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Northwest of Iran as an endemic area in terms of fluoride contamination: a case study on the correlation of fluoride concentration with physicochemical characteristics of groundwater sources in Showt

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ABSTRACT

Fluoride in drinking water is an important chemical species with regard to public health considerations, although a fluoride concentration of about 1.5 mg L^{-1} in drinking water can decrease tooth decay and higher concentrations cause health problems. The concentration of fluoride in water is affected by some chemical characteristics. This study aimed to evaluate the relationship between fluoride and other chemical parameters (pH, Turbidity, EC, TDS, TH, ALK, Cl^-, SO_4^{2-}, NO^{3-}, NO^{2-}, Ca^{2+}, Mg^{2+}, Na^+, K^+, PO_4^{3-}) in water bodies located in Showt, West Azerbaijan Province, Iran. Sample preparation and analysis were performed according to standard methods. A wide range of fluoride concentrations was measured in this study (0.0–5.5 mg L^{-1}). The results indicated that alkalinity and Na^+ have a significant positive relationship with fluoride concentrations in water wells. Nevertheless, total hardness (TH) showed an inverse relationship with fluoride concentration. In addition, for a one-unit increase in alkalinity and Na^+ concentration, the average amount of fluoride in water increased by 0.0028 and 0.16, respectively. Furthermore, for a one-unit increase in total hardness, the average amount of fluoride in water decreased by 0.005. In conclusion, fluoride concentration was affected by numerous factors such as hardness, alkalinity, and Na^+ concentration. The findings of this study suggest that a suitable treatment of the groundwater is needed in contaminated areas before its use for drinking.

Keywords: Fluoride; Physicochemical; Drinking water; Showt; Iran

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1. Introduction

Water pollution is a controversial problem and many water resources globally have been contaminated by numerous contaminants resulted from human activities and natural processes [1–4]. Groundwater is the main source of drinking water in many areas [5–10]. Fluoride is necessary for teeth, and so many water systems add small amounts to drinking water. World Health Organization (WHO) has considered an acceptable range of 0.5–1.5 mg L^{-1} for fluoride in drinking water. No detrimental health effects are reported in this range. Teeth decay may occur at concentrations below 0.5 mg L^{-1} [11]. However, consumption of water containing high concentrations (above 1.5 mg L^{-1}) of fluoride can disturb bones and cause teeth mottling (dental fluorosis), hyperthyroidism, and electrolyte and enzymatic disorders [12–14]. For example, concentrations of fluoride above 1 mg L^{-1} may cosmetically affect and discolor teeth, but this is not a health risk [15]. Acute exposure to concentrations above the permissible value can cause a condition called skeletal fluorosis [16–18]. Studies about the effect of fluoride on human health were initiated in the past century. Many of these studies have recognized fluoride as an important factor for dental health [4,19–21]. The major intake sources of fluoride are water, food, and toothpaste. However, the most important source is drinking water [22–24]. The impact of fluoride on human health is strongly associated with its concentration in drinking water [22]. According to a report by the World Health Organization (WHO), more than 200 million people worldwide are affected by high concentrations of fluoride [25]. The concentration of fluoride in water is affected by several factors [6], the most important of which is the type of bedrock on which water flows [26]. Groundwater flowing on crystalline, volcanic, sandstone bedrocks has a higher concentration of fluoride [27–29]. Other effective factors include climatic conditions, presence of Na–HCO_3^-, Ca^{2+} concentration, groundwater storage time, distance from the aquifer, and even the activity of some microorganisms [27,28,30,31].

Naturally fluoride concentration in groundwater varies from low amounts to above 25 mg L^{-1} [32]. The presence of some cations and anions adjusts fluoride concentrations in groundwater. For instance, some studies have indicated that bicarbonate anion can increase aqueous fluoride levels. In addition, aluminosilicate in the presence of bicarbonates in sandstone aquifers leads to fluorite dissolution, subsequently increasing fluoride concentration [33,34]. On the other hand, according to a study by Chandio et al., [35] pH is an important factor in dissolving fluoride ion, and the pH range of 5 to 6.5 will cause the least amount of fluoride in water. Hence, it can be stated that the behavior of fluoride with other cations and anions varies according to location and physicochemical parameters [35,36]. In order to obtain a model for predicting fluoride concentration based on other cations and anions, their relationships should be studied. Therefore, the novelty of the present study was to provide a quantitative comparison of fluoride concentrations in drinking water and to investigate the parameters affecting fluoride concentrations in water wells located in Showt, West Azerbaijan Province, Iran. This research could be of great importance for the prevention of risk to human health and for proper groundwater resource management to provide a safe drinking water for the communities.

2. Materials and methods

2.1. Study areas description

The Showt County was selected for the purpose of the present study. Showt covers an area of approximately 931 km^2 and is located in West Azerbaijan Province, Iran (39°13′09″N 44°46′12″E/39.21917°N 44.77000°E) (Fig. 1). Its population was approximately 52519 in 2011 census. The Showt has a local steppe climate. This climate is considered to be Cold semi-arid stands (BSk) based on the Köppen-Geiger climate classification [37]. The mean annual rainfall in Showt is 264 mm. This city has a moderate climate, and it's highest, lowest, and average annual temperatures are recorded to be 26°C,–2.5°C, and 12.4°C, respectively. People used water wells for drinking, bathing, washing, and watering plants.

2.2. Sampling and analysis

22 villages were selected for the purpose of this study. The criterion for selection of the Showt's villages was the presence of fluoride above 5 mg L^{-1} in drinking water samples in some areas. Three drinking water samples were collected from each village of the selected areas for a total of three times during 2018 and stored in clean, high density polyethylene bottles at 4°C before analysis. Samples were collected from wells. In the sampling locations, the bottles were thoroughly rinsed with sample water and tightly sealed and labeled. The studied villages were coded as 1–22.

Analysis was performed in two categories of system tests and titrimetric tests. Temporary and permanent hardness, calcium, magnesium, and chloride were determined through titrimetric tests, whereas pH (measured by pH-meter device model pHwtw), electrical conductivity (EC) (by Esi meter wbw), and turbidity (by device model HACH50161/co150, HACH P210, USA, precision = 0.01) were determined using system tests. The analysis of anions and cations including nitrate, nitrite, phosphate, and sulfate was also performed by a Hach Spectrophotometer (model DR 5000) in the Water and Wastewater Laboratory of Showt. Total hardness (TH) was determined by the EDTA titrimetric method, TDS was measured gravimetrically, and the fluoride concentration of water samples was determined using the SPADNS method using a spectrophotometer [38]. Final, the mean values were reported and used in this study.

2.3. Statistical analysis

Pearson correlation coefficient and simple linear regression were employed to examine the relationship between different parameters of water and fluoride concentrations. Then, to obtain the simultaneous relation among variables, multiple linear regressions were run. The variables with bivariate association P-value <0.2 entered the model. In the final step, non-significant variables were excluded using the backward likelihood ratio method to reach the most parsimonious model. All statistical analyses were performed in SPSS (IBM Corp., Released 2016; IBM SPSS Statistics for Windows, Version 24.0; Armonk, NY: IBM Corp.).

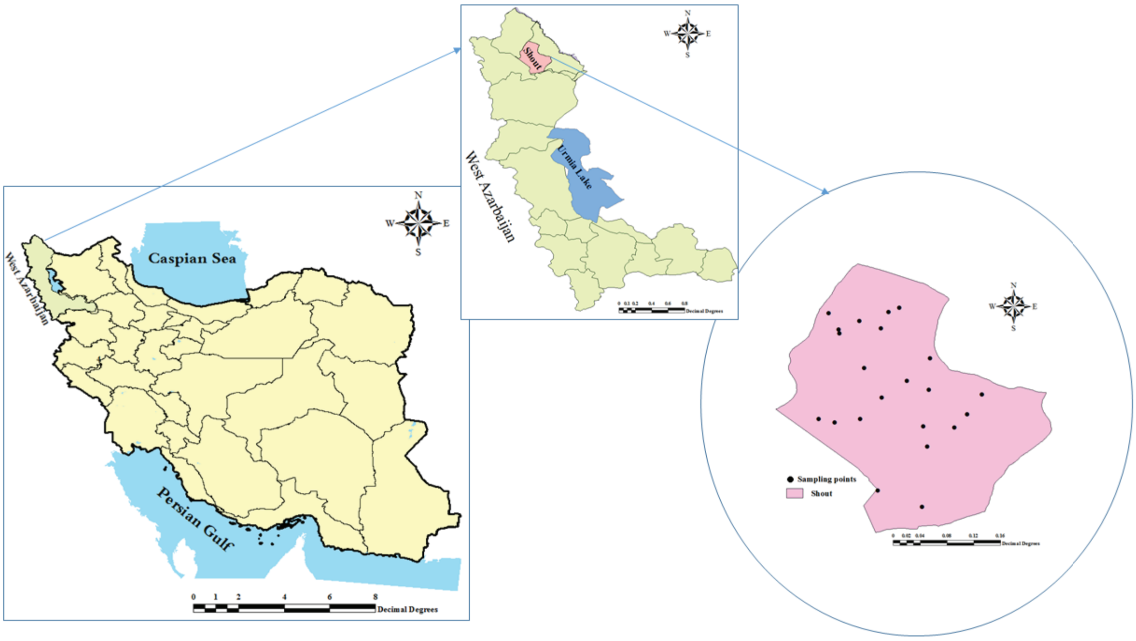

Fig. 1. Location of the study area in Showt, West Azerbaijan, Iran.

3. Results and discussion

3.1. Occurrence of fluoride and other selected parameters in groundwater samples

In this study, we analyzed the concentrations of fluoride in the groundwater in Showt's villages. Maps of fluoride concentrations for all sampling sites are shown in Fig. 2. The range of fluoride concentrations in the study area varied from 0 to 5.5 mg L^{-1}, respectively. 7 (31%) out of 22 villages had fluoride concentrations higher than the recommended value of WHO for fluoride (1.5 mg L^{-1}).

Descriptive statistics of fluoride and other characteristics of water wells from 22 villages are presented in Table 1. According to this table, a wide range of fluoride concentrations was observed.

Univariate analysis was utilized to investigate the relationship between the concentrations of fluoride and other characteristics of drinking water. These results are presented in Table 2. Variables with *P*-values <0.2 were entered into multivariate regression as the final model.

Finally, the ultimate multivariate regression model was obtained as follows (Table 3):

$$Yi = 0.0028\ ALK + (-0.005)\ TH + 0.16\ Na^{+}$$

According to this model, for a one-unit increase in alkalinity and Na^{+} concentration, the average amount of fluoride in water increases by 0.0028 and 0.16, respectively. Furthermore, for a one-unit increase in TH, the average amount of fluoride in water decreases by 0.005.

This study investigated the relationship between fluoride concentration and other physiochemical characteristics of drinking water (Table 4). The concentration of fluoride in drinking water greatly varied across the studied villages, such that it was zero in some villages, normal in some other villages (0.5–1.5 mg L^{-1}), and several times higher than the value recommended by the WHO guideline in some others (1.5 mg L^{-1}). This wide range of fluoride concentrations in groundwater sources has also been noted by other studies on the same region and other areas of West Azerbaijan Province. For example, in a study on Maku, the concentration of groundwater fluoride in various villages ranged from 0.3 to about 6 mg L^{-1} [39]. In another study, it was found that

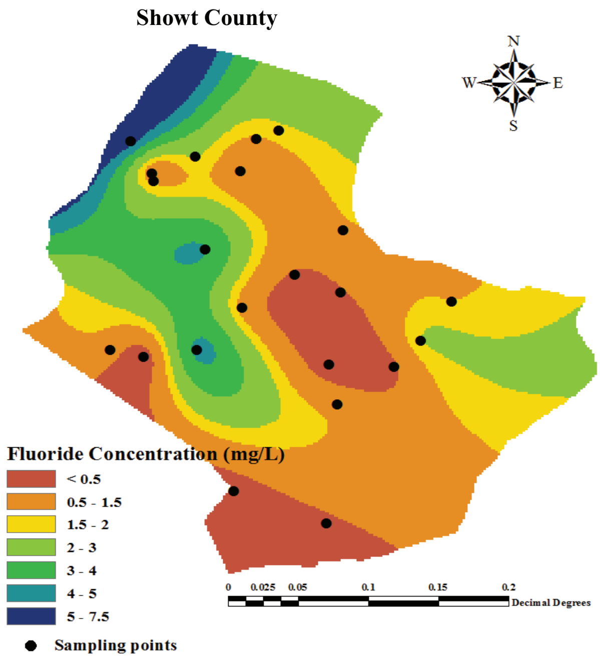

Fig. 2. Occurrence of fluoride in sampled villages of the study areas in Showt county.

Table 1
Analytical results of groundwater samples in 22 villages of Showt (F, TDS, CL^-, SO_4^{2-}, NO_3^-, Ca^{2+}, Mg^{2+}, Na^+, K^+, and PO_4^{3-} values are in mg L^{-1}, TH, ALK values are in mg L^{-1} as $CaCO_3$, EC value is in μmhos cm^{-1}, and turbidity value is NTU)

Village	F^-	pH	Turbidity	EC	TDS	TH	ALK	Cl^-	SO_4^{2-}	NO_3^-	NO_2^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	PO_4^{3-}
1	2	7.8	2	1,528	478	520	540	75	143	1	0.018	62	89	114	12.9	0.4
2	1.97	8.9	2.2	2,120	1,050	472	464	143	132	1.4	0.008	78	67	153	7	0.95
3	1.18	8.8	2.8	1,098	533	448	632	20	63	0.7	0.015	125	34	46	6.1	0.24
4	4.22	7.9	1.3	1,530	749	180	740	50	90	1.2	0.135	22	31	144	10.8	0.21
5	4	7.9	2.4	1,521	744	172	760	50	90	1.6	0.008	21	30	146	11	0.55
6	0	7.8	0.7	412	197	244	216	8	20	2.8	0.01	56	26	6	0.2	0.07
7	1	8	0.7	3,220	1,100	584	520	140	504	1.1	0.008	75	97	150	9.4	0.16
8	0.09	7.6	1	679	325	320	332	20	60	1.4	0.008	61	42	46	3.1	0.11
9	1.26	7.8	0.5	785	382	348	348	22	69	1.7	0.011	83	35	40	3.7	0.42
10	1.86	7.9	0.3	1,499	729	380	480	64	68	2.6	0.011	56	59	120	8.9	0.21
11	0.79	7.8	0.4	2,380	1,190	660	592	220	480	2.2	0.016	86	109	148	6.9	0.02
12	1.39	7.8	0.3	1,739	856	500	568	70	232	1.5	0.007	48	93	130	17.3	1.46
13	0.44	7.9	0.9	1,436	701	680	428	100	200	3.2	0.006	104	112	70	2.2	0.3
14	0	7.8	0.5	328	155	160	260	20	22	4.2	0.005	32	20	3	0	0.11
15	1.43	7.8	4.7	2,260	1120	580	148	150	448	1.8	0.005	56	107	156	6.7	0.41
16	1.52	8.2	0.6	2,170	1,100	440	420	155	376	2.3	0.006	72	63	160	5.4	0.36
17	0.38	7.8	1.3	918	444	440	220	35	100	5.4	0.007	88	54	44	1.9	0.28
18	1.01	7.8	0.5	1,316	643	520	660	55	104	1.7	0.008	96	68	103	7.3	0.45
19	5.5	7.9	0.5	1,517	742	240	648	45	76	1.5	0.009	32	107	147	11.6	0.58
20	0.37	7.9	1.2	890	429	420	368	30	69	4.3	0.006	48	73	43	2.1	0.71
21	0.45	7.8	1.3	1,021	497	360	240	40	480	5.9	0.005	64	49	85	0.9	0.36
22	1.38	7.8	1.9	2,300	1,140	600	500	145	488	2.1	0.005	48	116	156	6.7	0.7

fluoride concentration range was much wider in different sources of groundwater, varying from 0 to 10 mg L^{-1} [40]. High fluoride contamination is also reported in Poldasht, a city near Showt [41,42]. It seems that Northern regions of West Azerbaijan Province are an endemic area for fluoride contamination. The wide range of fluoride concentrations can be attributed to different minerals in the aquifer bed and also the physicochemical properties of water. On the other hand, in many parts of the West Azerbaijan Province, high concentrations of fluoride have frequently been observed. There were no industries and mining activities in this area. Showt County had geological formation including conglomerate sedimentary rocks, oligocene-miocene deposits, sand and maroon formations and several hot springs is with high Mg carbonated water observed due to volcanic activities. Weathering and leaching of fluoride-containing minerals can release fluoride into groundwater [43].

Several studies have investigated the relationship between fluoride concentrations and various characteristics of water. Some have suggested that pH range of 7–9 provides the most solubility. In addition, the relationship between Ca^{2+} and HCO_3^- ions and fluoride is reported to be direct by some studies but inverse by some others [28]. The results of the present study indicated that alkalinity and Na^+ have a significant positive correlation with fluoride concentrations in water wells. Nevertheless, TH showed an inverse association with fluoride concentration. The results of several studies confirm the relationship between alkalinity and Na^+ on the one hand and fluoride on the other, whereas some other studies have reported opposite findings [28]. In case of TH, the results of the present study were consistent with those of the study by Ramakrishnaiah et al. [44] which similarly reported an inverse relationship between TH and fluoride in water. This relationship can be attributed to the role of calcium in the precipitation of fluoride or, in other words, the negative role of calcium in the dissolution of fluoride. The negative effects of calcium and magnesium (hardness factors) on fluoride have been reported in numerous studies [27,45]. In general, studies have reported contradictory results which can be attributed to the difference among bedrocks in groundwater flows, different chemical compositions of water, and the interaction between chemical parameters. In a study conducted by Panezai and Ullah [46] on fluoride concentration in drinking water and its correlation with different physicochemical parameters in the selected areas of Quetta, Pakistan, results showed that the solubility of fluoride depends on the level of pH. Results also demonstrated that salts K^+, Na^+, Ca^{2+}, SO_4^{2-}, and Cl^- ions play a major role in attaining favorable

Table 2
Regression coefficients between fluoride and other physiochemical parameters

Parameter	*r*	β	95% CI		*P*-Value
			Lower	Upper	
pH	0.131	0.594	–1.502	2.692	0.561
Turbidity	0.121	0.163	–0.46	0.787	0.592
EC	0.266	0.0005	–0.0004	0.001	0.231
TDS	0.303	0.001	–0.0006	0.003	**0.17**[a]
TH	–0.365	–0.003	–0.007	0.0006	**0.095**[a]
ALK	0.67	0.005	0.003	0.008	**0.001**[a]
CL	0.021	0.0005	–0.011	0.012	0.925
SO_4^{2-}	–0.145	–0.001	–0.005	0.002	0.52
NO_3^-	–0.467	–0.467	–0.88	–0.054	**0.028**[a]
NO_2^-	0.447	2.344	1.562	4.532	0.037
Ca^{2+}	–0.492	0.026	–0.048	–0.004	**0.02**[a]
Mg^{2+}	0.09	0.004	–0.017	0.025	0.692
Na^+	0.597	0.016	0.006	0.025	**0.003**[a]
K^+	0.661	0.208	0.098	0.318	**0.001**[a]
PO_4^{3-}	0.234	1.014	–0.95	2.979	0.294

[a]*P*-Values <0.2

Table 3
Results of multivariate regression

Parameter	β	95% CI		*P*
		Lower	Upper	
ALK	0.0028	0.0006	0.005	0.016
TH	–0.005	–0.007	–0.0027	<0.001
Na^+	0.016	0.008	0.0236	<0.001
Intercept	0.708	–0.576	1.992	0.262

pH for the dissolution of fluoride-containing compounds in drinking water [46].

Another study reported that fluoride concentration in water has a significant positive correlation with Na^+ and SiO_2 and a significant negative correlation with Ca^{2+}, Mg^{2+}, HCO_3^-, alkalinity, and TH. Furthermore, findings suggested that groundwater with a high *F*- concentration is associated with Na–Ca–HCO_3, NaCaMg–HCO_3, and Na–Mg–Ca–HCO_3 types of water [24].

Rahman et al. [47] reported that *F* concentration has a positive correlation with electrical conductivity (EC), total dissolved solids (TDS), and the concentrations of Br, PO_4^{3-}, and SO_4^{2-}. In contrast, according to a study conducted by Dehbandi et al. [48] in central Iran, results revealed that *F* concentration in the groundwater has a negative correlation with pH and HCO_3^-.

According to the studies, physicochemical parameters in water affect fluoride concentration in water. Therefore, the geology and the presence of some anions and cations should be considered while investigating the concentrations of fluoride in water resources.

Table 4
Correlation matrix of the studied water quality parameters

Parameter	F^-	pH	TDS	Turbidity	EC	TH	ALK	NO_2^-	NO_3^-	Cl^-	SO_4^{2-}	PO_4^{3-}	Ca^{2+}	Mg^{2+}	Na^+	K^+
F^-	1															
pH	0.1311	1														
TDS	0.3034	0.2547	1													
Turbidity	0.121	0.3029	0.2321	1												
EC	0.2665	0.2211	0.9443	0.1985	1											
TH	−0.3654	0.0945	0.5765	0.1534	0.6032	1										
ALK	0.6705	0.2308	0.3386	−0.127	0.3457	−0.036	1									
NO_2^-	0.447	−0.0055	0.0278	0.0017	0.0166	−0.328	0.4054	1								
NO_3^-	−0.4669	–	−0.3781	−0.186	–	−0.103	0.6153–	−0.2395	1							
Cl–	0.0214	0.1657	0.9006	0.1672	0.8660	0.6938	0.1207	−0.0917	−0.235	1						
SO_4^{2-}	−0.145	−0.0874	0.7223	0.1834	0.7412	0.6193	−0.1000	−0.1594	0.0545	0.7692	1					
PO_4^{3-}	0.2341	0.1927	0.2721	0.0839	0.1935	0.1444	0.2065	−0.1712	−0.144	0.0683	0.0270	1				
Ca^{2+}	−0.4916	0.409	0.0455	0.0211	0.0624	0.6166	−0.0977	−0.3117	−0.023	0.1616	0.1095	−0.192	1			
Mg^{2+}	0.0896	−0.1121	0.6708	0.0899	0.6836	0.7780	0.1143	−0.2554	−0.165	0.6901	0.6068	0.3095	0.1269	1		
Na^+	0.5966	0.1545	0.901	0.2199	0.8700	0.3273	0.5111	0.1799	−0.495	0.7306	0.5720	0.3335	−0.216	0.5722	1	
K^+	0.661	0.0475	0.4819	0.0517	0.5280	0.1026	0.7139	0.2546	−0.664	0.2453	0.0907	0.5162	−0.279	0.3642	0.6987	1

4. Conclusions

This research was performed to evaluate the fluoride concentrations in water samples, which were collected from Showt's villages. The relation between fluoride and some physicochemical parameters, such as pH, Turbidity, EC, TDS, TH, ALK, Cl^-, SO_4^{2-}, NO_3^-, NO_2^-, Ca^{2+}, Mg^{2+}, Na^+, K^+, PO_4^{3-} were also determined. A wide range of fluoride concentrations (0.0–5.5 mg L^{-1}) was observed in Showt's villages. Generally, the fluoride concentrations in groundwater resources of some villages of Showt's has exceeded the maximum permissible limit (1.5 mg L^{-1}) set by WHO making it a health concern to residents who take groundwater for domestic uses. Results of the present study also indicated that alkalinity, sodium concentration, and TH can affect the concentrations of fluoride in groundwater resources. Since groundwater is the only dependable source for drinking water in this area, our research alerts that the contaminated water needs to be treated properly before supplying to the residents or alternative water resources should be used.

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Conflict of interests

The authors of this article declare that they have no conflict of interests.

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