Proc. IAHS, 379, 351–356, 2018
https://doi.org/10.5194/piahs-379-351-2018

Fluoride in groundwater: a case study in Precambrian terranes of Ambaji region, North Gujarat, India

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Received: 31 December 2017 – Revised: 20 February 2018 – Accepted: 26 February 2018 – Published: 5 June 2018

Abstract. Fluoride is one of the critical ions that influence the groundwater quality. World Health Organization (WHO, 1970) and Bureau of Indian Standards (BIS, 1991) set an upper limit of 1.5 mg L^{-1} in F^- concentration for drinking water purpose and above affects teeth and bones of humans. The presence of fluoride in groundwater is due to an interaction of groundwater and fluoride bearing rocks. Fluoride rich groundwater is well known in granitic aquifers in India and elsewhere. Generally, the concentration of F^- in groundwater is controlled by local geological setting; leaching and weathering of bedrock and climatic condition of an area. The main objective of the present study is to assess the hydrogeochemistry of groundwater and to understand the abundance of F^- in groundwater in hard rock terranes of Ambaji region, North Gujarat. A total of forty-three representative groundwater samples were collected and analyzed for major cations and anions using ICP-AES, Ion Chromatograph (Metrohm 883 Basic IC Plus) and titration methods. The F^- concentration in groundwater of this study area ranges from 0.17 to 2.7 mg L^{-1}. Among, twenty groundwater samples have fluoride exceeding the maximum permissible limit as per the BIS (1.5 mg L^{-1}). It is also noticed that residents of this region are affected by dental fluorosis. The general order of the dominance of major cations and anions are $Ca^{2+} > Mg^{2+} > Na^+ > K^+$ and $HCO_3^- > Cl^- > F^-$ respectively. Geochemical classification of groundwater shows most of the samples are the alkaline earth-bicarbonate type. The semi-arid climatic conditions of the region, the dominance of granitoid-granulite suite rocks and the fracture network in the disturbed and brittle zone has facilitated the development of potential aquifers and enrichment in F^- concentration in this area. The concentration of fluoride is due to high evaporation rate, longer residence time in the aquifer zone, intensive and long term pumping for irrigation.

1 Introduction

The usefulness of groundwater to human is great extent. Groundwater is considered to be the major source of drinking water in India. Groundwater chemistry is largely a function of the mineral composition of the aquifer through which it flows (Brindha and Elango, 2011). It depends upon physical and geological factors such as evaporation rate, residence time, aquifer media, rock–water interaction, recharge capacity, and anthropogenic activities. Fluoride in groundwater may be of geogenic (natural) or anthropogenic (Human induced). Fluoride is widely distributed in the environment and it ranks 13th among other elements in order of abundance in the earth's crust. Its natural abundance in the earth's crust is 0.06 to 0.09 % and 300 mg kg^{-1}. Generally, fluoride does not exhibit any colour, taste or smell when dissolved in water and hence, it is difficult to determine through physical examination. Fluoride is naturally present due to weathering of rocks rich in fluoride. Groundwater in crystalline rocks, mainly granites/granite gneisses are particularly vulnerable to fluoride because they often contain abundant fluoride bearing minerals (fluorite, apatite, hornblende, and biotite). Fluoride rich groundwater is well-known in different types of geological terranes and aquifers (Reddy et al., 2010). Several studies have mentioned an increase in dissolved F^- concentrations with increasing groundwater residence time in the aquifer systems (Nordstrom and Jenne, 1977; Apambire et al., 1997; Edmunds and Smedley, 2013). Prolonged rock-water interactions through weathering of fluoride-rich rocks enrich fluoride concentration in the groundwater systems. Consequently, fluoride is leached out and dissolves

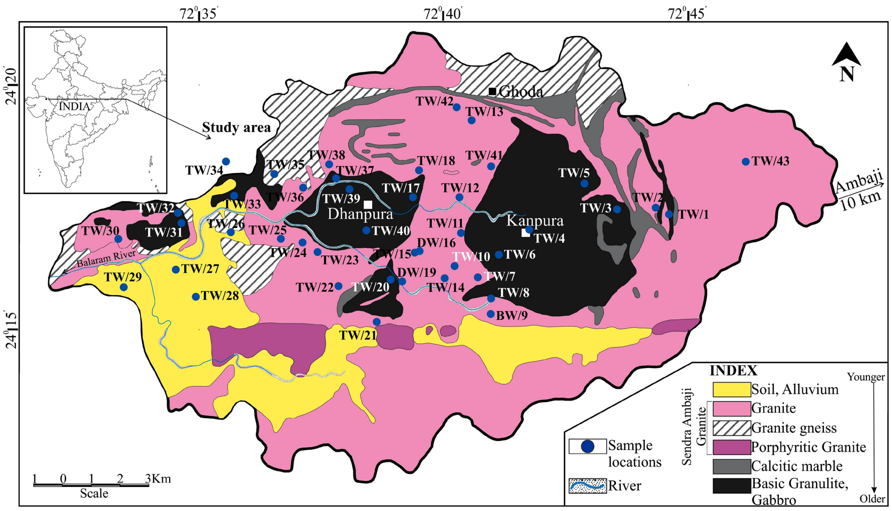

Figure 1. Geological map and sample locations of the study area (modified after Singh et al., 2010).

in groundwater and thermal gases (Deshmukh et al., 1995). Fluorite, apatite, mica (biotite) and hornblende are the minerals, which have the most effect on the geochemistry of F^- in groundwater (Dar et al., 2011) when they are interacted with groundwater and dissolved, leads to increasing F^- in groundwater (Rao, 2017). Groundwater interacting with crystalline rocks, mainly (alkaline) granites/granite gneisses (deficient in calcium) and hydrothermal solutions are probably to have reasonably high fluoride concentrations (Brunt et al., 2004). Ayenew (2008) studied that the higher concentration of F^- is due to low concentration of Ca^{2+} under an alkaline environment in Ethiopian groundwater. In different parts of China, Li et al. (2014, 2017) found that the high F^- was controlled by hydrogeological conditions, but also man-made activities and mixing of different recharge waters were also key reasons. High fluoride groundwater has been recorded as one of the most critical natural groundwater quality problems associated with hard rock areas in India (Handa, 1975; Reddy et al., 2010; Raj and Shaji, 2017; Shekhar et al., 2017). More than 200 million people from different nations suffer health problems due to high fluoride in groundwater (Ayoob and Gupta, 2006; Wu and Sun, 2016). In India, there has been an increase in the incidence of dental and skeletal fluorosis due to high fluoride concentration in drinking water. Dental fluorosis is common in 17 states of India with most pronounced in the states of Andhra Pradesh, Gujarat, Madhya Pradesh, Bihar, Punjab, Rajasthan, Tamil Nadu, Telangana, Uttar Pradesh and West Bengal (CGWB, 2015). World Health Organization (WHO, 1970) and Bureau of Indian Standards (BIS, 1991) set an upper limit of $1.5\,mg\,L^{-1}$ in F^- concentration for drinking water purpose and above affects teeth and bones of humans. In the literature, early researchers (Handa, 1988; Gupta, 1991; Sharma et al., 2005; Chinoy et al., 2005; Brindha and Elango, 2011; Raj and Shaji, 2017) discussed that fluoride and fluorosis were correlated with high concentration of fluoride ion in drinking water in different parts of India. In this paper, the data pertaining to fluoride in groundwater of Ambaji region of North Gujarat, India has been presented and discussed.

2 Materials and methods

2.1 Study area

The present area of study covers a part of Ambaji region (Ambaji-Amirgard-Dhanpura-Kanpura) situated in Banaskantha District of Gujarat state, India (Fig. 1). On the South, it is bounded by Mehsana district where high Fluoride in groundwater is reported. The area belongs to the Survey of India toposheet no. 45D/11, 45D/12 and 45D/15 (scale 1 : 50 000). The area has a diverse landscape and characterized by hilly upland with intermountain valleys, followed by piedmont zone with alluvium and residual hills and gently sloping vast alluvial plain. The area falls under semi-arid climate. Extreme temperatures (hot summer and dryness in the non-rainy seasons), erratic rainfall and high evaporation are the typical characteristic features of this type of climate. The annual rainfall of this region is 578.8 mm and is mostly received during the south-west monsoon season from June to September. Since it falls in a semi-arid type of climate, the rivers flowing through it are of ephemeral nature i.e. have water during monsoon only and dry up after monsoon period. The drainage network is constituted mainly by the Banas River and their tributaries (Balaram River). Geologically, the area belongs to the Meso-proterozoic South Delhi Terrane (1100–900 Ma), Aravalli craton. The rocks are pelitic-, calcareous-, calcareous and basic-granulites; these are intruded by three phases of Ambaji granites (G1, G2, G3). The rocks are extremely fractured along large-scale faults and shear zones (Singh et al., 2010).

2.2 Sampling and instrumentation

The area has been subjected to a high degree of deformation, which gives rise to fracture networks and this can be traced surficially and through remote sensing and GIS study. These intersecting fracture networks suggest that the water-yielding zones for phreatic and deeper aquifers are interconnected in hydraulic continuity. A total of forty-three ground water samples (Tube well/Bore well/Dug well) from different aquifers (Fig. 1) were collected in the month of May and June 2017 (Table 1) for the present study. In this area the depth of dug well ranges from 10 to 20 m b.g.l. with a diameter of 3 to 5 m and water level ranges from 3 to 15 m b.g.l. The depths of tube well are ranging from 60 to 90 m with 20 to 25 cm in diameter. Water samples were collected in HDPE bottles of 500 mL capacity. Samples collected in the field were filtered using 0.45 µm Millipore filter paper and Alkalinity of groundwater samples measured through titration methods. Water samples were brought to the laboratory for geochemical analysis. Groundwater field parameters (pH, EC, TDS, Temperature) were measured in the field using portable HANNA pH meter. Major cations and anions of water samples were determined using ICP-AES and Ion chromatograph (Metrohm 883 Basic IC Plus) with appropriate standards.

3 Results and discussion

Fluoride is one of the important ions that influence the groundwater quality. Fluorite (CaF_2) is the only principal fluorine mineral, mostly present as an accessory mineral in granites. The results of hydrogeochemical parameters of groundwater samples including fluoride concentration are presented in Table 1. Fluoride concentration in groundwater of Ambaji regions varied between 0.17 mg L^{-1} (Bhyla village) to 2.7 mg L^{-1} (Padni village). It was revealed that twenty groundwater samples have fluoride exceeding the maximum permissible limit as per the BIS (1.5 mg L^{-1}) and twenty-three groundwater samples are within the permissible limit (Table 2). Fluoride concentration in seven villages is very alarming in this region. The maximum concentration of fluoride was recorded in Padni (2.7 mg L^{-1}), Kengora (2.3 mg L^{-1}), Padliya (2.2 mg L^{-1}), Kothiavas (1.8 mg L^{-1}),Virampur (2 and 1.6 mg L^{-1}), Khemrajya (1.9 mg L^{-1}), Kanpura (1.5 (mg L^{-1}), Ajapur Mota (2 mg L^{-1}) and Dhanpura (1.8 mg L^{-1}). Wide variation of fluoride concentration is probably due to variation in chemical strata of different rocks.

Figure 2 shows the spatial distribution of fluoride concentration in different aquifers in Ambaji region. It was also found that high fluoride groundwater is concentrated mostly

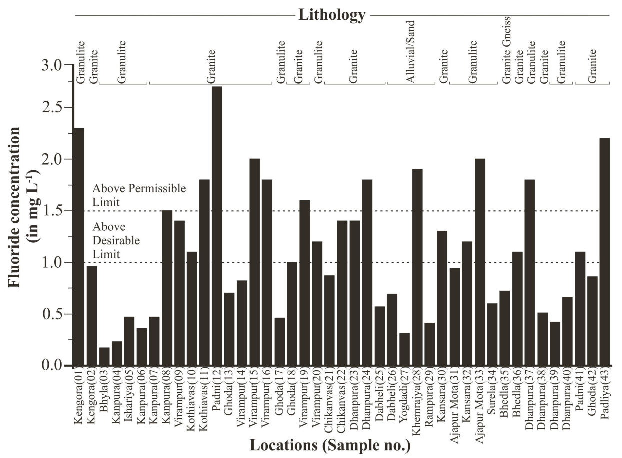

Figure 2. Spatial distribution of fluoride in groundwater of Ambaji region.

in the discharge areas than in the recharge areas with a trend of fluoride enrichment along the direction of flow. Other physico-chemical parameters such as pH, TDS, EC, major cations and anions of groundwater were analysed. Minimum, maximum and statistical values of water parameters are given in Table 3. The pH (H^+ ion in the solution) value is ranging from 6.44 to 7.45 in the study area and most of the groundwater samples found to be within the permissible limit. In the study area, Electrical conductivity (EC) ranged between 400 µS cm^{-1} (Dhanpura) to 2170 µS cm^{-1} (Padni) except TW-39 (Dhanpura) which is 4370 µS cm^{-1}. The high EC values of samples may be due to the primary minerals of earth's crust dissolved in water samples. All natural water contains different concentrations of Total Dissolved Salts (TDS) as a result of the dissolution of minerals within the rocks and soils. The essential ions that contribute TDS are carbonate, bicarbonate, sodium, potassium, calcium, magnesium, fluoride, chloride, nitrates and sulphates. TDS values varied from 200 to 1085 mg L^{-1} (Padni) except Dhanpura village (2185 mg L^{-1}) which is much higher than the permissible limit. The occurrence of other ions, mainly bicarbonate (HCO_3^-) and calcium (Ca^{2+}) ions also affects the concentration of fluoride in groundwater. Most of the samples are collected from aquifers of weathered and fractured zones occurring in the unconfined/semi unconfined conditions. Jacks (1990) studied that chemical weathering of minerals results in the formation of Ca- and Mg-carbonates which helps as good sinks for fluoride ions. But, it is the leachable state of fluoride ions that determine the water fluoride levels which is mainly due to (i) pH of the draining solutions and (ii) dissolved carbon dioxide in the soil. Dissolved fluoride in groundwater is possible only under favorable physico-chemical environments and with enough residence time (Handa, 1975).

Table 1. Physical and chemical analysis of groundwater of Ambaji region, North Gujarat, India.

Sample no.	Location	pH	Temp (°C)	EC (µS)	TDS ($mg\,L^{-1}$)	Fluoride ($mg\,L^{-1}$)	Lithology
TW/1	Kengora	6.87	27.6	780	390	2.3	Granulite
TW/2	Kengora	6.89	28.2	860	430	0.96	Granite
TW/3	Bhyla	6.92	29.7	1060	530	0.17	Granulite
TW/4	Kanpura	7.44	30	1090	545	0.23	Granulite
TW/5	Ishariya	6.96	30.1	1070	535	0.47	Granulite
TW/6	Kanpura	7.18	29.1	1820	910	0.36	Granulite
TW/7	Kanpura (Pedcholi)	6.88	29.8	1080	540	0.47	Granite
TW/8	Kanpura	6.97	29.7	1810	905	1.5	Granite
BW/9	Virampur (Golia)	7.04	29.9	1580	790	1.4	Granite
TW/10	Kothiavas	6.65	29.6	1580	790	1.1	Granite
TW/11	Kothiavas	7.11	28.7	940	470	1.8	Granite
TW/12	Padni	7.34	29.9	2170	1085	2.7	Granite
TW/13	Ghoda	6.89	29.5	770	385	0.7	Granite
TW/14	Virampur	6.77	29.2	1650	825	0.82	Granite
TW/15	Virampur	7.05	29.6	1010	505	2	Granite
DW/16	Virampur	6.85	28.8	1360	680	1.8	Granite
TW/17	Ghoda	6.86	30.1	1100	550	0.46	Granulite
TW/18	Ghoda	6.84	29.3	730	365	1	Granite
DW/19	Virampur	6.86	27.1	1980	990	1.6	Granite
TW/20	Virampur	7.15	28.6	1450	725	1.2	Granulite
TW/21	Chikanvas (Hadmana)	6.97	29	1100	550	0.87	Granite
TW/22	Chikanvas	6.84	29.4	950	475	1.4	Granite
TW/23	Dhanpura	6.66	29.3	1000	500	1.4	Granite
TW/24	Dhanpura	6.92	29.8	1160	580	1.8	Granite
TW/25	Dabheli	7.1	29.3	910	455	0.57	Granite
TW/26	Dabheli	7.07	30	1110	555	0.69	Alluvial/Sand
TW/27	Yogdadi	7.08	30.4	1050	525	0.31	Alluvial/Sand
TW/28	Khemrajya	7.08	29.6	1730	865	1.9	Alluvial/Sand
TW/29	Rampura	7.24	29.5	930	465	0.41	Alluvial/Sand
TW/30	Kansara	6.93	29.8	1360	680	1.3	Granite
TW/31	Ajapur Mota	7.25	30.7	1380	690	0.94	Granulite
TW/32	Kansara	7.08	29.9	1100	550	1.2	Granulite
TW/33	Ajapur Mota	7.45	28.6	1520	760	2	Granulite
TW/34	Surela	7.05	29.5	940	470	0.6	Granulite
TW/35	Bhedla	6.63	30.7	760	380	0.72	Granite Gneiss
TW/36	Bhedla	7.09	30	1050	525	1.1	Granite
TW/37	Dhanpura	7.24	29.6	1260	630	1.8	Granulite
TW/38	Dhanpura	6.44	30.2	400	200	0.51	Granite
TW/39	Dhanpura	7.09	29.6	4370	2185	0.42	Granulite
TW/40	Dhanpura	6.89	30.3	1010	505	0.66	Granulite
TW/41	Padni (Near School)	7.14	27.4	620	310	1.1	Granite
TW/42	Ghoda	7.27	29	790	395	0.86	Granite
TW/43	Padliya	6.77	29.2	900	450	2.2	Granite

Table 2. Comparison of fluoride content with drinking water standards (BIS, 1991 and WHO, 1970).

Parameter	BIS standard		WHO standard		Number of samples exceeding permissible limit	% of sample exceeding permissible limit
	Highest desirable ($mg\,L^{-1}$)	Maximum permissible ($mg\,L^{-1}$)	Highest desirable ($mg\,L^{-1}$)	Maximum permissible ($mg\,L^{-1}$)		
Fluoride	0.6–1.2	1.5	0.5	1.5	20	46

Table 3. Minimum, Maximum and Statistical values of water parameters.

Parameters	Units	Minimum	Maximum	Mean	Standard Deviation
EC	$\mu S\,cm^{-1}$	400	4370	1239.3	618.24
pH		6.44	7.45	6.99	0.2127
Ca^{2+}	$mg\,L^{-1}$	27	292.18	89.25	43.411
Mg^{2+}	$mg\,L^{-1}$	16.21	177.42	54.13	30.767
Na^{+}	$mg\,L^{-1}$	23.81	332.63	86.3	63.278
K^{+}	$mg\,L^{-1}$	0.16	14.62	3.22	2.478
CO_3^{-}	$mg\,L^{-1}$	0	0	0	0
HCO_3^{-}	$mg\,L^{-1}$	135	570	393.37	104.47
Cl^{-}	$mg\,L^{-1}$	29.99	769.76	121.47	128.63
F^{-}	$mg\,L^{-1}$	0.17	2.7	1.11	0.634
SO_4^{2-}	$mg\,L^{-1}$	13.05	266.97	44.16	42.223

In the present study, the general order of the dominance of major cations and anions are $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+}$ and $HCO_3^{-} > Cl^{-} > F^{-}$ respectively. Geochemical classification of groundwater shows most of the samples are of alkaline earth-bicarbonate type. These waters had temporary hardness because the concentration of Ca^{2+} and Mg^{2+} ions exceeds over Na^{+} and K^{+} ions. Simultaneously, values of CO_3^{2-} and HCO_3^{-} ions together were high as compared to SO_4^{2-} and Cl^{-} ions together. The groundwater samples are classified into Ca^{2+}-Mg^{2+}-HCO_3^{-}, Ca-Cl and Na-Cl hydrogeochemical facies which are due to lithology. The possible reason for the presence of fluoride-rich groundwater in the granitic area is due to release of fluoride from the fluoride-bearing minerals in the rocks. The alkaline water depleted in calcium is mainly responsible for the high concentration of F^{-} (Shaji et al., 2007). The long residence time of water in the aquifer system, caused by a high rate of evapotranspiration and a weathered zone of low hydraulic conductivity, are the additional factors that trigger to the dissolution of fluorine-bearing minerals for further increase of F^{-} content in groundwater. Geochemical behavior of groundwater from the study suggests that high fluoride groundwater contain low levels of Calcium and high alkalinity. High pH and HCO_3^{-}/Ca^{2+} suggest favorable chemical conditions for fluoride dissolution process.

Generally, the concentration of F^{-} in groundwater is controlled by local geological setting, leaching, and weathering of bedrock and climatic condition of an area. As the area falls under semi-arid climatic conditions, the dominance of granitoid-granulite suite rocks and the fracture network in the disturbed and brittle zone has facilitated the development of potential aquifers and enrichment in F^{-} concentration. Also, it is noticed that the concentration of fluoride is due to high evaporation rate, longer residence time and comparatively low rainfall in the aquifer zone, intensive and long term pumping for irrigation.

Therefore, to maintain the quality of ground water, it is recommended for continuous monitoring of physico-chemical parameters of water and can be used for drinking and other uses only after prior treatment.

Data availability. The data presented here are part of Rudra Mohan Pradhan's doctoral thesis work and yet, Ph.D. is not completed. However, individual data are available from the author(s) on request. Those interested in using this data may contact the corresponding author.

Competing interests. The authors declare that they have no conflict of interest.

Special issue statement. This article is part of the special issue "Innovative water resources management – understanding and balancing interactions between humankind and nature". It is a result of the 8th International Water Resources Management Conference of ICWRS, Beijing, China, 13–15 June 2018.

Acknowledgements. Authors are acknowledges to Ministry of Earth Sciences (MoES), Govt. of India and Department of Earth Sciences, Indian Institute of Technology Bombay for financial assistance. Rudra Mohan Pradhan thankful to Bhuban Mohan Behera and Abdul Rajik Khan for their help during field work. Thanks are due to the two anonymous reviewers for their valuable comments that improved the quality of this manuscript.

Edited by: Wenchao Sun
Reviewed by: two anonymous referees

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