the water is under-saturated dissolving $CaCO_3$ and will be considered as corrosive. The value close to zero states that the water is at saturation (equilibrium). Langelier saturation index is calculated by Eq. 10 (Kumar et al. 2009).

$$LSI = pH - pH_{(S)}. \quad (10)$$

The saturation pH can be calculated as Eq. 11 (Davit et al. 2009):

$$pH_{(S)} = (9.3 + a + b) - (c + d) \quad (11)$$

$$a = \frac{(\log_{10}[TDS] - 1)}{10} \quad (12)$$

$$b = -13.12 \log_{10}(^{\circ}C + 273) + 34.55 \quad (13)$$

$$c = \log_{10}(Ca^{2+} \text{ as } CaCO_3, \text{ mg/L}) - 0.4 \quad (14)$$

$$d = \log_{10}(\text{alkalinity as } CaCO_3, \text{ mg/L}). \quad (15)$$

Calculate the LSI value for groundwater samples in the study area indicated that 18.2% of the groundwater samples are supersaturated containing positive LSI index with a tendency to deposit $CaCO_3$ and 81.8% of the groundwater samples are under-saturated containing negative LSI index, accounts for its slight corrosive nature with a tendency to dissolve $CaCO_3$ as a result of low alkalinity and high free CO_2 content (Sivasankar and Ramachandramoorthy 2009). The LSI values are given in Table 4 and illustrated through histograms (Fig. 10).

Ryznar (1944) has designed an empirical method to determine stability index for predicting scaling tendencies of water.

This index can be calculated like LSI as follows Eq. 16 (Kannan 1991; Ravikumar and Somashekar 2012):

$$RSI = 2pH_{(S)} - pH \quad (16)$$

The result indicates that 53.2% of the samples are classified into aggressive category and 45.5% of the samples are classified into very aggressive category. Only one sample exhibits no scale forming tendency (Table 4; Fig. 11).

Larson–Skold index (L-S) is the ratio of sulfate and chloride to the alkalinity in the form of bicarbonate and carbonate (Jevaprabha et al. 2006; Ravikumar and Somashekar 2012). The L-S index can be calculated by Eq. 16 (Larson and Skold 1958).

$$L-S \text{ index} = \frac{(SO_4^{2-} + Cl^-)}{(HCO_3^- - CO_3^{2-})}. \quad (17)$$

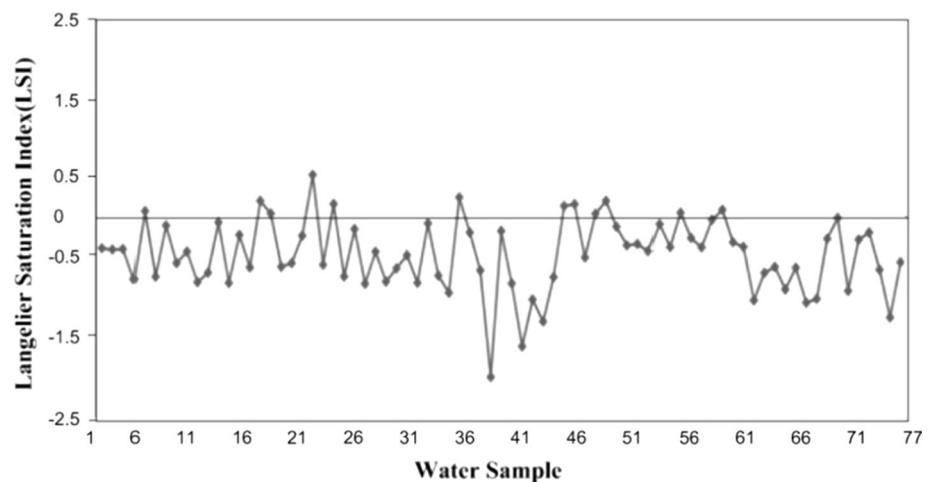
The value of L-S index below 0.8 indicates that chloride and sulfate do not interfere with natural inhibitor film formation while the value greater than 1.2 states the tendency towards high corrosion. The value of L-S index between 0.8 and 1.2 indicates that these ions may interfere with natural film formations (Rabbani et al. 2008). In the study area, chloride and sulfate interfere in 62.3% of the samples, do not interfere with natural film formation in 15.58% of the samples and may interfere with natural film formation in 22% of the samples (Table 4; Fig. 11).

The Puckorius scaling index (PSI) is used to account the buffering capacity and the maximum quantity of precipitation that can form in bringing water to equilibrium (Davit et al. 2009).

The PSI index is calculated in a manner similar to the Ryznar stability index, Puckorius has used an equilibrium pH rather than the actual system pH to account for the buffering effects. Conveniently, the PSI uses the same numbering systems and general interpretation as does the RSI (Sargaonkar and Deshpande 2003; Ravikumar and Somashekar 2012).

Table 4 LSI, RSI, L-S, and PSI index values of water samples in Ardabil plain

Parameters	Range	Indication	No. of samples	Percent
LSI (saturation capacity)	<0	Waters under-saturated with respect to CaCO ₃ and has a tendency to remove existing CaCO ₃ protective coatings in pipelines and equipment	63	81.8
	0	Water is saturated (in equilibrium) with CaCO ₃ . A scale layer of CaCO ₃ is neither precipitated nor dissolved	–	–
	>0	Water is supersaturated with respect to CaCO ₃ and scale forming may occur	14	18.2
RSI (scaling capacity)	<5.5	Heavy	–	–
	5.5–6.2	Scale	–	–
	6.2–6.8	No scale	1	1.3
	6.8–8.5	Aggressive	41	53.2
	>8.5	Very aggressive	35	45.5
L-SI index (interference of Cl and SO ₄)	<0.8	Not interfere	12	15.58
	0.8–1.2	May interfere	17	22
	>1.2	Interfere	48	62.3
PSI (scaling capacity)	<5.5	Heavy	2	2.6
	5.5–6.2	Scale	13	16.9
	6.2–6.8	No scale	15	19.5
	6.8–8.5	Aggressive	24	31.1
	>8.5	Very aggressive	23	29.9

Fig. 10 Evaluation of LSI for the groundwater samples in the study area

$$PSI = 2pH_{(S)} - pH_{eq} \quad (18)$$

The result indicated that 2.6% of the samples are classified into heavy category, 16.9% scale category, 19.5% no scale category, 16.9% aggressive category and 62.3% very aggressive category (Table 4; Fig. 11).

Conclusions

In the most groundwater samples of study area no cation–anion exceeding 50%, however the major cations are Ca²⁺, Na⁺ and the anions are HCO₃[−], Cl[−]. In the study area, the

dominant water types are Ca, Mg-HCO₃, Na-HCO₃ and Na-Cl. Results from the chemical analyses and minerals saturation index shows that chemical properties of groundwater in study area are controlled by natural geochemical processes such as mineral weathering, mixing, ion exchange and anthropogenic activities. Calculate the HPI and HEI heavy metal indices for determining the contamination of groundwater samples in the study area indicated that the groundwater of study area are not contaminated with heavy metals. Assessment of water samples according to exceeding the permissible limits prescribed by WHO standard for drinking purposes indicated that most of the groundwater in study area is chemically suitable for

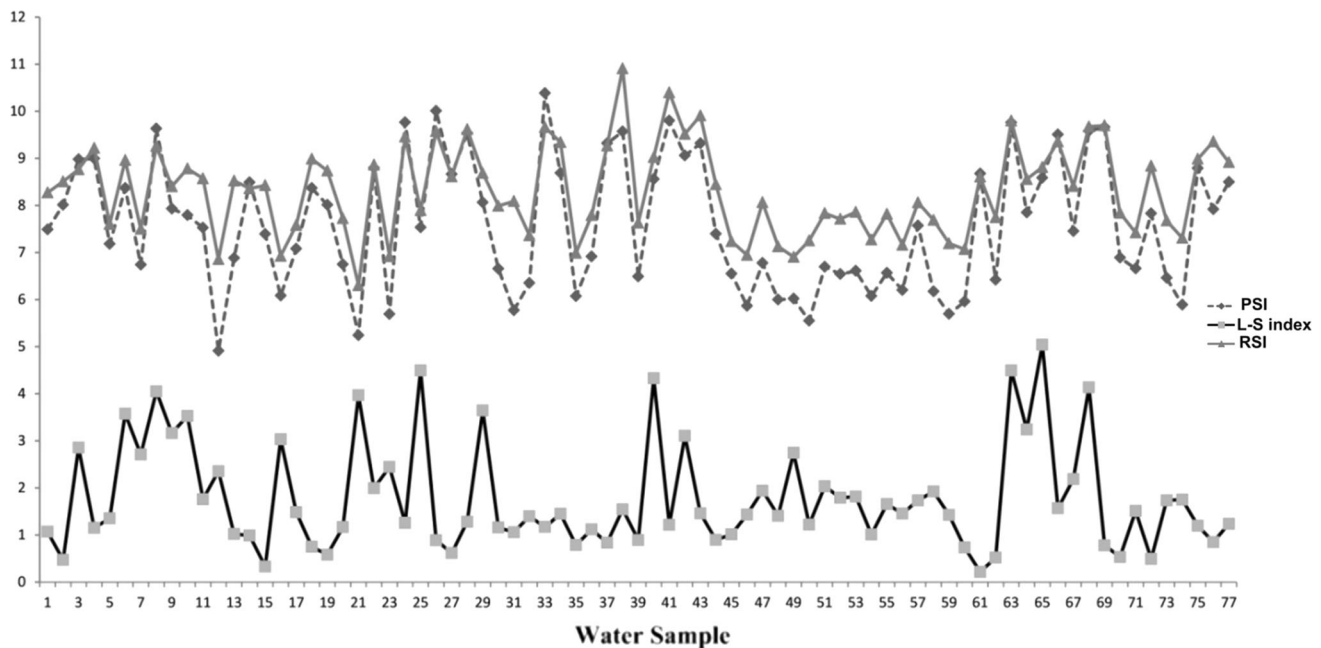


Fig. 11 Evaluation of RSI, L-S and PSI index for the groundwater samples in the study area

drinking uses. The suitability of groundwater for irrigation use was assessed from EC, SAR, RSC, Na%, and PI which varies from good to permissible, and indicated that most of the samples are suitable for this purpose. Calculating the LSI, RSI, L-S and PSI indices for determining the industrial water quality and the corrosive and scaling ability of groundwater samples in the study area indicated that most of the samples are corrosive and classified into aggressive category.

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