

## Study on Fluoride Content of Groundwater in Jind District, Haryana, India

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**Abstract:** The continuous usage of water carrying high amounts of fluoride may prove toxic to human, animal and plants. Excessive fluoride concentrations have been reported in groundwaters of more than 20 developed and developing countries including India where 19 states are facing acute fluorosis problems. In view of this, attempts were made to find out the fluoride content of groundwater of Jind district, Haryana and its relationships with the quality determining factors of irrigation waters. In March 2004, 446 representative tube-well water samples from 62 villages of two blocks of Jind district, Haryana were collected and analyzed for fluoride and various other water quality parameters. The analytical results indicated considerable variations among the analyzed samples with respect to their chemical composition. Results further revealed that fluoride content of these waters varied from 0.33 to 13.0 mg L<sup>-1</sup> with an average value of 2.08 mg L<sup>-1</sup> in Julana block and 0.22 to 5.8 mg L<sup>-1</sup> with an average value of 1.77 ppm in Pillu Khera block, 55.4% of the tested water samples were having fluoride content more than 1.5 mgL<sup>-1</sup> and hence unsuitable for drinking purpose. Overall water quality was found unsatisfactory for drinking purposes without any prior treatment. As far as irrigation is concerned, our data show that only 1% of the tube-well waters having fluoride content above 10 mg L<sup>-1</sup>, which is the safe limit for all types of crop plants. Thus, most of the waters tested in this investigation can safely be used for irrigation.

**Key words:** Drinking water • fluoride • irrigation water • safe limit • water quality

### INTRODUCTION

Water is an essential natural resource for sustaining life and environment that we have always thought to be available in abundance and free gift of nature. However, chemical composition of surface or subsurface is one of the prime factors on which the suitability of water for domestic, industrial or agricultural purpose depends. Freshwater occurs as surface water and groundwater. Though groundwater contributes only 0.6% of the total water resources on earth, it is the major and the preferred source of drinking water in rural as well as urban areas, particularly in the developing countries like India because treatment of the same, including disinfection is often not required. It caters to 80% of the total drinking water requirement and 50% of the agricultural requirement in rural India. But in the era of economical growth, groundwater is getting polluted due to urbanization and industrialization.

Presence of various hazardous contaminants like fluoride, arsenic, nitrate, sulfate, pesticides, other heavy metals etc. in underground water has been reported from

different parts of India [1]. In India, fluoride is the major inorganic pollutant of natural origin found in groundwater. Fluoride in minute quantity is an essential component for normal mineralization of bones and formation of dental enamel [2]. It is known to stimulate the growth of many plant species [3], but its essentiality for plant growth has not yet been established. On the other hand when fluoride is taken up in excessive amounts may prove toxic to plants and on feeding may cause clinical disturbances in animal and human being such as fluorosis, marked by changes in teeth and skeletal system. There are more than 20 developed and developing nations that are endemic for fluorosis. These are Argentina, USA, Morocco, Algeria, Libya, Egypt, Jordan, Turkey, Iran, Iraq, Kenya, Tanzania, S. Africa, China, Australia, New Zealand, Japan, Thailand, Canada, Saudi Arabia, Persian Gulf, Sri Lanka, Syria, India, etc. [4]. In India, it was first detected in Nellore district of Andhra Pradesh in 1937 [5] and the recent studies shows approximately 62 million people including 6 million children suffers from fluorosis because of consumption of water containing high concentrations of fluoride [6].

Table 1: \*USPHS recommendations for maximum allowable fluoride in drinking water

Annual average of maximum daily air temperature (°C)	Recommended fluoride concentration (mg L <sup>-1</sup> )			Maximum allowable fluoride concentration (mg L <sup>-1</sup> )
	Lower	Optimum	Upper	
10-12	0.9	1.2	1.7	2.4
12.1-14.6	0.8	1.1	1.5	2.2
14.7-17.7	0.8	1.0	1.3	2.0
17.8-21.4	0.7	0.9	1.2	1.8
21.5-26.2	0.7	0.8	1.0	1.6
26.3-32.5	0.6	0.7	0.8	1.4

\*United States public health service

According to WHO permissible limit for fluoride in drinking water is 1.5 mg L<sup>-1</sup> [7], whereas United States Public Health Service [8] has set a range of allowable concentrations of fluoride in drinking water for a region depending on its climate conditions because the amount of water consumed and consequently the amount of fluoride ingested being influenced primarily by the air temperature [9, 10]. Table 1 shows the maximum allowable fluoride concentrations as established by USPHS. Accordingly, the maximum allowable concentration of fluoride in drinking water in Indian conditions comes to 1.4 mg L<sup>-1</sup> while as per Indian standards it is 1.5 mg L<sup>-1</sup> [11].

Excess fluoride affects plants and animals also. The severity of injury is determined by duration of fluoride exposure and concentration. The fluoride concentrations in groundwater in India vary considerably. In some parts of India, the fluoride levels are below 0.5 mg L<sup>-1</sup>, while at certain other places, fluoride levels as high as 30 mg L<sup>-1</sup> have been reported [12]. This study was undertaken to assess the quality of underground water with special reference to fluoride content of two blocks of the Jind district in Haryana state, India.

## MATERIALS AND METHODS

**Study area:** Geography and geology: Survey and characterization of underground irrigation water of Julana and Pillu Khera blocks of Jind district in Haryana, India were undertaken during March 2004. Geologically, district Jind is a part of Indo-gangetic plain, mostly laid down by Indus system tributaries and other non-existent rivers. Soil of the district represents the alluvium, deposited by the Indo-gangetic river system and belongs to Pleistocene age. Medium soils, particularly loam are found in the district. Depth of water table generally ranges from 0.83 to 39.8 meter. Water table is the shallowest in the areas along canals and towards Ghaggar

River, whereas it is deep, generally resting below 30 meter, in the central parts of the district. In the areas where water table is closer to the land surface water logging and soils salination exist.

**Climate:** Jind district has a sub-tropical monsoon climate with a mean annual rainfall of 470 mm. The climate of the district is on the whole dry, hot in summer and cold in winter. The year may be divided into four seasons. The cold season from November to March is followed by hot season which lasts till the onset of the monsoon. Monsoon (rainy season) withdraws by 15<sup>th</sup> September and is followed by the post monsoon or the transition period or autumn season. The rainfall is mainly concentrated during the rainy season, which is inadequate and uncertain. The situation has considerably improved due to the availability of irrigation facilities. Rainfall increases from southwest to northeast. About 10% of the annual rainfall is also received during the winter months of December to February, in association with cyclones, which pass across the country. Winds are generally light, with some strengthening in force during the late summer and early monsoon season. Average maximum temperature for May and June is 44.5°C. Likewise, the average minimum temperature for January and February is 3.0°C.

**Collection of water samples:** Ground water samples (446) were collected from 38 villages of Julana block and 23 villages comprising of Pillu Khera block. From each village a minimum of two and a maximum of 15 water samples were collected in the month of March 2004. Running tube-wells distantly apart within each village were selected, randomly for collection of water samples. Each selected tube-well were run for three to four hours and then the samples were collected in thoroughly cleaned plastic bottles, properly labeled and brought to the laboratory for further chemical analysis.

**Analysis of water samples:** Water samples were analyzed using standard methods [13]. The pH was determined using a glass electrode connected to Elico, LI 610 pH meter, while EC was measured with Elico, CM 180 conductivity meter. Sodium and potassium were measured using a flame photometer (Systronics, 128) and calcium and magnesium were determined titrimetrically using versenate titration method. Carbonates and bicarbonates were determined by titrating a known volume of water against standard sulphuric acid (0.01 N). Chloride was measured by titration with silver nitrate. Sulphate was determined by turbidimetric method using barium chloride [14]. Fluoride was determined in water using an optimum buffer system with the fluoride selective electrode connected with an Orion ion analyzer.

**Reagents and standards:** Analytical grade chemicals were used throughout the study without further purification. To prepare all the reagents and calibration standards, double distilled water was used. All the experiments were carried out in triplicate. The results were reproducible within  $\pm 3\%$  error limit.

**Classification of groundwater on the basis of fluoride content:** Fluoride is classified as per WHO specifications for drinking water. According to this specification maximum permissible limit of fluoride for drinking water is  $1.5 \text{ mg L}^{-1}$  [28]. For irrigation purpose fluoride is classified according to criteria given by Leone et al. [15] who proposed a  $10 \text{ mg L}^{-1}$  limit for all type of plants [15].

## RESULTS AND DISCUSSION

**Major ion composition:** The dissolved component characteristics of the groundwaters from Julana and Pillu Khera blocks are summarized in Table 2. The groundwaters from the two blocks are alkaline, pH ranging between 7.04-9.90 in Julana block and 7.03-9.30 in Pillu Khera block. Alkalinity, expressed as  $\text{HCO}_3^-$ , ranges between 2.0 and 22.0  $\text{me L}^{-1}$  in Julana and 2.0 and 24.80  $\text{me L}^{-1}$  in Pillu Khera block.  $\text{Cl}^-$  is one of the dominant anion, with a concentration range of 0.60-99.0 and 0.80-59.6  $\text{me L}^{-1}$  in respective blocks,  $\text{SO}_4^{2-}$  ranges from 0.85 to 28.4 and 0.56 to 23.81  $\text{me L}^{-1}$  respectively, while  $\text{Na}^+$ , the most dominant cation varied from 0.55 to 65.3 and 0.55 to 48.0  $\text{me L}^{-1}$  in Julana and Pillu Khera blocks respectively. Calcium concentrations are notably low, ranging from 0.60 to 35.0 and 0.80 to 30.0  $\text{me L}^{-1}$  respectively. In Julana block the fluoride content varied from 0.33 to 13.0  $\text{mg L}^{-1}$  with a mean value of 2.08  $\text{mg L}^{-1}$  whereas in Pillu Khera block it varied from 0.22 to 5.8  $\text{mg L}^{-1}$  with a mean value of 1.77  $\text{mg L}^{-1}$ . The fluoride rich waters are characterized by high concentrations of  $\text{Na}^+$  and low concentrations of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . Low  $\text{Ca}^{2+}$  results from the intense cation exchange reaction between  $\text{Ca}^{2+}$  and  $\text{Na}^+$  [16]. High  $\text{HCO}_3^-$  concentrations and alkaline pH promote the precipitation of  $\text{Ca}^{2+}$  as calcite [16] and  $\text{Mg}^{2+}$  as dolomite and all of the studied ground waters are saturated with those minerals.

Fluoride content also varied from one well to another in the same village. For instance, in village Shamlo Kalan

Table 2: Quality of groundwater in Jind district, Haryana, India

S. No.	Parameters	Blocks					
		Julana			Pillu Khera		
		Min.	Max.	Mean	Min.	Max.	Mean
1	EC ( $\text{dSm}^{-1}$ )	0.27	11.00	3.61	0.26	7.80	2.71
2	PH	7.04	9.90	7.92	7.03	9.30	8.00
3	$\text{Na}^+$ ( $\text{me L}^{-1}$ )	0.55	65.30	17.01	0.55	48.00	11.96
4	$\text{K}^+$ ( $\text{me L}^{-1}$ )	0.02	2.90	0.48	0.02	2.72	0.42
5	$\text{Mg}^{2+}$ ( $\text{me L}^{-1}$ )	0.80	54.00	9.19	0.60	20.00	6.26
6	$\text{Ca}^{2+}$ ( $\text{me L}^{-1}$ )	0.60	35.00	10.87	0.80	30.00	9.29
7	$\text{CO}_3^{2-}$ ( $\text{me L}^{-1}$ )	0.00	2.00	0.74	0.00	2.40	0.83
8	$\text{HCO}_3^-$ ( $\text{me L}^{-1}$ )	2.00	22.00	10.04	2.00	24.80	9.33
9	$\text{Cl}^-$ ( $\text{me L}^{-1}$ )	0.60	99.00	20.18	0.80	59.60	14.00
10	$\text{SO}_4^{2-}$ ( $\text{me L}^{-1}$ )	0.85	28.40	5.98	0.56	23.81	4.28
11	$\text{F}^-$ ( $\text{mg L}^{-1}$ )	0.33	13.00	2.08	0.22	5.80	1.77
12	RSC ( $\text{me L}^{-1}$ )	0.00	14.00	2.71	0.00	7.20	2.26
13	SAR ( $\text{mmol L}^{-1}$ ) <sup>a</sup>	0.20	12.73	5.34	0.20	14.50	4.26

Table 3: Electrochemical properties and fluoride content in groundwater of Julana block in Jind district, Haryana

S. No.	Villages	No. of Samples	pH	EC dS m <sup>-1</sup>	RSC me L <sup>-1</sup>	SAR (mmol L <sup>-1</sup> ) <sup>1/2</sup>	F mg L <sup>-1</sup>	Frequency of F content in waters (%)			
								Suitable for drinking 0.0-1.49	Suitable for irrigation but unfit for drinking 1.5-4.99	5.0-10.0	Unsuitable for irrigation > 10.0 mg L <sup>-1</sup>
1	Kinana	9	7.54	3.71	4.20	5.63	1.05	67.0	33.0	*	-
2	Barar Khera	15	7.81	2.88	3.66	4.71	1.15	69.0	31.0	-	-
3	Anoopgarh	10	8.43	4.30	0.00	6.50	1.04	70.0	30.0	-	-
4	Shamlo Khurd	9	7.70	3.97	1.53	7.65	0.96	88.9	11.1	-	-
5	Khema Kheri	2	7.39	7.65	0.00	10.67	0.64	100.0	-	-	-
6	Shamlon Kalan	4	7.77	1.98	0.00	4.65	1.73	40.0	60.0	-	-
7	Ramkali	7	8.63	1.55	2.40	5.56	1.51	57.1	42.9	-	-
8	Karsola	10	8.12	3.16	4.00	6.54	1.47	70.0	30.0	-	-
9	Julana	8	7.80	2.19	2.10	4.73	2.49	25.0	75.0	-	-
10	Karela	10	8.11	3.26	3.92	5.71	2.16	30.0	70.0	-	-
11	Jamola	9	7.98	2.96	1.93	5.07	2.18	33.3	66.7	-	-
12	Kherabagta	7	7.78	4.83	1.20	7.09	1.87	42.9	57.1	-	-
13	Ghadwali Khera	7	8.06	2.32	0.00	3.52	1.88	28.6	71.4	-	-
14	Kharainti	9	7.84	3.47	4.20	4.08	1.95	55.6	44.4	-	-
15	Desh Khera	9	7.92	5.95	0.00	7.38	2.53	-	100.0	-	-
16	Malvi	5	8.46	1.86	3.88	4.24	0.84	100.0	-	-	-
17	Karnauch Khera	5	7.66	3.79	4.05	6.66	2.01	80.0	-	20.0	-
18	Raj Garh	7	7.86	3.53	2.97	5.64	2.64	14.3	85.7	-	-
19	Lijwana Khurd	11	7.55	6.08	0.00	5.88	1.67	36.4	63.6	-	-
20	Sirsa Kheri	8	8.09	4.84	4.20	6.02	5.24	-	62.5	25.0	12.5
21	Nand Garh	5	8.25	3.14	3.88	5.74	3.70	-	80.0	20.0	-
22	Fateh Garh	12	8.26	2.12	3.37	3.29	2.75	8.3	91.7	-	-
23	Lijwana Kalan	11	7.87	3.27	Nil	3.82	1.96	54.5	36.4	9.1	-
24	Mehrara	6	7.88	4.22	Nil	6.03	1.89	50.0	50.0	-	-
25	Akal Garh	4	7.95	1.89	Nil	1.92	2.73	-	100.0	-	-
26	Budha Khera	12	7.87	3.22	1.98	5.33	1.67	58.3	41.7	-	-
27	Deorar	11	7.70	5.21	2.50	6.50	2.76	-	90.9	9.1	-
28	Bamanwas	11	7.90	4.47	0.00	5.14	2.01	18.2	81.8	-	-
29	Shadipur Khera	4	7.27	6.73	Nil	8.39	2.78	25.0	75.0	-	-
30	Khudali	10	7.94	3.46	7.00	6.66	3.34	-	90.0	10.0	-
31	Paoli	7	8.07	1.91	2.27	4.09	5.16	-	71.4	14.3	14.3
32	Jai Jaivanti	6	7.94	1.77	1.95	4.38	1.68	50.0	50.0	-	-
33	Gatauli	6	8.11	1.97	1.15	4.73	2.47	16.7	83.3	-	-
34	Gosain Khera	6	8.08	3.00	1.73	5.26	1.64	50.0	50.0	-	-
35	Bura Deohar	7	7.62	4.43	0.40	6.67	1.25	71.4	28.6	-	-
36	Boana	6	7.46	4.68	Nil	4.94	1.01	83.3	16.7	-	-
37	Dhigana	8	8.15	4.30	1.20	4.54	1.31	50.0	50.0	-	-
38	Padana	3	7.41	4.87	Nil	3.63	2.37	-	100.0	-	-

\*Nil

of Julana block four water samples contained 0.8, 1.10, 2.0 and 3.0 mg L<sup>-1</sup> fluoride and in village Hadwa of Pillu Khera block three water samples contained 0.64, 0.85 and 3.20 mg L<sup>-1</sup> fluoride in groundwater. This variation can be attributed to the irregular distribution of fluorine bearing minerals in the soil, which are the main source of F in water [17, 18].

**Health impacts of fluoride:** Fluorine being a highly electronegative element has extraordinary tendency to get attracted by positively charged ions like calcium. Hence the effect of fluoride on mineralized tissues like bone and teeth leading to developmental alternations are of clinical significance as they have highest amount of calcium and thus attract the maximum amount of fluoride that gets

Table 4: Electrochemical properties and fluoride content in groundwater of Pillu Khera block in Jind district, Haryana

S. No.	Villages	No. of Samples	pH	EC dS m <sup>-1</sup>	RSC me L <sup>-1</sup>	SAR (mmol L <sup>-1</sup> ) <sup>1/2</sup>	F <sup>-</sup> mg L <sup>-1</sup>	Frequency of F content in waters (%)			
								Suitable for drinking 0.0-1.49	Suitable for irrigation but unfit for drinking 1.5-4.99	5.0-10.0	Unsuitable for irrigation > 10.0 mg L <sup>-1</sup>
1	Dathrath	10	8.20	1.21	0.80	1.27	1.32	70.0	30.0	*-	-
2	Mohammad Khera	7	8.13	2.57	2.36	5.27	1.39	71.4	28.6	-	-
3	Mandi Khurd	10	7.68	2.12	0.00	3.62	0.81	100.0	-	-	-
4	Kharak Gadian	6	8.10	1.60	3.44	5.18	1.91	33.3	66.7	-	-
5	Ahlanjogi Khera	6	7.82	2.71	2.60	1.99	0.93	83.3	16.7	-	-
6	Ratauli	5	7.97	1.85	2.40	4.04	1.21	80.0	20.0	-	-
7	Jamani	10	7.83	1.75	0.40	2.60	1.51	30.0	70.0	-	-
8	Rajana Kalan	4	7.93	1.30	0.90	1.11	2.26	25.0	75.0	-	-
9	Beri Khera	4	7.95	1.46	1.77	2.77	3.68	100.0	-	-	-
10	Budha Khera	3	7.99	2.85	2.30	5.29	0.99	66.7	33.3	-	-
11	Dharoli	10	8.07	4.46	4.00	7.10	1.80	40.0	60.0	-	-
12	Pillu Khera	4	8.24	2.19	0.20	3.27	2.80	25.0	75.0	-	-
13	Bhurain	6	7.89	2.05	2.60	4.65	0.87	83.3	16.7	-	-
14	Kalwa	10	7.88	4.47	Nil	6.38	2.82	20.0	70.0	10.0	-
15	Kalawati	2	7.99	4.85	Nil	8.49	1.31	50.0	50.0	-	-
16	Kharak Gagar	5	8.17	3.26	4.40	5.91	2.43	60.0	20.0	20.0	-
17	Gangoli	8	8.04	3.47	2.80	3.54	2.81	-	87.5	12.5	-
18	Bhirtana	10	7.98	3.68	0.80	4.74	1.62	50.0	50.0	-	-
19	Hadwa	3	8.20	4.97	7.20	6.65	1.56	66.7	33.3	-	-
20	Bhag Khera	9	7.82	3.22	3.93	4.71	1.64	44.4	55.6	-	-
21	Morkhi	5	8.01	2.46	0.20	3.27	0.99	80.0	20.0	-	-
22	Bhambhewa	10	8.38	1.33	1.71	4.60	2.34	50.0	40.0	10.0	-
23	Ludana	3	7.80	4.23	Nil	6.65	1.93	33.3	66.7	-	-

\*Nil

deposited as calcium-fluoroapatite crystals. Tooth enamel is composed principally of crystalline hydroxylapatite. Under normal conditions, when fluoride is present in water supply, most of the ingested fluoride ions get incorporated into the apatite crystal lattice of calciferous tissue enamel during its formation. The hydroxyl ion gets substituted by fluoride ion since fluoroapatite is more stable than hydroxylapatite. Thus, a large amount of fluoride gets bound in these tissues and only a small amount is excreted through sweat, urine and stool. The intensity of fluorosis is not merely dependent on the fluoride content in water, but also on the fluoride from other sources, physical activity and dietary habits.

If fluoride concentration in drinking water is more than 1.5 mg L<sup>-1</sup>, it would result in fluorosis (dental fluorosis and skeletal fluorosis) for human beings, especially for children and pregnant woman. This is a result of the destruction of metabolic calcium and phosphorus, inhibition of active enzymatic process in the

human body, thus the function of the endocrine system is interrupted, lead to fluorosis [19]. Our study shows that 60% of water samples from Julana (Table 3) and 47% water samples from Pillu Khera block (Table 4) have fluoride concentration more than 1.5 mg L<sup>-1</sup> and hence unsuitable for drinking purpose and may cause fluorosis if used for drinking, however, these well waters are the sole source of drinking water in most of the villages surveyed, so there is a need for the further investigation on the extent of fluorosis in these villages.

A safe limit of 10 mg L<sup>-1</sup> of fluoride has been proposed for all types of crop plants by Leone et al. (1948). Our data in Table 3 and 4 shows that 1% sample from Julana block crossed this limit of 10 mg L<sup>-1</sup> while in Pillu Khera block none of the water samples have found to cross this limit. This limit may not be equally applicable to the waters of arid and semi-arid regions where fluoride toxicity is associated with high salinity. A continuous application of such waters having toxic concentrations of

fluoride is likely to affect adversely the crop growth [20, 21]. Moreover, the harmful effect of fluoride will depend more on the actual fluoride concentration in the root zone and other soil-water-plant relationships that govern it rather than on the fluoride content in irrigation water alone [22].

**Defluoridation of water:** Defluoridation of drinking water is the only practicable option to overcome the problem of excessive fluoride in drinking water, where alternate source is not available. During the years following the discovery of fluoride as the cause of fluorosis, extensive research has been done on various methods for removal of fluoride from water and wastewater. These methods are based on the principle of adsorption, ion-exchange [23], precipitation-coagulation [24, 25], membrane separation process [27], electro dialysis [28] etc.

### CONCLUSION

Based on the findings of the current investigation, it can be concluded that most of the water samples, collected from the two blocks of the Jind district show a high amount of alkalinity in the groundwaters and the sampled groundwater do not meet the water quality standards for fluoride concentration and many other quality parameters. Hence it is not suitable for consumption, without any prior treatment. Further, some of the water samples analysed are having fluoride content beyond 10 mg L<sup>-1</sup> and hence unsuitable for irrigation purposes. Correlation matrix of fluoride with other water quality parameters showed that there do not exist any significant correlation, it appears that solubility of fluoride does not seem to be related much with the chemical characteristics of the irrigation waters probably due to the occurrence of various types of fluoride bearing minerals, their solubility and movement in soil and other environmental conditions.

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