



## Geochemical study of groundwater conditions with special emphasis on fluoride concentration, Iran

Hamed Biglari<sup>a</sup>, Afsaneh Chavoshani<sup>b</sup>, Nasibeh Javan<sup>c</sup>, Amir Hossein Mahvi<sup>d,e,\*</sup>

<sup>a</sup>School of Public Health, Gonabad University of Medical Science, Gonabad, Iran, Tel. +98 9017313467; Fax: +98 5157223814; email: [Biglari.h@gmu.ac.ir](mailto:Biglari.h@gmu.ac.ir)

<sup>b</sup>School of Public Health, Isfahan University of Medical Science, Isfahan, Iran, Tel. +98 5157225027; email: [chavoshani.afsane@yahoo.com](mailto:chavoshani.afsane@yahoo.com)

<sup>c</sup>School of Public Health, Zahedan University of Medical Science, Zahedan, Iran, Tel. +98 9037241514; email: [Nasibeh.Javan@gmail.com](mailto:Nasibeh.Javan@gmail.com)

<sup>d</sup>School of Public Health, Tehran University of Medical Sciences, Tehran, Iran, Tel. +98 9123211827; email: [ahmahvi@yahoo.com](mailto:ahmahvi@yahoo.com)

<sup>e</sup>Center for Solid Waste Research, Institute for Environmental Research, Tehran University of Medical Sciences, Tehran, Iran

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### ABSTRACT

The quality of groundwater at any point below the surface shows the combined effects of many processes in the groundwater flow path. Factors of chemical reactions such as precipitation, weathering, dissolution, ion exchange, and various biological processes commonly happen below the surface, influencing fluoride concentrations in groundwater. Due to the fluoride effects on human health and the fact that few studies have been conducted on the factors affecting low fluoride amounts, these factors were investigated in Sistan and Baluchestan province, Iran. Water samples from 270 open dug wells, located in nine cities, were collected within 10 years (from 2004 to 2014). They were analyzed for major ions according to the standard methods. In this study, parameters such as pH, specific electrical conductivity, temperature, total dissolved solids, sulfate, nitrate, fluoride, total alkalinity, total hardness, carbonate, bicarbonate, magnesium, calcium, sodium, and potassium were analyzed. It was revealed that the soil was saline with natural pH. The fluoride amount in groundwater was deeply influenced by the composite of granitic, NaF, and CaF<sub>2</sub> rocks for a prolonged period. The wells showed many changes of fluoride concentration from 0.125 to 1.71 mg/l and fluoride decreased toward the Southeast. Na<sup>+</sup>, Ca<sup>+</sup>, Cl<sup>-</sup>, and HCO<sub>3</sub><sup>-</sup> along with F showed a positive correlation; however, NO<sub>3</sub><sup>-</sup> plots in comparison with F showed a negative correlation. Obtained results from this study indicate that the hydrological condition of groundwater is also an important factor controlling the amount of fluoride in groundwater.

*Keywords:* Geochemical; Fluoride; Groundwater; Sistan and Baluchistan; Iran

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\*Corresponding author.

## 1. Introduction

The quality of groundwater at any place below the surface shows the combined effects of many processes in the groundwater flow path. Chemical reactions such as weathering, dissolution, precipitation, ion exchange, and various biological processes commonly happen below the surface. Hydrogeochemical study is a useful method to identify the processes that are responsible for groundwater chemistry. Water shortages have become a serious problem in Iran, especially in the arid and semi-arid areas of Iran [1–6].

Groundwater has become the main source of water supply for domestic, industrial, and agricultural sectors of many countries. Iran is located in a semi-arid area with an average annual precipitation less than one-third of that of the world [1,6]. Therefore, the geochemical study of groundwater ion, namely fluoride, is necessary in this country. Because of the considerable impact of fluoride on human health, its amount in groundwater for drinking and irrigation is very important worldwide [7]. The maximum limit of fluoride in drinking water determined by the World Health Organization (WHO) (1984) is 1.5 mg/l [8]. Drinking water with fluoride concentrations above 1.5 mg/l results in dental fluorosis [9]. Skeletal fluorosis may occur when fluoride concentrations in drinking water exceed 4–8 mg/l. Nonetheless, when consumed in insufficient quantities (less than 0.5 ppm), fluoride causes health problems like dental caries, the lack of formation of dental enamel, and the deficiency of mineralization of bones, especially among children [8,10]. Therefore, it is necessary to establish a method of determining fluoride resources and their mechanism in groundwater. Previous studies have shown that fluoride mostly exists as free fluoride ions in natural water, while fluoride compounds such as Al, Be, B, and Si are found under specific conditions. These studies have found that in many bedrocks, especially complex fluorophosphates such as apatite ( $\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$ ),  $\text{CaF}_2$  is a component of magmatic rocks; and worth to say that in villiumite ( $\text{NaF}$ ), fluoride presence is remarkable. Fluoride is also discharged to the environment by anthropogenic activities such as the application of fertilizers and aluminum smelting industry, which are a group of amorphous materials such as Al-hydroxides [1,11,12]. The usual fluoride content of most soils worldwide is 329 ppm. In general, the lowest  $F$  contents are found in sandy soils in humid climates, while higher  $F$  concentrations are found in heavy clay soils and in weathered rock soils [13]. Several studies have been conducted on the geochemistry of fluorite solubility in natural water; however, all of these studies were

related to high levels of fluoride, and few studies have been carried out on factors affecting the low fluoride amount. Moreover, the fluoride content of irrigation water is essential to detect since plants and/or animals are one of the sources of fluoride for people either directly or indirectly; however, information regarding the concentration of fluoride in cultivated soils is limited or unavailable [14]. Therefore, this study, with the purpose of better understanding fluoride geochemistry in the aquatic environment was conducted. In the future, this study may suggest new and more applicable methods to solve the problems of groundwater quality such as fluoride with respect to its geochemistry.

### 1.1. Study area

Sistan and Baluchestan province (Zabol, Zahedan, Kash, Iranshahr, Saravan, Nikshar, Sarbaz, and Chabahar) and its aquifers are located in South-East Iran between the latitudes  $25^{\circ}4'–31^{\circ}25' \text{ N}$  and longitudes  $58^{\circ}55'–63^{\circ}20' \text{ E}$ , encompassing an area of about 18,175 km<sup>2</sup> (Fig. 1). The region is a semi-flat plain with a gentle slope toward the south. The area has a warm, temperate climate, and the air's highest and lowest temperatures are 50°C and –7°C, respectively, with an annual average of 25°C. The climate of the study area is semiarid, and the range of precipitation is 70–130 mm per year with the rate of evaporation being 4,000 mm per year (four times as high as Iran's average). In the study area, rocks are a kind of gypsum, clay, marl, granite, lime, and marble.

## 2. Materials and methods

Water samples were collected from 657 out of 7,560 open dug wells, located in nine cities in Sistan and Baluchestan province (Fig. 1), within 10 years (from 2004 to 2014). They were analyzed for major ions, employing the standard methods [15]: Hydrogen ion concentration (pH) and specific electrical conductivity (SEC) were measured, using pH and SEC meters. Total dissolved solids (TDS) were obtained by multiplying the SEC by a factor (0.55–0.75), based on the relative ion concentrations. Total alkalinity (TA), as  $\text{CaCO}_3$ , carbonate ( $\text{CO}_3^{2-}$ ) and bicarbonate ( $\text{HCO}_3^-$ ), was estimated through titrating with HCl. Total hardness (TH), as  $\text{CaCO}_3$  and calcium ( $\text{Ca}^{2+}$ ), was analyzed by titration, using standard EDTA. Magnesium ( $\text{Mg}^{2+}$ ) was computed, taking the difference between TH and  $\text{Ca}^{2+}$  values. Sodium ( $\text{Na}^+$ ) and potassium ( $\text{K}^+$ ) were measured by flame photometer. Chloride ( $\text{Cl}^-$ ) was estimated by standard  $\text{AgNO}_3$  titration. Sulfate ( $\text{SO}_4^{2-}$ ),

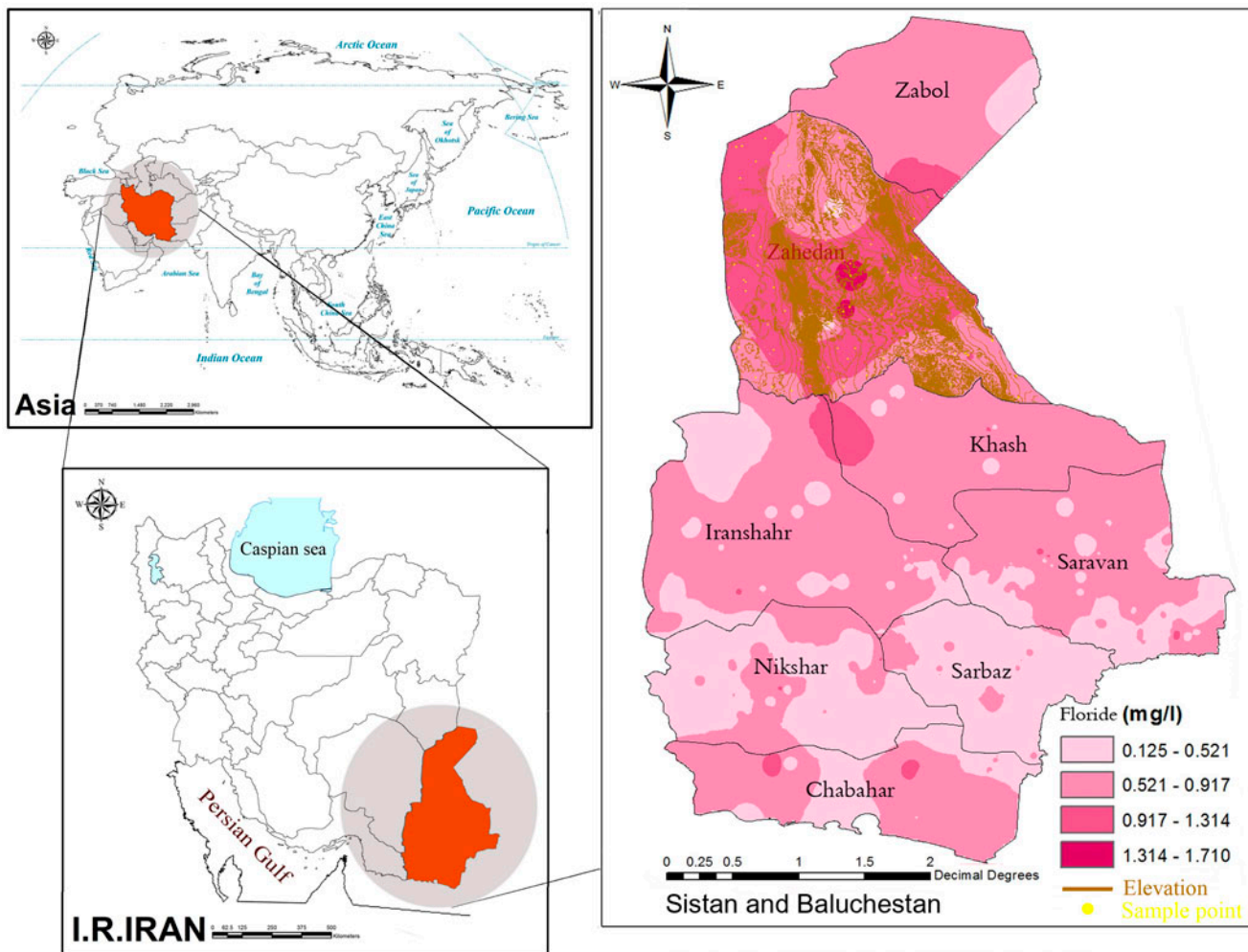


Fig. 1. Fluoride dispersal concentration from 2004 to 2014 (Drawn using GCS\_WGS\_1984 By Arc GIS v10.3 Software).

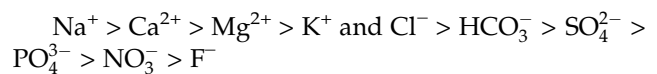
nitrate ( $\text{NO}_3^-$ ), and fluoride ( $\text{F}^-$ ) were analyzed using a spectrophotometer. All the parameters are expressed in milli-equivalent per liter (except pH). The analytical precision for the measurements of cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ , and  $\text{K}^+$ ) to anions ( $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{F}^-$ ) is indicated by the ionic balance error, which is observed to be within the stipulated limit of  $\pm 5\%$  [16].

### 3. Results and discussion

Various physicochemical parameters of water samples were analyzed, and the descriptive statistics of the analyzed parameters are given in Table 1. The EC values range from 468.153 to 9,860  $\mu\text{S}/\text{cm}$ . The average bicarbonate concentration in water samples is 549 mg/l.

Results indicate that the bicarbonate average concentration in the study area is higher than that of the

world averages, suggested by WHO (1984), and nitrate concentration is less than limits stated by WHO (50 mg/l) [17]. Moreover, hydrogeochemical concentrations of anions and cations are ordered as below:



In addition, changes in fluoride concentration in Sistan and Baluchestan province were as follows (Table 1): Zahedan > Khash > Iranshahr > Saravan > Chabahar > Nikshahr > Sarbaz.

According to Fig. 1, in Zahedan fluoride concentration ranges from 0.125 to 1.71 mg/l (0.0065–0.09 meq/l), and fluoride concentration decreases toward the southeast of the region. Subsequently, Piper triangular diagram and  $F$  plots, in comparison with other variables, were designed for Zahedan alone. The suggested fluoride concentration for fluoridated water supply system can be measured by Eq. (1) [18,19]:

Table 1  
Hydro geochemistry base concentrations from 2004 to 2014 (meq/l)

Location	T	pH	Na	K	Ca	Mg	SO <sub>4</sub>	HCO <sub>3</sub>	PO <sub>4</sub>	NO <sub>3</sub>	Cl	F	TH	TA
Zahedan	MIN	5.83	5.92	0.007	0.9	1.141	0.396	0.60	0.0	0.089	0.141	0.011	143.7	36.60
	MAX	8.28	23.45	0.333	8.75	9.7	31.25	8.87	0.033	1.081	0.141	0.082	1,567	541.4
	AVE	7.78	17.63	0.193	6.4	3.91	10.55	5.049	0.005	0.281	11.48	0.049	271.8	308.01
	SD	0.87	12.84	0.07	5.6	2.25	8.27	1.813	0.006	0.194	9.42	0.020	91.74	110.59
Iranshar	MIN	7.3	2.174	0.051	0.8	0.39	0.833	1.4	0.001	0.079	1.127	0.01	61.69	85.4
	MAX	7.16	34.32	0.769	8.92	5.115	20.83	11.36	0.019	0.790	26.873	0.091	608.54	704.9
	AVE	8.32	19.422	0.143	3.445	1.591	6.41	4.039	0.004	0.242	8.006	0.032	247.00	251.95
Chabahar	SD	1.09	9.73	0.109	1.64	0.87	4.72	1.63	0.003	0.097	5.76	0.015	99.92	71.76
	MIN	7.1	3.087	0.077	1.56	0.866	1.458	1.920	0.001	0.040	1.803	0.003	131.38	124.32
	MAX	8.3	33.78	0.333	11	22.462	23.958	7	0.007	1.161	34.085	0.077	2,085.5	427
	AVE	7.87	15	0.211	4.892	3.835	8.421	4.136	0.003	0.195	8.362	0.027	436.95	253.46
Saravan	SD	1.18	8.41	0.065	4.13	4.32	6.59	1.24	0.002	0.27	8.2	0.016	419.34	74.39
	MIN	7.14	2.609	0.077	1.76	1.180	2.083	2.720	0.001	0.129	1.746	0.01	188.72	175.52
	MAX	8.22	24.261	0.154	4.60	4.367	14.375	4.2	0.011	0.315	15.352	0.046	449.13	256.20
	AVE	7.919	13.435	0.127	2.37	2.664	3.229	3.082	0.003	0.191	2.987	0.025	237.17	288.53
Zabol	SD	1.24	5.59	0.073	3.14	1.115	4.14	1.38	0.002	0.144	3.46	0.012	73.50	84.28
	MIN	7.140	2.609	0.077	1.76	1.180	2.083	2.720	0.001	0.129	1.746	0.010	188.72	175.52
	MAX	8.22	24.261	0.154	4.6	4.367	4.375	4.200	0.011	0.315	15.352	0.046	449.13	256.20
	AVE	7.91	16.89	0.127	2.264	2.664	3.229	3.082	0.003	0.191	2.987	0.025	246.94	194.68
Khash	SD	0.92	4.98	0.022	0.68	0.88	2.87	0.380	0.002	0.042	3.12	0.10	62.62	21.92
	MIN	7.07	1.174	0.051	0.051	0.630	1.250	2.560	0.001	0.097	1.606	0.012	4,204.2	156.16
	MAX	8.3	36	0.769	13.00	7.948	15.625	11.960	0.017	0.589	25.439	0.089	119.52	729.56
	AVE	7.717	17.52	0.183	0.183	2.884	5.696	5.115	0.005	0.254	7.122	0.039	474.9	312.22
Kenarak	SD	1.26	6.60	0.118	10.02	1.74	3.71	2.11	0.004	0.135	4.64	0.015	110.62	128.97
	MIN	7.30	7.435	0.154	2.3	1.102	1.875	2.200	0.001	0.081	4.986	0.014	1,079.6	134.2
	MAX	8.150	25.73	0.333	9	9.010	16.667	4.200	0.007	0.376	18.986	0.041	171.20	256.2
	AVE	7.731	12.555	0.215	7.451	2.818	9.038	3.258	0.003	0.189	8.600	0.027	513.68	196.76
Sarbaz	SD	0.72	2.59	0.48	3.18	1.77	3.92	0.605	0.002	0.073	3.59	0.009	37.21	36.88
	MIN	7.420	3.96	0.051	0.27	0.708	0.0	2.200	0.001	0.081	1.127	0.008	64.876	134.2
	MAX	8.230	23.62	0.256	7.28	3.580	15.417	8.04	0.008	0.871	18.056	0.050	468.32	490.44
	AVE	7.814	10.25	0.117	3.181	1.335	2.158	3.832	0.001	0.251	2.782	0.021	225.96	233.76
Nikshar	SD	1.10	2.84	0.40	1.059	0.439	2.218	0.94	0.001	0.147	2.12	0.007	65.08	49.50
	MIN	7.2	3.56	0.026	1.32	0.878	0.625	2.320	0.00	0.081	1.155	0.005	105.45	2,214.7
	MAX	8.33	33.78	0.256	8.08	4.210	11.250	36.307	0.537	1.08	11.690	0.076	537.42	141.52
	AVE	7.75	14.567	0.116	3.64	1.694	3.160	4.564	0.008	0.234	4.302	0.024	266.89	278.41
SD	1.33	3.72	0.059	1.34	0.72	2.18	3.06	0.048	0.148	2.40	0.012	92.22	186.91	

$$F\left(\frac{\text{mg}}{\text{l}}\right) = \frac{0.34}{0.038} + 0.0062 T^{0f} \quad (1)$$

where  $T$  is the annual average temperature.

According to Piper triangular diagram, major groundwater types are NaCl and mixed Ca, and MgCl types (Fig. 2).

The pH of the soils was 7 on average. Moreover, according to obtained results from this study, amounts of  $\text{Na}^+$  and  $\text{Cl}^-$  are more than other cations and anions, respectively. Therefore, it can be suggested that the soils in these areas are in the neutral and saline range. The formation of NaCl and  $\text{CaCl}_2$  is the result of advanced water salinization. At higher salinity, the process of reverse cation exchange may create  $\text{CaCl}_2$  due to the removal of Na out of the solution for bound Ca. Additionally,  $\text{CaCl}_2$  could be the result of the process of mixing fresh water with saline water [20]. In

the study area, sodium chloride and calcium chloride in types of water were found due to the low velocity of groundwater, ion exchange, long-time deposits of water, and formations as well as the type of rocks [21]. With respect to high concentration of bicarbonate, it is possible that  $\text{Ca}(\text{HCO}_3)_2$  type of water can be found in this region. Water containing  $\text{Ca}(\text{HCO}_3)_2$  is found in areas of recharge (generally topographically higher areas) and NaCl water is found in discharging and static regimes (i.e. the lower lying areas). The intermediate water types are the result of hydrogeochemical processes that occur between two end members such as  $\text{Ca}(\text{HCO}_3)_2$  and NaCl [20,22]. This condition also affects some geochemical processes such as fluoride. The range of fluoride content in water samples, according to area temperature, is lower than the suggested amount by Iran's Department of Environment, EPA and WHO. In addition to the replacement of  $\text{OH}^-$  by

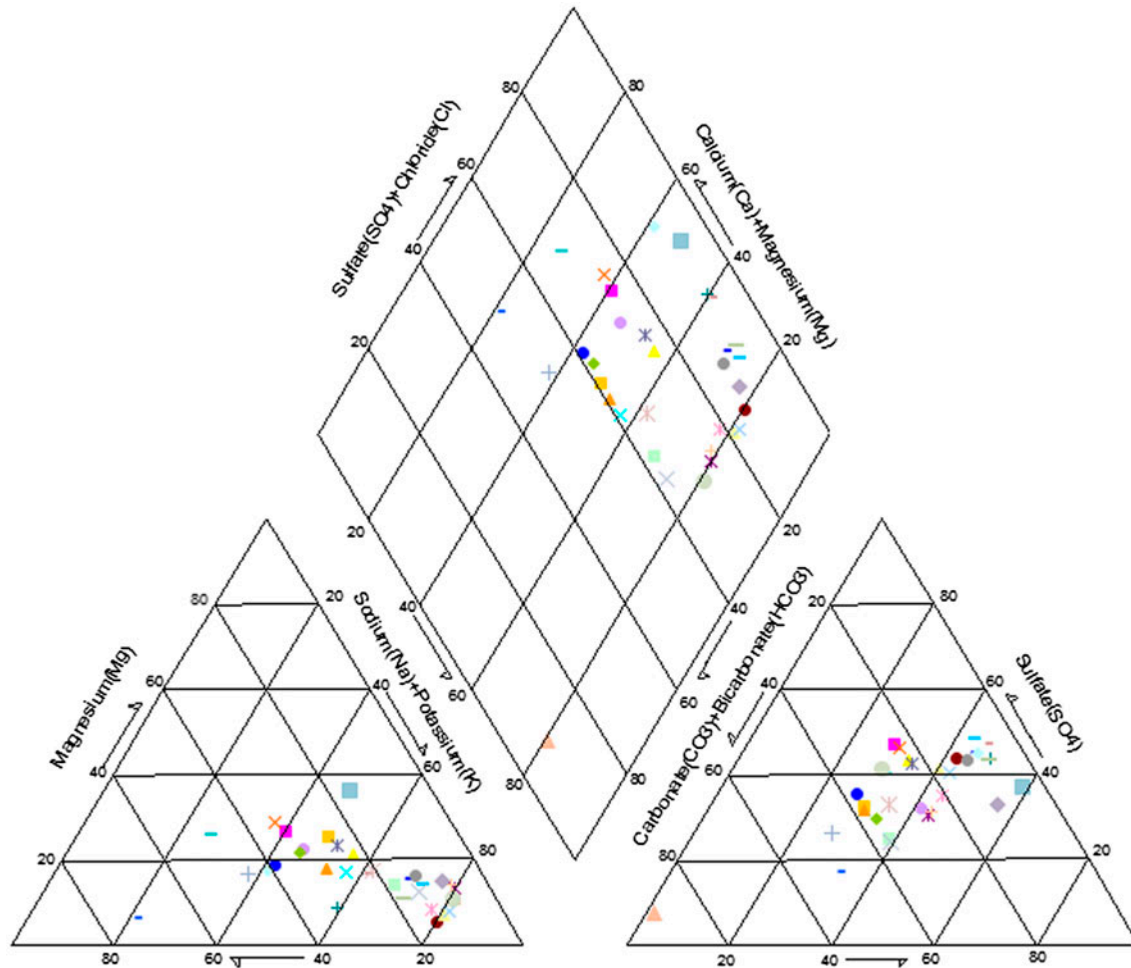


Fig. 2. Piper triangular diagram for water types in the study area.

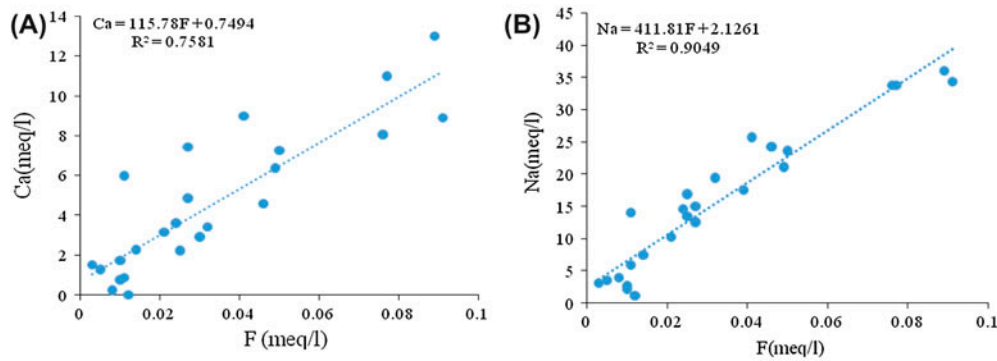


Fig. 3. Relation of Na (A) and Ca (B) with F.

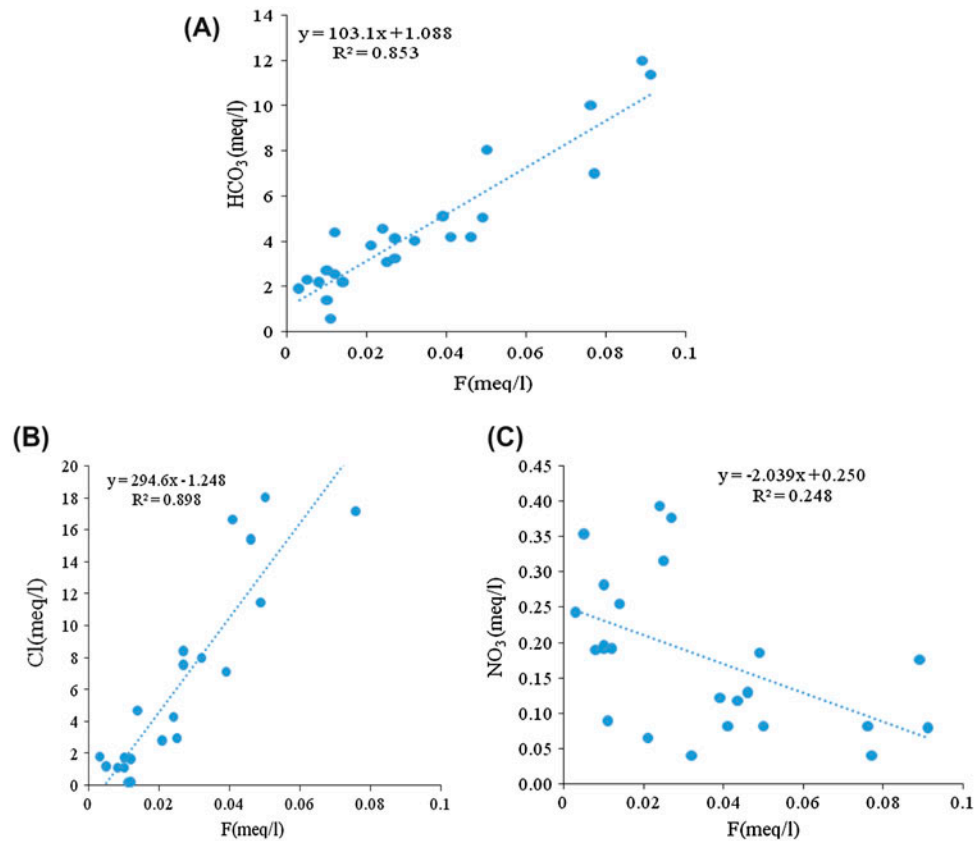


Fig. 4. Relation  $\text{HCO}_3^-$  (A),  $\text{Cl}^-$  (B), and  $\text{NO}_3^-$  (C) with F.

$\text{F}^-$  ion in mica, hornblende, and the soil consisting of clay minerals, sources of  $\text{F}^-$  in natural water are apatite and fluorite. According to low amounts of fluoride, it can be determined that rocks in Zahedan contain little fluoride. The obtained results of this study show that there is a significant relation between  $\text{Na}^+$ ,  $\text{Ca}^{+2}$ , and  $\text{F}^-$  (Fig. 3), and this can be used as an index of mineral weathering. The weathering caused by changing wet

and dry conditions of the semi-arid climate is responsible for  $\text{F}^-$  leaching from the minerals in the soils and the rocks [23]. In other words, in this region, the amount of fluoride in rock and soil is an important agent; therefore, climate changes have many effects on increasing groundwater fluoride. According to Fig. 3(A) and (B), the type of NaF rocks ( $R^2 = 9,049$ ) is more than  $\text{CaF}_2$  rocks ( $R^2 = 7,581$ ) [24,25].

In this study, there was not any significant correlation between  $F^-$ , TA, and TH (unknown data); however, there was a significant correlation between  $F^-$  with  $Cl^-$  ( $R^2 = 0.87$ ) and  $F^-$  with  $HCO_3^-$  ( $R^2 = 0.7581$ ) (Fig. 4).  $NO_3^-$  plots in comparison with  $F^-$  show a negative correlation. This suggests that controlling groundwater fluoride depends on the involvement of the ion exchange process and the involvement of other processes [25,26].

Seemingly, ion exchange process took place among these ions. It is believed that weathering granite rocks, containing fluoride, NaF, and  $CaF_2$ , are the main factors influencing fluoride content in Sistan and Baluchestan aquifers [10,25]. This indicates that the geological source of groundwater fluoride is related to the mineral compound of metamorphic rocks and granitites. Therefore, solubility and precipitation from pH changes do not have any effect on fluoride contents [25,27]. Since in some part of Iran, fluoride concentrations are high [28] and most people are used to drink black tea which has high  $F^-$  concentrations [29], it is recommended to use bottled water with low  $F^-$  or to remove fluoride by proper treatment processes [30].

#### 4. Conclusion

The results of the groundwater chemistry suggest generally low concentrations of physicochemical parameters as well as fluoride. The majority of samples fall within the natural water pH range. Most ion concentrations observed in this study were in the order of  $Na^+ > Ca^{2+} > Mg^{2+} > K^+$  and  $Cl^- > HCO_3^- > SO_4^{2-} > PO_4^{3-} > NO_3^- > F^-$  which indicated partial cationic and anionic characteristics of groundwater. The type of water in this area was saline. The obtained results demonstrate that fluoride content is influenced by weathering (the mineral compound of metamorphic rocks and granitites), ion exchanges of  $Na^+$ ,  $Ca^{2+}$ ,  $Cl^-$ , and  $HCO_3^-$  with  $F^-$  have a positive correlation. However,  $NO_3^-$  plots in comparison with  $F^-$  suggest a negative correlation. Therefore, the prevention of nitrogen fertilizers entering the groundwater is highly suggested. In the end, this study may help to suggest new and applicable methods to solve the problem of the presence of groundwater fluoride with respect to its geochemistry.

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