

# Occurrence of fluoride in ground waters of Saudi Arabia

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**Abstract** The presence of elevated levels of fluoride in groundwater is considered a global problem. Fluoride in water derives mainly from dissolution of natural minerals in the rocks and soils with which water interacts. The most common fluorine-bearing minerals are fluorite, apatite and micas. Anthropogenic sources of fluoride include agricultural fertilizers and combustion of coal. In the present research, a survey of wells ( $n = 1,060$ ) was undertaken in all the 13 regions of the Kingdom of Saudi Arabia to assess the contained fluoride (F) levels. The results indicated variation in fluoride levels from 0.10 to 5.4 mg/L as F throughout the kingdom. The average fluoride levels in milligrams per liter as F were as follows in descending order: 1.80 (Hadwood Shamalyah), 1.37 (Hail), 1.33 (Eastern Province), 1.16 (Al Jouf), 1.11 (Qassim), 1.01 (Riyadh), 0.90 (Madina Al Munnawara), 0.81 (Tabouk), 0.74 (Makkah Al- Mukaramma), 0.73 (Jizan), 0.66 (Asir), 0.64 (Najran), and 0.60 (Al Baha). The results indicated that fluoride levels exceeded the USEPA maximum contaminant limits for drinking water (4 mg/L) in several wells ( $n = 7$ ) in different regions of the kingdom and that 13.96 % of the wells exceeded the World Health Organization recommended levels (1.5 mg/L). The results were also compared with the secondary USEPA contaminant standards of 2.0 mg/L for fluorides.

**Keywords** Fluoride · Ground water · Saudi Arabia · Occurrence · Health effects

## Introduction

Fluoride is an essential element for both humans and animals, and its behavior in drinking water is vital. Optimum content (1.5 mg/L) of F in water is essential for growth of bones and formation of dental enamels. Higher contents ( $>1.5$  mg/L) pose a threat to human health, and can cause severe health problems such as dental and skeletal fluorosis, particularly in arid and semi-arid regions of the world. For example, F problems occur in some regions of India, China, the Korean Peninsula, East and North Africa, the United Kingdom, and the Western United States (Ayooob and Gupta 2006; Chae et al. 2007; Harrison 2005; Jacks et al. 2005; Kim and Jeong 2005; Reddy et al. 2010; Xiong et al. 2007; Zhu et al. 2007). Conversely, water fluoridation is necessary in Australia, New Zealand, Canada, and most South American countries, due to lack of natural fluoride. However, Scandinavian countries where dental awareness is very high, use alternative sources, such as fluoride-rich toothpaste (Lennon et al. 2004). Fluorine is available in soils and waters due to the weathering of fluoride-bearing minerals predominantly of igneous origin (Breiter and Kronz 2004; Breiter et al. 2006; Lukkari and Holtz 2007; Reddy et al. 2010; Totsche et al. 2000; Zhu et al. 2007). Apatite [ $\text{Ca}_5(\text{PO}_4)_3\text{F}$ ] and fluorite [ $\text{CaF}_2$ ] are the most common fluoride-bearing minerals. However, zircon [ $\text{ZrSiO}_4$ ] is also a possible source for fluoride in water or soil, as igneous zircons contain fluorine complexes (Farges 1996; Keppler 1993). Biotite [ $\text{K}(\text{Mg},\text{Fe})_3\text{AlSi}_3\text{O}_{10}(\text{F},\text{OH})_2$ ], hornblende [ $(\text{Ca},\text{Na})_{2-3}(\text{Mg},\text{Fe},\text{Al})_5(\text{Al},\text{Si})_8\text{O}_{22}(\text{OH},\text{F})_2$ ] and soils that consist mainly of clay minerals such as vermiculite [ $(\text{MgFe},\text{Al})_3(\text{Al},\text{Si})_4\text{O}_{10}(\text{OH})$ ], kaolinite [ $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ] and montmorillonite [ $(\text{Na},\text{Ca})_{0.33}(\text{Al},\text{Mg})_2(\text{Si}_4\text{O}_{10})(\text{OH})_2 \cdot n\text{H}_2\text{O}$ ] are also major sources of fluoride (Wodeyar and Sreenivasan 1996; Zhu et al. 2007).

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The problems associated with high fluoride content in groundwater in the dry zone of Sri Lanka are well known (Lennon et al. 2004). It has been found that both shallow and deep groundwater exceeded the World Health Organization (WHO) recommended levels in drinking water ( $>1.5$  mg/L). Agrawal (1997) reports that in Sri Lanka, fluoride has a strong geographical control linked to climatic conditions, with high fluoride waters being restricted to the dry zone on the eastern side of the island. In some parts, wells have fluoride content  $>10$  mg/L.

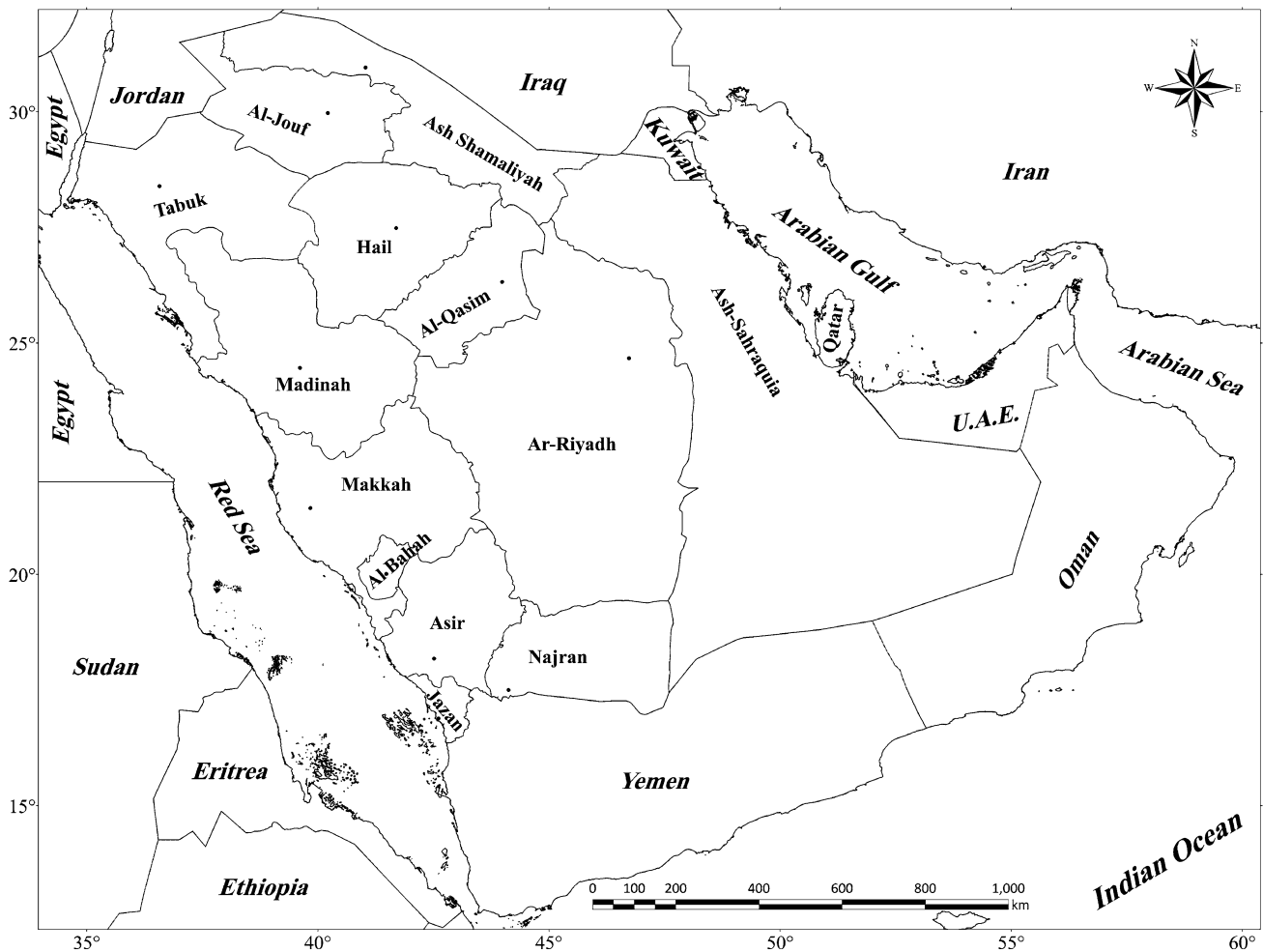
Generally, most groundwater sources have higher fluoride concentrations than surface water. As groundwater percolates through the weathered rock in the aquifers, it dissolves fluoride-bearing minerals thus releasing fluoride into solution (Falvey 1999). Because of the large number of variables, the fluoride concentrations in groundwater can range from well under 1 mg/L to more than 35 mg/L (WHO 1994). Studies have shown that the problem of high fluoride content in groundwater is very acute in mainland Tanzania (Mjengera and Mkongo 2002). The high fluoride content is attributed to some geological processes such as volcanic activities, thermal springs and the presence of minerals such as fluorite and apatite in rocks. For example, borehole water in South Sanya corridor was found to contain up to 96 mg/L fluoride. An area west of Ngorongoro Crater had fluoride content in spring water between 40 and 140 mg/L (Mjengera and Mkongo 2002). High fluoride groundwater has also been reported in Mexico (Alarcon-Herrera et al. 2001). In Guadiana Valley, drinking water supply is obtained from underground wells and is characterised by a high content of fluoride. Hurtado et al. (2000) reported that in Los Altos de Jalisco, situated in central Mexico, communities use water with a fluoride concentration of up to 12.97 mg/L. In northern China, high fluoride groundwater with fluoride concentration of up to 6.20 mg/L occurs in the Taiyuan basin (Qinghai et al. 2007). The study showed that high fluoride groundwater zones are mainly located in the discharge areas, especially in places where shallow groundwater occurs (the groundwater depth is  $<4$  m). Modeling results indicated that in the recharge and flow through area of the northern Taiyuan basin, interactions between groundwater and fluoride-rich minerals are the major factor for the increase of fluoride concentrations, whereas in the discharge area of the northern basin, evaporation as well as the mixing of karst water has greater contribution to the fluoride enrichment in groundwater. A study was carried out on fluoride distribution in Ethiopia (Redda Tekle-Haimanot et al. 2006). Of the total 1,438 water samples tested, 24.2 % had fluoride concentration above the 1.5 mg/L guideline concentration recommended by WHO. The highest fluoride levels were recorded in the Rift Valley, where 41.2 % of all samples exceeded the 1.5 mg/L level. Only 1.0 % of the samples

from the central and northwestern highlands and 10.0 % in the southeastern highlands exceeded 1.5 mg/L. Larger proportions of deep wells (50.0 %) and hot springs (90.0 %) than shallow wells (27.2 %) and cold springs (12.6 %) exceeded the 1.5 mg/L level. The highest fluoride concentrations were recorded for Rift Valley lakes Shala (264.0 mg/L) and Abijata (202.4 mg/L) and the lowest in Lake Tana, rivers, wells and springs in the highlands. Groundwater is the most widely used drinking water source in the upper regions of Ghana. In most parts of the upper regions, groundwater fluoride ranges from 0.11 to 4.60 mg/L (Apambire et al. 1998). About 28 % of the wells in the region have drinking water with fluoride concentration above the WHO recommended level. Apambire et al. (1998) recommended using geochemical health risk maps showing fluoride levels in groundwater, so that health and water officials should have information on fluoride occurrence in groundwater. Mgalela (1997) reported high fluoride groundwater in some parts of Gokwe district in Zimbabwe. He recommended a thorough mapping of all wells of this area to delineate those with fluoride content above optimum level. In Malawi, the Ministry of Health and Population conducted a survey in Machinga on occurrence of high fluoride groundwater (MOH 1996; Sibale et al. 1998). A few boreholes were sampled for fluoride determination in Machinga where fluoride content in water sources was as high as 8.6 mg/L (Sibale et al. 1998). In Nkhota Kota district, an area lying in the rift valley, water from hot springs has a fluoride content of up to about 17 mg/L and Chikwidzi spring is about 8 mg/L (Chapusa and Harrison 1975). Data on fluoride occurrence in groundwater in most parts of Malawi are not available because very little research work has been carried out on the occurrence of fluoride in drinking water supplies.

### Beneficial and toxic effects of fluoride

Fluoride is widely used in dental preparations to combat dental caries, particularly in areas of high sugar intake. These can be in the form of tablets, mouthwashes, toothpaste, varnishes or gels for local application. In some countries, fluoride may also be added to table salt or drinking water to provide protection against dental caries. The amounts added to drinking-water are such that final concentrations are usually between 0.5 and 1 mg/L (WHO 2011)

Elevated fluoride intakes can have more serious effects on skeletal tissues. Skeletal fluorosis (with adverse changes in bone structure) may be observed when drinking water contains 3–6 mg of fluoride per liter, particularly with high water consumption. Crippling skeletal fluorosis usually



**Fig. 1** Map shows different regions in the Kingdom of Saudi Arabia where well water sampling was undertaken

develops only where drinking-water contains over 10 mg of fluoride per liter (WHO 2011).

Exposure to excessive consumption of fluoride over a lifetime may lead to increased likelihood of bone fractures in adults, and may result in effects on bone leading to pain and tenderness. Children aged 8 years and younger exposed to excessive amounts of fluoride have an increased chance of developing pits in the tooth enamel, along with a range of cosmetic effects to teeth (USEPA 2012).

### MCL for fluoride concentrations

USEPA has set a maximum contaminant level (MCL) and also maximum contaminant goal limit (MCLG) of 4 mg/L for fluoride concentration in the drinking water. However, the secondary US EPA contaminant limit for fluoride is 2.0 mg/L (USEPA 2012). WHO has recommended a guideline limit of 1.5 mg/L of fluoride in drinking waters (WHO 2011).

## Materials and methods

### Study area

Groundwater samples were collected from wells ( $n = 1,060$ ) located in all 13 regions of Saudi Arabia (Fig. 1). The location of the wells was recorded using Global positioning system. The region wise breakup of sampled wells is shown in Table 1. The sampled wells were a mix of government and private property. Many of the sampled wells were the main source of drinking water for the local population.

### Collection of samples

Groundwater samples were collected in 1-L plastic bottles, which were previously thoroughly washed with tap water and rinsed with distilled water. Samples were stored in ice at the sampling locations and were refrigerated upon arrival in the laboratory until analyzed. Sampling protocol was

**Table 1** Number of Sampled wells in different regions of the Kingdom

Regions	No. of sampled wells
Eastern province	201
Riyadh	200
Qassim	108
Asir	87
Makkah Al-Mukaramma	85
Madina Al-Munnawara	71
Al Baha	61
Jizan	57
Hail	43
Najran	41
Hadwood Shamalyah	41
Al Jouf	34
Tabouk	31
Total Samples	1,060

designed in such a way that samples collected in one sampling schedule were analyzed in the shortest possible time.

#### Analysis of water samples for the determination of fluoride concentration

Water samples were analyzed according to the standard methods (APHA 1999). Water samples were analyzed for fluoride using ion chromatography (Dionex Ion Chromatograph model DX 500). The ion chromatograph was equipped with Dionex Ion Pac AS 14 analytical column with AG 14 guard column. The flow rate of the eluent (4.5 mM sodium carbonate/1.4 mM sodium bicarbonate) in the equipment was kept at 1.4 mL/min. The injection volume was 10  $\mu$ L. The system was calibrated with certified anion standards from Dionex.

#### Quality assurance and quality control program

To assess the precision and accuracy of results, replicate analysis of blank, standard, and samples was carried out. The relative standard deviations were determined to find the precision of the analysis. Recovery results were calculated for the determination of accuracy. Experiments were repeated till an accuracy of 95–105 % and precision of  $\pm 5$  % were obtained. Certified standards from Dionex were used for the calibration of the instrument. One standard with one set of sample was analyzed routinely.

#### Results and discussion

The minimum, maximum, and average fluoride concentrations in the different regions have been presented in

Table 2 and Fig. 2. Table 2 also shows the number of wells exceeding the maximum contaminant fluoride levels. The minimum and maximum fluoride concentrations detected were 0.31 and 1.70, 0.44 and 3.00, 0.29 and 0.88, 0.50 and 5.00, 0.80 and 2.40, 0.30 and 4.00, 0.60 and 1.10, 0.25 and 1.70, 0.65 and 2.00, 0.35 and 0.82, 0.10 and 5.40, 0.12 and 4.90, and 0.50 and 1.90 mg/L as F for Al Baha, Al Jouf, Asir, Eastern Province, Hadwood Shamalyah, Hail, Jizan, Makkah Al Mukaramma, Madina Al Munnawara, Najran, Qassim, Riyadh, and Tabouk regions of the Kingdom, respectively.

Only seven well water samples in the three regions of the Kingdom were found to exceed the fluoride maximum contaminant limits set by USEPA. Out of these seven wells, four were located in Qassim, two in the Riyadh region, and one in the Eastern Province area. In the Qassim region 3.7 % of the well water samples exceeded the USEPA limits whereas in Riyadh region 1 % well water samples exceeded the prescribed limits. In the Eastern Province 0.5 % well water samples exceeded the USEPA limits. Considering secondary USD EPA contaminant limits 6, 12, 10, 9, 11, and 12 well water samples from Al Jouf, Eastern Province, Hadwood Shamalyah, Hail, Qassim, and Riyadh regions, respectively exceeded the 2.0 mg/L fluoride limit. The land around these wells is mainly used for agricultural and residential purposes. However, some of the wells were located in the desert area. It can be said that source of fluoride is from agriculture practices, geological sources, and human activities.

Higher content of fluoride in drinking water can cause severe health problems, particularly in arid and semi-arid regions. This could be due to the prevalent climatic conditions which necessitate people to drink water in quantities higher than what people drink in temperate regions. To investigate the relationship between fluoride levels in well drinking water, the severity of dental fluorosis and dental caries in the Hail region of Saudi Arabia, 2,355 rural children aged 12–15 years were examined. Over 90 % of the children had fluorosed teeth and Chi square tests showed a strong association ( $P < 0.001$ ) between fluoride level (0.5–2.8 ppm) in well drinking water and severity of dental fluorosis (Akpata et al. 1997).

In setting national standards or local guidelines for fluoride or in evaluating the possible health consequences of exposure to fluoride, it is essential to consider the average daily intake of water by the population of interest and the intake of fluoride from other sources (e.g. from food and air). Where the intakes are likely to approach, or be greater than, 6 mg/day, it would be appropriate to consider setting a standard or local guideline at a concentration lower than 1.5 mg/L (WHO 2011).

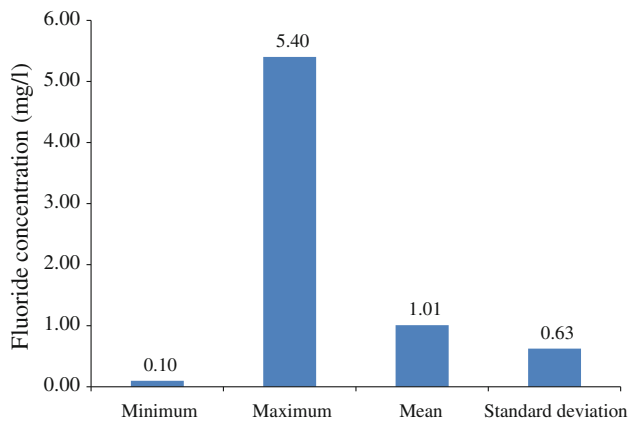
**Table 2** Concentration of fluoride in different regions of the kingdom of Saudi Arabia

Region	Fluoride concentration mg/L			Standard deviation	No. of samples exceeding the maximum contaminant limits			Percentage of wells exceeding the limits		
	Minimum	Maximum	Average		USEPA MCL	USEPA SMCL	WHO	USEPA MCL	USEPA SMCL	WHO
Al Baha	0.31	1.70	0.60	0.22	0.00	0.00	2	0.00	0.00	3
Al Jouf	0.44	3.00	1.16	0.66	0.00	6	7	0.00	17.6	20.5
Asir	0.29	0.88	0.66	0.12	0.00	0.00	0.00	0.0	0.00	0.00
Eastern province	0.50	5.00	1.33	0.44	1	12	26	0.5	6	13
Hadwed Shamalyah	0.80	2.40	1.80	0.35	0.00	10	35	0.00	24.4	85
Hail	0.30	4.00	1.37	1.02	0.00	9	12	0.00	22.5	30
Jizan	0.60	1.10	0.73	0.08	0.00	0.00	0.0	0.00	0.00	0.00
Makkah Al-Mukaramma	0.25	1.70	0.74	0.18	0.00	0.00	1	0.00	0.00	1.2
Madina Al-Munnawara	0.65	2.00	0.90	0.28	0.00	0.00	4	0.00	0.00	5.6
Najran	0.35	0.82	0.64	0.11	0.00	0.00	0.00	0.00	0.00	0.0
Qassim	0.10	5.40	1.11	1.01	4	11	23	3.7	10.2	21
Riyadh	0.12	4.90	1.01	0.69	2	12	36	1	6	18
Tabouk	0.50	1.90	0.81	0.39	0.00	0	2	0.00	0.00	6.5

USEPA primary maximum contaminant limits: 4.0 mg/L

USEPA secondary maximum contaminants limits (SMCL) for fluoride: 2.0 mg/L

WHO guideline limits: 1.5 mg/L



**Fig. 2** Minimum, maximum levels, mean and standard deviation of fluoride in Saudi Arabia

### Correlation of fluoride concentration with depth of the sampled wells

Fluoride concentrations were plotted against the depths of the sampled wells. Observed fluoride concentrations versus well depths in different regions of the Kingdom are shown in Fig. 3. It is known that depths of the deep wells can go up to thousands of meters. The figure shows that comparatively higher levels of fluoride were found in relatively less deep wells (1,000 meters). These relatively higher fluoride concentrations may be attributed to the geological

factors as well to the contamination from the external sources.

### Correlation of fluoride concentration with other cationic and anionic elements

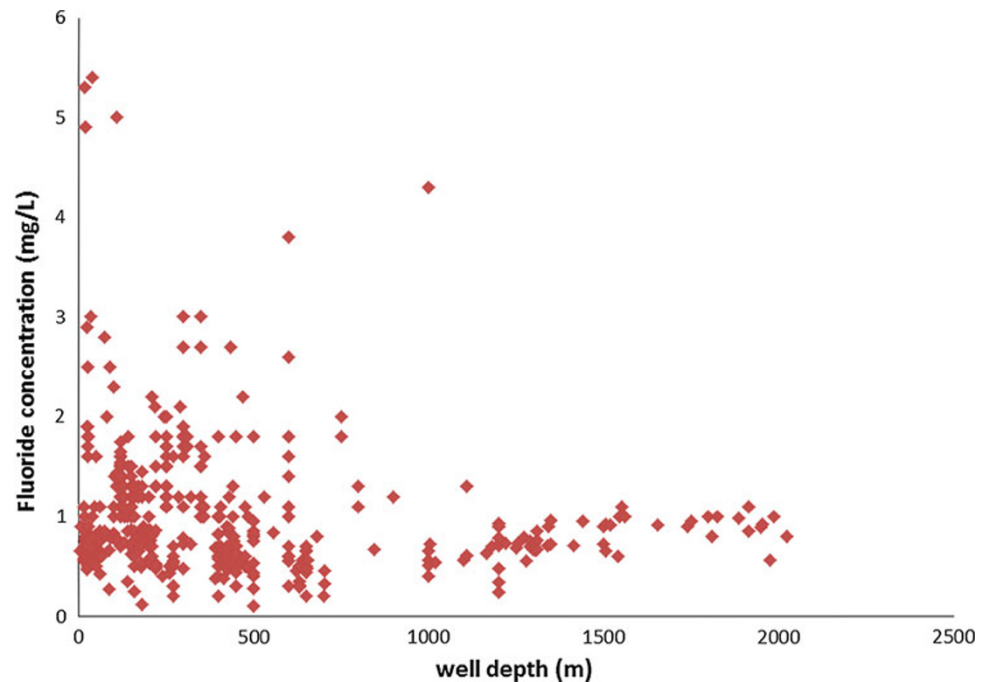
The correlation of determined fluoride concentrations in the well water samples collected from different locations in the kingdom with other major anions and cations has been presented in Table 3. The software SPSS was used to calculate the correlation between fluoride and other major cations and anions. The highest correlation was found with the parameters calcium (0.62) and sulphate (0.62) followed by sodium (0.54) and magnesium (0.52). The fluoride content was least correlated with the parameter potassium (0.26).

### Conclusions and recommendations

1. Well waters were collected in 13 regions of the Kingdom of Saudi Arabia to evaluate the fluoride concentrations. In all 1,060 wells were sampled.
2. The results indicated that fluoride levels exceeded the maximum allowed limits in considerable number of wells.



**Fig. 3** Correlation of fluoride concentrations with well depths



**Table 3** Correlation of Fluoride with major cations and anions

Parameters	Correlation factor
Calcium (Ca)	0.62
Magnesium (Mg)	0.52
Sodium (Na)	0.54
Potassium (K)	0.26
Chloride (Cl)	0.50
Nitrate (NO <sub>3</sub> )	0.35
Sulphate (SO <sub>4</sub> )	0.62

- The maximum percentage of wells exceeding the US EPA maximum contaminant limit of 4.0 mg/L fluoride limits was in Qassim (3.7 %).
- The maximum percentage of wells exceeding the secondary US EPA maximum contaminant limit of 2.0 mg/L fluoride was in the Hail region (22.5 %).
- The maximum percentage of wells exceeding the WHO guide line limit of 1.5 mg/L fluoride was in the Hadwad Shamalyah (85 %).
- It is recommended to control the possible sources of groundwater contamination by enforcing drinking water standards and monitoring agricultural activities and speedup completion of sewage system.
- It is also recommended to treat water in areas where fluoride levels have exceeded the prescribed limits. In addition, measures are needed to be taken to close down wells which contain very high fluoride levels.

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