Research Journal of Chemical Sciences _____ ISSN 2231-606X
Vol. **5(2),** 76-84, February **(2015)** *Res. J. Chem. Sci.*

Fluoride Contamination in Drinking Water and its Impact on Human Health of Kishanganj, Bihar, India

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Available online at: www.isca.in, www.isca.me
Received 1st February 2015, revised 16th February 2015, accepted 18th February 2015

Abstract

This paper is aimed to analyze the fluoride concentration in groundwater, their resources and monitoring of the impact of fluoride on human health in kishanganj district, Bihar, India. In the present study the fluoride concentration varied from 0.61-3.74 mg/l in different groundwater resources of various villages. Among 150 hand pump/tap water samples of all 5 blocks of Kishanganj district 110 (73.33%) sites (villages and town areas) are safe, 28(18.67%) villages are under dental fluorosis and 12(8.0%) villages are under skeletal fluorosis, whereas among 150 open/ring water samples, 121 (81%) sites are safe, 14(14%) villages are under dental fluorosis and 8(5.0%) villages are under skeletal fluorosis. The results were on the basis of the questionnaire survey conducted in five blocks of Kishanganj district. During the study, 2500 people were examined, of which 53.6% people were affected by dental fluorosis and 11.2% people were suffering from skeletal fluorosis. Fluorosis was found to be high in males compared to females. High pH, low calcium concentrations and high temperatures of the study area may contribute to high fluoride concentrations in groundwater. Questionnaire survey results generally indicate that majority of residents of rural area were suffering from dental and skeletal fluorosis due to unawareness and lack of precautionary measures against fluoride. If people become aware of impact of fluoride and use pure and fluoride free water then the probability of dental and skeletal problems may be minimized.

Keywords: Fluoride, Drinking water, impacts, human health, fluorosis, Kishanganj district.

Introduction

India has two major public-health problems induced by use of groundwater as a source of drinking water having excess fluoride and arsenic. Due to a variety of natural and human influences groundwater resources are under threat in few parts of the country[1- 5]. Small quantity of fluoride is an essential component for normal mineralization of bones and formation of dental enamel. However, excess concentration may result in slow, progressive scourge known as fluorosis. More than 20 developed and developing nations are suffering from fluorosis[6]. In India, fluoride in drinking water was first reported at Nellore district of Andhra Pradesh in 1937 and then considerable works have been done in different parts of India[7]. At present, it has been estimated that fluorosis is widespread in seventeen states of India indicating that endemic fluorosis is one of the most acute public health problems of the country. More than 66 million people are estimated to suffering from fluorosis, among which 6 million children are below 14 years of age in India alone[8]. Dipankar *et al*. reported that 20 out of 28 Indian states have some degree of groundwater fluoride contamination; impacting 85-97% of districts in some states[9].

Fluorine is widely distributed in earth's crust and exists in the form of fluorides. The natural concentration of fluoride in groundwater depends on the geological, chemical and physical characteristics of resources, types of the soil and rocks, temperature and the action of other chemical substances[10].

Fluoride is completely absorbed by the gastrointestinal tract and the absorbed fluoride is rapidly distributed throughout the body. Since fluorine is highly electronegative element, it has a strong tendency to get attracted by positively charged calcium ions in teeth and bones and excessive intake result in pathological changes in teeth and bones, such as mottling of teeth or dental fluorosis followed by skeletal fluorosis[11]. Thus, a large amount of fluoride gets bound in these tissues and only a small amount is excreted via urine, faeces and sweat. Low concentration of fluoride causes dental caries, both in children and adults[12]. Skeletal fluorisis is observed when drinking water contains 3-6 mg of fluoride per liter and developed as crippling skeletal fluorosis when drinking water contains over 10 mg of fluoride per liter. Other sources of fluoride poisoning are food, industrial exposure, drugs, cosmetics, etc[13]. WHO, has stated that India and China are the most affected countries to fluoride exposure. BIS (Bureau of Indian Standards) permissible limit for fluoride is 0.6-01.2 mg/l and WHO (International Standards for Drinking Water) is 1.5 mg/lit.

Material and Methods

Study area: Kishanganj district is situated at latitude of 25^0 20' to 26^0 30' north and longitude of 87^0 7' to 88^0 19' east. The climate of the district is humid with maximum temperature of

41^0C in May and minimum of 5^0C in January. The main rivers of the district are Mahananda, Kankai, Mechi, Ratwa, Ram Chandra, Donk and Kaul. In Kishanganj, two types of minerals are found; silica and iron. The forest area of the district is mainly situated in the two blocks of Thakurganj and Terhagachh. In the past, Kishanganj had recorded the highest rainfall in Bihar. The average rainfall is 2250 mm, of which 80 percent occurs during the monsoon.

Water samples were collected during summer (Mar-June), rainy (July-Oct) and winter (Nov-Feb) in the year 2011 and 2013 from 30 locations of municipal area of Kishanganj block, 30 villages from 10 Panchayats (3 villages from each panchayat), each of four blocks namely Bahadurganj, Kochadhaman, Teragachh and Thakurganj, covering entire Kishanganj district. The sampling points were confined to hand pumps, tap water, open wells, ring wells and PHE supply for drinking purposes. The research work was carried out in laboratory of P.G. Department of Chemistry, D.S. College, Katihar (Bihar), Shiva Test House, Bailey Road, Patna, Recognized as Environmental Laboratory by Central Govt. and laboratory of Kishanganj College of Engineering andTechnology Veriadangi, Kishanganj, Bihar.

Methodology: Temperature and pH were measured immediately at the spot after the collection of samples with the help of Celsius mercury thermometer graduated up to 110^0C and pH meter (Model digital pH Hmeter 335) respectively. Ion Selective Electrode (ISE) method described by Rong *et al*., recommended by APHA (1991)[14,15]. Calcium was estimated by EDTA Titration method as described by UNEP GEMS/Water Program (1985), and earlier referred by Environmental Canada 1974[16]. It was important to measure temperature, pH and calcium as they influence the concentration of fluorides in groundwater. The mean fluoride concentration for each sampling site was compared with the DWAF, BIS and WHO guidelines for domestic use to assess compliance[17-18,21].

Results and Discussion

Fluoride Distribution: All the sampling stations (the hand pump/tap, open/ring well wise concentration of fluoride) were categorized as follows[19]: Category I- Fluoride Concentration below 1.0 mg/l, Category II- Fluoride Concentration between 1.0-1.5 mg/l, Category III-Fluoride Concentration between 1.5-3.0 mg/l, Category IV- Fluoride Concentration between 3.0-4.0 mg/l.

The abstract of fluoride distribution in hand pump/tap and open/ring well water samples are shown in table-1 and 2 respectively. The colored comparative charts and pie charts are also prepared as shown figure- 2 to 5. Fluoride concentration in hand pump/tap water samples in the five blocks of Kishanganj district ranges from 0.65-3.74 mg/l. The minimum concentration (0.65 mg/l) was recorded from 2 sites [at S4 (K.L.Das) in Subhashpally and S10 (at B.K.Thakur) in Railway Colony] of Kishanganj municipal area while maximum concentration (3.74 mg/l) was recorded at S5 Haat Tola in Pawakhali village of Thakurganj block.

The present survey reveals that out of 150 hand pump/tap water samples in 63 villages (42%) have fluoride concentration ≤1.0 mg/l and fall in category-I. The concentration of fluoride is beneficial up to 1.0 mg/l, therefore there is no possibility of fluorosis in these habitations. Once fluoride is deposited into teeth, it reduces the solubility of the enamel and thereby provides protection against dental caries[20].

47 locations (31.3%) have fluoride concentration above 1.0 mg/l and ≤ 1.5 mg/l and fall in category-II, which is the maximum desirable limit of standards for drinking water (WHO). More than 4 mg per day fluoride is taken by the people of these habitations. Therefore, possibility of the first and second degree dental fluorosis is possible[22].

28 villages (18.7%) have groundwater with fluoride concentration above 1.5 mg/l and ≤ 3 mg/l and fall in category-III, which is above the maximum permissible limit of standards for drinking water (BIS,WHO). At this concentration, mottled enamel and in some cases, the pre stage of skeletal fluorosis has been observed[23,24]. In 12 villages (8%) fluoride concentration in groundwater is above 3.0 mg/l and ≤ 4 mg/l and fall in category-IV. The intake of fluoride per day by the population in this habitation is very high and cause dental as well as skeletal fluorosis[20,22].

Table-1
Abstract of Fluoride Distribution in Hand Pump/Tap Water Samples (Summer)

Block/Town	Fluoride content	No. of Water Samples with Percentage				Total no. of samples
		0-1.0 mg/l	1-1.50 mg/l	1.5-3.0 mg/l	3.0-4.0 mg/l	
Kishanganj	0.65-1.55	18 (60.0%)	10 (33.3%)	02 (8.7%)	----------	30
Bahadurganj	0.66-3.16	14 (46.7%)	12 (40.0%)	03 (10%)	01 (3.33%)	30
Kochadhaman	0.68-3.26	11 (36.7%)	13 (43.3%)	04 (13.3%)	02 (6.7%)	30
Teragachh	0.67-3.35	10 (33.3%)	08 (26.7%)	08 (26.7%)	04 (13.3%)	30
Thakurganj	0.74-3.74	10 (33.3%)	04 (13.3%)	11 (36.7%)	05 (16.7%)	30
----------	----------	63 (42.0%)	47 (31.3%)	28 (18.7%)	12 (8.0%)	150
----------	----------	63+47=110 (73.3%)safe		----------	----------	----------

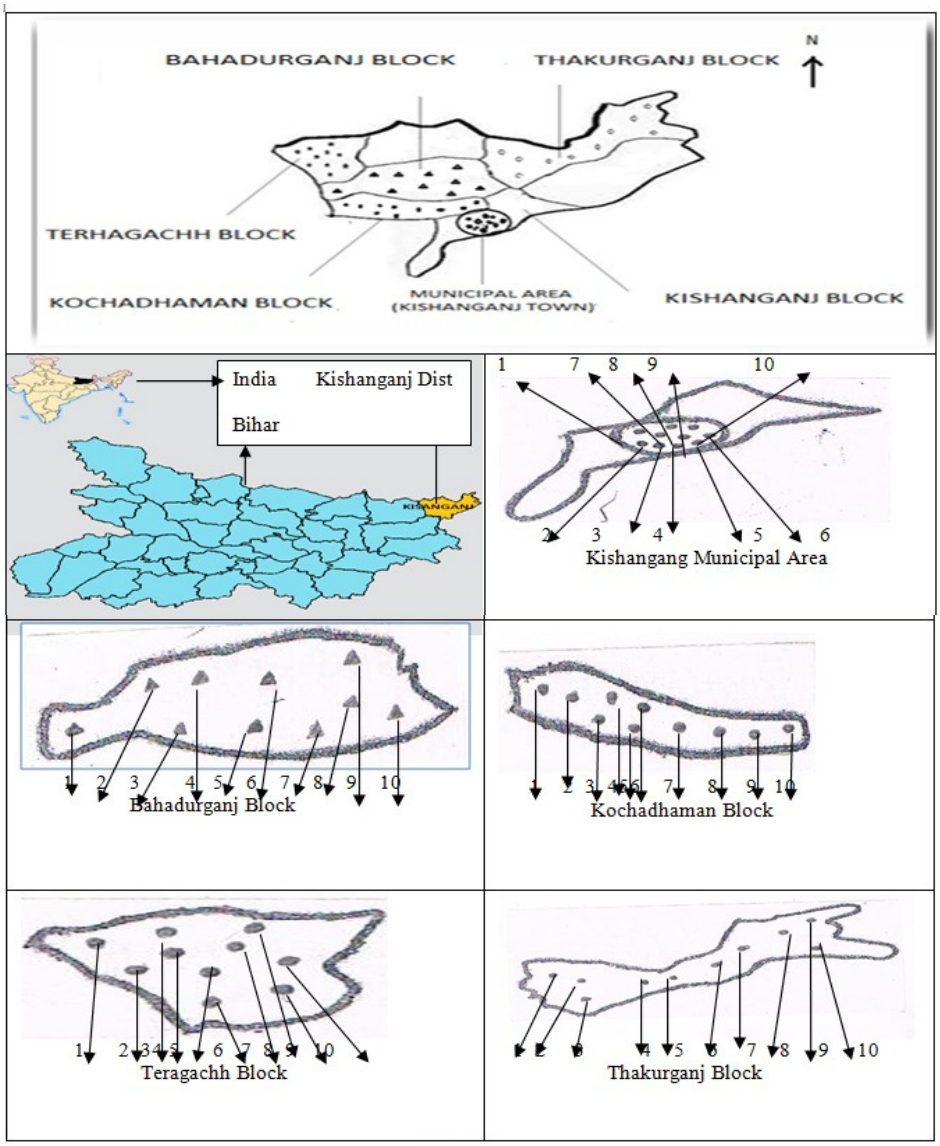
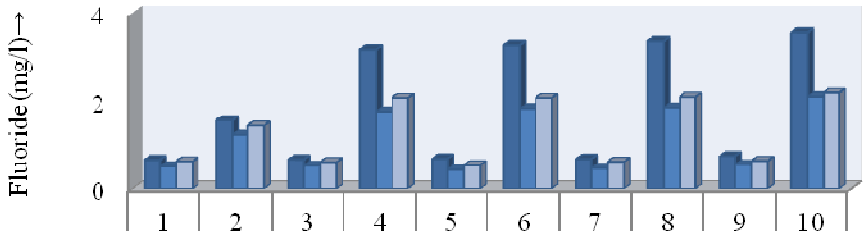


Figure-1
Sketch map of Kishanganj district showing five sampling blocks



	1	2	3	4	5	6	7	8	9	10
Summer	0.65	1.55	0.66	3.16	0.68	3.26	0.67	3.35	0.74	3.54
Rainy	0.5	1.22	0.52	1.75	0.43	1.82	0.46	1.84	0.54	2.09
Winter	0.62	1.43	0.6	2.06	0.53	2.06	0.61	2.08	0.63	2.18

Figure-2
Graph showing seasonal variations of fluoride in hand pump/tap water samples at different locations

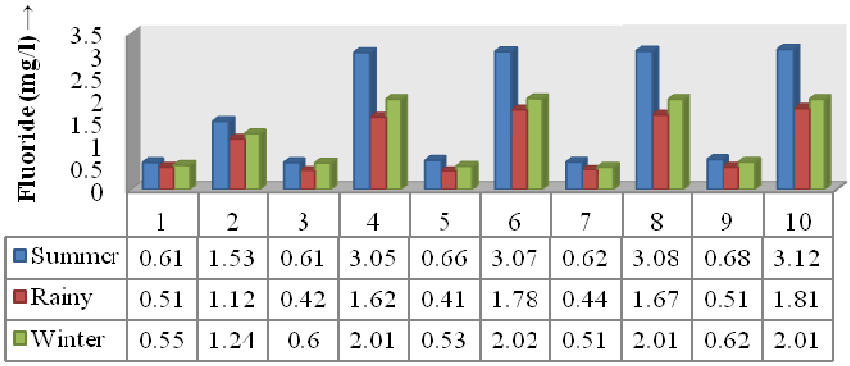
The results indicate that municipal area of Kishanganj block (as control) is safer than other block as shown in the result presented in table-1. From pie-chart as shown figure- 4, it is clear that among 150 locations of all 5 blocks of Kishanganj district 110 (73.33%) sites (villages and town areas) are safe, 28(18.67%) villages are under dental fluorosis and 12(8.0%) villages are under skeletal fluorosis.

From the abstract presented in the table-2, fluoride concentration in the open/ring well water samples ranges from 0.61-3.12 mg/l. In the Kishanganj municipal area fluoride content ranges from 0.61-1.53 mg/l, where as Bahadurganj, Kochadhamn, Teragachh and Thakurganj block is 0.61-3.05 mg/l, 0.66-3.07 mg/l, 0.62-3.08 mg/l and 0.6.8-3.12 mg/l respectively. Kishanganj municipal area of Kishanganj block has been selected as control. Among 30 samples of municipal area 28 (93.33%) have the fluoride content within the permissible limit and 2(6.67%) samples have above permissible limit.

In Bahadurganj, Kochadhamn, Teragachh and Thakurganj block have 27 (90%), 25 (83.3%), 23 (76.7%) and 18 (60%) have within the permissible limits and 3 (10%), 5 (16.7%), 7 (23.3%) and 12 (40.0%) samples respectively have above permissible limit. The results indicate that open/ring wells of municipal area of Kishanganj block is safer than other block as shown in the result presented in table-2. From pie-charts as shown in figure 4 and 5, it is clear that open well water samples are safer than hand pump water samples at the region of study area.

Table-2
Abstract of Fluoride Distribution in Open/Ring Water Samples (Summer)

Block/Town	Fluoride content	No. of Water Samples with Percentage				Total no. of samples
		0-1.0 mg/l	1-1.5.0 mg/l	1.5-3.0 mg/l	3.0-4.0 mg/l	
Kishanganj	0.61-1.53	18 (60.0%)	10 (33.3%)	02 (6.7%)	----------	30
Bahadurganj	0.61-3.05	16 (50.0%)	11 (36.7%)	03 (10.0%)	----------	30
Kochadhaman	0.66-3.07	13 (43.3%)	12 (40.0%)	04 (13.3%)	01 (3.33%)	30
Teragachh	0.62-3.08	12 (40.0%)	11(36.7%)	04 (13.3%)	03 (10.0%)	30
Thakurganj	0.68-3.12	10 (33.3%)	08 (26.7%)	08 (26.7%)	04 (13.3%)	30
----------	----------	69 (46.0%)	52 (34.7%)	21 (14.0%)	08 (5.3%)	150
----------	----------	69+52=121 (80.7%) Safe		----------	----------	----------



	1	2	3	4	5	6	7	8	9	10
Summer	0.61	1.53	0.61	3.05	0.66	3.07	0.62	3.08	0.68	3.12
Rainy	0.51	1.12	0.42	1.62	0.41	1.78	0.44	1.67	0.51	1.81
Winter	0.55	1.24	0.6	2.01	0.53	2.02	0.51	2.01	0.62	2.01

Figure-3
[Graph showing seasonal variations of fluoride in open/ring well water samples at different locations]

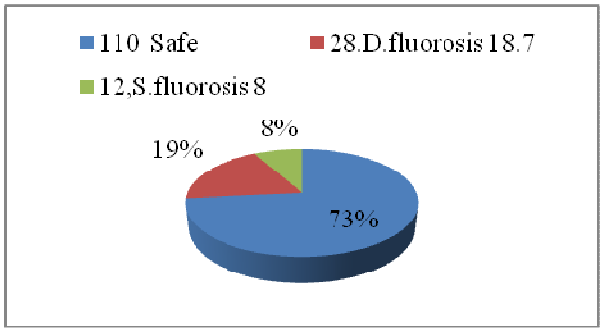


Figure-4
Fluoride toxicity in the hand/tap water samples]

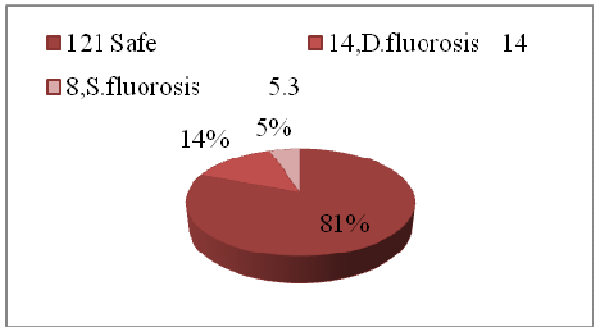


Figure-5
Fluoride toxicity in the open/ring well water samples

The comparative charts and figures show that fluoride concentration at all sampling sites (hands/tap, open/ring wells,) decreased during rainy season (July- Oct) and levels increased during winter season (Nov-Feb) and summer (March-June). The results show the effect of dilution and concentration associated with wet and dry season respectively

Chemical and Physical Controls: Correlation between calcium and fluoride concentration: The concentration of calcium is lower in summer season for mostly sampling stations as shown in figure- 6 and 7. The lower concentration of calcium increases the solubility of CaF_2, with increase in the concentration of fluorides in groundwater. The reaction between Ca^{++} and F^- ions indicates the fluoride concentration is controlled by equilibrium of fluorite[25]. When the water is saturated with respect to fluorite, low calcium concentration leads to higher fluoride concentration[26]. Higher fluoride concentration is therefore expected in ground water from aquifers with low calcium concentrations. The results generally show that increase in calcium is associated with decrease in fluoride and vice versa.

Correlation between pH and fluoride concentrations: From comparative study as shown in figure-8 and 9, pH values are higher in summer and lower in rainy.

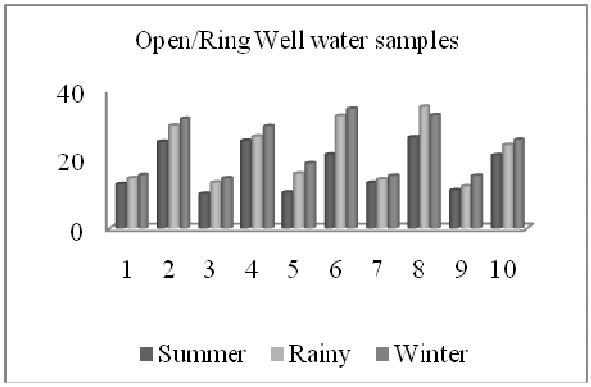


Figure- 6
Graph showing seasonal variations of Ca

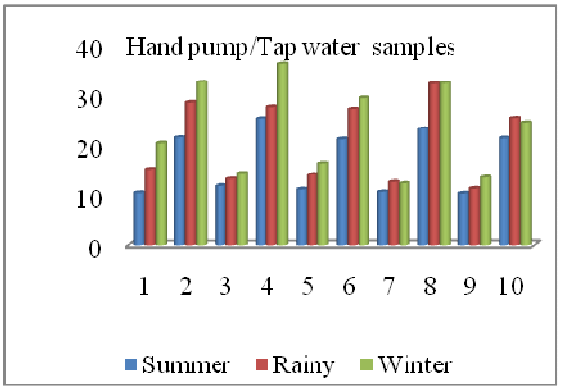


Figure-7
Graph showing seasonal variations of Ca

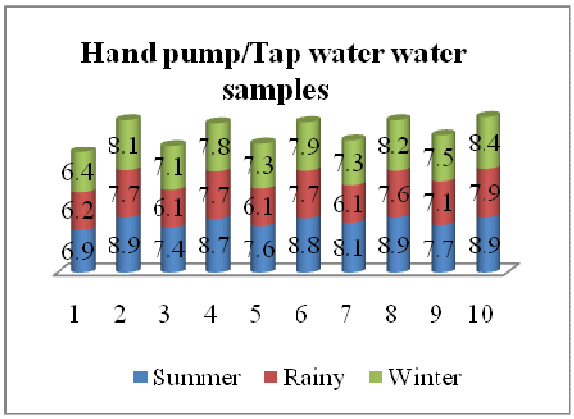


Figure-8
Graph showing seasonal variations of pH

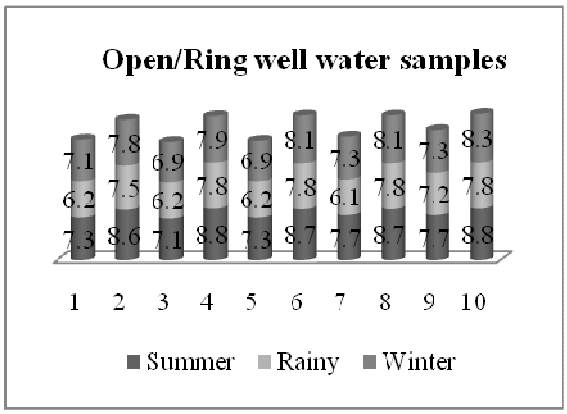


Figure-9
Graph showing seasonal variations of pH

When pH >8, fluoride concentrations commonly exceed 1.0 mg/l. Main rivers flowing through the Kishanganj district are Mahananda, Kanki, Mechi, Ritwa, Ramzan, Donk and Kaul. Due to weathering and leaching of fluoride minerals in rock formation under alkaline environments lead to the increase of fluorite in ground water[27]. Saxena and Ahmed reported that an alkaline pH ranging from 7.4-8.8 resulted in high fluoride concentrations (1.7-6.1 mg/l) in groundwater sources in India[28]. Alkaline pHs which increased the solubility of fluoride-bearing formations likely contributed to increased fluoride concentrations at hand pumps in this study.

Fluoride in Area of Intense Evaporation: The maximum average temperature in Kishanganj district during summer, rainy and winter for March-June, July-Oct, Nov-Feb were 38^0C, 30^0C and 24^0C respectively. Thermal waters especially those of high pH are rich in fluoride. Though the temperature values in summer are higher than those in rainy and winter season, their fluoride concentrations throughout the sampling period are generally high and comparable as shown in figure-2 (hand pumps) and figure-3 (open wells). This is likely to be due to the fact that their calcium concentration and pHs are also generally low and high respectively and comparable as shown in figure-6,

7, 8 and 9, resulting high fluoride concentration. Wet seasons have relatively low temperatures associated with low fluorides availability as observed. Similar trend of studies has also been reported by Madhnure et al.[29].

Evaluation of Impact of Fluorides on Human Health based on questionnaire: The results of the questionnaire survey was conducted on age group from 12 to 55 years' old people in the 10 locations of kishanganj town and 40 Panchayats of rural area in four block of kishanganj district to evaluate the impact of fluoride on human health. 2500 local people were examined for dental and skeletal fluorosis. The occurrence and severity of dental and skeletal fluorosis was recorded in table-3 and 4[23].

Dental fluorosis grade: Grade-0: Healthy teeth, Grade-I: Yellowish with white opaque, Grade-II: Brown stains, Grade-III: Chipped edges with brown stains, Grade-IV: Dark brown with loss of teeth.,

Skeletal fluorosis clinical grading: Grade-I: Mild joint pain, Grade-II: Restricted movement of joints due to stiffness, Grade-III: Deformities of bones leading to crippled or bed ridden

The present survey reveals that out of 2500 people 743 (29.7%) people were suffering from dental fluorosis of grade I, 392 (15.7%) people of grade II and 246 (9.84%) peoples of grade III on scale of grade I to IV, whereas 1160 (46.4%) people have healthy teeth as shown in table-3. The percent of skeletal fluorosis grade was found in 279 (11.2%) people of grade I, but no case was observed in grade II and III as shown in table-4. It was also observed that out of 2500 people 53.6% people were affected by dental fluorosis of grade 0-III, among which 694(27.8%) male and 646 (25.9%) female and 11.2% people are suffering from skeletal fluorosis (grade I), among which 169(6.7%) male and 110 (4.4%) female in all five blocks of Kishanganj district. The study shows that the percentage of males affected were greater than females. The similar case was observed by Saxsena and Narwaria in Karera block of Shivpuri district (M.P.)[30].

The results also indicate the persistence of high fluoric concentrations in groundwater even during the wet seasons. Hand pumps/taps and open/ring wells are the main sources of drinking water for Kishanganj township and villages of Kishanganj district, thus high fluoride concentration in groundwater from these sites S27 (Kishanganj town), S29, S30 (Bahadurganj block) S26, S27, S29 and S30 (Kochdhaman block), S3, S14, S18, S19, S23 and S24, (Teragachh block) and S5, S6, S9, S14, S15, S17 and S18 (Thakurganj block)] constitute a major health risk to the majority of Kishanganj district residents.

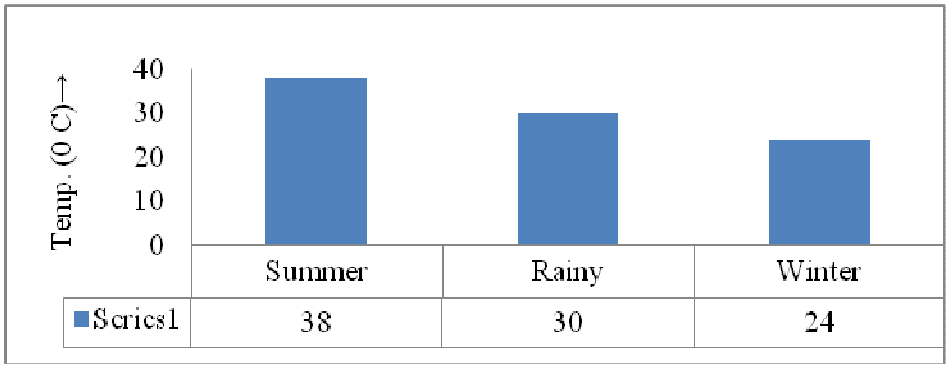


Figure-10
Graph showing mean temperature values

Table-3
Occurrence of Dental fluorosis

Examined Block /Town	Total No. of cases	Affected cases		Unaffected cases	Dental fluorosis (%)				Occurrence (%)
		Male	Female	0	I	II	III	IV	
Kishanganj	500	119	115	266 (53.2%)	139 (27.8)	60 (12)	35 (07)	--	46.8
Bahadurganj	500	138	129	233 (46.6%)	150 (30)	72 (14.4)	45 (09)	--	53.4
Kochadhaman	500	141	130	229 (45.8%)	133 (26.6)	89 (17.8)	49 (9.8)	--	54.2
Teragachh	500	145	134	221 (44.2%)	101 (20.2)	122 (24.4)	56 (11.2)	--	55.8
Thakurganj	500	151	138	211 (42.2%)	120 (24)	108 (21.6)	61 (12.2)	--	57.8
	Total 2500	694 27.8%	646 25.9%	1160 (46.4%)	743 (29.7)	392 (15.7)	246 (9.84)	--	53.6

Figure-A
Istaruh Nisha, Chakla panchayat

Figure-B
Rubina, kochadhaman Block

Figure-C
Amela Khatoon, near Engg. College

Figure-D
Jumatan Nisha, Jahangir Tola

Figure-E
K. L. Das, Subhashpally (Father), ***Residents in Kishanganj District, since 8 years***

Figure-F
Bittu Das, Subhashpally (Son), ***Residents in Kishanganj District, since 8 years***

Figure-G
Gauri, Thakurganj

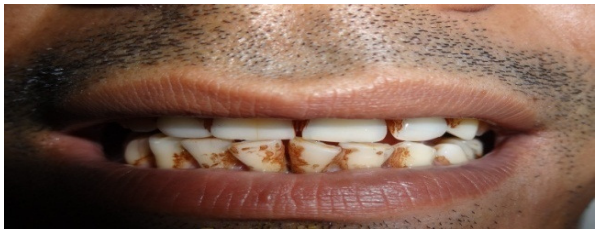


Figure-H
Pankaj at Pachimpali, Kishanganj

Figure-I
Arun Chowdhary at Keltax, Kishanganj
Figure-11
Showing dental fluorosis of different grads among villager of Kishanganj district

Conclusion

Finally it is concluded that in the present study the fluoride concentration varied from 0.61-3.74 mg/l in different groundwater resources of various villages in five blocks of Kishanganj district. The study also reveals that open well water samples are safer than hand pump water samples at the region of study area. High pH, low calcium concentrations and high

temperatures of the study area may contribute to high fluoride concentrations in groundwater. Questionnaire survey results generally indicate that majority of residents of rural area were suffering from dental and skeletal fluorosis due to unawareness and lack of precautionary measures against fluoride. If people become aware of impact of fluoride and use pure and fluoride free water then the probability of dental and skeletal problems may be minimized

Table-4
Occurrence of Skeletal fluorosis

Examined Block /Town	Total No. of cases	Affected cases		Unaffected cases	Skeletal fluorosis (%)			Occurrence (%)
		Male	Female	0	I	II	III	----------
Kishanganj	500	15	12	473 (94.6%)	27 (5.4)	----	----	5.4
Bahadurganj	500	27	17	456 (91.2%)	44 (8.8)	----	----	8.8
Kochadhaman	500	33	21	446 (89.2%)	54 (10.8)	----	---	10.8
Teragachh	500	41	29	430 (86.1%)	70 (14.0)	----	---	14.0
Thakurganj	500	53	31	416 (83.2%)	84 (16.8)	----	---	16.8
----------	Total 2500	169 (6.76%)	110 (4.4%)	2221 (88.8%)	279 (11.2%)	----	---	11.2

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