Fluoride Distribution in Aquatic Environment in Vicinity of Aluminum Industry and its Correlation with Rainwater Chemistry and Weather Parameter. A case study of Renukoot, District Sonbhadra,

Uttar Pradesh

6 Abstract:

Fluorine is a highly reactive element and readily hydrolyzes to form hydrogen fluoride and oxygen. Hydrogen fluoride reacts with many materials both in the vapor phase and in aerosols. Fluorides are naturally-occurring components of rocks and soil and are also found in air, water, plants, and animals. They enter the atmosphere through volcanic emissions and the resuspension of soil by wind. Volcanoes also emit hydrogen fluoride and some fluorine gas from aluminum industry. The resultant fluorides are typically nonvolatile, stable compounds. The chemical composition of rainwater is good indicators for assessment of rainwater quality that is greatly changed due to rapid industerization. To monitoring of fluoride content in precipitation and atmospheric pollution characteristics. Collect 57 rainwater samples in rainy season in pre-cleaned and sterilized polyethylene bottles of two litre capacity from roof of the local society of Hindustan Aluminum Company, of Renukoot, district Sonbhadra. The collected samples were analyzed for pH, Eect Conductivity and Fluorides.pH and EC were determined by pH, conductivity meter, and Fluoride through Ion Selective Electrode.

Keywords Fluoride Aquatic environment chemical composition of Rainwater chemistry

20 Introduction

Fluorides are naturally-occurring components of rocks and soil and are also found in air, water, plants, and animals. They enter the atmosphere through volcanic emissions and the resuspension of soil by wind. Volcanoes also emit hydrogen fluoride and some fluorine gas from industriz. Fluorine is a highly reactive element and readily hydrolyzes to form hydrogen fluoride and oxygen. Hydrogen fluoride reacts with many materials both in the vapor phase and in aerosols. Marine aerosols also release small amounts of gaseous hydrogen fluoride and fluoride salts into the air (Friend 1989). Anthropogenic fluoride emissions include the combustion of fluorine containing materials, which releases hydrogen fluoride, as well as particulate fluorides, into the air. Coal contains small amounts of fluorine, and coal-fired power plants constitute the largest source of anthropogenic hydrogen fluoride emissions. According to the Toxic Chemical Release Inventory (TRI), in 2001, the largest contributing industrial sectors were electrical utilities (TRI01 2003). Total air emissions of hydrogen fluoride by electrical utilities in 1998, 1999, 2000, and 2001 were reported as 64.1, 58.3, 58.3, and 55.8 million tons, respectively. Major sources of industrial fluoride emissions are aluminum production plants and phosphate fertilizer plants; both emit hydrogen

## UNDER PEER REVIEW

fluoride and particulate fluorides (EPA 1998b). Other industries releasing hydrogen fluoride are: chemical
production; steel; magnesium; and brick and structural clay products. Hydrogen fluoride would also be
released by municipal incinerators as a consequence of the presence of fluoride-containing material in the
waste stream. Hydrogen fluoride is one of the 189 chemicals listed as a hazardous air pollutant (HAP) in
Title III, Section 112 of the Clean Air Act Amendments of 1990. Maximum achievable control technology
(MACT) emission standards are being developed by the EPA for each HAP. Other anthropogenic sources
of fluoride in the environment are coal combustion causing air pollution, and waste production by various
industries, including steel, aluminum, copper and nickel smelting; and the production of glass, phosphate
fertilizers, brick and tile (Pickering, 1985; Skjelkvsle, 1994). Simultaneous air and groundwater pollution
by Heand As, due to coal combustion, causes serious health diseases over large areas of southern China
(Zheng et al., 1996; An et al., 1997; Finkelman et al., 2002) and Inner Mongolia (Wang et al., 1999;
Smedley et al., 2002), although Follows not coexist with As in polluted groundwater in most other areas.
In the atmosphere, gaseous hydrogen fluoride will be absorbed by atmospheric water (rain, clouds,
fog, snow) forming an aerosol or fog of aqueous hydrofluoric acid. It will be removed from the
atmosphere primarily by wet deposition. Particulate fluorides are similarly removed from the atmosphere
and deposited on land or surface water by wet and dry deposition. Atmospheric precipitation weathers
crustal rocks and soil, but dissolves out very little fluoride; most of the fluoride mobilized during
weathering is bound to solids such as clays. Upon reaching bodies of water, fluorides gravitate to the
sediment (Carpenter 1969). Fluorides have been shown to accumulate in some marine aquatic organisms
(Hemens and Warwick 1972). When deposited on land, fluoride is strongly retained by soil, forming
complexes with soil components. Fluorides in soils are transported to surface waters through leaching or
runoff of particulate-bound fluorides. Leaching removes only a small amount of fluorides from soils.
Fluorides may be taken up from soil and accumulate in plants. The amount of fluorides accumulated
depends on the type of plant and soil and the concentration and form of fluoride in the soil. Fluorides may
also be deposited on above-ground surfaces of the plant. Tea plants are particularly known to accumulate
fluoride, 97% of which is accumulated in the leaves (Fung et al. 1999). Fluoride accumulates primarily in
the skeletal tissues of terrestrial animals that consume fluoride-containing foliage. However, milk and
edible tissue from animals fed high levels of fluorides do not appear to contain elevated fluoride
concentrations (NAS 1971a).
In natural water, fluoride forms strong complexes with aluminum in water, and fluorine chemistry
in water is largely regulated by aluminum concentration and pH (Skjelkvale 1994). Below pH 5, fluoride is
almost entirely complexed with aluminum and consequently, the concentration of free F is low. As the pH

increases, Al-OH complexes dominate over Al-F complexes and the free F levels increase. Fluoride forms
stable complexes with calcium and magnesium, which are present in sea water. Calcium carbonate
precipitation dominates the removal of dissolved fluoride from sea water (Carpenter 1969). Fluorine is
incorporated into the calcium salt structure and removed from solution when the latter precipitates.
Fluoride occurs in soil in a variety of minerals and complexes with aluminum, iron, and calcium. Fluorides
occur predominantly as aluminum fluorosilicate complexes in acidic soils and calcium fluoride in alkaline
soils. The availability of these soluble complexes increases with decreasing pH (Fung et al. 1999;
Shacklette et al. 1974). This explains why acidic soils have both higher water-soluble fluoride and higher
extractable aluminum levels. The retention of fluoride in alkaline soils depends largely upon the aluminum
content of the soil.
Material and Methods
A. Site Description of the Study Aera
Sonbhadra is the largest district of Uttar PradeshIt has geographical area 6788.0 sq km, average height
from sea level 285 feet, average, rainfall 1036.6 mm and temperatures in summer 10-45°C in winter 8-25
<sup>0</sup> C Renukut is located at 24°12' of Northern latitude and 83°02' Eastern latitude. It has an average
elevation of 283 meters (931 feet). Summers peak in May and June. Renukut has the largest integrated
Aluminum plant in Asia. Monsoon generally sets in the First week of June and last up to last week of
September. 90-95 percent rainfall is received during June to September. The temperature begins to rise
from the first week February and reaches it maximum by the middle of May or end of June.
Sampling of Rainwater
Rain water was collected in rainy season around Hindustan Aluminum Company, Uttar Pradesh. The time
schedule for collection of rain water was 2 <sup>th</sup> June to 8 <sup>ht</sup> August 2008. The total samples were 33.In
Hindustan Aluminum Company Ltd the sample collector was placed about 10 m above the ground level
on the roof of the quarter of the employee. Each collector had a 19 cm diameter plastic fennel tightly fitted
to 2.5L borosilicate glass bottle. The rain water samples were then filtered and only aqueous phase was
analyzed, between two rainfall events collectors were properly rinsed with distilled water.
Methods of Analysis of cations and anions in Aquatic Environment
The pH was measured with a digital pH meter using reference (KCl) and glass electrodes standardized
with pH 4.0 and pH 9.2 reference buffers before and after pH determination. Since pH is influenced by
extreme values of rainfall amount, rainfall weighted mean values of pH were used instead of arithmetic
means. Electrical conductivity is used for determination of total concentration of soluble salts or ionized

#### JNDER PEER REVIEW

159

129 samples collected from Renukoot ranging from 4.9-8.1 with mean 6.5. More than 35% incidence of rainwater was observed acidic to the influence of industries (aluminum and thermal power plant). 130 Neutral pH was observed in latter incidences of rain of Renukoot. The Electrical conductivity of rain water 131 of Renukut ranged from 0.017-0.471 dSm<sup>-1</sup>. 132 **Anions Chemistry of Aquatic Environment** 133 Samples of rain water were collected on 1<sup>st</sup> week of June to 4<sup>th</sup> week of August, 2008 in Renukut, Uttar 134 Pradesh. The values of anionic composition viz., carbonate, bicarbonate, chloride, sulphate and residual 135 136 sodium carbonate (RSC) are given in Table 1.It was revealed from the data that the chloride and sulphate 137 were found in all the rain water samples in each locations; but carbonate was not found in rain water samples and Bicarbonate was found enriched with 100% water samples it is also revealed that in 138 comparison with other anions, HCO<sub>3</sub> was found in highest amount followed by chloride and sulphate. In 139 waters, containing high concentration of bicarbonate ions, there is a tendency for Ca<sup>2+</sup> and Mg<sup>2+</sup> to 140 precipitate as carbonates. This can be shown as RSC (residual sodium carbonate) =  $(CO_3^- + HCO_3^-) - (Ca_3^{2+} + HCO_3^-)$ 141 + Mg<sup>2+</sup>), where CO<sup>=</sup><sub>3</sub>, HCO<sup>-</sup><sub>3</sub>, Ca<sup>2+</sup> and Mg<sup>2+</sup> represents the concentrations in meq L<sup>-1</sup> of respective ions. 142 RSC from carbonate is more harmful than bicarbonate. But the values of RSC in most of the samples of 143 144 rain water in studies area were negative; thus precipitation problems will not occur in future. Moreover, 145 rainfall, soil texture and plant species to be grown has great impact in deciding the limits of RSC for suitability of irrigation water in a particular area. For example studies conducted at CSSRI has revealed 146 that water up to RSC 5.0 meg L<sup>-1</sup> can be used where rainfall is 700 to 900 mm per annum. The limit of 147 RSC (5.0 meg L<sup>-1</sup>) in rain water had not crossed in Renukut but only 10.5% rain water samples in Varanasi 148 149 had crossed the limit. 150 **Cations Chemistry of Aquatic Environment** 151 Results given in Table 1 presented the concentration of cations, viz., sodium, potassium, calcium and 152 magnesium in rain water samples during South West monsoon period. It was revealed from the data that K<sup>+</sup> and Mg<sup>2+</sup> were found in all the rain water samples account more than 75 %. The order of basic cations 153 (mean) found in rain water was as follows:  $Mg^{2+} \ge K^+ > Na^+ \ge Ca^{2+}$ . The mean total basic cations ( $Na^+ +$ 154  $K^+ + Ca^{2+} + Mg^{2+}$ ) (4.03 meg L<sup>-1</sup>) Thus, neutralization of acidic anions (viz., Cl<sup>-</sup>, SO<sub>4</sub> etc.) by basic anions 155 (CO<sub>3</sub> and HCO<sub>3</sub>) was noticed higher in water samples. The alkaline properties of the particulate matter in 156 157 rain water were responsible for neutralizing the acidic ions and consequently, for the observed increase in 158 pH. The cations are mainly of soil origin and predominantly present in giant size range. The range of

cations in rain water samples were as follows: 0.00-1.44 meg L<sup>-1</sup> of Na<sup>+</sup>, 0.2-2.3 meg L<sup>-1</sup> of K<sup>+</sup>, 0.00-1.44

### UNDER PEER REVIEW

meq L<sup>-1</sup> of Ca<sup>2+</sup> and 0.02-2.3 meq L<sup>-1</sup> of Mg<sup>2+</sup>. Thus, higher range of Na<sup>+</sup> and K<sup>+</sup> in rain water were observed in samples. The principal cations present in rain water are Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup> and K<sup>+</sup>. The alkali hazard involved in the use of water for irrigation determine the absolute and relative concentration of the cations. If the proportion of sodium is high, the alkali hazard is high. If the calcium and magnesium is high, the hazard is low. The Sodium Adsorption Ratio (SAR) of a solution or water is related to the adsorption of sodium by the soil. This is expressed by the equation:

SAR = 
$$\frac{Na^{+}}{\sqrt{\frac{Ca^{2+} + Mg^{2+}}{2}}}$$

Where all ionic concentration are expressed in meq  $L^{-1}$ . It was revealed from Table 1 that SAR values of rain water varied from 0.0-1.8. It was noticeable that SAR values were < 10 and EC were < 2.0 d Sm<sup>-1</sup> in all the samples of the rain water. Thus, this rain water is suitable for safe irrigation, without any sodicity problems.

#### Fluoride Distribution in Aquatic Environment

Presence of fluoride in pure rain water is depended on the atmospheric deposition from soil dust and industrial emission. Thus, the soluble fluoride in upper crust of the earth and the fluoride emission as a dust from industry, e.g. aluminum industry are the source of fluoride in rain water. It is not essential to crops, but it can cause toxicity of sensitive crops at higher level. The data on fluoride content in rain waters of Renukut have been presented in Table 1. The fluoride content in rain water ranged from from 0.02-0.45 meq L<sup>-1</sup>. Maximum fluoride in rain water was noticed in the month of July. The fluoride contamination was noticed in all the incidences of rain water samples of studies area. The mean value (0.17 meq L<sup>-1</sup>) of fluoride in rain water was represented in Fig.1

.Considerably higher centrations of fluoride around 0.3 mg L<sup>-1</sup> in rain water have been also reported from two sites in Uttar Pradesh (Satsangi *et al.*, 1988) and Madhya Pradesh (Singh *et al.*, 2001). Das *et al.* (1981) reported 0.1 mg L<sup>-1</sup> for monsoon rain water at Bhopal in Central India. According to these authors, a large fraction of the dissolved material in the rain water in their investigations, including the fluoride, may be derived from local soil dust and and Patel (1999) have presented an extensive investigation from eastern Madhya Pradesh comprising 13 sites with a mean volume-weighted concentration of fluoride in the precipitation ranging from 0.05 to 0.22 mg L<sup>-1</sup>, the latter being from a site close to an industrial Aluminum plant. Thus, the higher ount of fluoride in Renukut is obviously influenced by Al factory (Hindustan Aluminum Company, LTD).

#### Correlation Study of Fluoride with Chemical Composition and Weather Parameters

The data on correlation study of fluoride content in rainwater with chemical composition of rainwater and weather parameters of Renukut have been presented in Table 2 and Table 3 respectively. Fluoride content in rain waters were negatively correlated with Ca<sup>2+</sup> and Mg<sup>2+</sup>. An inverse relationship was also found between F and Ca<sup>2+</sup> in some investigations (Handa, 1975). It was revealed from the data that the fluoride content in rain water was negatively correlated with the atmospheric temperature and evaporation in study the regions of Renukut. Thus, due to increase in atmospheric temperature, rate of evaporation increases which leads to reduction of fluoride concentration in rain water.

197 Conclusion

The Research data indicated that the heavy air pollution was found around Hindustan Aluminum Company, LTD. which causes pollutant as well other contaminants in Aquatic Environment. So, the findings cleared that the Aluminum plant in Renukoot is the main sources of air pollution in atmosphere as well as in rainwate—he standard method to be used of analysis of Rainwater for monitoring of rainwater chemistry and fluoride. The fluoride in rain water was analyzed by ion selective electrode meter. Presence of fluoride in rain water is depended on the atmospheric deposition from soil dust and industrial emission. The fluoride contamination was noticed in all the rainwater samples of study area. Maximum fluoride contamination in rain water was observed in the month of July. The fluoride content in rain water of Renukut ranged from 0.02 0.45 meq L<sup>-1</sup> High fluoride contamination due to industrial emission from aluminum industry, chemical Industries, hydel power projects, thermal power projects. To name a few there exists HINDALCO India's largest aluminum company, and then in the same belt around some 40 km away is India largest NTPC plant. Higher fluoride concentration (3.23 mg/L) in rain water was observed in Renukut. Rainwater can be treate—applying appropriate technology to remove the impurities.

**Table 1: Rainwater Chemistry of Renukut** 

S. No.	Date of Sampling	pН	EC dSm <sup>-1</sup>	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>++</sup>	CO <sub>3</sub>	HCO <sub>3</sub>	CL-1	SO <sub>4</sub>	Fluoride	RSC	SAR
1	02/06/2008	6.4	0.107	0.00	2.00	0.00	0.0	0.4	1.0	0.01	0.18	-0.06	0.0
2	08/06/2008	7.3	0.093	0.00	0.79	0.00	0.0	0.5	0.4	0.00	0.13	-0.01	0.0
3	10/06/2008	5.7	0.112	0.00	1.10	0.00	0.0	0.6	1.2	0.03	0.20	-0.02	0.0
4	13/06/2008	7.1	0.158	0.00	2.00	0.00	0.0	0.8	1.3	0.17	0.23	-0.04	0.0
5	14/06/2008	8.1	0.227	0.00	2.20	0.00	0.0	1.9	1.5	0.12	0.20	-0.01	0.0
6	16/06/2008	7.1	0.099	0.00	0.90	0.00	0.0	0.4	1.1	0.19	0.22	-0.02	0.0
7	19/06/2008	7.9	0.175	1.12	1.40	1.12	0.0	1.1	2.1	0.11	0.07	-1.13	1.5
8	22/06/2008	7.7	0.166	0.00	1.60	0.00	0.0	1.2	2.5	0.09	0.06	-0.01	0.0
9	23/06/2008	7.9	0.194	0.00	1.50	0.00	0.0	0.3	2.1	0.02	0.21	-0.04	0.0
10	24/06/2008	5.8	0.078	0.80	0.75	0.80	0.0	0.2	1.9	0.02	0.34	-0.82	1.2
11	26/06/2008	5.3	0.074	0.32	0.15	0.32	0.0	0.4	1.6	0.06	0.28	-0.32	0.8
12	27/06/2008	5.0	0.055	1.28	0.18	1.28	0.0	0.3	0.7	0.02	0.29	-1.28	1.6
13	29/06/2008	4.9	0.085	0.48	0.41	0.48	0.0	0.4	1.6	0.07	0.26	-0.48	1.0
14	30/06/2008	5.8	0.056	1.44	0.02	1.44	0.0	0.4	1.3	0.06	0.26	-1.43	1.7
15	05/07/2008	6.5	0.044	1.28	0.49	1.28	0.0	0.3	0.4	0.01	0.26	-1.29	1.6
16	07/07/2008	5.8	0.047	0.48	0.11	0.48	0.0	0.4	0.4	0.08	0.26	-0.47	1.0
17	09/07/2008	5.3	0.023	0.96	0.81	0.96	0.0	0.3	0.0	0.01	0.26	-0.98	1.4
18	11/07/2008	5.8	0.017	0.64	0.41	0.64	0.0	0.4	0.0	0.01	0.26	-0.64	1.1
19	12/07/2008	5.8	0.054	0.96	0.59	0.96	0.0	0.3	1.9	0.10	0.13	-0.97	1.4
20	15/07/2008	5.5	0.024	0.32	0.14	0.32	0.0	0.3	1.8	0.01	0.08	-0.31	0.8
21	16/07/2008	6.8	0.129	0.8	0.36	0.80	0.0	1.2	1.3	0.02	0.07	-0.77	1.3
22	18/07/2008	7.3	0.074	0.96	1.10	0.96	0.0	0.3	2.2	0.01	0.10	-0.99	1.4
23	20/07/2008	6.8	0.040	0.80	0.12	0.80	0.0	1.9	1.8	0.01	0.09	-0.73	1.3
24	23/07/2008	6.4	0.037	0.32	0.42	0.32	0.0	0.2	2.0	0.01	0.11	-0.32	0.8
25	25/07/2008	6.6	0.054	1.12	0.97	1.12	0.0	0.4	1.9	0.08	0.45	-1.14	1.5
26	27/07/2008	6.2	0.023	0.48	0.57	0.48	0.0	0.5	2.0	0.03	0.04	-0.48	1.0
27	28/07/2008	6.2	0.040	1.44	0.78	1.44	0.0	0.6	0.2	0.08	0.09	-1.45	1.7
28	30/07/2008	6.3	0.025	0.64	0.12	0.64	0.0	1.1	0.2	0.03	0.07	-0.60	1.1
29	06/08/2008	6.5	0.017	0.32	1.90	0.32	0.0	0.4	1.8	0.00	0.08	-0.38	0.7
30	17/08/2008	6.4	0.019	1.44	1.50	1.44	0.0	0.2	0.8	0.00	0.06	-1.48	1.7
31	23/08/2008	6.8	0.069	1.28	2.30	1.28	0.0	1.2	1.8	0.03	0.02	-1.32	1.6
32	24/08/2008	6.7	0.034	1.60	1.20	1.60	0.0	0.8	0.9	0.03	0.12	-1.61	1.8
33	29/08/2008	6.8	0.038	0.32	0.56	0.32	0.0	0.3	1.3	0.03	0.09	-0.33	0.8
F	Range	4.9 -8.1	0.017-0.47	0.0-1.44	0.0-00	0.0- 1.44	0-0	0.2-1.9	0.20- 2.5	0.01-0.17	0.02-0.45	-0.011.61	0.0-1.8
N	Mean	6.5	0.080	0.65	0.67	0.65	0-0	1.30	0.55	0.01	0.170	0.050	0.95
	±SD.	0.83	0.056	0.53	75.73	0.52	0.0	0.71	0.23	0.01	0.102	0.048	0.62
	CV.	12.1	0.70	80.62	00	80.30	0.0	54.76	41.81	100.0	60.0	96.00	65.06

Table 2: Correlation Study of Fluoride with Chemical Composition of Rainwaters

Parameters	pН	EC	CO <sub>3</sub> <sup>2</sup> -	HCO <sub>3</sub> -	RSC	Cl	SO <sub>4</sub> <sup>2-</sup>	Ca <sup>2+</sup>	$Mg^{2+}$	Na <sup>+</sup>	$\mathbf{K}^{+}$	SAR	Fluoride
pН	1.000												
EC	0.637**	1.000											
$CO_3^{2-}$	0.315	0.382*	1.000										
HCO <sub>3</sub> -	1.000**	0.444*	-0.121	1.000									
RSC	0.236	0.455	0.215	0.087	1.000								
Cl	0.340	0.368*	0.215	0.163	0.204	1.000							
SO <sub>4</sub> <sup>2-</sup>	0.267	0.499**	-0.085	0.224	0.216	0.109	1.000						
Ca <sup>2+</sup>	-0.244	-0.462*	-0.224	-0.065	-0.999**	-0.209	-0.218	1.000					
$Mg^{2+}$	0.592**	0.524**	0.162	0.266	-0.999**	0.270	0.205	-0.206	1.000				
Na <sup>+</sup>	-0.244	-0.462	-0.224	-0.065	0.170	-0.209	-0.218	1.000	-0.206	1.000			
K <sup>+</sup>	0.592**	0.524**	0.162	0.266	0.170	0.270	0.205	-0.206	1.000	-0.206	1.000		
SAR	-0.361*	-0.581**	-0.276	-0.103	-0.950**	-0.166	-0.289	0.958*	-0.366	0.958**	-0.366*	1.000	
Fluoride	-0.358	0.051	0.072	-0.352	-0.007	-0.204	0.208	0.782*	-0.821*	0.002	-0.202	-0.016	1.000

<sup>\*</sup> Correlation is significant at the 0.05 level (2 tailed) \*\* Correlation is significant at the 0.01 level (2 tailed)

**Table 3: Correlation Study of Fluoride with Weather Parameters** 

Parameters	Rainfall	Tempe	rature	R.	H.	Sunshine	Evaporation	Fluoride	
		Max.	Mini.	Max.	Mini.		_		
Rainfall	1.000								
Temp. Max.	0.246	1.000							
Temp. Mini.	0.684*	0.782**	1.000						
R H Max.	0.203	-0.654	-0.298	1.000					
R H Mini.	0.641*	-0.211	0.306	-0.751*	1.000				
Sunshine	-0.469	0.616	0.062	-0.751*	-0.778**	1.000			
Evaporation	0.096	0.183	0.626	-0.919	-0.532	0.671*	1.000		
Fluoride	-0.456	0.893*	-0.343	-0.032	-0.324	0.596*	-0.646*	1.000	

<sup>\*</sup> Correlation is significant at the 0.05 level (2 tailed) \*\* Correlation is significant at the 0.01 level (2 tailed)

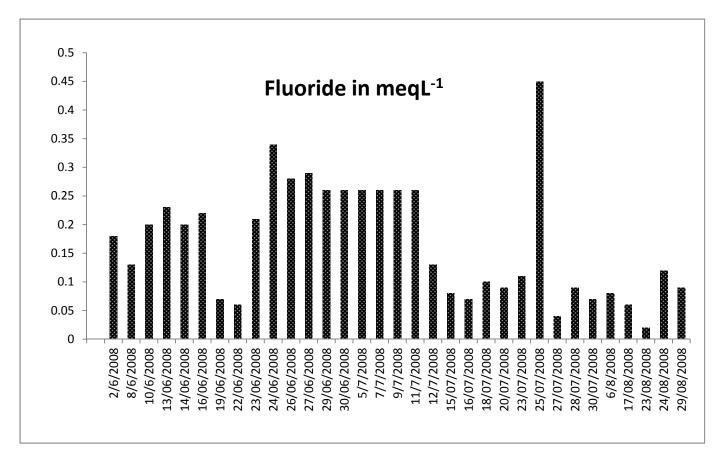


Fig.1 Fluoride Contamination in Rainwater of Renukoot

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