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THEMATIC ISSUE

Geochemical evaluation of fluoride and groundwater quality for drinking and irrigation purposes in a weathered gneissic rock aquifer of southern penisular India

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Abstract

Assessment of groundwater chemistry of a region is important to determine its usability. The objective of this study is to understand the hydrochemistry and groundwater quality especially with respect to fluoride in the Perambalur District, Tamil Nadu, India where groundwater is used without treatment for drinking and agricultural purposes. Groundwater samples were collected from 44 locations once every three months between September 2015 and July 2018. Mixed Ca–Mg–Cl, Ca–HCO_3, Na–Cl and mixed Ca–Na–HCO_3 types were the predominant hydrochemical facies. Fissile hornblende biotite gneissic rocks had more fluoride concentration than the charnockite rocks in the study area. These weathered rocks leach high concentration of fluoride to groundwater. Ion exchange processes played a major role in fluoride dynamics in groundwater. Fluoride concentration was comparatively high in the wells where the groundwater level fluctuation is at deeper levels. Groundwater in most of the wells were suitable for drinking purpose based on the ions analysed, but few wells were unsuitable for drinking and irrigation purposes. Based on health risk assessment, children are likely to be more affected than infants and adults through intake of high fluoride groundwater from the study area.

Keywords Groundwater · Drinking · Irrigation · Fluoride · Health risk · Perambalur · Southern India

Introduction

Groundwater quality varies due to rainfall, geology, hydrogeochemical processes, rock-sediment–water interaction, influence of anthropogenic sources, and from the impact of climate change. Understanding the variation in the concentration of dissolved constitutents in groundwater provides information on the origin, flow path, recharge mechanisms, geochemical reactions and also it helps to determine its suitability for specific purposes. Over-exploitation of groundwater can lead to negative impact on the groundwater quality, especially in the shallow aquifers (Khan et al. 2016; Pophare et al. 2014; Salameh 2008). Managing the groundwater resources from potential pollutants and contaminants is a difficult task and it requires long-term systematic monitoring of the water levels and quality as well as studying the stages of groundwater development in relation to the rate of replenishment. Though contamination of groundwater from human activities can be curtailed to a larger extent, geogenic contamination poses severe peril to freshwater demand and supply worldwide (Eawag 2015).

India, the world's largest groundwater user is facing crisis due to uncontrolled exploitation of the resource established by drying of wells and rapid lowering of water levels. Groundwater use is largely in the agricultural sector (89%), followed by the domestic (9%) and industrial sectors (2%) (World Bank 2010). About 50% of urban and 85% rural water requirements are met by groundwater (CGWB 2011). Common anthropogenic pollutants causing problems in

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Indian groundwaters are salinity (Manivannan and Elango 2019; Gopal et al. 2020; Biplab et al. 2021), nitrate (Karunanithi et al. 2020; Abdur et al. 2021), chromium (Kanagaraj and Elango 2019; Christina et al. 2021), lead, cadmium and other trace metals (Sharma et al. 2019; Monika et al. 2021). Key geogenic pollutants are arsenic (Sridharan and Nathan 2018; Sumant et al. 2021), fluoride (Jagadeshan et al. 2015a; Raju 2017; Chetan and Surindra 2019), iron and salinity (Kumar et al. 2019; Paul et al. 2019; Gopal et al. 2020; Dinesh Kumar et al. 2022). Among these natural contaminants, arsenic and fluoride are global threat to human health (Eawag 2015). Arsenic is more common contaminant in groundwater of the north eastern part of India and in the alluvial plains of the Ganges and Brahmaputra river basins (Chakraborti et al. 2017a, b; Jain et al. 2018). Fluoride contamination is more prominent throughout the nation (Brindha et al. 2011, 2016; Kalpana et al. 2018; Kanagaraj and Elango 2019; Ajaykumar et al. 2020).

Since adequate drinking water is not supplied through pipes to home in rural parts of India, the large rural population use groundwater without treatment. Hence, the periodical assessment of groundwater quality and hydrogeochemistry of major ions is necessary. One such region is located in the western part of Perambalur district, Tamil Nadu, India. Based on the groundwater availability and the gross groundwater pumped for various uses this region is categorised as over-exploited (CGWB 2017). Of the 11 administrative revenue blocks in Perambalur district, 6 are over-exploited, 3 are semi-critical and 2 are safe based on the current groundwater extraction practises (TWAD 2018**.** Thus, it is well-established that groundwater forms the major source of water for the rural population of this area for domestic use including drinking and agricultural needs.

Compared to the available literature on the hydrogeochemistry of other parts of Tamil Nadu, this region has not been throughly investigated till now. Previously, only the general groundwater quality assessment (Kasthuri et al. 2007) and the suitability for agricultural use (Ahamed et al. 2013) has been carried out. The excessive groundwater abstraction together with decifit in rainfall, reduced recharge and subsequent decline in groundwater level led to the identification and demarcation of the groundwater potential zones in the area for future groundwater development activities (Anbarasu et al. 2019). Uranium in the groundwater was used the health risk assessment to understand the presence of uranium and distribution based on rock types (RamyaPriya et al. 2021). These studies however did not draw out an understanding of the hydrogeochemistry, and the presence of fluoride and its dynamics in groundwater. Considering that groundwater is used for drinking and irrgation uses, it is necessary to assess the possible impact of groundwater use on the human health and plant growth. Hence, the present study aims at quantifying the fluoride abundance in groundwater, understanding the sources, factors controlling its occurrence, release meachanisms and its movement in groundwater in the hard rock aquifers of upper part of the Chinnar river basin which forms a part of the Perambalur district, Tamil Nadu, India. Health risk assessment is also carried out to understand the potential health hazards due to ingestion of groundwater on the rural population. This study also assesses the general hydrogeochemistry and the suitablity of groundwater for drinking and agricultural uses based on the major ions.

Methodology

Field work

Initially, a detailed hydrogeological field investigation was carried out and information on the geological outcrop, depth of the wells, its dimensions, purpose of use etc. were noted. The electrical conductivity (EC) and groundwater level in the wells were also measured in the field. Based on these information, forty-four representative wells (Fig. 1) which were spread uniformly throughout the study area were selected for long-term monitoring. Totally 436 samples were collected from the 44 open wells (diameter from 4 to

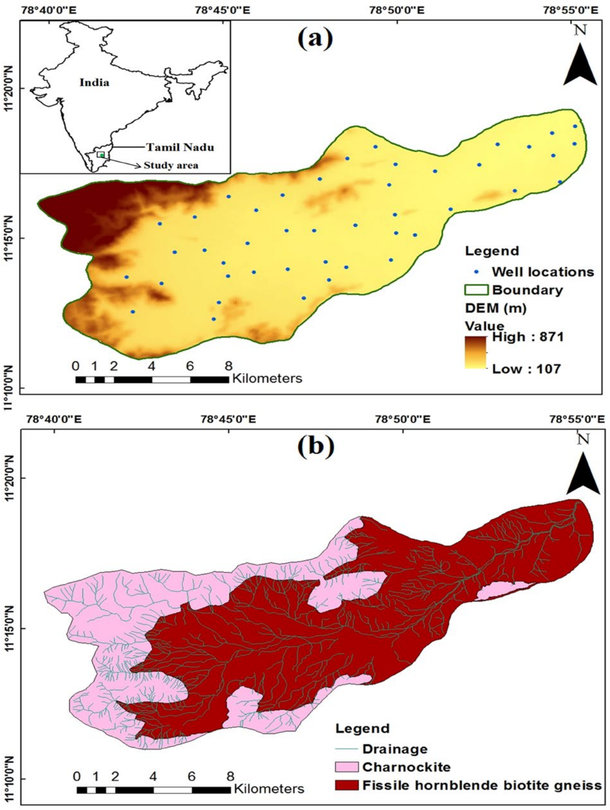


Fig. 1 **a** Study area, **b** geology and drainage map of study area

10 m) once every three months between September 2015 and July 2018. Water level indicator (Solinist101) was used to measure the water levels. Temperature, pH and EC were measured in situ using a portable meter (Oakton Pcstestr 35). Sampling bottles were cleaned with dilute HCl and then washed thoroughly with distilled water before sample collection. Groundwater samples were filtered using millipore filter paper (0.45 µm) and filled in the sample collection bottles without air bubbles, properly labelled, stored in a cooler and brought to the laboratory for further analysis.

Laboratory methods

Carbonate and bicarbonate concentrations were measured immediately after sample collection by volumetric titration against diluted H_2SO_4 using the Aquamerck test kit (1.11009.0001). Calcium and magnesium were measured by volumetric titration. Sodium, potassium and nitrate concentrations were determined by a flame photometer. Sulphate concentration was measured by UV–VIS spectrophotometer (Systronics UV–VIS 118). Chloride concentration was measured by using titrando 905. Fluoride in groundwater was measured by a fluorimeter (Extech F700) which can measure from 0.1 to 10 mg/l. All analytical methods were followed as per standard procedures (APHA 2012). Verification of efficiency of the analysis was done by testing blanks and standards. Ion balance error was calculated and was within the ± 5%.

Exposure dose of fluoride

Fluoride intake water was calculated by using the given formula (USEPA 2011).

$$\text{Exposure dose} = \frac{C \times \text{WI}}{\text{BW}}, \tag{1}$$

where, C is the measured fluoride concentration (mg/l), WI is the daily water intake (l/day) and BW is the body weight (kg). The water intake and body weight varies based on the different age groups. The water amount adopted for infants was 250 ml/day, for children was 1.5 l/day and for adults was 3 l/day (Planning Commission 2011). Infants, children and adult's body weight were considered as 6.9, 18.7 and 57.5 kg, respectively (ICMR 2009).

Description of the study area

The study area is located in the hard rock terrain of Perambalur district, Tamil Nadu, India (Fig. 1a) and it covers about 220 km^2. This region extends between the latitudes of 11° 8′ 16″ N and 11° 21′ 14″ N and longitudes of 78° 38′ 45″ E and 78° 57′ 35″ E. This region experiences semi-arid climate with temperature ranging from 20 to 42 °C. The average annual rainfall is about 900 mm with rainfall in the northeast monsoon, southwest monsoon and summer accounting for 60%, 25% and 15% of the rainfall respectively. The region is surrounded by the mountains of Eastern Ghats (Pachamalai) in the north, west and south, except for the eastern part. This area is characterized by discontinuous small with gentle uplands and the elevation ranges from 400 to 900 m msl. Subdendritic with trellis is the drainage pattern in this area (Fig. 1a). Agriculture is the primary activity in this area.

Geology and hydrology

The geology of the study area was demarcated based on the geological map obtained from the Geological Survey of India (GSI 1995) (Fig. 1b). This region comprises of fissile hornblende biotite gneisses and charnockite rocks. The gneissic and chanockite rocks are fractured over a thickness ranging mostly between 20 and 31 m and 18 and 25 m, respectively. The sand and silt occur in small quantity along the river course. Groundwater occurs in unconfined conditions in the weathered and fractured rocks. The aquifer thickness ranges between 15 and 35 m. Specific yield is between 80 and 210 lpm (litre per minute) and top soil thickness is from 0 to 6 m. Long-term fluctuation in groundwater table during the period from 1998 to 2007 indicates that there is no major change in its annual trend (CGWB, 2009).

Results and discussion

Statistical summary of the parameters analysed in groundwater are given in Table 1. The suitability of the groundwater for drinking purpose based on its properties are presented in Table 2.

Drinking water quality

Groundwater in this area is used for drinking and domestic purposes. Hence, the various hydrochemical parameters measured were compared with the prescribed values proposed for drinking water quality by the Bureau of Indian Standards (BIS 2012) and World Health Organisation (WHO 2011) (Table 2). Evaluation of water for drinking and domestic purposes depends on the parameters mentioned in the following sections.

Total dissolved salts and total hardness

The total dissolved salts (TDS) is the total amount of salts dissolved in water and comprises of major, minor ions and trace elements. TDS was calculated from the EC using the formula,

Table 1 Statistical summary of physicochemical parameters in the groundwater

Parameters	Unit	Pre-monsoon ($N=216$)			Post-monsoon ($N=220$)		
		Minimum	Maximum	Mean	Minimum	Maximum	Mean
pH	No unit	6.9	8.5	–	6.6	9.5	–
EC	µS/cm	446	3920	1449	232	3370	1442
TDS	mg/l	285	2508	927	148.5	2156.8	920
TH	mg/l	64.7	789.1	240	30	710	275
Calcium	mg/l	19	245.8	70	9	199	84
Magnesium	mg/l	4.2	66.6	17.5	1.8	68	23
Sodium	mg/l	21.5	470	107.6	13.1	537	108
Potassium	mg/l	0.4	5.7	2.6	1.1	15.1	5.1
Carbonate	mg/l	0	0	0	10	12.2	10.4
Bicarbonate	mg/l	158	597	282	55	647	312
Chloride	mg/l	17.6	589	170.5	16	579	184
Sulphate	mg/l	20	190	89	8	238	88
Nitrate	mg/l	6.9	37	15.7	0	24.8	8.6
Fluoride	mg/l	0.1	1.5	0.4	0.2	2.7	0.7
Na %	%	14.1	89	43.4	7	81.7	38
SAR	meq/l	0.4	31	3.1	0.3	17.4	3.8
RSC	meq/l	− 9.4	6.7	− 0.58	− 10.4	4.7	− 1.1
KR	meq/l	0.1	7.5	0.9	0.1	4.3	0.8
MH	meq/l	13.4	48.8	29.1	13.5	47.4	29.3
PI	%	21.2	120.7	69.4	21.7	124.3	64.2

EC electrical conductivity, *TDS* total dissolved salt, *TH* total hardness, *Na%* sodium percentage, *SAR* sodium adsorption ratio, *RSC* residual sodium carbonate, *KR* Kelly's ratio, *MH* magnesium hazard, *PI* Permeability Index

Table 2 Groundwater suitability for drinking purposes according to BIS (2012) and WHO (2011)

Parameters	Unit	References	Acceptable limit	Permissible limit	Number of samples exceeding acceptable limit	Number of samples exceeding permissible limit
pH	No unit	BIS (2012)	6.5–8.5	No relaxation	5	–
TDS	mg/l	BIS (2012)	500	2000	361	17
TH	mg/l	BIS (2012)	200	600	314	10
Ca	mg/l	BIS (2012)	75	200	232	2
Mg	mg/l	BIS (2012)	30	100	58	Nil
Na	mg/l	WHO (2011)	–	200	–	52
K	mg/l	WHO (2011)	–	12	–	8
CO_3	mg/l		–	–	–	–
HCO_3	mg/l		–	–	–	–
Cl	mg/l	BIS (2012)	250	1000	105	Nil
SO_4	mg/l	BIS (2012)	200	400	8	Nil
NO_3	mg/l	BIS (2012)	45	No relaxation	Nil	–
F	mg/l	BIS (2012)	1	1.5	44	28

$$\text{TDS in mg/l} = \text{EC in } \mu\text{S/cm} \times 0.64. \quad (2)$$

TDS was ranging between 148.5 and 2508 mg/l with an average of 959.3 mg/l (Table 1). Groundwater in this area was fresh based on the classification proposed by Freeze and Cherry (1979). BIS (2012) has proposed 500 mg/l and 2000 mg/l of TDS as the acceptable and permissible limit in drinking water. Groundwater in this area exceeds the maximum permissible limits for drinking water quality in

4% of the groundwater samples (Table 2). TDS decreases after the monsoon period due to dilution effect.

Calcium, magnesium and carbonate ions are the essential components determining the total hardness (TH) of groundwater. The TH in water was calculated from the given formula,

$$\text{Total hardness}\left(CaCO_3\right) = 2.5\left(Ca^{2+}\right) + 4.1\left(Mg^{2+}\right). \quad (3)$$

All values are expressed in mg/l. TH ranges from 30 to 789.1 mg/l with an average of 291 mg/l (Table 1). Groundwater of this area can be classified as moderately hard, hard and very hard water for drinking uses (Sawyer and McCarty 1978) (Fig. 2). About 16% wells were moderately hard, 34% wells were hard and 50% wells were very hard. About 71% of the samples exceed the acceptable limit and 2% of the samples exceed the permisbble limit (Table 2).

Cations and anions

In the study area, 52% of the samples exceeded the accepted limit for calcium (Table 2). The acceptable limit of magnesium is 30 mg/l in drinking water (BIS 2012) and its 13% of the groundwater samples exceeded the magnesium concentration in groundwater. Concentration of sodium exceeded the permissible limit (WHO 2011) in 12% of the samples and potassium exceeded the permissible limit (WHO 2011) in 2% of the groundwater samples. Carbonate content was low in groundwater. There were prescribed limits for carbonate and bicarbonate in drinking water. Chloride and sulphate also did not pose any threat to groundwater quality. About 24% of the groundwater samples exceeded the acceptable limit for chloride, but still were within the permissible limits. Sulphate in groundwater exceeded the acceptable limit in 2% samples (Table 2). However, it was within the permissible limit.

Minor ions

Nitrate concentration ranged up to 37 mg/l with an average of 10.8 mg/l in groundwater. There was no threat to groundwater quality and for drinking use based on nitrate (Tables 1, 2). Concentration of fluoride was ranging from 0.1 to 2.7 mg/l with average of 0.6 mg/l in the study area (Table 1). Table 3 shows that the fluoride concentration varied seasonally in the sampling locations.

The suitability of groundwater for drinking purpose in terms of TDS, TH, cations and anions was assessed as a combination of all these parameters. If all the parameters

Table 3 The temporal variation in the suitability fluoride for drinking

Month	Percentage of samples		
	< 1 mg/l	1–1.5 mg/l (permissible range)	> 1.5 mg/l
Sep-15	88	12	0
Jan-16	83	5	12
Jul-16	90	10	0
Oct-16	70	16	14
Jan-17	67	19	14
Apr-17	88	10	2
Jul-17	90	10	0
Oct-17	86	4	10
Jan-18	81	7	12
Jul-18	88	12	0
Total	83	10	7

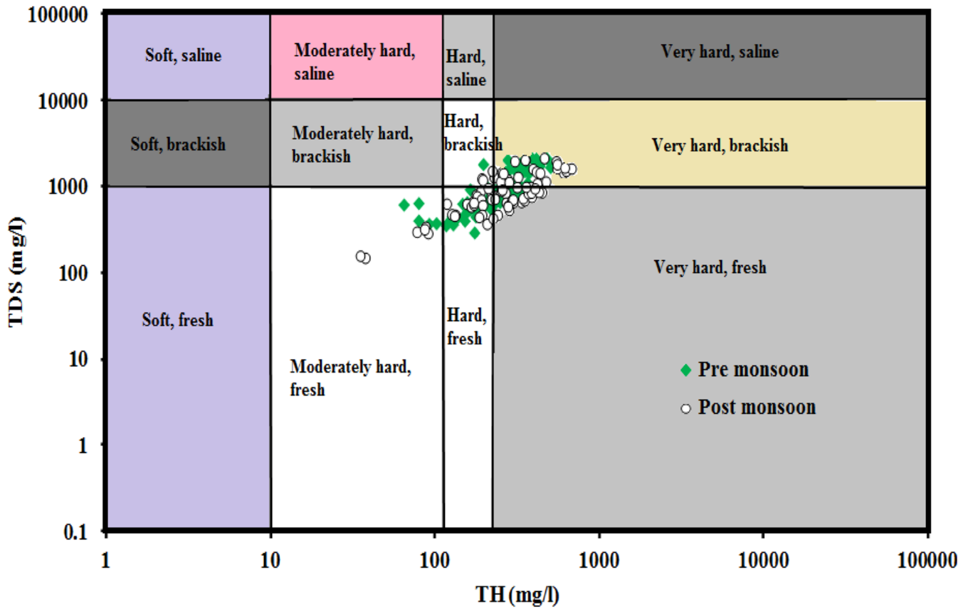


Fig. 2 Groundwater types based on total dissolved solids and total hardness

were within the permissible limits during three times of the four sampling times in a year, then the groundwater is considered as suitable for drinking. Based on the integration of all the parameters considered for drinking water quality analysis, 84% of the study area is suitable (Fig. 3).

Irrigation water quality

Groundwater in this area is widely used for irrigation purpose. Quality of groundwater for irrigation purpose was determined by various paramters as listed in following sections (Table 4).

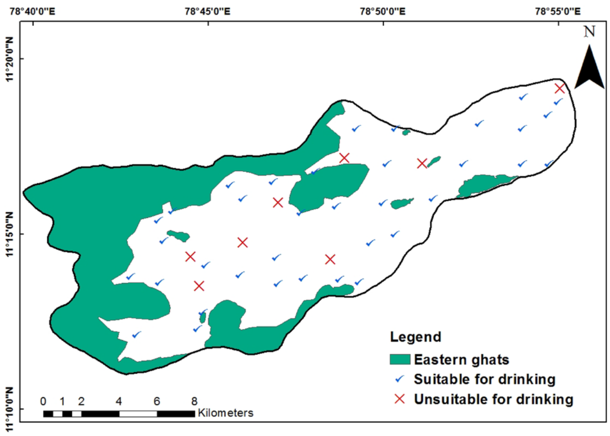


Fig. 3 Sampling locations indicate suitability for drinking use

Salinity and alkalinity hazard

High concentration of dissolved salts in groundwater can hinder soil permeability and may decrease the crop yield. The United States Salinity Laboratory (USSL) diagram (Richards 1954) classifies groundwater suitability for irrigation as two types: salinity hazard and sodium hazard. The salinity hazard is determined based on the EC and the sodium hazard is determined by using SAR equation. SAR determines the relative amount of sodium with respect of calcium and magnesium and is given as

$$\mathrm{SAR} = \frac{\mathrm{Na}^{+}}{\sqrt{\frac{\mathrm{Ca}^{2+}+\mathrm{Mg}^{2+}}{2}}}, \tag{4}$$

where the values are expressed in meq/l. Salinity hazard is further classified into four classes: C1, C2, C3, C4 and sodium hazard i.e. SAR is classified into S1, S2, S3, S4. SAR is plotted against EC in the USSL diagram. EC ranged up to 3920 μS/cm and classification based on EC show 16% of the samples were good, 70% were suitable and 14% were doubtful in the study area (Fig. 4a, Table 4). SAR values were between 0.3 and 31 meq/l with average of 3.5 meq/l (Table 1). Based on SAR, 93% of the locations were excellent and 7% of the locations were good for irrigation purpose (Fig. 4a).

Table 4 Classification of groundwater for irrigation uses

Parameters	Range	Classification	Percentage of wells exceeding the permissible limit
Na% (Wilcox 1955)	0–20	Excellent	07
	20–40	Good	47
	40–60	Suitable	36
	60–80	Doubtful	10
	> 80	Unsuitable	Nil
SAR (Richards 1954)	0–10	Excellent	93
	10–18	Good	07
	18–26	suitable	Nil
	> 26	Poor	Nil
RSC (Richards 1954)	< 1.25	Good	86
	1.25–2.5	Permissible	07
	> 2.5	Unsuitable	07
EC (Richards 1954)	< 250	Low salinity hazard	02
	250–750	Medium salinity hazard	14
	750–2250	High salinity hazard	70
	> 2250	Very high hazard	14
Kelley's ratio (Kelley 1957)	< 1	Suitable	82
	> 1	Unsuitable	18
MH (Szaboles and Darab 1964)	< 50	Suitable	100
	> 50	Unsuitable	Nil
PI (Donean 1964)	Class I	Max. permeability	77
	Class II	75% of Max. permeability	18
	Class III	25% of Max. permeability	05

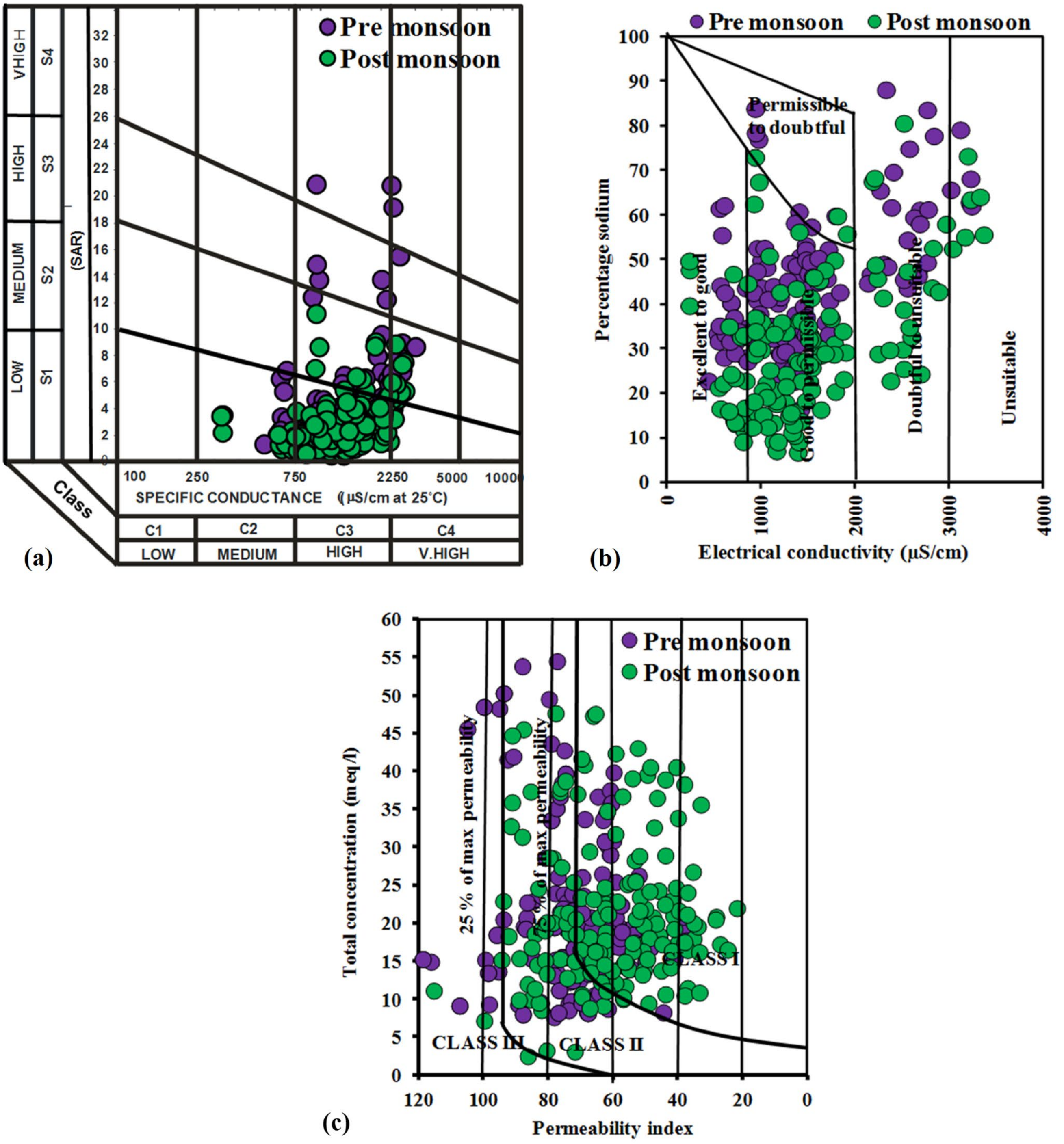


Fig. 4 Groundwater suitability for irrigation purpose based on **a** sodium and salinity hazard, **b** EC and sodium percent and **c** permeability index

Sodium percent

Sodium is an essential component in groundwater, which at high concentrations is not favorable for plant growth. If sodium is excess in groundwater, it will reduce the permeability of the soil and can clog the soil layer. Based the sodium content, Wilcox (1955) proposed an index known as the sodium percentage (Na%) for classifying the water quality for irrigation use. It is given as,

$$\text{Na\%} = \frac{\left(\text{Na}^{+} + \text{K}^{+}\right)}{(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^{+} + \text{K}^{+})} \times 100. \tag{5}$$

All concentrations given in meq/l. Na% was found to range from 7 to 89 meq/l with an average of 40.7 meq/l. Wilcox plot between EC and Na% (Fig. 4b) indicates that 54% of the groundwater samples had excellent to good quality, 36% were good to suitable and 10% of the samples has doubtful to unacceptable water quality for irrigation (Table 4). Figure 4b shows that most of the groundwater in the area is suitable for irrigation purposes.

Residual sodium carbonate

Alkalinity hazard in groundwater can be understood from the residual sodium carbonate (RSC). When sodium concentration is high in groundwater, calcium and magnesium will precipitate more in the soil and cause soil infiltration problems. RSC is calculated as (Ragunath 1987),

$$\text{RSC} = \left(\text{CO}_3^{2-} + \text{HCO}_3^{-}\right) - \left(\text{Ca}^{2+} + \text{Mg}^{2+}\right). \tag{6}$$

All concentrations given in meq/l. RSC of groundwater was between − 10.4 and 6.7 meq/l with average of − 0.8 meq/l (Table 1). Based on RSC, 86% of the groundwater samples were good, 7% were suitable and 7% were unacceptable for irrigation purpose (Table 4).

Permeability index

Calcium, sodium, magnesium, and bicarbonate ions affect the soil permeability. Hence, based on the concentration of these ions, the permeability index (PI) was proposed by Doneen (1964) to determine the quality of water for irrigation and. PI is calculated based on the formula given below

$$\text{PI} = \frac{\text{Na}^{+} + \sqrt{\text{HCO}_3^{-}}}{\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^{+}} \times 100. \tag{7}$$

All concentrations given in meq/l. PI in groundwater was ranging from 21.2 to 124.3 meq/l with average of 66.8 meq/l. The relation between PI and total concentration of ions in groundwater is plotted in Fig. 4c. The results show that 77% wells were good, 18% wells were within permissible limit, 5% wells were unsuitable for irrigation purpose (Table 4).

Kelly's ratio

Kelle's ratio (KR) to evaluate water quality for irrigation purposes was suggested by Kelly (1957) and is calculated as given below

$$\text{KR} = \frac{\text{Na}^{+}}{\text{Ca}^{2+} + \text{Mg}^{2+}}. \tag{8}$$

All concentrations given in meq/l. When sodium concentration is higher than the concentration of calcium and mangensium, it will affect the soil sailinity and is unsuitable for irrigation. So, if the KR is < 1, it is good for irrigation purposes. When KR is beyond 1, water is unacceptable for irrigation purpose. KR was found to range from 0.1 to 7.5 meq/l with an average of 0.9 meq/l (Table 1). About 18% of the groundwater samples were unsuitable for irrigation in the study area. Therefore, the results show that most of the groundwater wells were suitable for irrigation purpose.

Magnesium hazard

When exchangeable magnesium concentration is high in groundwater, it may cause infiltration issues. Szabolcs and Darab (1964) have proposed the following magnesium hazard (MH) index for irrigation purposes

$$\text{MH}(\%) = \frac{\text{Mg}^{2+}}{\text{Ca}^{2+} + \text{Mg}^{2+}} \times 100. \tag{9}$$

All concentrations given in meq/l. If MH is greater than 50%, it is considerd as harmful and unsuitable for irrigation purpose. MH ranges from 13.4 to 48.8 meq/l with an average of 28.6 meq/l in groundwater (Table 1). Based on MH, all sampling locations are within the maximum permissible limit. Hence, groundwater can be used for irrigation purpose with respect to MH.

The above indices determine the irrigation water quality based on each index at a particular point in time. An overall irrigation water quality map was prepared based on the EC, SAR, Na%, RSC, PI, KR and MH. If the groundwater quality based on these indices were suitable three times out of the

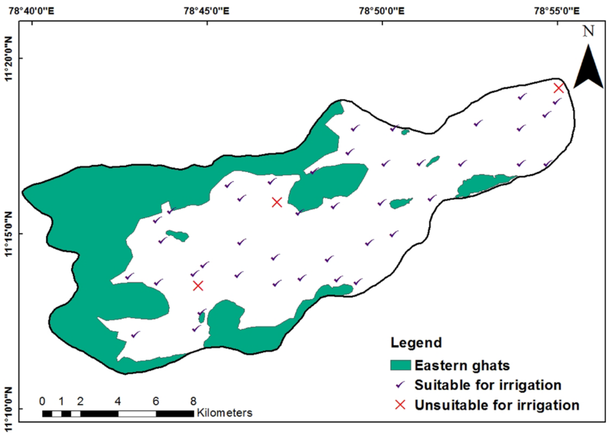


Fig. 5 Suitability of groundwater for irrigation use

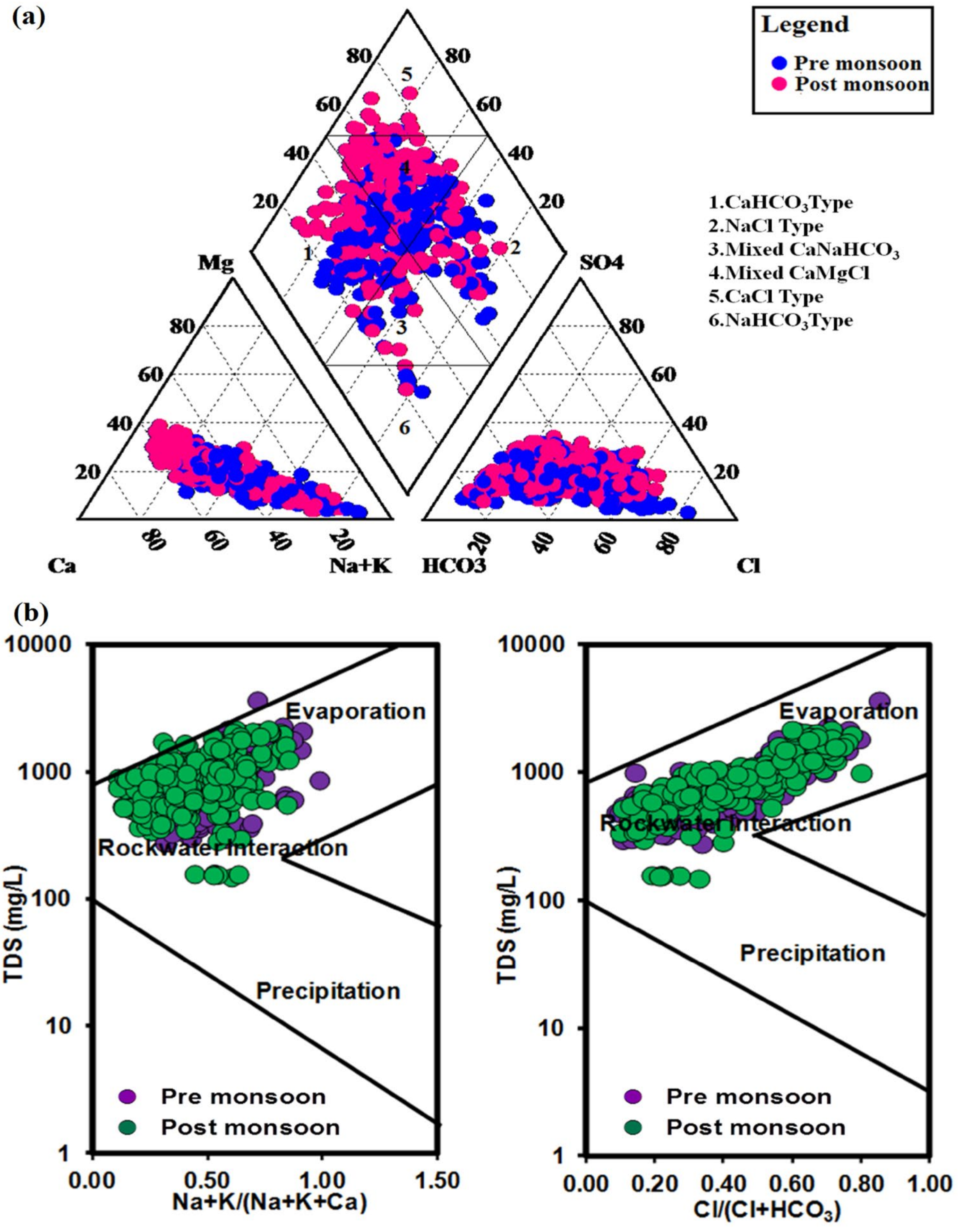


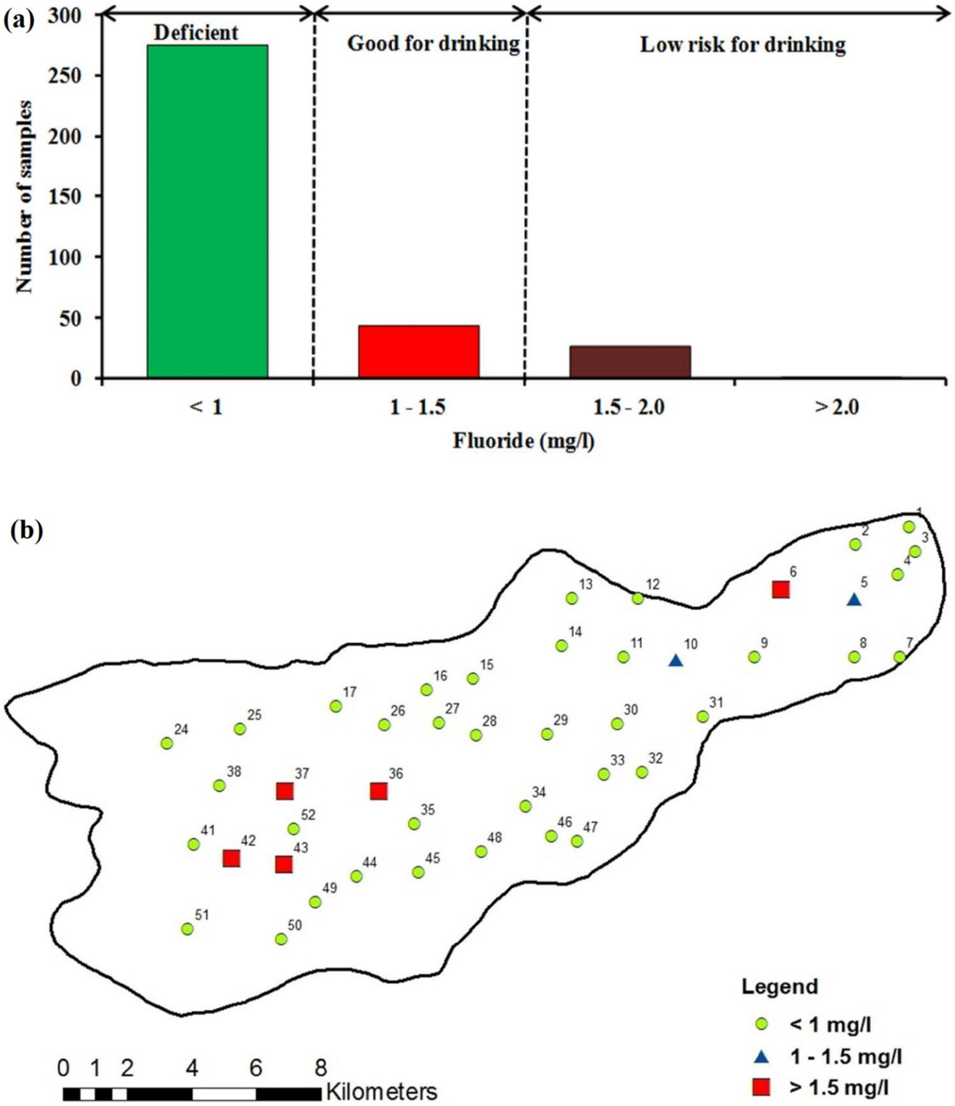
Fig. 6 **a** Major hydrochemical facies of groundwater, **b** major hydrological processes controlled the chemistry of groundwater

four sampling times in a year, the groundwater is considered suitable for irrigation. The irrigation water quality map prepared using this method is shown in Fig. 5. Based on the integration of all the irrigation water quality parameters, it was found that 93% of the study area has groundwater that can be utilised for irrigational purposes.

Hydrogeochemical facies

The pH of the groundwater was ranging between 6.6 and 9.5, which shows that it is slightly acidic to alkaline. EC in groundwater ranges from 232 to 3920 µS/cm with average of 1499 µS/cm (Table 1). Major ions of cations and anions in groundwater were in the order of sodium, calcium, magnesium and potassium, and bicarbonate, chloride and sulphate, respectively. Most of the groundwater samples exhibited mixed Ca–Mg–Cl, Ca–HCO$_3$ type and Na–Cl type (Fig. 6a). Key processes contributing to groundwater geochemistry were identified using Gibbs plot (Gibbs 1970). Figure 6b shows that rock water interaction and evaportaion are the domiant processes controlling the chemical composition of the groundwater.

Fig. 7 **a** Fluoride concentration in groundwater in different range, **b** average fluoride concentration in groundwater



Sources of fluoride

Figure 7a indicates that about 10% of wells exceeded the permissible limit of 1.5 mg/l, 18% of groundwater wells were within the permissible range of 1–1.5 mg/l which are good for drinking purpose and 72% of groundwater wells were below the permissible limit of (< 1 mg/l) in the study area. The average concentration of fluoride in the groundwater at different locations is shown in Fig. 7b. The percentage of samples exceeding fluoride concentration during this study is given in Table 3. The fluoride concentration exceeds 1.5 mg/l in many wells during October and January months i.e. post-monsoon. After monsoon, the concentration of ions increases in specific regions due to dissolution and leaching of minerals by the groundwater.

The sampling wells were differentiated based on the geology in which they were located. Groundwater samples collected from wells located in the fissile hornblende biotite gneiss had higher fluoride concentration in comparison with the wells located in the charnockite. Table 5 shows the range of fluoride concentration in the samples located in the two different geology. Fluoride concentration from wells in the fissile hornblende biotite gneissic rock ranges between 0.1 and 2.7 with average of 0.6 mg/l. The concentration of fluoride from samples located in charnockite areas range from 0.1 to 1.3 mg/l with an average of 0.35 mg/l. The key source of fluoride is mainly from the biotite, hornblende and amphibole minerals from the granite, gneissic and charnockite rocks of this area. The mobilization of these minerals by hydrogeological processes had led to high fluoride in groundwater. Jagadeshan et al. (2015b) reported maximum fluoride concentration in charnockite and epidote hornblende biotite gneiss as 68 mg/kg and 97 mg/l in a nearby region. The interaction

Table 5 Fluoride concentration in groundwater samples located in different rock types (10 times)

Sample no	Rock type	Minimum	Maximum	Average	Sample no	Rock type	Minimum	Maximum	Average
1	Fissile hornblende biotite gneiss	0.1	1.1	0.41	29	Fissile hornblende biotite gneiss	0.1	0.6	0.28
2	Fissile hornblende biotite gneiss	0.1	0.8	0.39	30	Fissile hornblende biotite gneiss	0.1	0.8	0.42
3	Fissile hornblende biotite gneiss	0.2	1	0.53	31	Charnockite	0.1	0.5	0.32
4	Fissile hornblende biotite gneiss	0.1	0.4	0.22	32	Fissile hornblende biotite gneiss	0.1	1	0.51
5	Fissile hornblende biotite gneiss	0.4	1.6	1.01	33	Fissile hornblende biotite gneiss	0.1	0.5	0.24
6	Fissile hornblende biotite gneiss	1.3	1.9	1.51	34	Fissile hornblende biotite gneiss	0.1	0.8	0.39
7	Charnockite	0.1	0.5	0.28	35	Fissile hornblende biotite gneiss	0.4	1.3	0.83
8	Fissile hornblende biotite gneiss	0.1	0.6	0.36	36	Fissile hornblende biotite gneiss	1.2	1.9	1.52
9	Fissile hornblende biotite gneiss	0.1	0.4	0.22	37	Fissile hornblende biotite gneiss	1.1	1.8	1.51
10	Fissile hornblende biotite gneiss	0.5	2.7	1.05	38	Fissile hornblende biotite gneiss	0.1	1.3	0.5
11	Fissile hornblende biotite gneiss	0.1	0.4	0.2	41	Charnockite	0.2	1.3	0.47
12	Fissile hornblende biotite gneiss	0.2	1.1	0.5	42	Fissile hornblende biotite gneiss	1	2	1.52
13	Fissile hornblende biotite gneiss	0.1	0.7	0.31	43	Fissile hornblende biotite gneiss	1	2	1.53
14	Charnockite	0.1	0.6	0.3	44	Fissile hornblende biotite gneiss	0.1	0.6	0.25
15	Charnockite	0.1	0.5	0.3	45	Fissile hornblende biotite gneiss	0.1	1	0.39
16	Fissile hornblende biotite gneiss	0.1	0.8	0.27	46	Fissile hornblende biotite gneiss	0.1	0.7	0.27
17	Fissile hornblende biotite gneiss	0.1	0.6	0.27	47	Fissile hornblende biotite gneiss	0.1	0.6	0.32
24	Charnockite	0.1	1.1	0.28	48	Fissile hornblende biotite gneiss	0.2	0.8	0.39
25	Charnockite	0.1	0.6	0.32	49	Charnockite	0.3	1.3	0.65
26	Fissile hornblende biotite gneiss	0.1	0.9	0.49	50	Charnockite	0.1	0.7	0.35
27	Fissile hornblende biotite gneiss	0.3	1.3	0.86	51	Fissile hornblende biotite gneiss	0.2	1	0.46
28	Charnockite	0.1	0.8	0.3	52	Fissile hornblende biotite gneiss	0.2	1.1	0.6

between groundwater and these fluoride bearing minerals over longer time period lead to the leaching and dissolution of these minerals in groundwater. About 2.9 and 1.1% of fluoride is present in hornblende and biotite minerals respectively (Jagadeshan et al. 2015a). The fissile hornblende gneissic rock is comparatively more weathered than the charnockites (Anbarasu et al. 2019) and hence the concentration of fluoride is higher in the former regions. Equations governing the weathering of these fluoride bearing minerals are given below

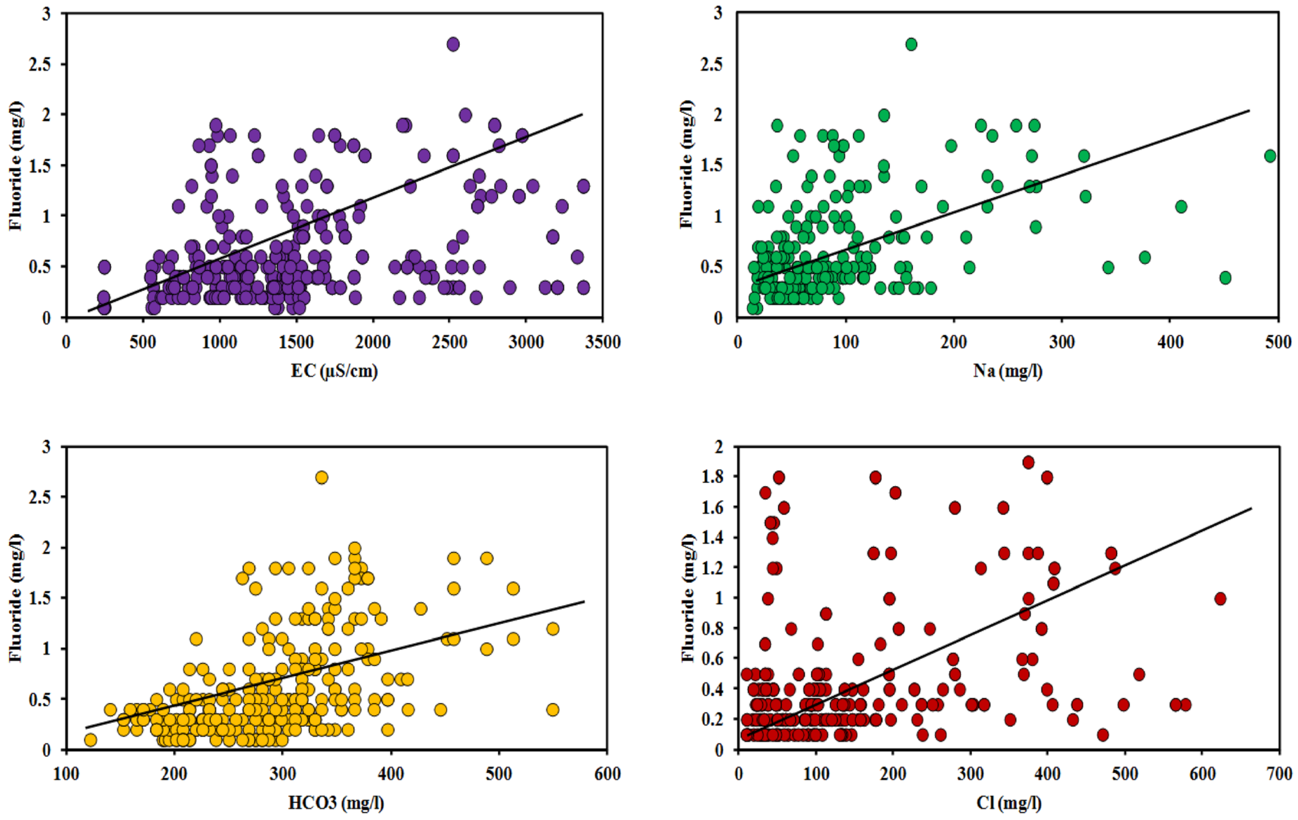


Fig. 8 The correlation between fluoride, EC and major ions shows that positive

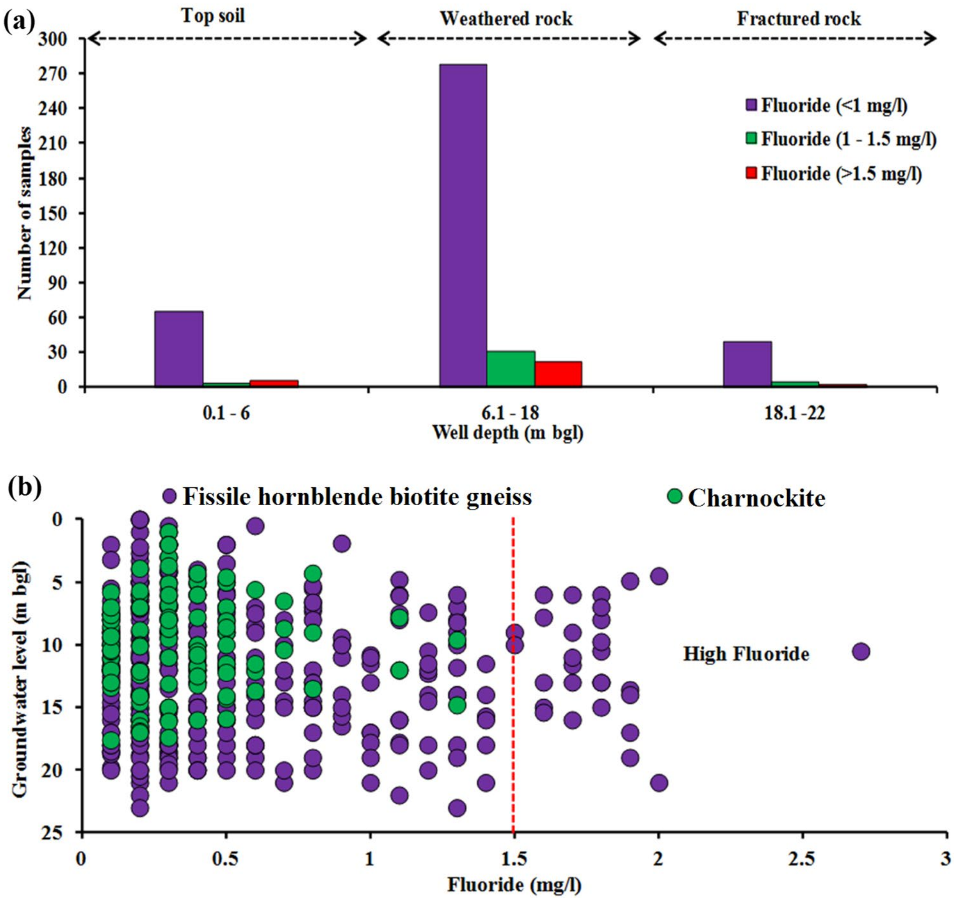
$$\begin{aligned}&CaNa_2(Mg)5\left(Si_8O_{22}\right)F_2(Hornblende) + 2OH^- \\ &\quad = Na(CaNa)\ Mg_5\left(Si_8O_{22}\right)(OH^-)_2 + 2F^-, \end{aligned} \tag{10}$$

$$\begin{aligned}&K(Mg)_3\left(AlSi_3O_{10}\right)F_2(Biotite) + 2OH^- \\ &\quad = KMg_3\left(Al_2Si_3O_{10}\right)(OH^-)_2 + 2F^-. \end{aligned} \tag{11}$$

The correlation between fluoride, EC and major ions is used to understand the source i.e. the weathering of rocks (Fig. 8). In general, fluoride shows a positive correlation with EC, sodium and bicarbonate. The relationship between the concentration of fluoride and well depth is shown in the Fig. 9a. When the groundwater level in wells was in top soil (0.1–6 m bgl) and fractured rock (18.1–22 m bgl), the concentration of fluoride in most of the wells was less than 1 mg/l. When the groundwater level in wells was in the weathered zone (6.1–18 m bgl), the concentration of fluoride was more than 1.5 mg/l in most of the wells. As the leaching of weathered gneissic rock is contributing more fluoride to groundwater, the wells in this area have high concentration of fluoride in groundwater. Comparison of the fluoride concentration with the groundwater level in different rock types is shown in Fig. 9b. This figure also confirms that groundwater in the fissile hornblende gneissic areas have high fluoride. Also, the fluoride concentration, irrespective of shallow or deep groundwater table was low in the charnockite areas. However, in fissile hornblende gneiss such differentiation could not be made as both shallow and deep water levels showed fluoride concentrations at all ranges. A clear understanding of the relationship between fluoride concentration and groundwater level could not be made from Fig. 9b as they were collectively plotted. The concentration of fluoride in groundwater varies temporally in the different rock types. It is evident that the groundwater fluctuation depth plays an important role in determining the fluoride concentration (Brindha et al. 2011, 2016; Fantong et al. 2009; Jagadeshan et al. 2015b). Hence, all the sampling wells were individually plotted to identify the relationship.

The temporal relationship between fluoride and groundwater level is classified as type I wells and type II wells. The shallow groundwater table increases during the rainy season, decreasing the fluoride concentration in groundwater due to dilution. During summer, fluoride concentration increases as the groundwater level declines due to evaporation and no natural recharge. This is the characteristic if

Fig. 9 Variation in fluoride concentration in groundwater **a** well depth, **b** groundwater level



type I wells (Fig. 10a). In type II wells, during the rainy season, infiltration of rainwater flushes the precipitated fluoride salts from the unsaturated zone and hence increase in groundwater levels also increases the fluoride concentration. With prolonged infiltration, the fluoride concentration will decrease in the deep wells too (Fig. 10b). This relationship has been reported in several regions with high fluoride concentrations (Brindha et al. 2011, 2016).

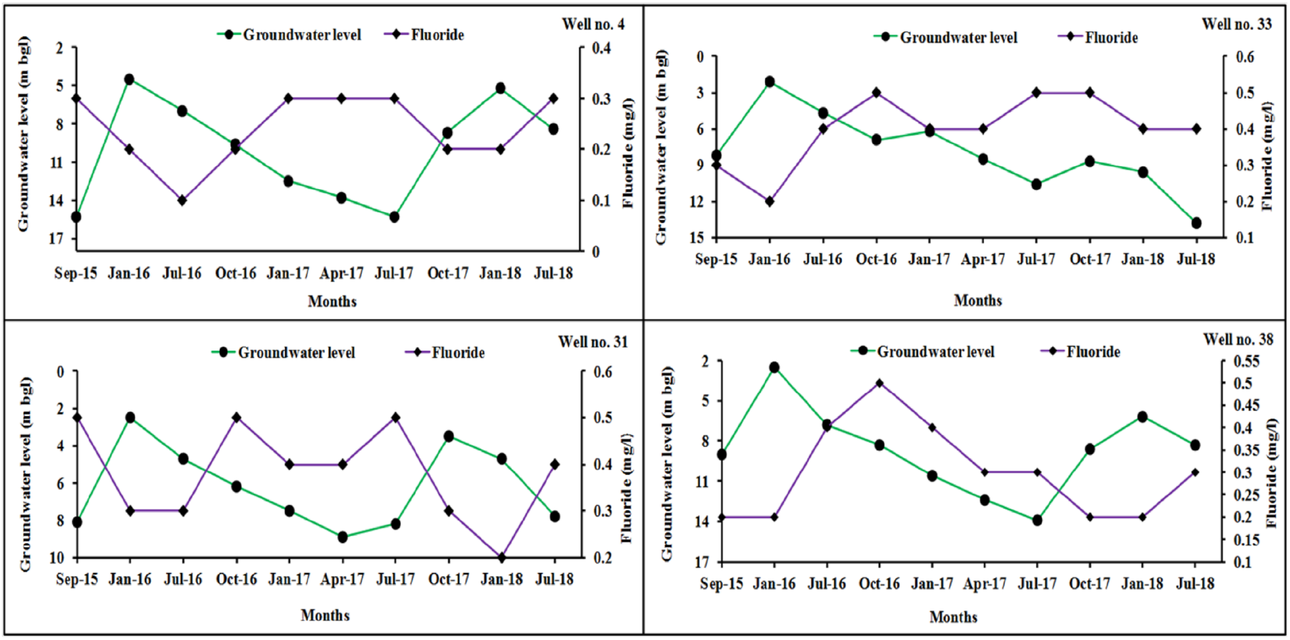
Health risk analysis

Groundwater with fluoride concentration between 1 and 1.5 mg/l as suggested by BIS (2012) is considered to be permissible for drinking use. When fluoride concentration in drinking water was deficient (< 1 mg/l), it can cause dental caries, especially in children (Raju 2017). Fluoride concentration above 1.5 mg/l in drinking water cause dental and skeletal fluorosis (Mandinic et al. 2010; Perumal et al. 2013). Fluoride exposure dose for infants ranges from 0 to 0.1 mg/kg/day, for children it ranges from 0.01 to 0.22 mg/kg/day and for adults, it ranges from 0 to 0.14 mg/kg/day. The results show that fluoride exposure dose was minimum risk for infants and adults but maximum risk for children (Fig. 11). The minimal risk level from oral ingestion based on chronic-duration studies is given as 0.05 mg fluoride/kg/day (ATSDR 2003). About 9% of the groundwater samples exceed the minimum risk level for infants, 21% of the samples exceeded for children and 13% of the samples exceed for adults. The fluoride exposure dose calculated for infants, children and adults shows that children have a high exposure dose than infants and adults.

Conclusion

The present study focused on the fluoride contamination, its sources and the associated human health risk due to the ingestion of contaminated groundwater. Mixed Ca–Mg–Cl, Ca–HCO_3, Na–Cl and mixed Ca–Na–HCO_3 types were the dominant groundwater facies. Groundwater is generally slightly acidic to alkaline nature. The suitability of groundwater for drinking purpose was estimated by TDS, TH, major and minor ions, and 84% of the study area wells

(a) Type I wells



(b) Type II wells

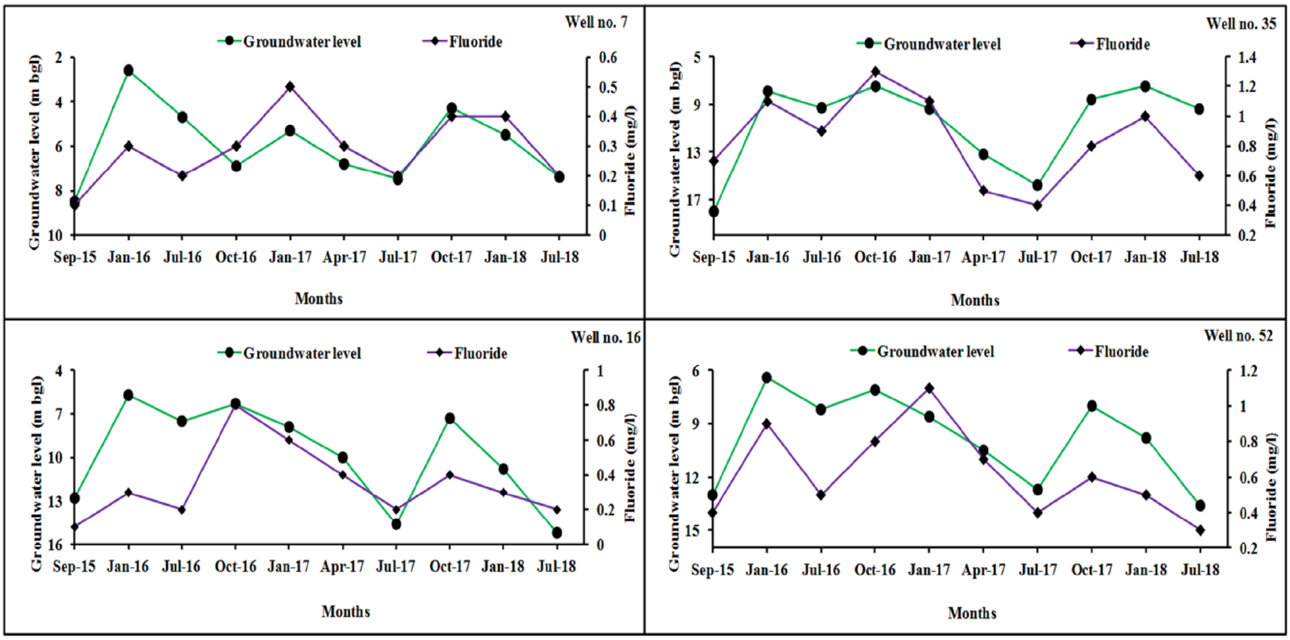


Fig. 10 The temporal variation of groundwater level and fluoride **a** type I wells, **b** type II wells

were good for drinking purpose. Except a few of well, about 93% of the groundwater wells were good for irrigation purpose based on integrating of Na%, SAR, RSC, KR, MH and PI values. 10% of groundwater samples were exceeding the permissible limit of fluoride and 31% of groundwater samples were within the permissible limit for drinking purposes based on BIS and WHO. Fluoride concentration was released more to groundwater due to recharge of rainwater and weathering processes during the monsoon and post-monsoon. Fluoride was showing positively good relation with EC, sodium and bicarbonate ion. Weathering of minerals, dissolution, evaporation and dilution were the major hydrogeochemical processes controlling high fluoride in groundwater. High fluoride

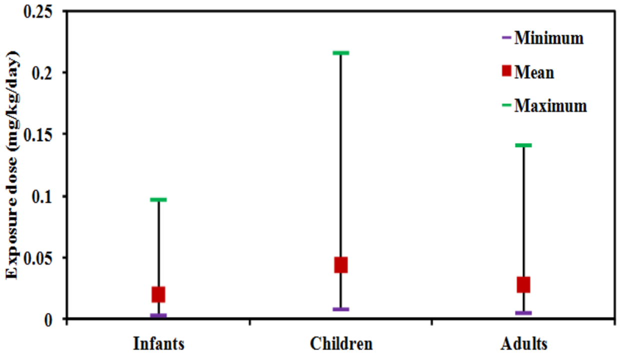


Fig. 11 Fluoride exposure dose for infants, children and adults

concentration occurred mostly in regions with weathered fissile hornblende biotite gneissic rocks than in charnockite rocks. The health risk assessment indicates that children are affected more than infants and adults through intake of high fluoride groundwater. Suitable management of water resources especially to overcome fluoride contamination is required in this area.

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Declarations

Conflict of interest All authors declare that they have no conflicts of interest.

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