

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, India

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This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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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. Fluorine gas pollutes the atmospheric environment originating from aluminium smelting plant operating at Renukoot, district Sonbhadra. Volcanic emissions also emit hydrogen fluoride. A detailed investigation undertaken during 2008-2012 to objective fluoride Distribution in Aquatic Environment in Vicinity of Aluminum Industry and its Correlation with Rainwater Chemistry and Weather Parameter. For this investigation Collect 57 rainwater samples in rainy season in pre-

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cleaned and sterilized polyethylene bottles of two litre capacities from roof of the local society of Hindustan Aluminum Company Renukoot, district Sonbhadra. The collected samples were analyzed for pH, Electrical Conductivity, Sodium, Potassium, Calcium, Carbonates, Bicarbonates, Chlorine, Sulphate, Fluoride, Residual Sodium Carbonates, and Sodium Adsorption Ratio with standard procedures.

Keywords: Fluoride; aquatic environment; rainwater; Uttar Pradesh.

1. 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 industrialization. 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 [1]. 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 [2]. 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 fluoride and particulate fluorides [3]. 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 [4,5]. Simultaneous air and groundwater pollution by F₋ and As, due to coal combustion, causes serious health diseases over large areas of southern China [6] An et al., 1997; [7] and Inner Mongolia [8,9], although F₋ does 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 [10]. Fluorides have been shown to accumulate in some marine aquatic organisms [11]. 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 [12]. 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 [10]. 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 [12,13]. 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.

2. MATERIALS AND METHODS

2.1 Site Description of the Study Area

Sonbhadra is the largest district of Uttar Pradesh. It 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°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 its maximum by the middle of May or end of June. The location map of the study area has been shown in Fig. 1.

2.2 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 2nd June to 8th 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 funnel 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.

2.3 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 constituents in water. It is related to the sum of cations and anions as determined chemically. The E.C. of water was measured with Conductivity Bridge using standard potassium chloride solution for calibration and determination of cell constant. Since it is influenced by temperature, maintenance of temperature is important on account of the fact that specific conductance is increased by 2% per degree centigrade rise of temperature. The temperature in water samples were maintained in water bath at 25°C. Calcium (Ca²⁺), magnesium (Mg²⁺), carbonate (CO₃²⁻), bicarbonate (HCO₃⁻) and chloride (Cl⁻) were analyzed by volumetric titration methods, sodium (Na⁺) and potassium (K⁺) were measured using the flame photometer, sulphate (SO₄²⁻), were determined by spectrophotometric technique as per the methods described by the American Public Health Association [14]. The analyses were completed within a week from the date of collection of the water samples at the Department of Soil Science and Agricultural Chemistry, Institute of Agricultural Sciences, Banaras Hindu University, Varanasi. Table 1 provides physico-chemical data of rainwater samples of the present study.

2.4 Analysis of Fluoride in Aquatic Environment

Fluoride content in water was determined electrochemically, using the direct ion selective electrode method. In this method, 25 mL of water sample and 25 mL of the TISAB solution (total ionic strength adjustment buffer) were taken in a 100 mL plastic beaker. The ratio of aliquot and TISAB Solution should be 1:1. After

proper calibration, the fluoride electrode was dipped in the sample and concentration in mg/L was displayed on the screen.

Preparation of TISAB solution for F⁻ 58 mL of glacial acetic acid and 12g of sodium citrate were added to 300 mL distilled water and pH of the solution was adjusted to 5.2 using 6N sodium hydroxide and then cooled and diluted to 1000 mL.

3. RESULTS AND DISCUSSION

The present investigation entitled "Fluoride Distribution in Aquatic Environment in Vicinity of Aluminum Industry and its Correlation with Chemistry of Rainwater and Weather Parameter. A case study of Renukoot, District Sonbhadra of Uttar Pradesh" was carried out in the Department of Soil Science & Agricultural Chemistry, Institute of Agricultural Sciences, Banaras Hindu University to investigate the status of fluoride in rainwater, Renukoot (Sonbhadra district).

3.1 Electrochemical Characterization of Rain Water

The pH value of tested water samples refers to the intensity of the acidic or alkaline condition of a solution [15]. The data on important electrochemical properties of rain water, viz., pH and EC of rain water of Renukoot, collected in different days have been presented in the Table 1. The pH values of rain water samples collected from Renukoot ranging from 4.9-8.1 with mean 6.5. More than 35% incidence of rainwater was observed acidic due to the influence of industries (aluminum and thermal power plant). Neutral pH was observed in latter incidences of rain of Renukoot. The Electrical conductivity of rain water of Renukoot ranged from 0.017-0.471 dSm⁻¹.

3.2 Anions Chemistry of Aquatic Environment

Samples of rain water were collected on 1st week of June to 4th week of August, 2008 in Renukoot, Uttar Pradesh. The values of anionic composition viz., carbonate, bicarbonate, chloride, sulphate and residual sodium carbonate (RSC) are given in Table 1. It was revealed from the data that the chloride and sulphate 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

comparison with other anions, HCO₃⁻ was found in highest amount followed by chloride and sulphate. In waters, containing high concentration of bicarbonate ions, there is a tendency for Ca²⁺ and Mg²⁺ to precipitate as carbonates. This can be shown as RSC (residual sodium carbonate) = (CO₃⁼ + HCO₃⁼) - (Ca²⁺ + Mg²⁺), where CO₃⁼, HCO₃⁼, Ca²⁺ and Mg²⁺ represents the concentrations in meq L⁻¹ of respective ions. RSC from carbonate is more harmful than bicarbonate. But the values of RSC in most of the samples of rain water in studies area were negative; thus precipitation problems will not occur in future. Moreover, 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 that water up to RSC 5.0 meq L⁻¹ can be used where rainfall is 700 to 900 mm per annum. The limit of RSC (5.0 meq L⁻¹) in rain water had not crossed in Renukoot but only 10.5% rain water samples in Varanasi had crossed the limit.

3.3 Cations Chemistry of Aquatic Environment

Results given in Table 1 presented the concentration of cations, viz., sodium, potassium, calcium and magnesium in rain water samples during South West monsoon period. It was revealed from the data that K⁺ and Mg²⁺ were found in all the rain water samples account more than 75%. The order of basic cations (mean) found in rain water was as follows: Mg²⁺ ≥ K⁺ > Na⁺ ≥ Ca²⁺. The mean total basic cations (Na⁺ + K⁺ + Ca²⁺ + Mg²⁺) (4.03 meq L⁻¹). Thus, neutralization of acidic anions (viz., Cl⁻, SO₄⁼ etc.) by basic anions (CO₃⁼ and HCO₃⁼) was noticed higher in water samples. The alkaline properties of the particulate matter in rain water were responsible for neutralizing the acidic ions and consequently, for the observed increase in 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 meq L⁻¹ of Na⁺, 0.2-2.3 meq L⁻¹ of K⁺, 0.00-1.44 meq L⁻¹ of Ca²⁺ and 0.02-2.3 meq L⁻¹ of Mg²⁺. Thus, higher range of Na⁺ and K⁺ in rain water were observed in samples. The principal cations present in rain water are Ca²⁺, Mg²⁺, Na⁺ and K⁺. 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:

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

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 $< 2.0 \text{ d Sm}^{-1}$ in all the samples of the rain water. Thus, this rain water is suitable for safe irrigation, without any sodicity problems.

3.4 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^{-1} . 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^{-1}) of fluoride in rain water was represented in Fig. 1.

Considerably higher concentrations of fluoride around 0.3 mg L^{-1} in rain water have been also reported from two sites in Uttar Pradesh [16] and Madhya Pradesh [17]. Das et al. [18] reported 0.1 mg L^{-1} 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. Chandrawanshi and Patel [19] 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^{-1} , the latter being from a site close to an industrial Aluminum plant. Thus, the higher amount of fluoride in Renukut is obviously influenced by Al factory (Hindustan Aluminum Company, LTD).

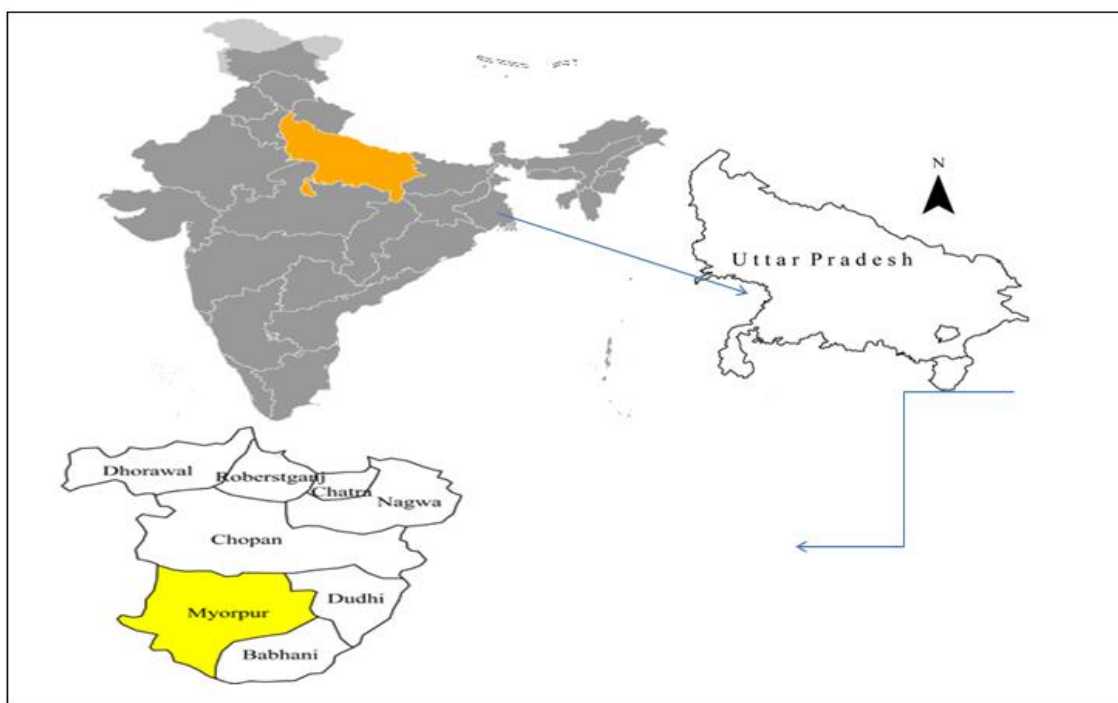


Fig. 1. Location map of the study area

Table 1. Rainwater chemistry of Renukut

S. No.	Date of sampling	pH	EC dSm ⁻¹	Na ⁺	K ⁺	Ca ⁺⁺	CO ₃ ⁻	HCO ₃ ⁻	CL ⁻¹	SO ₄ ⁻	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
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	-0.01 --1.61	0.0-1.8
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	pH	EC	CO ₃ ²⁻	HCO ₃ ⁻	RSC	Cl ⁻	SO ₄ ²⁻	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	SAR	Fluoride
pH	1.000												
EC	0.637**	1.000											
CO ₃ ²⁻	0.315	0.382*	1.000										
HCO ₃ ⁻	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 ₄ ²⁻	0.267	0.499**	-0.085	0.224	0.216	0.109	1.000						
Ca ²⁺	-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 ⁺	-0.244	-0.462*	-0.224	-0.065	0.170	-0.209	-0.218	1.000	-0.206	1.000			
K ⁺	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

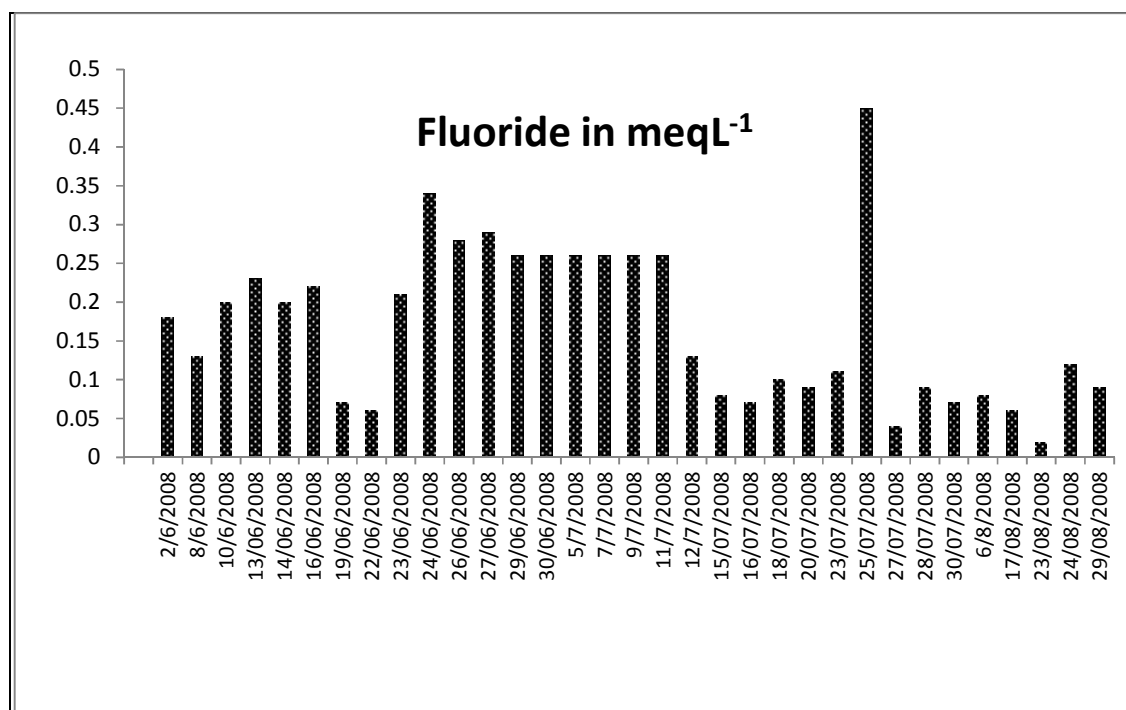
* Correlation is significant at the 0.05 level (2 tailed)

** Correlation is significant at the 0.01 level (2 tailed)

Table 3. Correlation study of fluoride with weather parameters

Parameters	Rainfall	Temperature		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

* Correlation is significant at the 0.05 level (2 tailed) ** Correlation is significant at the 0.01 level (2 tailed)

**Fig. 2. Fluoride contamination in rainwater of renukoot**

3.5 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 Renukoot have been presented in Table 2 and Table 3 respectively. Fluoride content in rain waters were negatively correlated with Ca^{2+} and Mg^{2+} . An inverse relationship was also found between F^- and Ca^{2+} in some investigations [20]. 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 Renukoot. Thus, due to increase in atmospheric temperature, rate of evaporation increases which leads to reduction of fluoride concentration in rain water.

4. 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 rainwater. The 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⁻¹. 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 treated by applying appropriate technology to remove the impurities.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Friend JP. Natural chlorine and fluorine in the atmosphere, water and precipitation. United Nations Environmental Programme/ World Meteorological Association. Scientific Assessment of Stratospheric Ozone: 1989. Alternative Fluorocarbon Environmental Acceptability Study Report; 1989.
2. TRI01. TRI explorer. Providing access to EPA's toxics release inventory data. Washington, DC. Office of Information Analysis and Access. Offices of Environmental Information. U.S. Environmental; 2003.
3. EPA. Background Report: AP-42 Section 12.1. Primary aluminum; 1998b. Available: <http://www.epa.gov/ttnchie1/ap42/ch12/bgdocs/b12s01.pdf>
4. Pickering WF. The mobility of soluble fluoride in soils. Environmental Pollution 1985;(Series B)9:281e308.
5. Skjelkvale BL. Factors influencing fluoride in Norwegian lakes. Water Air and Soil Pollution. 1994;77:151e167.
6. Zheng B, Yu X, Zhand J, Zhou D. Environmental geochemistry of coal and arsenic in Southwest Guizhou, P.R. China. 30th International Geological Congress Abstracts. 1996;3:410.
7. Finkelman RB, Orem William, Castranova Vincent, Tatu Calin A, Belkin Harvey E, Zheng Baoshan, Lerch Harry E, Maharaj Susan V, Bates Anne L. Health impacts of coal and coal use: Possible solutions. International Journal of Coal Geology. 2002;50:425e443.
8. Wang XC, Kawahara K, Guo XJ. Fluoride contamination of groundwater and its impacts on human health in Inner Mongolia area. Journal of Water Services Research and Technology e Aqua. 1999;48:146-153.
9. Smedley PL, Nicolli HB, Macdonald DMJ, Barros AJ, Tullio JO. Hydrogeochemistry of arsenic and other inorganic constituents in ground waters from La Pampa, Argentina. Applied Geochemistry. 2002;17: 259-284.
10. Carpenter R. Factors controlling the marine geochemistry of fluorine. Geochim Cosmochim Acta. 1969;33:1153-1167.
11. Hemens J, Warwick RJ. Effects of fluoride on estuarine organisms. Water Res. 1972; 6:1301-1308.
12. Fung KF, Zhang ZQ, Wong JWC, et al. Fluoride contents in tea and soil from tea plantations and the release of fluoride into tea liquor during infusion. Environ Pollut. 1999;104:197-205.
13. Shacklette HT, Boerngen JG, Keith JR. Selenium, fluorine, and arsenic in surficial materials of the conterminous United States. Washington, DC: U.S. Department of the Interior. Geological Survey Circular. 1974;692.
14. APHA. Standard methods for the examination of water and wastewater. American Public Health Association, Washington, DC; 1995.
15. Murhekar GH. Determination of physico-chemical parameters of surface water samples in and around Akot City. International Journal of Research in Chemistry and Environment. 2011;1(2): 183-187.
16. Satsangi GR, Lakhani A, Khare P, Singh SP, Kumari SS, Srivastava SS. Composition of rain water at a semi-arid rural site in India. Atmos. Environ. 1998;32: 3783-3793.
17. Singh SP, Khare P, Satsangi GS, Lakhani A, Kumari SS, Srivastava SS. Rainwater composition at a regional representative

- site of a semi-arid region of India. Water Air Soil Poll. 2001;127:93–108.
18. Das DK, Burman GK, Kidwai AL. Chemical composition of monsoon rainwater over Bhopal, Madhya Pradesh during 1977 and 1978. Mausam. 1981;32:221–228.
19. Chandrawanshi CK, Patel KS. Fluoride deposition in Central India. Environ. Monitor. Assess. 1999;55:251–265.
20. Handa BK. Geochemistry and genesis of fluoride containing ground waters in India. Groundwater. 1975;13:275-281.

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