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GEOLOGY

Hydrogeochemistry and origin of fluoride in groundwater of Hidhran & Alburayhi Basin, northwest Taiz City, Yemen

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ABSTRACT: Thirty-one groundwater samples were collected in the Hidhran and Alburayhi basin, northwest of Taiz city, from Tertiary age aquifers. A hydrogeochemical investigation was conducted in the study area. The fluoride ions along with other chemical parameters were analyzed to determine the hydrochemical characteristics, water types and evaluate the main source of fluoride in groundwater of the aquifer.

Fluoride ion concentrations in the groundwater in excess of WHO recommended limit of 1.5 mg/l for drinking water occurs throughout the area. Of the 31 well samples to be contained with fluoride concentration in excess of the recommended limit, the hydrogeochemical investigation indicates that water-rock interaction is probably the main reason for the high concentration of ions in the groundwater. The saturation indices (SI) of fluorite (CaF2) and calcite (CaCO3) in the groundwater samples showed that the majority of the samples are oversaturated with respect to calcite; and undersaturated with respect to fluorite.

The population of the study area is at a high risk due to excessive fluoride intake, especially when they are unaware of the amount of fluoride being ingested.

Key Words: Hydrogeochemistry, Fluoride, Groundwater, Hidhran and Alburayhi Basin, Taiz city, Yemen.

INTRODUCTION: Fluorine is the most electronegative and reactive of all elements, and is present as fluoride (F-) ions in drinking water. It occurs as Fluoride ion naturally in soils and natural waters due to chemical weathering of some F- - containing minerals (Totsche et al. 2000). 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), Turkey (Oruc 2003), India (Subba Rao and Devadas 2003; Gupta et al. 2005, Jacks et al. 2005), southeastern Korea (Kim and Jeong 2005) northern China (Guo et al. 2006) and recently in Yemen (Al-Amry 2009). Fluoride is commonly associated with volcanic or fumarolic gases, and in some areas, these may be important sources of fluoride for natural water (Hem, 1985).

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). The Yemeni Standard specifies the desirable and permissible limits for fluoride in drinking water as 1.0 and 1.5 mg/l, respectively.

Location:

Hidhran & Alburayhi Basin located north west Taiz city within the lower part of upper Wadi Rasyan, where the downstream sectors of of all other subcatchments converge (Fig.1). Morphologically, Hidhran & Alburayhi Basin is a flat or slightly undulating terrain located at altitude from 1400 to 800 m. Average rainfall is the lowest of the whole upper Wadi Rasyan and likely dose not exceed 400 mm/year (NWRA 2008).

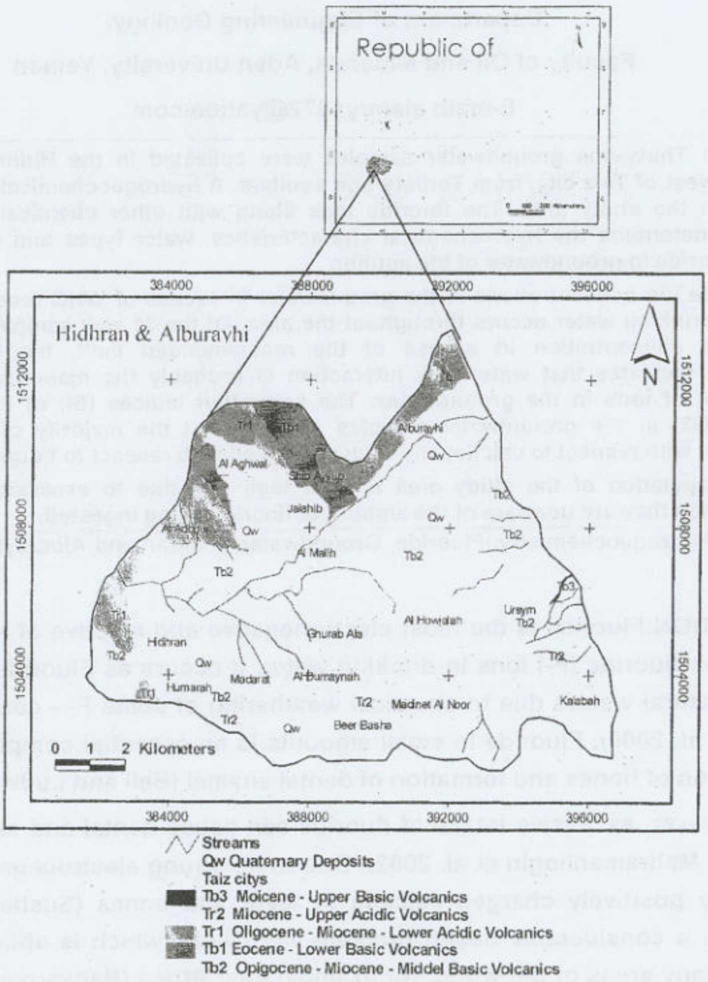

Fig.1: Location and geology map of Hidhran & Alburayhi Basin (modified after Rebertson Group, 1990)

Geology:

The geological map of Hidhran & Alburayhi Basin is given in Fig.1. it can be seen from the geological map that, the older stratigraphic unit exposed in the area is the lower basalt volcaniclastics (Tb1) which composed of greenish, fine-grained flood basalts and consist of horizons of compact fine grained basaltic volcanics (Alkali basalt, basanite, olivne nephelinite) interbeded with grey ash and some rhyolithic layers. The middle acidic volcanics (Tr2) are the most predominant volcanic units in the study area. This unit consists of fine grained flood basalt layers intercalated with grey ash having thickness in the range of tenths of meters. It partly overlies the (Tr1) or cut as

dykes through the older rocks. The middle acidic volcanics (Tr2) exposed in the south of study area surrounding Taiz city. This sequence consists of red coloured ignimbrite intercalated with black obsidian and some layers of green ash with total thickness of about 500 m. It has only limited distribution, exposed as small separated hills surrounding Taiz city and adjacent to the northern and southern slopes of J.Sabir. The upper basic volcanics (Tb3) outcrops as small hills in Al Hawban area to the south of Taiz-Sana road followed by the last rhyolitic eruptions of white or pinkish ignimbrite. The volcanic activity in the late Tertiary (Miocene to Pliocene Time) led to the eruptions of the upper basic volcanics (Tb3) which composed of compact basaltic lava flows intercalated with red and white ash layer.

Quaternary deposits (QW) consist of sands and gravels of various grain sizes. These deposits are derived from the surrounding volcanic rocks the thickness in the upstream of wadis does not exceed 10 m and may reach 60 m in the down streams of the wadis.

Hydrogeology:

Hidhran & Alburayhi Basin carachtrised by two types of aquifers:

1- Qaternary deposits aquifer: Consists of alluvoium of different grain sizes which ranges from silt to boulders and forms potential aquifer in the valley floor areas. The total thickness of this aquifer ranges from 10 to 35 m and depth to water table ranges from 5 to 30 m (NWRA 2008). The hydraulic conductivity and transmissivity of this aquifer ranges from 199 to 0.3 m/d and 596 to 9 m2/d respectively. According to NWRA/Taiz report (2008), this aquifer conceder to be poluted and not suitable for human consumption.

2- Fractured Volcanics Aquifer: it consists of tertiary volcanics, where the weathering of basaltic flows has helped in development of weathered basaltic aquifers in the study area. According to NWRA (2008) study, the depth to watre table ranges from 50 to 300 meter, the yeld of this aquifer about 5 l/s and transmissivity 7.2 m2/d.

Objectives and Methodology:

The main objectives of this study are to determine the hydrochemical characteristics, water types and evaluate the main source of fluoride in groundwater of the aquifers in the Hidhran and Alburayhi basin.

A total of 31 representative groundwater samples were collected (summer, 2008) and chemically analyzed from all parts of Hidhran & Alburayhi Basin. The sample localities were selected to represent the study area (Fig. 4). The temperature, pH, and electrical conductivity were measured in-situ utilizing portable meters. Exact sampling locations were marked with the help of GPS and the coordinates of the topographic map. The major anions (HCO3, Cl, SO4 and NO3) and major cations (Ca, Mg, Na, and K) in addition to fluoride were measured according to APHA methods (APHA, 1989). The analytical precision for the measurements of cations and anions 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% (Mandel and Shiftan 1980; Domenico and Schwartz 1990).

Results and discussion

Hydrochemistry of water:

The results of hydrochemical analysis data of groundwater samples collected from Hidhran & Alburayhi Basin in the summer of 2008 are given in Table 1. The pH of groundwater varied from 6.94 to 8.12 with a mean of 7.43, indicating alkaline groundwater in nature. The TDS content ranged from 1023 at well 25 to 5683 mg/l at well 8. All groundwater samples are brackish with dissolved concentrations grater than 1000 mg/l. Among the cations, the concentrations of Ca, Mg and Na ions ranged from 40 to 450, 18 to 360 and 131 to 1254 mg/l with a mean of 179, 154 and 524 mg/l, respectively. The major ion chemistry data revealed that Na and Ca are the most predominant cationic constituents followed by Mg. The dissolved anions of SO4, Cl, HCO3 and NO3 ions ranged from 96 to 480, 160 to 2034, 414 to 1525 and 0 to 33 mg/l with a mean of 236, 881, 913 and 10 mg/l respectively. The sequence of both anions and catins in the investigated water samples has the following order:

SO4 › Cl › HCO3 › NO3 and Na › Ca › Mg

Based on Piper diamond diagram (Piper, 1953) shown in Fig.2, the following groundwater groups could be differentiated into three groups:

The first group (I) occupies the middle lower part of the diamond shape, where water has sodium bicarbonate water type.

The second group (II) occupies the upper right side of the diamond shape. Its chemical properties are sodium-chloride water type prevails.

The third group (III) is characterized by advanced mineralization with magnesium-sulphate water type.

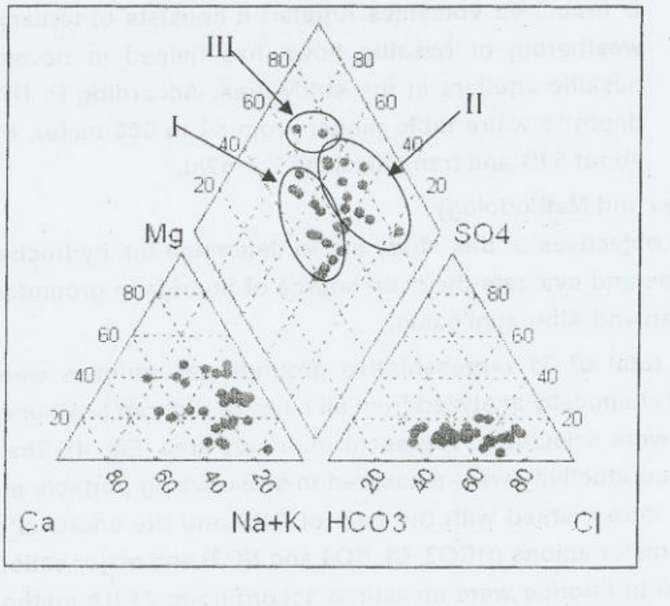

Fig.2: Piper's Diagram (1953) of water samples from Hidhran & Al Burayhi Basin

Geochemical modeling:

The chemistry of groundwater is the result of interaction between rain and the rock near the earth's surface. In order to study the chemical equilibrium existing in groundwater and identify the source of high fluoride concentration in groundwater from the study area, the concept of speciation modeling has been used. The most important results of speciation calculations are saturation indices (SI) for minerals, which indicate whether a mineral should dissolve or precipitate.

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). The saturation indices (SI) of fluorite (CaF2) and calcite (CaCO3) in the groundwater samples were calculated using PHREEQC Interactive, a computer programme of U.S. Geological Survey, version 2.8 (2003). Results are shown in Table 2 and plotted in Fig.3. The aquifer is saturated with respect to calcite and undersaturated for most water samples 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 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 udersaturation 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. Hence calcite and fluorite are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater of Hidhran & Alburayhi Basin.

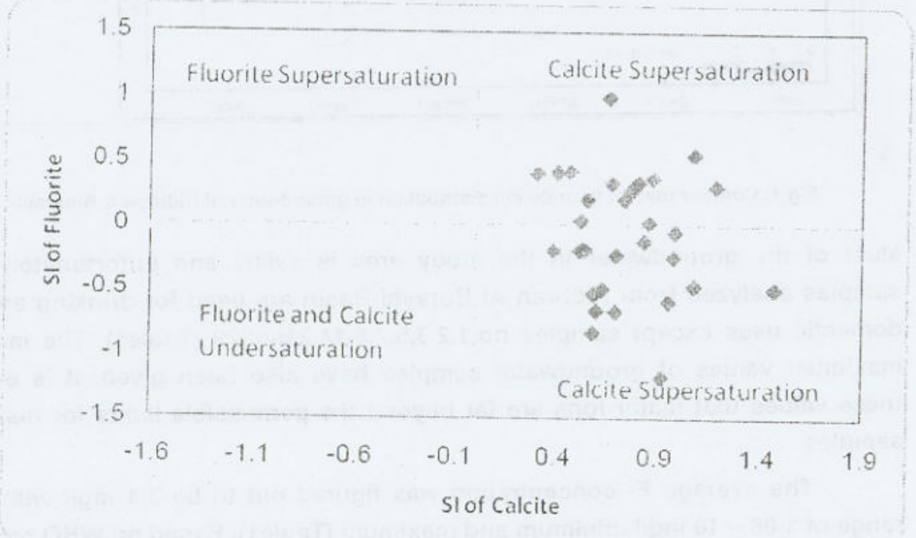

Fig.3: Plot of calcite saturation index versus fluorite saturation index from Hidhran & Al Burayhi Basin samples

Special Variation in the Fluoride Concentration:

The special variation in the fluoride ion concentration in the groundwater from Hidhran & Alburayhi Basin was evaluated. The special variation contour map in F^- from water samples of the study area is given in Fig.4. Generally, most of the water samples showed enhanced concentrations with general increase trends to the northwest spots of Hidhran & Alburayhi Basin area. The largest concentrations were observed southwest Shib Ayyob and Al Adhnur villages with concentrations greater than 5 mg/l. All the groundwater samples collected from Al Adhmur and Al Mirfid villages were found severely contaminated by the presence of fluoride ion. However, the situation is more aggravated around the Ghurab Al Asfaal locality (Fig. 4).

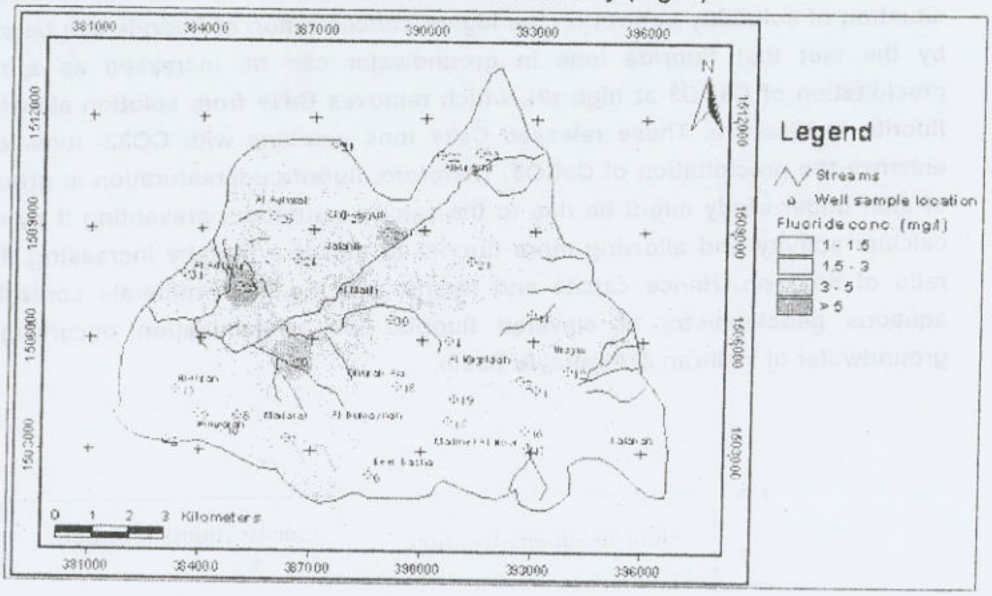

Fig.4: Contour map of fluoride ion distribution in groundwater of Hidhran & Alburayhi Basin

Must of the groundwater in the study area is saline and unfortunately all the 31 samples analyzed from Hidhran Al Burayhi Basin are used for drinking and the other domestic uses except samples no.1,2,3,6,7,8,14,21and29 (Table1). The minimum and maximum values of groundwater samples have also been given. It is evident from these values that major ions are far beyond the permissible limits for majority of the samples.

The average F- concentration was figured out to be 3.4 mg/l within the wide range of 1.08 – 10 mg/l minimum and maximum (Table1). Based on WHO recommended guidelines for fluoride in drinking water, 83.9 % (n = 26) out of total 31 groundwater samples were above the optimum level of 1.5 mg/l. 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 Ghurab Al Asfaal, 5.8 mg/l from Al Adhmur village and 5.4 mg/l in groundwater samples collected from Al Gail village.

For convenience in description, groundwater samples have been grouped into four categories according to their concentration of F- and associated risk to human population (Table 3). A total of 16.12% (n = 5) groundwater samples were found to be

within prescribed WHO limits (0.0–1.5 mg/l), 32.27% (n = 10) within 1.5–3.0 mg/l whereas, 35.48 % (n=11) within 3-5 mg/l and 16.13 % (n = 5) above 5 mg/l (Table 3).

In order to understand the vertical distribution of the fluoride ion concentration if any, the type of the sample water (Dug, Bore) evaluated separately. The following histogram represents the maximum, minimum and average of F- concentration for each well type form Hidhran & Alburayhi Basin (Fig.5).

The high average F- concentration values observed in the dug wells higher than the bore wells from the study area. It can be concluded that, the shallow aquifers reflects higher fluoride contamination than the deeper aquifers.

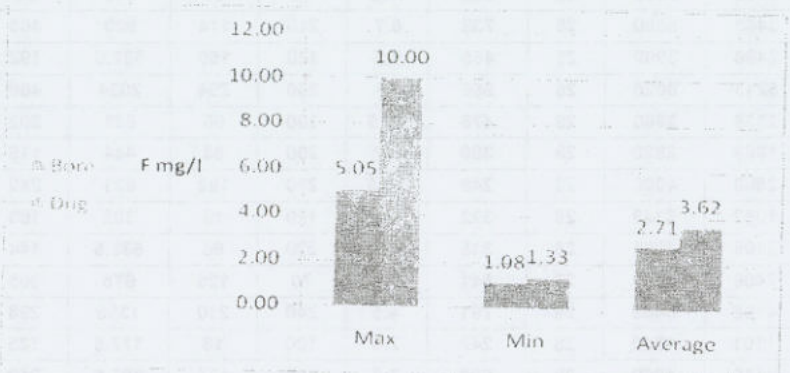

Fig.5: Variations of F- concentrations of Dug and Bore wells from Hidhran & Al Burayhi Basin

Conclusions:

The dominant cations from the study area are sodium and calcium, while the bicarbonate and chloride are the dominant anions. The dominant hydrogeochemical facies studies concluded that the high Na in the groundwater may be related to the plagioclase which is the main constituent of the basalts and which can release Na+ into groundwater.

The geochemical modeling indicates that all of the samples from the study area are oversaturated with respect to calcite whereas; all 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.

The calcite and fluorite are the main minerals controlling the aqueous geochemistry of elevated fluoride ion contamination occurring in the groundwater.

It's concluded that, the shallow aquifers reflect higher fluoride contamination than the deeper aquifers from Hidhran & Al Burayhi Basin.

Dental fluorosis are the widely fluoride disease observed in the affected areas.

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Sample No.	pH	TDS (mg/l)	E.C (µs/cm)	T (°C)	Na (mg/l)	K (mg/l)	Ca (mg/l)	Mg (mg/l)	Cl (mg/l)	SO4 (mg/l)	NO3 (mg/l)	HCO3 (mg/l)	F (mg/l)
1	7.5	3891	6080	28	766	19.07	140	240	1154	384	5.27	1220	4.008
2	7.1	4371	6830	28	596	15.13	240	360	1456	326	15.128	1220	4.864
3	7.8	3821	5970	28	644	24.02	340	168	1456	96	8.06	1005	1.33
4	7.1	3334	5209	28	435	8.03	450	126	1154	206	8.8	915	3
5	7.3	2605	4070	28	161	5.8	360	198	888	120	6.944	854	1.672
6	7.4	4032	6300	28	407	10	320	348	1456	317	16.12	915	2.66
7	7.4	4339	6780	28	683	5	200	336	1598	355	8.74	915	4.921
8	7.3	5683	8880	28	1254	28	330	204	1953	384	33.11	1525	5.05
9	7.6	1203	1880	28	143	3.28	110	84	266	101	5.33	549	1.425
10	7.3	1242	1941	28	131	8.03	180	54	266	106	8.06	579.5	1.33
11	8.12	1472	2300	28	322	1.68	40	84	249	96	8.06	854	1.37
12	7.6	3443	5380	28	738	6.7	240	114	923	408	6.45	1159	3.819
13	7.3	2496	3900	28	455	8.5	130	150	532.5	192	9.92	1201.7	2.66
14	7.3	5213	8020	28	966	45	250	294	2034	480	21.95	732	10
15	7.05	2336	3650	28	476	10.5	200	66	621	202	9.92	884.5	2.66
16	7.1	1869	2920	28	380	6.7	200	30	444	115	3.16	866	1.083
17	7.13	2560	4000	28	248	27.5	250	192	621	240	7.75	1049.2	3.33
18	7.4	1382	2159	28	322	4.2	120	18	302	163	5.02	579.5	2.261
19	6.94	2106	3291	28	315	8.3	220	96	532.5	144	11.22	884.5	3.99
20	7.7	2406	3760	27	541	7.5	70	125	675	365	8.866	641	5.8
21	7.24	4102	6409	28	791	4.5	240	210	1385	298	10.7	1128.5	4.33
22	8	1101	1720	28	242	7.5	100	18	177.5	125	0.56	579.5	1.729
23	7.3	3116	4869	28	658	7.5	100	174	887.5	240	7.75	1098	3.325
24	7.6	3206	5009	28	690	3.28	100	180	886.5	264	6.63	1177	3.667
25	7.4	1023	1598	29	225	3.3	66	32	159.7	96	7	549	5.8
26	8	2451	3830	28	557	4.1	50	138	533	192	9.49	1159	2.793
27	7.9	1235	1930	28	214	0.33	40	96	213	144	12.1	610	1.995
28	7.5	2803	4380	28	465	3.3	70	240	710	216	9.5	1159	2.793
29	7	4922	7690	28	1001	11.7	210	271	1864	394	13.21	958	5.45
30	7.7	2701	4220	27	805	7.5	100	24	1030	298	0	414	2.81
31	7.4	2577	4000		628	5.6	100	90	781	365	13.45	610	3
Max	8.12	5683.00	8880.00	29.00	1254.00	45.00	450.00	360.00	2034.00	480.00	33.11	1525.00	10.00
Min	6.94	1023.00	1598.00	27.00	131.00	0.33	40.00	18.00	159.70	96.00	0.00	414.00	1.08
Average	7.43	2838.33	4429.83	27.97	516.43	9.75	180.87	150.67	868.47	234.93	9.77	892.36	3.36

Table 1: Chemical analysis of water samples from Hidhran & Al Burayhi Basin (Summer of 2008)

Sample No.	X	Y	F mg/l	Sum of Anions meq/l	Sum of Cations meq/l	SI (Calcite)	SI (Fluorite)
1	393166	1504660	3	39.96104	39.85588	0.388	-0.1968
2	392700	1504888	2.81	42.19401	42.17257	0.5319	-0.192
3	391314	1505502	5.45	76.94215	76.61985	0.4072	0.416
4	390731	1505969	2.793	43.79015	43.553	0.5879	-0.5373
5	383134	1503222	1.995	19.26532	19.21264	0.5832	-0.8412
6	388626	1502325	2.793	38.29745	38.18385	0.9514	-0.5942
7	386364	1503312	5.8	15.89709	15.79815	0.3057	0.4073
8	385038	1503944	3.667	50.07282	49.89928	0.8319	-0.1252
9	383961	1503951	3.325	48.30266	48.12156	0.5172	-0.1958
10	384599	1502996	1.729	17.20636	17.18969	1.0775	-0.4743
11	392990	1506693	4.33	64.13435	63.77874	0.8008	0.3376
12	393986	1505185	5.8	37.56781	37.50317	0.5529	0.2011
13	383367	1504621	3.99	32.8698	32.79223	0.4658	0.4205
14	383944	1503844	2.261	21.59492	21.58315	0.5429	-0.1922
15	390750	1503800	3.33	39.9861	39.76565	0.736	0.2033
16	392852	1501554	1.083	29.20981	29.14962	0.6026	-0.669
17	392934	1503091	2.66	36.48774	36.38493	0.5217	0.0364
18	389343	1504746	10	80.1766	79.83786	0.6537	0.997
19	390903	1504434	2.66	38.98166	38.8393	0.6951	-0.2311
20	386444	1505422	3.819	53.81306	53.63003	1.1816	0.309
21	391321	1508115	1.37	23.19495	22.95746	0.9203	-1.1823
22	390441	1511441	1.33	19.38195	19.32958	0.6268	-0.5024
23	391497	1511056	1.425	18.74793	18.7055	0.6926	-0.6754
24	386597	1508155	5.05	88.77073	88.51669	1.0711	0.5506
25	385027	1507397	4.921	67.83682	67.46592	0.7739	0.2843
26	385960	1509309	2.66	63.01542	62.56424	0.9795	-0.0512
27	387573	1511269	1.672	41.72055	41.40937	0.9743	-0.2469
28	387848	1509362	3	52.10859	51.95129	0.8745	0.374
29	389032	1508898	1.33	59.71292	59.41804	1.4702	-0.4948
30	389130	1506557	4.864	68.30481	67.91173	0.6718	0.3337
31	383572	1507772	4.008	60.82332	60.54231	0.8557	0.0227

Table 2: The calculated saturation indices (SD) of fluorite (CaF2) and calcite (CaCO3) in the groundwater samples from Hidhran & Burayhi Basin

Table 3: Classification of groundwater samples from Hidhran & Alburayhi 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	5 (3,9,10,11,16)	16.12
Low risk areas	1.5 – 3	Dental fluorosis	10 (5,6,13,15,18, 22,26,27,28,30)	32.27
High risk areas	3 – 5	Dental and mild skeletal fluorosis	11(1,2,4,7,12,17,19, 21,23,24,31)	35.48
Very high risk areas	› 5	Severe skeletal fluorosis	5 (8,14,20,26,29)	16.13
Total			31	100.00

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هيدروجيوكيميائيه وأصل تركيزات أيون الفلوريد فى المياه الجوفيه لحوض حيدران-البريهي - شمال غرب مدينه تعز، اليمن

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تشمل هذه الدراسه جمع31 عينه من المياه الجوفيه من ابار مختلفه من حوض حيدران-البريهى الواقع فى الجزء الشمالى الغربى لمدينه تعز غ\فى اليمن، وذلك لتقييم الخواص الهيدروجيوكيميائيه والتعرف على مصدر التركيزات العاليه لايونات الفلوريد فى المياه الجوفيه، والذى يغطى بصخور بركانيات العصر الثلاثى. اجريت الدراسات الهيدروجيوكيميائيه على العينات المائيه وقد تم تحليل ايونات الفلوريد مع العناصر الكيميائيه الاخرى واستخدام الرسم التخطيطى بلبير لتصنيف المياه الى سحنات هيدرولوجيه.

تراوحت تركيزات أيون الفلوريد بين 1.08 - 10 ملجم/ لتر بمتوسط 3.4 ملجم/ لتر واستنتج أن عينات المياه ملوثه بتركيزات عاليه من أيون الفلوريد حيث أن غالبيه العينات لديها تركيزات أعلى من تلك المحدده وفقا لمعايير منظمه الصحه العالميه (1.5ملجم/لتر) لمياه الشرب. كما استنتج من الدراسه الهيدروجيوكيميائيه ان مصدر التركيزات العاليه لايونات الفلوريد يعود على الارجح الى التفاعل بين المياه الجوفيه والصخور البركانيه الحاويه لها مستدلا على ذلك بنتائج النمذجه الجيوكيميائيه التى أظهرت أن معظم معاملات التشبع لمعدن الكالسيت كانت موجبه (أى أنه فى حاله الترسيب) بينما معظم معاملات التشبع لمعدن الفلوريت كانت سالبه (أى أنه فى حاله الاذابه).

نفيد علما أن سكان منطقه الدراسه فى خطر كبير بسبب الفلورايد المفرطه, وخاصه عندما لا يدركون مقدار الفلوريد الذى يتم تناوله يوميا.