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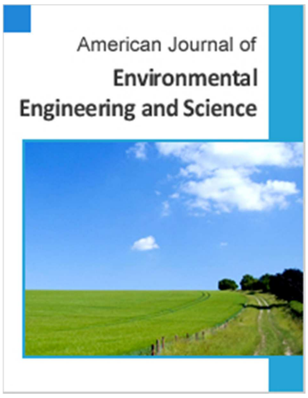

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Fluoride Analysis of Groundwater Around Tea-Gardens in Kishanganj Block, Bihar, India: Correlation with Physicochemical Parameters

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Abstract
The use of pesticides, weedicides and agrochemicals by tea growers and farmers for the growth and to enhance the rate of production, would have generated a lot of concern for public health and environmental pollution. Therefore, assessment and monitoring of drinking water sources located in the tea garden areas were almost necessary. In this paper monitoring of the fluoride content in groundwater was determined near 12 small tea gardens of Kishanganj block of Kishanganj district, Bihar, India. 36 samples were collected from different locations and analyzed for the determination of various physicochemical parameters. Fluoride concentration varied from 0.18-4.20 mg/L. Correlation study revealed that most of the samples were within the permissible limit but fluoride content was found to be positively correlated with pH, conductance, Cl^-, SO_4^{2-} and total alkalinity and negatively correlated with total hardness, Ca^{2}, Mg^{2+}, NO_3^- and PO_4^{3-}. During the study, 600 people were examined, of which 325 (54.2%) people were affected by dental fluorosis, whereas 65 (10.83%) people were suffering from skeletal fluorosis. Fluorosis was found to be high in males compared to females. Questionnaire survey also revealed that large numbers of the people living in and around the tea gardens engaged in physical labor were illiterate and unaware of the adverse effects of fluoride contaminated water. Therefore this work will be helpful for students, teachers, scientists, researchers and governmental and non governmental agencies to take correct and appropriate steps by creating awareness among the illiterate people and make them free from fluoride borne diseases.

1. Introduction

Water is an important part of our environment. All the living creatures depend upon water in one way or the other but there are instances that civilizations have disappeared due to shortage of fresh water or due to water borne diseases [1]; [2]. The main problem now before the world is that of, availability of safe drinking water which is fast assuming alarming proportions. Contamination of groundwater in an area depends on the physical and chemical parameters, which greatly affect the geological formation and human activities [3]. The quality of groundwater is the function of physicochemical and biological parameters through which it passes [4].

Some elements within limits are essential for aquatic organisms, plants as well as human beings but are toxic at higher concentrations. Fluorine is one among them [5]. Fluoride concentration in the range of 0.6-1.0mg/L is beneficial to prevent, dental caries and osteoporosis [6]. Excess fluoride intake (>1.5 mg/L) may lead to fluorosis which result in thyroid problems, growth retardation and kidney damage [7]. Excess fluoride greater than 2mg/L may lead to an increase of demineralization leading to the precipitation of calcium phosphate and calcium fluoride. Thus excess fluoride can initiate an erratic calcium metabolism which results in deformation and mottling of teeth. In fluorosis, due to substitution of active hydroxyl group by fluoride, some enzymes like enolase and pyrophosphatase etc. become inactive. Fluoride concentration greater than 3mg/Lin drinking water may cause dental fluorosis, which is shown by weakening of tooth enamel, development of brown or yellow pitches on teeth [8]. Intake of fluoride above 4-8mg/L may lead to severe toxicity like osteo sclerosis is in which 50% hydroxyl groups in hydroxyapatite are replaced by fluoride in bones. It leads to skeletal fluorosis and hypercalcification. More than 10mg/L of fluoride content in groundwater can cause crippling due to skeletal fluorosis [9]; [10]. Therefore, types and severity of fluorosis depends on the presence of fluoride concentration in drinking water, climatic conditions, continuity and duration of exposure.

The source of fluoride in groundwater is fluoride-bearing minerals such as fluorspar, fluorite, cryolite, hydroxylapatite and fluoapatite. The bedrock of Indian peninsula contains a number of these minerals like fluorite, apatite, rock phosphate and phosphorite. When the bedrock weathers, fluoride leaches into the water and soil. The disposal of industrial effluent and municipal sewage lead to the contamination of rivers with fluoride along with toxic elements [11]. The persistence of fluoride in groundwater is also influenced by physicochemical parameters, availability and solubility of fluoride minerals in water and velocity of flowing water [12]. The occurrence of fluoride exposure in human being is mainly through drinking water, food, industrial exposure, drug, cosmetic, pharmaceuticals and pesticide residues [13]. Among them drinking water is the major source of daily intake [14]. It was reported that leaching of chemical fertilizers and pesticides residues from the soil are responsible for high fluoride content in water system [15]

More than 20 developed and developing nations are suffering from fluorosis [12]. Rural Development Ministry has estimated (April, 2010) that 33,303 villages in 18 states of our country have high level of fluoride in groundwater. There is endemic fluorosis in these habitations. Severe dental fluorosis has been observed in Rajasthan, Haryana, and A. P. In 1930, it was observed that the bullocks used for ploughing in A. P. were unable to walk and this is how skeletal fluorosis was identified for the very first time in India. In 1937, some people complained of stiff joints and were unable to walk. According to Bihar Government 8,188 villages in 11 districts have excess fluoride in water (Feb 2010). The districts are Nalanda, Rohtas, Kaimur, Aurangabad, Gaya, Nawada, Bhagalpur, Banka, Munger, Jamui and Sheikhpura. Kishanganj is the only district in Bihar state where tea is grown. Due to lack of awareness and inappropriate use of chemical fertilizers and pesticides by tea growers, the groundwater of rural areas is gradually degrading [16]; [17]. As per literature review, there has been no published report concerning the contamination in groundwater sources in tea garden areas of Kishanganj district. Keeping the above facts in mind, we decided to analyze the fluoride concentration and some physiochemical parameters in different groundwater sources around the tea garden areas in Kishanganj district.

Therefore the objective of this research work is to analyze the potability of groundwater in tea garden areas at Kishanganj block of Kishanganj district with reference to fluoride levels and correlation with some other physiochemical parameters, which will provide information about extensive effect of fluoride contamination in water, sources of contamination and health consequences for population of Kishanganj district.

2. Materials and Method

2.1. Study Area

Kishanganj district is located in the north-east of the state of Bihar, at latitude of 25° 20’ to 26° 30’ north and longitude of 87° 7’ to 88° 19’ east (Figure 1). The total area of it is 1,884 square km. and total irrigated area of 262.2 square km. Kishanganj district, West Bengal, Nepal and Bangladesh are at border line of it. Mahananda, Kankai, Mechi, Ratwa, Ramzan and Donk are major rivers that flow through Kishanganj district. The climate of the study area was humid with maximum temperature 42°C in May-June and minimum of 5°C in January. The average rainfall is 2250 mm of which 80% occurs during monsoon. The climatic conditions of this district are suitable for tea cultivation. The huge potential of tea plantation (Kishanganj, Pothia and Thakurganj blocks) in this district is, as it is on the foothills of the Himalayas and the vicinity of Darjeeling district of West Bengal. Location of study area is as shown in Figure 1.

Tea-gardens in Kishanganj district certainly sound amazing, but it’s a reality. There are acres and acres of lush gardens in Taluka Motihara, Halamala and Belwa Panchayat about 10-12 Kms from Kishanganj city. Tea plantation has made quantum jump in Kishanganj district in the early part of 1990s. Now Kishanganj is only district in Bihar (India), where tea plantation has developed and today, Bihar has been put on the tea map of the country. The development of tea plantation in this district is not to originate very large employment opportunities, but to invert the migration of workers form Bihar and increase the income level of the rural people. Today the green tea leaves grown in the district is in high demand and has thrown a challenge to the states of west Bengal and Assam which are traditional tea growing states of India.

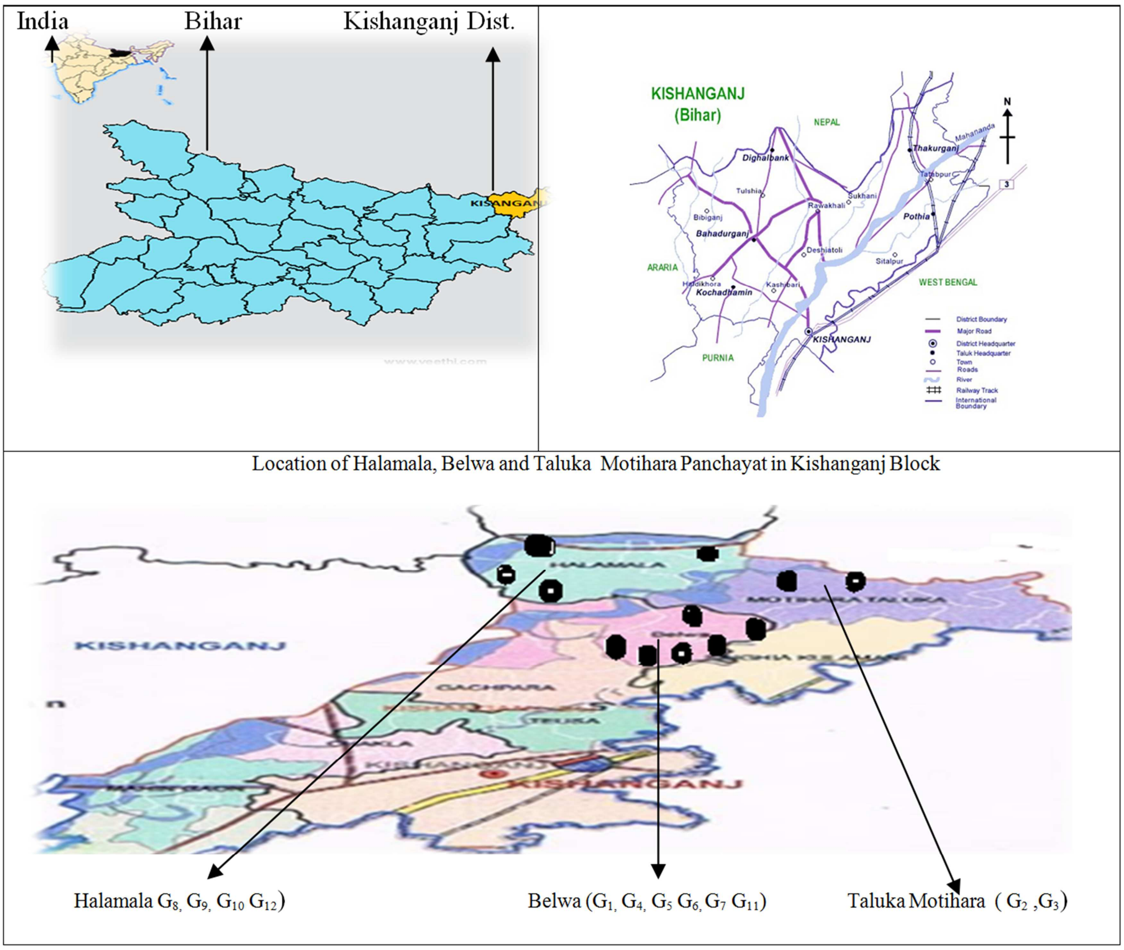

Figure 1. *Location of Study Area.*

2.2. Site Selection and Sample Collection

The present study was designed to determine the physicochemical parameters in groundwater samples of various locations of small tea-gardens in Taluka Motihara, Halamala and Belwa Panchayat of Kishanganj block of Bihar, India

About one liter of groundwater samples were collected from each of the 36 locations (open wells, ring wells, hand pump and tube well) around 12 tea garden areas (3 sites from each tea garden) during 2013 and 2014 of Kishanganj district, in screw capped high density pre cleaned polythene bottles in accordance with described sampling methods [18]; [19] and stored in ice box. Before the collection of samples, tube wells and hand pumps were operated at least 5-10 minutes to flush out the stagnant water inside the tube to get fresh groundwater. Each sample bottle was rinsed twice or thrice with the same water to avoid any impurity from exogenous sources. Preservation of samples was done as per standard procedure (APHA). Sample from each location was collected two times at 30 days interval to determine the average value of the results, which were compared with drinking water standards (BIS and WHO)

2.3. Determination of Physicochemical Parameters

Various physicochemical parameters in groundwater like pH, electrical conductivity, fluoride, chloride, sulfate; nitrate, phosphate, total alkalinity, total hardness, calcium and magnesium were determined by standard procedures as given below.

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°C and pH meter (Model digital pH meter 335) respectively. The specific conductance was measured by a conductivity meter [20]; [21].

Total hardness of water (mainly due to Ca^{++} and Mg^{++}) was analyzed by EDTA titrimetric method in presence of metal ion indicator erichrome black-T (EBT). About 100 mL sample water was first concentrated through evocation to about 5 mL. The carbonate precipitate (thus formed) was dissolved by dilute HCl solution and the CO_2 was boiled off. Then the solution was neutralized with dilute NaOH solution and to the 1 mL of the buffer (prepared by dissolving 17.5 g NH_4Cl in 14 mL of concentrated NH_4OH and then diluted to

250 mL with distilled water) solution and 3-4 drops of EBT indicator solution (prepared by dissolving 0.2g of indicator in 15 mL triethanol amine and 5mL of absolute alcohol) was added. The solution was titrated with 0.01 (M) Na_2H_2 edta [22].

The alkalinity in drinking water is mainly due to the presence of carbonates and bicarbonates. Acidimetric titration first converts CO_3^{2-} to HCO_3^- and in the next step, it converts HCO_3^- to H_2CO_3. The first step was detected in presence of an indicator like phenolphthalein that works in alkaline pH range (>8.2) while the second step was detected by indicator like methyl orange that responds to acidic pH range (>4.0) [23].

Sulfate was measured by gravimetric method. A water sample was filtered in the field through a 0.45 µm membrane filter paper and preserved at 4°C. The sulfate in the sample aliquot was precipitated in an HCl medium as barium sulfate by the addition of barium chloride. After digestion, the precipitate was filtered, washed, ignited, and weighed as $BaSO_4$ [24]; [25]. The chloride content of the water samples was also measured by gravimetric method. In this method chloride anions were precipitated as AgCl by addition of silver nitrate solution in the presence of dilute HNO_3 [26].

Phosphate was measured by acid-extraction method. A water sample was filtered through a glass fibre paper. The filter paper was ignited at low temperature and then the acid was extracted to dissolve the particulates. The extract was treated with ammonium moylbdate and ascorbic acid to form a molybdenium blue complex, which was measured calorimetrically at 660 nm and compared to identically-prepared standard and blank solutions [27].

Nitrate was estimated by using phenol disulphonic acid (PDA) method. 40 mL of water sample was evaporated with 2 mL of the phenol disulphonic acid (PDA) solution and cooled to room temperature. Subsequently 4.0 mL of concentrated H_2SO_4 was added to dehumidify the entire residue and allowed to stand for 10 minutes. The whole solution was then quantitatively transferred to a 100 mL volumetric flask. 10 mL NaOH was added and the volume was adjusted to 100 mL with distilled water. After 10-15 minutes, the absorbance was measured at 420 nm against the blank solution prepared in the same way. The calibration curve was constructed by using the standard solutions of KNO_3 in the concentration range of 2-25 mg/L [28].

The presence of fluoride in drinking water was determined by Ion Selective Electrode (ISE) method described by Rong et al., recommended by APHA (1991). This method utilizes a fluoride selective membrane, which is typically a lanthanum fluoride crystal. The fluoride activity is measured according to the following equation. $E = Eo - RT/F \ln [F^-]$ (Fluoride activity). The method is sensitive to pH. At low pH, fluoride can form hydrofluoric acid, which lowers the measurement. At high pH, hydroxide can also respond to the ISE and increase the measurement. A Total Ionic Strength Adjustment Buffer (TISAB) solution is added to the sample to adjust the solution pH to an optimum value of 5.0 ~ 5.5. The relatively high ionic strength from the TISAB can also minimize the liquid junction potentials and provide constant ionic strength for samples and standards [29]; [30].

Magnesium was estimated by EDTA Titration method. A water sample was filtered in the field through a 0.45 µm membrane filter and preserved at 4°C. The pH of the sample was adjusted to 10.0 ± 0.1 with a buffer (NH_4Cl, NH_4OH and Mg EDTA salt) solution; an indicator (Eriochrome Black T) was added and then slowly titrated with EDTA within five minutes to avoid precipitation. [31].

Calcium was also determined by EDTA Titration method. A water sample was filtered in the field through a 0.45 µm membrane filter paper and preserved at 4°C. The pH of the sample was adjusted between 12 -13, with a 1N NaOH solution to precipitate the magnesium to its hydroxide form. Calver II indicator was added and then titrated with a standardized EDTA solution. The color changed from pink to purple when calcium was removed. The samples were compared to identically-prepared standard and blank solutions [32]. The mean fluoride concentration for each sampling site was compared with the BIS and WHO guidelines for domestic use to assess compliance [33]; [34].

3. Results and Discussion

3.1. Distribution of Fluoride in Groundwater

Fluoride concentration in water sample around tea gardens of Kishanganj district varied from 0.18 - 4.2 mg/L as shown in Figure 2. The minimum concentration (0.18 mg/L) was recorded from 2 sites at S1and S2 of garden no. 1 (Anwar Alam), while maximum concentration (4.2 mg/L) was recorded at S31 and S32 around garden no. 11 (MustakAlam). The present survey reveals that out of 36 water samples in 9 samples (25%) have fluoride concentration 0.18 - 1.0 mg/L, in which fluoride concentration is ≤1.0 mg/L, maximum desirable limit of standard for drinking water recommended by BIS. There is no possibility of fluorosis in these habitations because this concentration of fluoride is beneficial for calcification of dental enamel especially in children below 10 years of age. Once fluoride is incorporated into teeth, it reduces the solubility of the enamel under acidic conditions and thereby provides protection against dental carries [35].

Out of 36 samples in 3 sites (5%) have fluoride concentration above 1.0 mg/L and ≤1.5 mg/L. The maximum permissible limit of fluoride, standard for drinking water is 1.5 mg/L as recommended by WHO. The population of these habitations, fluoride intake through drinking water is more than 4 mg/day in an individual. Therefore, an incidence of the first dental fluorosis is possible in local residents of this habitation [36].

15 locations (41%) have groundwater with fluoride concentration above 1.5 mg/L and ≤3 mg/L, which is above maximum permissible limit as recommended by BIS (1991) and WHO (2006). At this concentration, teeth lose their shiny appearance and chalky black, grey or white patches develop known as mottled enamel [37]. In some cases, the pre stage

of skeletal fluorosis may occur after age of 45 [38]. In 7 sites (19%) fluoride concentration in groundwater is above 3.0 mg/L and ≤ 4 mg/L. The intake of fluoride /day by the population in this habitation is very high and cause dental as well as skeletal fluorosis [35]; [36]. 3 locations around the tea garden, fluoride content was found more than 4 mg/L which may be the cause of II-grade of skeletal fluorosis i.e. restricted movement of joints due to stiffness. Among 36 sites around the tea garden of Kishanganj district 11 (30.0%) sites are safe, 15 (42.0%) locations under dental fluorosis and 10 (28.0%) sites are under skeletal fluorosis (Fig. 3).

The persistence of high concentration of fluoride in drinking water at other areas of Kishanganj district has been reported in our previous work [5]. High level of fluoride in West Bengal adjacent to Kishanganj district has also been reported by several researchers [39-41]. It is noted that fluoride in fluoride affected areas and other areas adjacent to Kishanganj district is in the same geological set up. It is assumed that fluoride bearing minerals may contribute high fluoride content in groundwater during the course of rock type minerals such as fluorspar, fluorite, apatite, rock phosphate and phosphites. The use of heavy amount of chemical fertilizers, weedicides, pesticides and other chemicals by tea growers contribute high fluoride concentration in water resources. It has been found that in some tea garden areas the concentration of fluoride is low, due to dilution by rain water, as this area is a rain shadow area within North-East India.

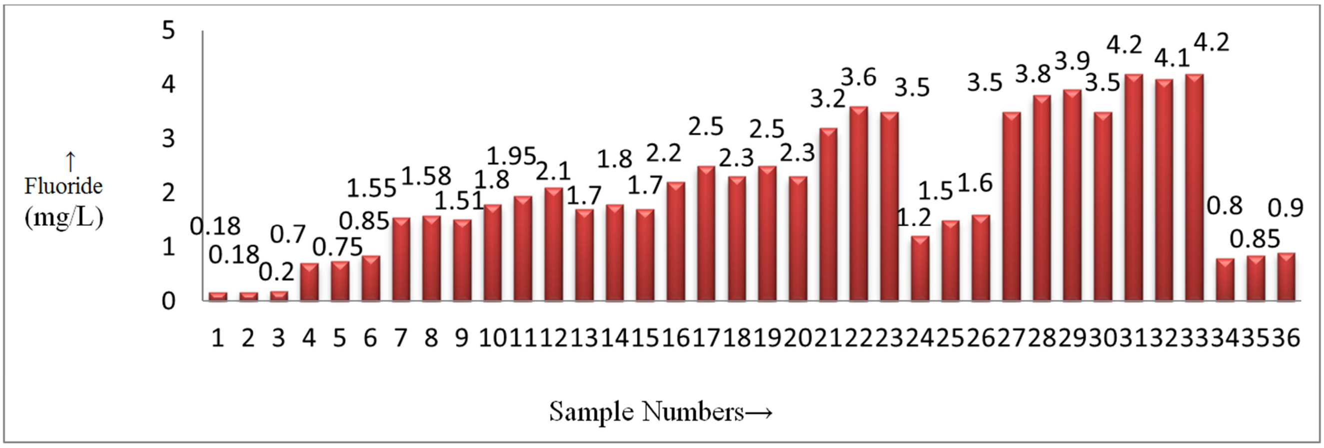

Figure 2. *Fluoride concentration in water sample around tea gardens.*

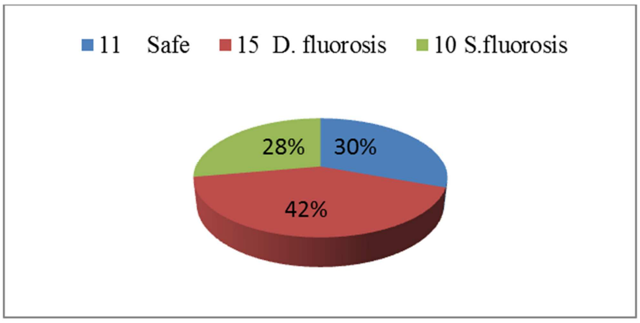

Figure 3. *Fluoride toxicity in the water samples around tea gardens.*

The correlation study revealed that high level of fluoride in water was associated with low calcium content and high pH values. The high pH of water displaces fluoride ions from mineral surface [42]. From correlation study it was observed that Ca^{2+}, Mg^{2+} and total hardness showed negative correlation with fluoride. This was probably due to low solubility of fluoride with these ions [43] and negative correlation of TH with fluoride because the total hardness was due to the presence of calcium and magnesium carbonate and bicarbonate. The decrease in hardness, resulting in higher fluoride concentration contributed to calcium complexion effect. Nitrate and phosphate ions also showed negative correlation with fluoride in groundwater. The phosphate ion in the water samples was due to anthropogenic activities. geogenic formation and dilution of minerals from rock and soil [43]. The drinking water contaminated by nitrate ion in water samples resulted from the leaching of nitrate present on the surface of percolating [44].

Fluoride showed positive correlation with pH, EC, Cl^{1-}, SO_4^{2-} and alkalinity. The correlation coefficient were 0.94, 0.95, 0.89, 0.97and 0.88 respectively. Chloride in water sources resulted from agricultural activities, industries and chloride rich rocks. High chloride concentration was due to invasion of domestic waste and disposal by human activities [45]. High chloride concentration in drinking water may lead to laxative effect [46]. Agricultural activities were also sources of high SO_4^{2-} ion in groundwater in and around tea garden areas. High concentration of fluoride in groundwater may also be the cause of increase in solubility of CaF_2 with increase of TA, according to the following equations

$$CaF_2 + CO_3^{2-} \rightarrow CaCO_3 + 2F^-$$

$$CaF_2 + 2HCO_3^- \rightarrow CaCO_3 + 2F^- + H_2O + CO_2$$

3.2. Physical and Chemical Controls

The mean value of physicochemical parameters of ground water samples collected around the tea garden areas with BIS and WHO standards is presented in Table 1.

3.2.1. Correlation Between pH and Fluoride

The pH values ranged from 6.4-8.6 is as shown in Figure 4a. The results revealed that 9 (25%) water samples had pH value in between 6-7, this may be due to use of fertilizers ammonium sulfate and supper phosphate by tea growers, 15 (41.7%) water samples have pH in between 7-8, while 12 (33.3%) water samples have pH values more than 8. Due to weathering and leaching of fluoride minerals in rock formation under alkaline environments lead to the increase of fluorite in the groundwater [47]. When pH >8, fluoride concentration commonly exceed 1.0 mg/L. 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 [48]. Alkaline pHs which increased the solubility of fluoride-bearing formations likely contributed to increased fluoride concentration at S29 (3.9 mg/L), of garden no. 10 (Sanjay Thakur) and at S31 (4.2 mg/L), at S32 (4.1 mg/L), and at S33 (4.2 mg/L) of garden no. 11 (Mustak Alum). The value of pH showed the positive correlation (r = 0.94) with fluoride as shown in Figure 4b. Similar positive correlation was also shown by several researchers [49]; [50].

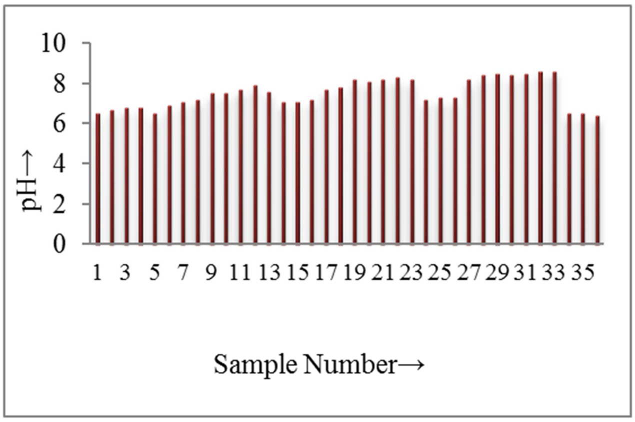

Figure 4a. *Variations of pH.*

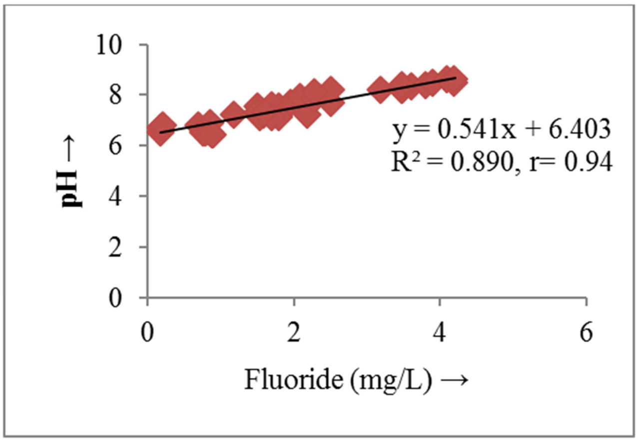

Figure 4b. *Correlation between pH and fluoride.*

3.2.2. Correlation Between EC and Fluoride

The values of electrical conductivity varied from 308-542 µS/cm. The maximum value of 542 µS/cm was found at S32 and S33 of garden no. 11 (MustakAlam) whereas minimum 308 µS/cm at S1 of garden no. 1 (Anwar Alam) (Fig. 5a). All the locations had low value of EC than permissible limit as recommended by WHO standard of 600 µS/cm. Figure. 5b shows positive correlation of EC with fluoride (r = 0.95).

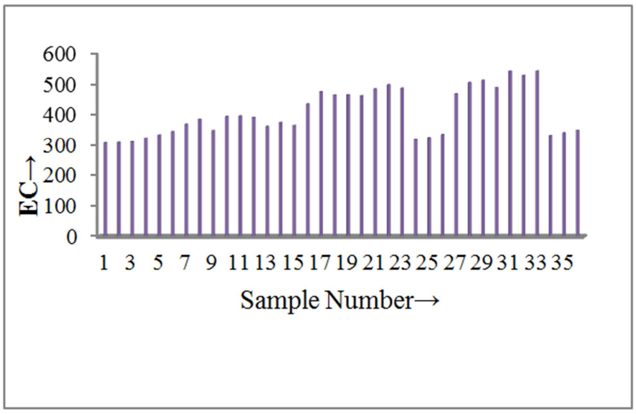

Figure 5a. *Variations of EC.*

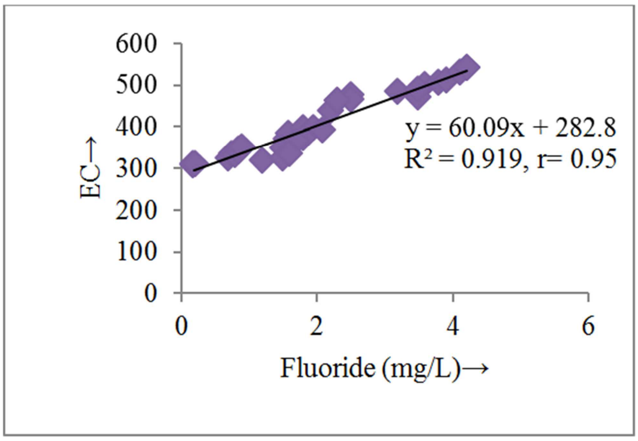

Figure 5b. *Correlation between EC and fluoride.*

3.2.3. Correlation Between TH and Fluoride

The value of total hardness in water around the tea gardens varied from 115-402 mg/L. The maximum value of 402 mg/L was found at S2 of garden -1 (Anwar Alam) and minimum value 115 mg/L was found to be at two locations S31 and S32 of garden no. 11 (Mustak Alam) (Fig. 6a). The results revealed that out of 36 samples only 10 (27.8%) locations had TH values above the desirable limit (300mg/L) as recommended by BIS. The data as shown in Figure 2 and 6a showed that concentration of fluoride was less than 1.5 mg/L for most of the sampling stations whereas value of TH was less than 200 mg/L [51]. The TH showed negative correlation (r = - 0.867) with fluoride (Fig. 6b). The dissolved salts and minerals present in the soil around tea gardens might increase the concentration of total hardness in water bodies near the gardens. The uses of chemical fertilizers by tea growers directly or indirectly affected the concentration of inorganic chemicals in water which effected the concentration of TH in water [49]. The total hardness showed positive correlation with Ca^{2+} and Mg^{2+} ions as shown in Figure 9.

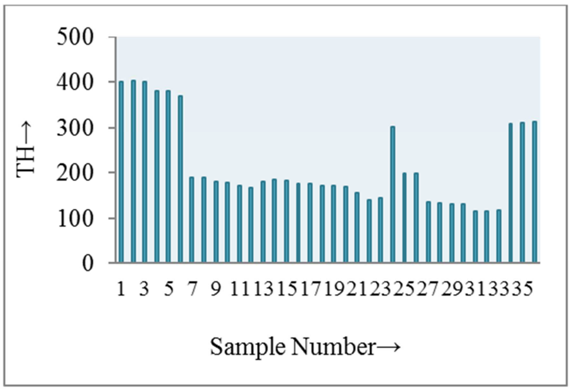

Figure 6a. *Variations of TH.*

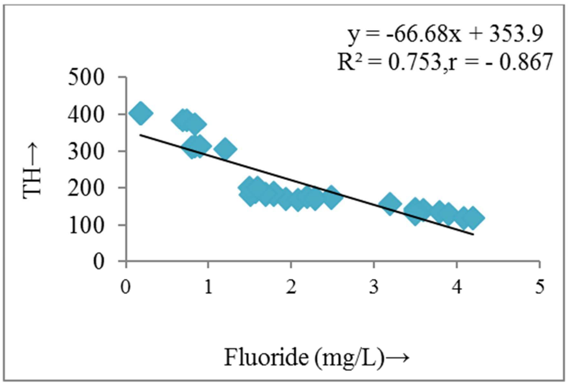

Figure 6b. *Correlation between TH and fluoride.*

3.2.4. Correlation Between Calcium and Fluoride

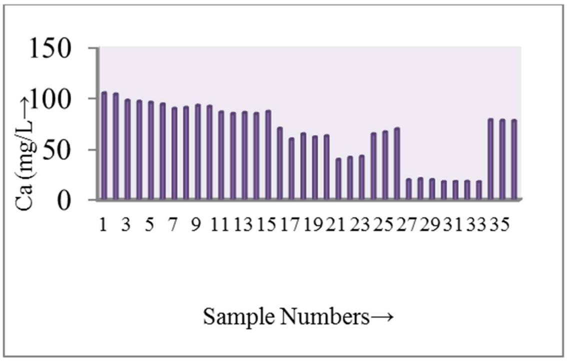

Figure 7a. *Variations of Ca^{2+} ion.*

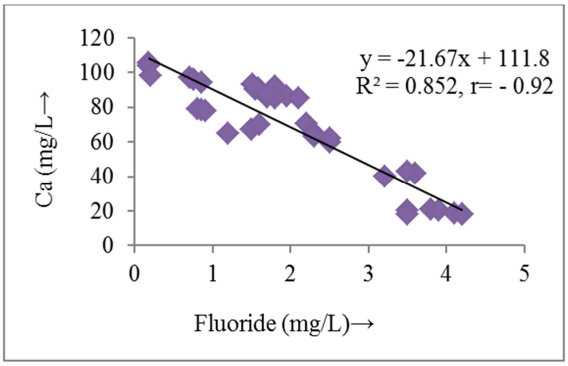

Figure 7b. *Correlation between Ca^{2+} and fluoride.*

The concentration of Ca^{2+} ions ranged from 18-105. 2 mg/L (Fig. 7a) which was below the maximum permissible limit 200 mg/L as recommended by BIS. The maximum value 105.2 mg/L was found at S1 in the garden no. 1 (Anwar Alam) and minimum 18 mg/L was found S32 in garden no. 11 (Mustak Alam). Figure7b showed negative correlation of calcium with fluoride (r = - 0.92) which was supported by various researches [49]; [50]. Lower concentrations of calcium increased the solubility of CaF_2, with increase in the concentration of fluorides in groundwater. The reaction between Ca^{++} and F^- ions indicated that the fluoride concentration was controlled by equilibrium of fluorite [52]. When the water was saturated with respect to fluorite, low calcium concentration lead to higher fluoride concentration [53]. Higher fluoride concentration was therefore expected in groundwater from aquifers with low calcium concentrations. The results generally showed that increase in calcium was associated with decrease in fluoride and vice versa.

3.2.5. Correlation Between Magnesium and Fluoride

The magnesium content varied from 11.3-55.2 mg/L (Fig. 8a) which was lower than maximum permissible limit of BIS (100 mg/L). The negative correlation between Mg^{2+} and F^- ions (r = - 0.97) as shown in Figure 8b might be the due to low solubility of fluoride of these ions [54].

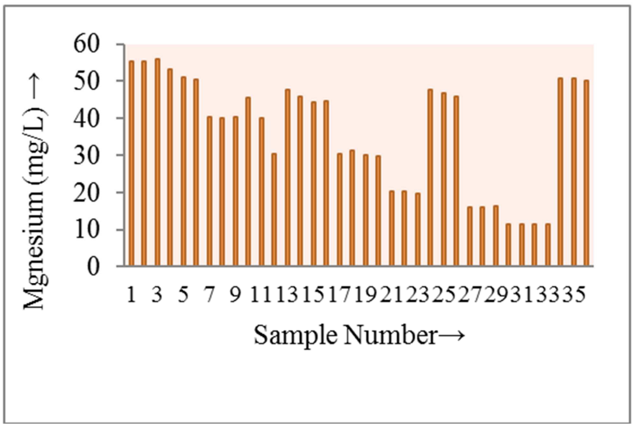

Figure 8a. *Variations of Mg^{2+} ion.*

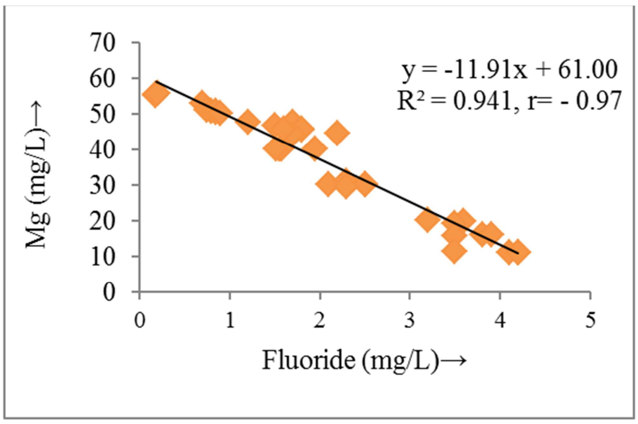

Figure 8b. *Correlation between Mg^{2+} and fluoride.*

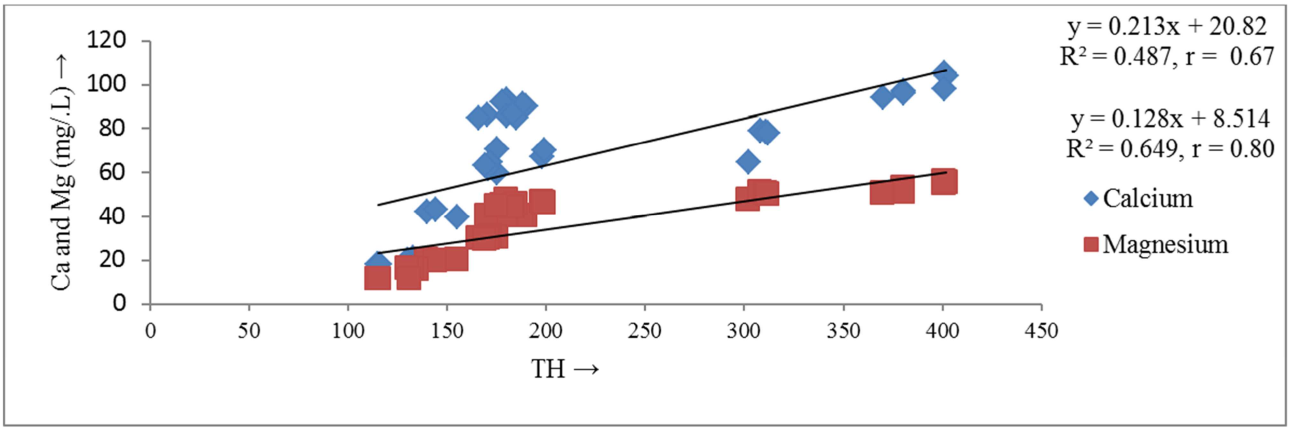

Figure 9. *Graph showing Correlation of TH with Ca^{2+} and Mg^{2+} ion.*

3.2.6. Correlation Between Nitrate and Fluoride

The range of nitrate content varied from 3.4-8.7 mg/L (Fig. 10a) which was below the permissible limit (45 mg/L) of WHO and BIS. The highest concentration of nitrate (8.7 mg/L) was recorded in the two sites S1 and S3 of garden no. 1 (Anwaer Alam) and lowest 3.4 mg/L was observed in the sample no. S33 of garden no. 11 (Kadir Alam). The occurrence of nitrate content near the water samples of the tea gardens might be due to leaching into the water sources from chemical fertilizers used in the tea gardens [54]. The presence of nitrate in water around the gardens showed negative correlation (r = - 0.95) with fluoride (Fig. 10b). Nitrate also showed negative correlation with Mg^{2+} ion which supported the persistence of magnesium nitrate in groundwater [55].

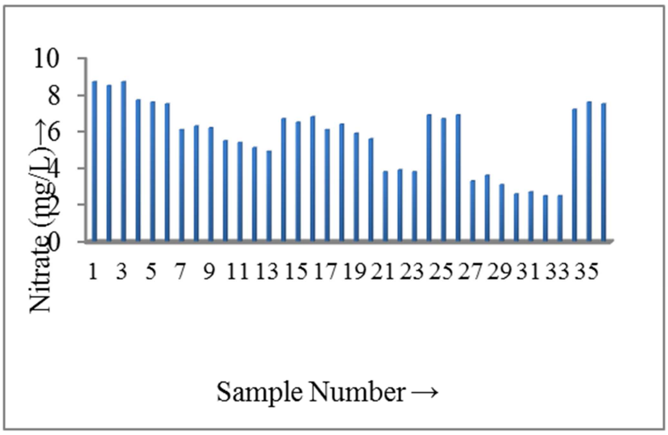

Figure 10a. *Variations of NO_3^- ion.*

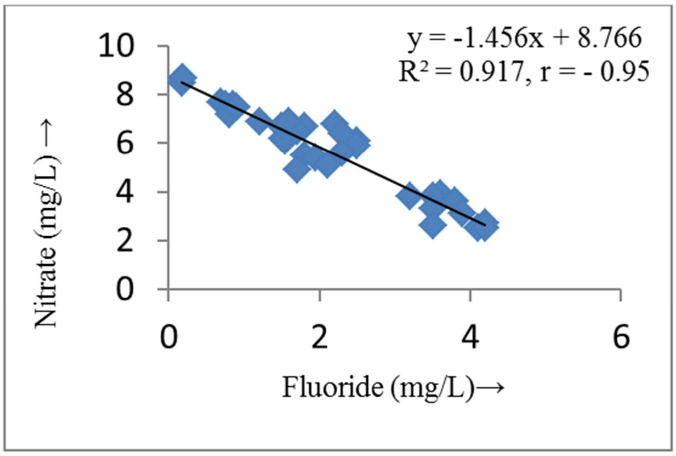

Figure 10b. *Correlation between NO_3^- and fluoride.*

Table 1. *Mean value of Physicochemical Parameter with BIS and WHO.*

Parameter	Garden-1	Garden-2	Garden-3	Garden-4	Garden-5	Garden-6	BIS (DL)	WHO (PL)
F (mg/L)	0.187	0.767	1.547	1.95	1.73	2.33	1	1.5
pH	6.67	6.73	7.27	7.7	7.27	7.57	6.5-8.5	7.0-8.5
EC (µS/Cm)	309.3	334	367.3	394	367.3	458	----	600
TH (mg/L)	401.3	376.7	186	171.3	182.7	174	300	----
Ca^{2+} (mg/L)	102.4	95.77	91.33	87.83	86	65.13	75	----
Mg^{2+} (mg/L)	55.43	51.47	40.2	38.57	45.93	35.37	30	----
NO_3^- (mg/L)	8.63	7.6	6.2	5.33	6.03	6.43	45	45
PO_4^{3-} (mg/L)	0.903	0.837	0.657	0.563	0.487	0.373	---	5
Cl^- (mg/L)	10.57	12.93	45.57	41.87	56.2	73.87	250	250
SO_4^{2-} (mg/L)	2.63	5.72	10.22	11.61	11.67	12.9	200	500
TA (mg/L)	236.7	268.3	279	308.33	263.3	377.7	200	600

Table 1. *Continue.*

Parameter	Garden-7	Garden-8	Garden-9	Garden-10	Garden-11	Garden-12	BIS (DL)	WHO (RL)
F (mg/L)	2.66	2.77	2.2	3.73	4.17	0.85	1	1.5
pH	8.17	7.9 0	7.6	8.43	8.56	6.46	6.5-8.5	7.0-8.5
EC (µS/Cm)	470.3	434.3	376	501.7	537.3	340.7	----	600
TH (mg/L)	165	195.5	177.3	131.3	115.3	310.3	300	----
Ca^{2+} (mg/L)	55.03	50	52.33	19.67	18.2	78.43	75	----
Mg^{2+} (mg/L)	26.67	29.6	36.13	14.5	11.33	50.57	30	----
NO_3^- (mg/L)	5.1	4.87	5.63	3.1	2.57	7.433	45	45
PO_4^{3-} (mg/L)	0.297	0.313	0.43	0.213	0.113	0.613	---	5
Cl^- (mg/L)	73.87	79.7	69.03	115	147.3	46.6	250	250
SO_4^{2-} (mg/L)	17.05	16.8	13.45	22.33	25.13	41.66	200	500
TA (mg/L)	419.3	378	335.7	442.3	679	271.3	200	600

DL- Desirable limit, PL-Permissible limit

3.2.7. Correlation Between Phosphate and Fluoride

The variation of phosphate content in all 36 water samples (0.11-0.91mg/L) was recorded which is shown in Figure 11a. The phosphate content in all locations was found below the permissible limit. The maximum value of phosphate 0.91mg/L was found in the two samples S2 and S3 of garden no. 1 while minimum value 0.11mg/L was recorded in sample no S31and S32.

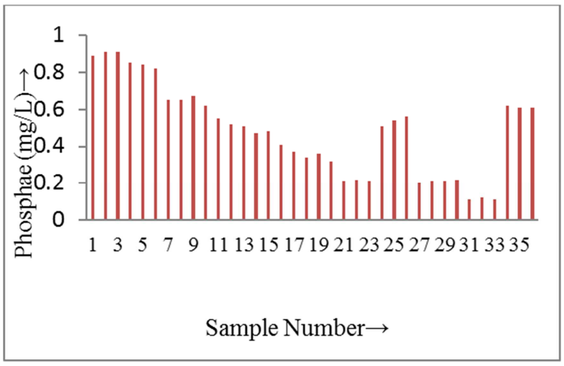

Figure 11a. *Variations of PO_4^{3-} ion.*

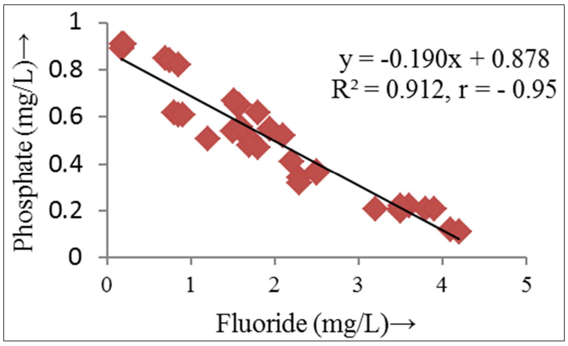

Figure 11b. *Correlation between PO_4^{3-} and fluoride.*

The persistence of phosphate in water system around the tea gardens was due to phosphatic fertilizers used by tea growers and decomposition of organic substances near the tea gardens [54]; [55]. The presence of phosphate showed negative correlation (r = -0.95) with fluoride (Fig. 11b).

3.2.8. Correlation Between Chloride and Fluoride

Variation of chloride in all analyzed samples was under the permissible limit of 250 mg/L as recommended by WHO. The maximum concentration of chloride 150 mg/L was observed at S30 of garden no. 10 (Sanjay Thakur) and minimum value (10.5 mg/L) at two sites S1and S2 of garden no. 1 (Fig. 12a). The presence of Chloride in water around the gardens showed positive correlation (r = 0.89) with fluoride as shown in Figure 12b.

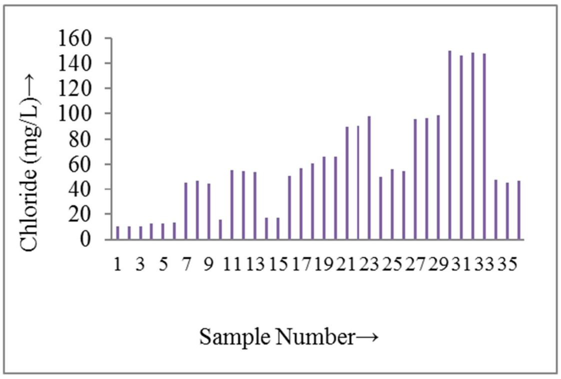

Figure 12a. *Variations of Cl^- ion.*

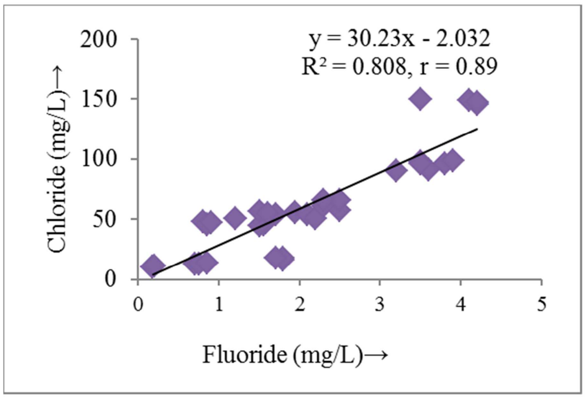

Figure 12b. *Correlation between Cl^- and fluoride.*

3.2.9. Correlation Between Sulfate and Fluoride

The range of sulfate concentration varied from 2.45-25.7 mg/L (Fig. 13a). The result revealed that concentration of sulfate in all sampling stations was below the desirable limit of 200 mg/L (BIS). The maximum value of 25.7 mg/L was found at S32 of garden no. 11 and minimum value 2.45 mg/l at S1 of garden no. 1. Figure 13b showed that positive correlation (r = 0.97) with fluoride ions. Due to oxidation of sulfur compounds used by tea growers was the cause of sulfate in water samples [55].

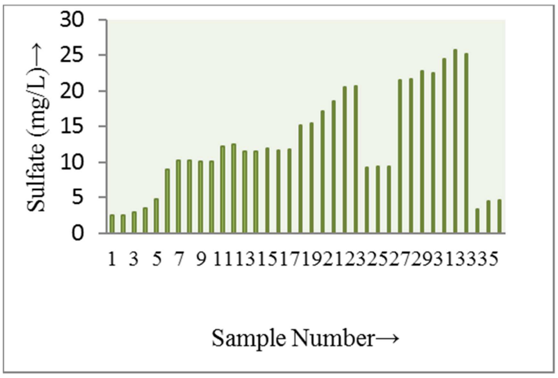

Figure 13a. *Variations of SO_4^{2-} ion.*

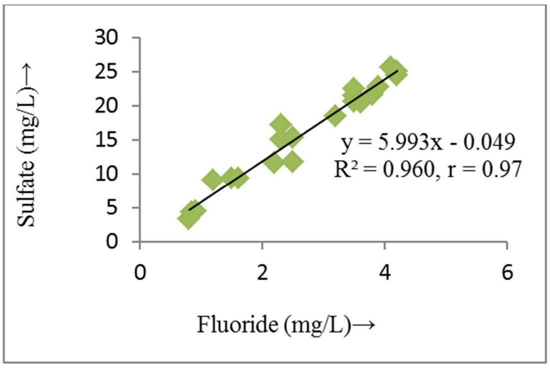

Figure 13b. *Correlation between SO_4^{2-} and fluoride.*

3.2.10. Correlation Between TA and Fluoride

Most of the sampling stations of the study area analyzed revealed the total alkalinity varied from 231-688 mg/L (Fig. 14a), higher than desirable limit of 200 mg/L while slightly more than permissible limit of 600 mg/L as set by WHO**.** Maximum value of alkalinity (688 mg/L) was recorded at S33 of garden no. 11 and minimum value (231 mg/L) was found at S1 of garden no. 1. Alkalinity showed positive correlation (r = 0.88) with fluoride (Fig. 14b). Positive correlation of TA with fluoride ions may increase the solubility of CaF_2 with increase in concentration of TA [56]. The dissolution of polyvalent metallic ions from soil minerals and sedimentary rock might also be the cause of persistence of alkalinity in groundwater of locations S32, S33 and S34 around the tea garden.

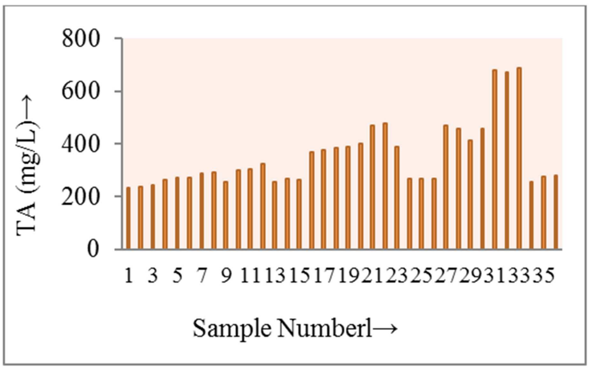

Figure 14a. *Variations of TA.*

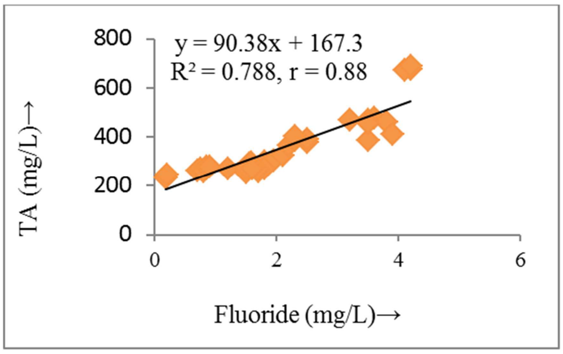

Figure 14b. *Correlation between TA and fluoride.*

3.3. Evaluation of Impact of Fluorides on Human Health Based on Questionnaire

The present survey reveals that out of 600 people, 200 people from each panchyat near the tea-gardens, Belwa, Halamaala and Taluka Motihara were examined. Among the different clinical signs and symptoms of dental fluorosis, 184 (30.7%) people were suffering of grade-I had faint and yellow teeth, 83 (13.8%) people of grade-II had brown stained teeth, 40 (6.7%) people of grade-III had brown pitches on teeth and 18 (3.0%) people of grade-IV had loss of teeth, whereas 275 (45.8%) people were having healthy teeth as shown in Table 2. Similarly skeletal fluorosis was also found on the basis of different clinical signs and symptoms, 38 (6.3%) people of grade-I had joint pains, 8 (4.0%) of grade-II had joint and back pains, 10 (1.7%) of grade-III had difficulty to walk and 9 (1.5%) people of grade-IV had knock - knees and were in bed ridden state as shown in Table 3. The result also revealed that out of 600 people 325 (54.2%) people were affected by dental fluorosis among which 177 (29.8%) were males and 148 (24.6%) were females, whereas 65 (10.83%) people were suffering from skeletal fluorosis among which 41 (6.83%) were males and 24 (4.0%) were females. All the individuals who were affected with fluorosis had resided in the respective villages since birth. The occurrence of mild grade of skeletal fluorosis like joint and back pains was more prevalent in males, whereas knock-knees were equally prevalent both among males and females

Dental fluorosis of different grading:

Grade 0: Normal, translucent, smooth, glossy teeth

Grade I: White opacities, faint and yellow line

Grade II: Changes as in Grade I with brown stains

Grade III: Brown line, pitting and chipped off edges

Grade IV: Brown black and/ loss of teeth.

Skeletal fluorosis of different grading:

Grade I: Mild-Generalized bone and joint pain,

Grade II: Moderate-Generalized bone and joint pain, Back pains and Stiffness of back

Grade III: Stiffness and rigidity, restricted movement of the spine and joints

Grade IV: Deformities of spine and limbs, knock-knees, crippled or in bed ridden state

Table 2. *Prevalence of dental fluorosis.*

Panchayat	Total No. of Survey	Affected cases (%)		Dental fluorosis (%)				Occurrence
		Male	Female	Grade- I	Grade- II	Grade-III	Grade -IV	(%)
Belwa	200	62 (31.0)	53 (26.5)	63 (31.5)	29 (14.5)	16 (8.0)	7 (3.5)	57.5
Halamala	200	59 (29.6)	49 (24.5)	61 (30.5)	28 (14.0)	13 (8.7)	6 (3.0)	54
Taluka Motihara	200	56 (28.0)	46 (23.0)	60 (30.0)	26 (13.0)	11 (5.5)	5 (2.5)	43.5
Total	600	177 (29.5)	148 (24.6)	184 (30.7)	83 (13.8)	40 (6.7)	18 (3.0)	54.2

Table 3. *Prevalence of skeletal fluorosis.*

Panchayat	Total No. of Survey	Affected cases (%)		Skeletal fluorosis (%)				Occurrence
		Male	Female	Grade- I	Grade- II	Grade-III	Grade -IV	(%)
Belwa	200	15 (7.8)	09 (4.5)	14 (7.0)	3 (1.5)	4 (2.0)	3 (1.5)	12
Halamala	200	14 (7.0)	08 (4.0)	13 (6.5)	3 (1.5)	3 (1.5)	3 (1.5)	9.5
Taluka Motihara	200	12 (6.0)	07 (3.5)	11 (5.5)	2 (1.0)	3 (1.5)	3 (1.5)	9.5
Total	600	41 (6.83)	24 (4.0)	38 (6.3)	8 (4.0)	10 (1.7)	9 (1.5)	10.83

From the present study, the overall prevalence of dental fluorosis was found to be 54.2%. Earlier studies have proved the direct link between the degree of dental fluorosis and the amount of fluoride in groundwater in the respective communities and countries. Choubisa [57] reported prevalence of dental fluorosis to be (45.7%), Bharthi et al. [58] (35%), Shourie [59] (36.5%), Vacher [60] (51.57%), Thaper [61] (59.1%), Ramchandran et al. [62] (66.2%) and Tiwari and Chawala [63] (81.6%). Similarly the prevalence of skeletal fluorosis was found to be 10.83%. Several studies in the past have also showed the effect of fluoride as skeletal fluorosis. Choubisa [57] (22%), Bharthi et al [58] (17%) and Majumdar [64] (23.8%), showed various degree of skeletal fluorosis

The study revealed that rock water interaction is the major source of fluoride in groundwater and very much influenced by local lithology. Besides the weathering processes, anthropogenic activities (usage of chemical fertilizers and agricultural pesticides in tea gardens) also play a significant role in the occurrence of fluoride in groundwater in study areas. The samples (S21, S22, S23, S27 S28, S29, S30, S31, S32, S33) in and around tea garden are the evidences to excess fluoride concentration in groundwater.

4. Conclusion

Finally it is concluded that in the present study, fluoride concentration varied from 0.18-4.2 mg/L in different groundwater resources near tea gardens of Kishanganj district. Linear correlations of fluoride with other physicochemical parameters were very useful to get fairly accurate idea of the quality of drinking water. The continuous and uncontrolled use of different chemicals in the tea garden areas of the region might increase the pollution rate which may lead to cause an adverse health effect to the tea garden community. So, awareness among the small scale tea growers and workers is to be imparted regarding use of fertilizers and pesticides. Questionnaire survey results generally indicated that majority of residents of the tea gardens were suffering from dental and skeletal fluorosis due to unawareness and lack of precautionary measures against the, impact of fluoride. Therefore this work will be helpful for students, teachers, scientists, researchers and governmental and non governmental agencies to take correct and appropriate steps by creating awareness among the illiterate people and make them free from fluoride borne diseases.

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