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SPATIAL DISTRIBUTION OF FLUORIDE IN DRINKING WATER IN DHAMAR CITY, YEMEN

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Abstract

The fact that groundwater is the only source of drinking water in Yemen mandates strict monitoring of its quality. The aim of this study was to measure the levels of fluoride in the groundwater resources of Dhamar city. Dhamar city is the capital of Dhamar governorate located in the central plateau of Yemen. For this purpose, fluoride content in the groundwater from 16 wells located around Dhamar city was measured. The results showed that 75% of the investigated wells contain fluoride at or below the permissible level set by the World Health Organization (0.5 – 1.5 mg/L), whereas 25% of the wells have relatively higher fluoride concentrations (1.59 – 184 mg/L). The high levels of fluoride have been attributed to the anthropogenic activities in the residential areas near the contaminated wells. Interestingly, some wells contain very low fluoride concentrations (0.30 – 0.50 mg/L). Data were statistically treated using the principal component *analysis* (PCA) method to investigate any possible correlations between various factors. PCA shows a high correlation between well depth and its content of fluoride. On the other hand, health problems dominating in the study area necessitate further studies to investigate any correlation with imbalanced fluoride intake.

Keywords: Groundwater; Fluoride levels; Water quality; Dhamar; Yemen

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Introduction

Fluorine is a common element distributed in Earth's crust and exists in the form of fluorides in a number of minerals, such as fluorite, cryolite and fluorapatite (Kapp, 2005). Fluoride traces are present in many drinking waters with higher concentrations often associated with groundwater (Edmunds & Smedley, 2005). However, for accurate estimation of fluoride uptake, inputs from other sources shall not be overlooked. These sources are food, pharmaceutical drugs and toothpaste (KheradPisheh et al., 2016; Ozsvath, 2009).

Naturally, vegetation contains some fluoride absorbed from soil and water. In dry tea, for instance, fluoride concentration may exceed a level of 100 g/kg (Hammer, 1986; World Health Organization, 2017). In aqueous media, fluoride usually exists in the form of CaF_2, $(AlF_6)^{3-}$, MgF_2, $(A1F_6)^{3-}$, $(A1F)^{2+}$ and MgF_2. However, aqueous fluoride may, also, occur in the form HF at a pH value below 3.5.

Although fluoride is a very important dietary element, out of range doses taken through food or water give rise to consequences on health such as dental caries (Hammer, 1986). Other serious health consequences include crippling skeletal fluorosis, an increased risk of bone fractures, and discolored, brown stained or blackened, mottled or chalky white teeth (Azami-Aghdash et al., 2013; Das & Mondal, 2016; Pizzo et al., 2007), skeletal deformities (Kasim & Choudhary, 2017; Yeung, 2008), decreased red blood cells, and osteoporosis (Hillier et al., 1996), decreased thyroid function (Susheela et al., 2005), oxidative stress (Nabavi et al., 2012), nervous system impairment (Valdez-Jiménez et al., 2011), periodontal disease (Bhattacharya et al., 2016), renal disease (Xiong et al., 2007), hypertension (Sun et al., 2013), decreased fertility in animals and humans, and low birth-weight infants (Aghaei et al., 2015). As much as 60–90% of adolescents in Moldova and Ukraine living in areas where fluoride content in groundwater ranges from 2 to 7 mg/L suffer from dental fluorosis (Fordyce et al., 2007).

As groundwater is the sole source of drinking water in Dhamar City, its quality must be monitored to ensure public health safety. One of the important parameters of groundwater intended for drinking is its fluoride content. Fluoride has been classified by WHO as one of the ten chemicals of major public health concern (World Health Organization, 2017). Treatment of groundwater with high fluoride levels is complicated and expensive. In contrary to surface water, groundwater usually contains higher fluoride levels as a result of weathering (Chuah et al., 2016; Irigoyen-Camacho et al., 2016; Wasana et al., 2017).

Average fluoride concentrations of 0.85 mg/L to 2.83 mg/mg/L were reported in the southern part of the upper valley Rasyan of Taiz governorate in Yemen (Ramzy Naser et al., 2020). Another study showed that the concentration of fluoride in the area of Al-Howban Basin, Taiz governorate in Yemen, ranged from 0.89 mg/L to 3.60 mg/mg/L (Aqeel et al., 2017). According to the same study, more than 80% of population in Al-taiziah district in Taiz governorate suffers from fluorosis with a level of risk varies from mild to moderate. This district (Al-taiziah district) depends on wells dug in Al-Howban Basin as a sole source of drinking water.

A study conducted in Dhamar volcanic province showed that concentrations of fluoride vary greatly and range from 0.07 mg/L to 2.49 mg/mg/L (Minissale et al., 2013). Al-Akwa and Al-Maweri conducted a study to assess the prevalence of dental caries among 17,599 schoolchildren (9623 boys and 7976 girls aged 6-12 years) in the urban and rural districts of Sana'a governorate in Yemen (Al-Akwa & Al-Maweri, 2018). The results showed that 67.6% of the children suffer from dental caries in the areas with very low (0.00-0.40 mg/L) or very high (>2 mg/L) fluoride content in drinking water. Moreover, the same study reported a negative significant correlation between prevalence of dental caries and fluoride level in drinking water ($P < 0.05$). A study conducted in Dhamar governorate in Yemen and involved 332 schoolchildren (173 boys and 159 girls aged 13–16 years old) revealed that the prevalence of dental fluorosis was 16.99% within the boys and 30.23% within the girls (Ajrab et al., 2008).

In some Yemeni districts, fluoride concentration can be as high as 10 mg/L (Aldeghaither, 2018). In the southern part of upper Rasyan Aquifer located in Taiz governorate (Yemen) fluoride content ranged from 0.10 mg/L to > 6 mg/L (Ramzy Naser et al., 2020). 130 wells that are used as a main source of drinking and irrigation water in the volcanic aquifers in the rural highlands of Yemen were investigated for their fluoride content, and the results showed that fluoride contents exceed the maximum permissible levels (MPL) recommended by WHO (Al-Mikhlafi, 2010). Comparable levels of fluoride in drinking water were reported in the district of Sinjar in Iraq and the district of Thamar in Yemen (2.05 − 2.22 mg/L and 1.80 − 2.20 mg/L, respectively) (Ajrab et al., 2008).

In this study, we measured the concentration of fluoride ion in the groundwater wells that are used as a source of drinking water supplied to Dhamar city, Yemen. In this area, only scarce investigations on its water quality have been conducted so far. For making authoritative conclusions, 16 representative wells were carefully chosen so that a clear picture about the concentration levels of fluoride ion in the area of Dhamar city could be drawn. These 16 wells are the main part of the drinking water supply project in the city.

Study area

This study was conducted in Dhamar governorate located south and southeast of Sana'a (the capital of Yemen). The study area is close to the center of Dhamar city (the capital of Dhamar governorate) from where drinking water is supplied to the population of the city and its suburbs. The study area lies between longitude 44.433116 and 44.445743 (about 100 Km south of Sanaa), Figure 1. The topography of the study area varies from level plain to slopes at elevation between 2400 to 2700 m above sea level. The climate in the study area is arid and semi-arid. The average rainfall in the study area during the period 1999 to 2016 was 408 mm and the area receives significant recharge from runoff of surrounding mountains. The average temperature for this period ranged from 21.4C◦ in December to 27.7C◦ in June (Al-Aizari, Achaouch, Fadli, & Al-Kadasi, 2018;

Al-Aizari, Achaouch, Fadli, & Al-Mashreki, 2018). The geology of Dhamar region is predominantly volcanic with Precambrian metamorphic rocks, tertiary volcanics, Paleocene and late Miocene, cretaceous sandstone and quaternary volcanics as the main formations in the area (Sporry, 1991).

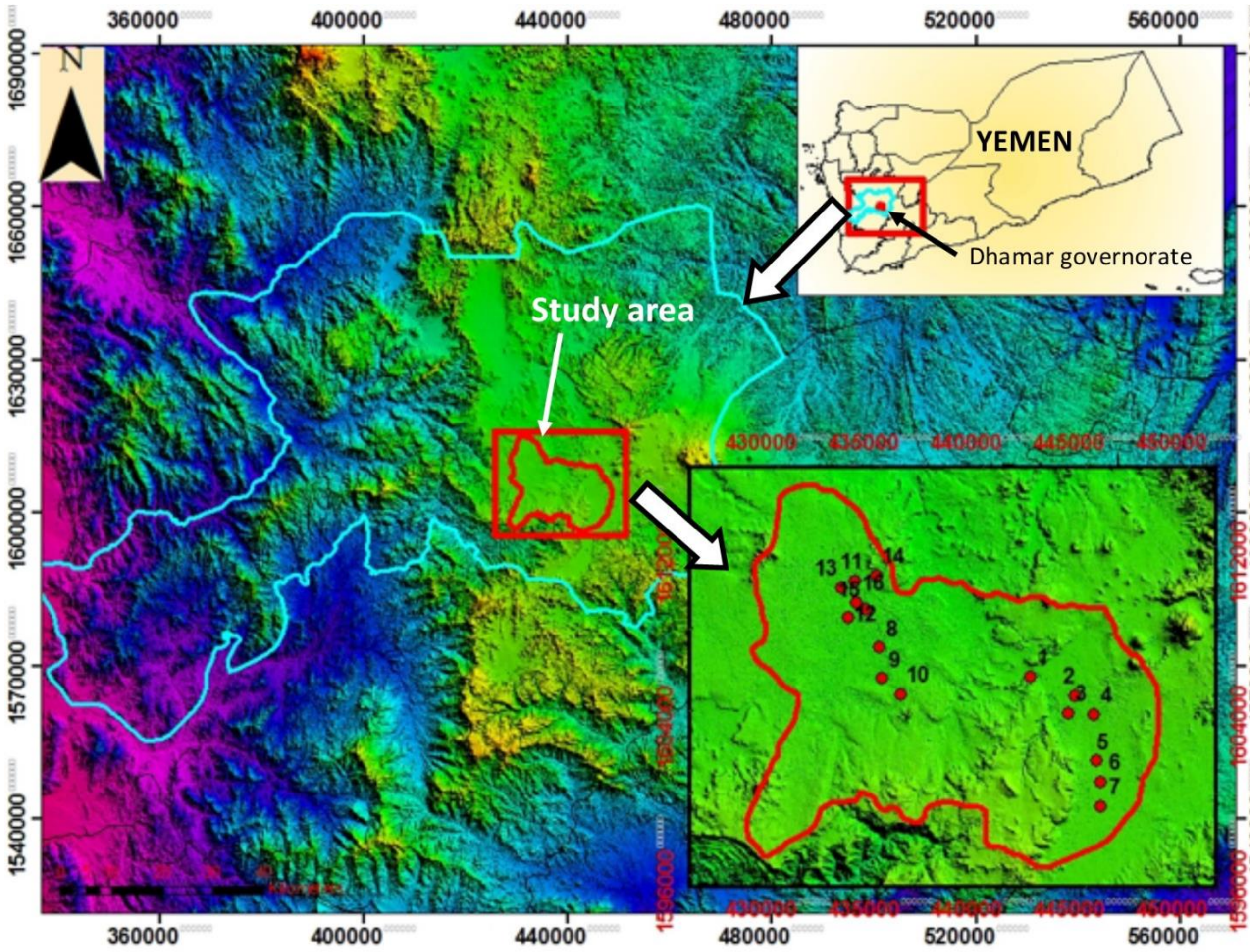

Figure 1 | A map of the study area in Dhamar city, Yemen

Materials and methods

Sixteen wells were selected for this study. Water samples were collected during the month of April 2017. Sampling points depicted in Figure 1 were selected based on topographic maps and photos aerial and readjusted in the field. Location and elevation of the selected sampling sites were determined in the field using Garmin GPS devices.

Groundwater sampling was conducted as per guidelines issued by U.S. Environmental Protection Agency (US-EPA, 2015). Clean and dry polyethylene bottles were used to collect the samples. Groundwater was allowed to pump out of the wells for about 10 min before collecting the samples. The electrical conductivity (EC) of the samples was measured in situ using a session conductivity meter (model CEL/850, HACH, USA).

All the samples were tagged and kept immediately in portable coolers at a temperature of about +4°C until analysis.

Samples were analysed following the guidelines of the American Public Health Association (Baird et al., 2017). Fluoride concentrations were measured using a colorimeter (model DR/890, HACH, USA). The ions PO_4^{3-}, SO_4^{2-}, NO_3^{-1}, F^{1-}, Ca^{2+} and Mg^{2+} were measured using standard methods (Baird et al., 2017). Statistical analysis was carried out using OriginPro v8.6. Spatial points on the location maps were drawn using ArcGIS system (version 10.3.1., 2015).

Results and discussion

A summary of the direct results of the targeted 16 wells is presented in Table 1.

Table 1 | Summary of the measurements of some ionic species in the 16 wells of drinking water in the city of Dhamar, Yemen

Variable	Min	Max	Mean	Median	WHO Guideline
Depth (m)	120	250	217.90	235	-
EC (µS/cm)	340	690	411.90	374	200
Ca (mg/L)	55	130	97.50	99	150
Mg (mg/L)	23	60	38.30	39.50	50
F (mg/L)	0.30	1.84	1.11	1.070	1.50
SO_4 (mg/L)	9	48	21.80	17.50	-
NO_3 (mg/L)	5.50	82.28	19.06	12.24	0.30

The detailed fluoride concentrations in all the studied wells are listed in Table 2. Figure 2 illustrates the spatial distribution of fluoride concentrations in the study area where different concentrations are represented with different colours.

Table 2 | Fluoride concentrations in the 16 wells of drinking water in the city of Dhamar

Sample ID	Station ID	Latitude	Longitude	F (mg/L)
w1	W- 2	14° 36' 20.4192" N	44° 26' 32.352" E	1.19
w2	W- 18	14° 36' 17.0784" N	44° 26' 40.1244" E	**1.71***
w3	W-20	14° 36' 13.932" N	44° 26' 38.9004" E	0.86
w4	W- 23	14° 36' 13.6512" N	44° 26' 43.2996" E	0.94
w5	W- 24	14° 36' 5.5692" N	44° 26' 43.926" E	1.33
w6	W- 25	14° 36' 1.9656" N	44° 26' 44.6748" E	**1.59**
w7	W-26	14° 35' 57.588" N	44° 26' 44.6748" E	**1.84**
w8	Qusoor well	14° 36' 25.4556" N	44° 26' 5.694" E	1.14
w9	Mahalah Well	14° 36' 19.9656" N	44° 26' 6.1224" E	0.30
w10	Aljeded well	14° 36' 17.2152" N	44° 26' 9.4884" E	0.38

w11	Alkhabra well	14° 36' 37.206" N	44° 26' 1.5324" E	1.30
w12	Anassr Well	14° 36' 30.6288" N	44° 26' 0.2832" E	0.98
w13	Asmaa Well	14° 36' 35.9604" N	44° 25' 59.2176" E	0.50
w14	Herran Well	14° 36' 38.0376" N	44° 26' 5.2656" E	**1.75**
w15	Almjmaa Well	14° 36' 32.2668" N	44° 26' 3.3504" E	0.90
w16	Alhamsah Well	14° 36' 33.3828" N	44° 26' 1.6476" E	1.00

**Bold values are higher than MPL*

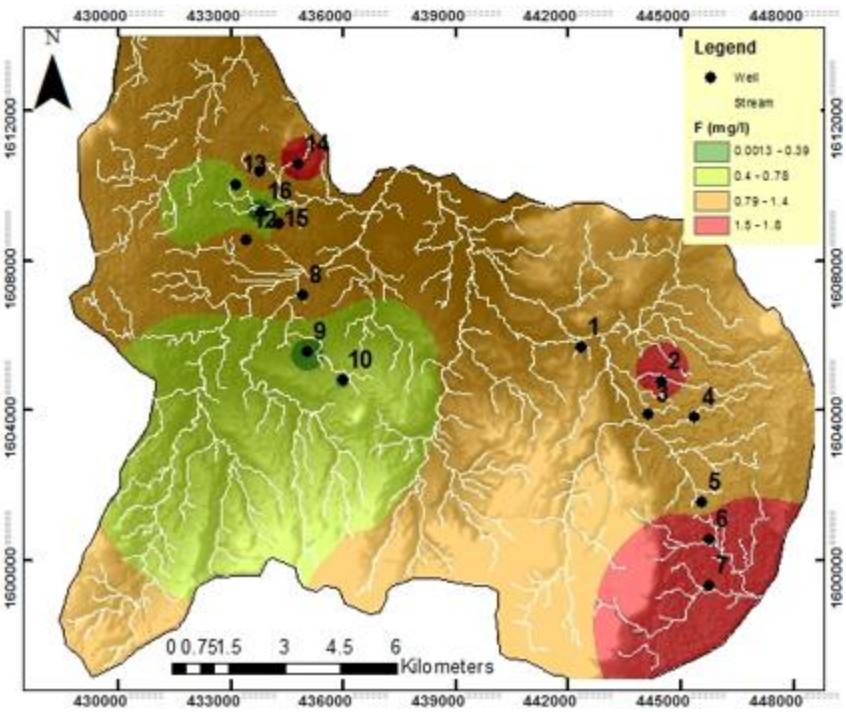

Figure 2 | Spatial distribution of Fluoride in the study area.

The results showed that fluoride concentrations lie in the range of 0.30 to 1.84 mg/L and that levels of fluoride in 4 wells (out of the 16 target wells) slightly exceed the maximum permissible limit of 1.5 mg/L recommended by the WHO (World Health Organization, 2017). Table 3 presents a comparison of fluoride's concentrations in some Yemeni regions. It is obvious that fluoride concentration in Yemen varies greatly. Although this variation could be attributed to the geological structures of these regions, only a comprehensive study can reveal the hidden causes and correlate these levels with the health of those living in these areas.

Table 3 | A comparison of fluoride's concentrations in some Yemeni regions.

Location	Fluoride range (mg/l)	Ref
Al-Howban Basin, Taiz-Yemen	0.5 to 1.5	(Aqeel et al., 2017)
The upper valley Rasyan, Taiz, Yemen	0.85 to 2.83	(R. Naser et al., 2016)
Hidhran & Alburayhi Basin, Taiz-Yemen	1.08 to 10.00	(Abdulmohsen Saleh Al-Amry, 2009)
Al-Dhala basin, Yemen	0.31 to 18.30	(Abdulmohsen S. Al-Amry et al., 2020)
Dhamar city, Yemen	0.30 to 1.84	This study

The relationship between various elements was studied using the Pearson correlation coefficient. The correlation matrix of the original data for the measured parameters is listed in Table 4.

Table 4 | Correlation matrix Table

Variables	**Depth**	**EC**	**Ca**	**Mg**	**F**	**SO_4**	**NO_3**
Depth	**1**						
EC	**0.436***	**1**					
Ca	-0.309	0.353	**1**				
Mg	-0.291	0.171	**0.728**	**1**			
F	**-0.563**	**-0.526**	0.257	**0.465**	**1**		
SO_4	**0.511**	**0.614**	0.207	0.140	-0.295	**1**	
NO_3	0.218	**0.900**	**0.498**	0.205	-0.305	**0.561**	**1**

p-value<0.05. p-value of the normalized covariance; 95% confidence intervals.
**Bold values indicate a significant correlation (positive or negative)*

The statistical treatment of the results showed a significant negative correlation between depth of the well and its fluoride content. This observation of decreasing fluoride content as the depth of the well increases comes in agreement with previous reports from other countries such as Thiland (Chuah et al., 2016; KheradPisheh et al., 2016), and could be attributed to the geological structure of the study area (Al-Mikhlafi, 2010). According to the WHO recommendations (World Health Organization, 2017), fluoride concentration in the range of 0.5 to 1.5 mg/L is considered optimal.

However, although 75% of the wells included in this study contain fluoride at levels lower than the maximum permissible level (MPL) recommended by WHO, it should be noted that the WHO guidelines give a relaxation regarding the set values of MPL by stating that each country should consider other factors such as climate, water consumption and diet when setting the MPL for fluoride (Fawell et al., 2006). In the current study, the concentration of fluoride in two out of the sixteen locations investigated was less than 0.5 mg/L; and, according to previous studies, fluoride content lower than 0.5 mg/L increases the risk of dental caries (Gao et al., 2013; World Health Organization, 2017). On the other hand, our results showed the presence of fluoride at higher than MPL in four wells (25%).

For children, an optimum fluoride intake in drinking water was proposed by Galagan and Vermillion (Galagan & Vermillion, 1957) who correlated fluoride concentration in drinking water to its temperature by the following equation:

$$\text{Optimal upper level for fluoride concentration} = \frac{0.022}{(0.0104\ +\ 0.000724\ \times\ \text{AMMTºC})}$$

Where AMMT stands for annual mean maximum temperatures. In Dhamar province, Yemen, AMMT equals to 28.4 ℃; so, using the above equation, optimal upper level for fluoride concentration will be 0.70 mg/L. It is obvious that more than 80% of the fluoride concentrations reported in this study are higher than the calculated optimal value. However, further validation of this equation is necessary. A published study by Kadir et al. showed that fluorosis occurred in 30.8% of the study's population of Yemeni adolescents, which was attributed to high levels of fluoride in drinking water (Kadir & Al-Maqtari, 2010). The risk of high level of fluoride is a serious issue in Dhamar governorate as is the case for other Yemeni cities.

Principal component analysis (PCA) was used to extract relevant information from the dataset collected in this study. The data matrix had the dimension of 16 monitoring locations × 10 measured variables. PC and its eigenvalues identify the most important gradients in the dataset. In this study, the first run of PCA showed negligible contributions from Fe, NO_2 and PO_4 to the level of fluoride in the analysed groundwater; hence, these variables were dropped. In the second run of PCA, the first two PCs were the most significant components, representing 77% of the total variance of the examined parameters PC1: 43.7 %, PC2: 33.2%, Figure 3.

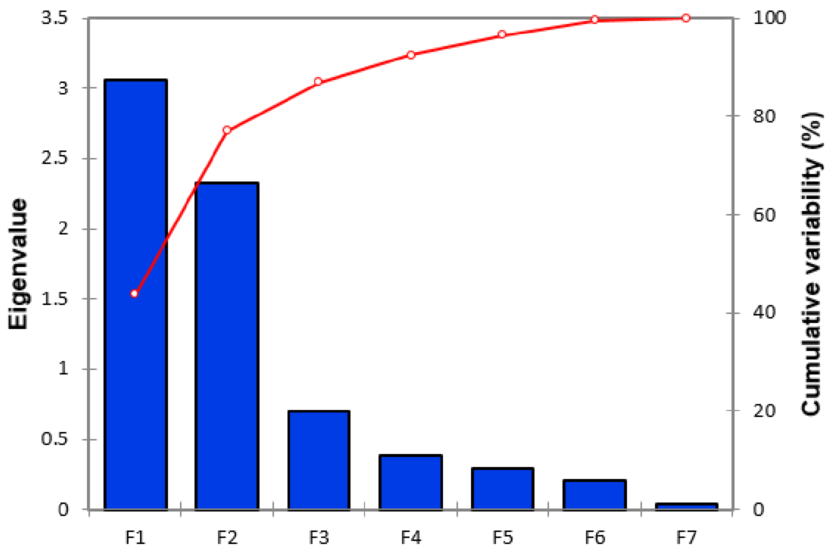

Figure 3 | The scree plot between eigenvalues and factors

As listed in Table 5, PC1 has high positive loadings of depth (0.574), EC (0.950), SO_4^{2-} (0.787) and NO_3^{1-} (0.869). The combination of these parameters on PC1 suggests that the level of fluoride was significantly affected by well depth and ionic strength. PC2 accounts for 33.2% of the total variance, and has high loadings of Ca (0.857), Mg (0.873). From the loadings' signs, it can be suggested that PC1 negatively affects the level of fluoride, whereas PC2 positively affects the level of fluoride. Scores plot in Figure 4 was applied to cluster the monitoring points along the study area according to common characteristics.

Table 5 | Loadings of the principal component analysis for the groundwater parameters

Variables	Loading	
	PC1 (ionic strength)	**PC2** (hardness)
Depth (m)	**0.574***	**-0.593**
EC (µS/cm)	**0.950**	0.072
Ca (mg/l)	0.340	**0.857**
Mg (mg/l)	0.129	**0.873**
F (mg/l)	-0.569	**0.635**
SO_4 (mg/l)	**0.787**	0.001
NO_3 (mg/l)	**0.869**	0.263
Eigenvalue	3.061	2.327
Variability (%)	43.735	33.239
Cumulative (%)	43.735	76.974

**Large loadings having an absolute value ≥ 0.5 are boldfaced.*

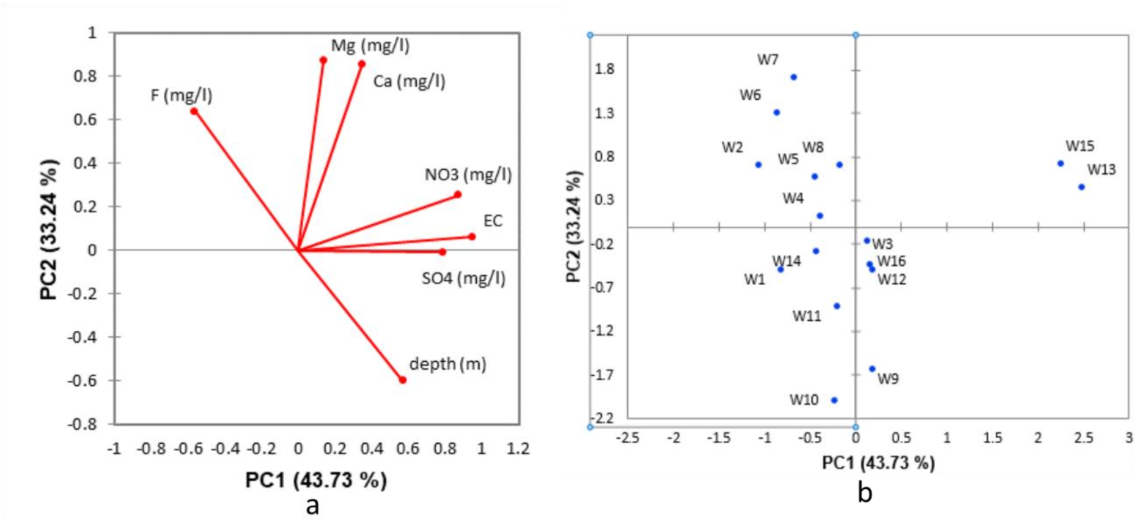

Figure 4 | Principal components analysis: a) Loading plot; b) score plot

As regards PC1, two groups can be characterized; group-1 (the wells 1, 2, 4, 5, 6, 7, 8, 10, 11, and 14) was distributed on the left direction of PC1 and was characterized by high fluoride content 0.94 – 1.84 mg/L), except for W10 which could be considered an outlier of this group. Group-2 (the wells 3, 9, 12, 13, 15, and 16) was distributed on the right direction of PC2 and was characterized by low fluoride content (≤1 mg/L). These results further show that PC1 clusters the data set according to fluoride content. It is also clear that this grouping was affected by well depth. On the other hand, PC2 clusters data according to hardness level. Group-3 (the wells 2, 4, 5, 6, 7, 8, 13 and 15) was distributed on the positive side of PC2 and was characterized by high hardness content; Ca: 93 – 130 mg/L, and Mg: 39 – 60 mg/L. Group-4 (the wells 1, 3, 9, 10, 11, 12, 14, and 16) was distributed on the negative side of PC2 and was characterized by low hardness content; Ca: 55 – 102 mg/L, and Mg: 23 – 42 mg/L. The weak correlation between fluoride and magnesium (R= 0.46) could be attributed to the release of fluoride from magnesium-bearing minerals such as biotite, hornblende, or weathering of apatite/hydroxyapatites found in the well geological structure (Manikandan et al., 2014). From biology's point of view, fluoride and magnesium ions have counter effects as each of them counters the biological activity of the other (Mackoy-Mokrzynska A, 1995). The negligible correlation between Ca and fluoride concentration (Table 3) confirms previously reported findings (Chae et al., 2006; Manikandan et al., 2014).

However, several factors including sampling design, number of constituents analyzed, data quality, and data pre-treatment may affect the effectiveness of principal component analysis (PCA) for identifying important environmental factors in a given study (Olsen et al., 2012).

Conclusions

Data generated in this study showed that concentration of fluoride in most of the investigated groundwater sources fall within the permissible levels set by the World Health Organization. Within the limitations of this study, statistical treatment of these factors using PCA method suggests that the level of fluoride in the investigated wells is significantly affected by well depth and ionic strength which, in turn, reflect the geological structure of the area.

However, other factors such as seasons, climate, and temperature need to be investigated along with a comprehensive and systematic risk assessment.

It must be emphasized, however, that health risk for Dhamar city's residents exposed to fluoride from drinking water should not be estimated on the basis of water consumption only without taking into consideration other inputs that may significantly contribute to the effective fluoride daily intake. These inputs include beverages, foods and fluoride supplements.

Finally, the authors urge for further studies in the region to investigate any correlation between the abnormal content fluoride and population health, especially the increase in kidney and renal diseases reported by several public health agencies.

Conflict of interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Author contribution statements

Hefdhallah Al Aizari: Designed the experiments and wrote the first draft; Ali Al Aizari: Conducted the experiments; Rachida Fegrouche: analyzed the data and wrote the first draft; Saeed S. Albaseer: Analyzed the data and wrote the final draft.

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