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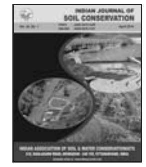

Hydrochemical characterization and spatial distribution of fluoride in groundwater of Delhi state, India

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ABSTRACT

The Fluoride (F) concentration along with other hydrochemical parameters of Delhi groundwater was analysed to characterize F pollution and its relation with other ions. Groundwater quality varied from fresh to saline with neutral to alkaline pH. The F concentration varied from 0.20-5.12 (0.89±0.89) mg L^{-1} and the concentration increased from eastern to western part of the study area. Out of total analysed samples, 24.3% contained F level lower than 0.5 mg L^{-1}, 55.4% from 0.5 to 1.0 mg L^{-1}, 9.5% from 1.0 to 1.5 mg L^{-1} and only 10.8% contained >1.5 mg L^{-1}. The molar ratio of different ions present in the groundwater system has identified the dominant reaction pattern occurring within the system. F was independent on other quality parameters except for pH. Positive correlation between F and pH indicated that alkaline condition improved solubility of F.

1. INTRODUCTION

Fluoride has long been recognised as one of the most significant natural groundwater-quality problems affecting arid and semi-arid regions of India. In India, approximately 62 million people including 6 million children suffer from fluorosis because of consumption of water with high F concentrations (Susheela, 1999). The F content in groundwater is governed naturally by climate, composition of the host rock, and hydrogeology and anthropogenically by use of phosphatic fertilizers, pesticides, sewage and sludges for agriculture, depletion of groundwater (Ramanaiah *et al*., 2006). An inventory of F concentration in drinking groundwater is thus important toward curbing the spread of fluorosis.

Delhi, a rapidly growing city is facing groundwater quality and quantity problems, forced to meet 50% of its water requirement from groundwater. Growing urbanization, exploding population and intensive agriculture are just some of the contributing factors. Delhi's groundwater level has gone down approximately by 8 meters within a span of 10 years (CGWB, 2006). Recently, a newspaper reported high F in groundwater of Delhi (The Hindu, 2005). Therefore, an intensive study was undertaken to characterize the groundwater contamination with special reference to F within Delhi state and its relation with other ions present in groundwater.

2. MATERIALS AND METHODS

Delhi is situated between the Himalayas and Aravali ranges in the heart of the Indian sub continent and lies between 28˚24′15″ and 28˚53′00″ N latitudes and 76˚50′24″and 77˚20′30″ E longitudes. The Delhi state have six administrative blocks, namely Najafgarh, Mehrauli, Alipur, Nangloi, Shahdara and main City block covering an area of 1484 km^2 with 16.753 million population (Census, 2011). The Alwar quartzites exposed in the area belong to Pre-Cambrian age. The quartzites are pinkish to grey in colour, hard, compact, highly jointed, fractured and weathered. These occur with interbeds of gneiss and are intruded locally by pegmatites and quartz veins. The basement or hard rock occurs at greater depth around 300 m below ground level. The climate is subtropical semiarid with average annual rainfall is about 700.8 mm and the annual evaporation is about 2565 mm. Out of total rainfall, 84% is received from south-west monsoon during the months of July, August, and September and the rest is received in the form of winter rain. The monthly mean temperature ranges from 21ºC to 41ºC, while the annual mean temperature recorded is 31.5ºC.

In May 2006, seventy four representative groundwater samples from different land use of the study area were collected (Fig. 1). Geographical coordinates and elevation of each sampling location was recorded using a handheld global positioning system (GPS). A few locations were also cross-checked with a differential GPS. Collected samples were analysed in the laboratory to measure the concentration of the hydrochemical parameters using standard procedures (APHA, 1995). F was measured by Alizarin S visual method (Crosby *et al*., 1968), using a Systronics UV-VIS Spectrophotometer. The spatial variation map of F was prepared using kriging technique in Arc GIS environment.

3. RESULTS AND DISCUSSIONS

Groundwater quality varied from fresh to saline with evidence of nitrate, chloride and F ions. The descriptive statistics of hydrochemical parameters along with water table depth are given in Table 1. The pH varied from 7.0 to 8.7 indicating neutral to alkaline in reaction, which was supported by the presence of large amount of Ca^{++} and Mg^{++}. The Na^{+} and K^{+} concentration varied from 21-2240 mg L^{-1} and 0.3-380 mg L^{-1}, respectively with irregular spatial distribution. The EC varied from 0.43-14.40 dS m^{-1}. The

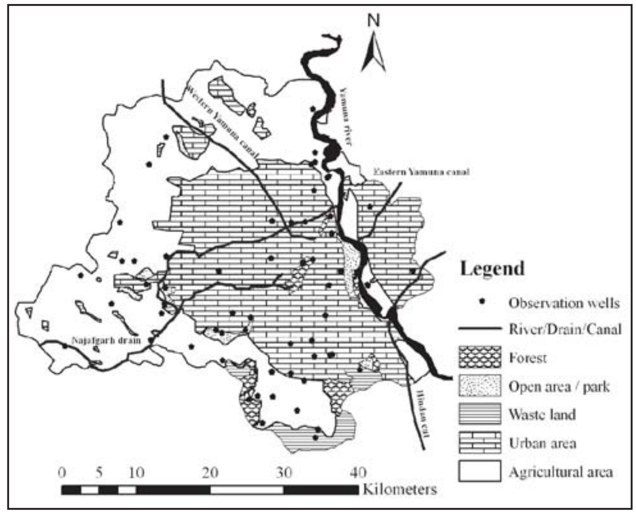

Fig. 1. Map of study area showing observation well locations in different land uses

bicarbonate concentration of groundwater ranged between 112 and 985 mg L^{-1} and mainly precipitated as calcium and magnesium bicarbonate. Chloride was mainly concentrated to the industrial and landfill sites and in the areas where the sewage water was used for irrigation purpose. The sum of the cations and anions were mainly Ca-Mg-HCO_3, Ca-Mg-Cl-SO_4, Na-K-HCO_3 and Na-K-Cl-SO_4 type.

The F concentration varied from 0.205.12 (0.89±0.89) mg L^{-1}. Out of total analysed samples, 24.3% contained F level lower than 0.5 mg L^{-1}, 55.4% from 0.5 to 1.0 mg L^{-1}, 9.5% from 1.0 to 1.5 mg L^{-1} and only 10.8% contained >1.5 mg L^{-1}. F distribution map indicated that F concentration

Table: 1
Descriptive statistics of groundwater quality parameters across the study area

Parameters	Range	Average	SD	Skewness	Kurtosis
Depth (m)	1.03-51.92	11.14	10.68	2.14	4.67
pH	7.01-8.71	7.75	0.37	0.35	-0.30
EC (dS m^{-1})	0.43-14.40	2.03	2.41	3.57	14.71
Hardness (mg L^{-1})	68-6731	533.82	842.68	5.98	41.49
Na (mg L^{-1})	21-2240	247.65	325.89	3.94	20.02
K (mg L^{-1})	0.32-380.01	28.98	74.31	3.92	15.35
Ca (mg L^{-1})	8.20-1584.12	109.47	190.99	6.65	50.15
Mg (mg L^{-1})	6.48-675.62	63.38	95.32	4.63	25.18
HCO_3 (mg L^{-1})	112-985	419.01	147.94	1.07	2.13
Cl (mg L^{-1})	25-5451	419.61	855.88	4.21	20.29
SO_4 (mg L^{-1})	0.80-1600	124.01	202.24	5.55	39.27
NO_3 (mg L^{-1})	0.50-140	31.02	28.14	1.69	3.98
F (mg L^{-1})	0.22-5.12	0.89	0.89	3.29	11.66

increased from eastern towards western part of the study area (Fig. 2). According to the guidelines of World Health Organization, water containing >1.5 mg L^{-1} fluoride is not suitable for drinking purpose (WHO, 2004). Nearly 11% of the study area comprising south-west part of South West block and western fringe of West and North West blocks showed F toxicity. Rest of the Delhi state is safe from groundwater F toxicity. The main source of F was the fluorite mineral found in quartzite, gneiss and pegmatite. The process of weathering of rock released F in soil and ground water. The alkaline water could mobilize F from fluorite with precipitation of calcium carbonate because the solubility of CaF_2 increased with increase in $NaHCO_3$ (Jha *et al*., 2013). In acidic form, F was absorbed in clay, and in alkaline form, it was desorbed, hence, alkaline pH was more favourable for F dissolution activity (Tirumalesh *et al*., 2007) and could be represented as:

$$CaF_2 + 2NaHCO_3 = CaCO_3 + 2Na^+ + 2F^- + H_2O + CO_2$$

The molar ratio of different ions present in the groundwater system can suggest the possible combinations; hence act as a relic to identify the dominant reaction pattern. The ratio of different ions (Table 2) in the groundwater indicated that Na and HCO_3 control the reaction dynamics. HCO_3/Cl ratio was found in the range of 0.02-12.71 (2.57 ± 2.42) indicate dominance of carbonate ions in the region. The HCO_3/Ca ratio observed in the range of 0.04-21.53 (2.61 ± 3.03) indicate favourable chemical condition for F dissolution (Saxena and Ahmed, 2003). The Na/Cl ratio was observed in the range of 0.18-9.26 (1.94 ± 1.50). The decrease in ratio indicated modification of sodium carbonate water by dissolution or mixing with sodium chloride, whereas increase in chloride may be due to local recharge (Dhiman and Keshari, 2006). The ratio value of

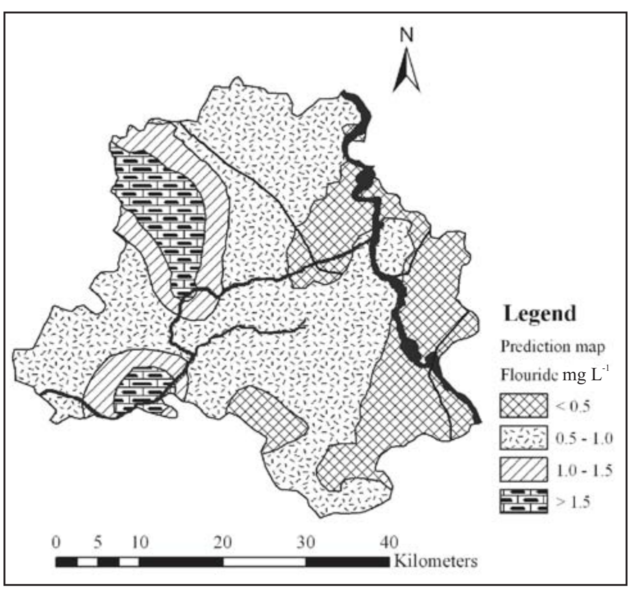

Fig. 2. Spatial variation of F in the groundwater of study area

Table: 2
Ratio values of different water quality variables across the study area

Sl.No.	Possible combinations	Ratio values Range	Average	SD
1.	HCO_3/Cl	0.02-12.71	2.57	2.42
2.	HCO_3/Ca	0.04-21.53	2.61	3.03
3.	Na/Cl	0.18-9.26	1.94	1.50
4.	Na/Ca	0.26-95.65	4.06	11.31
5.	Na/(Ca+Mg)	0.18-5.47	1.21	1.02

Na/Ca was observed in the range of 0.25-95.65 (4.06 ± 11.31) and the ratio value of Na/(Ca+Mg) was observed in the range of 0.18-5.47 (1.21 ± 1.02). The increase in ratio value may be due to lowering calcium activity whereby high sodium activates the dissolution of F bearing minerals at higher pH in groundwater system (Shaji *et al*., 2007). The ionic radius of F (0.136 nm) was same as that of hydroxyl ion which can be easily substituted for one another from water at high pH (Sreedevi *et al*., 2006).

4. CONCLUSIONS

The results of this investigation suggested a definite pattern in distribution of fluoride content (F) in Delhi. The river sides always showed low F concentration. The spatial variation map of F will be helpful for the planners and decision makers to start with developmental activities. The molar ratio of different ions present in the groundwater system has proved to be a efficient method to identify the dominant reaction pattern occurring within the groundwater system.

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