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Research Article

FLUORIDE CONTENT OF GROUNDWATER AND HEALTH IMPLICATIONS IN THE SOUTHERN PART CATCHMENT AREA OF THE UPPER VALLEY RASYAN, GOVERNORATE OF TAIZ, YEMEN

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ABSTRACT

Fluorosis continues to be an endemic problem in Yemen. More areas are being affected by fluorosis in different parts of the country. The present study tries to study the source offluoride in groundwater of the affected areas through an analytical study of three areas in the southern part of the upper valley Rasyan, governorate of Taizin Yemen. 93 wells based on the inventory of wells in the study area, which included samples of all types of aquifers. The results of the analyzed parameters formed the attribute data base for geographical information system (GIS) analysis and final output maps. Fluoride average concentration ranges from 0.85 mg / L as a minimum value in the samples taken from Al-Dabab to 2.83 mg / l as a maximum value of the samples taken from Central region. 82.76% and 75% of the groundwater samples respectively in Central and Al-Hawban area containing fluoride concentration that exceed the WHO drinking water guidelines value of 1.5 mg/l. The sewage and other wastes are the main anthropogenic source of water contamination with natural contamination by the water rock interaction in the studied areas, the exacerbates of the problems are the lack of proper treatment of the areas sewage, the lack of good drainage system.

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INTRODUCTION

Fluoride (F⁻) is an inorganic ion which is found in all type of water from low to high concentrations and it occurs in rocks, soil, air, water, plants and animals as well as in the human body. The subsurface water, most of which originates from rainfall or surface water bodies, gains minerals during its transport and residency period of earth's crust (Raju, 2012; Varol et al., 2013; Raju et al., 2014; Singh et al., 2015; Xiao et al., 2015; Patel et al., 2016). In groundwater, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the temperature and the action of other chemical elements (Tahaikt et al., 2008).

Fluoride ion in drinking water is known for both beneficial and detrimental effects on health. Fluoride in small amounts is an

essential component for normal mineralization of bones and formation of dental enamel (Bell and Ludwig, 1970). However, excessive intake of fluoride can cause dental and skeleton fluorosis (Sorg 1978; Mahramanlioglu et al., 2002). Due to its strong electronegativity, fluoride is attracted by positively charged calcium in teeth and bones (Susheela et al., 1993). Fluorosis is a considerable health problem worldwide, which is afflicting millions of people in many areas of the world, for example East Africa (Nanyaro et al., 1984; Gaciri and Davies, 1993; Gizaw, 1996), India (Subba and Devadas, 2003; Gupta et al., 2005; Jacks et al., 2005). According to World Health Organization (WHO) Guidelines for Drinking Water Quality (WHO, 2006) the limit value for fluoride is 1.5 mg/l. The value of 1.5 mg/l is a guiding value, which may be changed based on climatic conditions like temperature, humidity, volume of water intake, fluoride from other sources etc for different regions of the world (Viswanatham, 2008).

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Fluorosis continues to be an endemic problem in Yemen. More areas are being are affected by fluorosis in different parts of the country. Recently, a report from General Authority of Rural Water Projects (GARWP) indicates markedly increasing in fluoride content in groundwater (Between 2000 and 2006) in districts of some governorates such as Sana'a, Ibb, Dhamar, Taiz, Al-Dhalei and Raimah. The highest fluoride concentration in drinking water was reported in some districts of Sana'a governorate, especially Sanhan (UNICEF, 2008).

Most Yemenis dwelling in rural areas use deep well water for drinking and household works-and a large number of these wells are contaminated with fluoride in a concentration of 2.5 to 32 milligrams (Viswanatham, 2008).

Unfortunately there are no published literature available on fluoride and fluorosis aspects of Taiz area.

The present study tries to identify the intensity and the spatial extent of the existing groundwater contamination by fluoride in the study area and try to identify sources pollution responsible for the current pollution of the affected areas through an analytical study in the southern part of the upper valley Rasyan of Taiz governorate in Yemen.

MATERIALS AND METHODS

Study area

The study area is a southern part of upper Wadi Rasyan catchment, it's densely populated and includes Taiz city which represented the third largest and important cities in Yemen (Figure 1). Taiz, a city located in the republic of Yemen, 100 km east of the red sea, and away from the city of Sana'a is estimated at a distance of 253 km to the south, surrounded by highlands, especially from the south, such as mount Sabir, which rise up to 3,000 meters above sea level, while Taiz city center is located at an altitude of 1300 meters from almost sea level.

The study area contains three sub-basins as shown below:

- TAIZ CITY FIELD (Central sub-basin) (213.698 km2).
- AL-HAWBAN FIELD (146.08 km2).
- AL-DABAB FIELD (112.42 km2).



Geomorphology of study area

The relief is controlled by volcanic rocks and tectonic movements. The structural features of the area are primarily the results of an intensive igneous volcanic activity which began in the late cretaceous period and continued until the quaternary period. Most of the volcanic rocks consist of basalts, andesite, trachyte and tuffs (Figure 2). During the intervals of little or no igneous activity, sediments of fluvial or lacustrine origin have been deposited. Thus the geological formations in the Taiz area consist of a Precambrian crystalline unit, overlain by consolidated sediments, reported to range from Paleozoic to Mesozonic in age, capped by the Mesozoic and tertiary Yemen volcanic. Quaternary alluvial deposits fill up the valleys in the area. The alluvium is about 20 m thick in the upper Wadi channels but reach some 70 m or more in the alluvial plains. the high level alluvial areas are usually unconfined, but the downstream areas are semi-confined by a clayey layer or some 5-10 m thick. Static water levels in the alluvium vary from 5-15m. Nowadays, the dominant morphological feature of the area is Sabir Mountain, a post-volcanic granite intrusion while quaternary basalt flows, river terraces, alluvial fans and wind deposits make up the present landscape (Van Der Well, 1997).

Sampling

In order to evaluate the extent of groundwater contamination with fluoride, was selected 93 wells based on the inventory of wells in the study area (Figure 3), which included samples of all types of aquifers, and the type of wells, groundwater samples were collected in polyethylene bottles of 1000-ml capacity after rinsed with distilled water and the water of the well, through months in August, September and October 2014. The fluoride concentration of groundwater samples was determined using DR 2800 spectrophotometer.

Spatial distribution of fluoride

The spatial distribution of fluoride in groundwater samples in the study area is represented as a thematic layer using Map info GIS software.

Statistical methods

The Fisher test was used when comparing dichotomous data separately and Pearson's correlation coefficient for continuous variables. On the other hand, the Kruskal-Wallis H. test and box plot were used in the analysis of the data.



The Kruskal-Wallis test is a non-parametric test used to compare three or more samples. It is used to test the hypothesis that the different samples in the comparison were drawn from the same distribution or from distributions with the same median (Büyüköztürk, 2004). In the study, Al-hawban, Central and AL-Dabab areas were all different samples. After, we performed logistic regression analysis. fluoride was included as a dichotomous variable (lower or greater than 1.5mg/l). Other variables with p-values < 0.2 in the univariate analysis, were entered into the multivariate logistic regression model. We investigated the association between fluoride which was included as a dichotomous variable (lower or greater than 1.5mg/L) and each studied variable through odds ratio (OR) computed by regression binary logistic. Model 1 includes only the univariate association between dichotomous and each studied variable (crude model). Model 2 includes the simultaneous multivariate analysis of independent variables for fluoride.

In order to assess the accuracy of the estimates, we have indicated the 95% confidence interval (IC to 95%) of the average data. A p-value of less than 0.05 at 95% confidence level was considered as statistically significant.

RESULTS

Fluoride concentration in groundwater of the study area

In order to enable sustainable development of groundwater resources, it is necessary to delineate the safe and unsafe zones with reference to fluoride content; hence spatial distribution of high fluoride concentration was mapped in the three regions of Taiz (Figure 4).69% of groundwater samples in the central region have between 1.5 and 4 mg/L of fluoride, 15% between 4.1 and 10 mg/L, remaining only 17.24% groundwater samples have less than 1.5 mg/L fluoride. In Al-Hawban region these results are almost the same while AL-Dabab region was safer (Figure 5).



Figure 4Spatial distribution of fluoride concentration of in the study area.



Figure 5 Concentration distribution of fluoride (mg/l) by region

This figure is a box plot with upper and lower quartiles of the data defining the top and the bottom of a rectangle box. The line inside the box represents the median value and the size of the box represents the spread of the central value (TaheriTizro and Voudouris, 2008). In majority of the samples fluoride in Al-Hawban area is above the median in the plot, whereas in the majority of the samples fluoride in Central and AL-Dabab area are below the median to minimum level. Based on the Kruskal-Wallis test for the various areas, the level of significance was (p<0.05). This result illustrated that there were significant differences in groundwater fluoride. The geochemical trend of groundwater fluoride in the study area (Figure 6) demonstrates that fluoride in Central and Al-Hawban area is the dominant, (Central (F-)> Al-Hawban (F-)> AL-Dabab (F-). A multiple comparison of median concentration among these areas in fluoride shows that, Al-Hawban and Central region reach higher fluoride content which is more than 1.5 mg/L and similar but significantly higher than the region AL-Dabab; Al-Hawban-Central: (p>0.05); Al-Hawban- AL-Dabab: (p<0.0001); AL-Dabab-Central: (p<0.0001) (Figure 6).



Figure 6 Box plot for the maximum, minimum and average of the fluoride content in groundwater of three areas.

The results of studied samples given in the table 1, shows that the distribution of fluoride content by region in three well types is not significantly different.

 Table 1 Distribution of fluoride content by region in three well types

Degion	Well type -	Fluoride content*		
Region		Normal	Abnormal	
Al-Hawban	Borehole	4 (30,8%)	9 (69,2%)	
	Dug well	6 (21,4%)	22 (78,6%)	
	Spring	2 (40,0%)	3 (60,0%)	
Central	Borehole	1 (33,3%)	2 (66,7%)	
	Dug well	1 (20,0%)	4 (80,0%)	
AL-Dabab	Borehole	1 (100,0%)	-	
	Dug well	6 (100,0%)	-	
	Spring	1 (100,0%)	-	

*:The difference is not significant between the three types of wells between Central and Al-Hawban region.

The distribution of fluoride concentration according to water type in the groundwater samples showed more variation in the AL-Dabab, Central or Al-Hawban region. Normal concentration of fluoride was for AL-Dabab region Ca-HCO3 and Mg-HCO3 type groundwater, as compared to that for Central or Al-Haw ban region. However, abnormal concentration of fluoride was more prevalent with Na-Cl, Na-HCO3 in Central and Ca-HCO3, Na-So4, Ca-So4 and Mg-Cl type groundwater in Al-Hawban area (Figure 7).

On the other hand, high fluoride waters are Na-HCO3, Na-So4, Na-Cl, Ca-So4, Mg-Cl, Ca-Cl type groundwater and low fluoride waters are Mg-HCO3 and Ca-HCO3 type groundwater (Table 2).



Figure 7 Box plot for the maximum, minimum and average of the fluoride content in groundwater of three areas according to water type.

 Table 2 distribution of abnormal concentration fluoride according to type groundwater

Water type	Normal concentration N(%)	Abnormal concentration N(%)	P-value	
Mg-HCO3	6(85.7%)	1(14.3%)	1	
Ca-HCO3	6(60.0%)	4(40.0%)	0.3	
Mg-SO4	1(33.3%)	2(66.7%)	0.10	
Na-HCO3	8(28.6%)	20(71.4%)	0.01	
Na-SO4	3(27.3%)	8(72.7%)	0.02	
Na-Cl	9(25.0%)	27(75.0%)	0.01	
Ca-SO4	1(25.0%)	3(75.0%)	0.05	
Mg-Cl	1(16.7%)	5(83.3%)	0.02	
Ca-Cl	1(16.7%)	5(83.3%)	0.02	

The distribution of type groundwater according to pH showed that from Mg-So4 type through a Na-HCO3, Na-So4 and Na-Cl type groundwater to Mg-Cl and ultimately Ca-Cl when the pH range between 7.3 and 8 was more lower than pH \neq [7.3-8] (Figure 8)and the comparison of the groups« pH [7.3-8]» and « pH \neq [7.3-8]» showed a significant difference in fluoride concentration (p<0.05).



Figure 8 Distribution of water type according to pH

Results of the multivariate analysis are shown in table 3. In the two logistic regression model, after adjusting for pH, water

type and region (ORa=0,366,176-0,76), the water type (OR_a=1,71; CI: 1,261-2,32) remained dependently associated with abnormal fluoride concentration (Table 3).

 Table 3 Logistic regression univariate and multivariate model for fluoride variation

	OR _{crude}	CI for 95%	p-value	OR _a	CI for 95%	p-value
pH[7.3-8]	0,417	0,164-1,062	0,067	0,373	0,139-0,99	0,050
Water type	1,755	1,295-2,378	0.000	1,711	1,261-2,322	0,001
Region	0,389	0,194-0,778	0.008	0,366	0,176-0,760	0,007

ORa: Odds ratio adjusted

Al-Hawban region: reference

The comparison of the abnormal and normal fluoride concentration according to TDS (Total dissolved solids) in the three areas, showed a significant differences between the three areas p<0.0001) and positive relationship between the fluoride concentration and TDS(r=0.5; p<0.0001). A multiple comparison of median concentration among these areas in fluoride shows that, Al-Hawban and Central region reach higher fluoride content which is more than 1.5 mg/L and similar but significantly higher than the region Al-Dabab; Al-Hawban -Central: (p>0.072); Al-Hawban-Al-Dabab: (p<0.003); Al-Dabab-Central: (p<0.0001).Moreover, this increase in TDS has been found only at two regions when the fluoride is abnormal content (Figure 9).

The results of the study samples of electrical conductivity and TDS according to study areas given in the figure 10.



Figure 9 Box plot for the maximum, minimum and average of the fluoride content in groundwater according to TDS of three areas.



Figure 10 Box plot for the maximum, minimum and average of Electrical Conductivity and total dissolved solids according to study areas.

The topography of the study area, which determine the direction of the runoff, slope degree, factory distribution and contamination source inventory, population distribution and fault according to three areas are shown in figure 11 and 12.



Figure 11 Topography of the study area, slope degree and factory distribution according to three areas.



Figure12 Contamination source inventory, population distribution and fault in the three areas.

pH≠[7.3-8]: reference

Mg-HCO3 water type: reference

DISCUSSION

Much of the fluoride entering the body is from water and hydrogeochemistry of fluoride in water is therefore of major concern. The fluoride is found in the atmosphere, soil and water. It enters the soil through weathering of rocks, precipitation or waste runoff. Understanding of the fluoride occurrence is important in the management of the fluoride related epidemiological problems. It's clearly observed that the Central and Al-Hawban areas have the highest concentration of fluoride ion in the chemistry of water. Fluoride average concentration ranges from 0.85 mg / L as a minimum value in the samples taken from AL-Dabab to 2.83 mg / l as a maximum value of the samples taken from Central region. 82.76% and 75% of the groundwater samples respectively in Central and Al-Hawban area containing fluoride concentration that exceed the WHO drinking water guidelines value of 1.5 mg/l. Even though the WHO guidelines have recommended an upper limit of fluoride concentration in drinking water as 1.5 mg/l, in many tropical countries where there is a high sweat loss and a high intake of water due to the hot weather such an upper limit may be unsuitable (Brouwer et al., 1988). Based on WHO recommended guidelines for fluoride in drinking water. The population living in these areas is very dense and thus susceptible to higher dental and chronic skeletal fluorosis. Highest concentrations were found to be 10 mg/l from Central area, 5.81 mg/l from Al-Howban unlike the Dabab area which remains unaffected by the contamination fluoride of groundwater. According to report of RYMW (2009) in the selected villages, the dental fluorosis is the widely fluoride disease observed in the affected areas, there is a positive relationship between fluoride in water and the occurrence of dental fluorosis in Taiz region.

In order to understand the vertical distribution of the fluoride ion concentration from the water of the study area, the type of the sample water (Dug, Borehole and Spring) evaluated separately. There was no significant difference between the three well types dug well sample, springs and bore wells. It can be concluded that shallow aquifers do not reflect higher fluoride contamination than deeper aquifers.

It's observed that most of the water samples showed enhanced concentrations with generally increasing trends to the low elevated area (Central area) while the high elevation shows low concentration of fluoride (AL-Dabab area). All the water samples collected from the uphill zones of Dabab area were exhibited low fluoride concentration.

Compared with Na-HCO3 type groundwater, Ca-HCO3 type groundwater is known to generally contain lower fluoride (Lee *et al.*, 1997). Its hydrochemistryis characterized by increased Ca²⁺ ion concentration with increasing total dissolved solid due to the gradual dissolution of carbonate minerals or Ca²⁺ bearing plagioclase in aquifer materials (Yun *et al.* 1998a and b; Sujatha 2003). The Na-HCO3 type groundwater are generally enriched in fluoride and sodium ions, due to the dissolution of silicates as well as the removal of Ca²⁺ by calcite precipitation and cation exchange (Chae *et al.*, 2005; Kim and Jeong, 2005). This situation of solubility control on the higher concentration of fluoride can be explained by the fact that fluoride ions in

of fluoride can be explained by the fact that fluoride ions in groundwater can be increased as a result of precipitation of CaCO3 at high pH, which removes Ca2+ from solution allowing more fluorite to dissolve.

These released Ca2+ ions combine with CO3²⁻ ions to further enhance the precipitation of CaCO3. Therefore, fluorite under saturation in groundwater of area under study might be due to the calcite saturation, preventing it by reducing calcium activity and allowing more fluorite to dissolve there by increasing the F/Ca ratio of solution. Hence calcite and fluorite are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater of Al-Hawban Basin (RYMW, 2009). The solubility limits for fluorite and calcite provide a natural control on water composition in a view that calcium, fluoride and carbonate activities are interdependent (Kundu *et al.*, 2001).

Electrical Conductivity(the concentration of dissolved salts in these wells, especially in Al-Hawban wells because of the swamps salt in the area. In addition to the effect of those areas by different liquid waste by runoff and sewage disposal, the heavy pumping of well water is also contributed because of the scarcity of water which leads to the increase of the concentration of salt in the water).

TDS levels ranged widely from 291 to 6188 mg/L with most stations levels above 400 mg/L and many of the samples studied were higher than the permissible limit of 1500 mg/l according to WHO (2003). This wide variation in TDS values indicates that the area hydrochemistry is influenced by diverse processes such as water_rock interaction and anthropogenic pollution. Fluoride concentrations frequently are proportional to the degree of water-rock interaction because fluoride primarily originates from the geology (Gizaw 1996; Banks *et al.*, 1995; Dowgiałło, 2000; Frengstad *et al.*, 2001; Carrillo-Rivera *et al.*, 2002).

Due to the high rainfall, rugged topography, factories, lack of total coverage per sewerage network, population density and faults in the study areas could also explain this high fluoride content by Runoff and infiltration of chemical fertilizers in agricultural areas, Septic and sewage treatment system discharges with fluoridated water supplies and liquid waste from industrial sources The topography of the study areas varies from level plain to steep slopes. Study area ranges in elevation between 900 to 3000 m above sea level. Taiz plain receives about 500 mm/yr of rainfall and significant recharge form runoff of surrounding mountains (Abdulaziz, 2005).

In addition to this groundwater fluoride pollution that can affect human health, there have been indications that uptake of fluoride from other sources like food, dust and beverages may be many times higher than that of water (Chowdhury *et al.*, 1990).

About the Fluorosis in selected Villages of Taiz Governorate, the percentage of children with fluorosis was very high. Not only because of drinking water, various foods habits (like drinking black tea and Chewing Qat) indicated a high contribution of fluoride to food. In AL-Hawban area, some of children, especially from Jabal Sabir area, usedto chew Qat daily, and the Qat are cultivated in the man-made terraces of Jabal Sabir alkali granite, where it expected to be the main source of F⁻reach minerals like Fluorite (RYMW, 2009). On the other hand, the use of fluoridated water for cooking increases the fluoride content significantly especially in dry foods like maize flour which absorbs much water during cooking. It has been reported that fluoride availability may be influenced by simultaneous intake of food and fluoride containing compounds in a positive or negative manner depending on the food type, mode of administration and type of fluoride compound (Trautner and Einwag, 1989).

CONCLUSION

Al-Hawban and Central are the worst areas affected by fluoride contamination in drinking water where 69.5 % of F concentration above the permissible limit and alternate water sources will be difficult. Therefore, defluoridation of drinking water is the only practicable option to overcome the problem of excessive fluoride in drinking water in these areas. More refined studies however need to be done before any long-term intervention efforts can be planned. In the meantime, there is a critical need to educate young Yemenis about fluorosis and simple intervention measures to avoid long term health problems. Other studies in the region are urged studying the cause and effect relationship between the abnormal content fluoride and population health.

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