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RESEARCH ARTICLE

Judging the sources of inferior groundwater quality and health risk problems through intake of groundwater nitrate and fluoride from a rural part of Telangana, India

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Abstract

Evaluation of groundwater quality and related health hazards is a prerequisite for taking preventive measures. The rural region of Telangana, India, has been selected for the present study to assess the sources and origins of inferior groundwater quality and to understand the human health risk zones for adults and children due to the consumption of nitrate (NO_3^-)- and fluoride (F^-)-contaminated groundwater for drinking purposes. Groundwater samples collected from the study region were determined for various chemical parameters. Groundwater quality was dominated by Na^+ and HCO_3^- ions. Piper's diagram and bivariate plots indicated the carbonate water type and silicate weathering as a main factor and man-made contamination as a secondary factor controlling groundwater chemistry; hence, the groundwater quality in the study region is low. According to the Groundwater Quality Index (GQI) classification, 48.3% and 51.7% of the total study region are excellent (GQI: < 50) and good (GQI: 50 to 100) water quality types, respectively, for drinking purposes. However, NO_3^- ranged from 0.04 to 585 mg/L, exceeding the drinking water quality limit of 45 mg/L in 34% of the groundwater samples due to the effects of nitrogen fertilizers. This was supported by the relationship of NO_3^- with TDS, Na^+, and Cl^-. The F^- content was from 0.22 to 5.41 mg/L, which exceeds the standard drinking water quality limit of 1.5 mg/L in 25% of the groundwater samples. The relationship of F^- with pH, Ca^{2+}, Na^+, and HCO_3^- supports the weathering and dissolution of fluoride-rich minerals for high F^- content in groundwater. They were further supported by a principal component analysis. The Health Risk Index (HRI) values ranged from 0.20 to 20.10 and 0.36 to 30.90 with a mean of 2.82 and 4.34 for adults and children, respectively. The mean intensity of HRI (> 1.0) was 1.37 times higher in children (5.70) than in adults (4.16) due to the differences in weight size and exposure time. With an acceptable limit of more than 1.0, the study divided the region into Northern Safe Health Zone (33.3% for adults and 28.1% for children) and Southern Unsafe Health Zone (66.7% for adults and 71.9% for children) based on the intensity of agricultural activity. Therefore, effective strategic measures such as safe drinking water, denitrification, defluoridation, rainwater harvesting techniques, sanitary facilities, and chemical fertilizer restrictions are recommended to improve human health and protect groundwater resources.

Keywords Geochemistry · Groundwater Quality Index · Health Risk Index · Rural region · India

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Introduction

Groundwater is an important resource, especially for drinking and irrigation purposes (Reddy and Sakram 2014; Subba Rao et al. 2021a). More than 85% of the rural population depends on groundwater resources for their daily needs (Kulkarni et al. 2015). Although about 60 to 85% of groundwater in India is used for drinking and agriculture purposes (Sishodia et al. 2016; Sakram and Narsimha 2018), much of the groundwater is contaminated by natural processes and artificial activities (Alaya et al. 2014; Nadiri et al. 2018a, b; Keesari et al. 2020, 2021a, b). Natural contamination occurs under the influence of toxic components in soils as well as in rocks (example: fluoride-rich minerals), but artificial agents (example: poor drainage conditions, spillage of septic tanks, irrigation-return-flows, immense usage of agrochemicals) can damage groundwater quality and, consequently, health problems (Sakram et al. 2019; Subba Rao and Chaudhary 2019; Wu et al. 2020a).

In recent years, the researchers have focused their studies mainly on chemical quality of groundwater and related health risk issues, most notably nitrate (NO_3^-) and fluoride (F^-) ions being the most common toxins in groundwater (Qasemi et al. 2018; Deepali et al. 2021; Keesari et al. 2021a). Geogenic processes include mineral weathering, dissolution, ion exchange, and evaporation. These have a significant impact on groundwater chemistry (Subba Rao et al. 2017; Badana et al. 2018; Cao et al. 2018; Rajmohan 2020; Nawale et al. 2021). The anthropogenic activities include wastewater discharge and intensive agricultural practices with chemical fertilizers and irrigation-return-flows. These can mainly modify the existing natural groundwater quality (Lapworth et al. 2017; Silva et al. 2017; Shankaraiah et al. 2021). Therefore, the study of the assessment of dissolved ions in the groundwater system can explain the sources and origins of geogenic and anthropogenic activities present in an area that contribute to groundwater contamination. This poor quality of groundwater not only harms human health but also reduces agricultural production (Alaya et al. 2014; Subba Rao 2018; He et al. 2019; Li and Wu 2019; Aravinthasamy et al. 2020). To evaluate groundwater quality for drinking purposes, the Groundwater Quality Index (GQI) has been widely used in different parts of the world (Abbasniaa et al. 2018; Laxman Kumar et al. 2021; Ramachandran et al. 2021). Piper's trilinear diagram, bivariate diagrams, and principal component analysis have been widely used to assess the sources and origins of inferior groundwater quality (Manikandan, et al. 2020; Wu et al. 2020b; Kadam et al. 2022).

Some research studies have shown that the health risks of NO_3^- pollution and the dramatic increase in NO_3^- content in groundwater are mainly due to the impact of intense agricultural activity on the aquifer system, especially in arid and semi-arid areas, in many areas in the world (Barzegar et al. 2016; Serio et al. 2018; Soldatova et al. 2018; Wang et al. 2018; Barakat et al. 2019; Barakat 2020). In addition, it is the most commonly occurring ion in groundwater due to the irrigation-return-flows, untreated household wastes, sewage and septic tank leaks, nitrogen-rich soils, and animal waste (Li et al. 2017; Shukla and Saxena 2018; Zhang et al. 2018; He and Wu 2019; He et al. 2019; Karunanidhi et al. 2019; Subba Rao et al. 2021a, b, c). With the high solubility of NO_3^- in water and the low retention capacity of NO_3^- through soils, NO_3^- reaches the groundwater body, when it is not used properly by plants and leaches to subsurface soils (Barakat 2020).

In India, it has been observed that about 118 million people drink water with NO_3^- level ranging from 45 to 100 mg/L and more than 108 million people consume water with more than 100 mg/L of NO_3^- (Karunanidhi et al. 2020). It is well known that consumption of contaminated groundwater above 45 mg/L of NO_3^- can lead to *methemoglobinemia* (blue baby syndrome), where red blood cells reduce their ability to handle oxygen. This causes shortness of breath, heart attack, and even death, especially in children (WHO 2012). Sometimes, it leads to cancer also (WHO 2012).

About 75 to 90% of the F^- intake is mainly due to the drinking water consumption (Demelash et al. 2019). Approximately 200 million people suffer from high F^- content (> 1.5 mg/L) in the groundwater globally, especially in countries such as Africa, China, India, Iran, Nigeria, Pakistan, South America, and Sri Lanka (Wu et al. 2015; Craig et al. 2015; Chen et al. 2017; Satyanarayana et al. 2017; Subba Rao et al. 2020a). Granitic rocks are rich minerals with an F^- content of 500 to 1400 mg/k (Sajil Kumar 2017). Fluoride minerals (fluorite, apatite, biotite, and hornblende) occurring in the basement rocks (hornblende-biotite, gneiss, and granite) are the main sources of F^- contamination of groundwater, while agrochemicals (phosphate fertilizers) increase of F^- content as a secondary source in groundwater (Sajil Kumar 2017; Subba Rao 2017b; Deepali et al. 2020; Karunanidhi et al. 2019). High concentrations of F^- (> 1.5 mg/L) causes severe fluorosis (BIS 2012; WHO 2012). It has also been observed that children are more vulnerable to NO_3^- and F^- ions compared to adults (Zhai et al. 2017; Rezaei et al. 2019; Karunanidhi et al. 2020; Nawale et al. 2021). In India, the potential risk of groundwater contamination is a consequence of NO_3^- and F^- ions, where children are at a greater health risk than adults, leading to non-carcinogenic problems in children (Ding et al. 2020; Kaur et al. 2020).

The present study is a rural part of the Vikarabad district, Telangana, India (Fig. 1). Due to the lack of surface water supply in the present study area, local residents rely mainly on groundwater resources for their drinking needs. It involves intensive and long-term practice. Therefore, the effects of unlimited use of chemical composts (nitrate, phosphate, and potassium varieties), irrigation-return-flows, and animal wastes may be the most common phenomena on groundwater system. Furthermore, basic sanitation facilities such as disposal of household waste and leakage of septic tanks are in poor condition in the present study region. These factors have been identified as the most contaminated sources of groundwater, and hence, high levels of NO_3^- and F^- content have been observed from groundwater in the surrounding districts of Telangana (Sujatha and Reddy 2003; Roy et al. 2018; Sakram et al. 2019; Narsimha and Li 2019; Narsimha and Qian 2020, 2021; Shekhar et al. 2021; Subba Rao et al. 2021a, b, c). Keesari et al. (2014) studied radioactive elements in the groundwater of Nalgonda district in Telangana. However, in the present study region (Fig. 1), there is no research to date on the evaluation of sources and origins of degraded groundwater quality and health risk issues due to the use of NO_3^-- and F^--contaminated groundwater for drinking purposes. Therefore, the main focus of the present study is on (a) judging the groundwater quality for drinking purposes, using Groundwater Quality Index (GQI); (b) evolution of groundwater geochemistry, using Piper's trilinear diagram, bivariate diagrams, and principal component analysis; and (c) the assessment of health risk problems caused by NO_3^- and F^- contamination in groundwater, using Health Risk Index (HRI).

Assessing the sources and origins of poor groundwater quality and health hazard problems due to the high concentration of NO and F in drinking water consumption from rural areas can help in taking appropriate management measures to reduce the severity of health problems.

Study region

The present rural region is located in the southwestern part of Telangana, India (Fig. 1). It lies between the north latitudes 17° 23′–17° 25′ and the east longitudes 77° 45′–78° 50′, falling in the Survey of India toposheet numbers 56G/15 and 56G/16, and covering an area of about 633 km^2. The region has a semi-arid climate with an average annual temperature of 14 to 41 °C and an average annual rainfall (5 years) of 937 mm. The surface runoff was caused by the development of sub-dendritic drainage patterns in the study region.

The prominent rock exposures in the study region are basalt and granite (Fig. 2). Laterite patches also occur. Basalts are fine-grained and dark-colored volcanic rocks. They include mainly calcic plagioclase feldspars and clinopyroxene with olivine, quartz, hornblende, nepheline, and orthopyroxene minerals. The granites are generally medium- to coarse-grained. They contain mainly quartz, plagioclase and potassium feldspars, biotite, apatite, and hornblende minerals. Basically, they are hard rocks.

Basically, hard rocks are difficult to transmit and store the groundwater in the subsurface due to a lack of porosity and permeability. However, the occurrence of vesicular structures, cracks, and joints formed by primary and secondary porosities becomes aquifers in basalts, while the presence of weathered and fractured rocks developed by secondary porosity becomes water-bearing formations (aquifers) in granites. As a result, groundwater is transported from one place to another and stored depending on the rock permeability of the rocks. Laterites are porous, but they are slightly permeable to a limited area. Groundwater occurs under water table and also under semi-confined conditions. Groundwater table depth is 18 to 28 m below ground level. Groundwater quality in fieldwork generally appeared to be favorable for drinking. However, in some places, household waste, septic tank spills, irrigation-back-flows, chemical fertilizers, and animal waste seem to be useless for drinking due to the impact of non-geogenic sources on the groundwater system.

Materials and methods

Groundwater samples from 100 wells were collected in 1-L capacity polythene bottles from the present study region in May 2015 (Fig. 2). The containers were cleaned with 1:1 dilute hydrochloric acid and washed three times with distilled before collecting groundwater samples, according to the standard procedure (APHA 2012). Wells were pumped thoroughly before the collection of the samples to prevent stagnation in the wells.

The pH and electrical conductivity (EC) were measured in the field, using the Hanna H-198130 m. TDS was calculated by multiplying EC by a factor of 0.64 (Hem 1991; Subba Rao 2017a). Other chemical parameters (Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, Cl^-, SO_4^{2-}, NO_3^-, and F^-) were estimated, following the standard water quality methods of APHA (2012). The ions Ca^{2+}, Mg^{2+}, HCO_3^-, and Cl^- were determined, using the titration method. Na^+ and K^+ were measured, using a flame photometer (Elico CL-378). The ions SO_4^{2-} and NO_3^- were estimated, using a UV spectrophotometer (Phtolab-6600 WTW). The ion F^- was measured, using an ion-selective electrode (Orion). All ions are expressed in milligrams per liter (mg/L) and milliequivalents per liter (meq/L).

For calculation of chemical ionic balance error (IBE), total concentrations of cations (C) such as Ca^{2+}, Mg^{2+}, Na^+, and K^+ and total concentrations of anions (A) such as HCO_3^-, Cl^-, SO_4^{2-}, NO_3^-, and F^- were used (Eq. 1), which was from 4.15 to 4.85%, reflecting the reliability of the chemical data (Subba Rao 2017a).

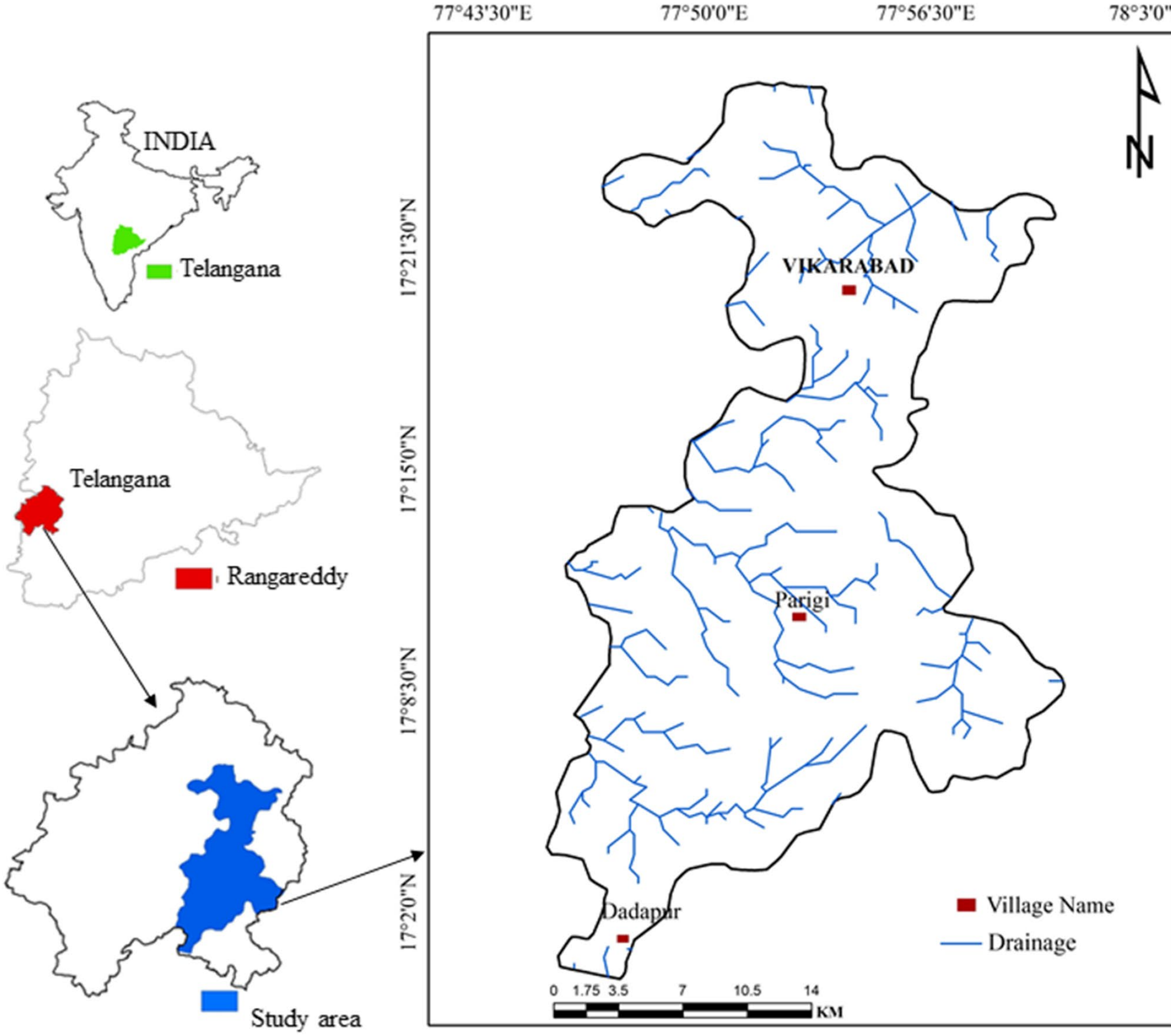

Fig. 1 Map showing the location of rural region of Telangana, India

$$\text{IBE} = \frac{\sum C - \sum A}{\sum C + \sum A} \times 100 \quad (1)$$

Comprehensive tool for utilization of groundwater quality for drinking

The Groundwater Quality Index (GQI) is a comprehensive technique for expressing overall drinking water quality in a single unit (Yidana and Yidana et al. 2010; Venkatramanan et al. 2016; Roy et al. 2018; Subba Rao et al. 2020a, b; Wu et al. 2020a; Ramachandran et al. 2021). For calculation of GQI, five steps were involved. The first step in this index was to assign unit weight (*U*) for each chemical variable (*i*) based on its relative significance on human health. In the second step, the relative weight (*W*) was computed for each chemical variable (Eq. 2). In the third step, the percentage of quality rating scale (*Q*) was calculated by dividing the concentration of chemical parameter (*C*) with its standard drinking water quality (*D*) for every chemical variable (Eq. 3). In the fourth step, the relative rating (*R*) was quantified by multiplying *W* with *Q* in each chemical parameter (Eq. 4). In the final step, the GQI was computed by adding all *R* values in each sample (Eq. 5).

$$W = \frac{U}{\sum_{i=1}^{n} U} \quad (2)$$

$$Q = \frac{C}{D} \times 100 \quad (3)$$

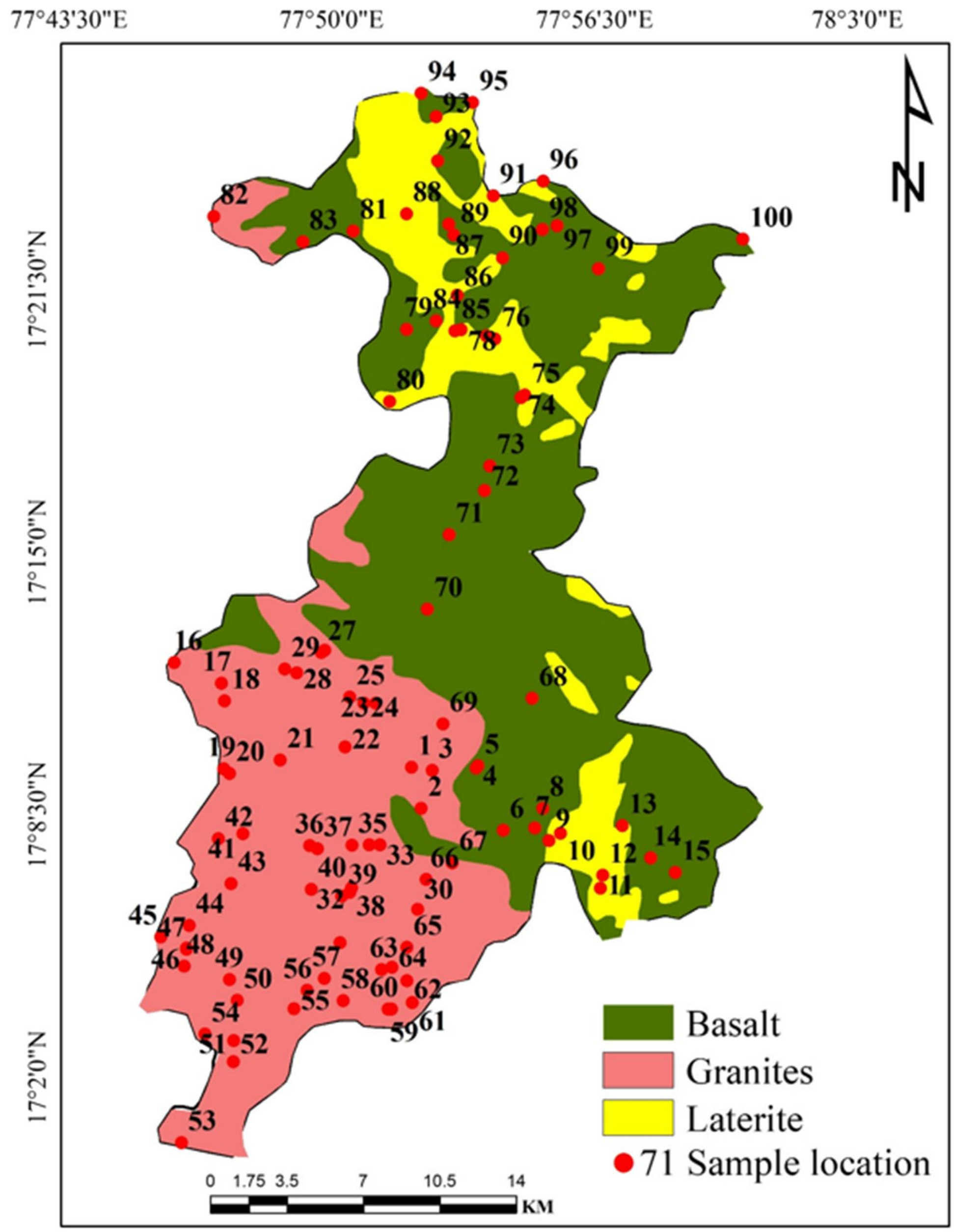

Fig. 2 Map showing the geology with groundwater sampling locations

$$R = W \times Q \tag{4}$$

$$GQI = \sum_{i=1}^{n} R \tag{5}$$

When the *GQI* is less than 50, it indicates an excellent water quality; when it is 50 to 100, it shows good water quality; when it is between 100 and 200, it specifies poor water quality; when it is from 200 and 300, it represents very poor water quality; when it is more than 300, it suggests unsuitable water quality for drinking purposes (Acharya et al. 2019).

Human health risk assessment

The NO_3^- and F^- ions have been selected for human health risk assessment. The oral intake procedure was selected for calculation of Health Risk Index (HRI) for adults and children (Li and Wu 2019; Li et al. 2019; Rezaei et al. 2019; Selvam et al. 2020; Wu et al. 2020b; Nawale et al. 2021; Razzagh et al. 2021). The hazard quotient (*HQ*) and *HRI* were calculated, using Eqs. 6 to 8 (USEPA 1991, 2006).

$$Dd = \frac{Ci \times Ir \times Ed \times Ef}{Bw \times Et} \tag{6}$$

$$HQ = \frac{Dd}{Rd} \quad (7)$$

$$HRI = \sum_{i=1}^{n} HQ \quad (8)$$

where *Dd* is the average daily dose of NO_3^- and F^- (mg/kg/day), *Ci* is the concentration of ions (NO_3^- and F^-) in groundwater (mg/L) and *Ir* is the intake rate (3 L/day and 1.5 L/day for adults and children), *Ed* is the exposure duration (66.4 years for adults and 12 years for children), *Ef* is the exposure frequency (365 days for both adults and children), *Bw* is the average body weight (65 kg for adults and 18.7 kg for children), *Et* is the average exposure time (24,236 days for adults and 4,380 for children), *HQ* is the hazard quotient, *Rd* is the recommended dose for chronic oral exposure (1.60 mg/kg/day for NO_3^- and 0.06 mg/kg/day for F^-) (ICMR 2009; UNDESA 2013; USEPA 2014; Brindha et al. 2016; Kadam et al. 2022), and *HRI* is the Health Risk Index (non-carcinogenic hazard).

The tolerable limit of HRI is 1.0 (USEPA 2014). If it is greater than 1.0, the non-carcinogenic risk of contamination is higher than the tolerable level. If it is less 1.0, the non-carcinogenic risk is within acceptable limit.

Principal component analysis (PCA)

Principal component analysis (PCA) provides a unique solution by reconstructing new results from the original data (Thivya et al. 2014; Subba Rao 2014; Li et al. 2019; Subba Rao et al. 2021b). According to the Kaiser Criterion in this analysis, the principal components (PCs) were extracted with the varimax rotation of loadings for maximum variance and the eigenvalues more than 1. Since TDS expresses the total dissolved concentrations of all ions (Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, Cl^- Cl^-, SO_4^{2-}, NO_3^-, and F^-), PCA was performed taking into account all ions. Therefore, the various combinations of these ions in terms of PCs can provide information about the sources and origins of the geochemical processes as well as the inferior groundwater quality.

Geographical information system (GIS)

The geographical information system (GIS) is a software-based technique for demarcating the spatial distribution of chemical quality of groundwater (Karunanidhi et al. 2020). ArcGIS software 10.7 was used to generate a spatial distribution of chemical parameters as well as a Groundwater Quality Index (GQI), using the inverse distance-weighted interpolation technique (Kadam et al. 2022; Subba Rao et al. 2021b).

Results and discussion

Groundwater characteristics

Groundwater pH ranged from 6.30 to 8.90 with a mean of 7.14 (Table 1), indicating that it is slightly acidic to highly alkaline in nature. Three percent of groundwater samples (17, 26, and 79) exceeded the safe limit of pH (6.5 to 8.5) in drinking water, which can damage mucous membranes (BIS 2012). TDS varied from 56 to 1,024 mg/L with a mean of 291 mg/L. About 15% of groundwater samples (1 to 5, 14, 16, 19, 20, 22, 29, 41, 43, 54, and 55; Fig. 2) were more than the recommended limit of 500 mg/L for drinking purposes, causing gastrointestinal irritation (BIS 2012).

Cations

The Ca^{2+} content was from 8.02 to 152 mg/L being a mean of 49.6 mg/L (Table 1). Fourteen percent of groundwater samples (41, 43, 49, 51, 53 to 55, 63, 64, 66, 68, 70, 90, and 98; Fig. 2) showed an unacceptable limit (75 mg/L) of Ca^{2+} (BIS 2012). Weathering and dissolution of plagioclase feldspars are the major sources of Ca^{2+} in groundwater (Subba Rao et al. 2017; Kadam et al. 2022; Deepali et al. 2021). The Mg^{2+} was between 2.43 and 92.4 mg/L with a mean of 23.5 mg/L, which was more than the tolerable limit of 50 mg/L in 6% of the groundwater samples (3, 55, 91, 94, 97, and 99; Fig. 2). The ion Mg^{2+} is mainly attributed to the dissolution of ferromagnesian minerals (olivine, pyroxene, biotite, etc.) occurring in host rocks, in addition to human-induced activities (Subba Rao 2021). The ion Na^+ was from 3 to 416 mg/L, with a mean of 54.1 mg/L. In 1% of the groundwater samples (43; Fig. 2), the Na^+ content was more than the threshold limit of Na^+ 200 mg/L (BIS 2012). Plagioclase feldspars in basement rocks are a major source and anthropogenic origin (household wastes, irrigation-return-flows, etc.) is another source of Na^+ in groundwater (Subba Rao 2021). The K^+ ranged from 1 to 118 mg/L and its mean was 6.20. It exceeds the desirable limit of 12 mg/L in 10% of groundwater samples (1, 16, 20, 29, 35, 45, 53, 88, 89, and 96; Fig. 2). Orthoclase feldspars are the main source and potassium compost is the secondary source of K^+ in groundwater.

Anions

The concentration of HCO_3^- was from 20.7 to 584 mg/L, with a mean of 147 mg/L (Table 1). This is formed by soil CO_2 due to the releasing from decay of organic decomposition (Subba Rao et al. 2017). The HCO_3^- was higher than the allowable limit of 300 mg/L in 4% of groundwater samples (1 to 29, and 31 to 100; Fig. 2) for drinking purposes

Table 1 Statistical summary of chemical composition of groundwater

Chemical parameters	Minimum	Maximum	Mean	BIS (2012)	
				Drinking water limits	% of samples exceeding limits
pH	6.30	8.90	7.14	6.50–8.50	3
TDS (mg/L)	56.0	1024	291	500	15
Ca^{2+} (mg/L)	8.02	152	49.6	75	14
Mg^{2+} (mg/L)	2.43	92.4	23.5	30	6
Na^{+} (mg/L)	3.00	416	54.1	200	1
K^{+} (mg/L)	1.00	118	6.20	12	10
HCO_3^-(mg/L)	20.7	584	147	300	4
Cl^- (mg/L)	17.7	425	128	250	10
SO_4^{2-}(mg/L)	30	166	97.9	200	-
NO_3^-(mg/L)	0.04	585	56.3	45	34
F^- (mg/L)	0.22	5.41	1.13	1.5	25

(BIS 2012). The concentration of Cl^- was between 17.7 and 425 mg/L with a mean of 128 mg/L. According to drinking water quality standards, the Cl^- was more than 250 mg/L in 10% of the groundwater samples (16, 41, 43, 54, 55, 63, 66, 70, 91, and 99; Fig. 2), causing salty taste and laxative effect. Non-lithological sources (domestic waste water, irrigation-return-flows, etc.) are the major contributors of Cl^- to groundwater body (Sarath Prasanth et al. 2012; Laxman et al. 2019). The SO_4^{2-} value was from 30 to 166 mg/L and its mean was 97.9 mg/L. It did not exceed its acceptable limit of 200 mg/L in all groundwater samples. There is no trace of sulfide-bearing minerals in country rocks. Since the present study belongs to the agricultural region, the application of gypsum appears to be a source of SO_4^{2-} in groundwater body, which can be used to increase soil permeability (Shankaraiah et al. 2021).

The NO_3^- ranged from 0.04 to 585 mg/L with a mean of 56.3 mg/L. In 34% of groundwater samples (2 to 6, 8, 9 to 11, 13, 14, 16, 18, 20, 22, 24, 25, 30, 33, 41 to 44, 46, 47, 49, 51 to 55, 63, 66, and 67; Fig. 2), it was more than the desirable limit of 45 mg/L, leading to blue baby disease (BIS 2012). The NO_3^- is formed by the biochemical transfer of urea or ammonium as biochemical through the impact of sewage waste, septic tank leakage, agricultural fertilizer, and animal waste on the aquifer system (Deepali et al. 2015; Zhang et al. 2018; He et al. 2019; Kadam et al. 2022; Singh and Craswell 2021). In the present study region, the F^- content was from 0.22 to 5.41 mg/L, with a mean of 1.13 mg/L. It exceeded 1.5 mg/L in 25% of groundwater samples (3, 7, 19 to 25, 28, 34, 36, 38, 39, 40, 42, 43, 50, 56, 59, 61, 64, and 72 to 74; Fig. 2), causing fluorosis. Fluoride-containing minerals such as fluorite, biotite, and hornblende are found in host rocks and the use of phosphate compost in agricultural areas is the major source of F^- in groundwater (Subba Rao et al. 2016, 2020a).

Ionic dominance

Ionic dominance is widely used to explain the diagnostic chemical aspect of groundwater solutions occurring in hydrologic systems (Wagh et al. 2019). It reflects the reaction of the chemical processes operating in the lithologic framework and also reflects the pattern of water flow and thus the change in groundwater quality (Manikandan et al. 2020). As mentioned above, the ionic dominant pattern is in the order of $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ and $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^- > F^-$ for cations and anions, respectively. The predominance of Na^+ and HCO_3^- ions between cations and anions, respectively, indicates the present state of the rock weathering and dissolution processes (Subba Rao et al. 2020a, b). However, the further dominance of Cl^-, SO_4^{2-}, and NO_3^- among anions obviously supports the influence of anthropogenic sources (household wastewater, septic tank spills, irrigation-rerun-flows, chemical fertilizers, etc.) on the aquifer system (Badeenezhad et al. 2020, 2021; Deepali et al. 2021; Kadam et al. 2022).

Groundwater quality assessment for drinking purpose

The Groundwater Quality Index (GQI) is a scale used to measure overall drinking water quality (Subba Rao et al. 2020a). The computed values of GQI ranged from 30 to 91 (Table 2). According to GQI's classification, 51% (30 to 50 with a mean of 40.2) and 49% (51 to 91 with a mean of 64.8) of groundwater samples come under excellent (GQI: < 50) and good (GQI: 50 to 100) water quality types, respectively, for drinking purposes. These water quality types covered 48.3% and 51.7% of the study area, respectively (Fig. 3). This indicates that the quality of groundwater is suited for drinking water needs without water purification. However,

Table 2 Classification of Groundwater Quality Index (GQI) for drinking purpose

Water quality	Range	Minimum	Maximum	Mean	Samples (%)
Excellent	< 50	30	50	40.2	51
Good	50 to 100	51	91	64.8	49
Poor	100 to 200	-	-	-	-
Very poor	200 to 300	-	-	-	-
Unsuitable	> 300	-	-	-	-

as mentioned earlier, the NO_3^- and F^- contents exceeded the drinking water quality limits of 45 and 1.5 mg/L in 34% and 25% of groundwater samples, respectively. Therefore, changing the quality of groundwater from excellent to good can occur due to the influence of anthropogenic sources on the geogenic origin.

Sources and origins of inferior groundwater quality

The trilinear diagram of Piper's has been widely used to identify the geochemical evolution of groundwater quality in terms of ion dominance (Piper 1944; Deepali et al. 2021). From the diagram (Fig. 4), 80%, 14%, 2%, 2%, 1%, and 1% of the total groundwater samples represent the carbonate water type (Ca^{2+}-HCO_3^-), mixed water type (Ca^{2+}-Mg^{2+}-Cl^-), non-carbonate water type (Ca^{2+}-Cl^-), non-alkali water type (Na^+-Cl^-), excess water type (Na^+-HCO_3^-), and mixed water type (Ca^{2+}-Mg^{2+}-HCO_3^-), respectively. The dominance of carbonate water type is significantly higher of alkaline earths (Ca^{2+} and Mg^{2+}) and weak acids (HCO_3^-) than of alkalis (Na^+ and K^+) and strong acids (Cl^- and SO_4^{2-}), indicating water–rock interactions (Badana et al. 2018; Kadam et al. 2022). The type of mixed water dominated by Ca^{2+}-Mg^{2+}-Cl^- ions indicates the movement of carbonate water type towards mixed water type due to the influence of anthropogenic sources (Nawale et al. 2021). The non-carbonate water type (Ca^{2+}-Cl^-) and the non-alkali water type (Na^+-Cl^-) clearly specify the domination of water–rock interactions over which the mad-made activities take place in aquifer system (Deepali et al. 2021). Excess water type (Na^+-HCO_3^-) and mixed water type (Ca^{2+}-Mg^{2+}-HCO_3^-) indicate that rock weathering and dissolution processes are controlled by groundwater chemistry (Deepali et al. 2015). Furthermore, the types of groundwater in the decreasing order are carbonate water > mixed water > non-carbonate water > non-alkali water > excess water.

The present study region is mainly underlain by granite and basalt rocks. The interaction of recharged water with soils and/or rocks (before reaching the groundwater body) releases Ca^{2+}, Mg^{2+}, Na^+, K^+, and HCO_3^- ions (Eqs. 9 to 12) through the rock weathering and dissolution processes (Subba Rao et al. 2012; Thivya et al. 2014; Nawale et al. 2021).

Calcium feldspar

$$CaAl_2Si_2O_8 + H_2O + 2H^+ \rightarrow Al_2Si_2O_5(OH)_4 + Ca^{2+} \quad (9)$$

Sodium feldspar

$$2NaAlSi_3O_8 + 2H^+ + 9H_2O \rightarrow Al_2Si_2O_5(OH)_4 + 4H_4SiO_4 + 2Na^+ \quad (10)$$

Potassium feldspar

$$2KAISi_3O_8 + 2H^+ + 9H_2O \rightarrow H_4Al_2Si_2O_9 + 4H_4SiO_4 + 2K^+ \quad (11)$$

Olivine

$$4Mg_2SiO_4 + 4H_2O + SiO_2 \rightarrow 2Mg_3Si_2O_5(OH)_4 + 2Mg^{2+} \quad (12)$$

Generally, the diagrams, Ca^{2+}/Na^+ vs HCO_3^-/Na^+, and Ca^{2+}/Na^+ vs Mg^{2+}/Na^+, are used to assess the chemistry of groundwater (Fig. 5), whether it is governed by evaporate dissolution or silicate weathering or carbonate dissolution (Manikandan et al. 2020; Subba Rao et al. 2021c). From Fig. 5, groundwater sample data were appeared to have been moved from the domain of evaporate dissolution towards the domain of carbonate dissolution via the domain of silicate weathering. Much of the groundwater sampling data falls within the domain of silicate weathering, where the ionic chemistry of groundwater is primarily controlled by the silicate weathering, and partly by the evaporate dissolution due to the soil CO_2 effect and the carbonate dissolution by the impact of gypsum applied for alteration of soil permeability (Subba Rao et al. 2021c). Broadly, these geochemical processes cause variation in groundwater quality.

Since agriculture is one of the major practices in the present study region, a significant portion of the applied agrochemicals (nitrogen fertilizers) is likely to penetrate into the soils/rocks, in addition to the impact of household wastewater, septic tank leakage, and animal excreta, and reach the aquifer by recharge water (Subba Rao et al. 2012; Manikandan et al. 2020). This is likely to increase NO_3^- levels in groundwater. However, NO_3^- levels of less than 45 mg/L were observed in 55.4% of the total study region (Fig. 6), which was mainly confined to the northern part and partly to the southern part as isolated pockets, where agricultural activity was minimal. The next high NO_3^- levels (45 to 90 mg/L, 90 to 135 mg/L, 135 to 180 mg/L, and > 180 mg/L) were found mainly in the southern part (44.6%), where agricultural activities were more. Therefore, the leaching effect of nitrogen fertilizer was assessed as the main source. Table 3 shows the concentration of NO_3^- (0.1 to 897 mg/L with a mean of 27.5 to 117 mg/L) in groundwater near the present study region. This ionic content significantly supports similar sources

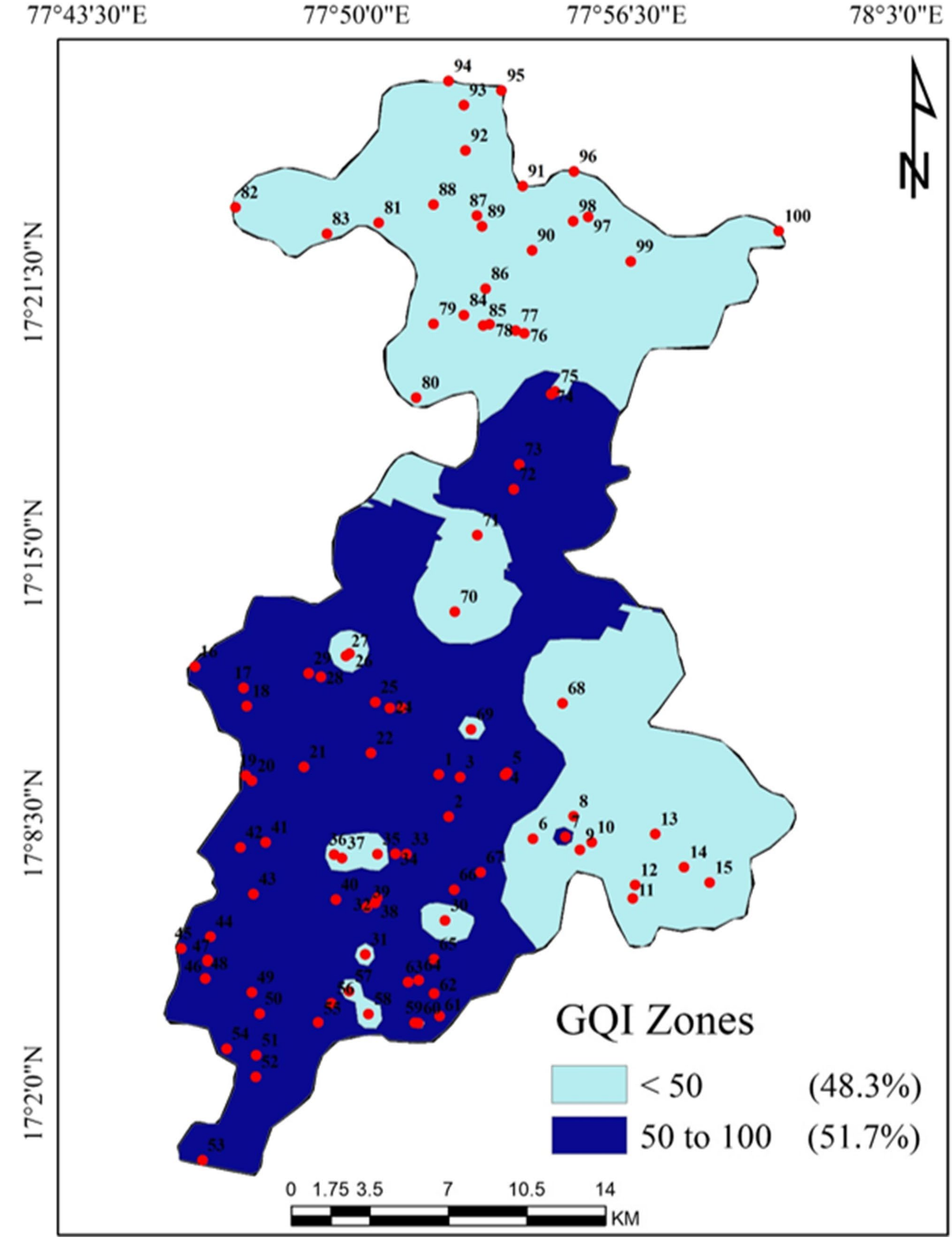

Fig. 3 Spatial distribution of Groundwater Quality Index (GQI) for drinking purpose

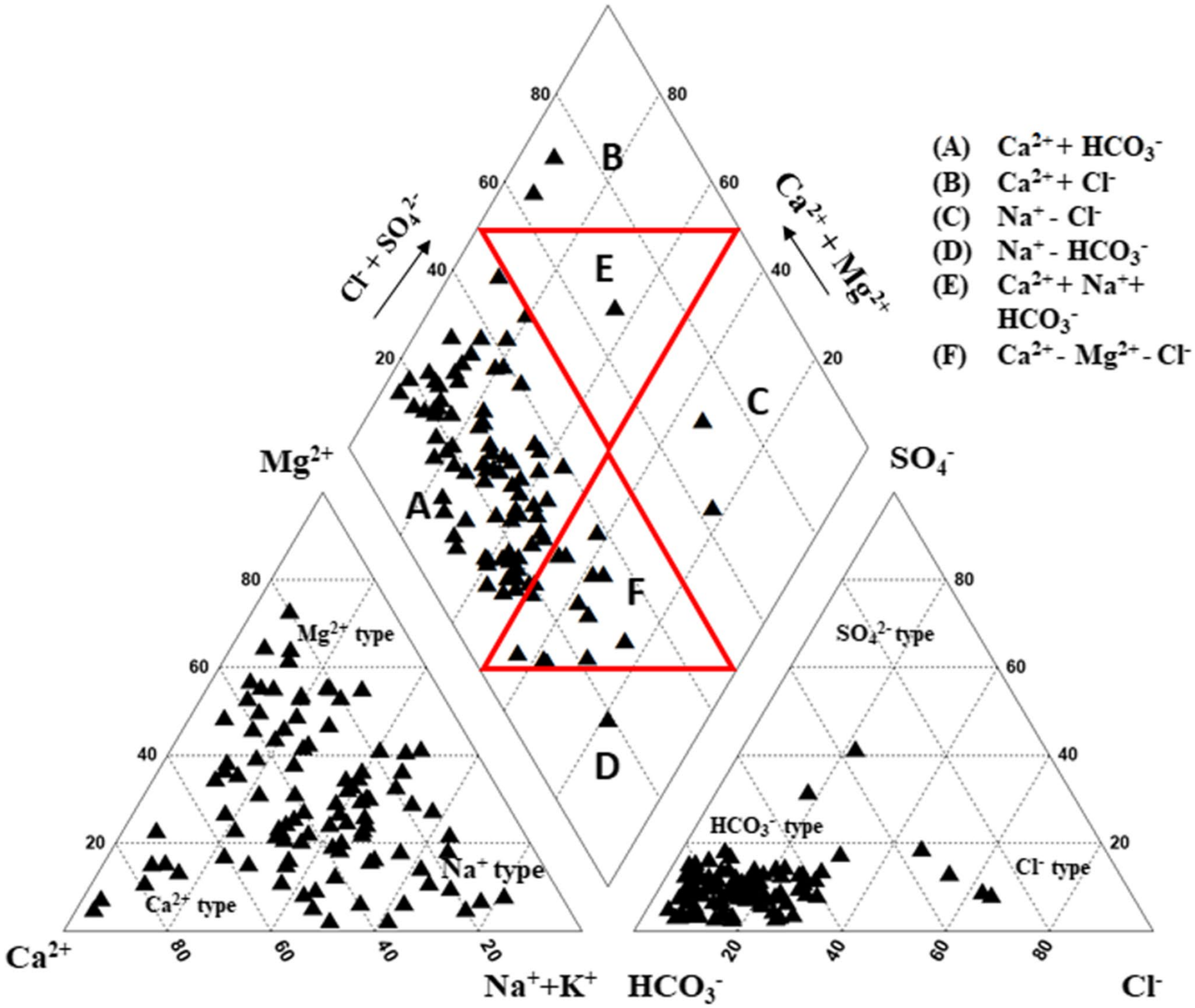

Fig. 4 Geochemical evolution of groundwater types (after Piper 1944)

for the occurrence of NO_3^- content (0.04 to 585 mg/L with a mean of 56.3 mg/L) in groundwater of the present study region. In addition, the effects of irrigation-return-flows and animal wastes are also secondary sources on the aquifer system in the agricultural region. Therefore, isolated patches of high concentration of NO_3^- may be the result of combined effect of both primary and secondary sources on groundwater. This hypothesis further supports an increase in NO_3^- (9.38 to 186 mg/L) with TDS (212 to 480 mg/L), Na^+ (39.8 to 89.2 mg/L), and Cl^- (113 to 179 mg/L; Table 4). In Fig. 7, a significant positive correlation coefficient (r) of NO_3^- with TDS ($r = 0.74$), Na^+ ($r = 0.55$), and Cl^- ($r = 0.51$) has been shown to further support human-induced contamination, as also reported in other regions by Manikandan et al. (2020), Deepali et al. (2021), and Kadam et al. (2022).

The higher alkalinity (pH and HCO_3^-) with Na^+ activates the leaching of fluoride minerals present in basement rocks and thereby increases the high F^- content in the groundwater system (Subba Rao et al. 2016, 2020a). Apart from this, the use of phosphate fertilizers can lead to an increase in the concentration of F^- in groundwater (Subba Rao et al. 2021a). The present study showed that spatial distribution of concentration of F^- less than 0.6 mg/L in groundwater was observed mainly from the northern part and partly from the southern part as isolated patches, which covers 20.4% of the total study region (Fig. 8). A safe limit of F^- (0.6 to 1.5 mg/L) was found in the central part (58.8%) of the study region. The next high F^- content (1.5 to 3.0 mg/L, 3.0 to 4.5 mg/L, and > 4.5 mg/L) was found to be isolated pockets (20.8%) from the total study region irrespective of agricultural activity. This means that the source of F^- content in

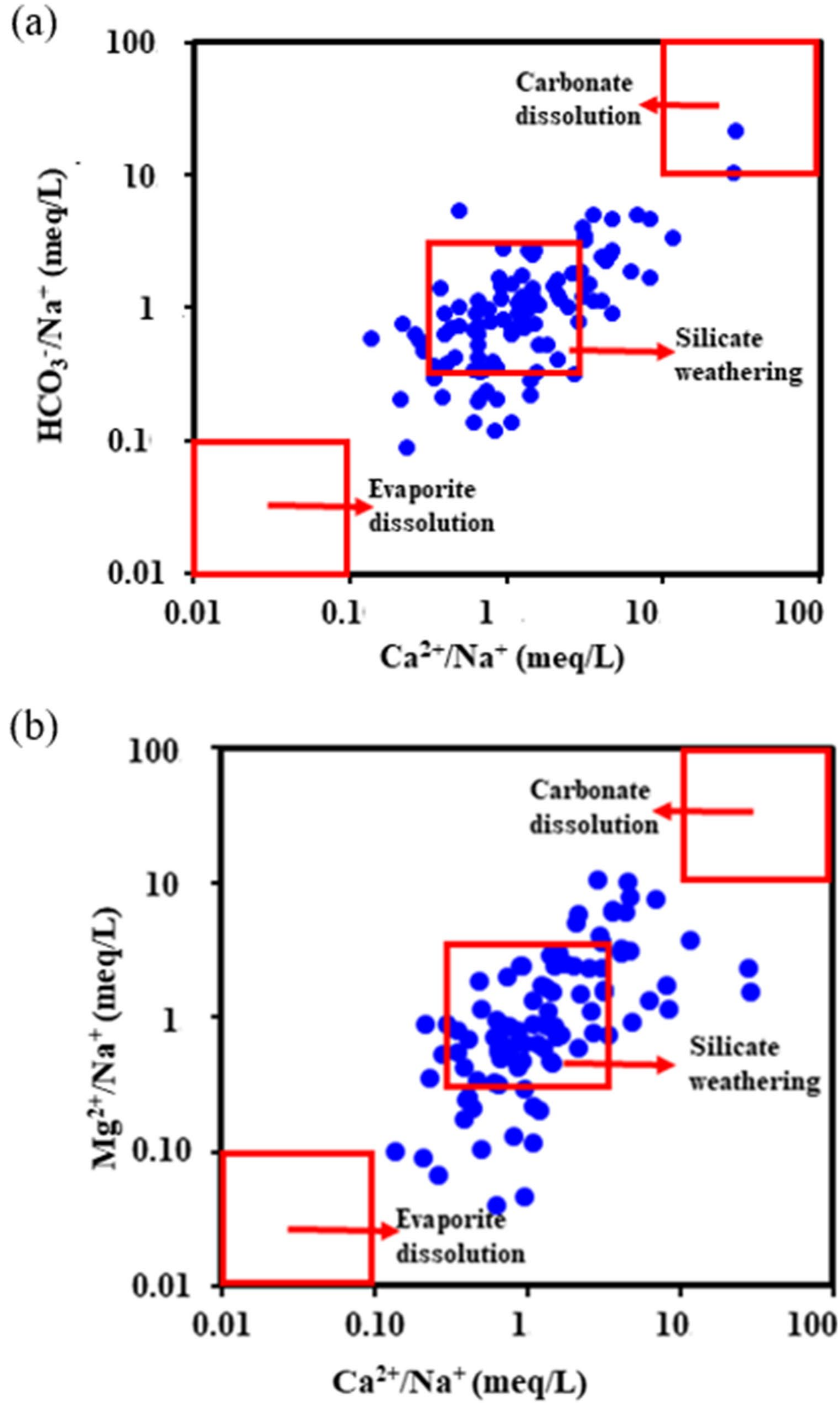

Fig. 5 Geochemical processes controlling the chemistry of groundwater

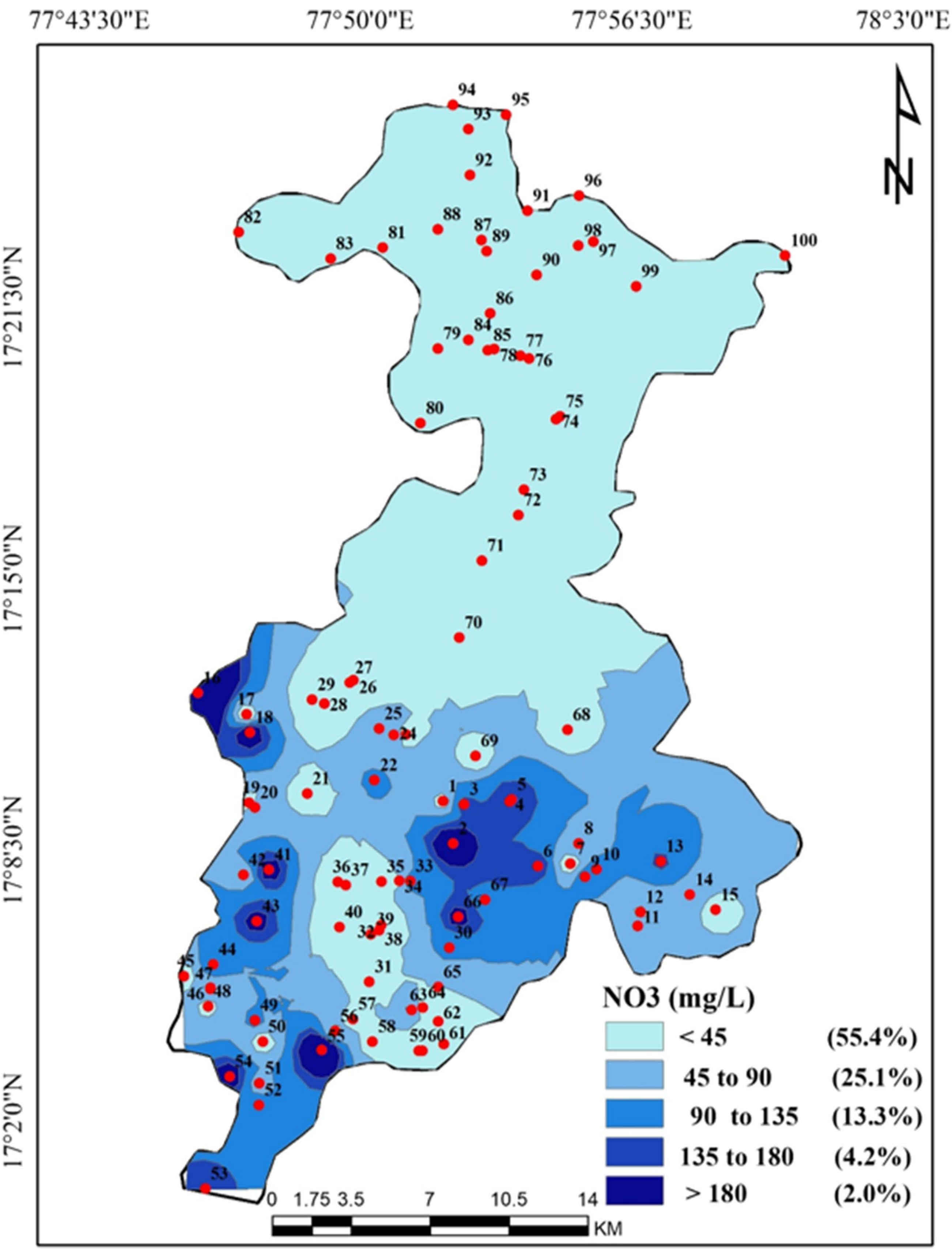

Fig. 6 Spatial distribution of NO_3^- content in groundwater

Table 3 Concentration of nitrate (NO_3^-) and fluoride (F^-) contents in groundwater near the present study region

Area/region	NO_3^-(mg/L)		F^- (mg/L)		Reference
	Range	Mean	Range	Mean	
Maheshwaram	3 to 200	62	0.31 to 3.03	1.43	Sujatha and Reddy (2003)
Yadadri-Bhuvanagiri	8.26 to 394	74.1	0.43 to 2.93	1.53	Shekhar et al. (2021)
Zaheerabad	1 to 50	27.5	0.13 to 2.63	0.89	Sakram et al. (2019)
Mulug-Venkatapur	0.1 to 897	117	0.28 to 5.48	1.26	Satyanarayana et al. (2017)
Chinnakodur	12 to 212	-	0.50 to 3.50	-	Narshima and Li (2019)
Rangareddy	7 to 300	-	0.50 to 4.50	-	Sujatha and Reddy (2003)
Present study area	0.04 to 585	56.3	0.22 to 5.41	1.13	-

groundwater is mainly the result of fluoride-rich minerals present in host rocks rather than the effect of phosphate fertilizers. As shown in Table 3, the F^- content in groundwater near the present study area varied from 0.13 to 5.48 mg/L with a mean of 0.89 to 1.53 mg/L. It has been observed that the F^- content (0.22 to 5.41 mg/L, with a mean of 1.13 mg/L) occurred in groundwater of the present study region, which significantly supports similar sources of F^-. As demonstrated in Table 5, the concentration of F^- (0.40 to 2.46 mg/L) was shown to increase with increasing pH (7.02 to 7.18), Na^+ (22.2 to 85.9 mg/L), and HCO_3^- (142 to 151 mg/L) and decreasing Ca^{2+} (54.7 to 40.2 mg/L). Figure 9 illustrates the positive correlation coefficient (r) of F^- with pH ($r = 0.29$) and Na^+ ($r = 0.52$), while the negative correlation with Ca^{2+} ($r = -0.20$) and HCO_3^- ($r = -0.03$). The positive correlation between F^- and pH indicates that water alkalinity promotes the leaching of fluoride-rich minerals, which affects F^- in groundwater (Brindha et al. 2016; Demelash et al. 2019). An inverse relationship between F^- and Ca^{2+} decreases by increasing Na^+ in the alkaline state supporting the positive correlation between Na^+ and F^- (Deepali et al. 2020). Since some groundwater samples belong to the deep aquifer, the negative correlation between F^- and HCO_3^- is usually due to the dissociation of alkaline water as carbonate and hydroxide (Madhnure et al. 2007; Salve et al. 2008). As a whole, it furthers supports the role of fluoride minerals contributing to the groundwater system.

The results of PCA are shown in Table 6, which provides information not only on the geochemical processes taking place in the aquifer system but also on the sources and origins of the inferior groundwater quality. High positive loadings of TDS (0.85), NO_3^- (0.81), Cl^- (0.77), and Na^+ (0.66) in PC1 were accounted for 28.5% of the total variance with a 3.14 eigenvalue. The combination of these ions with TDS reflects the salinity of the groundwater, which is the most common indicator primarily of man-made pollution on the aquifer system (Subba Rao et al. 2006; Ding et al. 2020). PC2 showed a high positive loading of F^- (0.73) with positive loadings of pH (0.52), Na^+ (0.55), and HCO_3^- (0.56), and a negative loading of Mg^{2+} (−0.56) with a total variance of 15.9% and eigenvalue of 1.75. This group indicates the effect of weathering and dissolution of fluoride-rich minerals occurring in host rocks over phosphate fertilizers (Karunanidhi et al. 2020; Narsimha and Qian 2020; Subba Rao et al. 2020a). In PC3, SO_4^{2-} had a high positive loading (0.65), while Ca^{2+} showed a negative loading (−0.59), which is 12.7% of the total variance with an eigenvalue of 1.40. These ions state the application of gypsum as an amendment to improve soil permeability (Subba Rao et al. 2017). The positive loading of Mg^{2+} (0.54) and the negative loading of K^+ (−0.63) with a total variance of 10.1% and eigenvalue of 1.11 in PC4 measured the effect of sewage on groundwater (Deepali et al. 2015). Therefore, the PCA further supports the role of geogenic and anthropogenic activities for variation in the chemical quality of groundwater taking place in the present study area.

Table 4 Mean values of TDS, Na^+, and Cl^- based on the classification of NO_3^-

NO_3^-(mg/L)		TDS (mg/L)	Na^+ (mg/L)	Cl^-(mg/L)	% of samples
Range	Mean				
<45	9.38	212	39.8	113	66
45 to 100	74.1	378	68.6	116	12
>100	186	480	89.2	179	22

Health risk index for nitrate and fluoride

When we look at the individual chemical parameters from Table 1, it becomes clear that all chemical parameters such as Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, Cl^-, and SO_4^{2-} (except NO_3^- and F^- ions) are above the highly desirable limits of 75, 30, 200, 12, 300, 250, and 200 mg/L, respectively, in less than 15% of the groundwater samples. But the ions NO_3^- and F^- are above the threshold limits of 45 mg/L and 1.5 mg/L in 34% and 25% of total groundwater samples, respectively (BIS 2012; WHO 2012). Furthermore, these two ions are more toxic than the rest of chemical parameters in drinking water because they can cause non-carcinogenic risk (USEPA 2014). Therefore, we have decided to assess the health risks

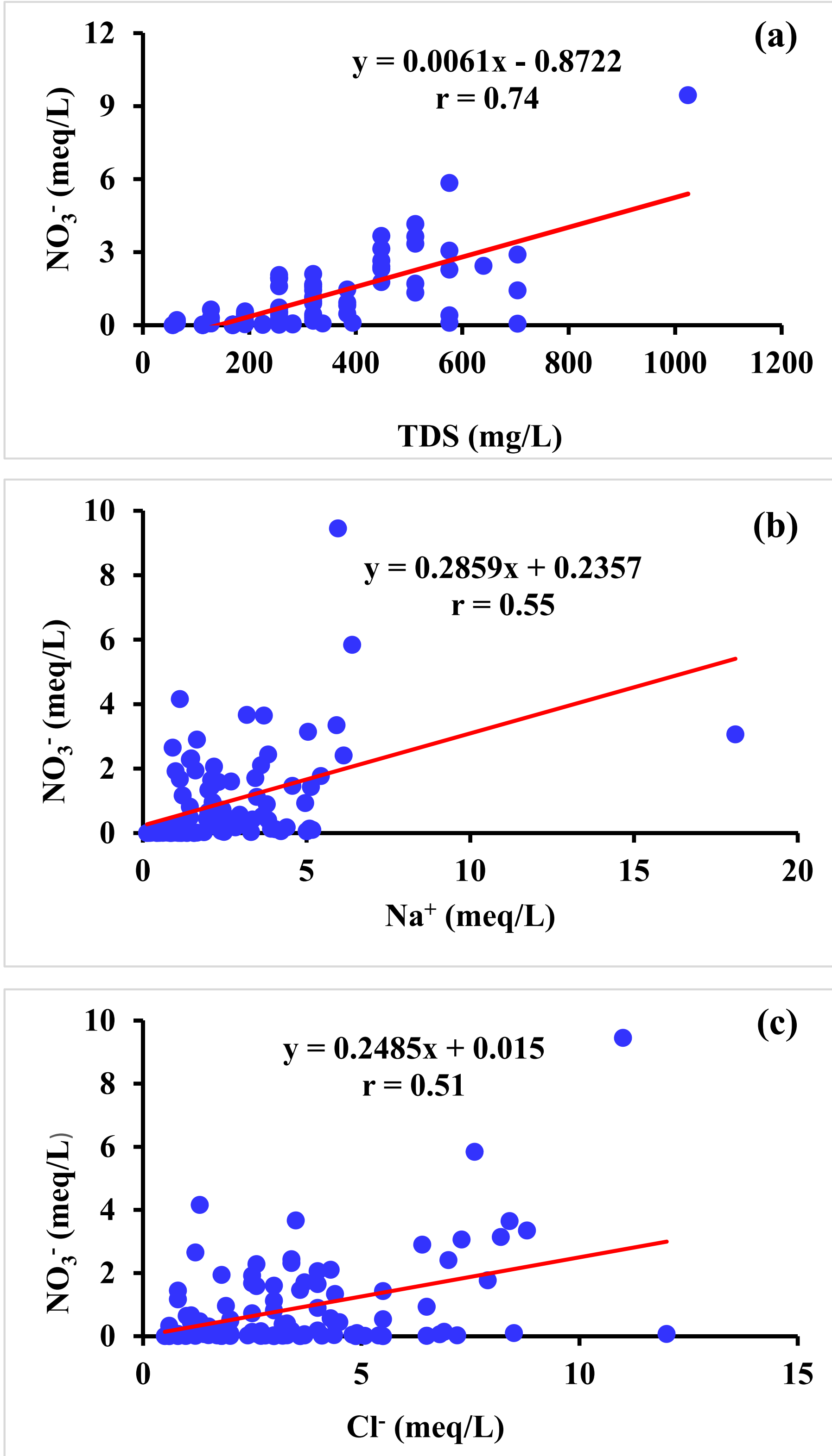

Fig. 7 Relationship between **a** TDS and NO_3^-, **b** Na^+ and NO_3^-, and **c** Cl^- and NO_3^-

of adults and children for NO_3^- and F^- pollutants in the present study.

According to the WHO (2012), high NO_3^- levels in drinking water affect the health of children and adults, while high F^- levels pose a health risk to people of all ages. For the

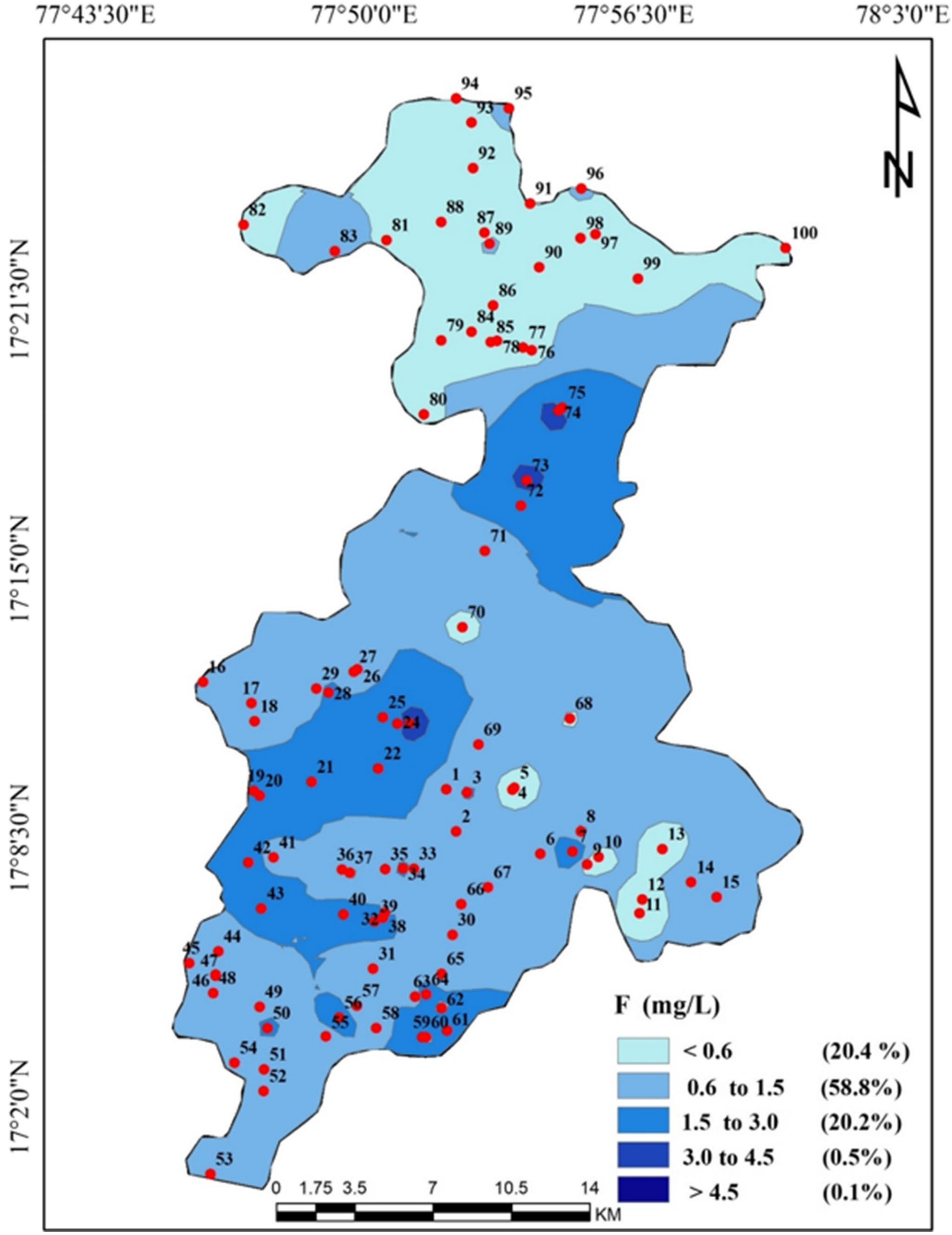

Fig. 8 Spatial distribution of F^- content in groundwater

Table 5 Mean values of pH, Ca^{2+}, Na^{+}, and HCO_3^- based on the classification of F^-

F^-(mg/L)		pH	Ca^{2+} (mg/L)	Na^+ (mg/L)	HCO_3^-(mg/L	% of samples
Range	Mean					
< 0.6	0.40	7.02	54.7	22.2	142	33
0.6 to 1.5	0.99	7.20	50.6	61.4	146	44
> 1.5	2.46	7.18	40.2	85.9	151	23

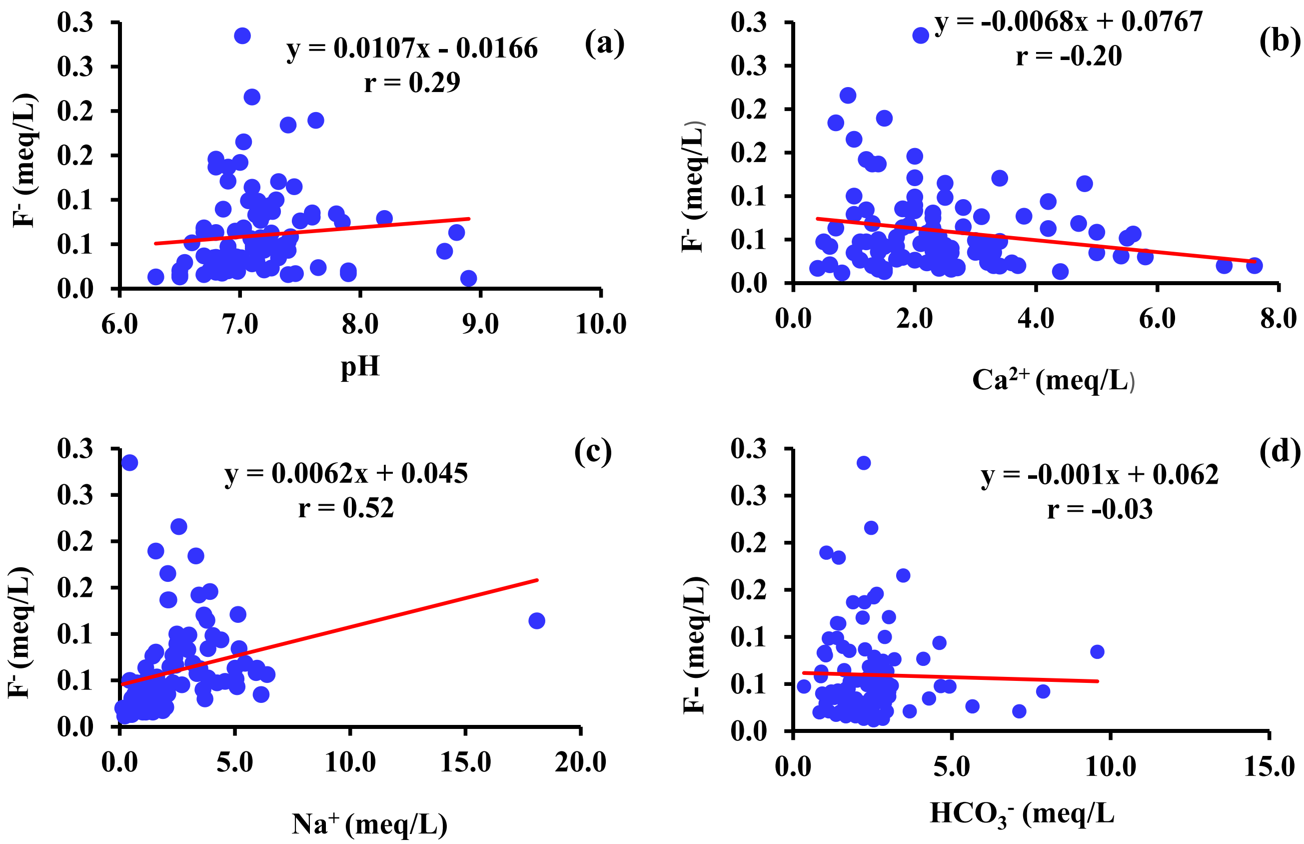

Fig. 9 Relationship between **a** pH and F^-, **b** Ca^{2+} and F^-, **c** Na^+ and F^-, and **d** HCO_3^- and F^-

calculation of HRI, the mean body weight of adults (65 kg) and children (18.7 kg) and the mean exposure time of adults (24,236 days) and children (4,380 days) were taken into account (ICMR 2009; USEPA 2014).

The values of hazard quotient (HQ) of nitrate ($HQ_{NO_3^-}$) varied from 0.01 to 19 for adults and from 0.01 to 29.3 for children, with a mean of 1.84 and 2.82, respectively (Table 7). Of the 100 groundwater samples, 40% and 48% of the samples showed a higher $HQ_{NO_3^-}$ of 1.0 for adults (4.27) and children (5.18), respectively (Table 8), which poses a health risk. The human health hazard quotient of fluoride (HQ_{F^-}) was between 0.19 and 4.70 for adults and between 0.29 and 7.23 for children with a mean of 0.54 and 1.78, in which 36% and 59% of groundwater samples had HQ_{F^-} more than 1.0 for adults (1.78) and children (2.15), respectively, causing health hazard. It was also observed from Table 7 that those children are at greater health risks due to NO_3^- than F^- compared to adults. This may be the result of groundwater contaminating with NO_3^- due to the impact of anthropogenic sources (household wastes, septic tanks leakage, irrigation-return-flows, nitrogen fertilizers, animal wastes, etc.) compared to the source of F^- (WHO 2012; Subba Rao et al. 2017, 2019).

Human health implications

To evaluate the overall implications of NO_3^- and F^- ions on human health, the Health Risk Index (HRI) was calculated, using Eqs. 6 to 8. HRI values varied from 0.20 to 20.1 for adults and 0.36 to 30.9 for children (Table 7). According to

Table 6 Principal component analysis (bold letters denote significant values > 0.50)

Chemical parameters	Principal components			
	1	2	3	4
pH	0.30	**0.52**	0.32	0.20
TDS	**0.85**	0.06	0.32	0.09
Ca^{2+}	0.44	− 0.13	**− 0.59**	− 0.03
Mg^{2+}	0.36	**− 0.56**	0.04	**0.54**
Na^{+}	**0.66**	**0.55**	− 0.01	0.13
K^{+}	0.47	0.03	0.40	**− 0.63**
HCO_3^-	0.22	**0.56**	− 0.45	0.34
Cl^-	**0.77**	− 0.27	− 0.02	− 0.11
SO_4^{2-}	− 0.20	− 0.10	**0.65**	0.43
NO_3^-	**0.81**	0.04	0.17	− 0.11
F^-	− 0.03	**0.73**	0.18	0.11
Eigenvalue	3.14	1.75	1.40	1.11
% total variance	28.5	15.9	12.7	10.1
Cumulative %	28.5	44.4	57.1	67.2

Table 7 Summary results of Hazard Quotient (HQ) and Health Risk Index (HRI)

Health hazards		Ions	Minimum	Maximum	Mean
HQ	Adults	NO_3^-	0.01	19.1	1.84
	Children		0.01	29.3	2.82
	Adults	F^-	0.19	4.70	0.54
	Children		0.29	7.23	1.78
HRI	Adults	$NO_3^- + F^-$	0.20	20.1	2.82
	Children	$NO_3^- + F^-$	0.36	30.9	4.34

the USEPA (2014), the recommended safe limit of HRI for non-cancer-causing hazard is 1.0 in drinking water. In the present study region, HRI was higher than 1.0 in 63% and 73% of total groundwater samples for adults and children, respectively. The mean HRI was 2.82 for adults and 4.34 for children (Table 7). This clearly indicates that the health risk is a threat to children rather than adults. This is not only due to the consumption of highly contaminated groundwater with a higher NO_3^- concentration than F^-, but also due to the smaller body weight and shorter exposure time (USEPA 2014) compared to adults.

To identify the intensity of human health risk zones, the spatial distribution of HRI for adults and children is shown in Fig. 10. Zones less than 1.0 and more than 1.0 of HRI covered 33.3% and 66.7% for adults and 28.1% and 71.9% for children of total study region, respectively. The former zone (< 1.0) was within the safe limit (mean HRI: 0.54 for adults and 0.68 for children), while the second one (> 1.0) was above the safe limit (mean HRI: 4.16 for adults and 5.70 for children) for non-cancer health problems (Table 8). Therefore, the intensity of the human health risk zone is 1.37 times higher in children than in adults. This study divides the region into Northern Safe Health Zone (33.3% for adults and 28.1% for children) and Southern Unsafe Health Zone (66.7% for adults and 71.9% for children), respectively, depending on the intensity of agricultural activity.

The effect of unlimited application of nitrogen fertilizers compared to fluoride minerals and phosphate compost seems to have formed the human health risk zone of the southern part. This fact was established by observing the spatial distribution of NO_3^- and F^- contents (Figs. 5 and 7), where the F^- ion showed a safer health zone compared to the NO_3^- associated with non-cancer risk. Li and Wu (2019) from China and Subba Rao et al. (2021a) from India stated that the major NO_3^- content is the result of impact of agricultural fertilizers in groundwater. Furthermore, due to the intensive agricultural practices in southern part, it is also important to consider the effects of return-irrigation-flows and animal wastes as a source of high NO_3^- in groundwater (Deepali et al. 2015). Therefore, the study helps to decipher the specific sites of HRI zones (> 1.0) for children (71.9%) and adults (66.7%; Fig. 10) to take preventive measures for stable health conditions.

Remedial measures

The intensity of susceptible zones that are likely to protect and manage groundwater resources from pollution is essential for making a healthy society for long-term growth. The present study region suggests some useful and easily applicable preventive measures. They include (a) supply of safe drinking water to maintain general health; (b) arrangement of denitrification and defluoridation tools to reduce NO_3^- and F^- content, respectively; (c)

Table 8 Mean values of Hazard Quotient (HQ) and Health Risk Index (HRI) based on acceptable limit of health hazard

Acceptable limit	$HQ_{NO_3^-}$		HQ_{F^-}		$HRI_{NO_3^- + F^-}$	
	Adult	Children	Adult	Children	Adult	Children
< 1.0	0.21 (60%)	0.18 (52%)	0.54 (64%)	0.61 (41%)	0.54 (37%)	0.68 (27%)
> 1.0	4.27 (40%)	5.18 (48%)	1.78 (36%)	2.15 (59%)	4.16 (63%)	5.70 (73%)

Values in bracket denote % of samples

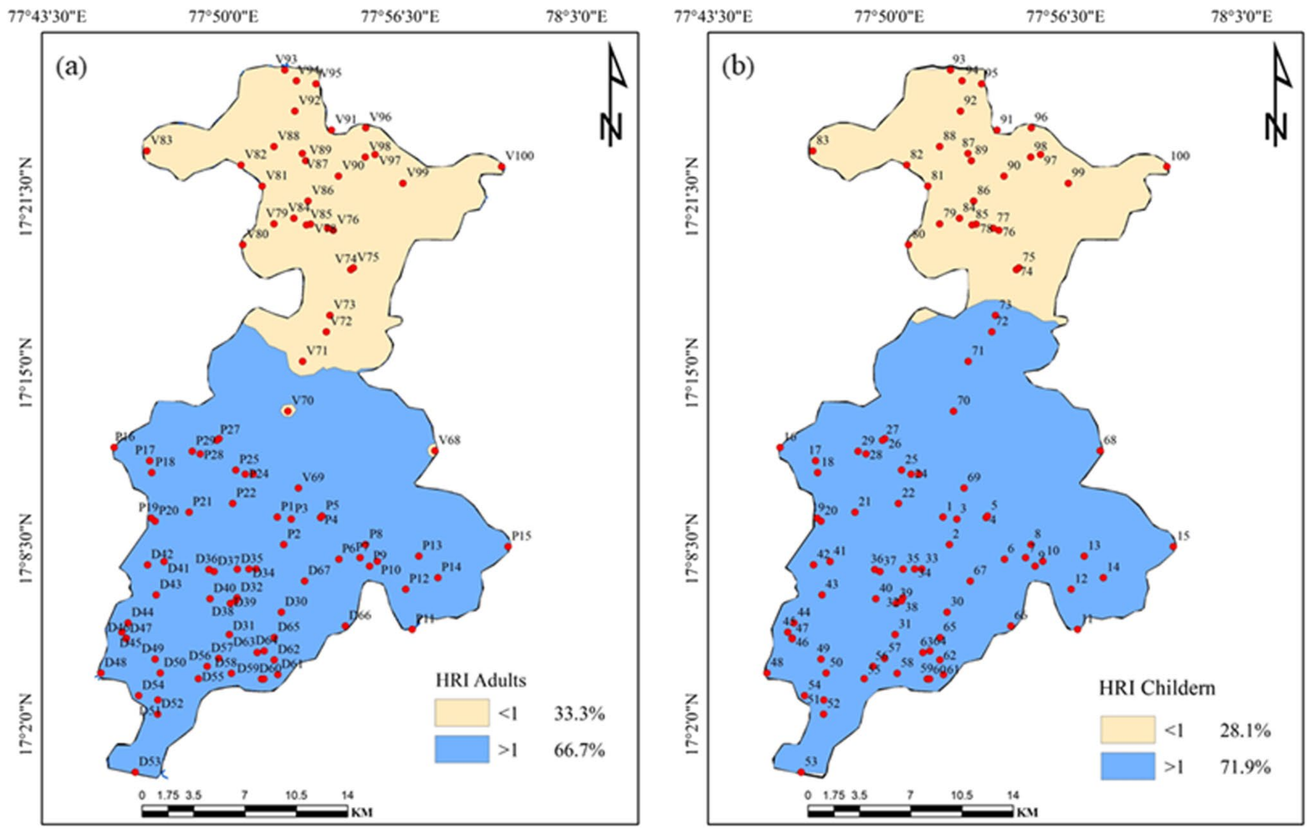

Fig. 10 Spatial distribution of Health Risk Index (HRI) with respect to **a** adults and **b** children

implementing rainwater harvesting methods to dilute the concentrations of NO_3^- and F^- ions in the groundwater system; (d) providing hygienic-sanitary facilities for clean surrounding habitats; and (e) using limited chemical fertilizers in accordance with soil conditions to prevent contamination activities.

Conclusions

The following conclusions were drawn, after examining Groundwater Quality Index (GQI), Piper's diagram, bivariate diagrams, principal component analysis, and Health Risk Index (HRI) issues related to groundwater quality from a rural region of Telangana, India:

- Groundwater quality showed Na^+ and HCO_3^- as the dominant ions. Piper's diagram and bivariate diagrams (Ca^{2+}/Na^+ vs HCO_3^-/Na^+ and Ca^{2+}/Na^+ vs Mg^{2+}/Na^+) illustrated that groundwater is primarily carbonate water type and controlled by silicate weathering, respectively.
- GQI indicated that the chemical quality of groundwater is suitable for drinking water needs. However, the NO_3^- content (0.04 to 585 mg/L with a mean of 56.3 mg/L) and the F^- content (0.22 to 5.41 mg/L with a mean of 1.13 mg/L) exceeded the consumption water quality limits of 45 mg/L and 1.5 mg/L in 34% and 25% of the total groundwater samples, respectively. Nitrate fertilizers are the main source of NO_3^- content, which is confirmed by the relationship of NO_3^- with TDS, Na^+, and Cl^-. Fluoride-rich minerals are the prime source of F^- content, which is confirmed by the relationship of F^- with pH, Ca^{2+}, Na^+, and HCO_3^-. Principal component analysis further supports these views.
- The values of HRI varied from 0.20 to 20.10 and 0.36 to 30.90 with a mean of 2.82 and 4.34 for adults and children, respectively. The severity of HRI was 1.37 times higher in children (5.70) than in adults (4.16) due to the differences in weight size and exposure time. According to the acceptable limit of more than 1.0, the study divided the region into Northern Safe Health Zone (33.3% for adults and 28.1% for children) and Southern Unsafe Health Zone (66.7% for adults and 71.9% for children). This is due to the intensity of agricultural activities.
- The present study recommended the effective management measures such as supply of safe drinking water, denitrification, defluoridation, rainwater harvesting tech-

niques, sanitary facilities, and limitation of chemical fertilizers not only to protect groundwater resources from pollution activities but also to improve health conditions of the locals.

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Author contribution Sakram Gugulothu: supervision, methodology, and original draft preparation.

N. Subbarao: writing, reviewing, editing.

Rashmirekha Das: literature collection, editing.

Laxman Kumar Duvva: literature collection, draft preparation.

Ratnakar Dhakate: statistical analysis, literature collection.

Declarations

Ethics approval Not applicable to this manuscript.

Consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare no competing interests.

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