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ORIGINAL ARTICLE

Assessment of fluoride contamination in groundwater from Basara, Adilabad District, Telangana State, India

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Abstract The major objective of this study was to locate the vulnerable areas in terms of fluoride contamination. A total of 34 groundwater samples were collected from major drinking water sources in rural areas of Basara, Telangana, and studied with reference to the distribution and hydrogeochemistry of fluoride. The geochemical trend of groundwater in the study area demonstrates that sodium is the dominant cation ($Na^+ > Ca^{2+} > Mg^{2+} > K^+$) and chloride is the dominant anion ($Cl^- > HCO_3^- > SO_4^{2-} > CO_3^{2-} > NO_3^- > F^-$). The fluoride concentration varied from 0.06 to 4.33 (1.13 ± 0.90) mg L^{-1} with the highest fluoride level at Karegaon village (4.33 mg L^{-1}). Seven locations showed the presence of fluoride in excess of permissible levels.

Keywords Groundwater · Fluoride · Granitic terrain · Basara · Telangana State

Introduction

Fluoride is an essential microelement for human health. However, if present in excess it contaminates groundwater. Fluoride is a major contaminant of groundwater reserves globally. Statistically, smaller quantities (<1.0 mg L^{-1}) in drinking water are usually considered to have a beneficial effect on the rate of occurrence of dental caries, particularly among children (WHO 2006), but excessive continuous exposure (>1.5 mg L^{-1}) to fluoride can give rise to a number of adverse effects, including dental fluorosis, skeletal fluorosis, increased rate of bone fractures, decreased birth rates, increased rate of urolithiasis (kidney stones), impaired thyroid function, and impaired development of intelligence in children (WHO 2006). Fluoride concentration is an important aspect of hydrogeochemistry, because of its impact on human health. The main potential health risks from fluoride are considered to be fluorosis or bone disease. Drinking water is a major source of human intake of fluoride, including its subsequent incorporation into food items. In skeletal fluorosis, high dose of fluoride replaces bone calcium in the form of calcium fluoride, and bones become soft, crumble and chalky white. In human health, high levels of fluoride are related to bone and teeth diseases as well as with negative effects on the reproductive and nervous systems (Edmunds and Smedley 1996). Fluorosis is a considerable health problem worldwide, which is afflicting millions of people in many areas of the world, for example East Africa, Turkey, India, southeastern Korea, China, Japan, Sri Lanka, Iran, Pakistan, Turkey, Algeria, Mexico, Korea, Italy, Brazil, Malawi, Jordan, Ethiopia, Canada, Norway, Ghana and Kenya.

In India, the excessive presence of fluorides in groundwater is present in nearly 177 districts covering 20 states and Telangana is one of them (Narsimha and Sudarshan 2016). In the early 1930s, fluorosis was reported only in four states of India, in 1986 it was 13, in 1992 it was 15, in 2002 it was 17, and now it is 20, indicating that endemic fluorosis has emerged as one of the most alarming public health problems of the country (Ayoob and Gupta 2006). Fluoride is present in the form of naturally occurring minerals as fluorite (CaF_2), apatite [$Ca_5(PO_4)_3F$], cryolite (Na_3AlF_6), topaz, tourmaline, muscovite, biotite, hornblende and villianmite (Handa 1975). Fluoride

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concentration in natural water depends on various factors such as temperature, pH, solubility of fluorine-bearing minerals, anion exchange capacity of aquifer materials (OH^- for F^-), and the nature of geological formations drained by water and contact time of water with a particular formation (Mithas Ahmad Dar et al. 2011). The fluoride in drinking water is mainly reported in the hard rock terrains in India (Handa 1975; Reddy et al. 2010). A number of cases of fluorosis have been reported mostly from the granite and gneissic complex of different states such as Telangana State (Ram Mohan Rao et al. 1993; Narsimha and Sudarshan 2013, 2016), Andhra Pradesh (Nagaraju Arveti et al. 2011), Odisha (Kundu et al. 2001). The present study region forms a part of the stable Dharwar Craton of the South Indian shield. It exposes rocks of peninsular gneissic complex (PGC), Dharwar Supergroup, and also of Deccan Traps. The PGC, which covers most of the area, comprises granites and gneisses. The Archaean crystalline rocks are represented by pink and grey granites and gneisses. Grey granites occupy dominant portion of the area. These rocks are composed of quartz, feldspars, biotite and hornblende. These are medium-grained to coarse-grained and equigranular in texture. The typical grey colour is due to the presence of the plagioclase feldspar and quartz. Pink granites are generally coarse and porphyritic and composed of potash feldspars, quartz, plagioclase, biotite and hornblende with some accessories. The main objective of this study is to assess fluoride concentration in groundwater and its relationship between the physico-chemical parameters.

Materials and methods

Groundwater samples of 34 locations around Basara, Adilabad District, Telangana State, were collected in pre-washed polyethylene narrow-mouth bottles (Fig. 1). Samples were collected only from the bore well/hand pumps, which are in regular use for drinking purposes. Groundwater was collected after pumping the wells for 5–10 min and rinsing the bottles for two to three times with water to be sampled. For sample collection, preservation, and

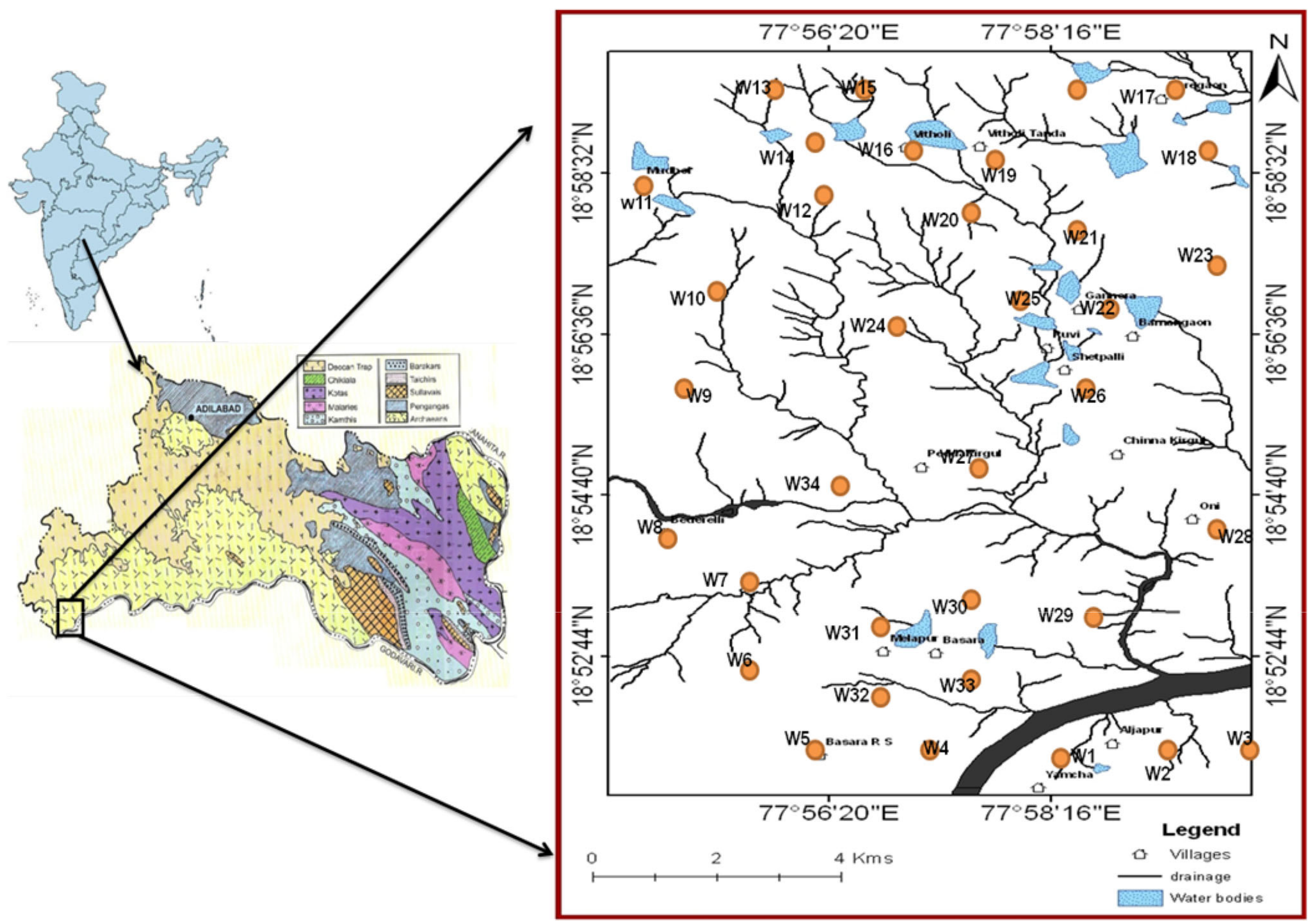

Fig. 1 Location map of the Basara area and its groundwater locations along with drainage pattern from Adilabad district, Telangana State, South India

Table 1 Analytical data for the groundwater samples from the study area (EC in µS cm^{-1}, remaining mg L^{-1}, except pH)

Sample ID	Longitude	Latitude	pH	EC	TDS	TH	CO_3^{2-}	HCO_3^-	Cl^-	SO_4^{2-}	NO_3^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	F^-	Ionic Balance
W1	77°58′11″	18°51′17″	6.61	392	251	160	0	85	128	42.5	22	42	13	76	4	0.37	2.6
W2	77°58′53″	18°51′42″	6.61	863	552	325	0	152	340	75	60	104	16	122	25	0.57	–8.0
W3	77°59′42″	18°51′31″	7.02	314	201	150	0	110	64	25	17.5	38	13	47	3	0.74	7.1
W4	77°56′49″	18°51′32″	6.57	235	150	115	0	85	50	5	4	30	9	15	8	0.46	2.0
W5	77°55′46″	18°51′29″	7.24	549	351	40	21	140	96	75	38	10	4	154	2	2.2	–1.9
W6	77°55′21″	18°52′32″	7.61	863	552	80	18	350	113	150	40	20	7	250	3	2.22	–3.0
W7	77°55′21″	18°53′33″	7.52	2039	1305	650	12	134	638	375	24	105	10	420	4	0.91	–8.2
W8	77°54′59″	18°54′13″	6.47	2118	1355	850	0	98	1010	300	80	339	1	370	4	0.06	–6.3
W9	77°55′6″	18°55′56″	6.77	1177	753	500	33	128	110	250	38	125	22	120	5	0.48	–0.0
W10	77°55′9″	18°57′6″	7.62	392	251	35	12	135	53	120	42	10	2	161	4	2.56	3.2
W11	77°54′55″	18°58′27″	8.03	471	301	85	15	128	50	80	40	8	16	121	4	2.48	5.7
W12	77°56′12″	18°58′16″	7.3	392	251	115	18	125	89	50	35	38	5	118	4	1.19	5.4
W13	77°55′36″	18°59′26″	6.9	471	301	160	15	98	124	50	15	36	17	102	1	1.12	5.3
W14	77°56′6″	18°59′1″	7.53	314	201	110	15	120	43	7.5	32	26	11	63	2	1.13	6.9
W15	77°56′30″	18°59′32″	7.62	314	201	150	12	110	74	12.5	13	40	12	49	2	0.96	4.1
W16	77°57′3″	18°58′48″	6.78	549	351	250	18	49	266	17.5	60	66	21	95	2	0.94	–5.3
W17	77°58′14″	18°59′35″	6.97	314	201	85	15	61	89	7.5	4	32	1	46	2	0.54	–6.3
W18	77°59′17″	18°58′51″	7.27	314	201	35	15	110	64	2.5	130	10	2	119	2	4.33	–3.0
W19	77°58′20″	18°58′41″	6.52	549	351	270	15	61	227	25	0.8	66	25	82	3	0.43	3.2
W20	77°57′37″	18°58′5″	7.42	471	301	75	18	125	96	27.5	42	18	7	129	3	1.46	4.0
W21	77°58′19″	18°57′45″	7.07	314	201	95	18	79	42	5	56	24	8	66	3	1.43	8.1
W22	77°58′41″	18°57′5″	7.28	471	301	150	21	103	120	37.5	35	20	24	110	3	2.44	4.8
W23	77°58′80″	18°56′85″	6.57	627	401	235	9	67	287	20	56	68	16	120	7	0.27	–3.4
W24	77°56′48″	18°56′49″	7.16	863	552	210	24	116	467	50	60	36	29	312	4	2.03	–0.1
W25	77°57′44″	18°57′9″	7.21	471	301	90	30	120	89	51	48	28	4	139	4	1.25	3.7
W26	77°58′27″	18°56′8″	7.18	314	201	125	15	85	85	15	2	38	7	61	4	0.76	6.0
W27	77°58′1″	18°54′57″	6.89	392	251	185	12	134	74	10	27	38	21	43	4	0.86	2.5
W28	77°59′22″	18°54′2″	6.91	706	452	160	15	165	227	32.5	35	42	13	169	5	0.78	–1.0
W29	77°58′35″	18°53′10″	7.15	627	401	210	30	183	252	25	32	85	6	120	39	0.26	–5.2
W30	77°57′20″	18°53′28″	6.84	392	251	170	18	104	156	37.5	8	50	10	92	4	0.75	–1.3
W31	77°56′34″	18°53′10″	6.71	549	251	315	24	122	308	17.5	60	102	14	94	16	0.71	–8.8
W32	77°56′45″	18°52′14″	7.73	235	150	95	30	73	89	27.5	25	22	10	89	5	0.54	2.0
W33	77°57′19″	18°52′24″	6.68	627	401	310	0	98	234	50	18	104	12	107	5	0.11	6.9
W34	77°58′19″	18°52′26″	7.44	549	351	120	18	250	110	50	42	18	18	191	4	1.02	6.3
Minimum			6.47	235	150	35	0	49	42	2.5	0.8	8	1	15	1	0.06	–8.8
Maximum			8.03	2118	1355	850	33	350	1010	375	130	339	29	420	39	4.33	8.1
Average			7.094	595.24	377.8	197	15.18	120.7	184	62.53	36.5	54.1	11.94	129	5.71	1.13	1.0

analysis, standard methods (APHA 1995) were followed. The water samples were analyzed for various hydrochemical parameters such as pH, electrical conductivity (EC), total hardness (TH) as $CaCO_3$, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), chloride (Cl^-), sulphate (SO_4^{2-}), nitrate (NO_3^-), fluoride (F^-). Using pH/EC/TDS meter (Hanna HI 9811-5), the EC and pH of water samples were measured in the field immediately after the collection of the samples. Total dissolved solids (TDS) were computed as per (Hem 1985) from EC values. Ca^{2+} and Mg^{2+} were determined titrimetrically using standard EDTA. Chloride was estimated by $AgNO_3$ titration. Na^+ and K^+ were measured using flame photometer (Model 130 Systronics Flame Photometer). SO_4^{2-} and NO_3^- were determined by colorimetry with an UV visible spectrophotometer. The fluoride concentration in water was determined electrochemically, using a fluoride ion-selective electrode (APHA 1995). This method is applicable to the measurement of fluoride in drinking water in the concentration range 0.01–1000 mg L^{-1}. The electrode used was an Orion fluoride electrode, coupled to an Orion electrometer.

Results and discussion

The various physico-chemical parameters obtained from analysis of water samples from Basara area are presented in Table 1. The geochemical trend of groundwater is shown in a box plot (Fig. 2) and demonstrates that sodium is the dominant cation ($Na^+ > Ca^{2+} > Mg^{2+} > K^+$) and chloride is the dominant anion ($Cl^- > HCO_3^- > SO_4^{2-} > CO_3^{2-} > NO_3^- > F^-$). The groundwater of the study area has pH values ranging from 6.47 to 8.03 (7.09 ± 0.40), which indicates that the groundwater is mildly acidic to alkaline in nature. Na^+ and Cl^- concentration varies in the range 15–420 and 42–1010 mg L^{-1}, respectively (Table 1). However, Na^+ and Cl^- show a good trend of increasing concentrations with increasing TDS (Fig. 3a, b), suggesting anthropogenic sources such as sewage, household waste, etc. (Williams et al. 1999; Subrahmanyam and Yadaiah 2000; Choi et al. 2005; Jalali 2009). The fluoride content of the groundwater is in the range 0.06–4.33 (1.13 ± 0.90) mg L^{-1} and the average value is 1.13 mg L^{-1} with the highest fluoride level at Karegaon village (4.33 mg L^{-1}) and lowest at Bederelli village (0.06 mg L^{-1}). The highest fluoride-bearing areas are found to be coincidental with the lowest calcium and high bicarbonate values (Table 1). In terms of % contribution, the fluoride level >1 was observed in 41% at 14 locations (Table 1; W5, W6, W10–W14, W18, W20–W25, W34) and 20% of the groundwater has more than 1.5 mg L^{-1} of fluoride at seven locations (Table 1; W5, W6, W10, W11, W18, W22, W24), which is not useful for drinking purpose. The spatial

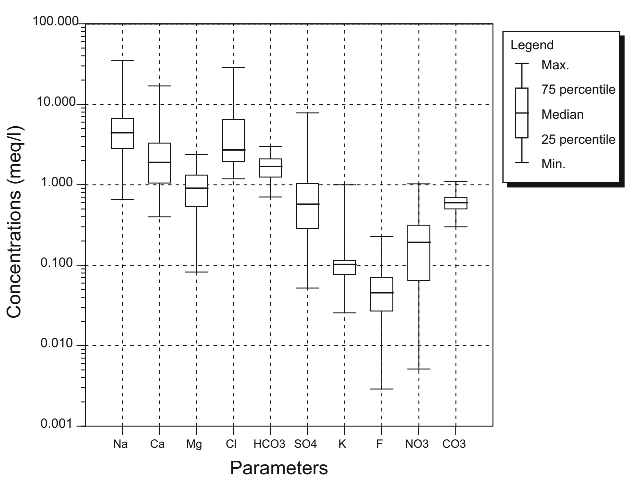

Fig. 2 Box plot for the maximum, minimum and average of the chemical constituents in groundwater of the study area, Adilabad district, Telangana State, India

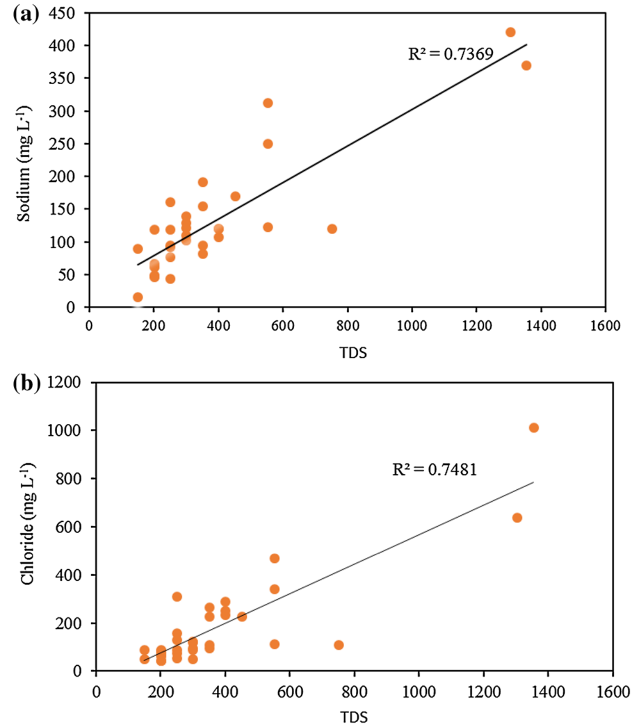

Fig. 3 Relationship between **a** TDS and sodium, **b** TDS and chloride in groundwater of the Basara region, Adilabad district, Telangana State, South India

distribution of fluoride in groundwater samples in the study area is shown in Fig. 4, with contour levels. Higher concentrations (>1.5 mg L^{-1}) of fluoride are noticed in the northeastern part of the study area where the geology of the study area is complex, comprising the pink granites. The lowest concentration is noted in the central to southern part of the study region. Few patches of fluoride with concentrations ranging between 1.8 and 2.4 mg L^{-1} are noticed in the corner of south-western, above middle part of the study province. However, major ion chemistry of groundwater is examined by using (Piper 1944) trilinear diagram (Fig. 5a) to identify chemical alteration in groundwater. The groundwater samples plotted in all the zones of the anions plot field (triangle on the right) revealed that the majority of the samples plotted towards the $HCO_3^- + CO_3^{2-}$ corner for only a few and remaining towards elevates the chloride type, indicating the predominance of this anion in the groundwater (Fig. 5a). Similarly, the samples plotted in all the zones of the cation plot field (triangle on the left) revealed that the majority of the samples are falling towards the $Na^+ + K^+$ corner and decreasing the calcium ions, indicating the prevalence of sodium and potassium. Furthermore, the diamond-shaped field of the Piper diagram can be classified into (1) Ca^{2+}–Mg^{2+}–Cl^-–SO_4^{2-} (2) Na^+–K^+–Cl^-–SO_4^{2-} (3) Na^+–K^+–HCO_3^- (4) Ca^{2+}–Mg^{2+}–HCO_3^-. It manifests that the lower concentrations of fluoride (<1.5 mg L^{-1}) are confined to Na^+–K^+–Cl^-–

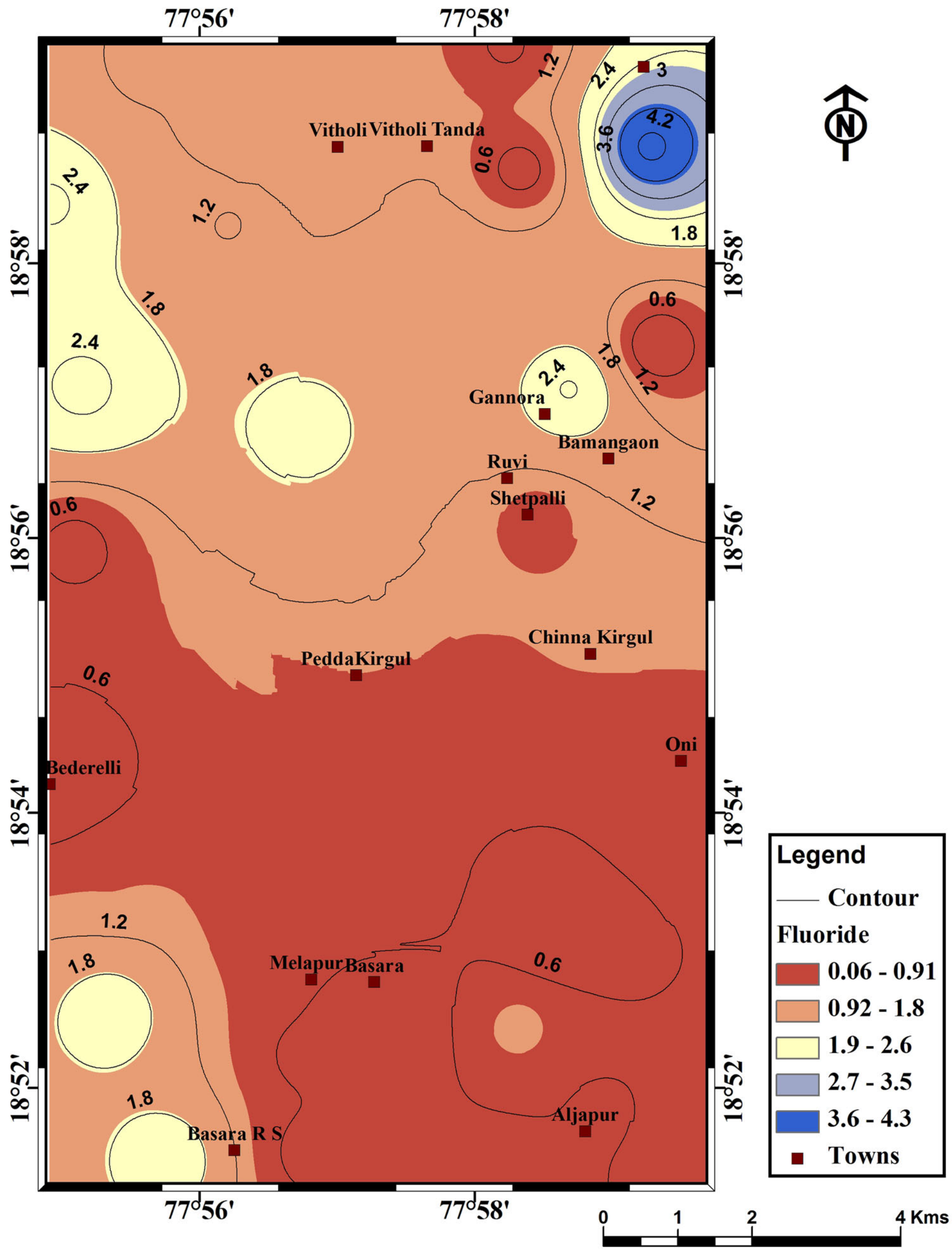

Fig. 4 Spatial distribution of fluoride in Basara region, Adilabad district, Telangana State, South India

SO_4^{2-}, Ca^{2+}–Mg^{2+}–HCO_3^- and Ca^{2+}–Mg^{2+}–Cl^-–SO_4^{2-} zones. The higher concentrations of fluoride (>1.5 mg L^{-1}) are associated with Na^+–K^+–HCO_3^-, indicating that the fluoride-rich water is normally associated with higher sodium and bicarbonate concentrations in the Basara region (Fig. 5a). It is also evident from Fig. 5b that the fluoride concentrations are increasing with increase of $Na^+/(Na^+ + Ca^{2+})$, since Na^+ replaces Ca^{2+} in a cation-exchange process, leading to enhancement of fluoride concentration in groundwater (Su et al. 2013; Li et al. 2014a, b).

Correlation analysis was employed to understand the interrelationship between fluoride and other physico-chemical parameters (Table 2). The results of this analysis indicate that only pH shows significant correlation with fluoride ($r = 0.564$; Table 2). The values of pH for the majority of the high-fluoride groundwater samples are within the range 7.16–8.03, indicating that the high-fluoride groundwater is commonly alkaline water. Thus, it is the higher alkalinity of groundwater which activates the leaching of fluoride resulting in its higher concentration (Tiwari et al. 2008). Several studies suggested that the increase in the fluoride concentration in groundwater was accompanied by increasing the sodium concentration as well as pH, and interpreted this as an indication that high sodium, pH values increase the release of fluoride or

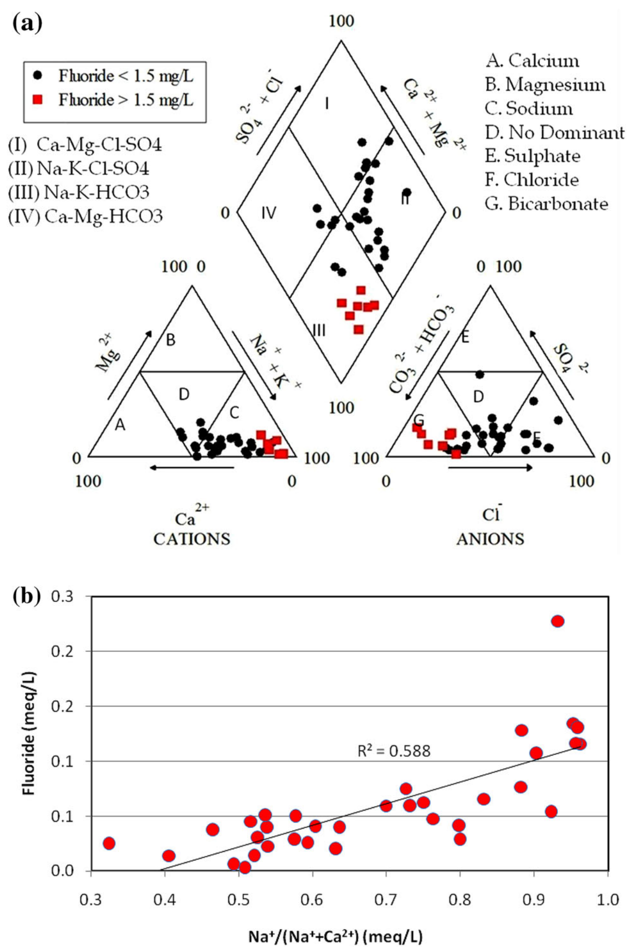

Fig. 5 **a** Piper (trilinear) diagram of groundwater samples in the study area. **b** Relation between fluoride concentration and $Na^+/(Na^+ + Ca^{2+})$ ratio for groundwater samples from Basara region

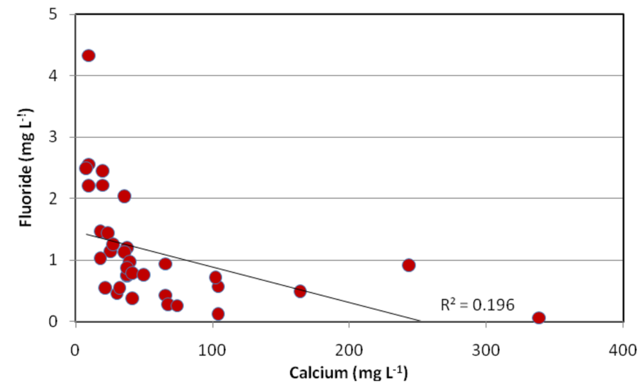

Fig. 6 Mutual relationship of fluoride and calcium

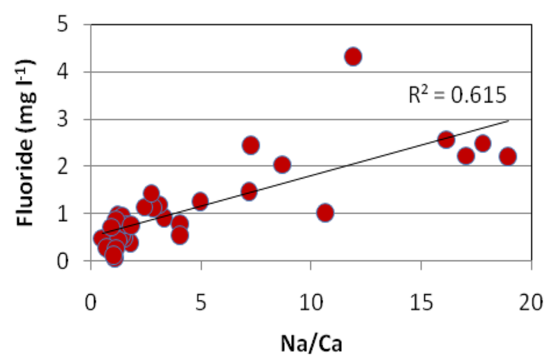

Fig. 7 Mutual relationship between fluoride and Na/Ca

exchanging of fluoride by OH^- (Moghaddam and Fijani 2008). Groundwater with high fluoride generally contains low levels of calcium. The calcium ion activity in the natural environment is controlled mainly by carbonate ion, which forms insoluble calcite. This is because the

Table 2 Correlation matrix of physico-chemical parameters of groundwater samples

	pH	EC	TDS	CO_3^{2-}	HCO_3^-	Cl^-	TH	Ca^{2+}	Mg^{2+}	Na^+	K^+	SO_4^{2-}	NO_3^-	F^-
pH	1.000													
EC	−0.169	1.000												
TDS	−0.158	0.998	1.000											
CO_3^{2-}	0.385	−0.112	−0.122	1.000										
HCO_3^-	0.172	0.283	0.276	0.038	1.000									
Cl^-	−0.357	0.873	**0.863**	−0.234	0.132	1.000								
TH	−0.429	0.897	0.887	−0.209	0.132	0.870	1.000							
Ca^{2+}	−0.392	0.903	0.893	−0.227	0.121	0.875	0.985	1.000						
Mg^{2+}	−0.212	0.006	0.002	0.099	0.068	0.011	0.126	−0.047	1.000					
Na^+	0.214	0.849	**0.850**	0.059	0.310	0.622	0.601	0.611	−0.023	1.000				
K^+	−0.188	0.083	0.067	0.107	0.498	0.180	0.146	0.154	−0.042	−0.092	1.000			
SO_4^{2-}	−0.054	0.933	0.935	−0.052	0.257	0.685	0.817	0.834	−0.061	0.875	−0.042	1.000		
NO_3^-	**−0.439**	0.363	0.343	−0.211	0.042	**0.605**	0.450	0.418	0.205	0.059	0.235	0.113	1.000	
F^-	***0.564***	−0.202	−0.196	0.021	***0.239***	−0.321	−0.464	−0.444	−0.124	***0.109***	−0.292	−0.162	−0.205	1.000

Values in bold indicate significant correlation

Bold italic indicate significant correlation with fluoride

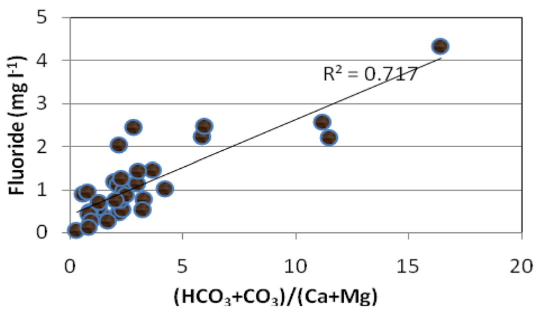

Fig. 8 Mutual relationship of fluoride and $(HCO_3^- + CO_3^{2-})/(Ca^{2+} + Mg^{2+})$

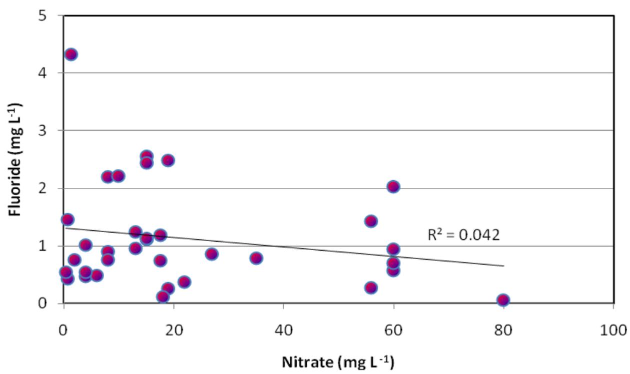

Fig. 9 Mutual relationship of fluoride and nitrate

correlation coefficient is very low ($r^2 = 0.196$), which is evident in Fig. 6, where most data points cluster in the low-Ca and low-F region. Low concentration of Ca^{2+} corresponding to high fluoride in the water has earlier been reported by Maina and Gaciri (1984), and also in agreement with earlier observations (Handa 1975) that elevated fluoride in the groundwater was generally associated with low calcium (Fig. 6) and a high amount of bicarbonates (Table 1) is in line with the findings of Chae et al. (2007), He et al. (2013), and Narsimha and Sudarshan (2016). The relation between $(HCO_3^- + CO_3^{2-})$, $(Ca^{2+} + Mg^{2+})$, Na^+, Ca^{2+} and F^- can be better explained by plots between Na^+/Ca^{2+} and $(HCO_3^- + CO_3^{2-})/(Ca^{2+} + Mg^{2+})$ vs F^-, respectively (Figs. 6, 7). It is observed that fluoride concentration increases with increase in the ratios of Na^+/Ca^{2+} and $(HCO_3^- + CO_3^{2-})/(Ca^{2+} + Mg^+)$ (Figs. 7, 8) and therefore, the findings of the present study are in close proximity with earlier studies conducted in Nalgonda, Pamber, Vaniyar and Vellore districts (Rao et al. 1993; Sajil Kumar 2012; Brindha et al. 2016). However, there is no appropriate correlation between fluoride and nitrate; consequently, it reveals that the fluoride is geogenic and nitrate is anthropogenic (Fig. 9). Eventually, the granite bedrock containing fluorine-bearing minerals like apatite, biotite and hornblende is responsible for the elevated concentration of fluoride in the groundwater of the Basara region. When these minerals come in contact with groundwater, they dissolve, which leads to the increase of fluoride in groundwater.

The Gibbs diagram is extensively used to assess the dissimilarity between groundwater controlled by rock-water interaction (i.e., leaching and dissolution), evaporation and precipitation processes (Gibbs 1970; Feth and Gibbs

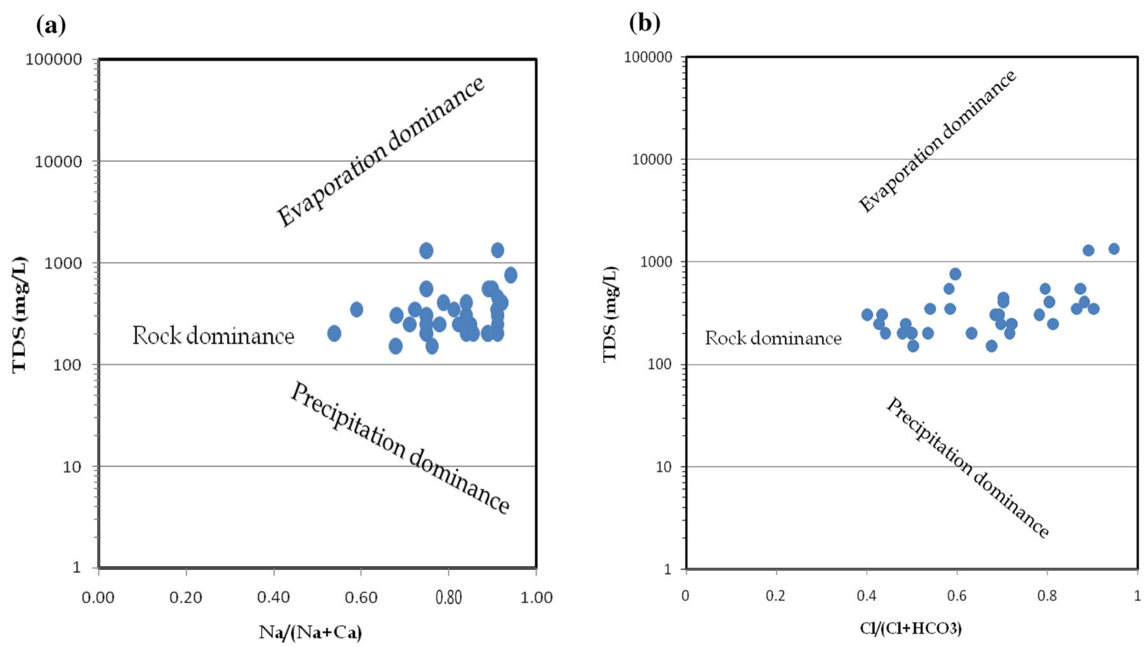

Fig. 10 Mechanism controlling the quality of groundwater which represents the ratio of (**a**) $Na^+/(Na^+ + Ca^{2+})$ and (**b**) $Cl^-/(Cl^- + HCO_3^-)$ as a function of TDS (Gibbs 1970)

1971; Narsimha and Sudarshan 2016; Reddy et al. 2016). The groundwater samples fall in the rock-water interaction dominance, few samples are under evaporation dominance zones and none of the groundwater sampling points lie in the precipitation dominance process (Fig. 10a, b). The predominance of rock-water interaction or weathering processes probably indicates that the geochemistry of groundwater over the area is due to influence of weathered rocks in groundwater fractured zones. Therefore, high concentration of fluoride in groundwater is derived from the rock-water interaction, i.e., weathering of minerals and accessory minerals present in the granite and gneissic rocks in the study area. Many investigations note that the relationship between F^- and TDS is due to the considerable influence of rock-water interaction on fluoride enrichment as well as other major components (Wang et al. 2009; Mamatha and Rao 2010; Li et al. 2014a; Narsimha and Sudarshan 2016). Thus, geochemical behaviour of groundwater from the study area suggests that the high-fluoride groundwater contains concentrations of Na^+ and Ca^{2+}, which are greatly influenced by the cation exchange and has high alkalinity. However, weathering of rocks and leaching of fluorine-bearing minerals are the major reasons which contribute to elevated concentration of fluoride in groundwater (Patel et al. 2014; Jagadeshan et al. 2015). It has to be considered that the groundwater with high-fluoride concentration was significantly affected by geogenic activity.

Conclusion

The study area of Basara region forms a part of Adilabad district of Telangana State, South India. The present study reveals that it is mildly acidic to alkaline in nature. It is observed that 20% of groundwater locations possess enrichment of fluoride concentrations above 1.5 mg L^{-1}; hence this groundwater is unsuitable for drinking. Fluoride concentrations are high in the northeastern part of the study area. The Gibbs diagram [log TDS vs $(Na^+/Na^+ + Ca^{2+})$ and log TDS vs $Cl^-/(Cl^- + HCO_3^-)$] reveals that the water-rock interaction is the primary source for the high fluoride concentration in the groundwater. Furthermore, high sodium and low calcium concentration favoured the release of fluoride into groundwater during rock-water interaction. Water type is identified as $Na^+–K^+–Cl–SO_4^{2-} > Na^+–K^+–HCO_3^- > Ca^{2+}–Mg^{2+}–HCO_3^- > Ca^{2+}–Mg^{2+}–Cl^-–SO_4^{2-}$. The $Na^+–K^+–HCO_3^-$ type of groundwater favours the higher fluoride content and the calcium-depleted alkaline water under high pH conditions is effective in releasing fluoride from the fluorine-bearing minerals.

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