Hydrogeochemistry of fluoride-enriched groundwater in Khaled-Abad basin, semi-arid region of Central Iran

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Abstract
Due to the decrease in rainfall in Iran and the lack of water consumption, especially in arid and semi-arid regions of the country, groundwater is of special importance as the only source of water in these areas. Groundwater samples were sampled from 28 wells stations and observe turned, to determination the fluoride value in groundwater of the Khaled-Abad basin. The variation in the quantity of fluoride in groundwater samples is adjustment from 0.3 to 8.6 mg/l and an average value is 2.8 mg/l. Generally, 75% of the samples contained fluoride concentrations more than the drinking water standard. The outcomes of this study infer that basement rocks embody epidote, biotite and apatite and the principal supply of fluoride inside the region’s groundwater sources is weathering of those minerals.

Keywords Groundwater · Fluoride · Concentration · Khaled-Abad

Introduction
Fluoride is wanted for the person’s body and may have dangerous health consequences of absorbing water (Jayawardana et al. 2012; Berger et al. 2016). The fantastic and useful consequences of low fluoride value on enamel teeth are properly known (Chae et al. 2007), but at greater concentrations of about > 1.5 mg/l (WHO 2008), it is a threat to human fitness, and may cause serious (Kim and Jeong 2005; Jayawardana et al. 2012; Barzegar et al. 2016). Fluoride may be discovered in numerous minerals, including: topaz Al2SiO4 (F,OH)2, fluorite (CaF2), apatite (Ca5 (PO4)3F) and cryolite (Na3AlF6) (Msonda et al. 2007; Raju 2016). Fluorite dissolves easily in water and can be a major source of groundwater fluoride in granite environments (Venkatayogi and Adimalla 2017). Various instances of excessive awareness of fluoride have been stated in groundwater in several sections of the world (Haimanot et al. 2006; Farooqi et al. 2007; Wang et al. 2009; Anshumali and Naaz 2015; Kumar 2017).

A critical aspect in the evaluation of water pollutants and control is the spatial distribution of water quality parameters (Kumar 2017). The fluoride danger type has been proposed with the aid of numerous authors (Maithani et al. 1998; Brindha et al. 2016). Fluoride deficiency can be motive dental caries (<0.6 mg/l); fluoride varies from 0.5 to 1.5 mg/l is optimal for dental health and for this reason beneficial, dental fluorosis can be created while fluoride exposure ranges among 1.5 and 4 mg/l which is determined in the low-risk area, 4–10 mg/l causes dental and skeletal fluorosis.

The purpose of this research is to present a scientific technique to evaluate the distribution of fluoride in the groundwater of the Khaled-Abad basin in Central Iran. In later years, the Iranian authorities and scientists have executed a good deal of studies on the evaluation and usage of water aid inside the arid and semi-arid basins. Most of the research activities have been related to the understanding the relationship between water and the environment, evaluation of natural water resources, water and development and water resources management (Jalali, 2006, 2007; Baghvand et al. 2010). However, no such research has been carried out in Central Iran, where fluoride-enriched happens is poorly understood.
Study area

The Khaled-Abad basin is located in Central Iran (Fig. 1). It lies between North latitudes 33°26′ and 34°2′ and East longitudes 51°28′ and 52°20′. The area has a warm temperate climate, with most summertime season temperatures (July) of approximately 48 °C and minimal iciness temperatures (January) of 6 °C, and the climate is semi-arid with the annual precipitation being approximately decrease about 130 mm. The availability of groundwater sources is scarce and groundwater is a critical useful water resource for agricultural, industrial, and drinking applications in the Khaled-Abad area. Low rainfall and overuse of groundwater sources in the latest decades (2002–2020) have caused an extensive groundwater level decline (17 m) on this plain, forbidding further improvement of the basin. Overall, the water level shows a regime of groundwater flow toward the central section of the basin (Fig. 1). In the south and north of the basin, Tertiary volcanic rocks, containing andesite and rhyolite with Eocene age, related to sandstone, limestone, dolomite and conglomerate with Paleocene age, are distributed. Volcanic rocks in the southern section of the basin have been stricken by granodiorite intrusions of post-Eocene age.

Materials and methods

Overall, 28 groundwater stations from the well were sampled during the duration of June 2017 to July 2017 inside the Khaled-Abad area. Figure 1 indicates the places of stations and the distribution of samples taken. After the bottles were washed with deionized water, three bases were washed with groundwater at the sampling station before sampling to prevent possible contamination. The groundwater samples from bore wells were sampled by pumping out water for approximately 10 min to clear stagnant water through the well. Instantly after sampling, dissolved oxygen concentration,
Table 1 Hydrochemical results of groundwater sample in the study area

| Sample | pH   | Eh (mv) | EC (μS/cm) | O₂ (mg/l) | TDS (mg/l) | Cl⁻ (mg/l) | SO₄²⁻ (mg/l) | CO₃²⁻ (mg/l) | HCO₃⁻ (mg/l) | Ca²⁺ (mg/l) | K⁺ (mg/l) | Mg²⁺ (mg/l) | Na⁺ (mg/l) | F⁻ (mg/l) |
|--------|------|---------|------------|-----------|------------|------------|--------------|--------------|--------------|-------------|-----------|-------------|-----------|----------|
| 1      | 7.8  | −48.2   | 5820       | 3.8       | 3080       | 2293.8     | 114          | 38.9         | 135.7        | 360         | 6         | 221.3       | 1350      | 7.1      |
| 2      | 8.5  | −67     | 3640       | 4.7       | 1945       | 1644.8     | 62           | 24.1         | 141.1        | 268         | 5.1       | 55          | 1145      | 1.6      |
| 3      | 8.6  | −72     | 4160       | 5.3       | 2200       | 401.3      | 900          | 22.7         | 119.8        | 360.8       | 4.8        | 57.1        | 1135      | 3.3      |
| 4      | 8    | −41     | 6570       | 5.5       | 3530       | 589.8      | 970          | 31.9         | 142.9        | 458         | 6.6        | 167.6       | 1850      | 5.3      |
| 5      | 8.3  | −58     | 1909       | 6.8       | 961        | 819.2      | 950          | 21.2         | 158.3        | 193.6       | 5.8        | 60.5        | 500       | 2.9      |
| 6      | 8.5  | −68     | 1834       | 4.8       | 922        | 641.8      | 720          | 24.8         | 115.9        | 109.7       | 8.9        | 41.3        | 516       | 2.4      |
| 7      | 8.2  | −54     | 2184       | 4.7       | 1094       | 130        | 190          | 31.1         | 160.5        | 183.7       | 5.8        | 64.8        | 537       | 3.4      |
| 8      | 8.4  | −60     | 1568       | 5.7       | 748        | 210        | 390          | 31.9         | 161.2        | 120.9       | 5.5        | 34.8        | 405       | 2.4      |
| 9      | 8    | −42     | 3660       | 4         | 1892       | 641.8      | 670          | 41.8         | 170.9        | 242.5       | 13.8       | 114.9       | 961.7     | 2.6      |
| 10     | 8.1  | −47     | 2330       | 6.2       | 1184       | 986.9      | 600          | 43.9         | 135.3        | 226.7       | 4.5        | 70.1        | 567       | 3.2      |
| 11     | 8.6  | −71     | 1540       | 5.9       | 769        | 622.6      | 580          | 40.4         | 203          | 99.9        | 5.5        | 48.3        | 446       | 3        |
| 12     | 8.1  | −47     | 1579       | 5.8       | 789        | 657.9      | 530          | 28.3         | 186.8        | 153.9       | 4.1        | 81.8        | 347       | 2.1      |
| 13     | 8.2  | −51     | 1535       | 5.9       | 766        | 589.8      | 480          | 38.2         | 183.9        | 186.4       | 3.7        | 98.1        | 225       | 1.8      |
| 14     | 8.2  | −54     | 1114       | 5.3       | 550        | 220        | 840          | 48.8         | 191.4        | 130.3       | 3.7        | 73.5        | 190.3     | 3.3      |
| 15     | 8.1  | −46.5   | 956        | 6.8       | 470        | 1013.4     | 560          | 73.3         | 105.3        | 153.2       | 3.9        | 77.7        | 159.7     | 1.6      |
| 16     | 8.1  | −46.5   | 956        | 6.8       | 470        | 205        | 340          | 66.4         | 110.3        | 148.8       | 4.1        | 76.7        | 156.9     | 1.6      |
| 17     | 8.2  | −48.2   | 663        | 4         | 223        | 360        | 400          | 84.9         | 33.9         | 107.8       | 3         | 16.1        | 130.3     | 1.4      |
| 18     | 7.7  | −23.1   | 738        | 5         | 360        | 63         | 60           | 40.3         | 78.8         | 106         | 1.9        | 10          | 172.3     | 1.1      |
| 19     | 8.5  | −69.8   | 1174       | 7.3       | 581        | 60         | 130          | 79.8         | 188.9        | 83.5        | 5.3        | 27.1        | 380.3     | 1.4      |
| 20     | 7.9  | −32.6   | 908        | 5.6       | 446        | 229.4      | 550          | 75.1         | 104.6        | 0.8         | 5.7        | 17.7        | 292.7     | 2.7      |
| 21     | 8    | −38     | 2320       | 5.3       | 1202       | 225        | 300          | 38.1         | 144.1        | 188         | 6.8        | 73.3        | 650.7     | 3.9      |
| 22     | 7.9  | −36     | 4660       | 6.9       | 2440       | 69         | 70           | 36.7         | 65.3         | 349         | 5.8        | 192.2       | 969       | 6.3      |
| 23     | 8.3  | −57.1   | 351        | 5.3       | 169.1      | 217.5      | 240          | 12.7         | 79.3         | 58          | 1.3        | 6.6         | 25.3      | 0.3      |
| 24     | 8    | −39.2   | 212        | 6         | 104.4      | 50         | 80           | 13.8         | 51.7         | 42.1        | 1.1        | 4.9         | 12.1      | 0.3      |
| 25     | 7.5  | −68.2   | 403        | 5.5       | 195.3      | 120        | 260          | 32.3         | 76.2         | 63.1        | 2.3        | 8           | 40.2      | 0.4      |
| 26     | 7.3  | −62.4   | 521        | 4.3       | 252        | 110        | 270          | 22.1         | 110.3        | 67.3        | 1         | 12          | 59        | 0.7      |
| 27     | 7.3  | −55.1   | 1796       | 5.3       | 1090       | 32         | 89           | 62.5         | 128.7        | 123.6       | 2.1        | 30.4        | 688       | 2.6      |
| 28     | 7.1  | −43.4   | 11,200     | 5.5       | 6170       | 24         | 70           | 0.3          | 103.2        | 646         | 15.9       | 190.6       | 3247.2    | 8.6      |
Eh, pH, total dissolved solids (TDS) and electrical conductivity (EC) were measured in the field with a portable multi-parameter device (HATCH). The pH became calibrated using two electrode buffers at every station. The general alkalinity (HCO$_3^-$ + minor CO$_3^{2-}$) is determined by titration techniques with HCl. The standard AgNO$_3$ titration method was used to determine the amount of chloride (Cl$^-$) and spectrophotometric turbidimetry for sulfate (SO$_4^{2-}$). Within 2 weeks after sampling, cations were analyzed by inductively coupled plasma and mass spectrometry (ICP–MS) in the filtered and acidified water samples. Fluoride was analyzed by an ion chromatograph. The analytical precision of the results of ions was calculated via way of means of the ionic balances, determined as $100 \times (\text{cations} - \text{anions})/(\text{anions} + \text{cations})$, that is commonly within ± 5%.

**Results and discussion**

The consequences of the hydrogeochemical evaluation of the groundwater samples are supplied in Table 1.

**General water quality**

The analytical outcomes of the groundwater samples gathered from the area display that the groundwater is normally...
alkaline in nature (pH 7.1–8.6). The electrical conductivity (EC) of water samples was discovered to be inside various 212.6–11,200 μS/cm with an average value of 2368 μS/cm. This excessive EC value of a few samples suggests the presence of saline groundwater. TDS represents the total dissolved solids, and is a crucial parameter that may be used to observe the effect of the main components on groundwater quality. The TDS of the groundwater samples varies from 104.4 to 6170 mg/l, with an average value of 1237 mg/l. 11 samples out of 28 were discovered to be exceeding the restriction for drinking (TDS < 1000 mg/l). The small quantity of soluble solids determined can be because of the presence of crystalline rocks along with diorite and granite which generally tend to sluggish the decomposition or brief residence time of the groundwater in the Khaled-Abad area (Raju, 2016). Dominating cations were of the following order: Na⁺ > Ca²⁺ > Mg²⁺ > K⁺ and the dominating anions had been of the order Cl⁻ > SO₄²⁻ > HCO₃⁻ > SO₄²⁻ inside the study area. Na⁺ is the overcoming ion, which varies from 12 to 3247.2 mg/l with an average value of 611.6 mg/l, followed by Ca²⁺ ranges 0.8–646 mg/l with a mean of 96 mg/l, Mg²⁺ values vary between 4.9 and 221.3 mg/l with an average of 69 mg/l and K⁺ ranges 1–94.9 15.9 mg/l with an average value of 5.1 mg/l. Cl⁻ dominating ion ranges 24–2293.8 mg/l with an average value of 472.4 mg/l, followed by SO₄²⁻ ranges 60–970 mg/l with an average of 407.7 mg/l, HCO₃⁻ ranges 33.9–203 mg/l with an average of 128.1 mg/l and carbonate ranges 0.3–84.9 mg/l with an average of 39.5 mg/l in the study area (Table 1). A plotting of ionic concentrations of groundwater samples on the Piper diagram (Fig. 2) shows that groundwater in the Khaled-Abad basin was Mg–Na–HCO₃–Cl (11 sample), Ca–Mg–HCO₃ (8 sample), Na–HCO₃ (5 sample), Na–Cl (3 sample) types and one sample exhibited Ca–Mg–SO₄–Cl type.

Groundwater was divided into five groups, indicating the converting nature of groundwater parameters chemistry. This indicates that there can be a significant number of different procedures affecting groundwater chemistry in the Khaled-Abad area (Salifu et al. 2012).
Table 2  Correlation coefficient for groundwater parameters in the Khaled-Abad basin

|          | pH  | Eh (mv) | EC (μS/cm) | O₂ (mg/l) | TDS (mg/l) | Cl⁻ (mg/l) | SO₄²⁻ (mg/l) | CO₃²⁻ (mg/l) | HCO₃⁻ (mg/l) | Ca²⁺ (mg/l) | K⁺ (mg/l) | Mg²⁺ (mg/l) | Na⁺ (mg/l) | F⁻ (mg/l) |
|----------|-----|---------|------------|-----------|------------|------------|--------------|--------------|--------------|------------|-----------|------------|------------|-----------|
| pH       | 1   | 0.63    | 0.1        | 0.18      | −0.03      | 0.38       | 0.4          | −0.04        | 0.46         | −0.03      | 0.09      | −0.1       | −0.03      | −0.09  |
| Eh (mv)  |     |         |            |           |            |            |              |              |              |            |           |            |            | 0.22      |
| EC (μS/cm)| 0.1 | 0.15    |            |           |            |            |              |              |              |            |           |            |            | 0.91      |
| O₂ (mg/l)| 0.18| 0.05    | 0.15       | 0.98      | 0.32       | 0.02       | 0.23         | 0.12         | 0.33         | 0.76       | 0.39      | 0.76       | 0.97       | 0.27      |
| TDS (mg/l)| −0.03| 0.15 | 0.98      | 0.03      | 0.32       | 0.02       | 0.23         | 0.12         | 0.33         | 0.76       | 0.39      | 0.76       | 0.97       | 0.27      |
| Cl⁻ (mg/l)| 0.38| 0.03    | 0.32       | 0.02      | 0.33       | 0.02       | 0.23         | 0.12         | 0.33         | 0.76       | 0.39      | 0.76       | 0.97       | 0.27      |
| SO₄²⁻ (mg/l)| 0.4 | 0.23    | 0.12       | 0.11      | 0.08       | 0.02       | 0.23         | 0.12         | 0.33         | 0.76       | 0.39      | 0.76       | 0.97       | 0.27      |
| CO₃²⁻ (mg/l)| −0.04| 0.2    | −0.18      | 0.14      | −0.18      | 0.08       | 0.12         | 1            |              |            |           |            |            |          |
| HCO₃⁻ (mg/l)| 0.46| 0.31    | 0.33       | 0.33      | 0.36       | 0.41       | 0.12         | 1            |              |            |           |            |            |          |
| Ca²⁺ (mg/l)| −0.03| 0.22    | 0.91       | −0.09     | 0.37       | 0.14       | −0.18        | 0.24         | 1            |              |           |            |            |            |          |
| K⁺ (mg/l) | 0.09 | 0.18    | 0.78       | −0.08     | 0.3        | 0.24       | −0.07        | 0.34         | 0.6          | 1            |           |            |            |            |          |
| Mg²⁺ (mg/l)| −0.1 | 0.39    | 0.76       | 0.09      | 0.38       | 0.21       | 0.06         | 0.36         | 0.84         | 0.61        | 1            |           |            |            |            |          |
| Na⁺ (mg/l)| −0.03| 0.12    | 0.97       | −0.21     | 0.26       | 0.06       | −0.13        | 0.32         | 0.83         | 0.77        | 0.64        | 1            |           |            |            |          |
| F⁻ (mg/l) | −0.09| 0.27    | 0.84       | −0.05     | 0.19       | 0.23       | −0.02        | 0.33         | 0.73         | 0.75        | 0.73        | 0.81        | 1            |           |            |          |
Geochemistry and fluoride distribution

The amount of fluoride concentration in the collected samples was higher than the maximal tolerance level (1.5 mg/l) endorsed by the World Health Organization (WHO, 2008). The amount of fluoride varies from 0.3 to 8.6 mg/l, with an average of 2.8 mg/l (Table 1). Figure 3 shows the distribution of fluoride concentration in the samples separately. Results are presented that 21 samples out of 28 samples (75%) have a value greater than the permissible limit for drinking usage. Fluoride was deficient (<0.6 mg/l) in 11% of groundwater stations in the study area, and excessive fluoride content (4–10 mg/l) was recorded in 14% of them (Fig. 3).

Fluoride concentration is excellently positively associated with K⁺, EC, Na⁺, Ca²⁺, Mg²⁺ and moderately positively associated with Eh, Cl⁻, HCO₃⁻ and SO₄²⁻ however, negatively correlated with the pH (Table 2), represents that the fluoride value is dependent to reactions increasing Mg²⁺, Ca²⁺, Na⁺, and K⁺ and subtractive pH. Accordingly, the hydrochemical parameters of these can be used to clarify the geochemical mechanism dependent on F⁻ concentration in groundwater in this basin. Since the study area has neither industries nor many human settlements, there is no opportunity for the anthropogenic origin of F⁻ in groundwater and the excessive attention of ground F⁻ is geogenic or local hydrological conditions (Raju, 2016).

The relation between F⁻ and pH is negative (r = −0.09), indicating that the decrease alkalinity of groundwater promotes the leaching of fluoride, accordingly affecting fluoride value in groundwater. This confirms that growth mineral weathering and F⁻ leaching beneath pH conditions (Berger et al. 2016). It is observed that the relation between F⁻ and EC-TDS is significant (Table 2) and positive (r = 0.84). High TDS in groundwater can increase the ionic strength and lead to promoting fluoride solubility (Brindha et al. 2016). Table 2 shows a strong positive correlation was observed for F⁻ with Na⁺ (r = 0.81), K⁺ (r = 0.75), Mg²⁺ (r = 0.73) and Ca²⁺ (r = 0.73; as an uncommon phenomenon) that could be derived from silicate and carbonate weathering. Eventually, the hydrogeochemical process that enhances the fluoride concentration is entirely linked to a process that will increase those cations and hydrogeochemical types of groundwater are not strongly managed F⁻ concentration.

Sources of fluoride

The dominant geology of the region is volcanic, granite, conglomerates and sandstone protected by sand and alluvium. Fluoride in the groundwater of the Khaled-Abad basin is a geological source attributed to weathering of minerals and rock–water interaction. The petrographic studies of rocks in the area showed that there are a number of fluoride-rich minerals, including feldspar, epidote, apatite and biotite (Fig. 4) in the rocks of the area.
This shows that these minerals can be leached $F^-$ to the groundwater (Naseem 2010; Adimalla and Venkatayogi, 2017).

The existence of fluorine-rich minerals in basement rocks is possible to noticeably increase inside the fluoride concentration of the groundwater in the Khaled-Abad basin.

The PHREEQC programs are selected to compute the saturation index (SI) of minerals affecting the hydrochemistry of the study area. Although different factors, such as kinetic, the presence or absence of the mineral in contact with the water, will influence whether a mineral dissolves or precipitates, water has a thermodynamic tendency to precipitate phases with a calculated saturation index extra than 0, and to dissolve phases with a calculated saturation index lower than 0 (Plumlee et al. 1999; Pazand and Javanshir, 2015). The saturation index values obtained for fluorite and calcite are shown in Table 3, and determined nearly all samples are slightly sub-saturated for fluorite and super-saturated for calcite.

The results of Table 3 with the positive associate between $Ca^{2+}$–$F^-$, $Na^+$–$F^-$ and $Ca^{2+}$–$Na^+$ (Table 2), indicate that the main source for $F^-$ in groundwater in the study area emanates from the dissolution of fluorite, as groundwater with low $Ca^{2+}$ content is sub-saturated with respect to fluorite, and fluorite has an orientation to dissolve (Coertsiers et al. 2008).

**Conclusion**

The groundwater in the Khaled-Abad basin that is placed in a semi-arid place with excessive temperatures has been recognized to have a tremendous fluorine value and ranges up to 8.6 mg/l with a mean value of 2.8 mg/l. Generally, 75% of
the samples contained fluoride that transgresses the drinking water standard of 1.5 mg/l. The groundwater hydrochemistry of the area was studied with the aim of evaluating the parameters affecting the increase of fluoride concentration in groundwater resources. Fluoride concentrations were found to be chemically controlled by the Mg$^{2+}$, Na$^+$, K$^+$, Ca$^{2+}$ and pH, and those ions confirmed an excessive correlation with TDS and EC. According to the available information, it can be concluded that the geological origin of fluoride is in the Khaled-Abad area. A volcanic rock in the Khaled-Abad region comprises apatite, epidote and biotite and the weathering of those minerals is possible to be the primary source of fluoride in groundwater. The groundwater of this basin is unsuitable for drinking usage and for using must be remove the fluoride effect.

Author contributions The present authors participated in the preparation of the paper. Use must be made to remove the fluoride effect.

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Code availability Not applicable.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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