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Fluoride geochemistry and assessment of the exposure dose of fluoride from drinking water in Garhwa district of Jharkhand, India

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ABSTRACT

From the study area the groundwater samples mostly used for drinking and other domestic purposes were analyzed for fluoride (F) content and other water quality parameters such as pH, Electrical Conductivity (EC), Carbonate (CO_3^{2-}), Bicarbonate (HCO_3^-), Chloride (Cl^-), Sodium (Na^+), Calcium (Ca^{2+}), Magnesium (Mg^{2+}), and Residual Sodium Carbonate (RSC). The F content in the ground water of the study area was found to vary between 0.52 to 7.62 mg/L and in about 24 % of water samples the F concentration exceeded the maximum permissible limits of 1.5 mg/L as laid down by World Health Organisation. The correlation study of F indicated that it has a significant ($p < 0.05$) positive relationship with pH ($r = +0.46$), Na^+ ($r = +0.33$), Sodium Adsorption Ratio ($r = +0.43$), and RSC ($r = 0.35$). In infants (6 Kg body weight) the exposure dose of fluoride was found to vary from 0.17 to 2.57 mg/kg/day. F exposure dose in case of children (20 kg body weight) varied between 0.07 to 1.14 mg/kg/day, whereas in case of adults (60 kg body weight), it varied between 0.06 to 0.88 mg/kg/day, which is quite high against the standard value of 0.05 mg/kg/day, which is the minimum risk level (MRL) calculated by the Agency for Toxic Substances and Disease Registry (ATSDR).

Keywords: Correlation; Exposure dose; Fluoride geochemistry; Groundwater.

INTRODUCTION

The fluoride (F) contamination in the ground water and the resultant disease 'Fluorosis' has become a global problem. From countries like India, China, Sri Lanka, West Indies, Spain, Holland, Italy, Mexico, and North and South America, sporadic incidence of high fluoride contents in drinking water has been reported [1-4]. In India, one of the most important toxicological and geo-environmental issue is the occurrence of high concentration of fluoride in groundwater and the prevalence of fluorosis which has been reported from 22 states of India, affecting more than 40 million people [5-9]. World Health Organization's recommendation is that drinking water should not contain more than 1.5 mg/L of fluoride [10]. Previous studies, from different parts of the world reported the development of dental fluorosis in the population, consuming drinking water containing fluoride less than 1.0 mg/L [11-13], which indicates that the optimal fluoride level in drinking water may vary with various features like local climatic conditions [14], methods of food processing and cooking [3] amount of food and water intake and its fluoride and other nutrients level [15-17], and dietary habits of the community [18-19]. Therefore the prediction of optimum F level in drinking water should be different for individual areas by considering the factors which influence fluoride

consumption or fluorosis prevalence. The occurrence of dental and skeletal Fluorosis have been reported in Garhwa district of Jharkhand [20] and the cause ascribed to the high intake of fluoride contaminated drinking water. The present study was conducted to characterize the geochemistry of ground water and to assess the exposure dose of fluoride in the drinking water.

MATERIALS AND METHODS

The study area (Garhwa district) occupies the North–West Part of Jharkhand with a geographical area of 4044.10 Sq. Km. It is an under developed and drought prone district having diverse terrain. It lies between the North Latitude $23^0$34'50" to $24^0$30'00" and East Longitude $83^0$37'25" to $84^0$04'00". Whole district is underlain by hard rock belonging to the Precambrian period and recent alluvium along river courses. The geology and structure of underlying basement controls the occurrence and movement of groundwater. The groundwater occurs in secondary porosities like joints, fractures and their inter-connected extensions within weathered mantle of hard rocks. The source of groundwater recharge is entirely by rainfall. The district comes under the Ganga and Son Sub basin. The climate of the district is humid and sub tropical. The average rainfall of the district as a whole is 1193 mm. The study area is far away from major industrial activity.

From the study area samples of ground water were collected from major sources of water supply, used for drinking and other domestic purpose. The samples were collected in pre-cleaned sterilized bottles and were immediately carried to the laboratory for chemical analysis. The analysis was carried out according to standard methods [21] for various physico-chemical parameters. Analysed parameters were pH, EC, Na^+, Ca^{2+}, Mg^{2+}, CO_3^{2-}, HCO_3^- and Cl^- concentrations. The F concentration in the water was determined by fluoride ion specific electrode using ORION ion analyzer. The correlation analysis was performed to study the relationship of fluoride with other water quality parameters.

The exposure dose of fluoride in drinking water for infant (6 kg body weight), children (20 kg body weight) and adults (60 kg body weight) was calculated by following the generic equation

ED = (C/BW) x WI

Where
ED = Exposure dose (mg/kg/day)
WI = Water intake (L/day)
BW = Body weight
C = Concentration of fluoride (mg/L)

RESULTS AND DISCUSSION

In the ground water of the study area, the fluoride content was found to vary between 0.52 to7.62 mg/L (Table 1) out of which, the fluoride concentration in about 24% of water samples exceeded the maximum permissible limits of 1.5 mg/L as laid down by World Health Organisation.[10] The statistical study of the inter-relationship between fluoride and other water quality parameters (Table 2) indicates that F has a significant (p <0.05) positive relationship with pH (r = +0.46), which has been reported by several authors [22]. From the positive correlation of pH with F, it is evident that pH is important in determining fluoride concentration in the groundwater of the study area. An important character for assessing the suitability of irrigation water is RSC, which is also significantly and positively correlated with F (r = 0.35). The elevated concentration of fluoride in the ground water was generally associated with low calcium and high sodium which was in agreement with earlier observations [23]. High F concentration in the ground water of the study area is highly related to the geochemistry of the ground water and hence, geo-genic in origin. The main source of drinking water in the study area is groundwater. The presence of shallow ground water indicates the aquifer has not been disturbed due to over-exploitation of groundwater, which ruled out anthropogenic occurrence of fluoride. In general, it has been observed that the ground water contains a higher amount of F dissolved from geological conditions, while the surface water typically contains a lesser amount of Fluoride [24].

Table 1. Analytical results of the ground water samples of Garhwa

S No	pH	EC	CO_3^{2-}	HCO_3^-	Cl^-	Ca^{2+}	Mg^{2+}	Na^+	F	RSC	SAR
1	7.56	1.02	1.20	3.12	0.40	1.01	1.69	2.05	1.30	1.62	1.76
2	7.65	1.31	2.10	4.18	1.26	1.56	1.82	2.63	1.35	2.90	2.02
3	7.62	0.81	0.85	4.23	0.60	1.25	1.68	2.75	1.09	2.15	2.27
4	7.62	0.81	1.65	3.65	0.86	1.72	2.04	2.85	1.18	1.54	2.08
5	7.63	0.82	1.80	3.75	1.25	1.88	2.12	2.96	1.20	1.55	2.09
6	7.64	0.82	1.70	3.78	1.98	2.65	2.35	3.05	1.26	0.48	1.93
7	7.10	0.59	1.06	2.38	1.30	1.20	0.86	0.96	0.86	1.38	0.95
8	7.25	0.65	1.20	2.56	1.34	1.25	0.88	1.23	0.88	1.63	1.19
9	7.38	0.72	1.25	3.28	1.65	1.28	0.90	1.62	1.02	2.35	1.55
10	7.80	0.78	1.26	3.56	1.68	1.35	0.96	2.03	1.20	2.51	1.89
11	7.96	0.82	1.28	3.69	1.70	2.34	1.47	2.98	1.26	1.16	2.16
12	7.20	1.00	1.24	3.85	0.50	0.88	2.13	4.02	1.20	2.08	3.28
13	7.65	1.25	1.60	3.98	2.60	0.90	2.18	4.26	1.25	2.50	3.43
14	7.85	1.30	1.69	4.02	2.80	1.96	2.49	4.56	1.66	1.26	3.06
15	7.75	0.90	0.00	5.09	2.00	1.68	0.64	0.87	1.60	2.77	0.81
16	7.99	0.92	1.25	5.25	2.16	1.69	0.68	1.98	1.66	4.13	1.82
17	8.01	0.94	1.26	5.69	2.20	2.68	2.34	2.65	2.11	1.93	1.67
18	7.28	0.97	1.45	2.39	1.00	0.62	3.05	3.45	0.52	0.17	2.55
19	7.36	0.96	1.55	2.45	1.20	1.15	2.38	3.62	0.68	0.47	2.72
20	7.45	0.98	1.65	2.46	1.23	1.26	2.50	3.68	0.72	0.35	2.68
21	7.56	1.20	1.68	3.26	1.54	1.28	3.20	3.72	0.86	0.46	2.49
22	7.62	1.35	1.62	3.00	1.76	1.00	1.12	3.78	1.23	2.50	3.67
23	7.69	1.46	1.86	4.85	1.80	1.35	3.53	3.96	1.45	1.83	2.54
24	7.65	1.25	1.52	2.96	1.64	1.29	2.65	3.65	1.98	0.54	2.60
25	7.12	0.95	0.60	3.75	0.90	1.56	1.98	2.00	0.82	0.81	1.50
26	7.50	0.97	1.20	3.82	0.98	1.58	1.99	2.62	0.86	1.45	1.96
27	7.70	1.01	1.28	3.89	1.02	1.86	2.03	2.91	0.92	1.28	2.09
28	7.25	0.95	0.00	2.85	0.40	0.94	1.06	3.02	0.91	0.85	3.02
29	7.70	0.98	1.60	2.98	1.65	0.95	1.08	3.42	0.91	2.55	3.39
30	7.75	1.01	1.72	3.45	1.82	1.32	3.35	3.62	0.92	0.50	2.37
31	7.50	0.85	0.00	3.02	0.30	0.71	1.02	3.04	3.02	1.29	3.27
32	7.65	0.90	1.18	3.10	1.26	0.78	1.06	3.26	4.62	2.44	3.40
33	8.20	0.98	1.95	4.00	1.68	1.45	1.32	3.96	7.62	3.18	3.36
34	7.50	0.90	0.28	4.00	1.60	1.82	0.98	2.01	1.02	1.48	1.70
35	7.55	0.91	0.76	4.45	1.98	1.83	0.99	2.08	1.18	2.39	1.75
36	7.60	0.92	0.86	4.50	2.02	1.87	1.02	2.16	1.26	2.47	1.80
37	7.59	1.08	0.23	3.02	1.40	0.54	2.02	4.82	2.02	0.69	4.26
38	7.65	1.18	1.23	3.52	2.02	0.62	2.08	4.98	2.62	2.05	4.29
39	7.80	1.21	1.32	3.69	2.08	1.75	2.20	5.65	2.92	1.06	4.02
40	7.79	0.80	0.44	4.08	1.50	1.22	1.65	1.95	0.80	1.65	1.63
41	7.79	0.85	1.60	3.08	1.82	1.69	2.02	2.10	0.90	0.97	1.54
42	7.80	0.89	0.96	4.59	2.00	1.98	2.00	2.30	0.95	1.57	1.63
43	7.20	0.95	0.69	2.50	0.90	1.04	0.83	2.20	1.30	1.32	2.28
44	7.28	0.98	0.75	2.72	1.26	1.08	0.84	2.56	1.32	1.55	2.61
45	7.30	1.04	0.80	2.86	1.60	1.48	0.86	2.93	1.35	1.32	2.71

All values are in meq /L except pH, EC and F; values of EC are in dS/m and F in mg/L

Table 2. Correlation of fluoride with different water quality parameters

Parameters	pH	EC	CO_3^{2-}	HCO_3^-	Cl^-	Ca^{2+}	Mg^{2+}	Na^+	RSC	SAR	F
pH	1.00										
EC	0.19	1.00									
CO_3^{2-}	0.33	0.35	1.00								
HCO_3^-	0.61	0.13	-0.01	1.00							
Cl^-	0.56	0.29	0.30	0.45	1.00						
Ca^{2+}	0.44	-0.16	0.19	0.59	0.43	1.00					
Mg^{2+}	0.14	0.49	0.53	0.05	0.11	0.06	1.00				
Na^+	0.23	0.68	0.36	-0.11	0.18	0.25	0.57	1.00			
RSC	0.39	-0.01	0.04	0.54	0.26	0.03	-0.55	-0.24	1.00		
SAR	0.10	0.57	0.13	-0.27	0.02	0.51	0.21	0.90	0.06	1.00	
F	0.46	0.12	0.09	0.13	0.11	0.08	-0.14	0.33	0.35	0.43	1

Values greater than 0.29 are significant at $p<0.05$

Table 3. Estimation of exposure doses for fluoride in Daltoganj

Age group	Source of water	Average water consumption (L/day)	F in water	F (mg/L)	F intake (mg/kg/day)	
					Minimum	Maximum
Infant (6 kg)	HP Boiled*	1	0.52 (min) 7.62(max)	1.04[b] 15.24[†]	0.17	2.54
Children (20 kg)	HP	3	0.52(min) 7.62(max)	0.52 7.62	0.07	1.14
Adults (60 kg)	HP	7	0.52(min) 7.62(max)	0.52 7.62	0.06	0.88

For the calculation of F^- intake we used the minimum and maximum fluoride level of 0.52 mg/L and 7.62 mg/L
[*]The sources of boiled water for infants are the water used in the reconstitution of milk formula as well as drinking purposes
[†]Considering that in boiled water, fluoride level increases proportionally to the loss of volume[3] the concentration of fluoride in water was doubled

Table 3. reflects the exposure doses to fluoride from groundwater used for drinking and household purposes of study area for infants, children, and adults. Considering the local habits of the population for the consumption of drinking water, the exposure doses were calculated on 1 L, 3 L, and 7 L for infants, children, and adults respectively. Due to the prevailing hot climate with a mean annual temperature as high as 38^oC, water consumption by the local population is very high. The occupation of the majority of people living in the study area was farming, where many of the working adults consume an average of 7 L of water per day including water used for cooking. The native diet is semisolid and starchy, containing substantial amount of water. Cooking requires average 2 L of water day^{-1} with a staple Indian diet consisting mostly of cereals and pulses. The dose estimated for the infants was found between 0.17 to 2.54 mg/kg/day, against the standard value of 0.05 mg/kg/day which is the minimum risk level (MRL) calculated by the Agency for Toxic Substances and Disease Registry (ATSDR) [25]. At these levels a clear risk for fluorosis is evident. But in the study area, the occurrence of fluorosis among the infants is rarely reported. This may be due to the fact that majority of the infants are dependent on the mother's feed and those who do not take mother's feed, depend on the cattle milk diluted with the boiled water. Therefore, the exposure of fluoride to the infants may not be so severe due to drinking water [22]. The exposure dose for water was also calculated for children (20 kg body weight) and adults (60 kg body weight). For children the dose was found to vary between 0.07 to 1.14 mg/kg/day and in case of adults it varied from 0.06 to 0.88 mg/kg/day, when ground water of the study area was used for drinking. Therefore the maximum exposure dose to fluoride for adults in the study area is 17.8 times higher than the ATSDR's MRL. From this high exposure dose it is evident that there is a high possibility of occurrence of various forms of fluorosis and osteosclerosis among the population of the study area. This estimation, however does not take into account other sources of fluoride intake such as from food, beverages (tea) vegetables, fruits and milk. Therefore the real dose of exposure would be much higher than the estimated.

The major concern of pollution in Garhwa district of Jharkhand is of excess fluoride concentration in groundwater. In order to combat the fluoride problem in the study area, a community-based water defluoridation strategy should be adopted with an emphasis on domestic defluoridation with household activated alumina filters. Education and awareness among the population about the negative impact of fluoride should be initiated. Besides drinking water, grains, vegetables and milk are also significant sources of fluoride to man, their fluoride content should be taken into account while fixing the safe limit of fluoride in drinking water.

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