Total Water Quality And Total Human Health Hazard Indices Approach For Health Risk Evaluation With Oral Intake Pathway of Groundwater of Nitrate And Fluoride From A Rural Region of South India

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Total water quality and total human health hazard indices approach for health risk evaluation with oral intake pathway of groundwater of nitrate and fluoride from a rural region of South India

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Abstract

Evaluation of chemical quality of groundwater and associated health hazards is a prerequisite for taking remedial measures elsewhere. A rural region of South India was, thus, chosen for the present study to assess the total quality of groundwater and also to decipher the human health risk zones with respect to adults and children due to the groundwater pollution with nitrate (NO_3^-) and fluoride (F^-) ions. Groundwater samples collected from the study region were determined for various chemical parameters. According to the total water quality index, groundwater quality is suitable for drinking purposes. However, the NO_3^- (0.4 to 585.20 mg/L) and F^- (0.22 to 5.41 mg/L) ions exceed the drinking water quality limits of 45 mg/L and 1.5 mg/L in 34% and 25% of the groundwater samples, respectively. Nitrate fertilizers appeared as the chief source of NO_3^- , and the fluoride minerals as the main source of F^- in the groundwater body, which are further supported by principal component analysis. Total human health hazard index (THHHI) was observed to be higher than its tolerable limit of 1.0 in 63% and 73% of the groundwater samples in respect of NO_3^- and F^- of adults and children, respectively. The intensity of human health risk zones of THHHI (>1.0) was 1.37 times higher in children (5.69) than in children (4.15), which cover an area of 71.75% and 66.73%, respectively. Thus, the effective strategic measures were recommended for the

protection of groundwater resources from pollution and also for improving the human health conditions.

Keywords: Groundwater, Total water quality index, Total Human health hazard index, Human health risk zones, Rural region, India

Introduction

Because of the rapid climatic changes, groundwater is an essential source globally, especially for both drinking and irrigation purposes (Reddy and Sakram 2014). More than 85% of the rural population depends upon the groundwater resources for their daily needs (Kulkarni et al. 2015). Though about 60 to 85% of groundwater is utilized for drinking and agriculture purposes, respectively, in India (Sishidia et al. 2016; Sakram and Narsimha 2018), most of it has been contaminated by naturally processes and artificially activities (Alaya et al. 2014; Subba Rao et al. 2017). Natural contamination is due to the influence of toxic components existing in the soil and rocks (example: fluoride minerals), while the artificial agents such as poor drainage conditions, spillage of septic tanks, irrigation-return-flows, immense usage of agrochemicals, etc cause inferior quality of groundwater and consequently health problems (Sakram et al. 2018; Subba Rao and Chaudhary 2019; Wu et al. 2019a).

Recently the researchers are mainly focused their work on the chemical quality of groundwater and associated health problems with respect to nitrate (NO_3^-) and fluoride (F^-) ions, as they are the most common toxicities in the groundwater (Qasemi et al. 2018; Subba Rao et al. 2020a, b). Nitrate is the most common in the regions of agricultural activities (especially due to nitrogen fertilizers), irrigation-return-flows, flowing of untreated household wastes sewage and septic tank leakages onto the ground, nitrogen-rich soils, and animal wastes (Marghade et al. 2015; 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). About 118 million people drink water with a NO_3^- level ranging from 45 to 100 mg/L and more than 108 million people consume water with higher than 100 mg/L of NO_3^- in India (Karunanidhi et al. 2020). Approximately 200 million people suffer from high F^- content (> 1.5 mg/L) in the groundwater globally, especially from the countries, including Africa, China, Iran, Nigeria, Pakistan, South America, and Sri Lanka (Wu et al. 2015; Craig et al. 2015; Chen et al. (2017). The fluoride minerals (fluorite, apatite, biotite and hornblende) occurring in the basement rock such as hornblende-biotite, gneiss, and granites are the chief source of F^- contamination of groundwater and the agrochemicals like phosphate fertilizers lead to rise the concentration of F^- as secondary source in the groundwater (Subba Rao 2017a; Karunanidhi et al. 2019; 2020).

It is a well-known fact that the consumption of contaminated groundwater by NO_3^- more than 45 mg/L would cause methemoglobinemia (blue baby syndrome), where red blood cells reduce their ability to carry oxygen, which leads to shortness of breath, heart attack, and even death, especially in children. Sometimes, it leads to cancer also (WHO 2012). Whereas the F^- higher than 1.5 mg/L causes severe fluorosis (BIS 2012). Globally, it was observed that children are more vulnerable in comparison with adults in respect of NO_3^- and F^- ions (Zhai et al. 2017; Rezael et al. 2018; Karunanidhi et al. 2020). In India, the potential risk of groundwater is a consequence of contamination of NO_3^- and F^- ions, where children are at a greater health risk than adults, hiking the non-carcinogenic problems in children (Ding et al. 2020; Kaur et al. 2020; Subba Rao et al. 2021a).

The present study region is a part of Telangana State, India, which is a rural area (Fig. 1). It has intensive and long-term practice. So, the influences of unlimited usage of chemical

composts (nitrate, phosphate, and potassium verities), irrigation-return-flows, and animal wastes are the most common phenomena on the groundwater system. Basic sanitary facilities like disposal of household wastes, flowing of leakage of septic tanks, etc are poor conditions in the study region, which are known as the most contamination sources of groundwater. However, there is no scientific study so far on the human health risk, especially due to the drinking of NO_3^- and F^- polluted groundwater. People are depending upon the groundwater resources for their drinking purpose due to the lack of surface water supply. Therefore, the main focus of the present study is on the assessment of health risks caused by NO_3^- and F^- contaminations in the groundwater. The study facilitates the decision-making authorities for the protection of the groundwater quality from the pollutants, and thereby for improving the health conditions of the local community.

Figure 1 should be placed here

Study region

The present rural region is located in the southwestern part of Telangana, India. It lies between north latitude $17^\circ 23'$ - $17^\circ 25'$ and east longitudes $77^\circ 45'$ - $78^\circ 50'$, falling in Survey of India toposheet number 56G/15 and 56G/16 and covering a geographical area of 632.45 sq kms (Fig. 1). The region comes under semi-arid climate with the annual average temperature varying from 14°C to 41°C and the annual average rainfall (5 years) of 937 mm. The surface runoff has resulted from the development of sub-dendritic drainage pattern in the study region.

The prominent rock exposures in the study region are basalts and granites (Fig. 1). Laterite patches also occur. The basalts are fine-grained and dark-colored volcanic rocks, which are composed of calcic plagioclase feldspars and clinopyroxene with olivine, quartz,

hornblende, nepheline, orthopyroxene, etc. The granites are generally medium- to coarse-grained. They contain the quartz, plagioclase and potassium feldspars, biotite, apatite, hornblende, etc. Basically, they are hard rocks. However, the occurrence of vesicular structures, cracks, and joints become aquifers in the basalts, while the presence of weathered and fractured rocks are the water-bearing formations in the granites. The laterites are porous, but they are slightly permeable with a limited areal extent. Groundwater occurs under water table to semi-confined conditions. Depth to groundwater table varies from 18 m to 28 m below ground level. The quality of groundwater is generally appeared to be potable in the fieldwork.

Materials and methods

A hundred bore wells were observed from the study region during May 2015 (Fig. 1). The groundwater samples were collected from them in one-liter capacity polythene bottles, which were cleaned with 1:1 dilute hydrochloric acid and wash away with distilled water three times before collecting the water samples, following the standard procedure (APHA 2012).

The pH and electrical conductivity (EC) were measured in the field, using their meters (Table 1). TDS were calculated by multiplying EC with 0.64 factor. The other chemical parameters (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , HCO_3^- , Cl^- , SO_4^{2-} , NO_3^- and F^-) were estimated, using the conventional procedures such as titration (Ca^{2+} , HCO_3^- , and Cl^-), calculation (Mg^{2+}), flame photometer (Na^+ and K^+), UV spectrophotometer (SO_4^{2-} and NO_3^-), and ion-selective electrode (F^-), and following the standard water quality methods (APHA 2012). For the computation of analytical error, the total cations ($\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+$) and the total anions ($\text{HCO}_3^- + \text{Cl}^- + \text{SO}_4^{2-} + \text{NO}_3^- + \text{F}^-$) were used, which was observed to be $\pm 5\%$ (Subba Rao 2017a), reflecting the reliability of the chemical data.

Comprehensive tool for utilization of groundwater quality for drinking

The total water quality index (TWQI) is a comprehensive technique to express the overall drinking water quality in a single term (Subba Rao et al. 2020; Wu et al. 2020a). The first step in this index is the calculation of relative weight (W_i), after assigning the unit weight (w_i), for each chemical parameter on the basis of its relative significance on the human health (Eq. 1). The second step is the computation of rating of water quality (q_i), which is divided by the concentration of chemical parameter (C_i) with national drinking water quality standard (S_i) for each chemical parameter (Eq. 2). The third step is the assessment of SI_i by multiplication of q_i with W_i to each chemical parameter (Eq. 3). The last step is the computation of TWQI by adding all SI_i in each sample (Eq. 4).

$$W_i = \frac{w_i}{\sum_{n=1}^n w_i} \quad \dots\dots \quad (1)$$

$$q_i = \frac{C_i}{S_i} \times 100 \quad \dots\dots \quad (2)$$

$$SI_i = q_i \times W_i \quad \dots\dots \quad (3)$$

$$TWQI = \sum_{i=1}^n SI_i \quad \dots\dots \quad (4)$$

When the TWQI is less than 50, it indicates an excellent quality of quality; when it is from 50 to 100, it shows a good quality of water; when it is between 100 and 200, it specifies a poor quality of water; when it is from 200 and 300, it represents a very poor quality of water; when it is more than 300, it suggests an unsuitable quality of water for drinking purpose.

Human Health risk assessment

The NO_3^- and F^- ions were chosen for the human health risk assessment. The oral intake procedure was selected for the computation of total human health hazard index (THHHI)

with respect to adults and children (Li et al. 2019a, b; Wu et al. 2020). The THHHI was calculated as shown in Eqs. 4 to 6 (USEPA 1991, 2006).

$$MDD = CGW \times IR \times ED \times EF / MBW \times MET \quad \dots \quad (4)$$

$$HHHQ = MDD / RfD \quad \dots \quad (5)$$

$$THHHI = \sum_{i=1}^n HHQ_i \quad \dots \quad (6)$$

where MDD is the mean dosage of daily of NO_3^- and F^- (mg/kg/day), CGW is the concentration of NO_3^- and F^- in the groundwater (mg/L), IR represents the ingestion rate (3 L/day for adults and 1.5 L/day for 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), MBW is the mean body weight (65 kg for adults and 18.7 kg for children), MET is the mean exposure time (24,236 days for adults and 4,380 for children), HHHQ is the hazard quotient, RfD is the reference dose for chronic oral exposure (1.60 mg/kg/d for NO_3^- and 0.06 mg/kg/d for F^- (ICMR 2009; UNDESA 2013; USEPA 2014; Brindha et al. 2016), and THHHI is the total human health hazard index (non-carcinogenic hazard).

The tolerable limit of THHHI is 1.0 (USEPA 2014). If it is above than 1.0, the non-carcinogenic risk of the pollutant is greater than the tolerable level. If it is below 1.0, the non-carcinogenic risk is at acceptable limit.

Results and discussion

Groundwater characteristics

The pH in the groundwater ranged from 6.30 to 8.90 with a mean of 7.14 (Table 1), which indicates that it is slightly acidic to highly alkaline nature. Three groundwater samples are

exceeding the safe limit of pH (6.5 to 8.5) in drinking water, which causes damage to the mucous membranes (BIS 2012). The TDS was from 56 to 1024 mg/L with a mean of 291 mg/L. About 15% of the groundwater samples are higher than its the recommended limit of 500 mg/L allowing for drinking purposes, causing gastrointestinal irritation (BIS 2012).

Table 1 should be placed here

Cations

The Ca^{2+} content was from 8.02 to 152.30 mg/L being a mean of 49.60 mg/L (Table 1). Fourteen percent of the groundwater sample showed a non-acceptable limit (75 mg/L) of Ca^{2+} (BIS 2012). Weathering and dissolution of plagioclase feldspars are the chief sources of Ca^{2+} in the groundwater (Subba Rao et al. 2017). The Mg^{2+} was between 2.43 and 92.42 mg/L with a mean of 23.53 mg/L, which is more than the tolerable limit of 50 mg/L in 6% of the groundwater samples. The Mg^{2+} is mainly attributed to the dissolution of ferromagnesian minerals (olivine, pyroxene, biotite, etc.) occurring in the host rocks, in addition to human-induced activities (Subba Rao 2021). The Na^+ was from 3 to 416 mg/L, with a mean of 54.13 mg/L. Only one groundwater sample shows Na^+ more than the threshold limit of Na^+ 200 mg/L (BIS 2012). The plagioclase feldspars in the basement rocks are the prime source and the anthropogenic origin (household wastes, irrigation-return-flows, etc) is another source of Na^+ in the groundwater (Subba Rao 2021). The K^+ ranged from 1 to 118 mg/L and its mean is 6.20. This exceeds the desirable limit of 12 mg/L in 10% of the groundwater samples. The orthoclase feldspars are the prime source and the potassium composts are the secondary source of K^+ in the groundwater.

Anions

The HCO_3^- was from 146.40 to 2,538 mg/L, with a mean of 1,014.31 mg/L (Table 1), which is caused by soil CO_2 . This is released from the decay of organic decomposition (Subba Rao et al. 2017). The HCO_3^- is greater than its allowable limit of 300 mg/L for drinking purposes in 99% of the groundwater samples (BIS 2012). The concentration of Cl^- was between 17.73 and 425.40 mg/L with a mean of 127.52 mg/l. According to the drinking water quality standards, the Cl^- is more than 250 mg/L in 10% of the groundwater samples, causing a salty taste and laxative effect. Non-lithological source (domestic waste waters, irrigation-return-flows, etc) is the prime contributor of Cl^- in the groundwater (Sarath Prasanth et al. 2012; Laxman et al. 2019). The value of SO_4^{2-} was from 30 to 166 mg/L and its mean is 97.90 mg/L. It is not more than its acceptable limit of 200 mg/L in all groundwater samples. Since no traces of sulphide minerals in the country rocks, utilization of gypsum for alternation of conditions is the only source of SO_4^{2-} in the groundwater body (Subba Rao et al. 2017).

The NO_3^- ranged from 0.4 to 585.20 mg/L, being a mean of 56.27 mg/L. In 34% of the groundwater samples, it is above the desirable limit of 45 mg/L, causing a blue baby disease (BIS 2012). The NO_3^- is a result of the influence of sewage wastes, septic tanks leakage, agricultural fertilizers and animal wastes on the aquifer system (Marghade et al., 2015; Zhang et al. 2018; He et al. 2019). In the present study region, the F^- was from 0.22 to 5.41 mg/L, with a mean of 1.13 mg/L. It exceeds 1.5 mg/L in 25% of the groundwater samples, causing fluorosis. The occurrence of fluoride containing minerals like fluorite, biotite, hornblende, etc. in the host rocks and the usage of phosphate composts in the agricultural area are the main sources of F^- in the groundwater body (Subba Rao et al. 2016, 2020a).

Total groundwater quality assessment for drinking purpose

The total water quality index (TWQI) is a scale to measure the overall drinking water quality (Subba Rao et al. 2020a). The computed values of TWQI were from 30 to 91 with a mean of 52.72 (Table 2). According to the classification of TWQI, 51% and 49% of the groundwater samples come under the excellent (TWQI: < 50) and good (TWQI: 50 to 100) water quality types for drinking purposes, respectively. It suggests that the quality of groundwater is fit very well for drinking purposes without any water treatment.

Table 2 should be placed here

However, when we observed the individual chemical parameters from Table 1, it is apparent that all chemical parameters such as Ca^{2+} , Mg^{2+} , Na^+ , K^+ , HCO_3^- , Cl^- and SO_4^{2-} , (excepting NO_3^- and F^- ions) are more than highest desirable limits of 75, 30, 200, 12, 300, 250, and 200 mg/L, respectively, in less than 15% of the groundwater samples. But in the case of NO_3^- and F^- ions, they are above the safe limits of 45 mg/L and 1.5 mg/L in 34% and 25% of the total groundwater samples, respectively (BIS 2012; WHO 2012). Further, these two ions are the most common toxicities than the rest of the chemical parameters in the drinking water because of their potential production of non-carcinogenic risk (USEPA 2014).

Since agriculture is one of the main practices of the present study region, a considerable portion of applied agrochemicals (nitrogen fertilizers) is expected to penetrate the soils/rocks, in addition to the influences of household waste waters, septic tank leakages, and animal excretions, and reaching the aquifer body through the recharge water. This is likely to increase the NO_3^- levels in the groundwater. However, NO_3^- levels less than 45 mg/L were observed from 54.69% of the total study region (Fig. 2a). They were mainly confined to

the northern part and however were also found as very limited isolated pockets in the southern part, where the agricultural activities are comparatively less. Next higher NO_3^- levels (45 to 100 mg/L and > 100 mg/L) were mainly observed in the southern part in 46.31% of the total study region, where the agricultural activities are more. Therefore, the influences of utilization of nitrogen fertilizers, irrigation-return-flows, and animal wastes are expected on the aquifer system. This hypothesis is further substantially supported by the increasing of NO_3^- (9.38 to 186.12 mg/L) along with TDS (212.33 to 480 mg/L), Na^+ (39.82 to 89.18 mg/L) and Cl^- (112.70 to 178.54 mg/L), and also by the higher loading of NO_3^- (0.813) along with TDS (0.851), Na^+ (0.660) and Cl^- (0.768; Tables 3 and 4), which are the most common indicators of human-induced pollution (Subba Rao et al. 2017).

Figure 2 should be placed here

Tables 3 and 4 should be placed here

The higher alkalinity (pH and HCO_3^-) with Na^+ activates the leaching of fluoride minerals occurring in the basement rocks and thus increases the higher F^- content in the groundwater system (Subba Rao et al. 2016, 2020a). Apart from this, usage of phosphate fertilizers may also lead to an increase of concentration of F^- in the groundwater (Subba Rao et al. 2021a). Spatial distribution of concentration of F^- ion in the groundwater of the present study region showed that the F^- level less than 0.6 mg/L was observed mainly from the northern part and also as very limited isolated patches in the southern part, which covers an area of 20.36% of the total study region (Fig. 2b). A safe limit of F^- (0.6 to 1.5 mg/L) was found in the study region (58.47%). The next higher F^- content (> 1.5 mg/L) was found as isolated pockets (21.17%) from the entire study region irrespective of the agricultural activities. As demonstrated in Tables 3 and 4, the concentration of F^- (0.40 to 2.42 mg/L)

shows an increase with an increase of pH (7.02 to 7.30), Na^+ (22.24 to 83.83 mg/L), and HCO_3^- (863.98 to 1099.78 mg/L), and the higher loading of F^- (0.732) also shows the higher loadings of pH (0.519), Na^+ (0.552) and HCO_3^- (0.555), which obviously indicates that the spatial distribution of F^- content could be mainly caused by the influence of weathering and dissolution of fluoride-bearing minerals occurring in the host rocks rather than that of phosphate fertilizers on the groundwater system.

According to WHO (2011), excessive NO_3^- levels in the drinking water affects the health of children and adults, whereas the high F^- levels provide a health risk to people of all ages. For computation of THHHI, the mean body weight is 65 kg for adults and 18.7 kg for children, and the mean exposure time is 24,236 days for adults and 4,380 days for children are taken into consideration (ICMR 2009; USEPA 2014). From this point of view, we have decided to evaluate the health risks between adults and children with respect to NO_3^- and F^- pollutants in the present study.

Human health risk with respect to nitrate and fluoride

The values of the human health hazard quotient of nitrate ($\text{HHHQ}_{\text{NO}_3^-}$) varied from 0.01 to 19.08 for adults and from 0.01 to 29.34 for children, with a mean of 1.83 and 2.82, respectively (Table 2). Out of 100 groundwater samples, 39% and 47% of the samples showed $\text{HHHQ}_{\text{NO}_3^-}$ more than 1.0 with respect to adults (4.26) and children (5.68), respectively, causing a health risk. The human health hazard quotient of fluoride (HHHQ_{F^-}) was between 0.19 and 4.70 for adults and between 0.29 and 7.23 for children, with a mean of 0.99 and 1.52, in which 36% and 58% of the groundwater samples had HHHQ_{F^-} more than 1.0 with respect to adults (1.78) and children (2.14), respectively, causing a health hazard. It

is also significantly observed from Table 2 that children are more threatening to health risks due to the NO_3^- rather than the F^- compared to adults. According to WHO (2011) and Subba Rao et al. (2017, 2019b), the groundwater could have been more contaminated with NO_3^- due to the influences of anthropogenic sources (household wastes, septic tanks leakage, irrigation-return-flows, nitrogen fertilizers, animal wastes, etc) compared to the source of F^- .

Total human health implications

In order to evaluate the total implications of NO_3^- and F^- ions on the human health, the total human health hazard index (THHHI) was computed, following the Eqs. 4 to 6. The values of THHHI varied from 0.27 to 20.4 for adults and 0.26 to 19.6 for children (Table 2). According to the USEPA (2014), the recommended safe limit of THHHI for the non-cancer-causing hazard is 1.0 in the drinking water. In the present study region, the THHHI was more than 1.0 in 63% and 73% of the total groundwater samples with respect to adults and children, respectively. It is also further observed that the mean THHHI is 2.82 for adults and 4.34 for children. It obviously suggests that the danger is a threat to children than to adults. This appears to be caused not only by the consumption of more contaminated groundwater with a higher concentration of NO_3^- than that of F^- , but also by the smaller weight body and the lesser exposure time of children compared to adults (USEPA 2014).

For the identification of intensity of human health risk zones, the spatial distribution of THHHI for adults and children was demonstrated in Fig. 3. The zones with less than 1.0 and more than 1.0 of THHHI were covered by 33.27% and 66.73% for adults and 28.25% and 71.75% for children of the total study region, respectively. The first zone was in safe limit (average THHHI: 0.54 for adults and 0.66 for children) with respect to non-cancer health risk, while another one was in the unsafe limit (average THHHI: 4.15 for adults and

5.69 for children) of non-cancer health problem (Table 3). Thus, the intensity of human health risk zone appears to be 1.37 times higher in children than in adults. These two zones have obviously divided the region into the northern safe health zone and the southern unsafe health zone, respectively, depending upon the intensity of the agricultural activities.

Figure 3 should be placed here

The human health risk zone of the southern part appears to be caused by the influence of the unlimited application of nitrogen fertilizers compared to that of fluoride minerals and phosphate composts. This fact is established by observing the spatial distribution of NO_3^- and F^- contents (Fig. 2), where the F^- ion shows mostly the safe health zone compared to the NO_3^- associated non-cancer risk. Li et al. (2019a) from China and Subba Rao et al. (2021a) from India stated that the main NO_3^- content is a result of the influence of agricultural fertilizers of the human-induced sources in the groundwater. Further, because of the intensive agricultural practices in the southern part, it is also important to consider the influences of return-irrigation-flows and animal wastes as a source of higher NO_3^- in the groundwater (Marghade et al., 2015). Therefore, the study helps for deciphering the specific sites of the human health risk zones ($\text{THHHI} > 1.0$) in the case of children (71.75%) and adults (66.73%; Fig. 3) for taking remedial measures for sustainable health conditions.

Remedial measures

The intensity of susceptible zones for the purpose of protecting and managing groundwater resources from pollution is essential for making health society for long-term growth. Since the groundwater is the only resource for the locals, the present study suggests some useful

and easily applicable remedial measures such as (a) supply of safe drinking water to maintain the normal health, (b) arrangements of denitrification and defluoridation tools to reduce the NO_3^- and F^- contents, (c) implementation of rainwater harvesting techniques to dilute the groundwater salinity, including the concentrations of NO_3^- and F^- ions, (d) providing the hygienic-sanitary facilities for clean surrounding residential locations, (e) utilization of limited chemical fertilizers, according to the soil conditions, to arrest the contamination activities, and (f) education of the local population on environmental protection and management.

Conclusions

The following conclusions were summarized, after observing the chemical quality of groundwater and associated health problems in respect of NO_3^- and F^- ions at age groups of adults and children, using total water quality index (TWQI) and total human health hazard index (THHHI), from a rural region of Telangana State of India:

➤ The TWQI suggested that the chemical quality of groundwater is suitable for drinking purposes. However, the NO_3^- varying from 0.4 to 585.20 mg/L and F^- from 0.22 to 5.41 mg/L exceed 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 and the fluoride minerals as the prime source of F^- content in the groundwater. They are further supported by principal component analysis.

➤ According to the THHHI, 63% and 73% of the total groundwater samples associated adults and children were more than its safe limit of 1.0 with respect to NO_3^- and F^- , respectively. The intensity of human health risk zones of THHHI was 1.37 times more in

children (5.69) than in children (4.15), covering an area of 71.75% and 66.73%, respectively.

- The present study was recommended effective management measures not only for the protection of groundwater resources from pollution activities but also for improving the health conditions of the locals.

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Captions to Figures

- Fig. 1. Map showing the location of the rural region of the present study
- Fig. 2. Map showing the spatial distribution of (a) NO_3^- and (b) F^- ions
- Fig. 3. Map showing the spatial distribution of total human health hazard index (THHHI) with respect to (a) adults and (b) children

Table 1: Statistical summary of chemical composition of groundwater

Chemical parameters	Minimum	Maximum	Mean	BIS (2012)	Samples exceeding the drinking water quality limits		Analytical methods	Reference
					Percent	Sample numbers		
pH	6.30	8.90	7.14	6.5 - 8.5	3	17, 26, and 79	pH/EC/TDS meter	APHA (2012)
TDS (mg/L)	56	1024	291.0	500	15	1 to 5, 14, 16, 19, 20, 22, 29, 41, 43, 54, and 55.	EC x 0.645	Hem 1991
Ca ²⁺ (mg/L)	8.02	152.30	49.60	75	14	41, 43, 49, 51, 53 to 55, 63, 64, 66, 68, 70, 90, and 98.	EDTA Titrimetric	APHA (2012)
Mg ²⁺ (mg/L)	2.43	92.42	23.53	50	6	3, 55, 91, 94, 97, and 99.	Calculation (TH-Ca ²⁺)	APHA (2012)
Na ⁺ (mg/L)	3	416	54.13	200	1	43	Flame photometric	APHA (2012)
K ⁺ (mg/L)	1	118	6.20	12	10	1, 16, 20, 29, 35, 45, 53, 88, 89, and 96	Flame photometric	APHA (2012)
HCO ₃ ⁻ (mg/L)	146.40	2538	1014.31	300	99	1 to 29, and 31 to 100	Titrimetric	APHA (2012)
Cl ⁻ (mg/L)	17.73	425.40	127.52	250	10	16, 41, 43, 54, 55, 63, 66, 70, 91, and 99.	AgNO ₃ titrimetric	APHA (2012)
SO ₄ ²⁻ (mg/L)	30	166	97.90	200	-	-	UV Visible spectrophotometer	APHA (2012)
NO ₃ ⁻ (mg/L)	0.04	585.20	56.27	45	34	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.	UV Visible spectrophotometer	APHA (2012)
F ⁻ (mg/L)	0.22	5.41	1.13	1.5	25	3, 7, 19 to 25, 28, 34, 36, 38, 39, 40, 42, 43, 50, 56, 59, 61, 64, and 72 to 74.	Ion selective electrode	APHA (2012)

Table 2: Computed values of total water quality index (TWQI), human health hazard quotient (HHHQ) and total human health hazard index (THHHI) (Bold letters denote exceeding the recommended limit)

Sample No.	TWQI		HHHQ _{NO³-}		HHHHQ _{F⁻}		THHHI _{NO³- + F⁻}	
	Value	Classification	Adult	Children	Adult	Children	Adult	Children
1	62	Good	0.81	1.25	0.87	1.34	1.68	2.59
2	53	Good	8.39	12.90	0.78	1.20	9.18	14.11
3	89	Good	4.92	7.57	1.39	2.14	6.31	9.71
4	66	Good	4.61	7.08	0.26	0.40	4.87	7.48
5	60	Good	5.85	9.00	0.35	0.53	6.20	9.53
6	41	Excellent	5.35	8.23	0.61	0.94	5.96	9.16
7	58	Good	0.06	0.10	3.04	4.68	3.11	4.78
8	39	Excellent	2.35	3.62	0.43	0.67	2.79	4.29
9	33	Excellent	3.87	5.96	0.26	0.40	4.13	6.36
10	37	Excellent	3.39	5.21	0.26	0.40	3.65	5.61
11	47	Excellent	1.65	2.54	0.43	0.67	2.08	3.21
12	34	Excellent	1.33	2.05	0.35	0.53	1.68	2.59
13	48	Excellent	4.68	7.19	0.35	0.53	5.03	7.73
14	50	Excellent	2.70	4.15	0.70	1.07	3.39	5.22
15	42	Excellent	0.66	1.02	0.78	1.20	1.45	2.22
16	67	Good	19.08	29.34	1.04	1.60	20.13	30.94
17	62	Good	0.12	0.18	1.04	1.60	1.16	1.78
18	63	Good	7.40	11.38	1.13	1.74	8.53	13.12
19	73	Good	0.20	0.30	1.39	2.14	1.59	2.44
20	70	Good	2.88	4.43	2.00	3.07	4.88	7.51
21	63	Good	0.55	0.85	1.30	2.01	1.86	2.86
22	69	Good	3.44	5.29	2.35	3.61	5.79	8.90
23	80	Good	0.95	1.46	3.57	5.48	4.51	6.94
24	62	Good	1.94	2.98	2.26	3.48	4.20	6.45
25	55	Good	2.91	4.48	2.26	3.48	5.17	7.95
26	50	Excellent	0.14	0.21	0.70	1.07	0.83	1.28
27	40	Excellent	0.04	0.06	0.78	1.20	0.82	1.26
28	63	Good	0.07	0.11	1.65	2.54	1.73	2.65
29	55	Good	0.12	0.18	0.78	1.20	0.90	1.39
30	33	Excellent	3.33	5.12	0.58	0.90	3.91	6.01
31	47	Excellent	0.89	1.37	0.72	1.10	1.61	2.47
32	55	Good	0.18	0.27	1.26	1.94	1.44	2.21
33	58	Good	4.16	6.40	1.07	1.64	5.23	8.04
34	53	Good	1.08	1.66	1.48	2.27	2.56	3.93
35	42	Excellent	0.15	0.23	0.83	1.27	0.98	1.50
36	50	Excellent	0.86	1.32	1.43	2.21	2.29	3.53
37	42	Excellent	0.32	0.49	0.28	0.43	0.59	0.91
38	58	Good	1.14	1.76	1.37	2.11	2.52	3.87
39	59	Good	0.76	1.18	1.63	2.51	2.40	3.69
40	70	Good	0.24	0.37	1.63	2.50	1.87	2.87
41	74	Good	6.76	10.39	0.97	1.48	7.72	11.87

42	73	Good	1.81	2.78	1.90	2.91	3.70	5.69
43	91	Good	6.18	9.51	1.89	2.90	8.07	12.41
44	54	Good	4.25	6.53	0.66	1.02	4.91	7.55
45	60	Good	0.28	0.43	0.71	1.10	0.99	1.53
46	66	Good	1.88	2.89	0.79	1.22	2.67	4.11
47	60	Good	2.96	4.54	0.81	1.24	3.76	5.79
48	60	Good	0.85	1.31	0.95	1.46	1.80	2.76
49	72	Good	3.20	4.92	1.27	1.95	4.47	6.87
50	70	Good	0.36	0.55	1.41	2.17	1.76	2.71
51	54	Good	2.27	3.49	1.03	1.59	3.30	5.08
52	51	Good	3.23	4.96	0.75	1.15	3.98	6.11
53	67	Good	4.86	7.48	0.57	0.88	5.44	8.36
54	78	Good	7.36	11.32	0.50	0.76	7.86	12.08
55	85	Good	11.79	18.13	0.93	1.43	12.72	19.56
56	67	Good	1.07	1.65	1.99	3.06	3.06	4.71
57	44	Excellent	0.98	1.50	0.80	1.23	1.78	2.73
58	42	Excellent	0.62	0.95	1.05	1.62	1.67	2.57
59	61	Good	1.30	1.99	2.73	4.20	4.03	6.19
60	44	Excellent	0.42	0.65	0.57	0.88	1.00	1.54
61	61	Good	0.27	0.42	2.41	3.70	2.68	4.12
62	67	Good	0.13	0.20	1.23	1.90	1.37	2.10
63	70	Good	3.57	5.49	1.13	1.74	4.70	7.23
64	63	Good	0.36	0.55	1.55	2.38	1.91	2.93
65	57	Good	1.46	2.25	1.10	1.68	2.56	3.93
66	63	Good	6.34	9.75	0.85	1.31	7.19	11.06
67	52	Good	3.92	6.02	0.89	1.36	4.80	7.39
68	36	Excellent	0.15	0.23	0.51	0.79	0.66	1.02
69	45	Excellent	0.06	0.10	0.58	0.90	0.65	1.00
70	43	Excellent	0.19	0.30	0.33	0.51	0.52	0.81
71	46	Excellent	0.01	0.01	0.81	1.24	0.82	1.26
72	56	Good	0.02	0.03	1.33	2.05	1.35	2.07
73	72	Good	0.01	0.01	3.13	4.81	3.14	4.82
74	77	Good	0.01	0.01	4.70	7.23	4.71	7.24
75	36	Excellent	0.01	0.02	0.57	0.88	0.58	0.90
76	32	Excellent	0.05	0.07	0.34	0.52	0.38	0.59
77	40	Excellent	0.02	0.04	0.35	0.53	0.37	0.57
78	42	Excellent	0.02	0.04	0.28	0.43	0.30	0.47
79	41	Excellent	0.01	0.01	0.19	0.29	0.20	0.30
80	38	Excellent	0.01	0.01	0.33	0.51	0.34	0.52
81	47	Excellent	0.05	0.07	0.50	0.78	0.55	0.85
82	37	Excellent	0.05	0.08	0.46	0.71	0.51	0.79
83	45	Excellent	0.05	0.07	0.75	1.15	0.79	1.22
84	30	Excellent	0.05	0.08	0.22	0.33	0.27	0.41
85	32	Excellent	0.02	0.02	0.33	0.51	0.35	0.53
86	42	Excellent	0.01	0.01	0.39	0.60	0.40	0.61
87	38	Excellent	0.04	0.06	0.38	0.59	0.42	0.65
88	36	Excellent	0.02	0.03	0.49	0.75	0.51	0.78
89	38	Excellent	0.00	0.00	0.59	0.91	0.59	0.91
90	33	Excellent	0.02	0.03	0.22	0.33	0.24	0.36

91	47	Excellent	0.05	0.08	0.29	0.44	0.34	0.52
92	34	Excellent	0.04	0.06	0.30	0.47	0.34	0.52
93	32	Excellent	0.01	0.02	0.33	0.51	0.34	0.53
94	38	Excellent	0.03	0.05	0.45	0.70	0.48	0.74
95	39	Excellent	0.01	0.01	0.70	1.07	0.71	1.08
96	38	Excellent	0.05	0.08	0.58	0.90	0.63	0.97
97	32	Excellent	0.09	0.14	0.34	0.52	0.43	0.66
98	43	Excellent	0.02	0.03	0.33	0.51	0.35	0.54
99	43	Excellent	0.14	0.21	0.32	0.49	0.46	0.71
100	40	Excellent	0.12	0.19	0.38	0.59	0.50	0.78
Mean	52.72		1.83	2.82	0.99	1.52	2.82	4.34
< 1.0	-	-	0.21	0.17	0.53	0.6	0.54	0.66
>1.0	-	-	4.26	5.68	1.78	2.14	4.15	5.69

Table 3. Mean values of pH, TDS, Na⁺, HCO₃⁻, and Cl⁻ based on the classification of NO₃⁻ and F⁻

NO ₃ ⁻ (mg/L)		TDS (mg/L)	Na ⁺ (mg/L)	Cl ⁻ (mg/L)	Percent of samples
Range	Mean				
<45	9.38	212.13	39.82	112.70	66
45 to 100	74.56	378.67	68.58	115.51	12
>100	186.12	480.00	89.18	178.54	22

F ⁻ (mg/L)		pH	Na ⁺ (mg/L)	HCO ₃ ⁻ (mg/L)	Percent of samples
Range	Mean				
<0.6	0.40	7.02	22.24	863.98	33
0.6 to 1.5	0.98	7.11	62.02	1030.62	43
>1.5	2.42	7.30	83.83	1099.78	24

Table 4. Principal component analysis

Chemical parameters	Principal Component Analysis			
	1	2	3	4
pH	0.302	0.519	0.316	0.201
TDS	0.851	0.055	0.317	0.094
Ca ²⁺	0.439	-0.125	-0.585	-0.026
Mg ²⁺	0.364	-0.560	0.043	0.543
Na ⁺	0.660	0.552	-0.010	0.130
K ⁺	0.469	0.030	0.404	-0.634
HCO ₃ ⁻	0.224	0.555	-0.449	0.339
Cl ⁻	0.768	-0.267	-0.023	-0.113
SO ₄ ²⁻	-0.195	-0.150	0.654	0.430
NO ₃ ⁻	0.813	0.043	0.166	-0.113
F ⁻	-0.033	0.732	0.176	0.107
Eigenvalue	3.14	1.75	1.40	1.11
% Total variance	28.50	15.94	12.69	10.07
Cumulative %	28.50	44.44	57.13	67.20

Fig. 1. Map showing the location of the rural region of the present study

Fig. 2. Map showing the spatial distribution of (a) NO₃⁻ and (b) F⁻ ions

Fig. 3. Map showing the spatial distribution of total human health hazard index (THHHI) with respect to (a) adults and (b) children