Journal of Urban and Environmental Engineering, v.9, n.2, p.119-126

ISSN 1982-3932
doi: 10.4090/juee.2015.v9n2.119126

Journal of Urban and Environmental Engineering

www.journal-uee.org

STATISTICAL APPRAISAL OF FLUORIDE ENRICHMENT IN AREAS OF MALDA AND SOUTH DINAJPUR DISTRICT, WEST BENGAL, INDIA

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Received 10 February 2014; received in revised form 17 November 2015; accepted 19 November 2015

Abstract: The present study was conducted in some randomly selected area of Malda and South Dinajpur district of West Bengal to find out the level of fluoride in groundwater and its association with other inorganic constituents through statistical modeling and chemical indices. Results revealed that the considerable variation of different parameters, among them 7.90% sample showed F^- levels beyond the WHO recommended value. Moreover, F^- shows positive correlation with Na^+ ($p < 0.467^*$) and negative correlation with Ca^{2+} ($p < -0.289$) which is again well supported by factor analysis study. From the results of Multiple linear regression model and piper diagram revealed the significant relationship of F^- with Na^+, alkaline pH and Ca^{2+} and dominancy of F^- due to high Na-HCO_3 enrichment respectively. The higher level of F^- in groundwater is the consequence of fluoride containing sedimentary rock weathering.

Keywords: Fluoride; Groundwater; Contamination; Statistical modeling; Weathering.

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INTRODUCTION

Ground water quality is prime concern now a day because it's a soul source for drinking water. About 80% of human disease and 30% of infant mortality in developing countries originating from ground water pollution (Chakraborty, 1999). In the era of global development ground water being contaminated due to industrialization, urbanization and from various other sources. Although ground water contributes only 0.6% of total water resources on earth, from which 80% accounts rural domestic water needs and 50% contribute urban water needs in developing countries like India (Karthikeyan *et al.,* 2010; Meenakshi & Maheswari, 2006).

Ground water contamination by fluoride was emerging as one of the severe regional problem in India (Karthikeyan *et al.,* 2010). The occurrence of fluoride in ground water depends on geological formation, rock type, solubility of fluorine-bearing minerals, pH, carbonate and bicarbonate alkalinity, temperature, depth of the aquifer and ion exchange capacity of aquifer materials of that particular area. Excluding these environmental factor some anthropogenic activities such as use of phosphatic fertilizers, pesticides, sewage and sludge for agriculture also implicate incidence of high fluoride in groundwater. Area where human interference is negligible, fluoride bearing mineral such as fluorite, fluoro appetite and mica is reported as a main source of fluoride in ground water (Kafri *et al.,* 1989; Lahermo *et al.,* 1991; Apambire *et al.,* 1997; Rafique *et al.,* 2008). Therefore the area with such dominating types of rock can be suspected as the cause of occurrence of high fluoride (Gupta *et al.,* 2012). The possible mechanism of dissolution of fluoride in water are due to water mineral interaction and water retention time in clay by leaching of fluoride from hydroxyl position of aluminium containing silicate minerals such as mica, chlorite and amphiboles (Kundu *et al.,* 2011).

Presence of fluoride in ground water in terms of low or high has long being linked to the incidence of dental and skeletal carries (Mondal *et al.,* 2012; Dissanayaka, 1991). Assimilation of F^- in human body through water is portable up to the level of 1.0 mg/L for development of bones and dental carries (WHO, 1996). Exposure F^- at higher level (> 1.5 mg/L) are resulting in fluorosis characterized by initial opaque white patches followed by molting and pitting of teeth's. Skeletal fluorosis may occur when exposed concentration of fluoride in water was exceed 4.0 mg/L (Mondal *et al.,* 2012; Kundu *et al.,* 2011; Dissanayaka, 1991).

About 65 million people including 6 million children of India, particularly from West Bengal have symptoms of fluoride poisoning from ground water. Survey results highlighted that 60 blocks of eight districts of West Bengal viz., Bankura, Bburddhaman, Birbhum, Purulia, Midnapur, Malda and West Dinajpur are fluoride affected (Chatterjee *et al.,* 2008). In view of this attribute it is of great fundamental interest for research community to introduce exhaustive research on these reported areas. Except Birbhum, still now no exhaustive study on occurrence and behavior of fluoride in ground water was previously introduced in any research report. Therefore, it is immense important to study the ground water survey and the present communication was dedicated to find out the ground water quality of Malda and South Dinajpur district, with special emphasis on fluoride occurrence and level of significant of enrichment with the help of multivariate statistics and chemical indices. In the light of above interest a comprehensive study was done in the target area of Malda and South Dinajpur district.

MATERIALS AND METHODS

Study area

Malda and South Dinajpur is a juncture between north and south Bengal. The study is mainly conducted in north to north-east part of Malda district and middle to south, south-east and south-west part of the South Dinajpur district. The study area is confined within $88^{o}44'$ to $88^{o}35'$ E and $25^{o}35'$ to $24^{o}40'$ N (**Fig. 1**) with an average elevation of 41 meter from average sea surface level. The climate of that area enjoy the pleasant temperature, varies between less than $10^{o}C$ to $35^{o}C$ in winter and summer along with average annual rainfall above 1500 mm.

The topography of this area is mainly of low-low lying plains sloping towards the south in respect with undulating areas in north-east. The main land is characterized by rolling topography, oxidized clayey sediments (yellowish brown soil) enrich with moderate to high iron concentration. Mature yellowish brown soils are belonging from oldest quaternary formations. The alluvial plain which is dominated by sediments, contain mainly sand, silt and clay. The area is devoid of hard rock which contains fluorite and fluoro-apetite found in granitic crystalline complexes is long being linked with fluoride incidence in this area.

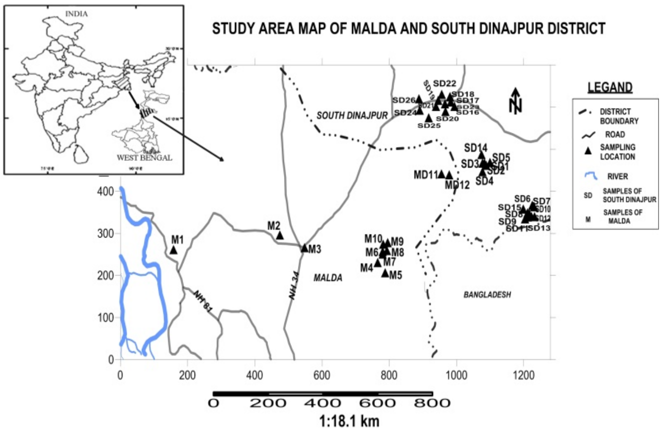

Fig. 1 Study area along with sampling location of Malda and South Dinajpur districts.

Sampling and analysis

Samples were picked up randomly chosen 38 sites of 16 villages (seven villages of Malda and nine villages of South Dinajpur districts) from Malda and South Dinajpur district of study area (**Fig. 1**). All the samples were collected in 300 ml sterilized polypropylene (Turson made) bottles and stored in freezing condition (temperature < 10^{o}C), by following standard protocols. Geographical positions of all the sampling sites are recorded during the sampling by using GPS (Model: GARMIN GPS 12). Study area map with sampling location points and post map with fluoride distribution was constructed by using SURFER 9.

As per standard method pH and electrical conductivity (EC) was measured in in-situ condition by PCSTestr 35 (EUTECH). Rest of the parameters were measured in the laboratory of the Department of Environmental Science, University of Burdwan after preservation and transportation. Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, HCO_3^-, CO_3^{2-}, Fe^{2+}, F^- and TDS are measured by following the standard method of APHA, (2005) 21th edition.

Fluoride content in ground water was measured by using Ion selective electrode (model: Thermo Scientific Orion 4-Star). During the measurement the ionic strength of fluoride was adjusted by buffer (TISAB II), and concentration of fluoride was estimated in correspondence with used standard known fluoride solutions. The accuracy of the chemical analysis was verified by calculating charge balance errors (viz., cationic and anionic charge balance) and it was found that the errors were below ±5%, an added proof of precision of analytical data.

Data management

Experimental Data were analyzed statistically through Xl stat 2012 for analysis of correlation matrix; principal component analysis and multiple regression analysis.

RESULTS AND DISCUSSION

Statistical summary of analyzed water parameters with WHO guideline value were represented in **Table 1**. The variations in parameters in terms of their concentration are quite noticeable (**Table 1**). Electrical conductivity ranges from 260 µS/cm to 2700 µS/cm, from which 92% of samples are below the WHO guideline value and only 8% samples are exceeding the range of permissible limit. Among the cationic species, Na^+ dominates the ground water chemistry, although Na^+ showed the concentration less than the WHO guideline value. TH of all the observed samples are just below the permissible limit (WHO, 2004), but only 2.6% of this (TH) parameters are exceeding WHO recommended value. Whereas 94.74% of overall water samples exceeded the permissible limit of Fe^{2+}. But interestingly 7.90% of water are exceeding the permissible limit of F^- (>1.5 mg/L). The higher level of fluoride (1.5 mg/L) was recorded at South Dinajpur district (**Fig. 2**).

Table 1. Statistical summary of observed results along with WHO guideline value for drinking water

Paramter	Mean	Min.	Max.	±SD	WHO Guideline value 2004
pH	7.55	7.02	8.8	0.34	6.5−8.5
EC	995.79	260	2700	495.76	1000
TA	244.32	60	464	97.38	--
CO_3^{2-}	68.00	8	320	65.20	--
HCO_3^-	173.63	12	400	83.43	--
TH	193.63	42	490	118.47	500
Ca^{2+}	17.93	3.52	48.96	10.73	--
Mg^{2+}	36.16	3.79	112.46	24.10	--
Cl^-	60.08	1.6	419.9	77.51	250
Na^+	41.34	16	69	14.41	200
K^+	8.95	1	69	15.49	--
F^-	0.87	0.4	1.7	0.30	1.5
Fe^{2+}	2.53	0.07	21.72	3.49	0.3
SO_4^{2-}	27.59	4.57	57.17	9.34	250
TDS	363.14	96.5	862	156.91	1000

(EC=Electrical conductivity, TA=Total alkalinity, TH=Total hardness, TDS=Total Dissolve Solid, All units are mg/l except pH and EC µS/cm).

The inorganic constituents like TA, CO_3^{2-}, HCO_3^-, Mg^{2+} and Cl^- showed positive relationship ($P<0.05$) with EC. Whereas, T.H and Ca^{2+} shows significant positive relationship with EC ($P<0.01$). The high level of electrical conductivity is might be due to the presence of higher level of dissolve salts. On the other hand pH of the studied water samples showed linear relationship with F^-. This is quite possible, because under alkaline condition fluoride can be leached from minerals in to ground water (Gupta *et al.*, 1986; Mondal *et al.*, 2012).

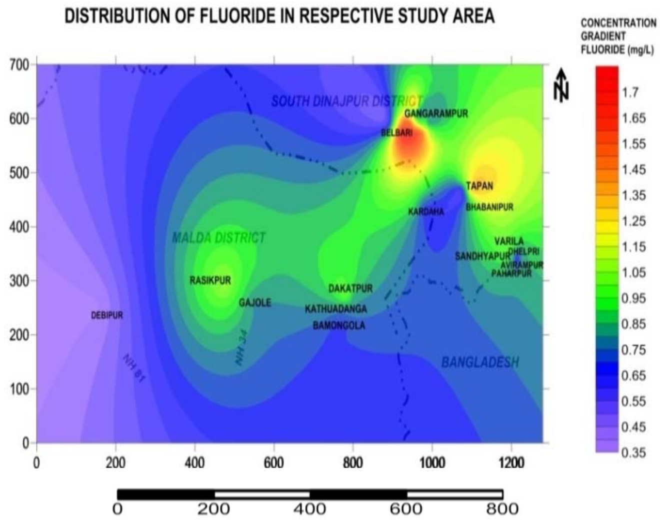

Fig. 2 Distribution of fluoride concentration on the study area.

Table 2. Correlation matrix of observed variables

	pH	EC	TA	CO_3^-	HCO_3^-	TH	Ca^{2+}	Mg^{2+}	Cl^-	Na^+	K^+	F^-	Fe^{2+}	SO_4^{2-}
EC	-0.019													
TA	-0.034	0.585*												
CO3	-0.012	0.388	0.518											
HCO3	-0.009	0.446*	0.735	-0.12										
TH	-0.122	0.750**	0.611	0.296	0.512									
Ca	-0.154	0.830**	0.635	0.308	0.55	0.774								
Mg	-0.104	0.672*	0.559	0.271	0.462	0.985	0.654							
Cl	-0.076	0.446*	0.055	0.132	0.024	0.257	0.345	0.213						
Na	-0.071	0.302	0.259	0.237	0.050	0.183	-0.046	0.231	0.178					
K	0.322	0.222	0.097	0.147	0.160	-0.046	0.002	-0.056	0.088	-0.079				
F	0.126	-0.055	-0.067	-0.101	0.000	0.016	-0.289	0.097	-0.043	0.467*	-0.22			
Fe	-0.364	-0.095	-0.266	-0.123	-0.199	-0.126	-0.076	-0.129	0.064	-0.073	-0.084	-0.185		
SO4	0.100	0.278	-0.158	-0.044	-0.019	0.014	0.100	-0.010	0.126	0.112	0.465	-0.167	0.025	
TDS	-0.021	0.974	0.649	0.396	0.523	0.772	0.848	0.693	0.429	0.296	0.248	-0.096	-0.104	0.278

(*P<0.05, **P<0.01)

Under such alkaline medium bicarbonate plays a crucial role (**Eq. 1**) to aggravate the F^- level in water (Mondal *et al.,* 2012).

$$K_{cal-fluor} = \frac{a_{HCO_3^-}}{a_{H^+}(a_{F^-})^-} \tag{1}$$

According to Kim *et al.* (2011) & Handa (1975), both fluorite and calcite are in contact with water (**Eq. 1**). Therefore equilibrium constant $K_{cal\text{-}fluor}$ explains that any increase or decrease in bicarbonate concentration is accompanied by corresponding increase or deficiency in fluoride concentration. This equation (**Eq. 1**) accounts for best fitted to depict the inverse relationship between Ca^{2+} and F^-. Again HCO_3^- shows a linear in turn equilibrium relationship with F^- (**Fig. 4**). Present study revealed that that at relative alkaline pH (7.3 to 7.7) the concentration of F^- and HCO_3^- are high which support the equilibrium constant (**Eq. 1**) (**Figs 4**, **5** and **7**).

Figures 6–10 depicts a negative correlation in contrast of fluoride with Ca^{2+} and positive relationship ($P < 0.05$) with Na^+. Therefore F^- shows some greater affinity to Na^+, means F^- increase with Na^+ and consequently Ca^{2+} is precipitated as carbonate (Gaciri & Davis 1993). Probably ion exchange is principal mechanisms behind the dissolution of F^- in association with Na^+ during the time of water movement through weathering zone (Rao & Devadas, 2003). High concentration of Na^+ may also increase the solubility of F^- in water (Yidana *et al.,* 2012) resulting in common ion effect which in turn suppress the dissolution of Ca^{2+} when the concentration of F^- is above the limit of Ca^{2+} solubility.

Correlation study also supported that the negative relationship of F^- and pH with Fe^{2+} which in turn demonstrates the availability of Fe^{2+} in the medium (**Table 2**). This is probably because at relatively high pH fluoride is leached from mineral surface and Fe^{2+} can converted to $Fe(OH)_3$ and subsequently precipitated out and demonstrate the low availability of Fe^{2+} in aqueous medium.

Multivariate Statistical Analysis

Factor analysis was performed to determine the magnitude of variables of all 38 observations, at the same time simplify the data structure by extracting the principal components by varimax rotation, from original

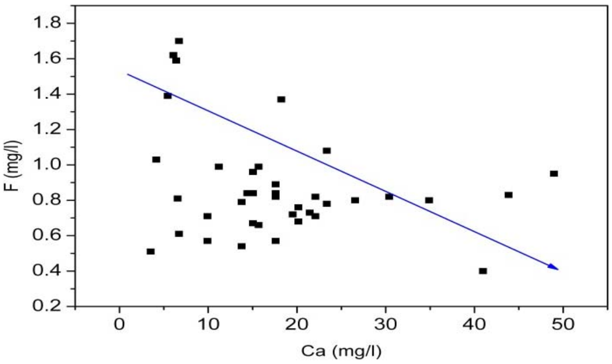

Fig. 3 Scatter plot F Vs Ca.

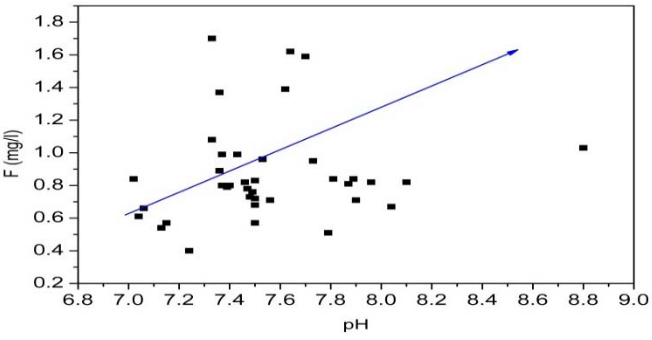

Fig. 4 Scatter plot F Vs pH.

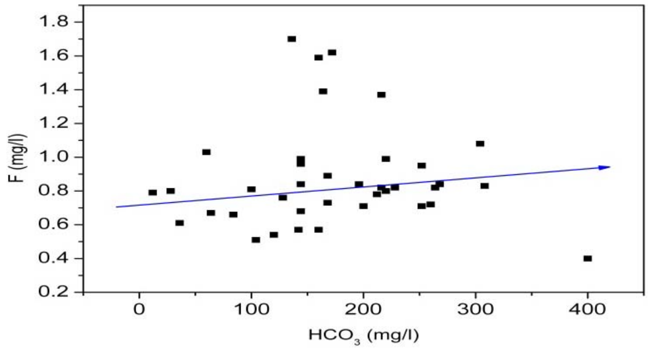

Fig. 5 Scatter plot F Vs HCO_3.

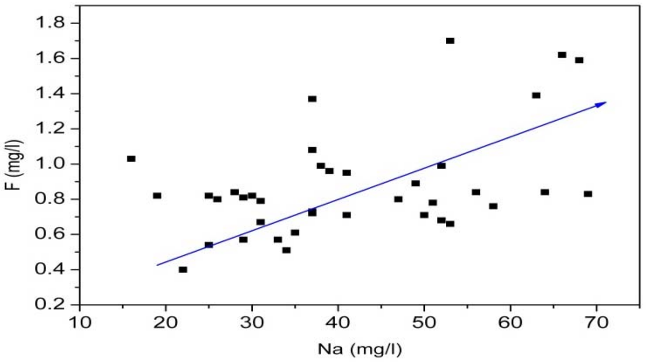

Fig. 6 Scatter plot F Vs Na.

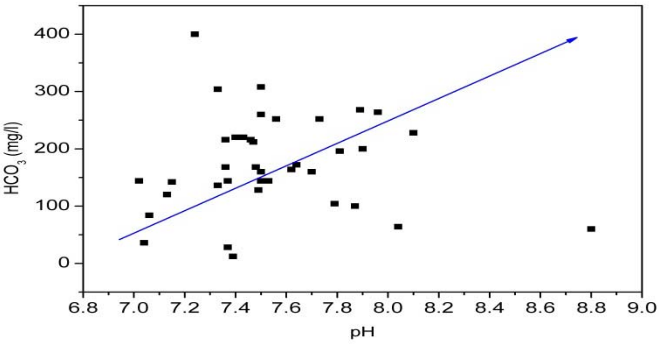

Fig. 7 Scatter plot HCO_3 Vs pH.

set of data. Kaiser-Meyer-Olkin (KMO) and Bartletts sphericity test results 0.524 and 967.730 respectively, indicating the significant ($P < 0.000$) reduction of dimensionalities of all observed data. After varimax rotation five components were extracted with total variance of 77.90%.

The component 1 (C1) constitute highly positive significant variables with EC, T.A, HCO_3^-, T.H, Ca^{2+}, Mg^{2+} and TDS with total variance of 34.44 % and Component 2 (C2) showed higher loading with SO_4^{2-} (0.842) and K^+ (0.689) which explain 12.38% of total variance. In spite of C1 and C2 component 3(C3) is significantly dominated by F^- (0.840) and Na^+ (0.816) with total variance 10.99 %. Whereas component 4 and 5 are dominated by pH (0.738), Fe^{2+} (-0.730) and CO_3^{2-} (0.942) with total variance of 10.84 % and 9.25% respectively.

Again factor loading plot (**Fig. 8**) showed a strong correlation in Group 1 with T.A, HCO_3^-, T.H and Mg^{2+}, where as in Group 2, EC shows strong positive correlation with TDS; which means EC shows proportional relationship with TDS. Significant correlation between SO_4^{2}- and K^+ (Group 3) are observed, while others are not so significantly correlated in case of C2. In turn off C3 the strength of correlation between F^- and Na^+ (Group 4) showed very much significant because there location point in loading plot are greater than +0.8 which is much nearer to +1 of the corresponding axis (**Fig. 8**).

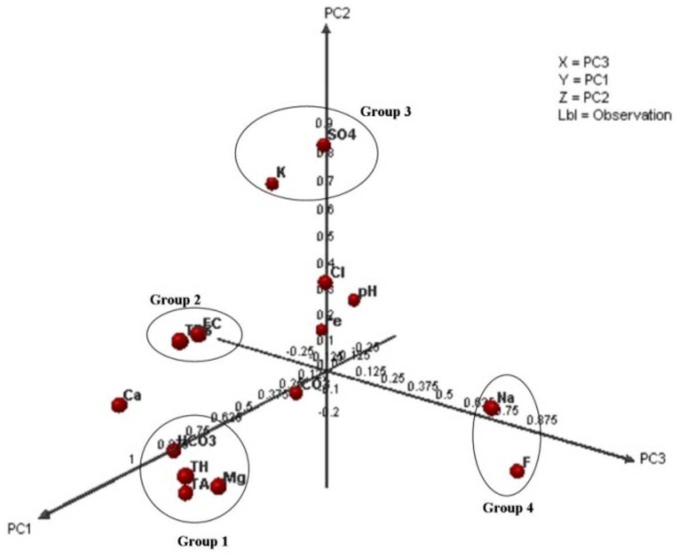

Fig. 8 Factor loading plot after varimax rotation.

Multiple linear regression

Moreover, multiple correlation results with F^- as dependent variable and other studied parameters as independent variable showed that F^- is inversely dependent on Ca^{2+} ($-19\times10^{-3}Ca^{2+}$) and proportionally dependent on pH (111×10^{-3}pH) and Na^+ ($8 \times 10^{-3}Na^+$). The output results of linear regression model is well adjusted with correlation matrix (**Table 2**).

Piper diagram

The output of Piper diagram (Piper, 1944) (**Fig. 9**) demonstrated that the overall chemical composition of studied water samples is dominated by Ca-Mg-HCO_3 (60.53%) followed by Na-CO_3 (29.95%), Ca-Mg-So_4 (7.89%), and Na-Cl (2.63%) type. Meanwhile it was observed 5.26 % water samples (SD20, SD24, SD26) which containing fluoride concentrations above 1.5 mg/L are derived from the Na-HCO_3 type of water. The occurrence of F^- in this kind of water defined by dissolution of F^- bearing mineral such as Fluorite and Fluoro-apatite found in granitic crystalline complexes due to dominancy of hard rock (Mahapatra, 2009). Several researchers documented that the concentration of F^- in ground water is accelerated by sodium concentration, alkalinity and pH values. Increasing concentration of Na^+, alkalinity and pH is dependent on retention time of water in clay minerals and enriched by water rock interaction. Enrichment of Na^+ concentration and alkaline pH is an indication of high F^- in water by replacement and release of fluoride from the hydroxyl positions of minerals (Guo *et al.,* 2007; Rafique *et al.,* 2008; Kim *et al.,* 2011).

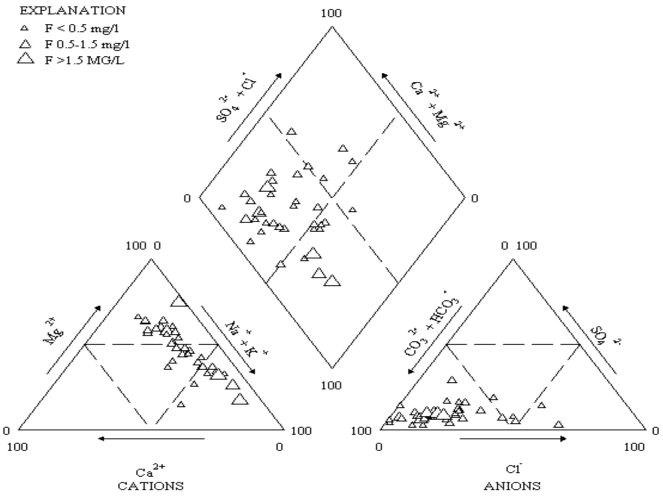

Fig. 9 Classification of groundwater on the basis of origin.

Saturation index of fluorite and calcite

Fluoride concentration in ground water is often controlled by solubility of fluorite and calcite among all other geochemical processes (Rafique *et al.,* 2008; Kundu *et al.,* 2011). The solubility limit of fluorite and calcite also provide a natural control on water composition, such that calcium, fluoride and carbonate activities are interdependent (Kundu *et al.,* 2011). The saturation index of fluorite and calcite is calculated by using following formula (Stallard & Edmond, 1987):

$$SI_F = \log(a_{Ca} \times a_F^2) - \log(K_{SPfluorite}) \quad (2)$$

$$SI_c = \log(a_{Ca} \times a_{CO3}) - \log(K_{SPcalcite}) \quad (3)$$

where a is the activity or mole concentration term and K_{sp} is the solubility product. Log K_{sp} values for fluorite and calcite at 25°C are 3.45×10^{-11} and 4.8×10^{-9} (Smyshlyaev & Edeleva, 1962). The saturation indices of fluorite (SI_F) and calcite (SI_C) are depicted in **Fig. 10**. From the SI_F value it has been observed that all the water samples are over saturated with respect to calcite. The over saturation of calcite in water is the indication of availability of calcium in water. Whereas only 5.36% (SD24 and SD26) of all samples are nearer to fluorite and calcite under saturated zone.

The under saturation in respect of fluorine in most of the water is mainly due to deficiency of fluorine source in some location or due to calcite saturation preventing it by reducing calcium activity. In contrast of **Fig. 11** high fluoride containing water samples showed low calcium content relative to other, that means when over saturated, calcium is precipitated as $CaCO_3$ and make the water under saturated to fluorite thus there is always net balance of fluoride in solution and it prevent to precipitate, consequently increases the chance of fluoride contamination in water (Kundu *et al.,* 2011). In present study negative SI values of fluorite and positive SI values for calcite are obtained from SI, resulting of it attributed to mineral richness of that particular area.

The HCO_3^-/Ca^{2+} ratio is found to be greater than 1 in all the places which make favorable condition for fluoride dissolution processes. Therefore Na^+/Ca^{2+} (89.50 % sample representing the ratio > 1) ratio explain that the low calcium activity of respective study area, which is suitable for dissolution of fluoride from water. Again Na^+/Na^++Cl^- ratio representing the value greater than 0.5 which explain that the source of ground water is dominated by sodium. Moreover $HCO_3^-/\sum$anions ratio showed greater than 0.02 which corresponds the area is dominated by rock weathering. Problem of F^- concentration in ground water was a toxicological and geoenvironmental issues emerging in 25 countries throughout the world, without excluding India. Occurrence of elevated concentration of F^- in ground water depends on weathering of fluoride containing rocks or leaching of fluoride from fluoride bearing minerals in water .Therefore water chemistry of the studied area is dominated by rock weathering which again simultaneously supported by Gibbs diagram (Gibbs, 1970) (**Figs 11–12**). On other dimension dissolution of fluoride in water was also accelerated by bicarbonate ratio through dissolution of calcium fluorite in water.

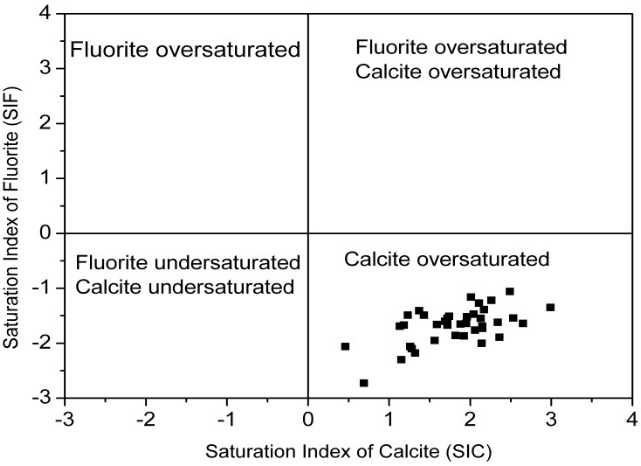

Fig. 10 Saturation index of fluorite and calcite.

In contrary of high F^- concentration (> 1.5 mg/L) in water is long being linked with several health hazards dental fluorosis, skeletal fluorosis and indigestion in metabolic system. In contrast of high F^- concentration samples from Belbari contain high F^- concentration > 1.5 mg/L, which was not acceptable for drinking as per WHO, (2004) recommendation and it's may be the main reason for prevalence of dental fluorosis in this particular area. Therefore as per WHO, (1996) recommendation, the whole experimental samples has been stratified in to four classes (**Table 3**).

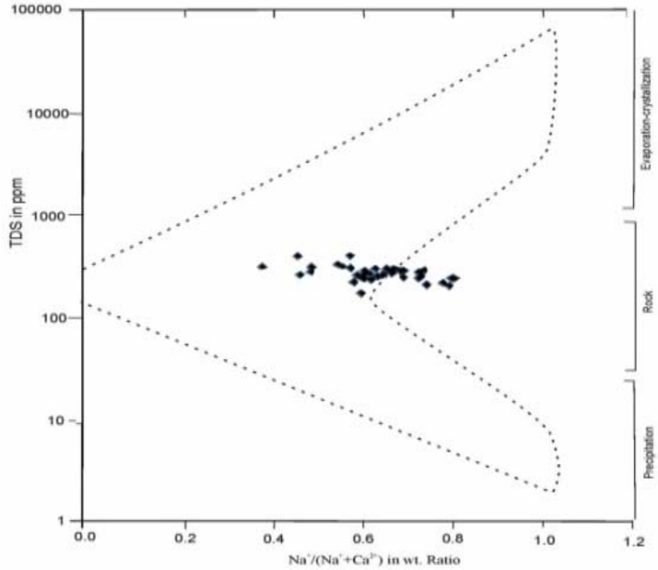

Fig. 11 Gibbs diagram, TDS Vs Na/(Na+Ca) ratio.

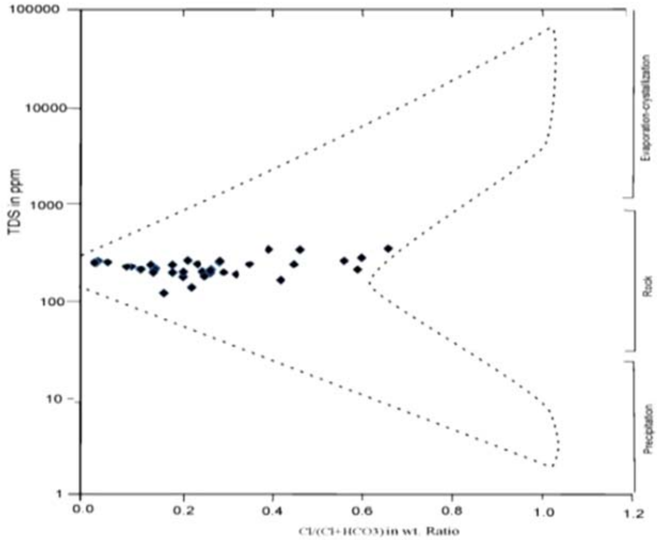

Fig. 12 Gibbs diagram, TDS Vs Cl/(Cl+HCO_3) ratio.

Table 3. Classification of ground water samples according to WHO, (1996) in presence of F^-concentration and probable health effect

F^- (mg/l)	Effect on human health	Representing Samples	Total no. of simple
<0.5	Dental caries	M1	1
0.5-1.5	Prevents tooth decay	M2-M12, SD1-SD19, SD21-SD23, SD25	34
1.5-4.0	(Dental Fluorosis)	SD20, SD24, SD 26	3
>4.0	Pain in neck bones and back and Crippling Fluorosis	--	--

CONCLUSION

From the entire study it has been concluded that the contamination of fluoride in ground water is mainly due to weathering of fluoride bearing rock, and the source is dominated by sodium. All the statistical modeling explains that F^- has statistically strong significant association with Na^+, alkaline pH and Ca^{2+}. The water samples which containing high F^- in ground water are derived from Na-HCO_3 type water. Moreover the ratio of HCO_3^-/Ca^{2+}, Na^+/Ca^{2+}, Na^+/Na^++Cl^-, $HCO_3^-/\sum$anions and Gibbs diagram suggest that occurrence of fluoride contamination of the particular area are mainly due to weathering or dissolution of fluoride containing rock.

Belbari is emerging as a fluoride endemic area of South Dinajpur rather than other studied villages in both Malda and South Dinajpur districts. The peoples belongs from F^- endemic areas are need awareness about fluoride endemism's and should find alternative source of uncontaminated water for their living concern.

Acknowledgement Authors express their sincere thanks to all villagers for their unconditional help to complete this particular work. Authors are also extending their sincere gratitude to Professor (Dr.) J. K. Datta, Department of Environmental Science, The University of Burdwan, for his moral support and valuable suggestion for preparing this manuscript.

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