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Temporal and spatial variation of drinking water quality in a number of Divandareh villages, Iran: With emphasis on fluoride distribution

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Original Article

Abstract

Fluoride is found in all water resources at different concentrations and drinking water is the major source of fluoride exposure. Thus, because of the adverse effect of fluoride in low and high concentrations, the evaluation of its content in drinking water is necessary. In the present study, the temporal variations and spatial distribution of fluoride concentrations in the drinking water of villages in Divandareh (Kurdistan, Iran) were determined. Thus, 30 villages were selected and 180 groundwater samples were taken in 2 dry and wet seasons in the year of 2013. The concentrations of fluoride and other anions were measured using the ion chromatography (IC) method. Geospatial analysis of the data was performed using the ArcGIS geographical information system (GIS) software. The results showed that the average fluoride concentration in drinking water ranged from 0.136 to 0.736 mg/l; 90.56% of samples had a concentration less than 0.50 mg F/l, and the rest had concentrations between 0.51 and 1.0 mg F/L. Based on the results of the nonparametric Wilcoxon test, a significant difference was found between the concentrations of fluoride in the two-stage sampling ($P < 0.01$).

KEYWORDS: Drinking Water, Fluoride, Geographic Information Systems, Groundwater, Water Resources

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Introduction

Groundwater is one of the most important sources for drinking and agricultural uses, and its quality is mainly affected by both natural and human factors.[1] In other words, the chemical composition of groundwater is a complex function of several variables including geological structure and mineralogy of the watersheds and aquifers, hydro-geological conditions, the evaporation of the water table, hydro-chemical processes within the aquifer, lithology, groundwater velocity, interaction of water with soil and rock, precipitation, and human activities.[2,3] The interaction of these factors leads to different types of water. Thus, the chemical composition of water in each area is different and can affect the health aspects of drinking water quality.[4] For this reason, It is necessary to monitor the groundwater quality, especially in areas where water quality is not desirable, the values of cations and anions (such as fluoride) in

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water are above the permitted standards, and water-borne diseases have been reported.[5,6]

According to the World Health Organization (WHO) guidelines for drinking water quality, fluoride, arsenic, and nitrate are key chemicals that cause large scale health effects through drinking water exposure.[5,7] Fluoride is necessary for human life and drinking water is the main source of its intake. However, excessive intake of fluoride can cause a wide range of adverse health effects such as dental mottling and fluorosis.[8-10] The concentration of fluoride in water resources is a function of the factors mentioned above; therefore, determination of fluoride concentration in drinking water and its correlation with other drinking water parameters is important. Therefore, the aim of this study was to determine the concentration of major anions and cations, their correlation with F content of groundwater, and the temporal variations and spatial distribution of fluoride concentration in Divandareh villages, Kurdistan, Iran.

Materials and Methods

This research was a cross-sectional study performed to determine the quality of drinking water of 30 villages in Divandareh, Kurdistan, Iran. A total of 180 samples were taken in 2 dry and wet seasons and were analyzed according to standard methods.[11] Fluoride concentration and other anions were measured with ion chromatography (IC) method using a Metrohm 882 compact IC plus (Metrohm AG, Switzerland). Descriptive statistics and the Piper, Schuler, and Wilcox diagrams were used to interpret the results. In order to compare the results of the 2 phases of the study, SPSS software (version 16, SPSS Inc., Chicago, IL, USA) and Wilcoxon test were used. To determine the correlation between physical and chemical characteristics of water quality, Pearson correlation coefficient was used. RockWorks software (RockWare Inc., Golden, CO, USA) was used to analyze the results of chemical analysis in the studied samples. Using the results obtained from previous phases, groundwater type was determined and its application for drinking, agricultural, and industrial purposes was assessed. Moreover, Esri's ArcGIS, a geographical information system (GIS) (version 10, Esri, Redlands, CA, USA) was used to study the spatial variation of F.

Results and Discussion

Table 1 shows the average concentrations of major anions, cations, and other physicochemical parameters in the studied groundwater samples. The variation of fluoride concentration in the studied water supplies (in dry and wet seasons) is provided in figure 1 As can be seen in figure 1, the average fluoride concentration varied from 0.136 mg/l (sample no. 17) to 0.736 mg/l (sample no. 1). About 90.56% of samples showed a fluoride concentration below 0.5 mg/l and 9.44% of them had a fluoride concentration between 0.5 and 1 mg/l. These results are in accordance with another study in Iran which showed a low concentration of fluoride in drinking water.[12] Compared to drinking water quality standards for fluoride concentration, set by the Institute of Standards and Industrial Research of Iran (ISIRI)[13] and the WHO[5], only 9.44% of water samples have a fluoride concentration in the permissible range and 90.56% of them have lower concentration than the permissible limit (0.5 mg/l), indicating a high probability of dental caries in the study area. Therefore, supplying fluoride through other sources, such as foodstuff, tea, and toothpastes, is recommended. Indeed water fluoridation is not recommended because of its cost especially in rural areas. Cartona's study comes to the same conclusion.[14] On the other hand, the disadvantages of high concentrations of fluoride in water are much greater than that of its scarcity. According to the Iranian Fluoride Scientific Association, fluoride concentration exceeding 0.7 mg/l in drinking water may increase disadvantages rather than desirable effects of caries prevention.[10] Thus, although the lack of fluoride is a risk for consumers, it is essential to monitor its concentration in water, and tooth decay especially in children.

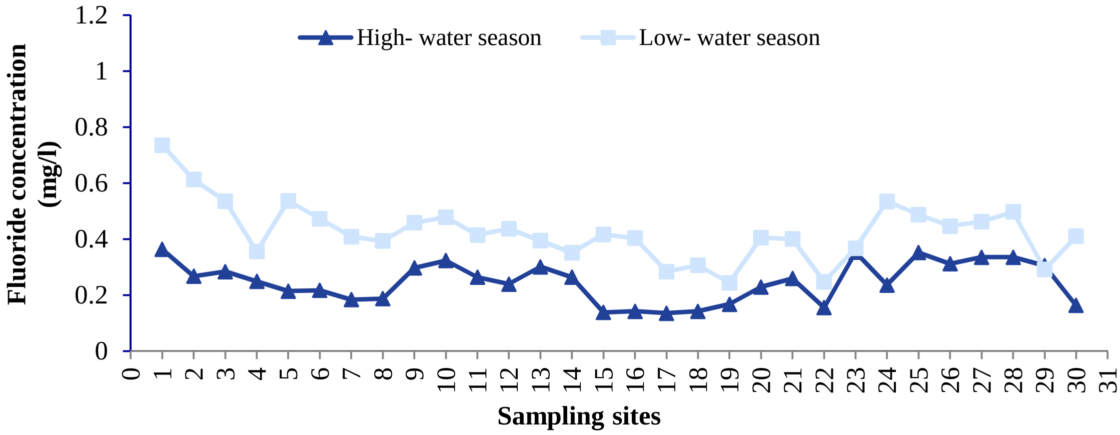

Figure 1. Fluoride concentration of different sampling sites in low- and high-water seasons

Table 1. Average values of physicochemical parameters of groundwater in rural areas

Village	Site Code	Ca	Mg	K	Na	NO_3	SO_4	F	Cl	pH	TDS*	EC†	Alkalinity	TH‡
Haydardideban	1	106.0	20.8	7.5	34.0	53.3	20.6	0.55	22.5	7.7	625	943	314	346
Kanichay	2	61.6	4.8	0.3	12.0	67.5	6.4	0.44	2.9	7.7	312	455	145	172
Kasnazan	3	76.7	14.6	0.2	16.0	42.5	8.5	0.51	13.3	7.7	428	624	215	248
Zarineh	4	94.0	23.0	0.9	48.5	1.6	26.1	0.30	25.9	7.1	626	951	330	325
Kalkan	5	121.6	15.8	18.1	41.0	56.3	74.8	0.35	35.5	7.5	707	1060	281	334
Kanisefid	6	69.2	14.3	0.2	18.5	72.1	9.2	0.36	8.6	7.5	398	586	171	226
Ghalerotaleh	7	115.5	20.3	4.8	23.0	121	30.6	0.40	36.5	7.5	648	938	231	368
Gavsheleh	8	97.8	19.2	3.5	44.5	5.2	20.7	0.29	25.5	7.6	613	928	328	318
Darehasb	9	103	21.0	4.4	71.0	9.2	41.6	0.38	66.0	7.8	726	1096	336	339
Jafarabad	10	103	20.7	1.9	38.5	6.8	16.2	0.40	19.8	7.6	625	932	344	340
Rashidabad	11	93.4	9.2	2.9	14.5	63.3	21.1	0.34	5.1	7.4	448	665	199	267
Koleh	12	87.9	18.0	3.0	34.0	5.3	33.2	0.34	16.8	7.8	537	808	282	290
Ghajar	13	73.0	13.4	0.7	24.0	25	9.1	0.35	5.0	8.0	425	618	228	234
Kapak	14	87.7	11.9	0.4	21.0	5.0	12.1	0.30	4.3	7.8	451	674	259	264
Kool	15	66.0	12.5	0.85	10.5	5.9	7.5	0.26	2.3	7.9	354	526	207	213
Dozakhdareh	16	55.0	7.5	0.4	8.5	2.5	5.2	0.27	1.3	7.9	269	402	160	165
Ghezelbelagh	17	64.5	9.5	0.6	8.5	4.3	7.1	0.20	2.0	7.8	322	478	187	198
Hezarkanian	18	80.0	10.4	3.1	17.0	4.8	16.8	0.22	14.2	7.8	413	618	219	240
Zagheolya	19	100.0	17.0	2.5	16.0	8.9	14.7	0.20	7.7	7.8	525	780	293	315
Zaghesufla	20	100.0	20.6	1.1	21.5	9.5	13.0	0.39	10.5	7.9	554	836	315	332
Nesareolya	21	78.5	17.8	3.7	45.0	22.5	28.1	0.33	23.3	7.7	534	757	254	266
Ghalerehaneh	22	74.3	21.6	0.5	9.5	50.0	18.9	0.20	12.0	7.8	427	648	198	270
Kahrizeh	23	87.0	10.6	0.1	33.5	28.2	7.5	0.36	5.6	7.7	493	720	267	254
Sarghaleh	24	47.0	12.3	0.9	43.5	23.5	11.0	0.40	4.3	8.0	387	571	205	166
Berkeh	25	65.4	12.1	0.4	42.0	27.5	9.6	0.42	6.2	7.9	448	657	239	210
Aghajari	26	47.5	13.3	1.0	50.0	21.2	5.9	0.43	4.9	7.9	443	650	254	222
Baghcheleh	27	87.7	20.3	1.5	77.5	18.9	41.5	0.41	16.8	7.8	699	1030	361	324
Dalan	28	101.5	19.0	4.5	81.5	15.3	76.2	0.44	27.2	7.8	761	1123	339	326
Ahmadabad	29	72.3	12.9	0.4	19.5	22.3	8.5	0.32	4.3	8.0	402	595	221	226
Aldareholya	30	50.5	15	0.2	11.5	18.5	13.2	0.35	3.1	7.9	300	461	159	185
National Standard of Iran (Max. Permissible) [13]		-	-	-	200	50	400	1.5	400	-	1500	-	-	500

* Total Dissolved Solids; † Electrical Conductivity; ‡ Total Hardness
All parameters are expressed in mg/l, except for pH, EC (µS/cm), Alkalinity (mg/l $CaCO_3$), and TH (mg/l $CaCO_3$)

Other physicochemical parameters such as pH, Ca^{2+}, Mg^{2+}, NO_3, total dissolved solids (TDS), and total hardness (TH) were also studied. An important parameter affecting the solubility of fluoride is pH. According to Saxena and Ahmed, in acidic pH, fluoride remains in the soil; however, in alkaline pH, it is released into the water.[15] According to the results, the pH value varies from 7.1 to 8 and represents an alkaline condition that is appropriate for the solubility of fluorine-bearing minerals. However, the low fluoride concentrations in the studied groundwater indicate that there are small quantities of fluorine-bearing minerals in the soil. On the other hand, fluoride has a unique chemical ability to replace other anions. Ca^{2+}, Na^+, OH^-, and some complexing ions may change fluoride concentration in groundwater.[16,17] Therefore, fluorite dissolution is limited when the concentration of Ca^{2+} exceeds the limit of fluorite solubility.[18] A strong reverse correlation between F and Ca^{2+} in the Ca^{2+}-containing groundwater, with concentrations higher than that required, has been observed by Raju et al. for the solubility of fluoride minerals.[18] The Ca^{2+} and Na^+ concentrations in groundwater vary from 47 to 121.6 mg/l and 8.5 to 81.5 mg/l, respectively. According to table 2, the average concentration of Ca^{2+} (82.35 mg/l) was higher than Na^+ (31.3 mg/l), which may be the reason for the low fluoride concentrations in the groundwater.

Electrical conductivity (EC) and TDS showed good correlation with fluoride concentration compared to other studied physicochemical parameters. Minimum EC of groundwater samples was around 700 µS/cm (Table 2), which placed them in the C2-S1 and C3-S1 classes; classified as slightly salty and usable brackish waters for agriculture, respectively.[19] The amount of TDS in water samples ranged between 260 and 790 mg/l. The major ions contributing to TDS are calcium, magnesium, sodium, potassium, carbonate, bicarbonate, chloride, fluoride, sulfate, and nitrate.[20] According to Gaillardet et al., variation in TDS may be related to land use and pollution.[21] Hence, animal waste, agricultural fertilizers, and industrial and municipal wastewater are the main source of nitrate, sulfate, sodium, and chloride ions in the water environment and can be related to the TDS variation.[3,22]

Based on the results, TH in the groundwater samples is carbonate hardness with a mean concentration higher than 250 mg/l $CaCO_3$. Dutta et al. have stated that water with a fluoride concentration higher than 1.5 mg/l has a hardness less than 200 mg/l.[23] Considering the amount of fluoride and hardness in this study, the results seem reasonable. A positive correlation was observed between fluoride and Ca^{2+}, Mg^{2+}, and Na^+. Similarly, a direct correlation was also observed between fluoride and bicarbonate (R^2 = 0.28) followed by chloride (R^2 = 0.16), sulfate (R^2 = 0.11), and nitrate (R^2 = 0.08). The results are in agreement with the findings of other researchers.[4,8]

Table 2. Descriptive statistics of elemental concentration for the studied parameters

Parameter	N (in each season)	Low-water season				High-water season			
		Mean	SD	Min	Max	Mean	SD	Min	Max
EC	90	782.00	217	396.00	1180.0	700.00	194.0	399.0	1100.0
TDS	90	525.00	146	261.00	784.0	468.00	130.0	275.0	737.0
pH	90	7.70	0.29	7.00	8.1	7.70	0.3	7.0	8.1
Ca	90	88.90	22.00	49.00	131.0	75.80	20.5	45.0	126.0
Mg	90	15.70	6.40	4.90	25.0	14.90	4.5	4.8	21.7
Na	90	31.80	21.20	9.00	83.0	30.60	19.7	8.0	80.0
K	90	2.60	4.30	0.20	22.8	2.10	2.8	0.1	13.4
HCO_3	90	328.00	81.00	189.00	512.0	285.00	75.2	140.0	410.0
Cl	90	15.40	16.10	1.00	75.3	13.40	12.1	1.5	56.0
SO_4	90	20.80	18.40	3.50	79.0	19.90	17.9	4.8	82.2
F	90	0.25	0.08	0.14	0.4	0.43	0.1	0.2	0.7

N: number of samples; SD: Standard deviation; EC: Electrical conductivity; TDS: Total dissolved solids

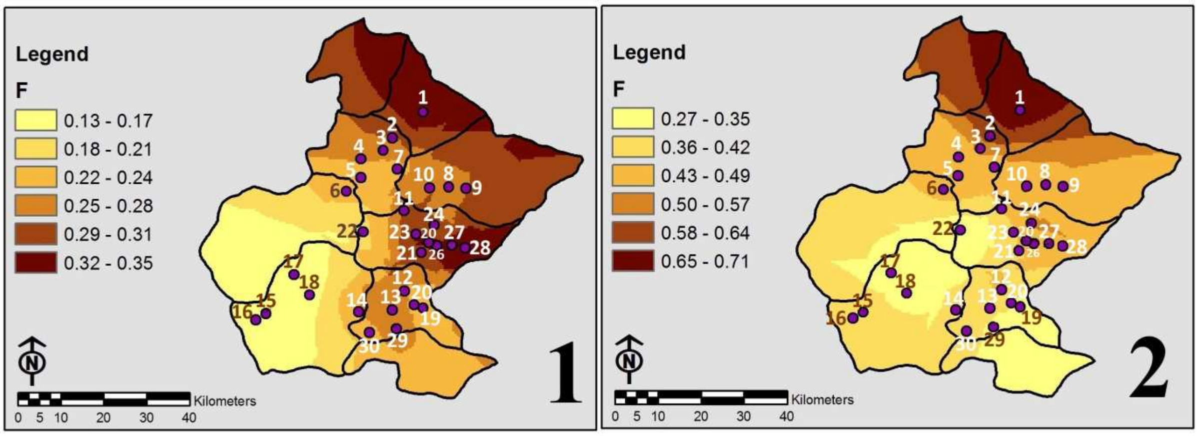

Figure 2. Spatial distribution of fluoride in groundwater in 1: low-water seasons and 2: high-water seasons

According to figure 2, the concentration of fluoride is related to the North and Northeast part of the aquifer, which is an agricultural area with a high concentration of Ca^{2+}, TDS, and TH. According to the Wilcoxon test and its correlation coefficient, there is a significant difference between the fluoride ions concentration in samples taken in the dry and wet seasons ($P < 0.01$). However, due to the low concentration of fluoride in both periods, this difference is not important.

The average concentrations of Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+} were 62.8%, 11.7%, 23.8%, and 1.8% of all cations, respectively (Table 1). Average concentration of HCO_3^-, NO_3^-, SO_4^{2-}, and Cl^- were 83%, 7.5%, 5.6%, and 3.9% of all anions, respectively (Table 1). NO_3^- concentration in around 20% of water samples was higher than the recommended standard set by ISIRI.[13] This is mostly due to agricultural activities in the area; application of different fertilizers and pesticides. Geochemical facies showed that HCO_3^- and Ca^{+} are the dominant anion and cation in the studied samples, respectively, introducing calcic-bicarbonate as the water type. Results obtained from RockWorks software showed that the rock bed in the studied villages is mainly made up of limestone, dolomite, and feldspar. Distribution of carbonate rocks in the study area and dissolution of carbonate minerals are the main source of Ca^{2+} and Mg^{2+} ions in the water. The high concentration of HCO_3^- ions in the water is due to erosion and weathering of carbonate and silicate minerals. Correlation coefficients between the different chemical parameters measured in the studied villages showed that the highest correlation exists between HCO_3^- and Ca^{2+} (Table 3). The Piper diagram also confirmed that the facies of water was calcic bicarbonate, showing that the main chemical composition of water is $Ca(HCO_3)_2$. Considering the presence of other cations and anions in the water and existence of the correlation coefficient between them, other chemical compounds present are $CaSO_4$, $CaCl_2$, $Mg(HCO_3)_2$, $MgSO_4$, $MgCl_2$, $NaHCO_3$, Na_2SO_4, NaCl, and KCl (depending on the specific terms of the ratio of Ca^{2+} to Mg^{2+} and Na^{+} to Ca^{2+} in each of the studied sources).

Conclusion

This study provides an overview of groundwater quality with emphasis on fluoride concentration in rural areas of Divandareh County. It was found that groundwater is slightly alkaline and hard in nature. Fluoride concentration in 90% of groundwater samples was less than the permissible limit set by ISIRI. It is evident from the results that the consumers in study area are at risk of dental caries. Therefore, it is essential to monitor the fluoride concentration of water and tooth decay especially in children.

Table 3.Correlation matrix of studied water quality parameters

	Ca	Mg	Na	K	F	HCO_3	Cl	SO_4	NO_3	TH	TDS	EC	pH
Ca	1												
Mg	$0.332^{\pounds}$	1											
Na	$0.307^{\pounds}$	0.480^{**}	1										
K	0.476^{**}	$0.244^{\pounds}$	$0.293^{\pounds}$	1									
F	$0.266^{\pounds}$	$0.072^{\pounds}$	0.554^{**}	$0.005^{\pounds}$	1								
HCO_3	0.685^{**}	0.644^{**}	0.747^{**}	$0.253^{\pounds}$	0.276	1							
Cl	0.593^{**}	0.595^{**}	0.637^{**}	0.542^{**}	$0.162^{\pounds}$	0.571^{**}	1						
SO_4	0.509^{**}	0.467^{**}	0.646^{**}	0.757^{**}	0.112^{3}	0.489^{**}	0.715^{**}	1					
NO_3	$0.258^{\pounds}$	-0.059^{3}	$0.184^{\pounds}$	$0.219^{\pounds}$	$0.079^{\pounds}$	$0.206^{\pounds}$	$0.141^{\pounds}$	$0.037^{\pounds}$	1				
TH	0.873^{**}	0.678^{**}	0.489^{**}	0.477^{**}	$0.304^{\pounds}$	0.846^{**}	0.699^{**}	0.571^{**}	$0.174^{\pounds}$	1			
TDS	0.794^{**}	0.654^{**}	0.782^{**}	0.499^{**}	0.450^{*}	0.904^{**}	0.787^{**}	0.730^{**}	$0.100^{\pounds}$	0.907^{**}	1		
EC	0.786^{**}	0.687^{**}	0.781^{**}	0.514^{**}	0.427^{*}	0.906^{**}	0.805^{**}	0.739^{**}	$0.059^{\pounds}$	0.911^{**}	0.996^{**}	1	
pH	-0.628^{**}	$-0.267^{\pounds}$	$-0.069^{\pounds}$	$-0.342^{\pounds}$	$-0.116^{\pounds}$	$-0.320^{\pounds}$	-0.374^{*}	$-0.232^{\pounds}$	-0.401^{*}	-0.562^{*}	-0.450^{*}	-0.454^{*}	1

* Correlation is significant at the 0.05 level; ** Correlation is significant at the 0.01 level; $^{\pounds}$ Non-significant; EC: Electrical conductivity; TDS: Total dissolved solids; TH: Total hardness

Conflict of Interests

Authors have no conflict of interests.

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