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RESEARCH ARTICLE

FLOURINE CONTAMINATION IN GROUNDWATER OF AMARAVATHI RIVER BASIN, TAMILNADU, INDIA

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ABSTRACT

Fluoride with high concentration in groundwater has been reported from many parts of India. However, a systematic study is required to understand the presence of fluoride in natural water in terms of local hydrogeological setting, climatic conditions, and agricultural practices etc. The present study reveals to assess hydrogeochemistry of groundwater in parts of Amaravathi river basin in Tamil Nadu to understand the fluoride abundance in groundwater and to deduce the chemical parameters responsible for the dissolution activity of fluoride. The study area is geologically occupied by partly sedimentary and partly crystalline formations. A total of 60 dug cum borewell-water samples, representing an area of 8280 Sq.km. The results of the chemical analyses in June 2012 show fluoride abudance with the range of 2.1to4.5mg/l. Presence of fluoride-bearing minerals in the host rock, chemical properties like decomposition, dissociation, and dissolution, and their interaction with water are considered to be the main causes for fluoride in groundwater. Chemical weathering with relatively high alkalinity favours high concentration of fluoride in groundwater. Villagers who consume nonpotable high fluoride water may suffer from yellow, cracked teeth; joint pains; and crippled limbs and also age rapidly.

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INTRODUCTION

Groundwater is a preferred source of human water supply. It is estimated that approximately one third of the world's population uses groundwater for drinking purpose (UNEP) 1999) for most rural and small communities, groundwater is still the only source of drinking water. Generally, ground water carries higher mineral content than surface water, because of slow circulation and longer period of contact with rich rocks or sediment mineral. Fluoride (F−) occurs in almost all waters from trace to high concentration. Fluoride concentration in natural waters depends on various factors such as temperature, pH, solubility of fluorine bearing minerals, anion exchange capacity of aquifer materials (OH− for F), and the nature of geological formations drained by water and contact time of water with a particular formation. Groundwater quality varies due to changes in chemical composition of the underlying sediments and aquifers (Jameel 2002). Fluoride is one such naturally acquired constituent of groundwater. The main source of fluoride in groundwater is fluoride-bearing minerals such as Fluorspar (CaF2), Cryolite (AlF3, 3NaF) and Fluroapatite [Ca5 (PO4)3F] in rocks (Farooqi *et al*. 2007). Fluoride is among the substances for which there are both lower (0.6 mg/l) and upper (1.2 mg/l) limits of concentration in drinking water, with identified health effect and benefits for human beings (Indian Standard Institute [ISI]). Very lowdoses of fluoride (<0.6 mg/l) in water promote tooth decay. However, when consumed in higher doses (>1.5 mg/l), it leads to dental fluorosis ormottled enamel, and excessively high concentration (>3.0 mg/l) of fluoride may lead to skeletal fluorosis. In general, fluoride content in water between 1.5 and 2.0 mg/l may lead to dental mottling, which is characterized initially by opaque white patches on the teeth, and, in advanced stages, leads to dental fluorosis (teeth display brown to black staining) followed by pitting of teeth surfaces. High manifestations of dental fluorosis are mostly found in children up to the age of 12 years, and skeletal fluorosis (Apambire *et al*. 1997) may occur when fluoride concentrations in drinking water exceed 4–8 mg/l. Crippling skeletal fluorosis can occur when the water supply contains more than 10 mg/l of fluoride (Boyle and Chagnon 1995). The severity of fluorosis depends on the concentration of fluoride in the drinking water, daily intake, continuity and duration of exposure, and climatic conditions. In India, an estimated 62 million people, including six million children, suffer from fluorosis because of consuming fluoride-contaminated water. Though fluoride as a contaminant can enter the body through food, water, industrial exposure, drugs, cosmetics etc., drinking water is the major contributor (75-90% of daily intake) (Sarala and Rao 1993). The cause fluorosis is mainly due to the drinking of fluoride contaminated groundwater. It has been now confirmed that the villagers are

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drawing water from the dug wells and the shallow hand pumps for their drinking purposes and other domestic uses (Mukherjee *et al*.1995). The high fluoride concentration manifests as an increase in bone density leading to thickness of long bones and calcification of ligaments. Symptoms include mild rheumatic/arthritic pain in the joints and muscles to severe pain in the cervical spine region along with stiffness and rigidity of the joints. The disease may be present in an individual at subclinical, chronic, or acute levels of manifestation.

A number of cases of fluorosis have been reported mostly from the granite and gneissic complex of different states such as Andhra Pradesh (Rao 2003; Rao and Devadas 2003; Sreedevi *et al*. 2006), Bihar (Ray *et al*. 2000), Delhi (Susheela *et al*. 1996), Gujarat (Chinoy *et al*. 1992), Karnataka (Wodeyar and Sreenivasan 1996; Sumalatha *et al*. 1999), Kerala (Shaji *et al*. 2007), Madhya Pradesh (Chatterjee and Mohabey 1998; Nawlakhe *et al*. 1995), Maharashtra (Deshmukh and Chakravarti 1995),Orissa (Kundu *et al*. 2001; Das *et al*. 2000), Rajasthan (Muralidharan *et al*. 2002; Choubisa *et al*. 1996), and Tamil Nadu (Handa 1975; Apparao and Karthikeyan 1986).

Study Area

Amaravathi river is one among the major tributaries of the Cauvery river system in southern India. It originates from Anaimalai hills at an altitude of 1827m which drains from south to the north east and confluences with the main river, Cauvery at Thirumukudalur which is situated around 10km east of Karur,(Karur) district, Tamilnadu. This basin is located between latitudes 10º 8´ N to 11º 1´N and longitudes 77º3´E to 78º 8´E covering an area of about 8280sq.km.The river is of north flowing type. Topographically, the basin is enclosed by coimbatore uplands, on the west and by the Anaimalais, palani and sirumalai hills on the south and southeast. . The northern part of the basin has an undulating plain with an average elevation of 300m. The valleys represent a low relief from those of the hills with a difference in height ranging from 1500 to 2500m. The study area experiences tropical monsoonal climate. The temperature on the hills differs from that of the plains, which lies between 15ºC and38ºC on the low land and 10ºC to 18ºC on the hills. The average annual rainfall is about 855mm.Strong winds from the southwest prevail during June,July and August. The hills are covered with dry monsoon forests. At some places, medicinal shrubs have also grown within the palani hills.

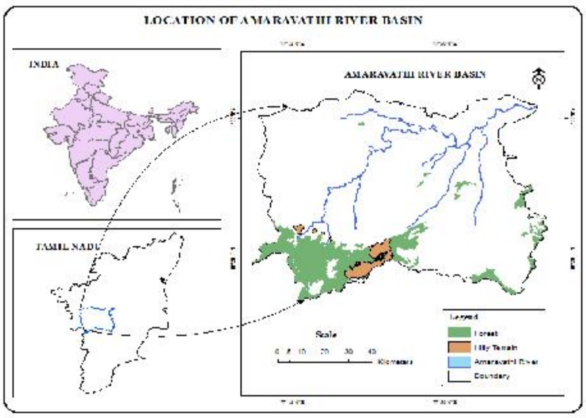

Fig. 1 Location map of the study area

METHODOLOGY

In the present study, water samples were collected from 60 dug cum bore wells in the study area (Fig. 2). The study is carried out with the help of topographic sheets, Garmin GPSMAP76, ArcGIS 9.3, and fieldwork. Toposheets are used to prepare the base map and the drainage map and to understand the general nature of the study area. The global positioning system is used to map the location of each sampling well, and finally, the results were taken to the geographic information system (GIS) for further analysis. The samples collected were analyzed for various parameters by Soil Testing Laboratory, Trichy, Tamil Nadu. (Table 1)

These parameters include pH, chloride (Cl), nitrate (NO3), calcium (Ca), magnesium (Mg), and fluoride (F). Spatial analyst, an extended module of ARCGIS 9.3, was used to find out the behavior of the groundwater quality parameters.

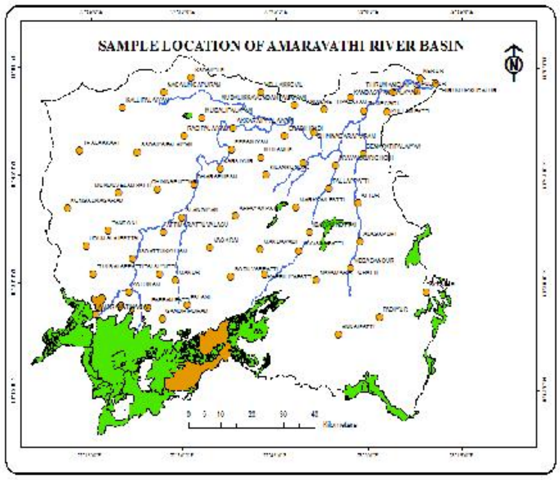

Fig .2 Sampling stations of Location map

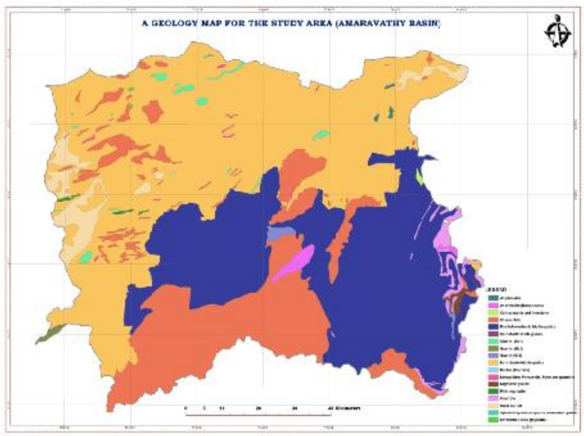

Fig. 3 Geology of Amaravatahi River Basin

Geology

Several digital image processing techniques, including standard color composites, intensity-hue saturation (IHS) transformation, and decorrelation stretch (DS) were applied to map rock types. Better contrast was obtained due to color enhancement, and this facilitated visual discrimination of various rock types.

Table 1 Drinking water specification of the study area in comparison with WHO (1984), ISI (1983) Minimum, Maximum and Mean Ion concentration

Parameters	Minimum	Maximum	Mean	WHO Standards 1984	US Environmental Protection agency secondary dirking water standards	ISI (1983)
TDS	262	627	449	500	500	500
EC	410	980	656	-	-	-
pH	7	7.89	7.5	6.5-8.5	6.5 - 8.5 on scale	6.5 - 8.5 on scale
co^3	0	0	0	-	-	-
HCO_3	115	362	211	-	-	-
Cl	72	168	108	200	250	250
SO_4	34	78	57	200	250	150
NO_3	0	0.62	0.07	< 0.1	1	-
Ca	80	224	129	75	-	75
Mg	41	108	67.3	< 30 If SO4 is 250 mg/l upto 150 Mg/l if SO4 is < 250 Mg/l	-	3
Na	51	152	84	200	-	-
K	1	6	3	12	-	-
F	2.1	4.5	2.9	1.5	-	-

Eighteen rock types were mapped and could be distinguished by distinct colours in the processed images. They are Amphibolite, Anothosite (Basic rocks),Calc-granulite and limestone, Charnockite, Fissile hornblende blotite gneiss, Garnet-sillimanite gneiss, Granite (Gr1),Granite (Gr2),Granite (G3),Hornblende-blotite gneiss, Kankar (Calcrete),Pyroxenite, Migmatite gneiss, Pink migmatite, Quartzite, Sand and silt, Syenite and Ultramafic rocks. A map of the interpreted distribution of rock types in the study area is shown in Fig. 3

Hydrochemistry

The use of water analyses in groundwater hydrology is to produce information concerning the water quality. Understanding the groundwater quality is important, as it is the main factor determining its suitability for drinking and domestic, agricultural, and industrial purposes. The minimum, maximum, and mean concentrations of physicochemical parameters of water quality such as pH, electrical conductivity (EC), total dissolved salts (TDS), and major anions and major cations are presented in Table 2. During the present investigation, pH value as low as 7.0 in was recorded in Kolumum and the highest was found in idaiyakkottai, with a value of 7.89. In general, the distribution of pH did not show any specific trend within the study area. EC is measured in microsiemens per centimetre and is a measure of salt content of water in the form of ions.

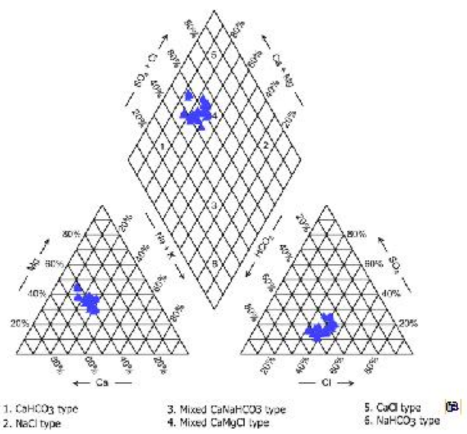

Fig .4 Piper diagram

In the present study, EC values ranged from 410 to 980 µS/cm. The distribution of TDS values clearly shows that the entire study area falls within this range. The values ranged between 262 and 627 mg/L (Table 2). The chloride concentration varies between 72 and 168 mg/l with an average value of 108 mg/l (Table 2). The nitrate ion concentration varies from 0 to 0.62 mg/l with an average value of 0.07mg/l. The sulfate concentration in groundwater of the study area is within the permissible limit in all the sample locations.

The piper diagram is extensively used to understand problems concerning the geochemical evolution of groundwater. The diagram consists of three distinct fields-two triangular fields and a diamond-shaped field. The percentage equivalents per mole values are used for the plot. The overall characteristic of the water is represented in the diamond-shaped field by projecting the position of the plots in the triangular fields. Different types of groundwater can be distinguished by their plotting position, occupying certain subareas of the diamond-shaped field. Piper–trilinear plots were made for the samples collected during the june 2012 field visits. The AquaChem software is used for the plotting of piper–trilinear diagrams (Scientific Software Group, Utah, 1998). From the plots, it can be found in Fig. 4 that calcium and magnesium ions are the dominant cations. A perusal of hydrochemical character from the piper–trilinear diagram indicates that alkalis exceed alkaline earths and strong acids exceed weak acids in groundwater (Table 3).

Fluoride

The anion and cation concentrations, except fluoride, are within the desirable limits (BIS 1991). The analysis results indicate that the water is generally alkaline in nature. F− concentration values are higher than the permissible limit, i.e., 1.5 mg/l (BIS 1991; WHO 1984). However, all the samples are recorded F^- concentration values are higher than the maximum permissible limit.

Fluoride concentration in samples has been depicted in an isofluor map (Fig. 5) for a better understanding of the distribution and the behavior.

Table 2 Hydrochemistry data for the groundwater samples from the study area

Sl.No	St. Name	pH	Ca	Mg	Na	K	HCO3	Cl	F	No3	so4	CO3	TDS	EC
1	Amaravatinagar	7.25	85	45	56	3	182.0	79.0	2.56	0.62	39	0.0	333	520
2	Tumbalappatti	7.12	82	46	52	2	186.0	85.0	2.36	0.62	38	0.0	358	560
3	Kolumam	7.06	89	45	56	2	189.0	86.0	2.12	0.62	34	0.0	326	510
4	Pappampatti	7.13	85	48	51	1	182.0	82.0	2.32	0.62	35	0.0	346	540
5	Gandhipuram	7.42	86	43	53	2	185.0	74.0	2.1	0.62	36	0.0	339	530
6	Palani	7.45	165	102	58	3	258.0	148.0	2.56	0.62	62	0.0	525	820
7	Manur	7.56	158	105	53	2	253.0	146.0	2.48	0	68	0.0	550	860
8	Vagarai	7.48	164	103	56	5	248.0	142.0	2.65	0	63	0.0	538	840
9	Talaiyuttu	7.62	160	102	59	5	263.0	140.0	2.5	0	65	0.0	531	830
10	Attimarattuvalasu	7.54	167	105	64	4	248.0	148.0	2.42	0	64	0.0	544	850
11	Madattukkulam	7.34	165	82	75	3	243.0	89.0	2.45	0	58	0.0	397	620
12	Udumalaipettai	7.38	162	84	74	3	238.0	87.0	2.16	0	53	0.0	378	590.0
13	Tandoni	7.39	167	86	73	2	225.0	82.0	2.59	0	52	0.0	403	630
14	Kongalnagaram	7.34	169	87	70	2	241.0	86.0	2.48	0	59	0.0	397	620
15	Thalakkari	7.32	172	84	72	1	236.0	80.0	2.38	0	52	0.0	410	640
16	Kallipalaiyam	7.82	212	105	136	3	360.0	158.0	2.89	0	59	0.0	570	890
17	Sadaiyapalaiyam	7.79	213	102	135	4	358.0	168.0	2.87	0	58	0.0	525	820.0
18	Munduvelampatti	7.8	223	103	132	4	349.0	159.0	2.56	0	53	0.0	550	860
19	Chinnaputtur	7.75	224	104	134	5	342.0	160.0	2.48	0	56	0.0	538	840
20	Rasipalaiyam	7.72	216	108	120	4	362.0	164.0	2.96	0	54	0.0	512	800.0
21	Nagalingapuram	7.78	125	56	112	1	186.0	112.0	2.58	0	59	0.0	435	680.0
22	Kadaiyur	7.74	123	54	114	2	185.0	118.0	2.62	0	63	0.0	410	640
23	Mudalipalayam	7.85	124	58	115	2	187.0	123.0	2.57	0	65	0.0	397	620
24	Dharapuram	7.84	120	56	112	2	183.0	124.0	2.54	0	68	0.0	416	650
25	Alangiyam	7.79	128	53	110	1	182.0	125.0	2.61	0	61	0.0	403	630
26	Appayampatti	7.56	85	48	69	4	128.0	75.0	3.25	0	62	0.0	307	480.0
27	Boduvarpatti	7.54	86	46	63	5	126.0	74.0	3.16	0	63	0.0	288	450.0
28	Kappiliyapatti	7.46	82	42	64	3	125.0	78.0	3.25	0	65	0.0	275	430
29	Mandavadi	7.5	84	47	67	4	120.0	79.0	3.14	0	68	0.0	288	450
30	Kilankundal	7.52	80	49	62	3	134.0	72.0	3.2	0	69	0.0	267	420
31	Karaiyur	7.63	128	75	89	4	245.0	110.0	3.56	0	58	0.0	487	760.0
32	Peramiyam	7.64	125	76	87	3	241.0	113.0	3.45	0	52	0.0	499	780.0
33	Akkaraippalaiyam	7.69	123	78	82	5	263.0	115.0	3.51	0	54	0.0	480	750.0
34	Mulanur	7.62	127	74	83	5	254.0	118.0	3.42	0	53	0.0	461	720.0
35	Vellakkovil	7.65	126	73	84	4	248.0	116.0	3.5	0	57	0.0	474	740
36	Erachipadi	7.69	168	78	125	6	312.0	156.0	4.26	0	75	0.0	614	960
37	Lakshminayakkanvalasu	7.1	167	79	123	5	321.0	158.0	4.23	0	78	0.0	627	980
38	Aravakkurichchi	7.12	164	75	125	5	315.0	162.0	4.51	0	74	0.0	602	940
39	Pallappatti	7.32	171	71	124	6	324.0	163.0	4.2	0	72	0.0	589	920.0
40	Markkampatti	7.25	163	76	152	5	316.0	157.0	4.3	0	73	0.0	576	900.0
41	Idaiyakkottai	7.89	125	63	79	4	182.0	85.0	3.25	0	58	0.0	365	570.0
42	Javvadupatti	7.84	124	65	75	2	189.0	87.0	3.16	0	52	0.0	378	590.0
43	Navamarattupatti	7.82	123	68	78	2	184.0	82.0	3.48	0	54	0.0	371	580.0
44	Annaipatti	7.87	120	64	88	3	187.0	86.0	3.15	0	56	0.0	346	540
45	Padiyur	7.81	128	68	81	3	192.0	80.0	3.21	0	53	0.0	339	530
46	Vedasandur	7.36	116	56	74	2	186.0	102.0	2.58	0	54	0.0	435	680
47	Ayyalur	7.32	112	58	85	1	185.0	106.0	2.62	0	52	0.0	454	710.0
48	Alagapuri	7.42	105	52	84	1	182.0	105.0	2.57	0	58	0.0	467	730.0
49	Attur	7.39	108	50	82	2	180.0	112.0	2.61	0	59	0.0	442	690.0
50	Senapatipalaiyam	7.42	110	54	79	3	189.0	108.0	2.5	0	52	0.0	461	720.0
51	Tumbivadi	7.22	85	52	74	3	115.0	86.0	2.51	0	50	0.0	288	450.0
52	Chinnadarapuram	7.25	80	51	72	3	118.0	82.0	2.45	0	48	0.0	262	410.0
53	Mudalikkavundanpalaiyam	7.24	84	56	76	3	123.0	84.0	2.54	0	46	0.0	288	450
54	Andichettipalayam	7.23	86	50	71	2	120.0	79.0	2.65	0	43	0.0	269	420
55	Kandaswamipalayam	7.21	84	51	73	4	121.0	80.0	2.4	0	48	0.0	294	460
56	Kullampatti	7.62	82	46	59	2	125.0	86.0	3.12	0	57	0.0	326	510
57	Thirumanelaiyur	7.61	84	45	52	2	123.0	84.0	3.05	0	59	0.0	333	520
58	Nerur	7.51	86	42	54	2	120.0	82.0	3.07	0	53	0.0	365	570.0
59	Puliyur	7.62	80	41	57	4	124.0	87.0	3.16	0	51	0.0	339	530.0
60	Thirumukkudalur	7.68	87	43	53	3	120.0	83.0	3.22	0	54	0.0	365	570.0

Occurrence of fluoride is quite sporadic, and marked differences in concentrations occur even at very short distances. A perusal of premonsoon (June 2012) fluoride distribution shows that 32 samples have F− concentration below 2.7mg/l. Similarly, an F− concentration between 2.7 and 3.3 mg/l is observed in 17 samples of the study area. Higher concentrations above 3.3 mg/l have been noticed in 11 samples.

These high values are confined to all parts of the study area, which form the discharge areas having weathered schistose and gneissose rocks. Those samples showing an F− concentration above 3.0 mg/l in groundwaters can be considered as highly problematic as drinking water. This is in agreement with the observations made by Gupta *et al*. (1993). The area where groundwater is enriched in fluoride is underlain by different types of rocks. charnockites, and shales with alluvial formations. The concentration of F− in groundwater depends on the abundance of fluoride-bearing minerals in the rock types and their decomposition, dissociation, and dissolution activities along with residence time of the chemical reaction. High fluoride concentration in groundwater is common in areas where rocks contain fluoride-bearing minerals (Handa 1975; Wenzel and Blum 1992). The subsurface rocks in an area control the zones in which weathering affects the host rocks in minerals. High concentration of fluoride in water is common in pegmatite-rich fractured hard rock

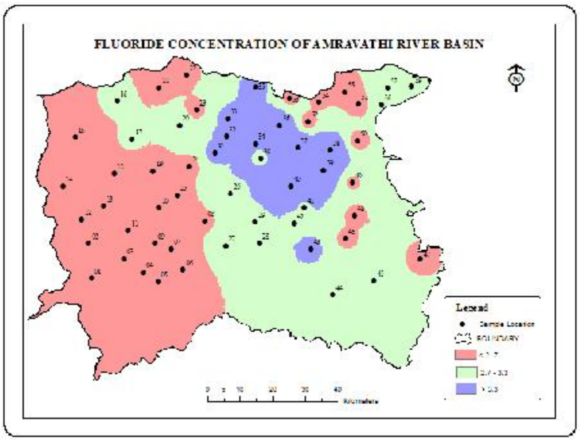

Fig .5 Isoflour map of Amaravathi River Basin

terrains, which contain minerals like fluorite, topaz, fluorite, fluorapatite, villiamite, cryolite, and fluoride-replaceable hydroxyl ions in ferromagnesium silicates (Ramesham and Rajagopalan 1985). Fluoride ions from these minerals leach into the groundwater and contribute to high fluoride concentrations. In ome cases, micas (muscovite and biotite) also contribute to fluoride content (Handa 1975). Fluorspar (fluorite [CaF2]) occurs in structurally weak planes like shear fractures, joints, and host rock–vein quartz interface. Chemical weathering (hydrolysis) of minerals results in formation of Ca and Mg carbonates that serve as good sinks for fluoride ions (Jacks *et al*. 1980). However, it is the leachable state of fluoride ions that determines the water fluoride levels, which is mainly governed by (1) pH of the draining solutions and (2) dissolved carbon dioxide in the soil. Presence of dissolved fluoride in groundwater is possible only under favorable physicochemical conditions and with a sufficient residence time (Kullenberg and Sen 1973; Handa 1975). In the present case, the study area comprised partly sedimentary and partly crystalline formations (Fig. 3). They are unconformably overlain by sandy and clayey soils of recent to subrecent age. The sedimentary rock types include limestone and quartzite and sand.. The crystalline formations are charnockite and granitic gneiss. Crystalline charnockite and granitic gneiss of Archean age have been intruded by amphibolites, dykes of dolerite, and occasionally, veins of quartz and pegmatites. The gneisses of this area have quartz, feldspars (potash feldspars and albite), hornblende, biotite, etc. The acid charnockite of this area has quartz, k-feldspars, hypersthene, and biotite minerals of coarse-grained nature, which are potential sources of fluoride. Major fluoridebearing minerals that present in the igneous and metamorphic rocks are fluorapatite, biotite, hornblende, etc. Sedimentary horizons also have apatite as an assessor mineral, and fluorite also often occurs as cement in some sandstones Fluorite seems to be the most likely source along with minor contributions from hornblende gneisses. From natural sources, a considerable amount of fluoride may be contributed through anthropogenic activities. Phosphatic fertilizers, which are extensively used in agriculture, often contain fluoride as an impurity that can leach down to the saturated zone. Arid to semiarid climatic conditions are quite conducive for chemical weathering, which results in enhanced salinity and fluoride abundances in phreatic water system. An arid climate with low rainfall and high evapotranspiration and insignificant natural recharge cumulatively lead to salinization of groundwater and precipitation of calcite. Soils become more alkaline with a very high pH that affects the solubility of calcite (Ramasesha *et al*. 2002). These conditions lower the activity of Ca and increase the Na–Ca ratios, thus allowing fluoride to concentrate in the groundwater environment. Bedrock containing fluoride minerals is generally responsible for the high concentration of this ion in groundwater (Handa 1975). The fluoride concentration in ground water of the study area varies between 2.1 and 4.5 mg/l, with an average value of 2.98 mg/l. The maximum allowable limit of fluoride is 1.5 mg/l according to the WHO (1993).

Hydrochemistry of fluorine

Fluorine is 13th in the order of abundance of elements in the earth's crust. Fluoride is physiologically important, and its extremely higelectronegativity makes it highly reactive, and therefore, it occurs in a number of naturally combined forms. Its abundance in the continental crust is about 626 μg/g (Periakali *et al*. 2001). Chemically, OH− and fluoride are negatively charged and also possess almost similar ionic radii. Hence, during the chemical reaction, fluoride can easily replace OH− ions in many rock-forming minerals. A better understanding of fluorine geochemistry in the aquatic environment under specific geographic and geologic conditions is necessary for evaluating the contamination process. During the process of chemical weathering, dissolution of fluoride species in the natural water is controlled by calcium and governed by thermodynamic principles. The calcium ion activity in the natural environment is determined mainly by carbonate ion, which forms insoluble calcite. The equilibrium constant with respect to calcite can be evaluated from the following reactions.

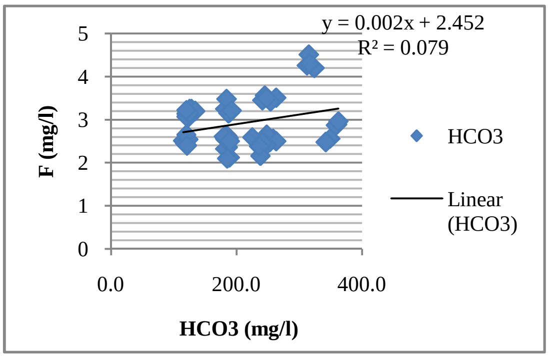

Fig .6 Relationship of fluoride and HCO[3]

$CaCO3 + H+ \rightarrow Ca2+ + HCO3-$ (1)

$$K_{CaCO3} = \frac{a(Ca^{2+})\cdot a(HCO_3^-)}{a(H^+)} = 97 \text{ at } 25^\circ C \qquad (2)$$

The fluoride concentration in groundwater is controlled bimineral fluorite as per the formula given below (Brown and Roberson 1977):

$$CaF2 = Ca2+ + 2F- \quad (3)$$

$$KCaF2 = a\,(Ca2+)\,.a(F^{-})^{2} = 10^{-10.58} \text{ at } 25°C \quad (4)$$

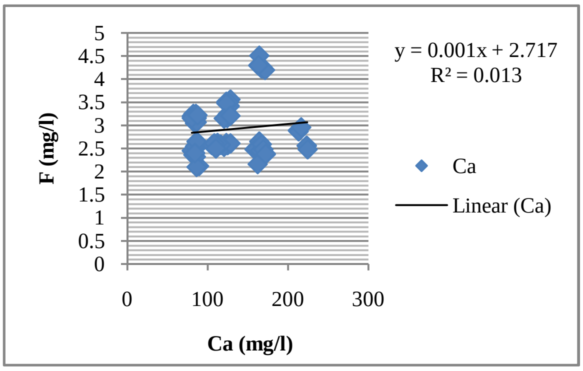

Fig .7 Relationship of fluoride and Ca

where *K* represents the solubility product constant and *a* denotes the activities of the corresponding ions. Thus, the activities of calcium and fluoride are negatively correlated. Minerals rich in calcite (CaCO3*)* also favor the dissolution of fluoride from fluoride-rich minerals. Decreasing Ca concentrations are found under alkaline conditions with a corresponding rise in Na. Therefore, fluoride can accumulate in water if soils and groundwater are low in calcium. In the present case, a negative correlation is seen between calcium and fluoride (Fig. 7).

In the study area, all groundwater samples have EC values within the permissible limits for drinking water standards (BIS 1991). Fluoride has a unique chemical behavior toward most of the anions and can be easily replaced even under normal pressure and temperature conditions (Wenzel and Blum 1992). The less soluble products of fluoride in the presence of Ca make the dissolution activity more effective. Fluoride shows a negative correlation with Ca and HCO3 as well as Na and a positive correlation with pH in the groundwater samples analyzed in the study area (Figs. 6, 7, and 8). Thus, the ranges of such ionic species may suggest favourable chemical conditions for the fluoride dissolution process in the study area.

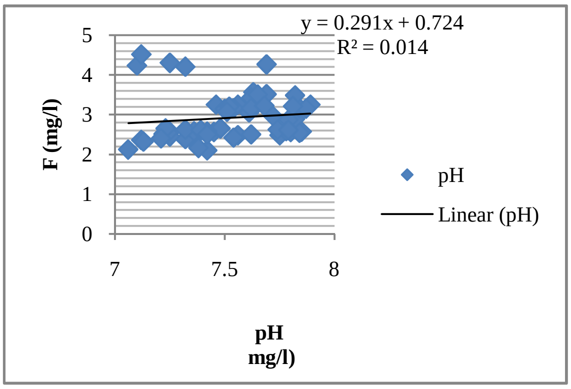

Fig .8 Relationship of fluoride and pH

Impact on human health

Fluoride in water can be a blessing or a hazard depending on the concentration levels. The Bureau of Indian Standards (BIS) and the Indian Council of Medical Research prescribe a fluoride concentration of 1.0 g/l as the desirable limit and 1.5 mg/l as the maximum permissible limit in drinking water, if there is no alternate source. These guidelines vary depending on the climate and the total fluoride intake from other sources, since the absorption of fluoride by body fluids depends on temperature. The study area falls in the climatic zone where an average summer temperature is greater than 25°C and the average drinking water consumption is higher than 4 l/day. For these population groups, drinking water containing less than 0.6 mg F/l is fit for consumption (Deshkar *et al*. 1999). It is a deadly disease with no cure so far. In the study area, villagers who consume nonpotable high fluoride content water may suffer from yellow, cracked teeth, joint pains, and crippled limbs and also age rapidly. Assimilation of fluorine by the human body from potable water at the level of 1 mg/l enhances bone development and prevents dental carries. It is found to cause fluorosis when it exceeds the limit of 1.2 mg/l (Kundu *et al*. 2001).

CONCLUSION

Presence of fluoride-bearing minerals in the host rocks and their interaction with water are considered to be the main causes for fluoride enrichment in groundwater. Decomposition, dissociation, and dissolution are the main chemical processes responsible for mobility and transport of fluoride into groundwater. Chemical weathering under arid to semiarid conditions with relatively high alkalinity and long residence time of interaction seem to have favored high concentration of fluoride in groundwater. Geochemical behavior of groundwater from the study area suggests that the high fluoride content groundwater contains low levels of Ca and has high alkalinity.

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