

Fluoride Contamination in Groundwater of Central Assam, India

R.K. Dutta^{*}, G. Saikia¹, B. Das, C. Bezbaruah², H.B. Das¹ and S.N. Dube¹

Department of Chemical Sciences, Tezpur University
Tezpur-784 028, Assam, India

¹Defence Research Laboratory, DRDO
Tezpur-784 001, Assam, India

²Kaliabor College, Kunworitol, Nagaon, Assam, India
✉ robind@tezu.ernet.in

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Abstract: Fluoride contamination of groundwater of the central region of the state of Assam, India, has been studied. The study area includes five districts, viz., Karbi Anglong, North Cachar Hills, Nagaon, Morigaon and Golaghat. A total of 388 groundwater samples have been analyzed for fluoride contamination and 25 of them have been analyzed for fluoride as well as other related parameters. High fluoride contamination has been observed in the groundwater of areas having ancient alluvial red soil and Precambrian metamorphic rock complex basement in the Kapili-Jamuna sub-basin, viz., the plains of Diphu and Hojai subdivisions of Karbi Anglong and Nagaon districts, respectively. Moderate fluoride has been found in groundwater of some places in Morigaon and Golaghat districts also. Besides fluoride, presence of high concentrations of SO_4^{2-} , much above the guideline values, have also been recorded in some samples. The fluoride contents, as a whole, have poor positive correlation with Na^+ , K^+ , pH and depth of source, and poor negative correlation with Mg^{2+} , Ca^{2+} and total hardness with cumulative data spread over entire area of study. The observed poor correlation has been attributed to the diversity of the study area. However, fluoride showed fairly good positive correlation with depth of sources when plotted separately for different localities.

Key words: Fluoride, groundwater, water pollution, central Assam, India.

Introduction

High level of fluoride contamination in groundwater is causing great problems in many parts of India and neighbouring Bangladesh (Murulidharan et al., 2002; Susheela, 2001; Subramanian, 2004). This is a matter of great concern because several physiological disorders are reported to be associated with consumption of water having excess fluoride (Susheela, 2001). Chakraborti et al. (2000) recently reported about severe contamination of fluoride in groundwater and occurrence of fluorosis in some areas of Karbi Anglong and Nagaon districts of Assam. Skeletal and dental fluorosis is prevalent in many parts of these two districts. The groundwater of parts of

the nearby Guwahati city has also been reported to be contaminated with fluoride above the WHO guideline value (Das et al., 2003; Sharma et al., 2005).

The presence of fluoride in groundwater can be attributed to geological reasons (Murulidharan et al., 2002; Subramanian, 2004). The fluoride distribution in the groundwater of the Guwahati city has been correlated with other relevant parameters including its geological set up (Das et al., 2003; Sharma et al., 2005). It is interesting to note that the areas in Assam, which have been so far reported to be contaminated with excess fluoride, i.e., parts of Karbi Anglong and Nagaon districts and the Guwahati city, share the same geological set up. Precambrian metamorphic rock complex forms the basement of these areas (Bhagawati et al., 2001; Goswami

* Corresponding Author

et al., 1998; Pathak et al., 2001; Taher et al., 2001). A detailed study of fluoride contamination in groundwater of the two highly affected districts, viz., Karbi Anglong and Nagaon is also necessary for proper management of the problem. Moreover, in the neighbourhood of these areas there are other areas with same geological set up where no systematic study about fluoride contamination has been reported so far. With this in mind it was thought worthwhile to conduct a detailed survey of fluoride contamination in groundwater of five central Assam districts and to correlate it with other related parameters including geological set up. The area under study is bound by north latitudes $25^{\circ}0'$ to $26^{\circ}7'$ and east longitudes $92^{\circ}0'$ to $94^{\circ}1'$.

Materials and Methods

The water samples were collected in PET bottles and closed tightly. All together 387 groundwater samples were

collected. Fluoride contents in all the samples have been determined whereas determinations of other ions and parameters have been carried out in only 25 representative samples with varying fluoride concentrations, different sources and locations. Fluoride was determined by SPADNS method (Dean, 1990). The results were reproducible within an experimental error limit of $\pm 5\%$. The SPADNS (2-(p-sulfophenylazo)-1,8-dihydroxy-3,6-naphthalein disulfonate) was obtained from E-Merck, Germany and SRL, India. Nitrate contents were determined by spectrophotometric method (Kenkel, 1994). Sulfate and chloride were determined by turbidimetric method and argentometric titrimetric method, respectively (Patnayak, 1997). Phosphate (orthophosphate) was determined by molybdenum blue method (Malati, 1999). For all spectrophotometric determinations a Hitachi U2001 uv-vis spectrophotometer fitted with thermostated cell holders was used. All metal ions were determined by using a

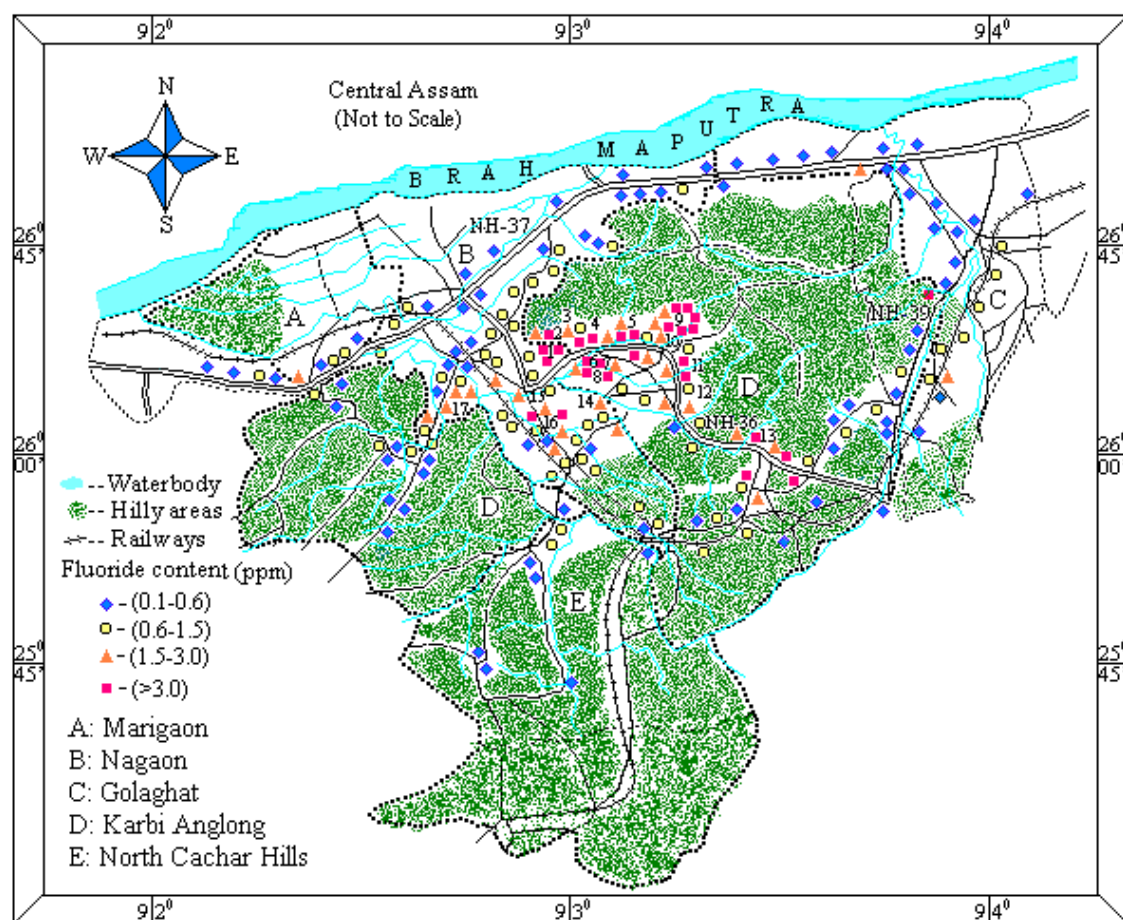


Figure 1: Map showing the distribution of fluoride in the central Assam districts. Highly fluoride affected areas: 1. Akashiganga, 2. Tapatjuri, 3. Haladhiati, 4. Parakhowa, 5. Dengaon, 6. Ratiagaon, 7. Samdingbay, 8. Thoitsutimung, 9. Baghpani (Tekelungjung), 10. Dokmoka, 11. Langhing, 12. Phulani, 13. Amlakhi, 14. Howraghat, 15. Doboka, 16. Nilbagan and 17. Kachua.

Chemito AA203 atomic absorption spectrophotometer. Total hardness (TH) was calculated from concentrations of Ca^{2+} and Mg^{2+} ions (Patnayak, 1997). Doubly distilled water was used for all purposes. Samples were collected in separate bottles for determination of iron. Iron was determined by atomic absorption spectroscopy after dissolving the precipitated iron oxide with nitric acid.

Results and Discussion

A total of 395 groundwater samples collected from the study area were analyzed for fluoride contents. The results of analysis of the samples are given in Table 1. 18.60% of the samples were found to contain fluoride above 1.5 ppm—the guideline value of WHO—whereas 54.26% of the samples are deficient in fluoride, i.e., have fluoride below 0.6 ppm (WHO, 1984). Five samples had fluoride concentrations above 10 ppm, 18 samples between 5 and

10 ppm and 13 samples between 3 and 5 ppm, which are alarmingly high. These data are actually underestimated as a large number of the samples were collected from areas where the groundwater is either deficient or normal in fluoride concentration.

The distribution of fluoride in groundwater of the area under study is shown in Figure 1. The localities where the groundwater contains more than 1.5 ppm of fluoride are listed in Table 2. Several samples were collected from each of these localities. Various locations are based on repeat data as well as one time observation. It can be seen that very high fluoride contamination has been found in the plains of the Diphu subdivision of Karbi Anglong and the adjacent southern plains of Nagaon District. Other areas where moderately high fluoride content has been recorded are some tiny valleys in the hilly areas of Karbi Anglong district, Dhansiri valley of Golaghat district situated to the east of Karbi Anglong and foot hill areas

Table 1: Distribution of Fluoride in Groundwater of Five Districts of Central Assam

District	Source type	No. of samples	% of samples having fluoride (ppm) in different ranges					
			0.1-0.6	0.6-1.5	1.5-3.0	3.0-5.0	5.0-10.0	>10
Entire region	A	388	54.63	30.92	3.60	2.06	7.47	1.03
	B	223	23.70	21.40	2.60	1.8	6.92	1.03
	C	55	8.76	3.86	0.51	0.26	0.26	-
	D	87	17.78	4.12	0.51	0.26	-	-
	E	23	4.38	1.54	-	-	0.26	-
Karbi-Anglong	A	148	52.02	28.37	1.35	2.70	15.54	0.67
	B	86	12.83	18.91	-	2.70	14.18	0.67
	C	19	7.3	2.4	-	-	0.6	-
	D	48	23.3	4.9	1.2	-	-	-
	E	11	5.5	1.2	-	-	0.6	-
Nagaon	A	175	50.85	36.5	4.57	2.28	3.42	1.71
	B	120	31.42	26.28	4.00	1.71	3.42	1.71
	C	20	6.28	4.00	0.57	0.57	-	-
	D	25	10.2	4.00	-	-	-	-
	E	10	3.42	2.28	-	-	-	-
Marigaon	A	30	66.6	23.3	6.6	-	-	-
	B	14	23.3	16.6	6.6	-	-	-
	C	4	10	3.3	-	-	-	-
	D	11	33.3	3.3	-	-	-	-
Golaghat	A	30	66.6	26.6	6.6	-	-	-
	B	17	36.6	16.6	3.3	-	-	-
	C	13	30	10	3.3	-	-	-
North Cachar Hills	A	5	100	-	-	-	-	-
	D	3	60	-	-	-	-	-
	E	2	40	-	-	-	-	-

Type of Groundwater Sources: A – all sources, B – hand tube well or deep tube well, C – deep tube well installed by state public health department, D - crude well and E – stream or river.

Table 2: Fluoride Affected Areas where Groundwater Recorded above 1.5 ppm Fluoride in Different Districts of Central Assam

<i>District</i>	<i>Affected areas</i>
Karbi Anglong	Dengaon, Parokhowa, Amlokhi, Manza, Ratiagaon, Samdingbay, Dokmoka, Thoitsutimung, Terang-gaon, Ranghang, Hafjangaon, Tekelungju, Baghpani, Phulani, Nopakling, Kharguthi and Langhing
Nagaon	Akashiganga, Tapatjuri, Nilbagan, Haladhiati, Kachua, Singimari, Padumoni Tiniali, Sankar Dev Nagar, Kaki-I, Kaki Chariali, Nijparakhowa, Morajhar and Doboka
Marigaon	Nellie and Rupaibari Karbigaon
Golaghat	Sorupother and Garmurh

of southern Morigaon district. None of the samples collected from the North Cachar Hills district was found to have fluoride concentration above 1.5 ppm (Table 1). The two samples of this district, which were found to have fluoride above 0.6 ppm, were from places near the plains of Karbi Anglong and Nagaon districts.

The Karbi Anglong district and the southern part of Nagaon district have a common Precambrian metamorphic rock complex basement with mainly ancient red alluvial soil. Presence of limestone and Cretaceous alkaline-carbonatite igneous complexes in the Karbi plateau has been reported in certain places (Taher, 2001). Moreover, this is a rain-shadow area within North-east India (Bhagabati et al., 2001). On the other hand the geological set up of the North Cachar Hills district, where the groundwater is free from fluoride contamination, is different from that of the Karbi Anglong and in fact is an extension of the Himalayan ranges. It can be noted here that the Guwahati city which is adjacent to the west of Morigaon district also has the same geological set up with the fluoride affected areas under study (Bhagawati et al., 2001; Goswami et al., 1998; Pathak et al., 2001; Taher et al., 2001). Occurrence of high fluoride in groundwater of other adjacent areas, viz., Guwahati and the Darrang district, the latter being situated on the north bank of Brahmaputra opposite Morigaon district and the Guwahati city, can be noted here (Das et al., 2003; Sarma, 1999; Sharma, 2005). Thus the entire fluoride affected area in Assam has a geomorphological setup which is distinctly different from that of the other parts of Assam, viz., the Brahmaputra Basin, the part belonging to the Himalayan ranges and the Barak basin (Taher et al., 2001).

The water samples studied included one hot spring, nine streams and 11 rivers of the study area (Table 3). In

the hot spring, located in the Karbi Anglong district, water was found to have 9.5 ppm of fluoride. The highest fluoride concentrations recorded for streams and rivers were 0.88 and 0.63 ppm, respectively. The fluoride concentration of the Jamuna river was found to be 0.58 at Silveta in the upstream, whereas it was 0.40 at Dabaka, downstream. The fluoride concentration in the groundwater samples gradually decreased and became deficient in fluoride content as one goes from the most fluoride affected area towards the recent alluvial Brahmaputra basin. The fluoride concentration of the Brahmaputra river is less than 0.2 ppm (Das et al., 2003; Madhavan et al., 2001). This indicates that the source of fluoride is present in the basement of the affected areas.

Table 3: Fluoride Contents in ppm in Water of Springs and Rivers of the Fluoride Affected Areas of Karbi Anglong (Sl. No. 1 to 10), Nagaon (11 to 19) and North Cachar Hills District (20 and 21)

<i>Sl. No.</i>	<i>Name of source</i>	<i>Source type</i>	<i>Fluoride</i>
1	Garam pani, Nambor	Hot Spring	9.33
2	Dikharu, Dokmoka	River	0.48
3	Disoubi	River	0.68
4	Mondadisha	River	0.63
5	Parakhowa, Ratiagaon	Stream	0.53
6	Longpur Pam	Stream	0.25
7	Longni, Longnit	River	0.23
8	Diphu	River	0.53
9	Akashigonga	Stream	0.31
10	Jamuna, Silveta	River	0.58
11	Jamuna, Dabaka	River	0.40
12	Nimakhjuri, Lumding	Stream	0.88
13	Shukaan Juri	Stream	0.53
14	Shorulongpher	River	0.75
15	Siv Mandir, Rang Bang	Stream	<0.1
16	Bishnu Mandir, Rang Bang	Stream	~0.1
17	Budigaon	Stream	0.15
18	Borgaon	Stream	0.25
19	Nonoi	River	<0.1
20	Kapili	River	0.51
21	Doyang	River	0.28

The fluoride concentrations, for the entire study area when considered together, showed a poor positive correlation with depth. However, it is interesting to note that the approximate depths of the sources of the samples of three different fluoride affected localities, when plot separately for each locality, showed fairly good positive correlations with the fluoride concentration (Figure 2). The *R*-squared values for the correlation of fluoride concentration with depth at different localities were found

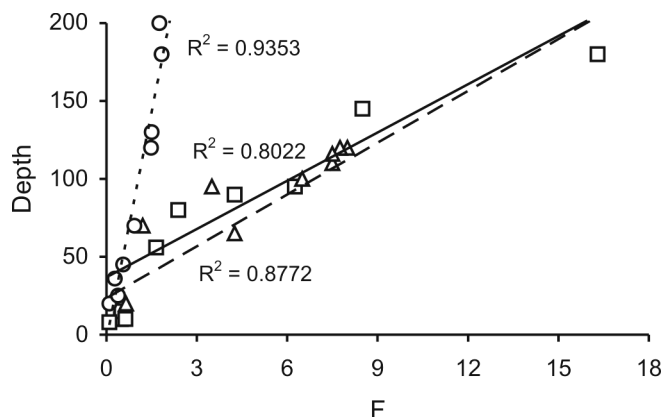


Figure 2: Correlation of depth in feet of groundwater source with fluoride concentration in ppm at three highly fluoride affected locations: O, dotted line–Kachua; □, broken line–Baghpani; Δ, solid line–Samdingbay.

to be 0.9353 at Kachua in southern part of Nagaon district and 0.8772 and 0.8022 at Baghpani and Samdingbay villages, respectively, both in the plains of Karbi Anglong

district. The variation in the slopes of the correlation curve with location can be attributed to the diversity of the study area. The positive correlation with depth indicates the source of fluoride to be fluorite or (and) apatite minerals present in the Precambrian granite or granitic-gneiss of the underground basement of the areas (Taher et al., 2001; Handa, 1973). The smaller slopes indicate greater severity of the fluoride contamination and location of the source of fluoride near that area. It can be further noted from the figure that the samples with depth less than 50 ft had fluoride concentration below 1 ppm. The highest fluoride concentration in the area under study (16.3 ppm) was recorded at Nopakling village of Karbi Anglong in a tube well of 180 ft depth, whereas the fluoride concentrations found in two wells of 10 ft depth in the same village were less than 0.5.

The results of various ions and other parameters of 25 representative samples (mostly having high F^- concentrations) of groundwater of areas covered are shown in Table 4. The concentrations of F^- in thirteen

Table 4: Chemical Analysis of Selected Groundwater Samples of Central Assam

Sample No.	Sample location	Source Type	Ca^{2+}	Mg^{2+}	Na^+	K^+	F^-	Cl^-	SO_4^{2-}	NO_3^-	PO_4^{3-}	TH	TA	pH
025	Nilbagan, NG	HT	11.5	10.0	104.4	5.4	2.50	6.0	60	3.5	0.9	70.0	425	8.5
036	Ratiagaon, KA	HT	2.4	5.4	60.6	10.6	9.48	8.0	140	1.4	0.2	28.2	171	7.5
037	Sumdingbay, KA	HT	2.0	5.9	58.0	2.5	1.80	10.0	110	4.0	3.2	29.3	229	7.5
048	Dengaon, KA	PH	4.1	4.8	58.6	4.8	3.50	6.0	150	1.6	0.4	30.0	215	8.1
049	Dengaon, KA	HT	3.2	6.2	61.4	4.1	2.88	8.0	80	3.0	0.9	33.4	237	8.2
058	Nopekling, KA	HT	2.3	1.0	113.1	2.8	16.30	47.6	4200	1.6	0.4	9.8	325	8.3
103	Borboha, NG	Well	4.9	34.9	55.4	8.1	1.30	21.8	61	1.3	0.2	155.8	332	8.2
104	Heera Basti, NG	HT	5.3	30.4	53.4	3.6	1.25	12.0	143	0.8	0.2	138.4	366	8.2
111	Kapili, NC	River	17.2	2.2	2.8	3.0	0.27	4.0	1040	0.7	0.1	52.1	12	7.4
138	Dengaon, KA	HT	3.5	4.9	57.3	5.5	3.88	6.0	80	1.8	0.9	28.7	220	7.8
149	Bokuliaghat, KA	HT	17.3	15.1	29.2	5.2	1.35	6.0	180	1.0	0.4	105.3	229	6.8
163	Amlokhi, KA	HT	3.2	3.1	225.0	12.8	2.25	16.0	175	5.5	0.4	20.8	771	8.1
168	Halfjan Gaon, KA	Well	3.2	21.6	776.0	12.6	2.00	61.8	4000	4.2	0.3	96.8	1074	7.8
181	Lumding, NG	Well	35.3	20.0	60.6	9.8	0.58	87.4	2150	43.6	0.5	170.4	254	8.0
199	Kaki, NG	PH	2.4	12.6	43.4	9.2	1.75	10.0	110	0.8	0.3	58.0	361	7.1
206	Chaparmukh, NG	HT	8.1	8.2	60.2	4.6	0.80	12.0	80	3.5	0.7	54.0	268	7.8
217	Sarupather, GH	PH	10.5	7.7	63.2	6.1	1.57	8.0	105	1.8	0.5	57.7	283	7.6
218	Barpather, GH	HT	3.2	3.5	91.5	6.6	0.70	49.6	70	0.9	1.2	22.5	390	8.1
245	Nilbagun, NG	HT	7.4	15.1	166.4	2.8	1.28	4.0	170	1.7	0.3	80.7	461	7.7
251	Hojai, NG	HT	33.3	19.0	71.2	15.5	0.27	79.4	2150	1.1	0.1	161.4	278	6.7
271	Borzari, NG	PH	2.4	7.7	78.4	5.4	1.32	6.0	150	3.5	3.1	37.6	317	7.5
274	Jamunamukh, NG	HT	2.3	7.9	80.6	5.2	1.50	6.0	135	3.1	2.8	38.1	249	8.0
307	Numaligarh, GH	PH	9.5	12.8	6.9	4.3	0.32	12.0	1050	1.6	0.1	76.5	83	7.2
316	Nambor, KA	Spring	18.0	1.0	360.4	17.2	9.50	651.2	190	0.9	0.3	49.1	68	6.9
329	Bokajan, KA	HT	2.3	8.1	27.1	10.4	0.28	10.0	255	0.8	0.2	39.0	229	6.8
—	WHO Standards	—	200.0	150.0	—	—	1.50	600.0	400	45.0	—	500.0	—	6.5–8.5

All quantities are in ppm except pH. Abbreviations: HT—hand tube well, DTW—deep tube well, PH—DTW installed by state public health & engineering department, NG—Nagaon district, KA—Karbi Anglong district, GH—Golaghat district, NC—North Cachar Hills district, TH—total hardness and TA—total alkalinity.

samples, SO_4^{2-} in six samples and Cl^- in one sample out of these 25 samples studied were above the recommended values of WHO (WHO, 1984). The SO_4^{2-} ion concentrations in the six samples are alarmingly high and needs appropriate attention. It can be noted here that the users of the well in Half-jan village in Karbi Anglong district reported about the water of a well (sample No. 168) to be highly saline where the Na^+ ion concentration also was found to be as high as 776 ppm.

The correlation of some select ions and some other parameters with F^- are shown in Figure 3. In general only weak correlation was observed with the ions and the parameters. The alkaline earth metal ions, viz., Mg^{2+} and Ca^{2+} and the total hardness showed poor negative

correlation with fluoride contents. Negative correlation of F^- with Mg^{2+} and Ca^{2+} is as expected due to low solubility of fluorides of these ions (Das et al., 2003; Handa, 1975; Smedley et al., 2002; Hounslow, 1995; Hem, 1991). It can be mentioned here that Chakraborti et al. (2000) reported observation of a positive correlation of fluoride with Ca^{2+} in the groundwater of parts of Nagaon and Karbi Anglong districts of Assam, which is unusual. The poor negative correlation of F^- with Mg^{2+} and Ca^{2+} observed in this study, as well as the unusual positive correlation observed by Chakraborti et al. (2000), can be attributed to the diversity in the situations of the different localities. A more detailed study at individual localities may show better negative correlation as has

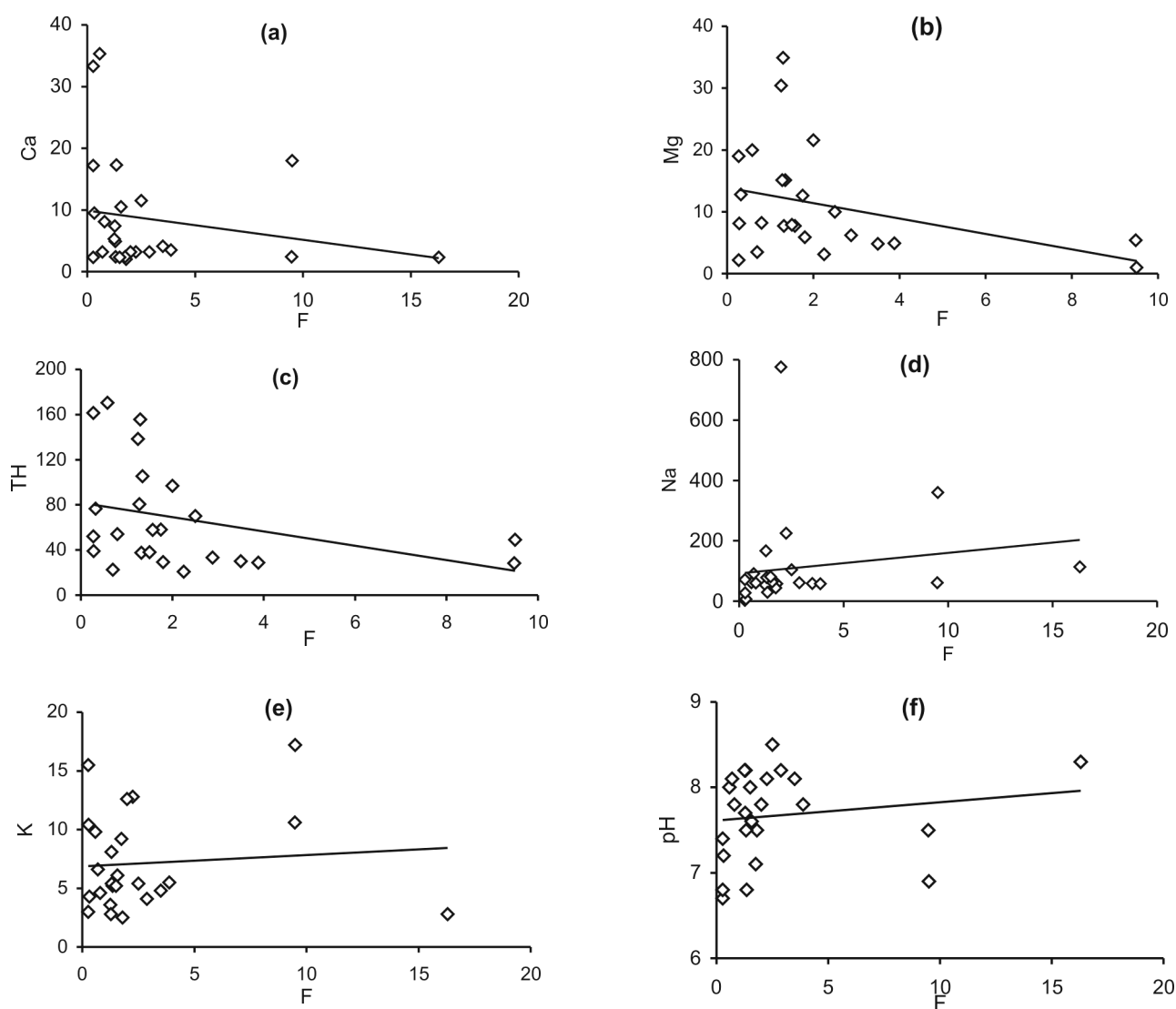


Figure 3: Correlation of different ions and parameters with fluoride concentration in selected groundwater samples: (a) Ca^{2+} vs. F^- , (b) Mg^{2+} vs. F^- , (c) TH vs. F^- , (d) Na^+ vs. F^- , (e) K^+ vs. F^- , (f) pH vs. F^- . The solid lines indicate trends. All quantities are in ppm except pH.

been observed in the case of correlation with depth of sources (Figure 2). Presence of limestone and Cretaceous alkaline-carbonatite igneous complexes in the Karbi plateau may have been a boon to the people living in the region as these Ca^{2+} containing minerals release high amount of Ca^{2+} in the areas which can reduce the concentration of F^- in groundwater (Taher, 2001; Heaman, 2002).

The alkali metal ions, viz., Na^+ and K^+ showed positive correlation with fluoride contents. Similarly, a poor positive correlation of F^- was observed with pH also. The total alkalinity and pH were moderate or within the WHO guideline values. A positive correlation of fluoride with Na^+ reported in groundwater of a nearby Guwahati city can be mentioned here (Das et al., 2003). The SO_4^{2-} ion, even though is present in very high concentrations in some locations, also did not show any correlation with fluoride. Nitrate and orthophosphate were found to be moderate or within the WHO guideline values. Cl^- also did not show any correlation with F^- content which suggests that evaporation is not a reason for high fluoride contamination of the groundwater of the region (Smedley et al., 2002). The presence of fluoride in the groundwater of the area necessitates a study of presence of other toxic elements since certain toxic elements, e.g., arsenic, boron have been reported to show positive correlation with fluoride (Smedley et al., 2002).

Conclusion

In view of the presence of fluoride and sulfate in alarmingly high concentrations in a large percent of the groundwater sources, testing of potability of the groundwaters of the area should be done before using for drinking purposes. This is particularly important for the southern part (Hojai sub-division) of Nagaon district and the plain areas of Diphu sub-division of Karbi Anglong district. Streams (except the hot spring) and rivers and groundwater sources of depth less than 50 ft are safe sources of drinking water for the affected areas. The safe depth, however, can be greater in some localities like Kachua of Nagaon district. Measures for fluoride removal should be taken where alternative source for direct use is not feasible.

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