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Fluoride Contamination Status of Groundwater in East Coastal Area In Tamilnadu, India

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Abstract: The objective of the study is to investigate the fluoride pollution of groundwater in the South east coastal regions of Tamil Nadu, India. Totally 36 ground water samples have been collected from open dug well, hand pump and bore wells in the east coastal region from Rameshwaram to Thiruvanmiyur during the year 2011. Samples were analysed to find out the fluoride ion along with physical and Chemical Parameters. The fluoride content of ground water ranges from 0.02 to 1.54ppm suggests domestically not suitable. All the samples observed low amount of fluoride (< 0.5mg/l) in the study area. Four out of 36 samples (Kottaipaattinam, Vedharaniyam, poombukar and Aaraiyankuppam) observed within the permissible WHO limit for safe drinking water (O.5-1.5 mg/l). Two samples (Adhirampattinam(1.54mg/l), Aalappakam(1.53mg/l)) exceed the limit for fluoride in drinking water set by the WHO.. Understand the hydro geochemistry of fluoride in groundwater by the mechanisms of mobilization of fluoride with correlation coefficient taken in the study area.

Keywords: Ground water, Fluoride, East Coastal, Correlation Coefficient

I. INTRODUCTION

Fluoride compounds are abundant in the earth's crust (0.06-0.09%) (Wedepohl,1974) and found in rocks, soils, salt, sea water and also present in rivers, lakes and almost all fresh ground water at varying concentrations. Fluoride in water exists in the dissociated form, i.e. the fluoride ion. The most common minerals are fluorite (CaF_2), Fluorapatite ($Ca_5F(PO_4)_3$) and Cryolite (Na_3AlF_6). Fluoride is an essential element, which is good for the teeth enamel and helps to prevent dental caries. In excessive doses, however, it will lead to a chronic fluoride poisoning (fluorosis). Fluoride contamination of groundwater is a growing problem in many parts of the world. High concentration of fluoride is reported both from hard rock (granites & gneisses) as well as alluvial aquifers[1]. In India more than 66million people are at risk of developing fluorosis and high fluoride concentration in groundwater (greater than 1 mg/l) is widespread in the arid to semi-arid western states of Rajasthan and Gujarat and in the southern states of Andhra Pradesh, Karnataka and Tamil Nadu.[2,3,4,5] People living in such areas were drinking high fluoride water without realizing its presence, which caused various bone diseases. The cause of high fluoride in ground water is geogenic being a result of the dissolution of fluoride bearing minerals. Fluoride in ground water is mainly influenced by the local and regional geological setting and hydro geological condition. However, soil consisting of clay minerals [6,7], the influence of local lithology, aided by other factors like semi-arid climate of the region may be responsible for higher concentration of fluoride in the groundwater of the region. In sea area, fluoride containing chemical components of Ca, Mg, Na, Cl, SO4, bicarbonate, Bromide, Phosphate, Iron, Aluminium etc., Locally used agricultural pesticides and anthropogenic contamination of surface water due to many rivers carry on particulate matter on rainy seasons. Therefore it is very important to understand the mechanisms of mobilization of fluoride to be able to mitigate the problem as effectively as possible. Tomas Blom and Elin Cederlund [8,9] have been used hydro geochemistry of the groundwaters of Alappuzha and Palakkad districts with an aim to understand the mechanisms of mobilization of fluoride. Physicochemical conditions like decomposition, dissociation, subsequent dissolution and agrochemicals are might be responsible for leaching of fluoride in to the drinking water sources. However, Joydev Dutta,2010, Kuldeep Krishan Sharma, 2013,[10,11] have been used to determine fluoride by correlation coefficients along with physical

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Vol. 3, Issue 3, March 2014

chemical parameters. Moreover, the Karl Pearson correlation coefficients between Fluoride and other water quality parameters Such an observation was also made by Ramamohana Rao et al. (1993), Sivasankar,v., 2011, Mamatha P.,

(Vol.3, issue.3, MARCH 2014)

2010.[12,13,14]. In recent years, there has been an increased F- concentration in ground water causes adverse impact of human health. Hence, the present study is focused on to investigate the fluoride contamination along with change of physico chemical characteristic of ground water in Southern East coastal region in Tamilnadu, India.

The objective of this study is

1. To investigate the detailed hydro geochemical characteristics of the groundwater to be able to understand the mechanisms of fluoride mobilization.
2. To determine the amount of fluoride present in ground water.

Health effects of Fluoride

The effects of fluoride on human health can be either positive or negative depending on the amount of fluoride that has been ingested. WHO recommends that drinking water should ideally contain 0.5-1.0 mg/L fluoride, as it helps to prevent dental caries. When the teeth are fully developed fluoride will still help to protect the teeth. It will dissolve in the saliva and help to repair teeth that have been attacked by dental caries. The fluoride can also attach to the surface of the teeth and then be released to help and protect the teeth when needed. Osteoporosis is an illness where bones become very fragile and breaks very easily. Fluoride can be used to treat that condition as it affects the enzyme that controls the production and degradation of bone. It will lead to a faster production than degradation and the bones will become less fragile[8]. Low concentration provides protection against dental caries, especially in children. This protective effect increases with concentration up to 2mg/l of drinking water. The minimum concentration of fluoride in drinking water required to produce the effect is approximately 0.6mg/l. High fluoride concentrations exert a negative effect on the course of metabolic processes and consequently individuals may suffer from dental fluorosis, skeletal fluorosis (This can cause joint pain, restriction of mobility, and possibly increase the risk of some bone fractures) and non-skeletal manifestations (see table 1). Excessive intake of fluoride affects the teeth and the bones. This is because fluorine is very electronegative and thus easily binds to the positively charged calcium ions in teeth and bone. In large quantities fluoride can also affect the kidneys and the thyroid gland and in the most extreme cases it can lead to death.

Table:1 Fluoride content in drinking water and various effects on human health

Fluoride content in mg/L	Corresponding effects on human health
< 0.5	Dental caries
0.5-1.0	Safe limit
1.0-3.0	Dental fluorosis
3.0-4.0	Stiff and brittle joints/bones
> 4.0	Deformities in knees; crippling fluorosis;

II. MATERIALS AND METHODS

Study area

Groundwater samples from different hand pumps, dug wells and bore well of thirty six sampling locations (Fig 1) from the South Eastern coastal region *viz.*, Ramanathapuram, Thiruvarur, Thanjavur, Puthukottai, Nagapattinam, Perambalur, Cuddalore, Viluppuram, Kancheepuram and Chennai in Tamilnadu (Fig.1) were analyzed during post monsoon and pre monsoon seasons-2011. Geographically it is located 9^0 17' 3.51" N-12^0 59' 0.45'' N latitude and 79^0 13' 32'' E-80^0 15' 32.63'' E longitude. In the Tamilnadu coastal region has two monsoon periods a south-western monsoon which is active, a south-western monsoon which is active from mid-June to mid-September and a north eastern monsoon which is active from mid-September to mid-January. More than 60% of rainfall received during the north-eastern monsoon (October-december). Annual rainfall ranges from 1000 to 1500mm. the annual mean

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Vol. 3, Issue 3, March 2014

temperature of the study area is 28^0 c. The soils of the coastal area are entirely alluvial, but vary in quality. Most of the soils are alluvial [15]. Annual rainfall was recorded in 1477mm in the study area at the year - 2011.

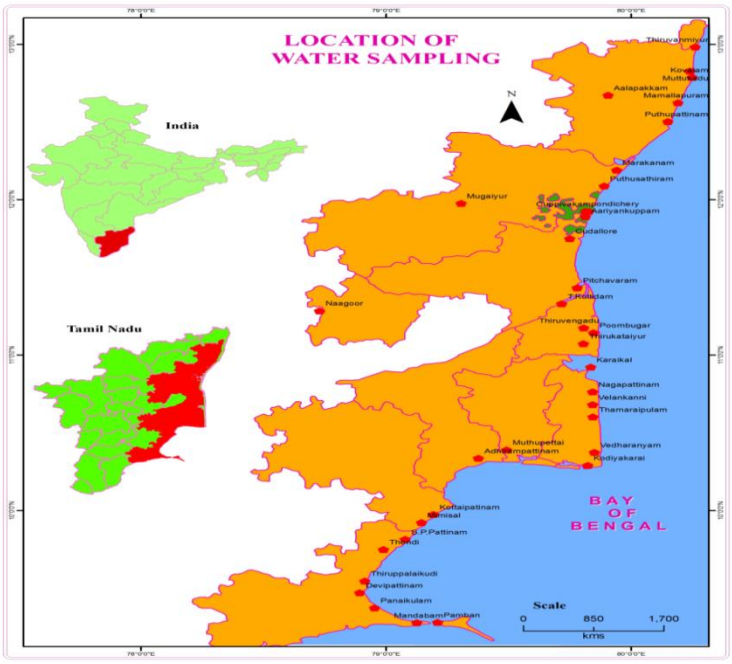

Fig.1 Location map in the study area

Materials and Methodology

The samples were collected in 1000 ml plastic bottles with necessary precaution. They were then carefully sealed, labeled. High pure (Anal R grade) chemicals and double distilled water was used for preparing solutions for analysis of fluorides along with physico-chemical parameters such as pH, TDS, TH, CO3, HCO3, Cl, Na, K, Ca, Mg and SO4 during the year 2011 in the month of may-11. The groundwater samples were subjected to physicochemical analysis using standard procedure by APHA [16] except pH and electrical conductivity were measured at time of collection of samples by using Elico pH meter and conductivity meter.

III. RESULT AND DISCUSSION

The Fluoride content in the groundwater samples in various locations of the study area has been shown in Table 3. From the results, all the samples have below the value of 0.5mg/l obtained except two samples have high concentration of fluoride value observed (Adhirampattinam(1.54mg/l), Aalappakam(1.53mg/l)), Sample location of Kottaipaattinam, Vedharaniyam, Poombukar and Aaraiyankuppam were obtained with in permissible the limits of 0.5mg/l to 1.0mg/l as recommended by BIS value (see table 2). Water temperature of ground water samples ranged from $27\text{-}28.9^0$c. The pH value in most of the water samples ranges from 7.8-9 which indicate alkaline in nature of the samples. In the study area, the sodium (Na+) which is dominant cation and Cl^- is dominant anion due to may be sea water intrusion in this coastal region. A high percentage of TDS is contributed by Na, Cl, Ca, Mg, CO_3, HCO_3 and SO_4 as it is clear from table (3). As the chemical composition of natural water is controlled by many interrelated processes, it follows that some understanding of these processes is needed. Pearson's correlation coefficient is used to measure and establish the strength of a linear relationship between two variables and the results are presented in the table (4).

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(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 3, March 2014

Table 2 The permissible limits of fluoride in drinking water by various organizations

S.no	Name of the organization	Permissible limit of fluoride (mg/l)
1	World Health Organization	1.5
2	Bureau of Indian Standards (BIS)	1.0
3	Indian Council of Medical Research (ICMR), Govt. of India	1.0

Table :3 Test results in the study area.

Parameter	Minimum	Maximum	Mean
pH	7.8	9	8.3
TDS	330	11646	1863
TH	97	6725	573
Ca	24	1046	117
Mg	8	1030	69
Na	46	1762	407
K	5	154	45
Cl	52	6779	728
SO4	7	636	97
CO3	0	182	57
HCO3	38	876	298
F	0.02	1.54	0.11

For thermodynamics equilibrium in ground waters which are in contact with calcite and fluorite solid phase, a combined mass law equation relating both the solute species may be used

$CaCO_3 + H^+ + 2F^- \rightarrow CaF_{2(s)} + HCO_3^-$ $\rightarrow 1$

$K_{cal\text{-}flu} = a_{Hco3} / a_{H+} \times (a_{F^-})^2$ $\rightarrow 2$

Where $K_{cal\text{-}flu}$ is a constant and in reasonably constant water, any increase or decrease in bicarbonate concentration is accompanied by corresponding changes of increase or decrease in the concentration/activity of fluoride ion concentration. This also appears to be a more appropriate mechanism to account for the observed inverse relationship between Ca and F, than the one suggested by Gaciri and Davies (1993)[17] on the basis of solubility of calcite and fluorite in water. Sodium concentration increases with fluoride, thereby increasing the solubility of fluorite in water [18]. Hydro geochemical relationships can be illustrated in a number of different ways. The correlation-coefficient helpful along with neither spatial variation nor plots is used to illustrate the hydro geochemical relationships in this study.

Table 4 correlation coefficient between different hydrogeochemical parameter in the study area

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(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 3, March 2014

correlation	pH	TDS	TH	Ca	Mg	Na	k	Cl	SO4	CO3	HCO3	F
PH	1											
TDS	-0.036	1										
TH	-0.162	.919**	1									
Ca	-0.189	.907**	.992**	1								
Mg	-0.143	.920**	.997**	.983**	1							
Na	0.059	.936**	.737**	.720**	.745**	1						
K	.401*	0.111	0.014	0.045	0.002	0.123	1					
Cl	-0.125	.978**	.953**	.940**	.958**	.888**	0.01	1				
SO4	-0.08	.883**	.794**	.780**	.784**	.826**	0.091	.825**	1			
CO3	.507**	-0.066	-0.212	-0.245	-0.195	0.07	.375*	-0.185	-0.043	1		
HCO3	0.15	0.2	0.064	0.131	0.031	0.23	.457**	0.083	0.146	0.021	1	
F	-0.13	0.106	-0.089	-0.102	-0.077	0.25	-0.31	0.078	0.129	0.014	-0.164	1

A negative correlation is seen between fluoride and bicarbonate (fig.6 and Table 4).When plotted, all samples show a negative correlation. According to equation (1) bicarbonate can react with fluoride to form calcite leaving fluoride ion in the ground water. The negative correlation coefficient means that if the concentration of bicarbonate decreases the fluoride ion concentration will decreases as well. Thus the low values of fluorides. The correlation between sodium and fluoride is a weak (a value of +0.25). This is also confirmed by the plot (fig.3) the small correlation that exists can be explained by evapotranspiration, when the water evapotranspirates the ions are left in

Solution. This increases the concentration of all the ions including sodium and fluoride. Thus, a weak positive correlation between fluoride and sodium can be observed. High fluoride and very low calcium in water may be due to the prior precipitation of CaCO3 from water with limited incorporation of fluoride in CaCO3 structure. Alternatively Ca might have replaced by Na in cation exchange reactions with increasing concentration of sodium the solubility of fluorite in water also increases [19]. This also confirmed by the relation from the plot Calcium, Magnesium, Chloride, pH against fluoride is a negative correlation indicates that there exists and low level of fluoride in the rock that can dissolve in the ground water (fig. 2,4,5,7 and Table.4). Moreover, the occurrence of low levels of fluoride in ground water from these areas may be due to either to the absence of fluoride bearing magmatic solutions or of fluoride containing minerals in the strata through which ground water is circulating. It could also be due to too rapid fresh water exchange with the result that the normal process of concentration through evaporation or evapotranspiration is not very effective in raising the fluoride content of the ground waters to high values prevalent in the study area [20].

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Vol. 3, Issue 3, March 2014

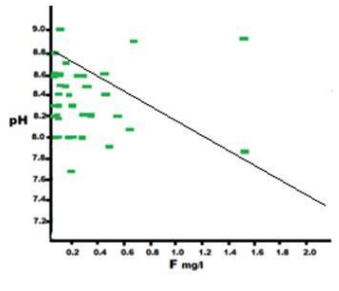

Fig.2 pH against fluoride

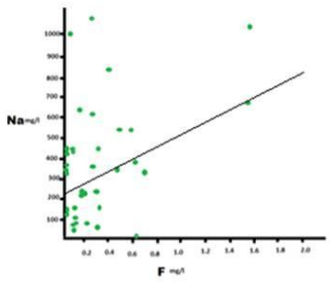

Fig.3 Sodium against fluoride

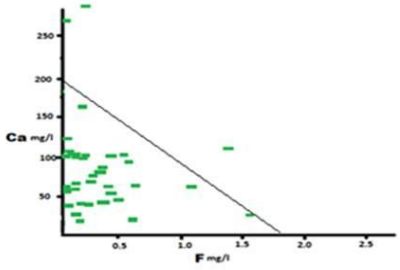

Fig.4 Calcium against fluoride

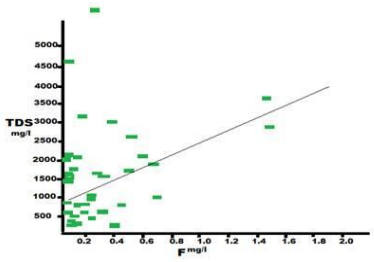

Fig.5 TDS against fluoride

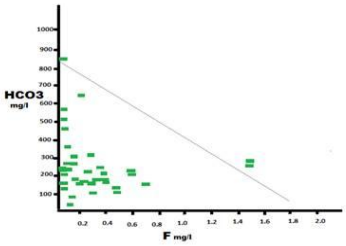

Fig.6 Bicarbonate against fluoride

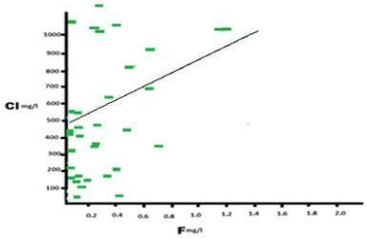

Fig.7 Cl against fluoride

IV. CONCLUSION

The present study investigates the hydro geochemistry of fluoride in groundwater from south east coastal regions in Tamilnadu, India with an aim to understand the mechanisms of mobilization fluoride. The groundwater characteristics by the following as ; The fluoride concentrations Within the WHO limit for safe ground water (1.5 mg/L) in four samples (Kottaipaattinam, Vedharaniyam, poombukar and Aaraiyankuppam). Two out of 36 samples (Adhirampattinam(1.54mg/l), Aalappakam(1.53mg/l)) exceed the limit for fluoride in drinking water set by the WHO and all the samples (30/36) observed low amount of fluoride in the study area. The fluoride originates from the bedrock, the high levels of sodium help to dissolve calcite and dolomite to precipitate and high level of calcium and magnesium decrease level of fluoride on water. Therefore the treatment system has to be locally available and easy to maintain and operate. Water fluoridation is the process of adding fluoride to a water source so that the level of fluoride in the water reaches the recommended level of fluoride for good dental health. Water fluoridation is the addition of fluoride to water supplies that are low in fluoride in order to prevent dental decay. The level shown scientifically to be best for each area depends on the climate.

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