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Monitoring of Fluoride Concentration In Ground Water of Small Tea Gardens in Sonitpur District, Assam, India: Correlation with physico-chemical Parameters

Joydev Dutta[1]*, Manoranjan Nath[2], Mridual Chetia[3],Abani Kr.Misra[4]

[*1]Department of Chemistry, Chaiduar College, Gohpur, Sonitpur, Assam, India,784 168

[2]North Eastern Regional Institute of Water and Land Management Tezpur, Sonitpur, India

[3]Defence Research Laboratory, Tezpur ,Sonitpur, Assam, , India

[4]Department of Chemistry, Gauhati University, Guwahati, Assam-781014, India

Corres.author: joydevghp@rediffmail.com
Ph. No. 03715292822 ; Mob. No. 6706372670

Abstract: Fluoride is very much essential for healthy growth of teeth and bones if it present between 0.6 ppm. to 1.5 ppm. in drinking water but if the level is higher than 1.5 ppm than causes dental and skeletal fluorosis, decalcification, digestive and nervous disorders occurs but less than 0.6 ppm. bones and teeth will be fragile. Fluoride concentrations in water samples (Surface and ground) were determined in fifteen small tea gardens of Sonitpur district, Assam, India. Fourty five samples were collected and analysed for Fluoride content along with pH, electrical conductivity (E.C), total hardness (T.H), sulphate (SO_4^{2-}), nitrate (NO_3^-), phosphate (PO_4^{2-}), chloride (Cl^-), calcium (Ca^{++}) and magnesium (Mg^{++}). Fluoride concentration is varied from 0.17 ppm. To 5.602 ppm. Fluoride concentration is not uniform of all the tea gardens area. Physicochemical conditions like decomposition, dissociation, subsequent dissolution and agrochemicals are might be responsible for leaching of fluoride in to the drinking water sources. From the correlation analysis F^{2-} contents was found to be negatively correlated with Ca^{2+} , Mg^{2+} and Total hardness and positively correlated with pH, SO_4^{2-} and Cl^-.

Keywords: Fluoride, Correlation, Physico chemical parameters, Water samples, Small tea garden, Sonitpur district.

Introduction

Fluoride is the lightest member of the halogen group of elements. In number of respects its behavior is quite different from other halogens and it is reflected in natural water also. Fluoride is the common element in the earth's crust as component of the rocks and minerals. Fluoride (CaF_2) is a common fluoride material. It is considered as one of the minor constituents of natural waters, but it is an important parameter in ascertaining the suitability of water for potable purposes. Intake of 1mg/l per day is very much essential for healthy growth of teeth, but level higher than the permissible limit of 1.5mg/l is dangerous to health[1]. Fluoride contamination of ground water has now become a major geo-environmental issue in many parts of the world due to its toxic effects even if consumed in trace quantities. Fluoride in ground water poses a great problem in most of the states of India[2,3]. Fluoride concentration in ground water of India varies widely ranging from 0.01ppm to 48ppm[4]. A high fluoride content in drinking water sources have been observed in 15 states of India[5]. Sever contamination of fluoride in ground water of Karbi Anglong and Nagaon districts of Assam, India and its manifestation in the form of fluorosis have been reported recently[6].

Deficiency of fluoride leads to dental caries and higher concentration leads to dental and skeletal

fluorosis[7]. Fluorosis was first reported from India by Short et al., in 1937. Fluoride also circulate in blood and effect foetus, nerves and heart. Fluoride reduces secretion of thyroid gland by affecting iodine in the body which may lead to monogolism. Apart form these, excess fluoride intake will also cause gastro intestinal problems like loss of appetite, nausea, vomiting, pain in abdomen, intermittent diarrohea, muscular weakness, excessive thirst etc[8]. High fluoride intake over a period of time can cripple one for life[9].

It is found that fluoride bearing well waters to be high in sodium and bicarbonate alkalinity and low in calcium hardness. High fluoride containing ground water is chemically distinctive in that, it is soft, has high pH and contains large amount of Silica[10]. The natural ores of fluoride are fluorapatite, cryolite, apatite and fluorospar. Fluorosis in India has surfaced as the varying scourage affecting twenty million people in ten states said Dr. Susheela as president of International Society for fluoride research an Professor at All India Institute of Medical Sciences, New Delhi, Till 1960.

In tea gardens area heavy amount of chemical fertilizers and pesticides, weedicides and other chemicals are used these chemicals have fluoride and other water contaminants substance e.g. fluoride is an impurities in normal and triple super phosphate. Which contaminate the drinking and other water sources. Bhuyan et. al. (2004) studied the water quality in tea gardens of Lakhimpur district, Assam, India[11]. There is no earlier statistics of drinking water contamination in small tea gardens of Sonitpur district Assam, India. So it is very important to monitor the drinking water quality specially fluoride concentration in the drinking water sources in the small tea gardens of Sonitpur district Assam, India.

Experimental

Study area

The district Sonitpur, (second largest district of Assam after Karbi Anglong in area.) which is taken as study area is located in the north east part of Assam. The total area of Sonitpur district is 5103 sq. kms. and lies 100 meter above the mean sea level[12]. It is surrounded by Arunachal Pradesh in north, the Brahmaputra river and Morigaon, Nagaon, Jorhat and Golaghat districts in south, Darrang district in the west, Lakhimpur district in the east and. Sonitpur district is located north bank of river the Brahmaputra within 26°2 and 26°6 N latitude and 92°2 and 93°5 E Longitude[12]. Located between mighty Brahmaputra River and Himalayan foothills of Arunachal Pradesh, the district is largely plain with some hills. Land use in the district is divided primarily among tropical semi evergreen, moist deciduous, riverain forest, grassland agricultural land and tea garden. The temperature ranges from 7°C in January to as high as 38°C in May. Sonitpur District falls in the Sub-Tropical climatic region, and enjoys Monsoon type of climate. Summers are hot and humid, with an average temperature of 29° C. The annual rainfall in the district is 2393mm. The climatic conditions of this area are very suit for the tea cultivation. It is interesting to note that the Monabari tea estate, the biggest tea garden in Asia is situated in Sonitpur district. According to the estimate of 2004, in Sonitpur district itself, there are 62 large tea gardens and 207 registered small tea gardens covering an area of 497.57 hectares[12] and based on this a numbers of small tea industries are growing day by day. The location map of Sonitpur district is shown in figure 1. The tea gardens are C.R.(Chitra Ranjan), A.S.(Adymoni Sarmah), R.P.(RamPrasad), G.D. (Gobinda Dahal), S.P.(Susil Payeng), S.G.(Santanu Goswami), R.U.(Rishiraj Upadhya), D.U.(Dilip Upadhya), B.B.(Bul Bul), B.D.(Bhatri Day), R.S.(Ratul Saikia), Jamuna tea garden, Manjeet tea garden, Shiva tea garden, and Trinayan tea garden.

Sampling methodology

The 45 water samples were collected from different locations of 15 small tea gardens of Sonitpur district. The sources of the water samples were shallow tube wells (hand tube well, Tara pump, mark tube well), ring well, ponds and drains. Tube well was operated at least 10 minutes before collection to flash out the stagnant water inside the tube and to get fresh ground water. The water samples were collected in clean 1L Poly propylene bottles and stored in an ice box[13]. Sample location map are shown in **Figure 1.**

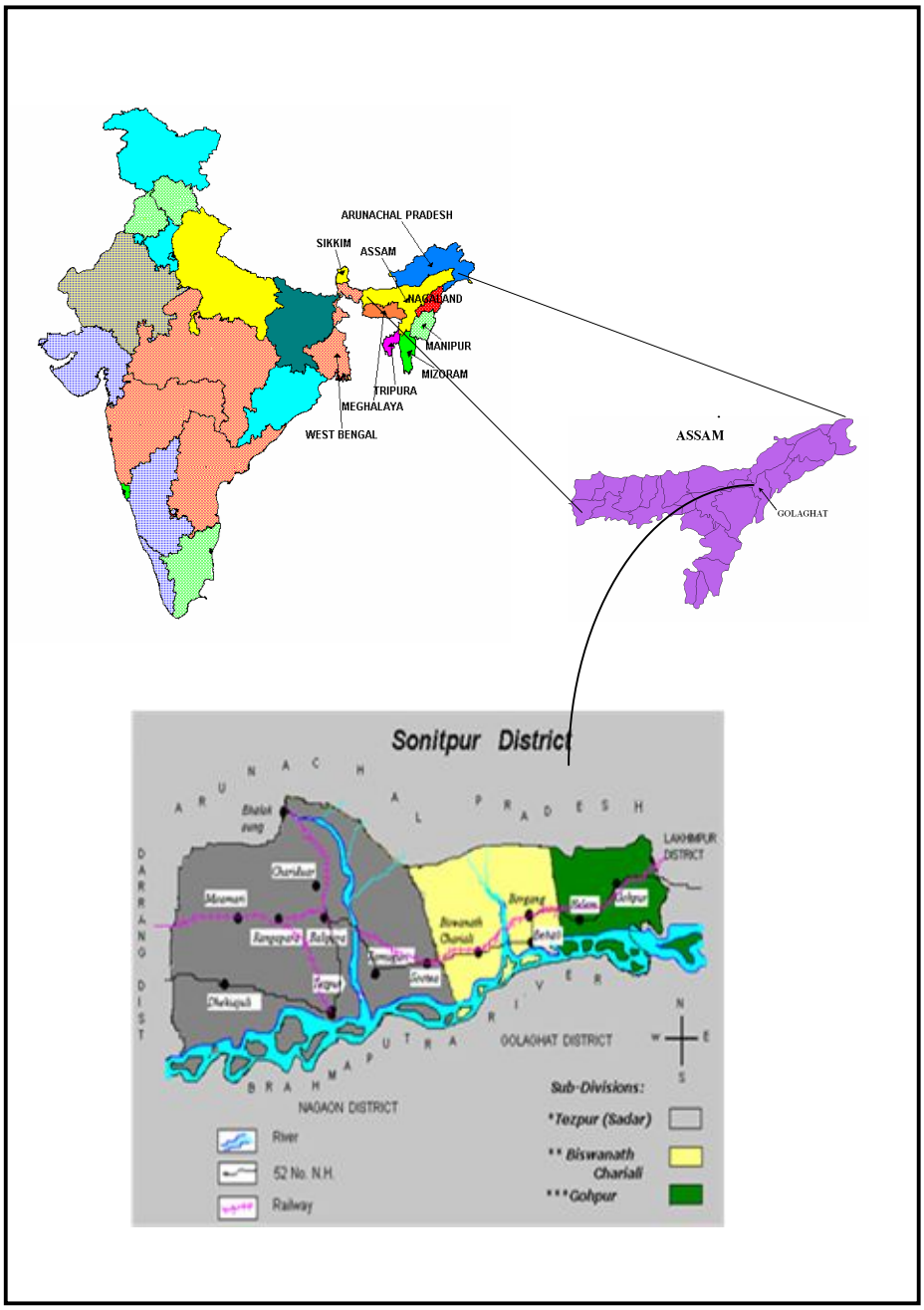


Figure 1. Location map of Sonitpur district.

Sample analysis

pH of the samples was measured at the site of collection by using Pocket pH meter (Merck, India). Fluoride contents were determined by SPADNS method[14]. SPADNS [2-(p-sulphophenylazo)-1, 8-dihydroxy-3,6-naphthalein disulphonate] was obtaine from E-Merck and SRL. Nitrate contents were determined by spectrophotometric method[15]. Sulphate and chloride were determined by turbidimetric and argentometric tritrametric method[16]. Phosphate was determined by molybdenum blue method[17]. For all spectrophotometric determination, SPECORD 40, Analytic Jena (Germany) was used. E.C, Total Hardness were determined by standard method (APHA 1995)[18]. Ca and Mg were determined by EDTA titrametric method.

Result and Discussion

All the water samples collected from different ground water and surface water sources of fifteen small tea gardens of Sonitpur district, Assam, India (Fig.1). The data obtained after analysis of all the samples was compiled and presented in Fig 2, 4 to 12 parameter wise. A total of 15 small tea gardens and from each garden 3 water samples is taken in the study area they are fro well water, surface water(pond, drain etc.) and deep tube well. Fluoride concentration in the study area is depicted in Fig.2. Fluoride concentration in the

study area varied from 0.17ppm to 5.602 ppm. Out of 45 samples above 1.5 ppm fluoride (above WHO guide line) is 17.7%, 24.4% is deficient of (below 0.6 ppm WHO)[19] fluoride and between 0.6 ppm to 1.5 ppm fluoride is 55.55%. the tea garden S.P (Susil Payeng) which have highest fluoride contents all the well, surface and deep tube well (4.926ppm, 3.136ppm, 5.602ppm) then Shiva tea garden (1.8ppm, 3.5ppm, 5.21ppm) then R.P (Ram Prasad) (1.71ppm, 1.7ppm, 1.7ppm), and the lowest fluoride contents is C.R (Chitra ranjan Upadhaya) (0.2ppm, 0.17ppm, 0.23ppm). Occurrence of high fluoride in ground water of other areas adjacent to Sonitpur district can be noted here. High fluoride level in ground water has been reportrd in Darrang district[20], Nagaon and Karbianlong district[21]. It is interesting that the entire fluoride fluoride-affected region is in the same geological set-up[22]. It seems more appropriate that rocks rich in fluoride minerals have contributed to the enriched fluoride content of ground water during the course of weathering of rock types fluorspars, rock phosphate and phosphites. In general relatively high pH conditions have a tendency to displace fluoride ions from the mineral surface[23]. From the correlation analysis it was observed that high fluoride concentration in water associated with low calcium concentration and high pH values. This correlation was also findings of Sanjay Kumar et al[24]. In these gardens synthetic agrochemicals is also used which increase the fluoride contents in the water sources. Modern agriculture practice, which involves the application of fertilizer coupled with pesticides contributes fluoride to ground water[25,]. In some small tea gardens the fluoride concentration in the water sources is deficient than the WHO guide line this is probably due to dilution by rain water, because this is a rain shadow area within North-east India[26].

The correlation of some selected ions and other parameters with fluoride is shown in Fig.3. The Ions Ca^{2+}, Mg^{2+} and Total Hardness showed negative correlation with fluoride contents. The correlation coefficients were -0.330, -0.109, and -0.280, respectively. The negative correlation of fluoride withCa^{2+} and Mg^{+} is as expected due to low solubility of fluorides of these ions[27]. However Chakraborti et al[21] reported positive correlation of fluoride with Ca^{2+} in ground Nagaon and Karbi Anglong districts of Assam. The positive correlation observed by these authors may be attributed to the presence of lime stone in those areas[22]. Total hardness show negative correlation with fluoride because total hardness is due to the sum of calcium and magnesium carbonate and bicarbonate . Generally water with fluoride more than 1.5ppm. has hardness less than 200ppm[23]. Which was found true in the present study. The phenomenon of decrease in hardness concentration contributing to higher fluoride concentration may be attributed to calcium complexing effect. Fluoride shows positive correlation with sulphate in Fig 3 (D). Phosphate show negative correlation with fluoride correlation coefficient is -0.211 and chloride shows positive correlation with fluoride and the correlation coefficients is 0.147.

Fig.2. Fluoride Concentrations in the water samples (dotted line indicates the permissible limit).

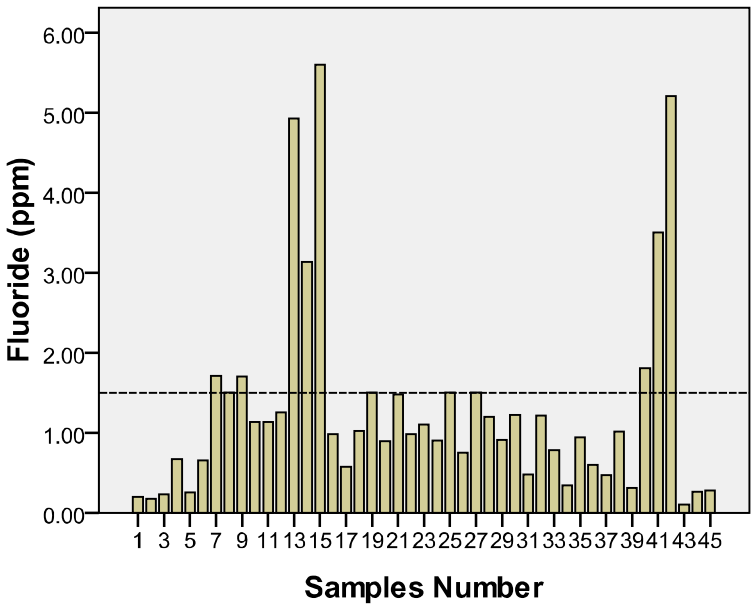
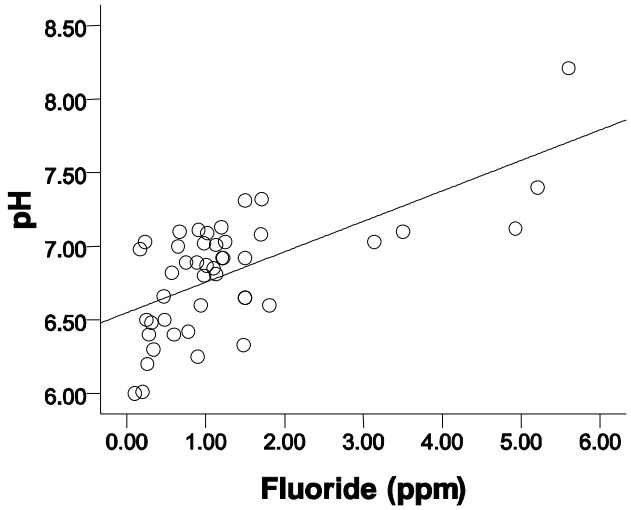
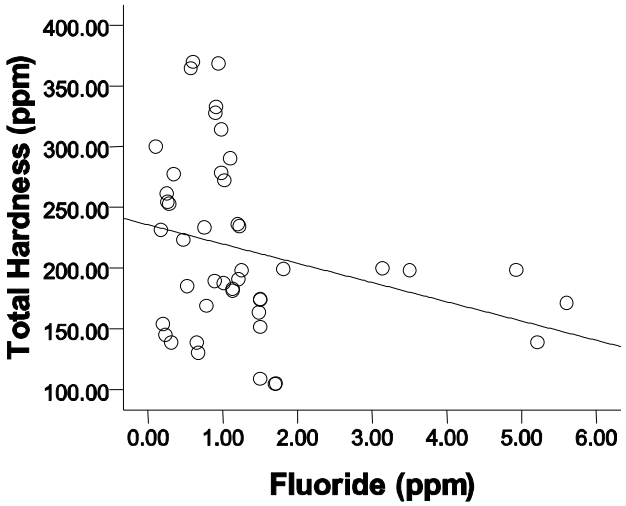


Fig.3. Correlation of different parameters and ions with fluoride concentration in water samples of selected small tea gardens (A) pH vs F^- , (B) E.C vs F^- ,(C). T.H. vs F^- ,(D) SO_4^{2-} vs F^- ,(E) NO_3^- vs F^- ,(F). PO_4^{2-} vs F^- , (G). Cl^- vs F^- ,(H) Ca^{2+} vs F^- , (I) Mg^{2+} vs F^- . The solid lines indicate trends.

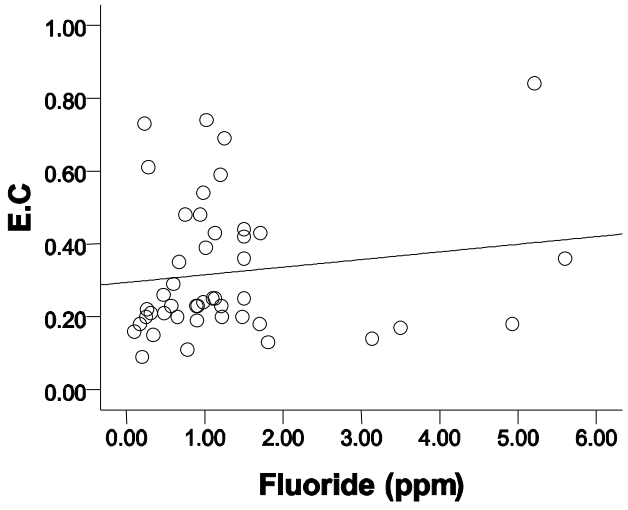
(A).



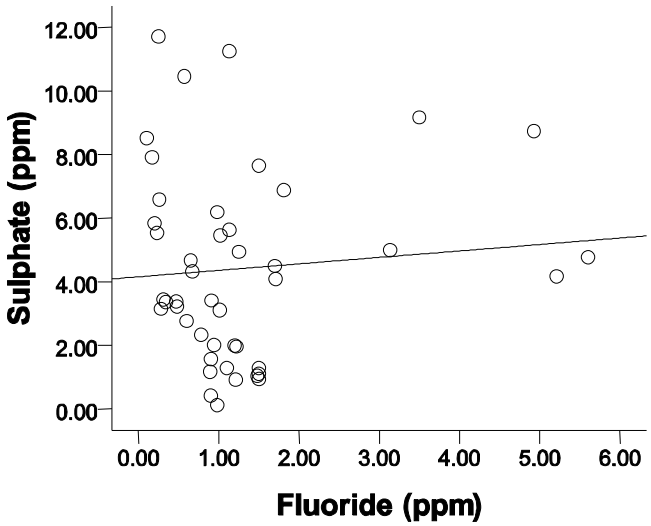
(C).



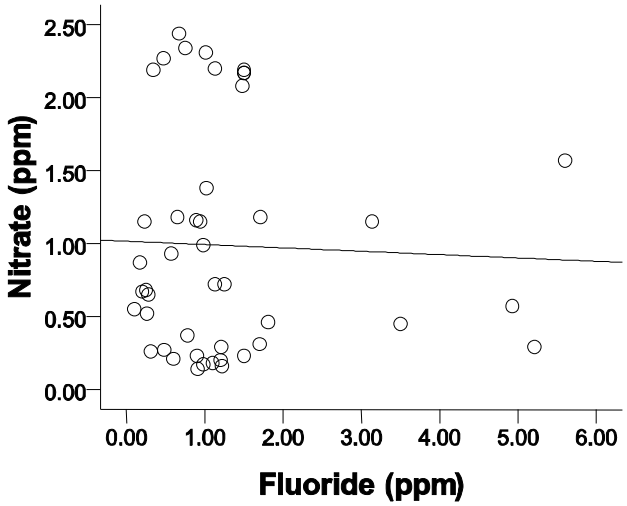
(B).



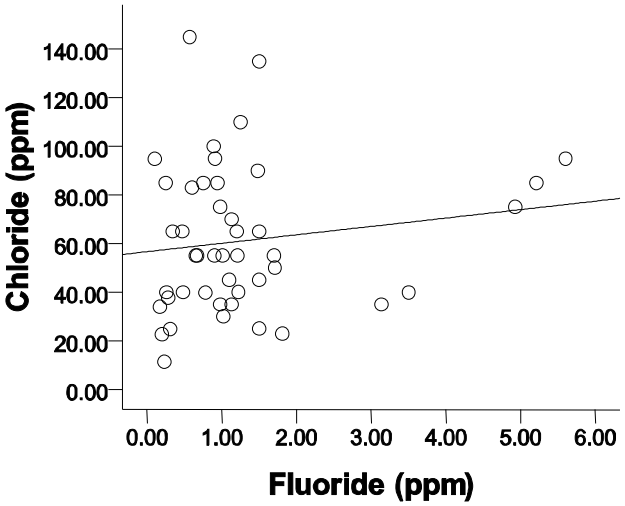
(D).



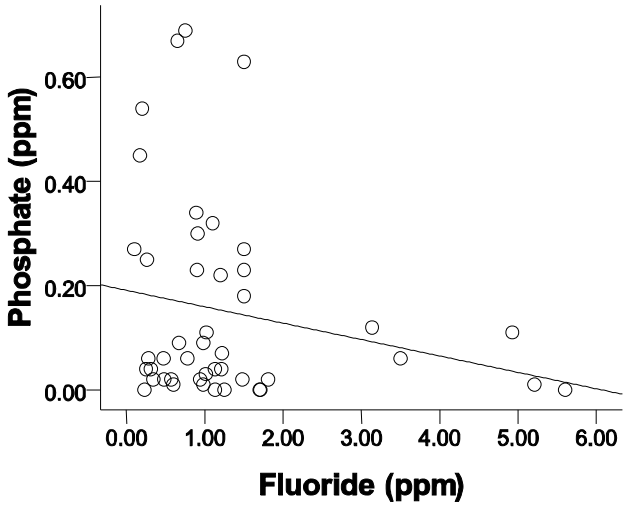
(E).


Nitrate (ppm)
2.50
2.00
1.50
1.00
0.50
0.00
0.00 1.00 2.00 3.00 4.00 5.00 6.00
Fluoride (ppm)

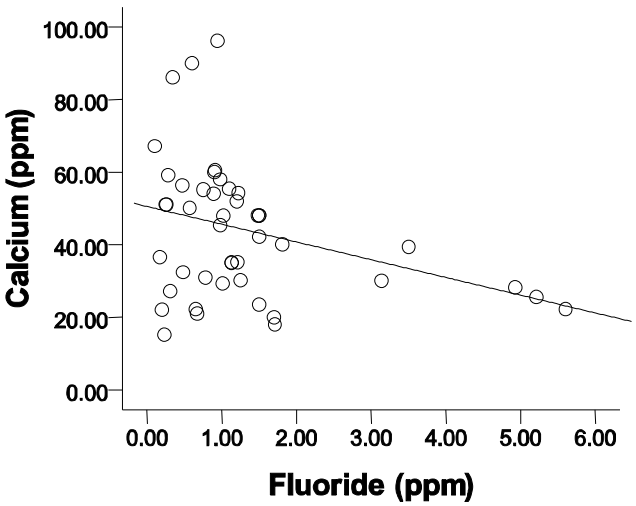
(G).


Chloride (ppm)
140.00
120.00
100.00
80.00
60.00
40.00
20.00
0.00
0.00 1.00 2.00 3.00 4.00 5.00 6.00
Fluoride (ppm)

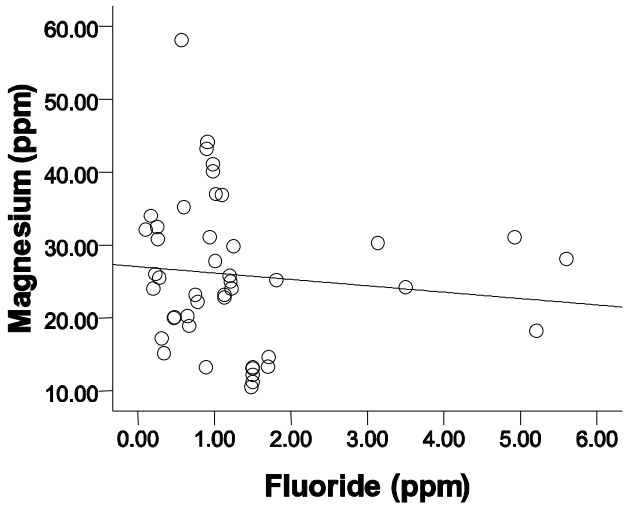
(F).


Phosphate (ppm)
0.60
0.40
0.20
0.00
0.00 1.00 2.00 3.00 4.00 5.00 6.00
Fluoride (ppm)

(H).


Calcium (ppm)
100.00
80.00
60.00
40.00
20.00
0.00
0.00 1.00 2.00 3.00 4.00 5.00 6.00
Fluoride (ppm)

(I).



The pH values varying from 6.00 to 8.21 **(Fig 4).** The permissible limit of pH in drinking water is 6.5 to 8.5 Indian Standard[1] . Maximum pH value 8.21 is found in sample number15 S.P (Susil Payeng) tea garden and minimum value is found 6.0 in sample number 43 of Trinayan tea garden. The pH of the water samples towards acidic it may be due to artificial agrochemicals used in tea gardens leaching to the water sources. Though pH has no direct effect on human health, all the biochemical reactions are sensitive to variation of pH. For most reaction as well as for human beings, pH value 7.0 is considered as best and ideal.

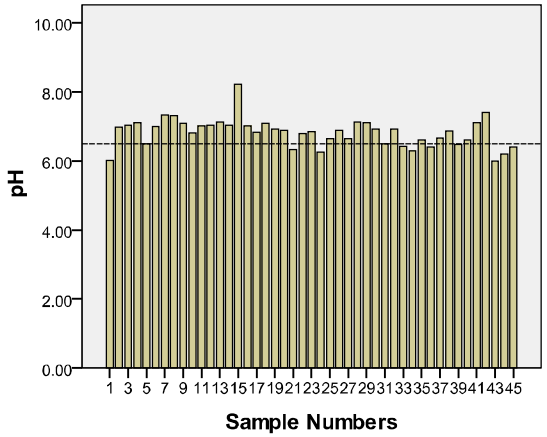


Fig.4. pH concentrations in the water samples (dotted line indicates the permissible limit).

The electrical conductivity varying from .09 mS/cm to 0.81mS/cm (**Fig 5**). All the samples showed E.C below the permissible limit. For drinking water (300 mS/cm) as per bthe ISI standard[1]. Maximum value found in sample number 42 in the Shiva tea garden. 0.81mS/cm and minimum value is 0.09mS/cm in sample number 1 of C.R (Chira Ranjan) tea garden. Conductance is a function of water, hence a standard temperature, usually 25^{O}C, is specified in reporting conductivity. High the concentration of electrolytes in water, the more is its electrical conductance. E.C is positively correlated with pH and correlation coefficient is 0.386.

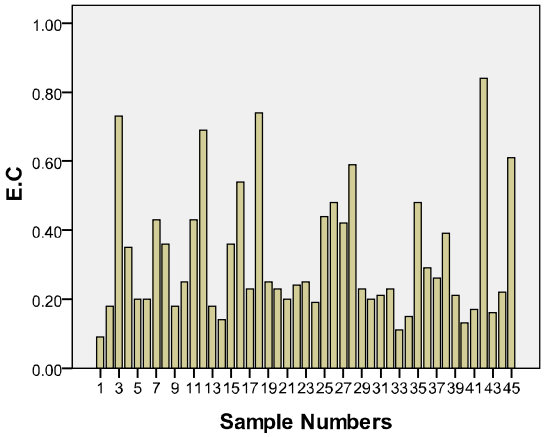


Fig. 5. Electrical Conductivity in the water samples

All the samples analysed showed the Total Hardness varying from 104.63 ppm. to 370.1 ppm **(Fig 6)**. The permissible limit of T.H. for drinking water is 200ppm[1]. The maximum value of T.H is 369.79 in sample number 36 of Jamuna tea garden and minimum value is 104.63 of R.P (Ram Prasad) tea garden. The hardness of water is not depend upon by a single substance but by a variety of dissolved polyvalent metallic ions, predominantly calcium and magnesium cation. The high concentration of T.H in some water samples is due to dissolution of polyvalent metallic ions from sedimentary rocks, seepage and run off from soil . In the present study T.H is positively correlated with calcium, magnesium and chloride the correlation coefficient is 0.832 , 0.784, 0.252.

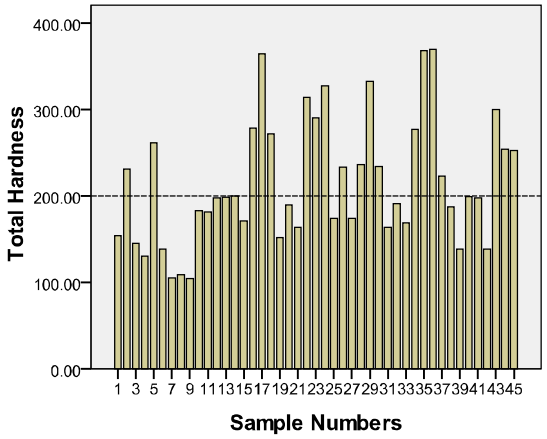


Fig. 6. Total Hardness in the water samples (dotted line indicates the permissible limit).

The Sulphate concentration is varied from 0.119 ppm to 11.71 ppm. The water samples analysed showed lower concentration of sulphates then the prescribed permissible limit for drinking water. The ISI permissible limit of sulphate is 200 ppm[1].Maximum concentration of 11.71 ppm sulphate was observed at the sample number 5 of the tea garden A.S. (Adyomoni Sarmah) and the minimum concentration 0.119 ppm is found in the sample number 22 of the tea garden D.U(Dilip Upadyaya). Sulphate shows weak negative correlation with Ca^{2+} and weak positive correlation with Mg^{2+}. Which indicates in the water samples Calcium Sulphate may be absent and magnesium sulphate may be present. Sulphate in the water samples is due to oxidation of sulfur compounds used in the tea gardens between aerobic and anaerobic environment.

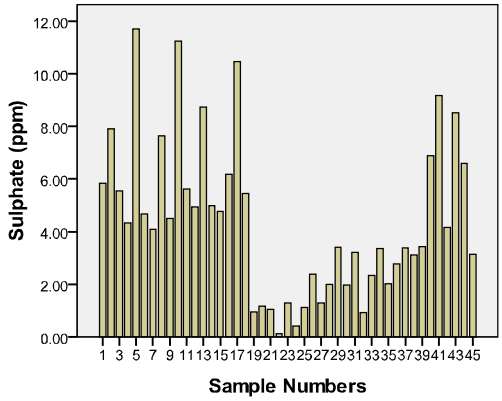


Fig. 7. Sulphate Concentrations in the water samples.

The nitrate concentration is varied from 0.144 ppm to 2.441 ppm **(Fig 8)** . The highest concentration of nitrate is 2.441 ppm in sample number 4 of tea garden A.S (Adyomoni Sarmah) and the lowest concentration of nitrate is found in the sample number 29 of the tea garden B.D (BhatriDay). All the sample have nitrate concentration is lower than the prescribed permissible limit of 45 ppm (ISI)[1]. Nitrate shows negative correlation with Mg^{2+} which indicates that the $Mg(NO_3)_2$ may be absent in the water samples. Nitrate ion may be leaching to the water sources from fertilizers used in the tea gardens.

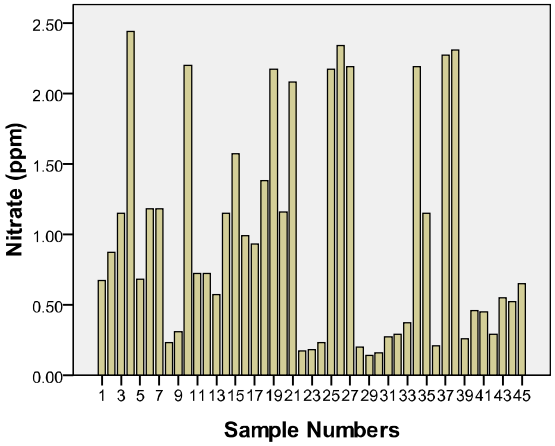


Fig. 8. Nitrate Concentrations in the water samples.

The variation of the phosphate concentration is shown in the **Fig.9.** The phosphate concentration is varied from N.D.(Non Detectable) to 0.688. The maximum concentration of the phosphate is found in the sample number 26 is 0.688 ppm in the tea garden B.B (Bul Bul) and the minimum concentration is non detectable found in the sample number 3, 7, 9, 11,12, 15 . The phosphate ion in the water samples is due to anthropogenic activities mainly input of fertilizers applied to tea gardens and decomposition of organic matter.

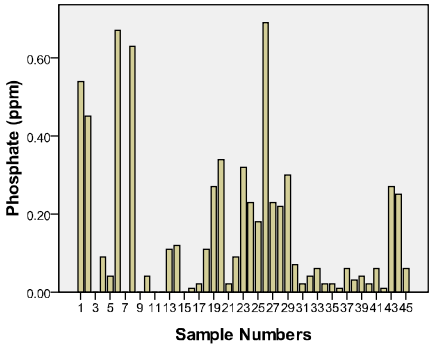


Fig. 9. Phosphate concentrations in the water samples.

Most of the water sample in the tea gardens the chloride concentration is below the maximum ISI permissible limit of drinking water, i.e. 250 ppm[1]. The range of chloride concentration is varied from 11.3 ppm to 144.95 ppm. The maximum concentration is found in the sample number 17 of the tea garden S.G (Santanu Goswami) and the minimum value is found in the sample number 3 of the tea garden C.R(Chitraranjan Upadhya).

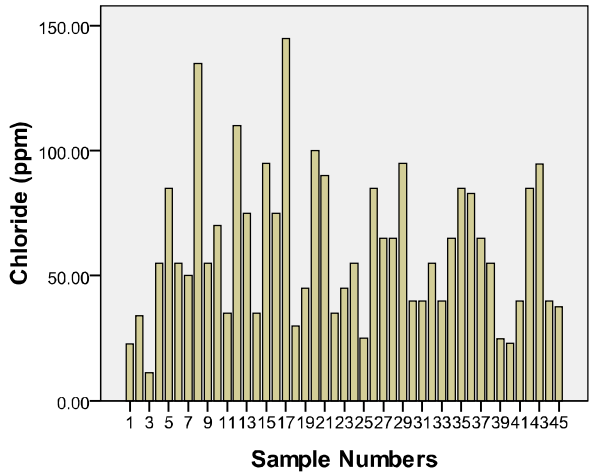


Fig. 10. Chloride Concentrations in the water samples.

The concentration of calcium varied from 15.2 ppm to 96.2 ppm. The highest concentration of calcium is found in the sample number 35 is 96.2 ppm in the jamuna tea garden. The concentration of the calcium ion is shown in the **Fig. 11.** The ISI permissible limit is 75 ppm. In fact 98% of all world ground water are dominated by Calcium and bicarbonate ions due to lime stone weathering in the catchments and under ground water beds[28].

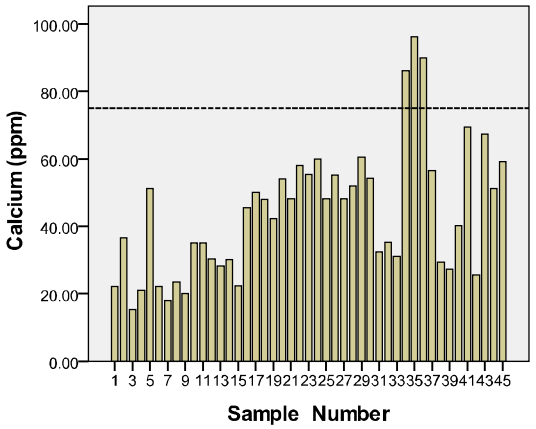


Fig. 11. Calcium Concentrations in the water samples (dotted line indicates the permissible limit).

The concentration of the magnesium is from 10.54 ppm to 58.1 ppm. The maximum concentration 58.1 ppm is found in the S.G.(Santanu Goswami) tea garden and the minimum concentration 10.54 ppm is found in the R.U. (Rishiraj Upadhya) tea garden. The ISI permissible limit is 30 ppm.

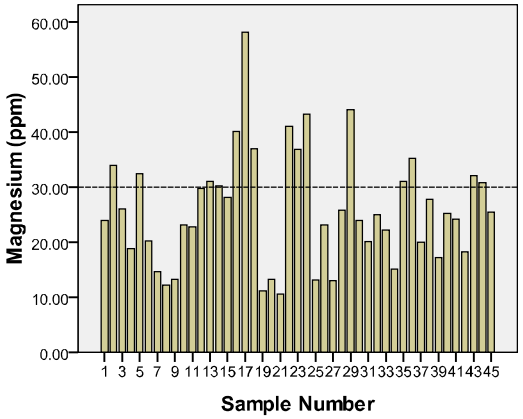


Fig. 12. Magnesium Concentrations in the water samples (dotted line indicates the permissible limit).

Conclusion

Fluoride distribution is associated with pH, calcium, magnesium phosphate and chloride. Positive correlation is observed with pH, E.C. and chloride and negative correlation is observed with total hardness, phosphate, calcium and magnesium. The linear correlation is very useful to get fairly accurate idea of the quality of drinking water or nature of water. In some small tea gardens area S.P (Susl Payeng) Shiva tea garden, the fluoride in drinking water sources is alarmingly high and some gardens area C.R.(Chitra Ranjan),Trinayan tea garden is very low than the ISI limit. Measures of fluoride monitoring should be taken where alternative source for direct use is not feasible and diet of rich calcium and phosphate are suggested where high level fluoride is found and the gardens where low level fluoride is found a ground water management program is suggested.

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