Geochemistry and multivariate statistical analysis for fluoride occurrence in groundwater in the Kuhbanan basin, Central Iran

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Abstract The groundwater is the only major source of drinking water in central part of Iran. The study was carried out to locate and decipher hydrogeochemical reactions responsible for elevated concentration of fluoride. In the Kuhbanan basin groundwater samples were collected at 24 sites and analyzed. The concentration of fluoride ranged from 0.05 to 10.8 mg/l in groundwater of study area. Statistical analyses were applied to a subgroup of the dataset to evaluate their usefulness to classify the groundwater samples, and to identify geochemical processes controlling groundwater geochemistry. Groundwater in the study area is mainly saline (Na–Cl type water), and statistical analyses show that weathering of the fluorapatite and silicate minerals is the common source for F^- in study area.

Keywords Groundwater · Hydrogeochemical · Fluoride · Statistical analysis · Kuhbanan

Introduction

Fluorine is the lightest element of the halogen groups, and has high electronegativity and solubility, and hence occurs as F^- in natural waters (Rango et al. 2009; Jayawardana et al. 2012). The main source of fluoride for human body is usually drinking water (Dey et al. 2012). Fluoride in drinking water has both positive and negative effects on human health. Small concentrations of fluoride (about 1 mg/l) have beneficial effects on the teeth by hardening

the enamel and reducing the incident of caries (Fung et al. 1999; Shomar et al. 2004). Intake of excess fluoride for long periods can, however, result in the incidence of dental and skeletal fluorosis as well as other adverse health effects including growth retardation, loss of mobility, changes in DNA structure, lowering of IQ of children and even death when doses reach very high levels (about 250 mg/l) (Salifu et al. 2012). According to the World Health Organization (WHO 2004), permissible limit for fluoride in drinking water is 1.5 mg/l. Considerable research has been carried out to understand various factors responsible for influencing the origin, status of fluoride in groundwater and relationship between fluoride concentration and water–rock interaction in various aquifers with different geologic settings (Handa 1975; Gaciri and Davies 1993; Saxena and Ahmed 2003; Amini et al. 2008). Potential sources of fluoride in groundwater include various minerals in rocks and soils, such as topaz ($Al_2(F, OH)SiO_4$), fluorite (CaF_2), fluorapatite ($Ca^{10}(PO_4)^6F_2$), cryolite (Na_3AlF_6), amphiboles $(Ca, Na, K)_{0-1}(Ca, Fe, Li, Mg, Mn, Na)_2(Al, Cr, Fe, Mg, Mn, Ti)_5(Al, Si, Ti)_8O_{22}(OH, F, Cl)_2$ and micas $(K, Na, Ca, Ba)(Al, Cr, Fe, Li, Mg, Mn, V, Zn)_{2-3}(Al, Be, Fe, Si)_4O_{10}(OH, F)_2$ (Chae et al. 2007). Clay minerals such as illite, chlorite and smectites also represent excellent anion exchange media that can contribute to fluoride enrichment of groundwater (Weinstein and Davison 2004). Iran is located in a semi-arid area with an average annual precipitation less than one-third of that of the world. Despite the importance of groundwater in Iran, little is known about the natural phenomena that govern the chemical composition of groundwater or anthropogenic factors that presently affect them. Increased knowledge of geochemical evolution of groundwater in these arid regions could lead to improved understanding of hydrochemical systems in such areas, leading to sustainable development of water

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resources and effective management of groundwater resource (Pazand and Fereidoni Sarvestani 2013). Even though groundwater remain the most important source for rural water supply in the Central region of Iran, little is known about the natural and any anthropogenic factors that control the groundwater chemistry, and hence the groundwater quality and fluoride contamination. The purpose of this study was to define the principal hydrogeochemical processes controlling groundwater quality and outline the relationships between the fluoride concentration in groundwater and geology and to explain the geochemical behavior of fluoride in groundwater using the hydrogeochemical study of groundwater samples.

Study area

The study areas are located in the Central Iran (Fig. 1). The climate of the study area is considered semi-arid to arid; the annual average precipitation being approximately 250 mm. Rainfall occurs from November to April, with maximum during January and February, each year. The mean monthly temperatures are high, varying between -2°C in December–January, 50°C in July. The Central Iran is a segment of

the Iran Plate, Which in turn, is part of the Cimmerian Continent collage (Sengor 1990). This microcontinent, which is a part of a magmatic island arc complex, separated from Gondwana at the beginning of the Triassic period to collide with Eurasia, more specifically with the Turan plate, in the Late Triassic. Sedimentation on the blocks during the Mesozoic was partly shallow marine, partly terrestrial in nature (Sengor 1990). The study area consists of different lithology with of Precambrian and Quaternary age. Most of the area is covered with alluvium of Quaternary age. The sedimentary rocks including shale, limestone, dolomite, marl, sandstone, conglomerate, gypsum and shale are the dominant lithology of study area that outcrops of Precambrian to Paleocene (Mahdavi and Huckriede 1996) (Fig. 1). Intrusive rocks with composition of diorite and metamorphic schist are found in the North region.

Materials and methods

A total of 24 groundwater samples were collected from existing saprings and wells in the study areas in March 2009. Water samples were filtered through $0.45\ \mu\text{m}$ Milipore filter paper (Except for analysis of alkalinity). The

Fig. 1 The sample location and geologic map of study area (after Mahdavi and Huckriede 1996)

samples were collected in three new 250 ml polyethylene bottles. Conductivity, pH, Eh and temperature were determined in the field using portable pH meter (HATCH, Germany). The cations were measured with inductively coupled plasma and mass spectrometry (ICP-MS). The anions (Cl^- , F^- , PO_4^{3-} and SO_4^{2-}) were analyzed using ion chromatography. Total dissolved solids (TDS) were measured by sample evaporation techniques and HCO_3^- was analyzed by potentiometric determination.

Results and discussion

General hydrogeochemistry

A statistical summary of physical and chemical parameters of the analyzed groundwater samples is presented in Table 1.

The concentration of fluoride in the groundwater samples ranged from 0.05 to 10.8 mg/l, with a median value of 1.06 mg/l and a standard deviation of 3.29 mg/l. A relatively high percentage (41.66 %) of the samples were found to have fluoride concentrations exceeding 1.5 mg/l, the WHO recommended maximum acceptable guideline value for drinking water (Fig. 2). The pH of the groundwater samples varied from 6.6 to 8.68 with an average value of 7.9 indicates the alkaline characteristics of the groundwater. The Eh can be used to find out the relative redox conditions of an aquifer. Eh in the study area ranged from 181.4 to 748.8 mV and therefore oxidization conditions prevail in the area (Pazand and Javanshir 2014).

The electrical conductivity of water samples found to be within a range of 291–16,160 $\mu\text{S}/\text{cm}$ with a median value of 1825.5 $\mu\text{S}/\text{cm}$. The high electrical conductivity in some of the samples could be due to prolonged and extensive agricultural practices such as irrigation coupled with the inherent geological conditions acquiring high concentrations of the dissolved material (Dey et al. 2012). The total dissolved solids (TDS) of the groundwater samples in the area ranged from 138.8 to 9100 mg/l. Fifty-six percent (50 %) of the groundwater samples were found to be fresh ($\text{TDS} < 1000$ mg/l) and 50 % were found to be brackish water ($1000 < \text{TDS} < 10,000$ mg/l). The concentration of major cations, e.g. Ca^{2+} (16.2–528 mg/l), Mg^{2+} (3.44–362 mg/l), Na^+ (11.8–2920 mg/l) and K^+ (1.18–26.8 mg/l) exhibit high variation in water samples. The major anions HCO_3^- (124–410 mg/l), SO_4^{2-} (12–2560 mg/l) and Cl^- (11.8–4885 mg/l) also have great variability in area.

Groundwater types

The groundwater samples were plotted onto Piper's diagrams for the classification of the groundwaters in the

Table 1 Statistical summary of various physical and chemical parameters for groundwater samples of Kuhbanan basin

Parameter	pH	Eh mV	EC µS/cm	TDS mg/l	HCO ₃ ⁻	PO ₄ ³⁻	SO ₄ ²⁻	F ⁻	Cl ⁻	Ca ²⁺	K ⁺	Na ⁺	Mg ²⁺	S	Fe	As µg/l	B
Min	6.6	181.4	291	138.8	124	<0.5	12	0.05	11.8	16.2	1.18	11.8	3.44	9	<0.1	<0.02	<1
Max	8.68	748.8	16160	9100	410	2	2560	10.8	4885	528	26.8	2920	362	1050	0.13	8.4	2513
Median	7.97	211.60	1825.50	939.81	173.52	0.40	172.44	1.06	264.50	88.10	4.19	289.50	46.50	119.50	0.02	0.65	226.50
	0.49	122.34	3562.57	2279.01	80.45	0.64	691.19	3.29	1042.15	133.19	5.84	631.34	73.50	262.04	0.02	2.23	595.70
Parameter	Ba µg/l	Co	Cr	Cu	Hg	Li	Mn	Mo	Ni	Pb	Sb	Se	Si	Sn	V	Zn	
Min	10.4	<0.02	<1	0.3	<0.1	5.4	0	2.1	<0.2	<0.1	<0.02	<0.02	2310	0.05	<0.2	1.8	
Max	87.53	2.2	11400	19.6	0.2	242	63.9	21.9	39.7	40.4	1.29	13.8	9404	0.24	11.3	1930	
Median	18.24	0.24	12.60	4.70	0.15	30.00	1.19	7.50	4.70	0.55	0.06	3.25	5985.00	0.09	0.90	18.00	
SD	19.43	0.47	2326.64	4.97	0.06	63.86	12.83	4.48	8.30	8.22	0.45	4.54	1794.26	0.07	3.42	390.09	

Fig. 2 Spatial distribution of F^- concentration in study area

study area basis of major cations and anions concentrations (Pazand and Javanshir 2013). It was observed that in the cation plot field the majority plotting towards the $Na^+ + K^+$ corner (Fig. 3), indicating the predominance of this cation in groundwater in the area. Similarly the samples plotted in all the zones of the anion plot field, with the majority of the samples plotted towards the HCO_3^- and Cl^- corners. Results show that groundwaters types are: Na–Cl, Ca–Mg– HCO_3 , Na– HCO_3 and Ca–Mg– SO_4 –Cl, that represent 70.83, 16.67, 8.33 and 4.17 % of the total number of water samples, respectively, reveals that most of the groundwater in the study area is classified as Na–Cl type water. Only two samples with high fluoride concentrations (>1.5 mg/l) plot outside the Na–Cl field. In Na–Cl and Na– HCO_3 types of groundwater, the fluoride exists in the form of a simple negative ion (F^-), which is most active. Although the combination of fluoride and sodium forms water-soluble salt, the content of fluoride ion increases relatively because, due to the content of sodium in groundwater and the opportunity for sedimentation of calcium fluoride (CaF_2) is greatly decreased (Yong and Hua 1991; Sujatha 2003).

Scattered plots

The different geochemical distribution diagrams of major ions are shown in Fig. 3 in order to determine the possible chemical reactions controlling changes in the chemical composition of groundwater. Due to the similar ionic radii of F^- and OH^- , which often substitute for each other within minerals, Fluoride is seldom detected in water having a pH value < 7.0 (Rafique et al. 2009). Clay minerals, such as kaolin, are able to hold F^- ions on their surfaces, but at high pH, OH^- ions can displace F^- ions, which are then released to groundwater (Sreedevi et al. 2006). It is observed that the relation between F^- and pH is positive ($r = 0.124$). Thus, higher alkalinity of groundwater activates the leaching of fluoride, therefore, affects the concentration of fluoride in the groundwater.

This is because of the similarity between the ionic radius of fluoride and hydroxyl ion thereby replacing each other at higher pH. The samples from the Kuhbanan area with the highest F^- concentrations are those with pH values above 7.5 (Fig. 3). The correlation coefficient between fluoride and Ca^{2+} and HCO_3^- ions is very poor (Fig. 3) and

Fig. 3 Piper diagram for the groundwater samples in the study area

between F^- and Na^+ and Mg^{2+} are slightly positive. It is observed that the relation between F^- and TDS is positive ($r = 0.081$). A higher concentration of TDS enhances the ionic strength, leading to an increase in the solubility of F^- ions in the groundwater (Rafique et al. 2009). The water-rock interaction in the aquifer is the governing process for F^- leaching. The correlations of F with TDS in study area indicated that groundwater F^- could be attributed mainly to the dissolution of F^- bearing minerals (Hu et al. 2013). This observation agreed well with our Factor analysis, and indicated that some F^- is associated with mineral

weathering and water-rock interactions in Kuhbanan aquifer, and water-rock interactions may not be the major reason for the occurrence of F^- in study groundwater. Groundwater F^- concentrations as a function of Ca^{2+} and Mg^{2+} are shown in Fig. 4. The results show that groundwater.

F^- concentrations decreased with increasing Ca^{2+} and Mg^{2+} concentrations. This observation suggested that the solubility of fluorite and calcite controls the groundwater compositions of components such as F , Ca and carbonate (Hu et al. 2013).

Fig. 4 Relationship between F^- content of groundwater and pH, TDS, Mg, Na, HCO_3 , Ca

At the same time, F^- concentrations decreased with the increase of Mg in groundwater, because of Mg^{2+} could easily precipitate with HCO_3^- and form dolomite ($CaMg(CO_3)_2$) in the presence of Ca^{2+} (Hu et al. 2013).

Arid and semiarid climate causes high evaporation of groundwater, which is an important factor in concentrating salts (including fluoride) in groundwater (Yong and Hua 1991).

Factor analysis

Factor analysis offers a powerful means of detecting similarities among the variables or samples with interpret the

structure within the variance–covariance matrix of a multivariate data collection (Reghunath et al. 2002). For a simple and easier interpretation, the rotated factor pattern after extraction using varimax rotation including six factors, which cumulatively explained 83.11 % of the data variance, is given in Table 2.

The first factor accounts for the highest variance usually represents the most important process or mix of processes controlling the hydrochemistry (Yidana et al. 2010). Factor 1 explains more than 37.47 % of total variance and contains large loadings on Mg^{2+} , Na^+ , K^+ , SO_4^{2-} , Cl^- , S, Cu, Mo, Si, Ni, Se, TDS, EC and HCO_3^- . It represents the weathering of halite, dolomite,

Table 2 Rotated factor pattern of three factors after varimax rotation

Variable	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6
EC	0.96	0.23	−0.03	0.08	0.05	−0.06
TDS	0.90	0.41	0.02	0.03	0.03	−0.08
pH	−0.01	−0.42	−0.54	0.39	0.04	0.19
Eh	−0.10	0.18	0.95	0.01	−0.03	0.08
PO ₄ ^{3−}	0.07	−0.05	−0.06	0.79	−0.21	−0.10
F [−]	0.25	−0.49	−0.12	0.44	0.00	−0.21
HCO ₃ [−]	0.49	0.35	−0.05	−0.05	−0.23	0.53
SO ₄ ^{2−}	0.87	0.47	0.03	−0.05	0.07	−0.06
Cl [−]	0.98	0.12	−0.01	0.01	0.00	−0.01
Ca ²⁺	0.90	0.37	0.02	0.07	0.01	−0.11
K ⁺	0.85	−0.11	0.07	0.18	0.03	−0.06
Na ⁺	0.97	0.19	−0.04	0.08	0.04	−0.03
Mg ²⁺	0.99	0.03	−0.05	0.04	0.03	−0.03
S	0.91	0.33	−0.04	0.13	0.10	−0.10
Fe	−0.16	−0.21	0.04	−0.09	0.07	0.69
As	0.28	0.92	0.05	0.01	−0.01	0.00
B	0.63	0.66	0.06	0.01	0.15	−0.12
Ba	−0.37	−0.08	0.02	−0.64	−0.23	0.01
Co	0.86	−0.40	−0.11	0.16	0.04	0.09
Cr	−0.11	−0.06	0.93	0.06	−0.04	−0.06
Cu	0.78	−0.29	−0.17	0.39	0.18	0.06
Hg	−0.12	−0.19	−0.15	−0.39	−0.17	−0.33
Li	0.87	0.45	−0.01	0.06	0.09	−0.09
Mn	−0.16	0.13	−0.17	0.22	−0.19	0.59
Mo	0.59	−0.39	−0.15	0.16	−0.14	0.35
Ni	0.85	−0.42	−0.13	0.13	0.05	0.10
Pb	0.17	−0.08	−0.05	−0.01	0.94	0.03
Sb	0.13	0.75	0.52	−0.24	−0.11	−0.15
Se	0.63	0.59	0.00	0.15	0.28	−0.16
Si	0.73	0.17	0.15	0.48	−0.16	0.15
Sn	−0.01	−0.18	0.00	−0.43	0.27	0.65
V	0.09	0.91	−0.01	0.05	−0.11	−0.05
Zn	0.05	−0.03	−0.05	−0.02	0.96	0.00
Percent of variance	37.47	17.59	7.62	7.21	7.07	6.14
Cumulative %	37.47	55.06	62.68	69.90	76.97	83.11

Bold values indicate strong loading

silicate, gypsum and maybe associated with weathering products of sulfide minerals from the underlying geology. This component represented major cations and anions resulting from the mineral weathering and water–rock interactions in the aquifer (Purushotham et al. 2011). Therefore, this natural process plays a significant role in the occurrence of F[−] in the study area. The igneous rocks in the study area contain silicate minerals such as amphiboles, and the weathering of these minerals is likely to be the one source of F[−] in groundwater of area.

Factor 2 explains 17.59 % of variance and has strong loadings of As, B, Sb and V with negative representation of

F[−], which may suggest different or multiple sources of As and F[−]. Factor 3 explains 7.62 % of total variance in the data set and consists of a strong loading of Eh and Cr and a negative factor loading of pH suggest utilization of dissolved oxygen in the ferrous to ferric conversion accompanied by H⁺ release into the system. A significant positive weight of F[−] with PO₄^{3−} and pH for the factor 4, suggests that probably these elements come from weathering fluoroapatite. The predominant processes represented by Factor 4, reflection of the similar source of Pb and Zn. Factor 6 represents the weathering carbonate minerals and contributes to 6.14 % of the total variance and is strongly associated with HCO₃[−], Fe and Mn. The negative

representation of HCO_3^- and pH could show that bicarbonate come from weathering and release of hydroxyl group of minerals. Fluorine in factor 1 shows significant positive loading.

When groundwater F^- is caused by water–rock interactions in the aquifer, F^- might be correlated with some major geochemical parameters. The results indicate that F^- occurrence in Inner Mongolia might be attributed to multiple complicated processes including agriculture, mining, and water–rock interactions in the aquifer (Kim et al. 2011).

Conclusion

The hydrogeochemistry of groundwaters in Kuhbanan basin has been studied in order to evaluation fluoride concentration in groundwater. From the investigation, it has been concluded that 41.66 % water samples of the study area exceed the WHO prescribed limit of 1.5 mg/l. The following major groundwater types were found in the study area : (1) Na–Cl (2) Ca–Mg– HCO_3 (3) Na– HCO_3 and (4) Ca–Mg– SO_4 –Cl. The concentration of fluoride in groundwater is not uniform in the area. This may be due to the differences in the presence and accessibility of fluorine-bearing minerals to the circulating water and also due to the weathering and leaching activities. The fluoride concentration was highest in Na–Cl type groundwater and lowest in Ca–Mg– HCO_3 type groundwater.

In addition, fluoride concentration was positively related to the Na^+ concentration, and negatively related to that of Ca^{2+} . This implies that the geochemical behavior of fluoride in groundwater is related to a geochemical process releasing Na^+ and removing Ca^{2+} ions. It is difficult to elucidate the exact pathways that explain changes of Ca^{2+} and F^- concentration affected by processes such as fluorite dissolution, calcite and fluorite dissolution, biotite dissolution, calcite precipitation and/or cation exchange of Ca^{2+} with Na^+ . Nevertheless, it can be concluded that Ca^{2+} concentration may plays a crucial role in fluoride behavior in deep thermal groundwater. Statistical analysis suggests that the weathering of the fluorapatite and silicate minerals is the common source for F^- in study area. Presence of hornblende diorite in study area can be attributed to the presence of higher fluoride concentration in the groundwater of the district.

The results of this study suggest that high fluoride concentrations in groundwater originate from geological sources and that fluoride can be removed by precipitation when high Ca^{2+} concentrations are maintained.

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