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RESEARCH ARTICLE

HYDROGEOCHEMICAL CHARACTERIZATION AND ENVIRONMENTAL IMPACT OF FLUORIDE CONTAMINATION IN GROUNDWATER FROM AL-DHALA BASIN, YEMEN

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Abstract

Groundwater is the only source of water in the Al-Dhala Basin, and its quality is important because it defines the suitability of the groundwater for drinking and other domestic uses. The main objective of the study was to obtain a better understanding of the factors controlling the high levels of F in groundwater samples. Groundwater samples from the study area were collected from 28 selected wells. The areas with high fluoride concentrations have been identified, and the possible causes for its variation have been investigated. The regional hydrogeochemical investigation indicates that water-rock interaction is probably the main reason for the high concentration of ions in groundwater. The geochemical modeling indicates that calcite and fluorite are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater of the study area.

The fluoride ion concentration in Al-Dhala Basin comprises 71% of the water used for the drinking purpose. Dental and skeletal fluorosis are present in people in the area who receive their drinking water sources at high levels of fluoride from the wells. The population of the study area is at high risk because of the excessive intake of fluoride, especially in the absence of knowledge of the fluoride consumption.

Keywords: Hydrogeochemistry, Fluoride, Groundwater, Al-Dhala Basin, Al-Dhala, Yemen.

1 Introduction

Fluoride is known to occur at high concentrations in various parts of the world and can have significant adverse effects on public health and wellbeing under such conditions. About one-third of the world's population uses groundwater for domestic, agricultural and industrial uses **[1]**. Safe drinking water supply and deterioration of groundwater quality are a major problem worldwide **[2,3]**. The main sources of Fluoride ions in groundwater are derived from the weathering and leaching of amphiboles, fluorite, apatite, and mica. It occurs as Fluoride ion naturally in soils and groundwater due to chemical weathering of some F-bearing minerals **[4,5]** (Sajil Kumar et al. 2015; Totsche et al. 2000). Thus, high F- ions are highly expected in the areas, where F-bearing minerals are abundant in the rocks. **[6,7]**

Fluorosis is an extensive health problem worldwide, which is affecting millions of people in many areas of the world, for example, India **[8-12]**, Pakistan **[13,14]**, East Africa **[15-17]**, Turkey **[18,19]**, southeastern Korea **[20],** northern China **[21,22]**, central region of Argentina **[23]**, Palestine **[24]** and in Yemen. **[25-27]**

The maximum permissible limit value of fluoride concentration in drinking water is 1.5 mg/l **[28]**, the guiding value of fluoride may be changed based on some factors like humidity, temperature, the volume of water intake fluoride from other sources, etc for different regions of the world. The Yemeni Standard specifies the desirable and permissible limits for fluoride in drinking water as 1.0 and 1.5 mg/l, respectively.

Fluorosis, especially the skeletal one is historically unknown in Yemen. It is known only about 8 to 10 years reported back. Clinically, it's developed due to the concentration of fluoride in bones. However, dental fluorosis is not new in Yemen, especially in Taiz governorate.

The main objective of this paper is to understand the control of geochemical processes on the F enrichment in groundwater in Al-Dhala, Yemen and its relationship with other major element concentrations and its health implications. The primary purpose of this paper is to obtain a better understanding of the factors controlling the high levels of F in groundwater samples.

2 Location

Al-Dhala Basin located in the south-west part of Al-Dhala governorate and extends into three districts; they are Al-Dhala, Qa'tabah, and Al-Husha districts and spread over an area of about 298 km^2. The main source of the basin is the drainage system of upper Wadi Tuban where the mean stream of the basin has its origin in the hilly regions at Ibb Governorate. The basin streams flow from north-west to low-lying alluvial plains in the downstream part of Al-Dhala, basin beyond Qarad Village (Fig.1).

3 Geology and hydrogeology

The oldest rocks, recognized in Al-Dhala basin are from Cretaceous age known as sandstone of Tawilah Group. The sandstone of Tawilah Group outcrops in the central part of the basin in small portion, in the west; elsewhere due to block faulting it has gone down and is covered by Tertiary volcanic (Fig. 1). Tertiary volcanic rocks (TV) comprising, Basalt, Rhyolite, Dacite, Ignimbrite, and Basaltic Pyroclastic deposits covers a major part of the basin border (Fig.1). Younger acidic or basic intrusives, as well as, dikes or sills, occur in the area, such as granitic rocks which exposed in the southeastern part of the basin. It is found between Wadi Siwat and Wadi Matar to the NW of Ad Dhala city**[29]**. Most of the area of the basin is covered by Quaternary deposits (Fig.1). It consists of sands and gravels of various grain sizes, the coarse-grained alluvium occurs in Wadi beds at the foot of the hills, the sands, silt and clay material occur in the wide Wadis in the study area. These deposits are derived from the surrounding volcanic rocks. The thickness in the upstream of Wadis does not exceed 5 m and may reach 10 m in the down streams of the Wadis. **[30]**

Hydrogeologically there are two aquifers, Shallow or Upper Aquifer and Deeper Aquifer. The upper aquifer comprises of Quaternary alluvial (QA) deposits or upper fracture volcanic (TV) or Tawilah sandstone or combination of either QA or TV or QA and sandstone. The Deeper aquifer comprises of Tertiary volcanic or Tawilah sandstone or combination of the two. **[30]**

Quaternary alluvial constitute the shallow aquifer in a major part of the basin. The width ranges from 5m to a few hundred meters and thickness is typically less than 20m and transmissivity ranges from 8-800 m2/day **[30]**. These deposits have extremely favorable recharge from Wadi floods. However, after the rainy season these drain quickly. Deeper aquifer receives recharge directly from the rain and surface runoff in areas where they outcrop especially in the high elevated area of the catchment area. The specific capacity of some wells has been evaluated by NWSA/Al-Dhala. It ranges between 0.06 to 3.5 m3/hr/m for TV aquifer and between 0.42 to 6 m3/hr/m for Tawilah sandstone aquifer. The Upper and Deeper aquifers are hydraulically interconnected. **[30]**

4 Materials and methods

A total of 28 representative groundwater samples were collected from all parts of Al-Dhala, basin so that full geographic representation has been made for the distribution of fluoride ion in the groundwater of the study area. Temperature, pH, and conductivity were measured at the sampling sites. Exact sampling locations were marked with the help of GPS and the coordinates were plotted on the map. Groundwater samples were analyzed for conventional water quality parameters according to APHA methods **[31]** in the laboratory of Local Corporation for Water Supply and Sanitation, Aden Governorate. The analytical precision for the measurements of cations (Ca, Mg, Na, and K) and anions (HCO_3, Cl, SO_4, NO_3, and F), indicated by the ionic balance error (IBE) was computed on the basis of ions expressed in meq/l. The value of IBE was observed to be within a limit of ±5%. **[32,33]**

5 Results and Discussion

5.1 Hydrochemistry of water

The results of the chemical analysis data of groundwaters from Al-Dhala, basin is presented in Table 1. The pH of groundwater varied from 6.61 to 8.60 with a mean of 7.37, indicating alkaline groundwater in nature. Concentration of TDS, a measure of quality, ranged from 529 to 2560 mg/l with a mean of 1379.5 mg/l. According to the TDS classification (Todd, 1985), 75 % of the samples of groundwater belonged to the brackish type (TDS> 1000 mg/l). Among the cations, the concentrations of Ca, Mg and Na ions ranged from 12 to 224, 2 to 204 and 145 to 1352 mg/l with a mean of 74.94, 32.64 and 451.14 mg/l, respectively. The major ion chemistry data revealed that Na is the most predominant cationic constituents followed by Ca and Mg. The dissolved anions of SO_4, Cl, HCO_3 and NO_3 ions ranged from 75 to 2130, 50 to 1400, 73 to 1098 and 2 to 132 mg/l with a mean of 480, 392, 472 and 15 mg/l respectively. For the major anions (SO_4, Cl, HCO_3, and NO_3), the sulphate and chloride are found to be the most predominant anions followed by bicarbonate and nitrate.

Table 1: Chemical analysis of water samples from Al-Dhala Basin

Sample NO.	pH	TDS mg/l	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	NO3 mg/l	SO4 mg/l	HCO3 mg/l	F mg/l
1	7.5	1281	34	3.6	412	16	195	4.4	260	644	2.57
2	8.2	1160	20.8	2.9	386	11	176	4	250	561	2.57
3	8.2	1000	19	4	354	9	186	6.2	180	549	2.47
4	8.27	1096	63	3.5	380	9	169	3.5	270	581	2.59
5	7.9	998	16	2.9	346	8	232	4.8	270	342	2.48
6	7.72	789	12	7	514	8	135	2	402	671	17.6
7	7.25	832	16	7	389	6	225	3	313	427	18
8	7.32	1335	38	5	497	9	217	6	656	506	15.3
9	7.51	1044	26	3	376	14	50	4	280	725	18.3
10	7.05	1805	144	55	278	16	585	22	240	427	3.8
11	7.8	1801	152	88	1352	22	1400	20	1450	927	1.5
12	7.5	2280	224	204	1089	18	1140	132	2130	537	1
13	8	1002	25	7	245	6	210	13	375	73	0.31
14	8.15	2500	59	60	405	9	523	22	235	272	8
15	7.4	1050	36	7	234	12	280	7	112	170	0.34
16	8	1456	51	15	180	9	175	12	75	252	4.85
17	8.6	1520	30	15	402	14	305	5	300	449	4.2
18	7.8	2370	89	8	478	21	680	45	488	98	0.59
19	7.3	1287	38	4	612	12	490	14	688	85	0.32
20	8	1069	25.6	15	178	18	70	6	110	385	1.24
21	8.33	837	152	2	145	21	152	5	86	520	0.76
22	7.33	940	132	88	312	17	315	49	350	885	0.6
23	7.42	1071	40	22	344	7	260	3	400	366	16.4
24	6.89	1162	96	44	611	11	800	2	340	573	14
25	6.61	2040	192	126	277	12	100	19	1200	415	14
26	7.8	2560	120	41	1065	16	900	4	1080	1098	11
27	7.11	1812	172	57	388	9	450	5	590	598	2.4
28	6.88	529	76	17	383	11	550	8	300	73	1.6
Min	**6.61**	**529.00**	**12.00**	**2.00**	**145.00**	**6.00**	**50.00**	**2.00**	**75.00**	**73.00**	**0.31**
Max	**8.60**	**2560.00**	**224.00**	**204.00**	**1352.00**	**22.00**	**1400.00**	**132.00**	**2130.00**	**1098.00**	**18.30**
Average	**7.64**	**1379.50**	**74.94**	**32.64**	**451.14**	**12.54**	**391.79**	**15.39**	**479.64**	**471.75**	**6.03**

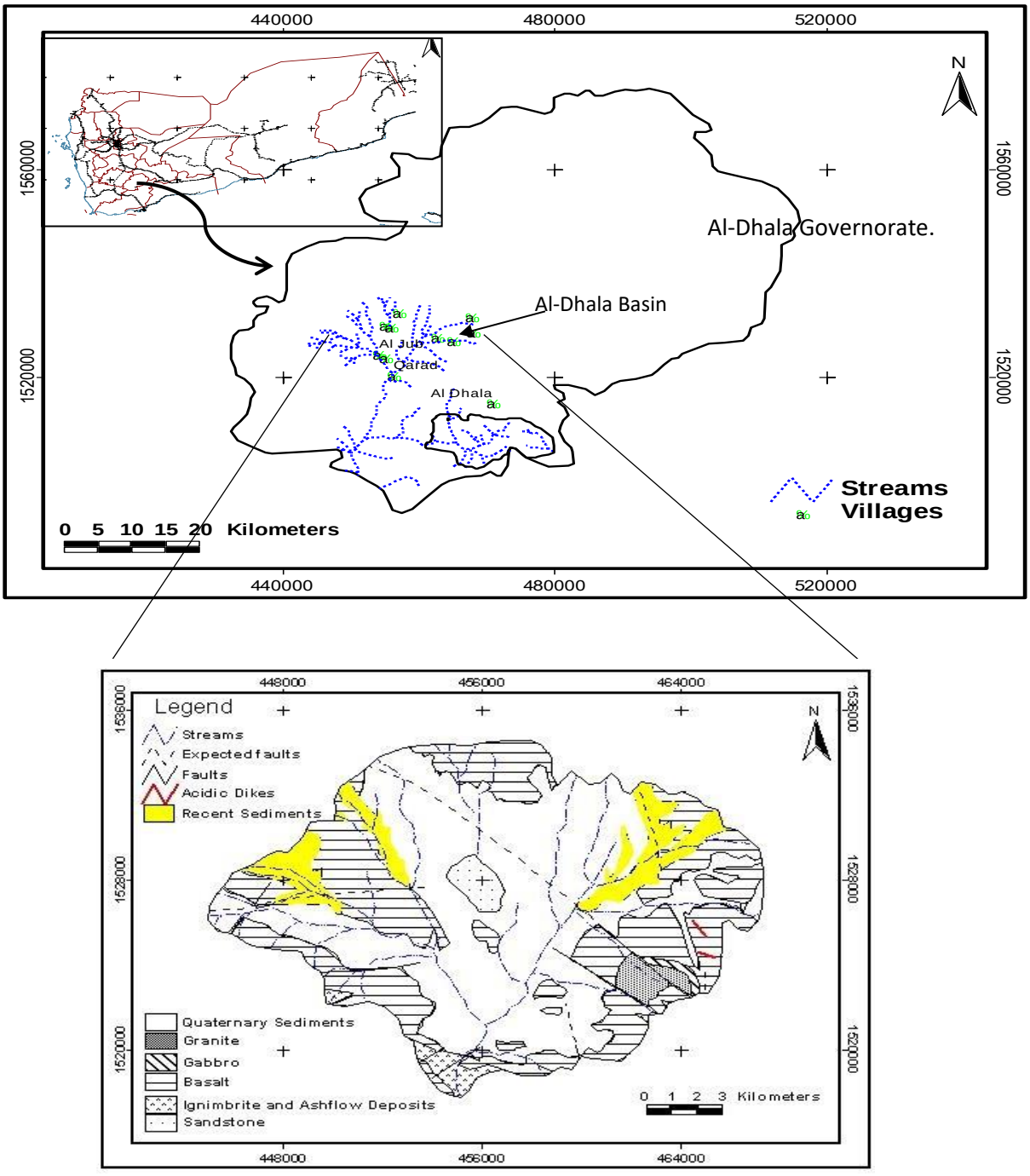

Figure1: Location & Geological map of Al-Dhala Basin (modified after Rebertson Group, (1990))

5.2 Hydrofacies

The hydrochemical facies analysis reflects the chemical processes operative in a certain lithological environment and under specific geochemical conditions. The general classification and trend of variation in the groundwater of the study area is exhibited in the Piper-diagram (Fig.2). In the cation triangle, all samples are Na-type except for one sample. The high Na in the groundwater may be related to the cation exchange operative in the aquifers **[22]**. The assumption also signifies from high Na/Ca ratios (12.6), which showed ion exchange between Na absorbed on the surface of clay minerals and Ca in the groundwater. **[34]**

In addition, diamond fields are classified into six types: 1= NaCl Type 2= $CaHCO_3$ Type 3= $NaCaHCO_3$ Type 4=

CaMgCl Type 5= CaCl Type 6= NaHCO3 Type (Fig. 3). NaCl type is found to be the dominant water type in the study area.

Sodium groundwater type is found supporting the dissolution of fluoride. Similar results have been found in the regions of Central Telangana, India, where sodium is high, fluoride is also high. **[35]**

5.3 Geochemical modeling

The method of speciation modeling has been used to study the chemical equilibrium occurring in groundwater and to identify the source of a high concentration of fluoride in groundwater from the study area. Saturation indices (SI) for minerals are the most important results of the speciation calculations which indicate whether a mineral

will dissolve or precipitate. In the view that calcium, fluoride, and carbonate activities are interdependent, the solubility limits for fluorite and calcite provide natural control over water composition **[36]**. The saturation indices (SI) of fluorite (CaF_2) and calcite ($CaCO_3$) in the groundwater samples were calculated using PHREEQC Interactive, a computer program of U.S. Geological Survey, version 2.8 (2003) (Table No.2) and are plotted in Fig.3, which shows that most of the samples are oversaturated with respect to calcite whereas, majority of samples have been found undersaturated with respect to fluorite. This situation of solubility control on the higher concentration of fluoride can be explained by the fact that fluoride ions in groundwater can be increased as a result of precipitation of $CaCO_3$ at high pH, which removes Ca^{2+} from solution allowing more fluorite to dissolve (Kumar et al., 2017). These released Ca^{2+} ions combine with CO_3^{2-} ions to further enhance the precipitation of $CaCO_3$. Thus, fluorite undersaturation 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 thereby increasing the F/ Ca ratio of solution.

Mattash **[29]** concluded that the fluorite and calcite minerals are wide distribute in the form of veins, particularly in association with quartz-carbonate veins or dikes as a result of hydrothermal process. Their main occurrences are found in Al-Dhala area and its vicinity. Therefore, it can be concluded that the calcite and fluorite which are widely distributed in the area are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater of Al-Dhala, Basin.

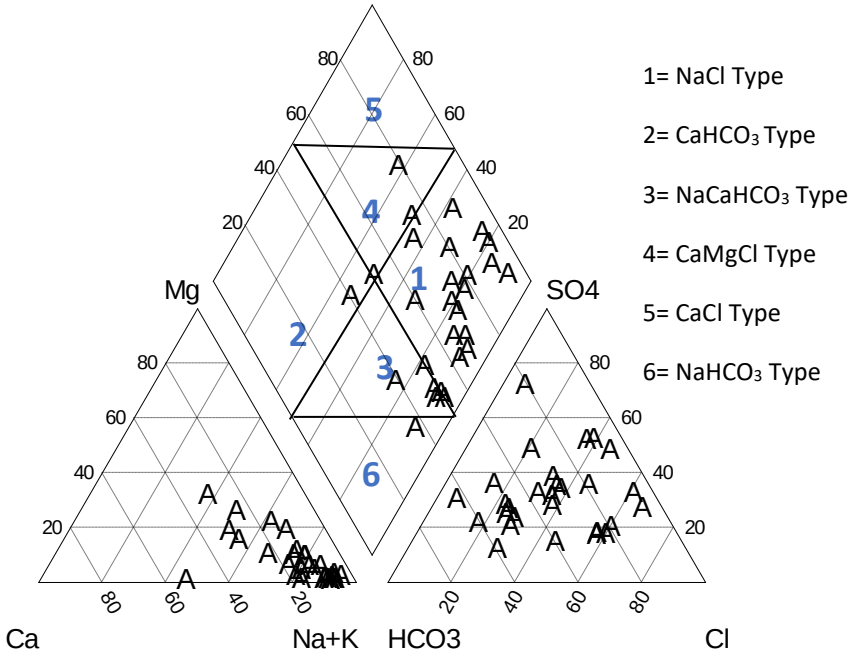

Figure2: Piper's Diagram of water samples from Al-Dhala, Basin

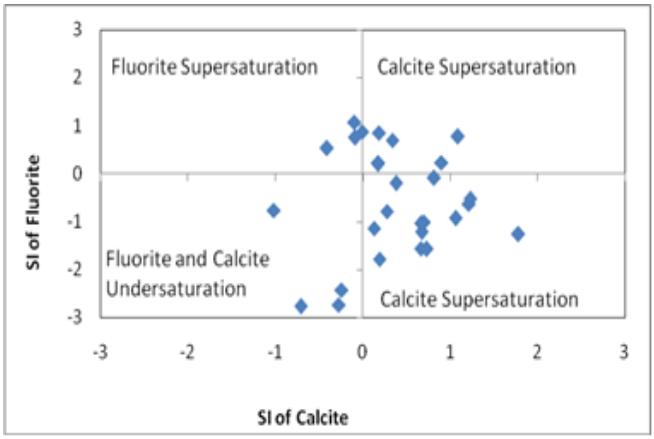

Figure3: Plot of calcite saturation index versus fluorite saturation index

6 Drinking Water Sources

The quality of water is very important to mankind because it has a direct link with human welfare. With the growth in the human population, there is an increase in urbanization too. This is not only putting pressure on the quantitative aspects but even in the quality of characteristics. The area under study is characterized by low and irregular rainfall, high temperature, high evaporation and eminent drought periods. In addition to that, human activities, such as overuse of the groundwater, have led to serious consequences, for example, intense mineralization of downstream waters, lowering of the regional water table and salinization. Therefore, it is essential to look into the quality aspect of groundwater.

Table.2: The calculated saturation indices (SI) of fluorite (CaF2) and calcite ($CaCO_3$) in the groundwater samples from Al-Dhala Basin

Sample No.	Sum of Anions	Sum of Cations	SI (Calcite)	SI (Fluorite)
1	21.66305	20.32317	0.2802	-0.7829
2	19.55338	18.34803	0.7013	-1.008
3	18.20339	16.90565	0.6665	-1.0431
4	20.09502	20.19115	1.2329	-0.5264
5	17.96598	16.29186	0.1326	-1.1445
6	24.13183	23.73723	0.1767	0.2281
7	20.85126	18.44848	-0.4085	0.5265
8	28.9625	24.15624	-0.0845	0.7475
9	20.14009	18.25751	0.3399	0.695
10	28.98094	24.21336	0.1749	0.1946
11	85.22536	74.19797	1.0622	-0.9255
12	87.07617	75.79414	0.6762	-1.2132
13	15.11524	12.63395	-0.2702	-2.7332
14	24.80877	25.72824	0.9036	0.2346
15	13.12492	12.85787	-0.2413	-2.4216
16	11.03776	11.83911	0.8147	-0.0908
17	22.49677	20.5755	1.2139	-0.6309
18	31.5612	26.4286	0.1938	-1.7811
19	29.74394	29.15291	-0.7071	-2.7645
20	10.71802	10.71478	0.6646	-1.5667
21	14.70537	14.59405	1.7824	-1.2622
22	31.34062	27.83452	0.7345	-1.5756
23	22.56705	18.94865	-0.0046	0.8732
24	39.80195	35.26969	0.1909	0.8539
25	35.60251	32.30531	-0.0967	1.0706
26	66.51115	56.09629	1.0902	0.771
27	34.97559	30.38092	0.3816	-0.2049
28	23.14679	22.13242	-1.0127	-0.7667

Table 3: Classification of groundwater samples from Al-Dhala Basin according to the concentration of fluoride and associated risk

Classification	F Conc. ranges (mg/l)	Associated risk	No. of samples	Percentage (%)
Safe Areas	0-1.5	Within WHO permissible limits	8	28.57
Low risk areas	1.5-3	Dental fluorosis	8	28.57
High risk areas	3-5	Dental and mild skeletal fluorosis	3	10.71
Very high-risk areas	> 5	Severe skeletal fluorosis	9	32.14
Total			28	100

For drinking purposes of water, certain minimum quality parameters requirements have been suggested by the World Health Organization **[28]**. All the 28 samples analyzed from Al-Dhala, Basin are used for drinking and domestic uses except samples no. 8,9,11 and 23 (Table No.1). The minimum and maximum values of groundwater samples from Al-Dhala, Basin have also been given. It is evident from these values that major ions are far beyond the permissible limits for the majority of the samples.

Out of 24 groundwater samples (using for drinking purposes), 17 samples have shown F- concentration values above the maximum permissible limit of 1.5 mg/l. Obviously, 71% of the water used for drinking purposes is contaminated by fluoride ion concentration in Al-Dhala, Basin.

The population living in these areas is thus susceptible to higher dental and chronic skeletal fluorosis. The highest concentrations were found to be 18.3 mg/l from Albaidhanyah, 18 mg/l from Habeel Altanami, 17.6 mg/l from Al Hada`a, 16.4 mg/l from Musadah and 14 mg/l from Al-Jub villages.

For the sake of convenience in description, groundwater samples have been grouped into four categories according to their concentration of F- and associated risk to the human population. A total of 28.57 % (n = 8) groundwater samples were found to be within prescribed WHO limits (0.0–1.5 mg/l), 28.57% (n = 8) within 1.5–3.0 mg/l, whereas, 10.71% (n = 3) within 3.0– 5.0 mg/l and 32.14% (n = 9) above 5.00 mg/l categories, respectively (Table 3).

Conclusions

This study indicated that the groundwater in Al-Dhala basin has high fluoride content. 71 percent of the groundwater samples tested were found to have fluoride concentrations above the maximum human consumption limit recommended by the WHO. The major ion chemistry data revealed that Na is the most predominant cationic constituents followed by Ca^{2+} and Mg^{2+}. The SO^{4-} and Cl^{-} are found to be the most predominant anions followed by HCO_3^{-} and NO_3^{-}. The geochemical modeling indicates that calcite and fluorite are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater of Al-Dhala, region. This situation of solubility control on the higher concentration of fluoride can be explained by the fact that fluoride ions in groundwater can be increased as a result of precipitation of $CaCO_3$ at high pH, which removes Ca^{2+} from solution allowing more fluorite to dissolve. These have been found through the present study in this particular area, which is supported, by the correlation study of fluoride with other constituents and solubility equilibrium diagram between calcite and fluorite. The hydrofacies analysis of major cations from the Piper diagram clearly indicates Na-type while the anions are mainly Cl-type with HCO_3 and SO_4.

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مقالة بحثية

التوصيف الهيدروجيوكيميائي والتأثيرالبيئي للتلوث بالفلورايد في المياه الجوفية في حوض الضالع، اليمن

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الملخص

تعتبر المياه الجوفية في حوض الضالع، المصدر الرئيسي والوحيد للمياه، كما أن نوعيتها مهمة كونها تحدد مدى ملائمة المياه الجوفية لأغراض الشرب أو الاستخدامات المنزلية الأخرى. لقد كان الهدف الرئيسي من هذه الدراسة، الحصول على فهم أفضل العوامل التي تتحكم في التركيزات العالية لأيون الفلوريد في عينات المياه الجوفية. لذا تم جمع عينات من المياه الجوفية من 28 بئراً وتم تحديد المناطق ذات التركيزات العالية من الفلوريد، واستخدمت البيانات الكيميائية في توضيح الأسباب المحتملة للتباين في تركيزات الفلوريد. أشار المسح الهيدروجيوكيميائي في المنطقة إلى أن التفاعل بين الماء والصخور ربما يكون السبب الرئيسي لتركيز الأيونات في المياه الجوفية. كما أوضحت النمذجة الجيوكيميائية إلى أن الكالسيت والفلوريت هما المعدنان الرئيسيان المتحكمان في هيدروجيوكيميائية التلوث الأيوني المرتفع للفلوريد في المياه الجوفية لحوض الضالع. بينت الدراسة إن مجموع ما نسبته 71٪ من مياه الآبار التي تم جمعها تستخدم لأغراض الشرب، لذا لوحظ إن الآثار الصحية لتركيزات الفلوريد العالية في حوض الضالع متمثلة في أمراض تفلور الأسنان بشكل شائع جدًا، وتفلور العظام بشكل محدود في بسبب الإفراط في تناول الفلوريد، خاصةً في ظل غياب التوعية.

الكلمات الرئيسية: هيدروجيوكيميائي, فلورايد, المياه الجوفية, حوض الضالع, الضالع, اليمن.