

# Fluoride Contamination of Groundwater and Skeleton Fluorosis in Central India

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# Abstract

In country like India, the groundwater is widely used for drinking purposes. In central India, the groundwater is hard and contaminated with F<sup>-</sup> and other elements above the permissible limits and found to be linked with prevalence of the fluorosis diseases. In this work, the groundwater quality Balod district, Chhattisgarh, India is investigated for assessment of water quality for drinking purposes. The concentration (n = 50) of F<sup>-</sup> was ranged from 1.5 - 14.0 mg/L with mean value of  $3.9 \pm 0.8$  mg/L. The causes of prevalence of skeleton fluorosis in human of the studied area are described.

# **Keywords**

Ground Water Quality, Fluoride, Skeleton Fluorosis

# **1. Introduction**

Ground water is an important resource for drinking agriculture purposes. Groundwater uses and applications are often related to its composition, which is increasingly influenced by human activities. In fact the water quality of groundwater was affected by many factors including precipitation, surface runoff, groundwater flow, and the characteristics of the catchment area. The over extraction of groundwater caused huge weathering of the metamorphic rocks. In several regions of the country, the groundwater was contaminated with  $F^-$  beyond permissible limit of 1.5 mg/L with linking of fluorosis diseases in human [1]-[15]. The Balod district, Chhattisgarh, India is a rice producing area, taking multiple crops by using the water resources. The water is hard with contamination of  $F^-$ ,  $CI^-$  and Fe at hazardous levels. In this work, the groundwater quality of Balod district (area  $\approx 4000 \text{ km}^2$ ) is described.

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# 2. Materials and Methods

### 2.1. Study Area

The Balod district included five blocks: Balod, Gurur, Gunderdehi, Dondi Lohara and Dondi. The area of the district is  $\approx$ 4000 km<sup>2</sup> with population of 1.3 million. The materials *i.e.* buildings, metals, pipes, etc. are corroded due to acidic nature of water. The residents of the studied area were suffering with the fluorosis diseases due to mineralization of F<sup>-</sup>. In present investigation, the Balod district of Chhattisgarh state, central India has been selected for groundwater quality studies to assess the contaminants.

# 2.2. Sampling and Hydrological Parameters

The groundwater sampling network, based on water uses and contamination sources is shown in **Figure 1**. The hydrological parameters (*i.e.* age and depth) of 50 tube wells were recorded in January 2016. The water sample was collected in the cleaned narrow polyethylene 250-mL bottle in duplicate during January 2016 [16]. The physical parameters *i.e.* temperature (T), pH, dissolved oxygen (DO), reduction potential (RP) and electrical conductivity (EC) were measured at the spot. The samples were dispatched to the laboratory for the analysis by subsequent refrigerating at  $-4^{\circ}$ C.

## 2.3. Analysis

The total dissolved solid (TDS) was determined by the evaporation method. The total hardness (TH) value of the

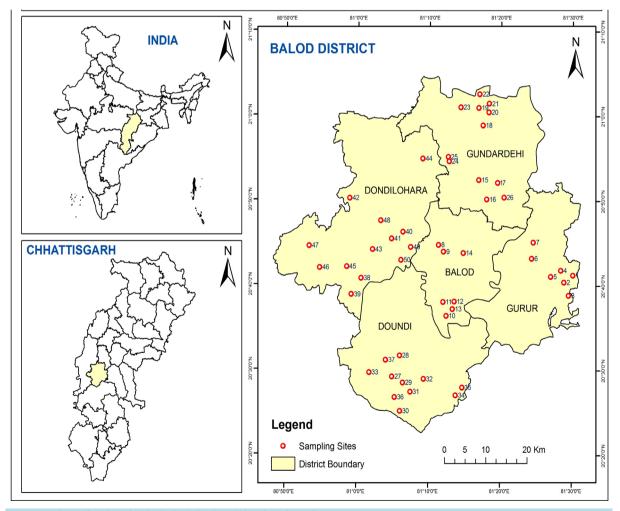


Figure 1. Sampling location in Balod district, Chhattisgarh, India.

water was analyzed by the titration method [17]. The  $F^-$  content of the water was monitored by the Metrohm ion meter-781 using the CDTA buffer in a 1:1 volume ratio. The content of other ions were measured by the Dionex ion chromatography-1100 equipped with appropriate anion and cation exchange columns and conductivity detector. The Fe content was analyzed by using the GBC flame AAS. The sources of the contaminants in the water were apportioned by the statistical analysis.

#### 3. Results and Discussion

#### 3.1. Physical Characteristics of Tube Well and Water

The physical parameters (*i.e.* age and depth of tube wells) and habitant population were recorded with the help of public health engineering department, and data are presented in **Table 1**. The total population residing in the studied is  $\approx$ 130000. The age and depth of tube wells were ranged from 4 - 25 Yr and 37 - 85 m with mean value of 11.6 ± 1.5 Yr and 50 ± 3 m, respectively. The most of tube wells of the studied area are young with shallow depth. The higher population density is observed in the Balod town. The value of T, pH, DO, RP, EC, TDS and TH of the water (n = 50) was ranged from 23°C - 28°C, 6.1 - 8.6, 10 - 12 mg/L, 146 - 298 mV, 101 - 1278  $\mu$ S/cm, 470 - 3018 mg/L and 60 - 732 mg/L with mean value of 25.0°C ± 0.3°C, 7.4 ± 0.2, 10.9 ± 0.2 mg/L, 212 ± 7 mV, 506 ± 70  $\mu$ S/cm, 1345 ± 180 mg/L and 210 ± 35 mg/L, respectively.

#### 3.2. Chemical Characteristics of Water

The chemical characteristics of the groundwater are presented in **Table 2** and **Table 3**. The concentration of  $F^-$ ,  $CI^-$ ,  $NO_3^-$ ,  $SO_4^{2^-}$ ,  $NH_4^+$ ,  $Na^+$ ,  $K^+$ ,  $Mg^{2+}$ ,  $Ca^{2+}$  and Fe was ranged from 1.5 - 14.0, 33 - 655, 4.0 - 106, 10 - 130, 1.0 - 2.9, 28 - 157, 3.0 - 48, 2.0 - 26.5, 24 - 216 and 0.4 - 4.5 mg/L with mean value of  $3.9 \pm 0.8$ ,  $190 \pm 44$ ,  $30 \pm 5$ ,  $47 \pm 10$ ,  $1.5 \pm 0.1$ ,  $79 \pm 9$ ,  $14 \pm 3$ ,  $6.5 \pm 1.4$ ,  $82 \pm 12$  and  $0.8 \pm 0.2$  mg/L, respectively. The ion concentration in the water was found to occur in following increasing order: Fe <  $NH_4^+$  <  $F^-$  <  $Mg^{2+}$  <  $K^+$  <  $NO_3^-$  <  $SO_4^{2-}$  <  $Ca^{2+}$  <  $CI^-$ . The F<sup>-</sup> content in the water of the studied area was found to be higher than reported in other parts of the country [1]-[15].

#### 3.3. Spatial Variation in Water Quality

The spatial variation in physiochemical parameters of the water is shown in **Figure 2** and **Figure 3**. The water was found to be acidic in the Gurur block. The remarkably high content of  $F^-$ ,  $CI^-$ ,  $Na^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$  was marked in the water of Gunderdehi block. However, the water of two blocks *i.e.* Dondi and Dondi Lohara becomes reddish when exposed to the air due to hydrolysis of the Fe(III) into oxides and hydroxides.

#### 3.4. Sources of Contaminants in Water

The correlation matrix of elements in the water of Gunderdehi block is summarized in **Table 4**. The content of F<sup>-</sup> ions had good correlation with the  $NH_4^+$ ,  $Na^+$ ,  $K^+$  and  $Mg^{2+}$  ions, indicating existence of F<sup>-</sup> in the geo media as Barberiite, bararite, ferruccite, sellatite, cryolite, hieratite, etc. Other ions *i.e.* Cl<sup>-</sup>,  $SO_4^{2-}$ ,  $Na^+$ ,  $K^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$  were partially correlated, showing origin from multiple sources *i.e.* geogenic, mining, agriculture, etc.

#### **3.5.** Toxicities

Three elements *i.e.*  $F^-$ , Fe and Ca<sup>2+</sup> were found in all locations above permissible limit of 1.5, 0.3 and 75 mg/L, respectively [18] [19]. In Gunderdehi block, Cl<sup>-</sup> was found at hazardous levels, >250 mg/L, may be due to mineralization. The skeletal fluorosis (*i.e.* hyperostosis, osteopetrosis and osteoporosis) was observed due to long term ingestion of  $F^-$  contaminated water by the residents of the studied area, Figure 4.

#### 4. Conclusion

The groundwater of the Balod district, Chhattisgarh, central India was found to be contaminated with Fe,  $F^-$ ,  $Cl^-$  and  $Ca^{2+}$  above permissible limits, which rendering water not suitable for the drinking purposes. The higher prevalence rate of fluorosis diseases was observed in the Gunderdehi and Balod blocks.

5. No.	Location	Block	Age of tube well, Yr	Depth of tube well, m	Population exposed to F <sup>-</sup> contamination
1	Chitoud	Gurur	10	46	2500
2	Mirritola		12	46	2000
3	Boridkala		8	55	2100
4	Userwara		10	61	2400
5	Kaneri		8	55	1800
6	Bhulandabari		25	46	2000
7	Dhaneli		8	49	2300
8	Ranitarai	Balod	12	40	2100
9	Balod		9	61	18,000
10	Madwapathra		7	46	1200
11	-		13	61	1300
	Malganv				
12	Malganv		10	46	1300
13	Malganv		17	76	1300
14	Umerdhah		12	40	2500
15	Shikosha	Gunderdehi	7	46	2500
16	Limora		20	46	1500
17	Rangakatora		25	55	2400
18	Kachandur		12	46	2600
19	Gureda		10	61	2800
20	Gureda		8	46	2800
21	Gureda		10	55	2800
22	Gureda		15	46	2800
23	Shikola		10 7	37	2100
24 25	Parna Parna		12	61 46	2400 2400
23 26	Naharkhapari		12	40 55	2500
20	Dondi	Dondi	15	37	4000
28	Kamta	Donar	5	46	1800
29	Awari		8	37	2000
30	Pusawad		13	55	1500
31	Kuwagondi		20	46	1300
32	Surdongar		8	85	1200
33	Dighwari		5	46	900
34	Chihero		4	61	1200
35	Aamadula		25	46	1900
36	Gudum		10	43	2300
37	Khairwahi		10	76	1100
38	Parsuli	Dondi Lohara	15	46	2500
39	Bharda		12	46	1200
40	Bhaihakuwa		5	46	1100
41	Jatadhah		8	55	1200
42	Sahganv		10	61	1000
43	Gainji		15	46	1200
44	Shikaritola		7	40	1500
45	Armurkasa		13	43	1800
46	Patratola		5	37	1700
47	Chikhalakasa		17	55	2300
48	Dalli-Rajhara		25	37	20,000
49	Borid		8	55	1500

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Table 2. Physical parameters of ground water in January, 2016.								
S. No.	T °C	pН	EC, μS/cm	TDS, mg/L	TH, mg/L	DO, mg/L	RP, mV	
1	23	7.4	802	599	183	11	196	
2	23	6.9	213	498	174	10	206	
3	24	7.2	530	1001	187	11	211	
4	25	7.2	417	1218	155	11	219	
5	25	7	310	1417	141	10	224	
6	24	7.5	833	1990	208	11	179	
7	24	7.7	535	1616	236	11	168	
8	24	7.9	535	470	201	11	225	
8 9								
	26	7.4	1171	650	495	11	298	
10	24	6.6	224	971	105	11	215	
11	25	6.1	271	1133	115	11	225	
12	23	6.6	345	2796	135	11	217	
13	25	7.2	490	1353	155	11	196	
14	25	7.9	491	1220	204	11	193	
15	25	7.4	338	677	176	10	188	
16	25	7.8	594	644	234	11	201	
17	24	7.7	884	1161	348	11	205	
18	24	8	412	735	184	10	198	
19	24	7.9	390	1027	155	12	250	
20	25	8	420	667	187	12	219	
21	25	8.1	509	1038	216	11	221	
22	25	7.4	490	1149	169	12	218	
23	25	7.4	1278	616	540	12	259	
23	23	8.1	702	901	345	11	146	
25	24	8.2	698	2110	295	10	201	
26 27	24 25	7.7 6.8	535 1069	866 816	198 395	10 11	198 296	
27	23 28	6.4	481	751	150	11	296	
28 29	28 24	6.5	212	910	75	11	200	
30	27	6.8	135	1640	145	11	202	
31	25	6.4	460	2288	205	11	219	
32	24	6.5	167	1085	75	11	232	
33	25	6.6	437	949	120	10	211	
34	25	6.1	141	2612	65	11	230	
35	25	7.2	413	1460	175	11	210	
36	23	7.4	520	852	247	11	211	
37	25	7.8	644	1440	185	10	184	
38	25	8.6	502	2066	732	12	248	
39	25	7.5	394	956	145	10	221	
40	26	6.9	215	1928	112	11	211	
41	25	7.5	529	1641	210	10	201	
42	25	7.6	862	2050	456	11	205	
43	25 24	8	425	1232	230	11	186	
44 45	24 26	7.7	371	1856	142	10	202	
45 46	26 26	7.7 7.7	515 487	2414 3018	185 160	11 11	215 217	
40 47	26 25	7.6	487 642	2083	180	11	217 199	
47	25 25	8	101	2381	60	10	205	
48 49	25	7.8	655	841	213	11	203	
50	25	7.3	509	1504	105	12	210	

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Table 3. Dist	Table 3. Distribution of ions in groundwater during January, 2016, mg/L.									
S. No.	$F^{-}$	Cl	$\mathbf{SO}_4^{2-}$	$NO_3^-$	$\mathrm{NH}_4^+$	$Na^+$	$\mathbf{K}^+$	$Mg^{2+}$	Ca <sup>2+</sup>	Fe
1	1.9	60	22	19	2.9	50	8	3.8	75	0.51
2	1.8	51	10	16	2.6	52	13	3.3	69	0.63
3	2.0	124	50	10	1.4	57	7	4.5	79	0.71
4	1.9	132	96	14	2.3	89	14	4.1	65	0.69
5	1.9	164	90	55	1.0	63	28	4.3	56	0.75
6	3.0	330	92	31	1.7	111	6	7.2	83	0.66
° 7	5.8	256	10	11	1.0	105	48	8.9	94	0.49
8	4.6	503	32	31	1.2	103	13	7.1	80	0.59
9	4.0	470	100	22	2.2	60	13	4.1	198	0.39
10			100							
	3.9	33		36	1.1	50	14	7.7 8	42	0.43 0.4
11	4.7	77	14	17	1.1	45	17		46	
12	6.5	111	36	37	1.3	60	8	11.3	54	0.46
13	8.6	102	48	41	1.4	89	15	11.9	62	0.42
14	7.2	73	96	55	1.2	153	20	3.5	83	0.51
15	3.6	80	12	17	1.2	89	8	4.5	70	0.52
16	2.7	230	44	19	1.0	89	8	3.4	94	0.49
17	1.9	372	86	42	1.0	40	5	2.9	139	0.51
18	2.4	107	34	36	1.1	57	5	3.2	74	0.74
19	2.1	460	18	12	1.0	73	12	3.5	62	0.66
20	7.6	260	20	18	1.4	142	26	8.3	75	0.72
21	3.1	340	114	45	1.2	77	16	4.2	84	0.68
22	12	90	20	26	2.5	129	39	26.5	68	0.78
23	11	99	30	54	2.6	153	32	23.9	216	0.64
24	4.2	413	54	17	1.2	157	15	6.4	138	0.48
25	4.0	655	68	19	1.0	105	11	5.9	138	0.51
26	4.1	410	52	24	1.1	110	9	6.3	79	4.23
27	1.8	529	108	24	1.2	94	14	3.1	158	0.59
28	3.8	210	50	29	1.5	81	10	7.2	60	0.81
29	1.5	59	10	66	1.6	45	11	2.8	30	0.76
30	3.2	40	10	37	1.1	48	12	6.5	58	0.69
31	4.0	165	42	22	2.3	52	12	8.1	82	1.51
32 33	2.0 2.0	40 130	10 32	106 23	1.0 1.0	40 93	11 11	6 3.2	30 48	0.72 0.53
33 34	2.0 1.9	68	32 24	23 41	1.0	93 48	11	3.2 2.6	48 26	0.53
34	5.4	101	34	19	2.3	48 64	44	2.0 8.5	20 70	0.31
36	1.7	65	16	33	1.5	76	3	6.7	70 79	0.65
37	4.9	147	24	35	1.3	105	8	8.9	74	0.43
38	4.9	136	17	52	1.8	86	4	23.4	178	0.49
39	1.8	74	130	41	1.0	45	31	2.3	58	0.57
40	2.0	56	12	16	1.0	60	11	3.4	45	0.66
41	5.5	108	46	4	1.4	36	8	8.9	84	0.45
42	3.5	288	92	31	1.2	93	7	6.7	182	0.51
43	3.4	61	30	23	1.2	68	7	4.1	92	0.42
44	2.1	85	24	22	2.1	73	5	3.9	57	2.04
45	3.4	40	36	24	1.1	64 77	5	4.0	74	0.78
46 47	2.2	91 240	34	26 31	2.0 1.0	77 57	6 14	3.1 2.8	64 73	4.53
47 48	1.7 1.8	249 510	118 114	31 16	1.0 1.5	57 81	14 14	2.8 2.0	73 24	0.87 0.65
48 49	2.2	155	58	23	1.5	28	14 7	3.6	24 85	0.83
50	3.3	110	34	10	2.2	101	12	3.9	42	0.62
	5.5	110	57	10	2.2	101	14	5.7	14	0.02

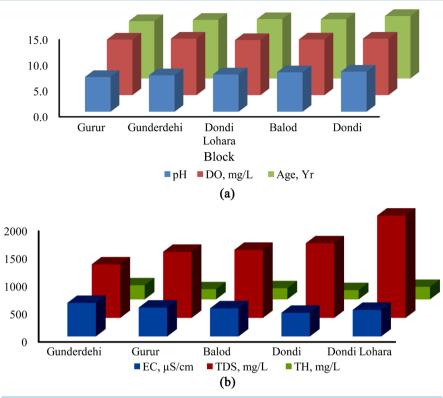


Figure 2. Spatial variation in physical parameters of water.

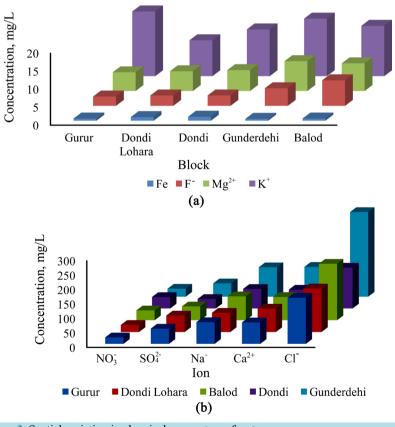


Figure 3. Spatial variation in chemical parameters of water.

Correlation matrix of elements in groundwater of Gunderdehi block.										
	$F^{-}$	Cl	$\mathrm{SO}_4^{2-}$	$\mathrm{NO}_3^-$	Na <sup>+</sup>	$\mathbf{K}^{+}$	$Mg^{2+}$	Ca <sup>2+</sup>	Fe	$\mathrm{NH}_4^+$
$F^{-}$	1.00									
Cl	-0.46	1.00								
$\mathbf{SO}_4^{2-}$	-0.41	0.49	1.00							
$NO_3^-$	0.23	-0.32	0.45	1.00						
$Na^+$	0.73	-0.09	-0.34	-0.13	1.00					
$\mathbf{K}^+$	0.96	-0.37	-0.31	0.20	0.70	1.00				
$Mg^{2+}$	0.96	-0.46	-0.35	0.33	0.62	0.91	1.00			
Ca <sup>2+</sup>	0.28	0.09	0.23	0.55	0.36	0.20	0.35	1.00		
Fe	-0.11	0.13	0.24	0.27	0.20	-0.10	0.01	0.09	1.00	
$\mathrm{NH}_4^+$	0.94	-0.58	-0.37	0.43	0.59	0.89	0.98	0.40	0.10	1.00



Figure 4. Skelton fluorosis in the studied area, Balod district, India.

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