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Fluoride Contamination of Groundwater and Health Hazard in Central India

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Abstract

The basic bed rocks of central India are contaminated with fluorite minerals. The overuse of groundwater for irrigation causes increased mineralization of F- in the groundwater. This contaminated groundwater is widely used for drinking and other household purposes. The excess F- is excreted through urine of animals. In this work, the exposure of contaminated groundwater in domestic animals of Dongargarh city, Chhattisgarh, India is studied. The symptoms of fluorosis diseases in the domestic animals *i.e.* cattle and buffalo are surveyed. The quality and sources of the contaminants of the groundwater are discussed.

Keywords

Groundwater, Fluoride, Exposure, Fluorosis

1. Introduction

Abnormal levels of F¯ in the groundwater are common in India due to weathering of the fractured hard rock pegmatite veins composing of minerals viz. topaz, fluorite fluorapatite, villuamite, cryolite, ferro magnesium silicate, etc. [1]. The F¯ contamination of groundwater in several states of the country was reported [2]-[21]. Millions of people and animals were exposed to excessive amount of F¯ through drinking water contaminated from geogenic and anthropogenic sources, suffering with various types of fluoride diseases [22]-[31]. The goal of this work is to study F¯ contamination of the groundwater and its exposure in domestic animals *i.e.* cattle and buffalo of Dongargarh city, India.

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2. Materials and Methods

2.1. Study Area

Dongargarh (21.18842°N and 80.75875°E) is a tourist city in central India with population of 0.1 million inclusive of neighboring villages. The town was settled near majestic mountains. The contaminated groundwater is widely used for drinking, cooking, washing and agricultural purposes. Four minerals *i.e.* oligoclase, rectorite, kaolinite and feldspar have been identified in the studied area. Feldspar is one of the most dominant mineral constituents of all the above-mentioned rocks, which is highly susceptible to chemical weathering and produces various types of clay minerals [32].

2.2. Sample Collection

Forty eight groundwater samples were collected from the tube wells of the Dongargarh city from $\approx 100 \text{ km}^{-2}$ area in the post monsoon (January) and pre monsoon (May) period, 2014 by using established method, Figure 1

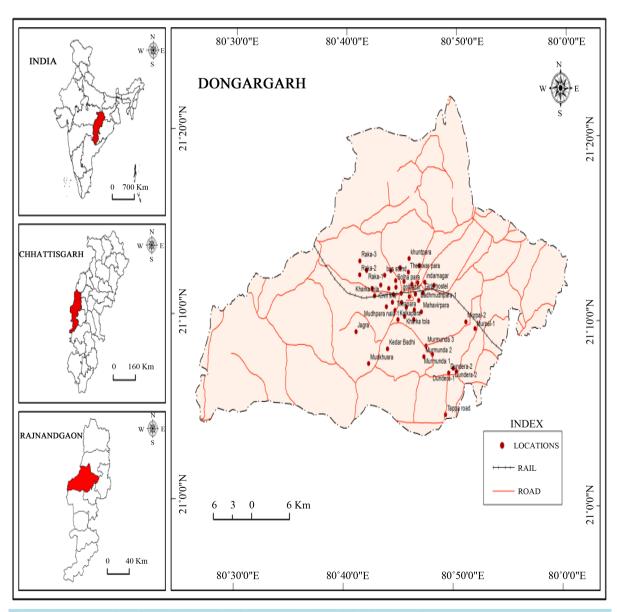


Figure 1. Representation of sampling locations in Dongargarh area, Chhattisgarh, India.

[33]. The groundwater sample was stored in 1-L cleaned polyethylene bottle. The physical parameters *i.e.* pH, temperature (T), electrical conductivity (EC), dissolved oxygen (DO) and reduction potential (RP) of the water were analyzed at the spot. The water samples were dispatched to the laboratory and preserved in the deep freezer.

The first morning urine sample (100 ml) was collected in plastic bottles containing 0.2 g EDTA. Total 40 urine samples of cattle and buffalo were collected in January, 2014. They were shipped to the laboratory in insulated container at about 4° C and refrigerated at -20° C until use.

2.3. Analysis

The total dissolved solid (TDS) value was determined by evaporation of the filtered water sample (through glass fiber filter) by drying at constant weight. The total hardness (TH) and total alkalinity (TA) values were analyzed by titration methods [34]. The F⁻ content was analyzed by using Metrohm ion meter-781 in the presence of 1:1 total ion strength adjustment buffer (TISAB). The buffer was prepared by adding 58 g NaCl + 5 g CDTA (trans-1, 2, NNNN, cyclodiamine tetra acetic acid) +57 ml glacial acetic acid and deionized water by adjusting pH value to 5.5 with 8 N NaOH in 1-L volumetric flask.

The ion (*i.e.* Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, Na⁺, K⁺, Mg²⁺ and Ca²⁺) content of the water was analyzed by Dionex-1100 ion chromatography equipped with the anion and cation columns.

The water quality index (WQI) of the groundwater was computed by using the weighed arithmetic method. The value of 6 parameters *i.e.* pH, DO, EC, TDS, TA and NO_3^- was used in calculation of the WQI with the help of following expression.

$$WQI = \sum q_n W_n / \sum W_n$$

where:

$$q_n = 100(V_o - V_{io})/(V_s - V_{io})$$

 q_n = Quality rating of the nth water quality parameter;

 V_n = Estimated value of the nth parameter of a given water;

 S_n = Standard permissible value of the nth parameter;

 V_{io} = Ideal value of the nth parameter of pure water (*i.e.*, 0 for all other parameters except pH and dissolved oxygen (7.0 and 14.6 mg/L, respectively);

 W_n = Unit weight for the nth parameter;

K =Proportionality constant.

Multivariate statistical analysis such as factor analysis (FA) was employed for the source apportionment. The windows statistical software Statistica-7.1 was used for the multivariate statistical calculation.

3. Results and Discussion

3.1. Geological Characteristics of Tube Well

The geological characteristics of the tube wells is summarized in **Table 1**. The tube wells of the studied area lie in the deeper zone, ranging from 45 - 110 m. The life was ranged from 2 - 50 Yr old. Tube wells are recharged by rain and runoff water during rainy season. The water table is varied from 20 - 50 m, depending on seasons and water uses. The higher T value for deeper tube well was observed due to geothermal energy. In turn, the higher DO value of shallow tube wells was marked.

3.2. Physical Characteristics of Groundwater

The physical characteristics of the groundwater in the post monsoon period is shown in **Table 1**. The value of pH, DO,T, RP, EC, TDS, TA and TH of groundwater located in 48 tube wells was ranged from 6.0 - 8.1, 8.4 - 9.2 mg/L, 20.0° C - 25.0° C, 237 - 330 mV, 221 - 1938 μ S/cm, 342 - 2598 mg/L, 128 - 659 mg/L and 99 - 687 mg/L with mean value of 7.2 ± 0.1 , 8.9 ± 0.1 , 23.0 ± 0.3 , 285 ± 5 mV, 861 ± 96 μ S/cm, 1138 ± 121 mg/L, 383 ± 47 mg/L and 344 ± 37 mg/L, respectively. Two ions *i.e.* Na⁺ and SO₄²⁻ were found to be responsible for contributing the EC value of the water. The DO, EC, TDS, TA and TH value of water was found to be higher than recommended value of 4.0 mg/L, 300 μ S/cm, 500 mg/L, 120 mg/L and 300 mg/L, respectively [35] [36]. The

S. No.	Location	Age, Yr	Depth, m	T°C	рН	EC, μS/cm	RP, mV	DO, mg/I
1	Mahavir Para	20	75	21	7.3	831	283	8.8
2	Goal Bajar	1	82	21	6.1	1017	310	8.9
3	Bhagat Shih Ward	3	75	21	8.1	967	237	8.6
4	Civil Line	7	69	22	6.2	991	311	8.8
5	Shubhash Ward	1	90	22	6.5	1013	319	8.9
6	Thana Chowk	7	69	22	6.4	1139	313	8.9
7	Kachari Chowk	12	75	21	6.6	603	304	8.8
8	Thethwar Para	12	82	21	6.5	344	309	8.7
9	BUS Stand	25	75	22	6.0	609	319	8.8
10	Solh Para	2	105	22	6.1	982	330	8.8
11	Ek-batti Char Rasta	10	66	23	8.0	733	265	9.1
12	Kumhar Para	15	54	22	6.5	385	269	9.0
13	School	10	69	22	6.3	276	297	9.2
14	Kumhar Para	1	75	22	6.9	349	298	8.8
15	Ambedkar Ward	1	90	23	7.3	613	254	8.9
16	Station	3	92	23	7.6	888	246	8.8
17	Bheem Nagar	4	105	22	7.3	1451	254	8.7
18	Utkarsh Nagar	8	110	22	7.3	1329	290	8.9
19	Kalka Para	10	75	22	7.2	1331	293	8.7
20	Indra Nagar	15	60	23	7.4	969	277	9.0
21	Rajiv Nagar	6	62	21	7.1	1178	304	8.7
22	Sanjay Nagar	10	66	22	7.2	1062	285	9.1
23	Khuta Para	12	72	22	7.2	951	302	8.4
24	Danteshwari Para	10	75	23	7.4	670	290	8.8
25	Kharka Tola	10	69	22	7.9	1033	282	9.0
26	Tikra Para	4	84	22	7.7	858	276	8.9
27	Kedar Badhi	23	75	21	7.6	1014	286	8.8
28	Badhiya Tola	30	82	23	7.1	1199	288	9.0
29	Raka Panchayat	24	90	22	7.2	637	275	9.0
30	Raka-2	50	75	21	7.2	322	291	9.1
31	Raka-3	3	92	21	7.1	1202	291	8.9
32	Murmunda-1	4	84	22	7.2	817	292	8.8
33	Murmunda-2	5	75	22	7.2	687	285	9.0
34	Murmunda-3	3	69	22	7.3	735	283	9.1
35	Murpal-1	10	45	22	7.3	539	276	8.9
36	Murpal-2	4	56	23	7.2	625	299	9.0
37	Murpal-3	6	64	23	7.4	1938	277	8.7
38	Jagra	2	75	22	7.4	221	272	8.9
39	Badhmudh Par-1	5	72	22	7.6	623	278	8.6
40	Badhmudhpar-2	21	72	21	7.5	707	276	8.7
41	Mudhpara Nala-1	7	75	22	7.8	1053	287	9.0
42	Mudhpara Nala-2	12	75	21	7.8	1057	272	8.9
43	Dundera-1	5	66	23	7.7	895	275	9.0
44	Dundera-2	20	69	22	7.6	1057	280	8.9
45	SBI Bank	10	90	22	7.2	605	294	8.9
46	Tappa Road	20	75	22	7.8	785	262	9.0
47	Mudhkhusra	10	66	21	7.3	681	269	9.1
. /	141uGiiKiiuSia	10	00	∠ 1	1.5	001	20)	7.1

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10

RP value of water was marked to be just of a half of recommended value of 600 mV.

3.3. Chemical Characteristics of Groundwater

The chemical characteristics of the groundwater in the post monsoon period is shown in **Table 2**. The concentration of ions *i.e.* F⁻, Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, Na⁺, K⁺, Mg²⁺ and Ca²⁺ in the groundwater of 48 tube wells was ranged from 2.0 - 10.3, 43 - 408, 48 - 152, 12 - 161, 10 - 144, 8.0 - 75, 3.0 - 25, 9.0 - 57 and 24 - 172 mg/L with mean value of 4.9 ± 0.5 , 121 ± 22 , 75 ± 8 , 47 ± 9 , 55 ± 11 , 41 ± 5 , 6.1 ± 1.5 , 28 ± 3 and 87 ± 9 mg/L, respectively. The F⁻ and NO₃ crossed the recommended limit of 1.5 and 45 mg/L, respectively in the water of all tube wells [35] [36]. However, Mg and Ca concentration was above recommended limit of 30 and 75 mg/L in the 33% and 67% tube wells [35] [36].

3.4. Seasonal Variation and Sources of Fluoride

The chemical data for the pre monsoon period, 2014 is presented in **Table 3**. The variation of physical and chemical parameters of the water in the pre monsoon period (May 2014) is presented in **Figure 2**. The value of pH, EC, TDS, TA, TH, F, Na, Mg and Ca was found to be increased \geq 30%, may be due to increase of water temperature, \approx 4°C, and deduction of water level up to 50 m. The F⁻ with the metals *i.e.* Na⁺, Mg²⁺ and Ca²⁺ had good correlation (r = 0.78 - 0.85), indicating origin from the rock weathering, **Figure 3**. Other ions (*i.e.* Na⁺, Mg²⁺, Ca²⁺, Cl⁻ and SO₄²⁻) among themselves had fare correlation, suggesting origin from multiple sources, **Table 4**. However, two ions *i.e.* NH₄⁺ and NO₃⁻ had good correlation, originating from similar anthropogenic sources.

3.5. Factor Analysis

The factor analysis of data has extracted six factors which explained 77.25% of the variance in the data set. The loadings of variables, eigenvalues and cumulative variance for each factor are shown in **Table 5**. Factor-1 accounted for 32.66% of the total variance with high positive loadings of Ca^{2+} , Mg^{2+} , F^- and TH. This factor suggests the role of dissolution/precipitation processes of some minerals such as CaF_2 and $CaCO_3$. Factor-2 accounted for 13.31% of representation with strong positive loading of pH which is negatively correlated with redox potential (RP). This factor suggests occurrence of redox processes which determine the acidic or alkaline nature of groundwater. Factor-3 represents 10.80% of the total variance with strong positive loadings of NH_4^+ and NO_3^- . This factor loadings shows the anthropogenic influences on these parameters. Factor-4 yielded 8.39% of the total variance with strong positive loadings of SO_4^{2-} and Na^+ suggesting mineral weathering. Factor-5 accounted for 6.32% of the total variance with high positive loading of K^+ . In groundwater, K could proceed from fertilizers or weathering of K-feldspar. Factor-6 accounted for 5.66% of the total variance with a high positive value of temperature (T). The T value is a variable which controls many reactions.

3.6. Water Quality Index

The WQI of the water in the post monsoon period was ranged from 22 - 226 with mean value of 97 \pm 12. The value of TDS, TA, TH, F⁻ and NO $_3^-$ in the water of all tube wells was found above permissible limits of 500, 120, 300, 1.5 and 45 mg/L, respectively [35] [36]. However, in the pre monsoon period, the value of F⁻, Mg²⁺ and Ca²⁺ crossed significantly the prescribed permissible limit of 1.5, 30 and 75, making water unsafe for drinking purposes.

3.7. Fluoride Toxicities

Chronic ingestion of fluoride water in endemic areas leads to development of fluorosis in the animal e.g. dental discoloration, difficulty in mastication, bony lesions, lameness, disability and mortality [37]. In lower age group, the lesion of teeth, skin, hair and nails were frequently observed. Fluoride enters the animal body mainly through the intake of water and quickly absorbed in the gastrointestinal tract. The excess F⁻ is excreted largely through the urine. The survey for the fluorosis in domestic animals (3 - 15 Yr) in the Dongargarh area, Rajnandgaon, Chhattisgarh, India was carried out in January 2014. A total of 40 domestic animals were screened for prevalence of various types of fluorosis *i.e.* lesion, dental, horn skin and toe fluorosis. The F⁻ concentration

Table 2. Chemical characteristics of groundwater in post monsoon period, 2014, mg/L.												
S. No.	TDS	TA	TH	F	Cl ⁻	NO ₃	SO ₄ ²⁻	NH ₄	Na ⁺	K ⁺	Mg^{2+}	Ca ²⁺
1	1187	293	414	5.5	89	95	50	90	32	3.1	30	108
2	1474	173	327	5.2	195	71	81	64	26	3.2	31	78
3	1411	275	411	6.2	160	66	74	57	34	6.4	29	108
4	1347	128	432	6.1	188	51	41	15	37	14	32	112
5	1308	189	291	4.1	142	107	55	99	28	3.3	25	72
6	1510	165	363	3.4	231	71	37	65	22	12	29	92
7	918	171	297	3.9	85	67	28	63	26	3.2	25	74
8	694	128	111	2.5	60	112	23	94	20	15	11	26
9	933	262	225	2.1	138	53	62	18	25	3.1	19	56
10	1719	177	426	6.1	195	145	63	133	33	3.2	36	106
11	886	275	123	2.2	92	53	68	39	39	12	11	30
12	537	153	171	2.9	60	59	18	34	26	3.4	15	42
13	550	153	99	3.4	56	72	17	69	34	3.5	9	24
13	545	153	120	3. 4 4.7	59	73	16	66	15	3.6	10	30
15	822	293	246	3.9	39 89	65	29	52	13 17	3.7	18	64
	982	293 549	246 297	4.3	89 85	70	34	52 61	43	3.7	23	76
16 17	982 1876	549 647	513	4.3 7.4	85 213	70 58	34 161	11	43 61	3.8	23 41	130
												88
18	1366	634	342	4.3	185	59	65	12	34	3.3	26	
19	1892	610	447	7.7	220	105	95	91	77	3.1	37	112
20	1204	659	357	7.4	67	89	63	77	73	5.1	33	86
21	1216	561	516	7.3	53	52	110	10	72	3.2	46	126
22	1348	580	360	4.4	174	55	44	49	37	3.2	28	92
23	1710	512	330	3.2	337	54	56	11	22	3.4	26	84
24	825	433	348	4.9	57	51	22	48	36	3.5	28	88
25	729	348	276	2.1	82	53	25	17	23	6.3	18	74
26	968	531	291	3.8	124	53	35	11	45	21	21	76
27	1287	549	330	3.9	149	105	26	94	45	3.2	24	86
28	991	378	438	6.2	56	68	47	57	37	3.3	36	110
29 30	957 342	305 189	282 126	4.1 2.0	75 58	89 53	41 35	85 12	17 16	9.1 3.4	24 10	70 32
31	342 1646	512	423	5.6	38 160	33 111	33	108	26	25	35	32 106
32	1152	140	300	3.1	114	48	74	42	22	17	26	74
33	1140	464	450	6.1	71	98	41	88	63	3.2	36	114
34	909	397	294	3.1	85	82	12	77	27	3.3	26	72
35	596	427	213	2.7	64	52	22	13	25	3.2	17	54
36	729	366	279	6.1	75	57	41	12	65	3.5	23	70
37	1188	390	435	6.1	75	49	104	11	63	6.2	39	106
38	2598	622	654	9.1	408	88	89	80	58	12	54	164
39	1042	512	534	7.4	43	67	12	56	69	6.3	38	140
40	1042	512	321	3.1	54	145	26	132	52	12	27	80
41	1200	427	315	4.1	107	99	26	86	31	3.7	27	78
42	722	458	285	5.6	71	53	22	10	46	3.1	25	70
42	1137	438 616	252	5.6 6.1	53	55 152	16	10	46 65	3.1	20	64
44	1525	470	687	10.3	138	65 52	48	47	85	3.4	57 26	172
45	657	299	348	5.1	57 124	53	15	17	58	3.4	26	90
46	1199	360	342	4.5	124	81	52	72	42	3.5	28	86
47	981	439	555	8.1	78	51	17	15	73 52	3.2	45	140
48	1638	500	492	5.9	234	53	74	13	53	6.2	42	122

7.7

7.7

7.6

7.8

7.8

8.0

7.9

8.2

8.2

8.1

8.0

7.6

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8.1

3.8

3.3

7.5

7.5

11.2

9.1

3.8

5.0

6.9

7.5

12.7

6.3

5.5

10.0

7.3

3.0

2.9

3.2

5.7

11.0

5.8

11.0

3.4

2.9

2.9

3.1

3.1

3.2

2.9

5.7

Table 3. Characteristics of groundwater in pre monsoon period, May 2014. F Cl⁻ NO, SO₄ NH_{4}^{+} Na⁺ K^{+} Mg^{2+} Ca^{2+} S. No. pН EC mg/L 7.7 6.8 2.9 6.4 6.4 2.9 8.5 7.6 5.9 7.5 12.9 6.5 6.8 5.0 3.0 6.7 4.2 11.0 6.9 4.8 2.9 6.8 3.1 13.8 6.3 2.6 2.9 6.4 7.5 2.9 8.4 2.7 11.0 6.8 3.6 3.1 6.6 4.2 3.2 7.2 5.8 3.3 7.7 4.8 3.4 8.0 5.3 3.5 7.7 9.1 2.9 7.7 5.3 3.0 7.6 9.5 2.9 7.8 9.1 4.7 7.5 9.0 2.9 2.9 7.6 5.4 7.6 3.9 3.1 7.8 6.0 3.2 8.3 2.6 5.8 8.1 4.7 19.3 8.0 4.8 2.9 7.5 7.6 3.0 7.6 5.0 8.4 7.6 2.5 3.1 7.5 6.9 23.0 7.6 3.8 15.6 7.5 2.9 7.6

Table 4. Correlation matrix of ions. F Cl⁻ $NO_3^ SO_4^{2-}$ NH_4^+ Na⁺ K^{+} Mg^{2+} Ca²⁺ F 1 Cl^- 0.24 1 NO_3^- 0.08 0.01 1 SO_4^{2-} 0.38 0.49 -0.141 NH_4^+ 0.05 -0.02 -0.20 1 0.94 Na^{+} 0.78 -0.010.06 0.29 -0.041 K^{+} -0.13 0.16 0.09 -0.010.11 -0.171 Mg^{2+} 0.85 0.42 0.02 0.48 0.00 0.66 -0.02 1 Ca^{2+} -0.03 0.97 0.83 0.43 -0.010.00 1

0.64

0.41

Table 5. Eigenvalues and factor loadings of Dongargarh groundwater samples.

Variable	Factor-1	Factor-2	Factor-3	Factor-4	Factor-5	Factor-6
Age	-0.18	0.07	-0.06	-0.15	-0.45	-0.66
Depth	-0.05	-0.13	0.19	0.45	0.30	-0.05
T	-0.09	0.17	-0.06	0.00	-0.25	0.82
pH	0.07	0.94	-0.04	-0.01	0.01	0.00
EC	0.24	0.17	-0.10	0.69	-0.16	0.11
RP	-0.07	-0.85	0.12	0.11	-0.09	-0.08
DO	-0.11	0.06	0.05	-0.51	-0.24	0.26
TDS	0.61	-0.04	0.21	0.65	0.23	0.09
TA	0.36	0.66	0.10	0.37	-0.18	0.09
TH	0.95	0.12	-0.02	0.25	0.03	-0.05
F ⁻	0.90	0.11	0.06	0.08	-0.11	0.12
Cl ⁻	0.36	-0.18	-0.04	0.60	0.32	0.12
NO_3^-	0.03	-0.03	0.98	-0.02	0.03	-0.02
SO_4^{2-}	0.30	-0.03	-0.24	0.73	-0.02	0.06
NH_4^+	0.02	-0.07	0.98	-0.03	0.08	0.00
Na^+	0.22	0.36	0.21	0.71	-0.21	0.16
\mathbf{K}^{+}	-0.05	0.07	0.06	0.04	0.82	-0.06
Ca^{2+}	0.94	0.14	-0.03	0.24	0.04	-0.06
Mg^{2+}	0.94	0.07	-0.01	0.26	-0.02	-0.02
Eigenvalue	6.21	2.53	2.05	1.59	1.20	1.10
% Total variance	32.66	13.31	10.80	8.39	6.32	5.77
Cumulative %	32.66	45.97	56.77	65.16	71.48	77.25

Significant factor loading in bold >0.7.

in their urine samples were measured and presented in Table 6. The concentration of F in the buffalo and cattle urines was ranged from 18 - 52 and 26 - 58 mg/L with mean value of 31 ± 4 and 41 ± 4 mg/L, respectively. At least 7 - 10 folds higher F content in the urine of animals was marked, may be due intake of higher dose of F contaminated water and food. The higher fluorosis prevalence rate was observed in the cattle than buffalo, Figures 4-6.

Table 6. Fluoride exposure in animals.

S. No.	Animal	Color	Age, Yr	F, mg/L	Fluorosis
1	Buffalo	В	6	32	DF
2	Buffalo	В	5	35	DF
3	Buffalo	В	13	29	DF
4	Buffalo	Br	8	40	DF
5	Buffalo	В	14	37	DF
6	Buffalo	В	5	35	DF
7	Buffalo	В	5	18	DF
8	Buffalo	В	7	29	DF
9	Buffalo	В	5	22	DF
10	Buffalo	В	8	45	SK
11	Buffalo	В	5	29	SK
12	Buffalo	В	5	21	DF
13	Buffalo	В	6	19	DF
14	Buffalo	В	5	22	DF
15	Buffalo	В	9	52	TF
16	Buffalo	В	7	19	TF
17	Buffalo	В	7	33	TF
18	Buffalo	В	6	35	TF
19	Buffalo	В	8	27	TF
20	Buffalo	В	15	31	TF
21	Cattle	W	10	58	SF
22	Cattle	W	8	40	SF
23	Cattle	Br	8	39	SF
24	Cattle	R	7	34	SF
25	Cattle	Gray	5	32	SF
26	Cattle	WG	6	36	HF
27	Cattle	WG	11	56	HF
28	Cattle	G	8	42	HF
29	Cattle	BW	8	39	HF
30	Cattle	WG	8	56	HF
31	Cattle	WG	9	46	HF
32	Cattle	WG	5	37	HF
33	Cattle	W	8	58	HF
34	Cattle	W	7	37	HF
35	Cattle	BW	6	32	L
36	Cattle	BW	5	26	L
37	Cattle	W	5	34	L
38	Cattle	W	6	29	L
39	Cattle	Br	7	39	L
40	Cattle	Br	9	47	L

 $DF = Dental \ fluorosis, \ SF = Skin \ fluorosis, \ TF = Toe \ fluorosis, \ HF = Horn \ fluorosis, \ L = Lesion.$

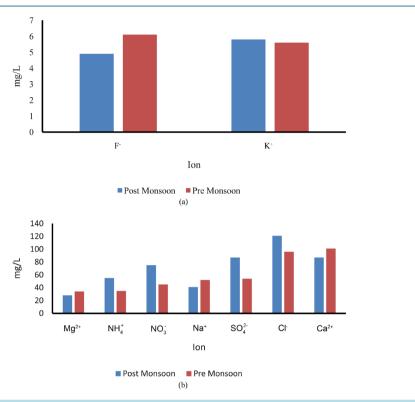


Figure 2. Seasonal variation of ionic concentration in post (a) and pre (b) monsoon period, 2014.

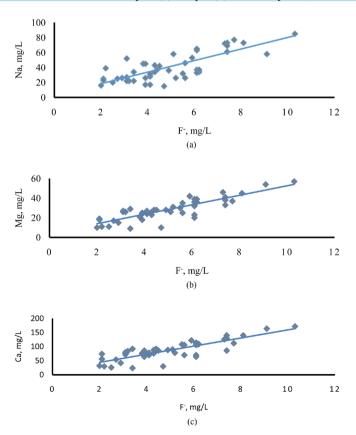


Figure 3. Correlation of F with Na⁺ (a), Mg²⁺ (b) and Ca²⁺ (c).



Figure 4. Skin lesson in cattle.



Figure 5. Horn fluorosis in cattle.



Figure 6. Dental fluorosis in buffalo.

4. Conclusion

The groundwater of Dongargarh is contaminated with F^- at dangerous levels due to mineralization of the bed rock F^- in the water. The WQI index of water was found to be ≈ 100 , making water unsafe for dinking purposes. The F^- levels in urine of cattle and buffalo were found several folds higher than recommended value of 4 mg/L. Around 5% domestic animals of the studied were suffered with different types of fluorosis.

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References

- [1] Saxena, V.K. and Ahmad, S. (2001) Dissolution of Fluoride in Groundwater: A Water-Rock Interaction Study. *Environmental Geology*, **40**, 1084-1087. http://dx.doi.org/10.1007/s002540100290
- [2] Datta, A.S., Chakrabortty, A., De Dalal, S.S. and Lahiri, S.C. (2014) Fluoride Contamination of Underground Water in West Bengal, India. *Fluoride*, **47**, 241-248.
- [3] Brindha, K., Rajesh, R., Murugan, R. and Elango, L. (2011) Fluoride Contamination in Groundwater in Parts of Nalgonda District, Andhra Pradesh, India. *Environmental Monitoring and Assessment*, 172, 481-492. http://dx.doi.org/10.1007/s10661-010-1348-0
- [4] Arveti, N., Sarma, M.R., Aitkenhead-Peterson, J.A. and Sunil, K. (2011) Fluoride Incidence in Groundwater: A Case Study from Talupula, Andhra Pradesh, India. *Environmental Monitoring and Assessment*, 172, 427-443. http://dx.doi.org/10.1007/s10661-010-1345-3
- [5] Reddy, A.G., Reddy, D.V., Rao, P.N. and Prasad, K.M. (2010) Hydrogeochemical Characterization of Fluoride Rich Groundwater of Wailpalli Watershed, Nalgonda District, Andhra Pradesh, India. *Environmental Monitoring and Assessment*, 171, 561-577. http://dx.doi.org/10.1007/s10661-009-1300-3
- [6] Rao, N.S. (2009) Fluoride in Groundwater, Varaha River Basin, Visakhapatnam District, Andhra Pradesh, India. Environmental Monitoring and Assessment, 152, 47-60. http://dx.doi.org/10.1007/s10661-008-0295-5
- [7] Suthar, S., Garg, V.K., Jangir, S., Kaur, S., Goswami, N. and Singh, S. (2008) Fluoride Contamination in Drinking Water in Rural Habitations of Northern Rajasthan, India. *Environmental Monitoring and Assessment*, **145**, 1-6. http://dx.doi.org/10.1007/s10661-007-0011-x
- [8] Vikas, C. (2009) Occurrence and Distribution of Fluoride in Groundwaters of Central Rajasthan, India. *Journal of Environmental Science and Engineering*, **51**, 169-174.
- [9] Rao, N.S. (2011) High-Fluoride Groundwater. Environmental Monitoring and Assessment, 176, 637-645. http://dx.doi.org/10.1007/s10661-010-1609-y
- [10] Hussain, I., Arif, M. and Hussain, J. (2012) Fluoride Contamination in Drinking Water in Rural Habitations of Central Rajasthan, India. *Environmental Monitoring and Assessment*, 184, 5151-5158. http://dx.doi.org/10.1007/s10661-011-2329-7
- [11] Kundu, M.C. and Mandal, B. (2009) Assessment of Potential Hazards of Fluoride Contamination in Drinking Groundwater of an Intensively Cultivated District in West Bengal, India. *Environmental Monitoring and Assessment*, **152**, 97-103. http://dx.doi.org/10.1007/s10661-008-0299-1
- [12] Jha, S.K., Nayak, A.K. and Sharma, Y.K. (2010) Potential Fluoride Contamination in the Drinking Water of Marks Nagar, Unnao District, Uttar Pradesh, India. *Environmental Geochemistry and Health*, 32, 217-226. http://dx.doi.org/10.1007/s10653-009-9277-y
- [13] Srikanth, R., Gautam, A., Jaiswal, S.C. and Singh, P. (2013) Urinary Fluoride as a Monitoring Tool for Assessing Successful Intervention in the Provision of Safe Drinking Water Supply in Five Fluoride-Affected Villages in Dhar District, Madhya Pradesh, India. *Environmental Monitoring and Assessment*, 185, 2343-2350. http://dx.doi.org/10.1007/s10661-012-2713-y
- [14] Avishek, K., Pathak, G., Nathawat, M.S., Jha, U. and Kumari, N. (2010) Water Quality Assessment of Majhiaon Block of Garwa District in Jharkhand with Special Focus on Fluoride Analysis. *Environmental Monitoring and Assessment*, **167**, 617-623. http://dx.doi.org/10.1007/s10661-009-1077-4
- [15] Singh, C.K. and Mukherjee, S. (2015) Aqueous Geochemistry of Fluoride Enriched Groundwater in Arid Part of Western India. Environmental Science and Pollution Researches International, 22, 2668-2678. http://dx.doi.org/10.1007/s11356-014-3504-5
- [16] Varadarajan, N. and Purandara, B.K. (2008) Fluoride Contamination in Ground Water of Malaprabha Sub Basin. Journal of Environmental Science and Engineering, **50**, 121-126.

- [17] Sajil Kumar, P.J. (2012) Assessment of Fluoride Contamination in Groundwater as Precursor for Electrocoagulation. Bulletin of Environmental Contamination and Toxicology, 89, 172-175. http://dx.doi.org/10.1007/s00128-012-0638-3
- [18] Vijay, R., Khobragade, P. and Mohapatra, P.K. (2011) Assessment of Groundwater Quality in Puri City, India: An Impact of Anthropogenic Activities. *Environmental Monitoring and Assessment*, 177, 409-418. http://dx.doi.org/10.1007/s10661-010-1643-9
- [19] Banerjee, A. (2015) Groundwater Fluoride Contamination: A Reappraisal. Geoscience Frontiers, 6, 277-284. http://dx.doi.org/10.1016/j.gsf.2014.03.003
- [20] Patel, K.S., Sharma, R., Sahu, B.L., Patel, R.K. and Matini, L. (2014) Ground Water Quality of Rajnandgaon City. Asian Journal of Water, Environment and Pollution, 11, 31-37.
- [21] Dutta, R.K., Saikia, G., Das, B., Bezbaruah, C., Das, H.B. and Dube, S.N. (2006) Fluoride Contamination in Ground-water of Central Assam India. Asian Journal of Water, Environment and Pollution, 3, 93-100.
- [22] Arif, M., Husain, I., Husain, J. and Kumar, S. (2013) Assessment of Fluoride Level in Groundwater and Prevalence of Dental Fluorosis in Didwana Block of Nagaur District, Central Rajasthan, India. *International Journal of Occupational and Environmental Medicine*, **4**, 178-184.
- [23] Yadav, J.P., Lata, S., Kataria, S.K. and Kumar, S. (2009) Fluoride Distribution in Groundwater and Survey of Dental Fluorosis among School Children in the Villages of the Jhajjar District of Haryana, India. *Environmental Geochemistry* and Health, 31, 431-438. http://dx.doi.org/10.1007/s10653-008-9196-3
- [24] Nayak, B., Roy, M.M., Das, B., Pal, A., Sengupta, M.K., De, S.P. and Chakraborti, D. (2009) Health Effects of Groundwater Fluoride Contamination. *Clinical Toxicology (Philadelphia)*, 47, 292-295. http://dx.doi.org/10.1080/15563650802660349
- [25] Suthar, S. (2011) Contaminated Drinking Water and Rural Health Perspectives in Rajasthan, India: An Overview of Recent Case Studies. *Environmental Monitoring and Assessment*, 173, 837-849. http://dx.doi.org/10.1007/s10661-010-1427-2
- [26] Nayak, B., Mohan Roy, M., Das, B., Pal, A., Sengupta, M.K., De, S.P. and Chakraborti, D. (2009) Health Effects of Groundwater Fluoride Contamination. *Clinical Toxicology*, 47, 292-295. http://dx.doi.org/10.1080/15563650802660349
- [27] Kotecha, P.V., Patel, S.V., Bhalani, K.D., Shah, D., Shah, V.S. and Mehta, K.G. (2012) Prevalence of Dental Fluorosis & Dental Caries in Association with High Levels of Drinking Water Fluoride Content in a District of Gujarat. *The Indian Journal of Medical Research*, **135**, 873-877.
- [28] Marya, C.M., Ashokkumar, B.R., Dhingra, S., Dahiya, V. and Gupta, A. (2014) Exposure to High-Fluoride Drinking Water and Risk of Dental Caries and Dental Fluorosis in Haryana, India. *Asia-Pacific Journal of Public Health*, **26**, 295-303. http://dx.doi.org/10.1177/1010539512460270
- [29] Panday, A. (2010) Prevalence of Fluorisis in an Endemic Village in Central India. Tropical Doctor, 40, 217-219. http://dx.doi.org/10.1258/td.2010.100032
- [30] Viswanathan, G., Gopalakrishnan, S. and Siva Ilango, S. (2010) Assessment of Water Contribution on Total Fluoride Intake of Various Age Groups of People in Fluoride Endemic and Non-Endemic Areas of Dindigul District, Tamil Nadu, South India, Water Research, 44, 6186-6200, http://dx.doi.org/10.1016/j.watres.2010.07.041
- [31] Viswanathan, G., Jaswanth, A., Gopalakrishnan, S., Siva Ilango, S. and Aditya, G. (2009) Determining the Optimal Fluoride Concentration in Drinking Water for Fluoride Endemic Regions in South India. *Science of the Total Environment*, **407**, 5298-5307. http://dx.doi.org/10.1016/j.scitotenv.2009.06.028
- [32] Bhattacharya, S., Majumdar, T.J., Rajawat, A.S., Panigrahi, M.K. and Das, P.R. (2012) Utilization of Hyperion Data over Dongargarh, India, for Mapping Altered/Weathered and Clay Minerals along with Field Spectral Measurements. *International Journal of Remote Sensing*, 33, 5438-5450. http://dx.doi.org/10.1080/01431161.2012.661094
- [33] Nielson, D.M. and Nielson, G. (2006) The Essential Handbook of Groundwater Sampling. CRC Press, Boca Raton. http://dx.doi.org/10.1201/9781420042795
- [34] Nollet, L.M.L. and De Gelder, L.S.P. (2007) Handbook of Water Analysis. 2nd Edition, CRC Press, Boca Raton.
- [35] BIS (2009) Drinking Water—Specification. 2nd Edition, Bureau of Indian Standards, New Delhi. http://bis.org.in/sf/fad/FAD25(2047)C.pdf
- [36] WHO (2011) Guidelines for Drinking-Water Quality. 4th Edition, World Health Organization, Geneva. http://www.haceclick.com.uy/documentos/GuIa OMS%202011 4aEd.pdf
- [37] Patra, R.C., Dwivedi, S.K., Bhardwaj, B. and Swarup, D. (2000) Industrial Fluorosis in Cattle and Buffalo around Udaipur, India. *Science of the Total Environment*, 253, 145-150. http://dx.doi.org/10.1016/S0048-9697(00)00426-5