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THEMATIC ISSUE

Hydrogeochemistry and Health Risk Assessment of groundwater and surface water in fluoride affected area of Yadadri-Bhuvanagiri District, Telangana State, India

Shekhar More[1,2] · **Ratnakar Dhakate**[1,2] · **G. Venkata Ratnalu**[1,2] · **G. Machender**[3]

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Abstract

Water is an indispensable and crucial component of the life sustenance system. According to the Central Ground Water Board (CGWB), groundwater in 276 Districts in 20 States is contaminated with Fluoride. Considering the fact that Yadadri-Bhuvanagiri district's majority depend on groundwater for drinking. Fluoride a menace to this source is a cause for concern. This study was carried out to understand the fluoride contamination and its health risk assessment. For this study 47 (28 groundwater, 19 surface water) and 45 (28 groundwater, 17 surface water) samples were collected during pre and post-monsoon seasons respectively and were analysed for fluoride and other major ions. The water quality analysis data shows a higher concentration of fluoride in groundwater and surface water samples which ranges from 0.43–2.93 to 0.37–3.48 mg/L for pre-monsoon while 0.60–3.56 mg/L and 0.90–3.21 mg/L for post-monsoon seasons, respectively. Among the collected samples about 46.80% and 51.11% samples of pre- and post-monsoon exceeded the permissible limit of fluoride. The water quality data and sources of the dissolved constituent were analyzed by Piper Trilinear Diagram, Gibbs Plot, and Chloro-Alkaline Indices. Besides these, Principal Component Analysis (PCA) and health risk assessments were carried out for different age groups. PCA result shows that the water chemistry is controlled by geogenic activities. The health risk assessment results divulged the hazard quotient via ingestion (HQ_{ing}) had a higher chronic hazard than the dermal pathway. Pre-monsoon HQ_{ing} percentage values of groundwater and surface water for the age group of 6–12 months are 92.85 and 97.73, respectively, and all the samples of post-monsoon have HQ_{ing} values greater than 1.

Keywords Health Risk Assessment (HRA) · Fluoride contamination · Principle component analysis (PCA) · Chinneru River Basin (CRB)

Introduction

Access to fresh and clean water for ingestion, cooking, domestic use, and some other reasons is an essential part of public well-being. Lacking the quantity and quality of water resources, expose individuals to preventable health hazards. About 2 billion people depend on water sources that are contaminated with faecal material for drinking purposes (Willi 2018). Groundwater is considered less contaminated than surface water because it has less contact with the external environment and also it undergoes natural filtration during percolation through the soil zone (Elumalai et al. 2017). The groundwater quality of an area contingent upon anthropogenic and natural factors like precipitation of minerals, dissolution, the interaction of groundwater with rock and soil, and the residence time (Batabyal and Gupta 2017). Fluoride (F^-) is a natural constituent of groundwater because of the

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✉ Shekhar More
shekharmore4444@gmail.com

[1] Academy of Scientific and Innovative Research (AcSIR), Ghaziabad 201 002, India

[2] CSIR-National Geophysical Research Institute, Uppal Road, Hyderabad 500 007, India

[3] Mahatma Gandhi University, Nalgonda 508 254, India

higher interaction of water with the rock in the aquifer (Hossain and Patra 2020).

Fluorine is a common element in nature but does not occur in the elemental state due to its high reactivity and is the most electronegative and reactive element in the halogen group (CGWB 2018; Hossain and Patra 2020). With the chemical analysis, we can determine the presence of fluoride but it is not easy with physical examination because it does not have smell, taste, or colour. Fluoride in the human body is required to make the bones and teeth strong but at a certain limit so there has been given a specific guideline by the World Health Organization (WHO). The permissible limit of fluoride concentration in the drinking water is 1.5 mg/L (WHO 2004; BIS 2012). The excess intake of Fluoride leads to skeleton deformation disease fluorosis. Densely populated countries like India and China are affected due to high dental and skeletal called fluorosis (Bera and Ghosh 2019).

Globally high occurrence of fluoride concentration in groundwater was reported in many countries like India, China, Italy, Iran, Bangladesh, Japan, Sri Lanka, Brazil, Canada, Norway, Turkey, Kenya, Jordan, Nigeria, Germany, and Mexico (Ayoob and Gupta 2006; Ali et al. 2016; Sudheer Kumar et al. 2017; Adimalla 2019; Bera and Ghosh 2019). Fluoride contamination in India was first detected in the Nellore district of Andhra Pradesh in the year 1937. About 62 million people all over the country suffer from fluorosis because of drinking water contaminated with high fluoride concentration (Srinivasamoorthy et al. 2012; Annadurai et al. 2014; Farooq et al. 2018; Adimalla et al. 2019). In the state of Telangana, nine districts are affected by fluoride contamination (CGWB 2018). Most of the region of Telangana is occupied by granite rock, which is rich in fluoride minerals is the main source of fluoride in groundwater (Sreedevi et al. 2006; Annadurai et al. 2014; Machender et al. 2014; Satyanarayana et al. 2017; Laxman Kumar et al. 2019; Adimalla et al. 2019). It is the second most fluoride-affected state in India. Nalgonda district is the most fluoride-contaminated area in the Telangana state. A fluoride concentration of 20 mg/L has been reported in Nalgonda (Ali et al. 2016; Farooq et al. 2018).

In recent years, there has been an increasing concern regarding the quality of groundwater and its impact on different age groups such as children to older adults. As a result, the United States Environmental Protection Agency (US-EPA) has developed policies, guidelines, and human health risk assessment (HRA) model (US-EPA 2011) to estimate and interpret the consequence of water on human health via different pathways by applying information from analytical data. Therefore, it is being widely adopted by researchers all over to delineate the adverse effects of fluoride and other contamination on human health (Ayoob and Gupta 2006; Narsimha and Rajitha 2018; Karunanidhi et al. 2020; Zango et al. 2019; Yuan et al. 2020).

The present study was carried out in the semi-arid region of Chinneru River Basin (CRB) in the Yadadri Bhuvanagiri district. In this context, the objectives of the study were to determine fluoride contamination in groundwater and surface water and the consequences of high fluoride concentration on human health via ingestion and dermal pathways. To know the health risk for different ages they are divided into seven age groups and they are 6–12 months, 6–11 years, 11–16 years, 16–18 years, 18–21 years, ≥ 21 years, and ≥ 65 years (Adimalla and Qian 2019) using the United States Environmental Protection Agency's Method.

Study area

Location

The Chinneru River Basin is located about 30 km away from the East of Hyderabad. The study area occupies the Survey of India's Toposheet No. 56K/15 and covers an area of 250 km^2 (Fig. 1). Some of the main towns covered in the area are Bibinagar in the north-west, Nandanam in the central and Sangem in the south of Yadadri Bhuvanagiri District, Telangana. The area slopes gently from the north-west to the direction of the south-east. The maximum elevation in the study area is in the north at about 450 m near Gudur and the lowermost is in the south which is about 340 m near Bollepalli.

Drainage and climatic condition

The Chinneru River Basin area is underlain by homogeneous material of granitic rock and the undulating topography makes it to be a dendritic type of drainage pattern with third-order streams. The Sharmirpet lake outflow originating from the Sharmirpet catchment area in the north after joining other streams grows into Chinneru River. Nearly 24 villages and twelve tanks are suffusing the complete drainage basin. Chinneru River flows from Gudur in the north and finally meets the Musi River near Sangem village in south of Yadadri Bhuvanagiri District.

The study area is located in the semi-arid region. It is mostly hot during summer, the rise in temperature is noticed from February month and it ranges maximum of about 40–45 °C during May. While the decrease in temperature is observed after May till December. The temperature during the winter ranges from 10 to 15 °C. The onset of monsoon (southwest monsoon) arrives in the first week of June till October. The highest rainfall occurs during the southwest monsoon between the months of June–September. The average yearly precipitation is 900 mm from both southwest and northeast monsoon. During the monsoon season, the study area also experiences high humidity.

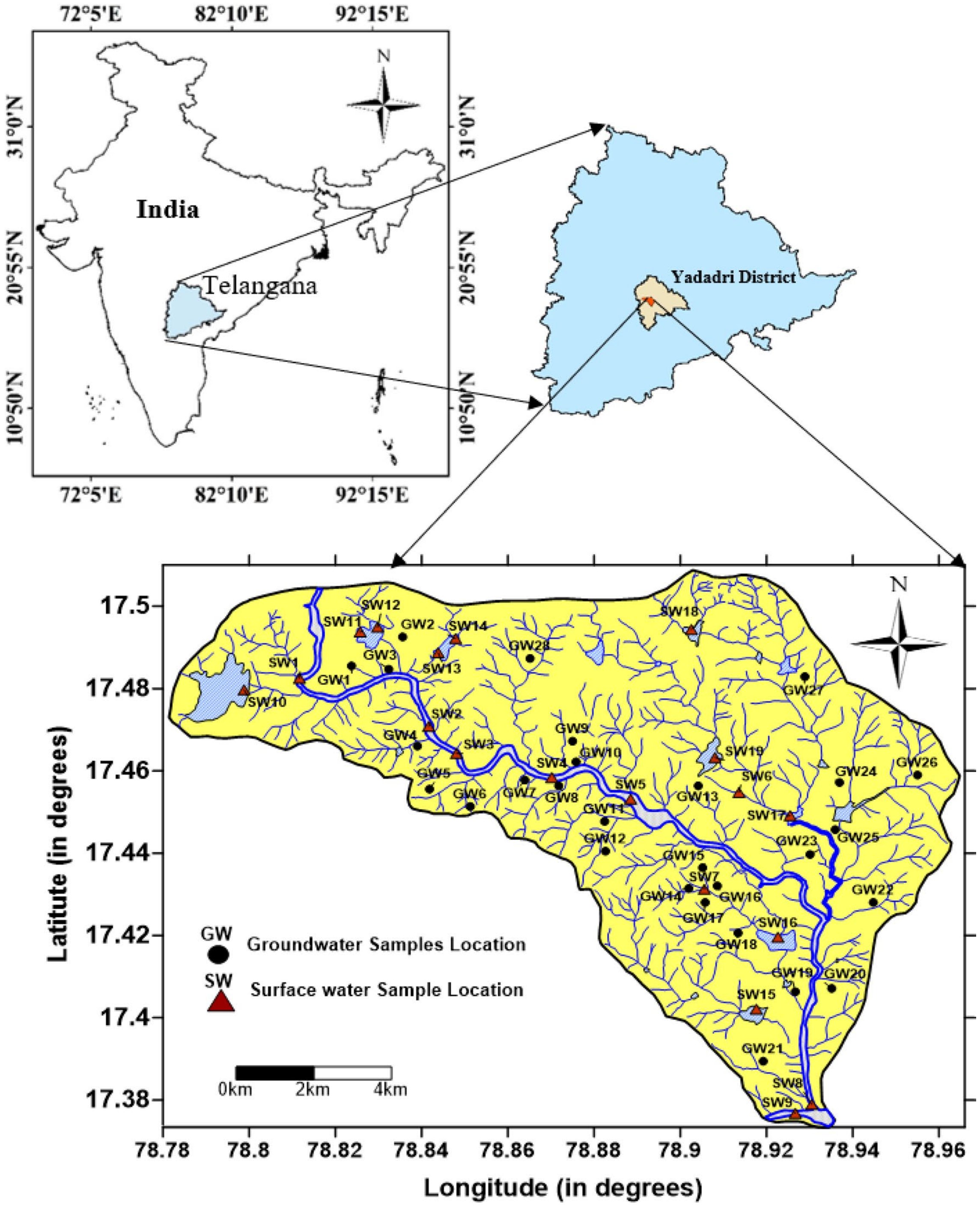


Fig. 1 Location map of study area showing sample locations

Geology of the area

The investigated area is in hard rock terrain of the northeastern part of Eastern Dharwar Craton (EDC). Pre-Cambrian granite rocks of grey and pink colour are covered in this region. The rocks with the composition of granite-granodiorite-tonalite gneiss are intruded by veins of pegmatite, quartz, dolerite dykes, and gabbro (Fig. 2). These rocks are generally hard, impermeable, and also non-porous. The conspicuous soil horizons in the study area are mostly red, black, and loamy soils. Red soils are present in the dry highland areas. These soils are formed as a result of the weathering of granitic rocks and mafic rocks.

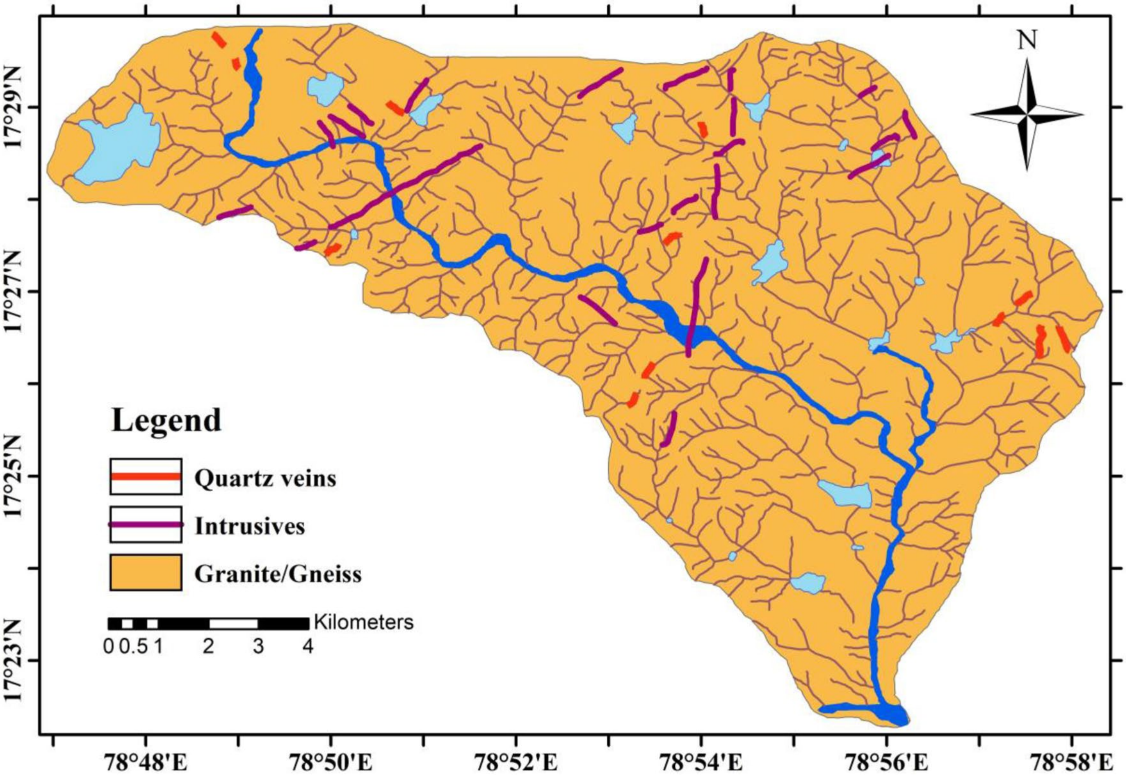


Fig. 2 Geological map of the study area

Materials and methods

Sampling

To assess water chemistry altogether 47 (28 groundwater, 19 surface water) and 45 (28 groundwater, 17 surface water) water samples were collected for pre-monsoon and post-monsoon seasons respectively. New polyethylene bottles of 1000 ml were used to collect the samples. Samples collected were from a combination of open wells, dug well, hand pumps, streams, and tanks. While collecting samples, the source was purged for 10 min, and bottles were three times rigorously rinsed with a water sample to be collected. Field filtration with the help of filter papers was carried out to eliminate suspended solids and then filled, closed to air-tight with bungs, and labelled. The parameters like hydrogen ion concentration (pH), Total Dissolved Solids (TDS), and Electrical conductivity (EC) were measured at each sampling location with the help of a portable pH/EC/TDS meter. Geographic coordinates of the sampling spots were noted by a GPS device and taken to laboratory at the CSIR-NGRI, Hyderabad.

Analytical techniques

The pre and post-monsoon samples brought to the CSIR-NGRI laboratory were analysed for major elements (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, HCO_3^-,CO_3^{2-}, NO_3^-, F^-) using standard procedures suggested by American Public Health Association (APHA 1995). The concentration of major ion Sulphate (SO_4^{2-}) was determined by the turbidity method using the Digital Nephelo-Turbidity meter 132 model of Systronics. Nitrate (NO_3^-) was determined by the spectrophotometric method using UV–visible spectrophotometer UV-1201 model of Shimadzu. Na^+ and K^+ were analysed by using the CL345 flame photometer of ELICO. Fluoride was analysed by the ion-selective electrode method using the Orion 290A + model of Thermo-electron Corporation. The classical method of analysis was used to determine Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Chloride (Cl^-), and Carbonate (CO_3^{2-}). The titration method was used for the bicarbonates. Replicate analysis was carried out on each sample to determine reproducibility and quality assurance. The precision of the analysis was cross-checked by calculating ionic balance error which is about ± 5. Used reagents were of analytical grade and all instruments prior to measurement were pre-calibrated.

Moreover, the human health risk assessment model for different age groups introduced by the United States Environmental Protection Agency was used to calculate the hazard quotient via different pathways, Gibb's Plot, Piper Trilinear Diagram, Chloro-alkali indices- (CAI-I and CAI-II) graphs were used to ascertain all the aspects which are changing the geochemistry of water and aquifer constituents to know its appropriateness.

Results and discussion

The statistical summary of the analytical results of pre and post-monsoon season samples for groundwater and surface water are shown in (Table 1). The groundwater and surface water are alkaline in nature. The pH of groundwater samples of pre and post-monsoon season ranges from 7.36 to 10.16 and 7.1–8.0 with mean values of 7.76 and 7.40 respectively. The pH range of surface water samples is 7.79–10.12 and 7.5–9.6 with a mean of 8.80 and 8.45 for pre and post-monsoon seasons respectively. EC of groundwater ranges from 204–1946 to 548.2–2472 µS/cm with mean values of 1061.35 µS/cm and 1363.47 µS/cm for pre and post-monsoon seasons, while the EC of surface water samples has a large variation ranging from 190.1–1709 to 330.7–2460 µS/cm with a mean of 636.96 µS/cm and 903.82 µS/cm for both the seasons. As per the TDS values, groundwater and surface water samples are classified as freshwater and very few samples with brackish water nature for both seasons. The TDS range for groundwater is 122.4–1167.6 mg/L and 304.1–1367 mg/L with a mean of 625.05 mg/L and 764.63 mg/L for pre and post-monsoon seasons respectively. Surface water TDS ranges for pre and post-monsoon seasons are 100.8–1025.4 mg/L and 188.5–1375 mg/L with mean values of 319.02 mg/L and 506.42 mg/L, respectively (Table 1).

The minimum, maximum and mean value concentration of Na^+, K^+, Mg^{2+}, Ca^{2+} (Cations) and Cl^-, SO_4^{2-}, HCO_3^-, CO_3^{2-}, NO_3^-, F^- (anions) obtained from the results of groundwater and surface water for both the season are summarized in Table 1. The mean dominance of cation in groundwater and surface water is $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ for both the seasons. The mean dominance of anions in groundwater for both the seasons is $HCO_3^- > Cl^- > NO_3^- > SO_4^{2-} > CO_3^{2-} > F^-$, whereas surface water anion concentration is $HCO_3^- > Cl^- > CO_3^{2-} > SO_4^{2-} > NO_3^- > F^-$ and $HCO_3^- > Cl^- > SO_4^{2-} > CO_3^{2-} > NO_3^- > F$ for pre and post-monsoon seasons respectively.

Fluoride distribution in the area

The results of fluoride concentration in the study area for pre and post-monsoon seasons of groundwater and surface water are presented in (Table 1). The fluoride concentration in groundwater ranged from 0.4–2.9 to 0.6–3.6 mg/L for pre and post-monsoon seasons with mean values of 1.5 mg/L and 1.7 mg/L, respectively. The permissible limit of fluoride in drinking is 1.5 mg/l (BIS 2012; WHO 2004). The groundwater and surface water samples of pre-monsoon having

Table 1 Statistical details of analytical data for groundwater and surface water sample for pre and post-monsoon seasons

Parameters	Pre-monsoon						Post-monsoon					
	Min		Max		Mean		Min		Max		Mean	
	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW
pH	7.36	7.79	10.16	10.12	7.76	8.80	7.1	7.5	8	9.6	7.40	8.45
EC (μ S/cm)	204	190.1	1946	1709	1061.35	636.96	548.2	330.7	2472	2460	1363.47	903.82
TDS	122.4	100.8	1167.6	1025.4	625.05	319.02	304.1	188.5	1367	1375	764.63	506.42
Na^+	37.01	21.81	476.39	280.31	164.38	129.19	35.80	44.76	428.02	466.84	175.52	157.24
K^+	1.71	1.34	116.48	42.69	10.12	8.34	0.70	1.35	113.88	15.54	11.87	6.21
Mg^{2+}	7.89	3.03	104.55	43.14	45.31	22.97	14.88	11.07	147.04	86.01	44.51	31.13
Ca^{2+}	6.51	6.04	234.64	75.13	67.96	24.89	28.60	9.03	181.75	95.04	80.56	39.38
Cl^-	11.27	11.32	710.99	428.98	206.36	105.67	13.81	27.36	521.45	496.21	189.41	137.70
SO_4^-	2.68	5.99	126.68	79.73	70.57	26.96	12.83	8.09	151.27	137.79	68.31	34.36
HCO_3^-	97.6	85.4	539.85	491.05	263.52	288.94	283.65	140.3	768.6	786.6	442.57	330.35
CO_3^{2-}	9	0	69	66	31.67	27.68	0	9	36	60	3.53	30.47
NO3$^-$	8.26	0	394.53	92.94	74.17	5.85	0	0	506.67	55.75	79.09	3.43
F^-	0.43	0.37	2.93	3.48	1.53	1.93	0.60	0.90	3.56	3.21	1.72	1.77

All the values are in mg/L except pH and EC

GW ground water, *SW* surface water

concentration < 0.5 mg/L are 7.14% and 5.26% respectively, ingestion of such water for a longer time promotes dental caries. The values of fluoride concentration in the surface water is ranged from 0.4–3.5 to 0.9–3.2 mg/L in the pre and post-monsoon seasons with mean values of 1.9 mg/L and 1.8 mg/L respectively (Table 1).

The concentration of Fluoride in groundwater and surface water were divided into three groups. Group-1 (concentration < 1 mg/L) is below permissible limit, Group-2 (concentration 1–1.5 mg/L) is within the permissible limit and Group-3 (concentration > 1.5 mg/L) is greater than the permissible limit. Groundwater samples of pre and post-monsoon seasons falling in Group-1 (concentration < 1 mg/L) is 32.14% and 21.42% of samples respectively (Table 2). Similarly, in Group-2 (concentration 1–1.5 mg/L) the values are 28.57% and 35.71% of samples of pre and post-monsoon and they are in the permissible range of fluoride for drinking standards (Table 2). Group-3 (concentration > 1.5 mg/L) have 39.28% and 42.85% in pre and post-monsoon seasons, respectively (Table 2) and ingestion of such high fluoride concentration of water can cause skeletal fluorosis (Ali et al. 2016; Kanduti et al. 2016; Toumba et al. 2019). Similarly, for surface water samples for pre and post-monsoon seasons, they are 10.52% and 11.76%; 31.57% and 29.41%; 57.89% and 58.82% for Group-1, Group-2, and Group-3 respectively. The spatial distribution of fluoride concentration in groundwater and surface water samples for pre and post-monsoon season is dissimilar due to the uneven distribution of the abundance of fluoride-bearing minerals in the study area (Fig. 3a and b). The pre-monsoon figures show high fluoride concentration (> 1.5 mg/L) in the north, north-west, and in the southern part, while the permissible concentration (1–1.5 mg/L) is noticed in the central as well as in the eastern region of the study area. The eastern part which was in the permissible limit during pre-monsoon season has turned out to have high fluoride concentration in the post-monsoon season due to the weathering mineral dissolution (Fig. 3b).

Hydrochemical facies analysis

Piper's trilinear diagram is an effectual method to know the hydrogeochemical regime of the study area by plotting the concentration of the major ions in the Piper diagram (Piper 1944). The diagram has two triangles at the base. The left triangle outlines the cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}), right triangle as anions (CO_3^{2-}, HCO_3^{-}, SO_4^{2-}, Cl^{-}) and different water type can be known by their position in rhombus (diamond) shaped field. The diamond-shaped field is divided into six types: (1) $CaCO_3$-type, (2) NaCl-type, (3) mixed $CaNaHCO_3$-type, (4) mixed CaMgCl-type, (5) CaCl-type, (6) $NaHCO_3$-type (Fig. 4).

In the study region, most of the groundwater samples of pre and post-monsoon seasons show alkaline earth

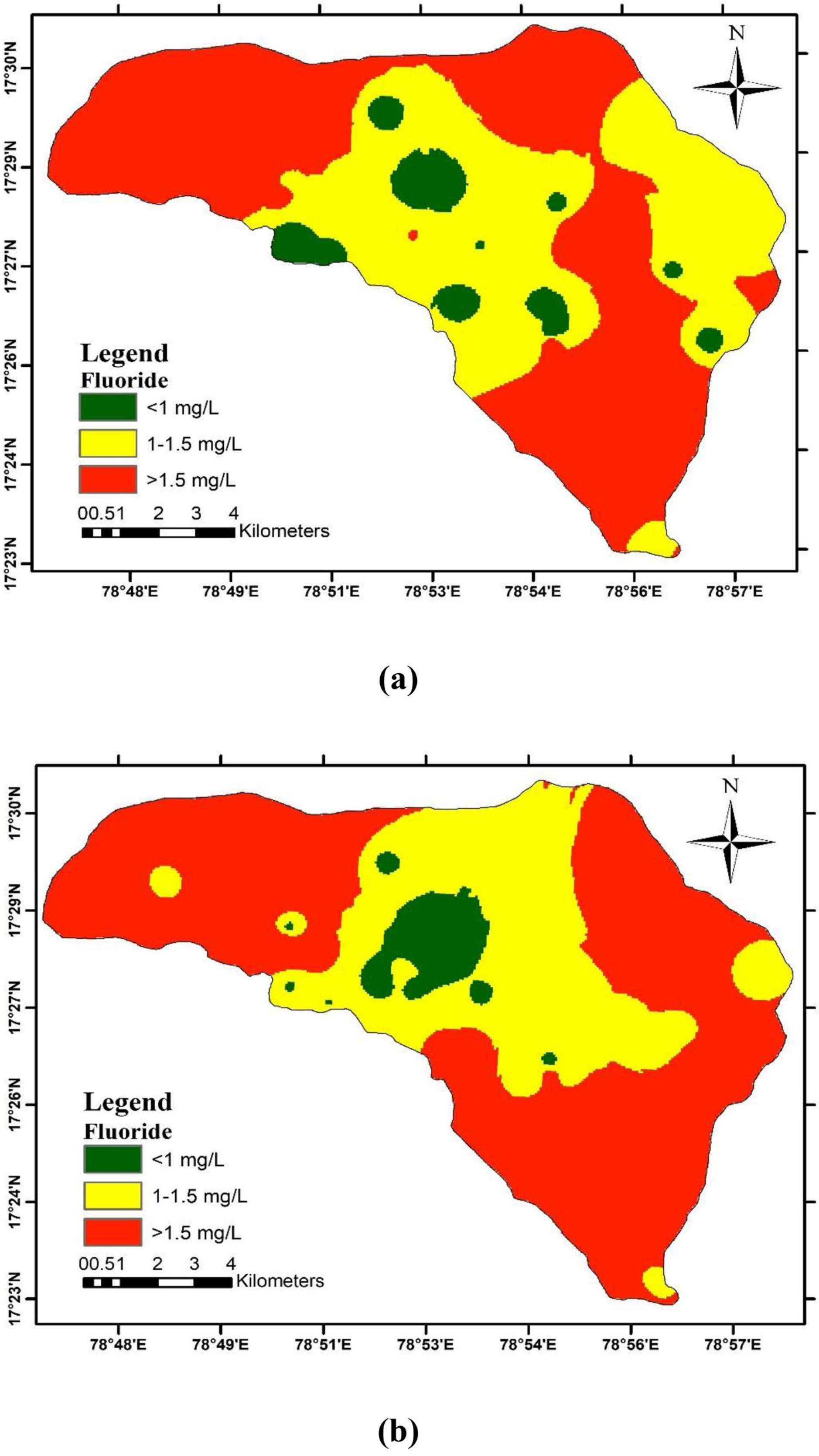
Table 2 Statistical summary based on fluoride concentration of ground and surface water for pre and post-monsoon seasons

Group	F conc. mg/L	Pre-monsoon										Post-monsoon									
		A		*B*		Minimum		Maximum		Mean		*A*		*B*		Minimum		Maximum		Mean	
		GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW	GW	SW
Group-1	< 1	9	2	32.14	10.52	0.4	0.4	0.9	0.9	0.7	0.6	6	2	21.42	11.76	0.6	0.9	0.9	0.9	0.8	0.9
Group-2	1–1.5	8	6	28.57	31.57	1.0	1.0	1.4	1.4	1.3	1.3	10	5	35.71	29.41	1.0	1.0	1.5	1.4	1.2	1.3
Group-3	> 1.5	11	11	39.28	57.89	1.6	1.6	2.9	3.5	2.4	2.5	12	10	42.85	58.82	1.6	1.6	3.6	3.2	2.7	2.2

A: no. of samples, B: % of samples

GW groundwater, *SW* surface water

Fig. 3 Spatial distribution map of fluoride **a** pre-monsoon season, **b** post-monsoon season



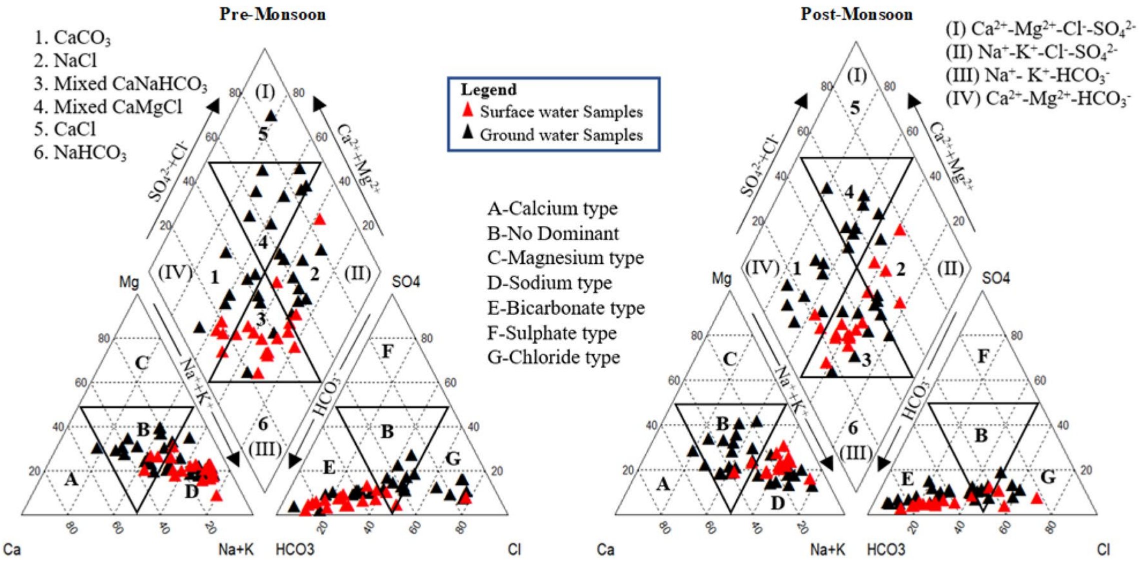


Fig. 4 Piper Trilinear diagrams of groundwater and surface water samples for Pre- and Post-monsoon season

($Ca^{2+} + Mg^{2+}$) exceeding alkalies ($Na^+ + K^+$). Strong acids ($SO_4^{2-} + Cl^-$) exceeds weak acids ($CO_3^{2-} + HCO_3^-$) for groundwater samples of pre-monsoon, while weak acids ($CO_3^{2-} + HCO_3^-$) exceed strong acids ($SO_4^{2-} + Cl^-$) for groundwater samples of post-monsoon (Fig. 4). The pre and post-monsoon samples of surface water show alkalies exceeding alkaline earth and weak acids exceed strong acids. It is also observed that most of the groundwater and surface water samples of both the seasons are in no dominant zone where the type of groundwater cannot be identified either as anion or cation dominant.

Among the samples of pre-monsoon, most of the groundwater samples are of NaCl-type, Mixed CaMgCl-type, and $CaCO_3$-type, while surface water samples are dominant in Mixed $CaNaHCO_3$-type and $CaCO_3$-type. Similarly, post-monsoon samples of surface water are dominant in the Mixed $CaNaHCO_3$-type and few in NaCl-type, but samples of groundwater are falling in dominance of Mixed CaMgCl-type, $CaCO_3$-type and Mixed $CaNaHCO_3$-type. The HCO_3^- and Na^+ ions are high in concentration because the entire area is of granitic hard rock terrain where silica minerals are generally present which leads to increasing the HCO_3^- and Na^+ concentrations in water (Sakram and Adimalla 2018; Adimalla and Qian 2019). This shows that most of the groundwater samples are unsuitable for drinking purposes.

Water–rock interaction (Gibbs's Plot)

Based on the chemical analytical data for pre and post-monsoon seasons of groundwater and surface water, the important three mechanisms controlling the chemistry are atmospheric precipitation, rock dominance, and evaporation crystallization process (Gibbs 1970). Gibbs plots are the ratio of cations [($Na^+ + K^+$)/($Na^+ + K^+ + Ca^{2+}$)] and anions [Cl^-/($Cl^- + HCO_3^-$)] against TDS. Gibbs plot states that the majority of samples from both the seasons of the study area fall in the rock dominance region followed by evaporation dominance (Fig. 5a and b). The extent of rock dominance interaction depends on factors like residence time of water in aquifer, region occupied by hard rock terrain, low rainfall, and high temperature. Few samples in evaporation dominance are due to an increase in Na^+ and Cl^- ions and an inferable increase in TDS (Gupta et al. 2009; Krishna Kumar et al. 2014; Madhav et al. 2018; Adimalla and Qian 2019; Kumara et al. 2020).

Chloro-Alkaline Indices (CAI)

The chloro-alkaline indices (CAI) are used to know the ion exchange process. The ion exchange process within the groundwater and the host rock is one of the important responsible process for the concentration of ions in groundwater (Scholler 1967). The computation of chloro-alkaline indices (CAI-I and CAI-II) and concentration of cations and anions is done by using the equations given below

$$\mathrm{CAI-I} = \frac{\mathrm{Cl^-} - \left(\mathrm{Na^+} + \mathrm{K^+}\right)}{\mathrm{Cl^-}}, \tag{1}$$

$$\mathrm{CAI-II} = \frac{\mathrm{Cl^-} - \left(\mathrm{Na^+} + \mathrm{K^+}\right)}{\left(\mathrm{SO_4^{2-}} + \mathrm{HCO_3^-} + \mathrm{CO_3^{2-}}\mathrm{NO_3^-}\right)}. \tag{2}$$

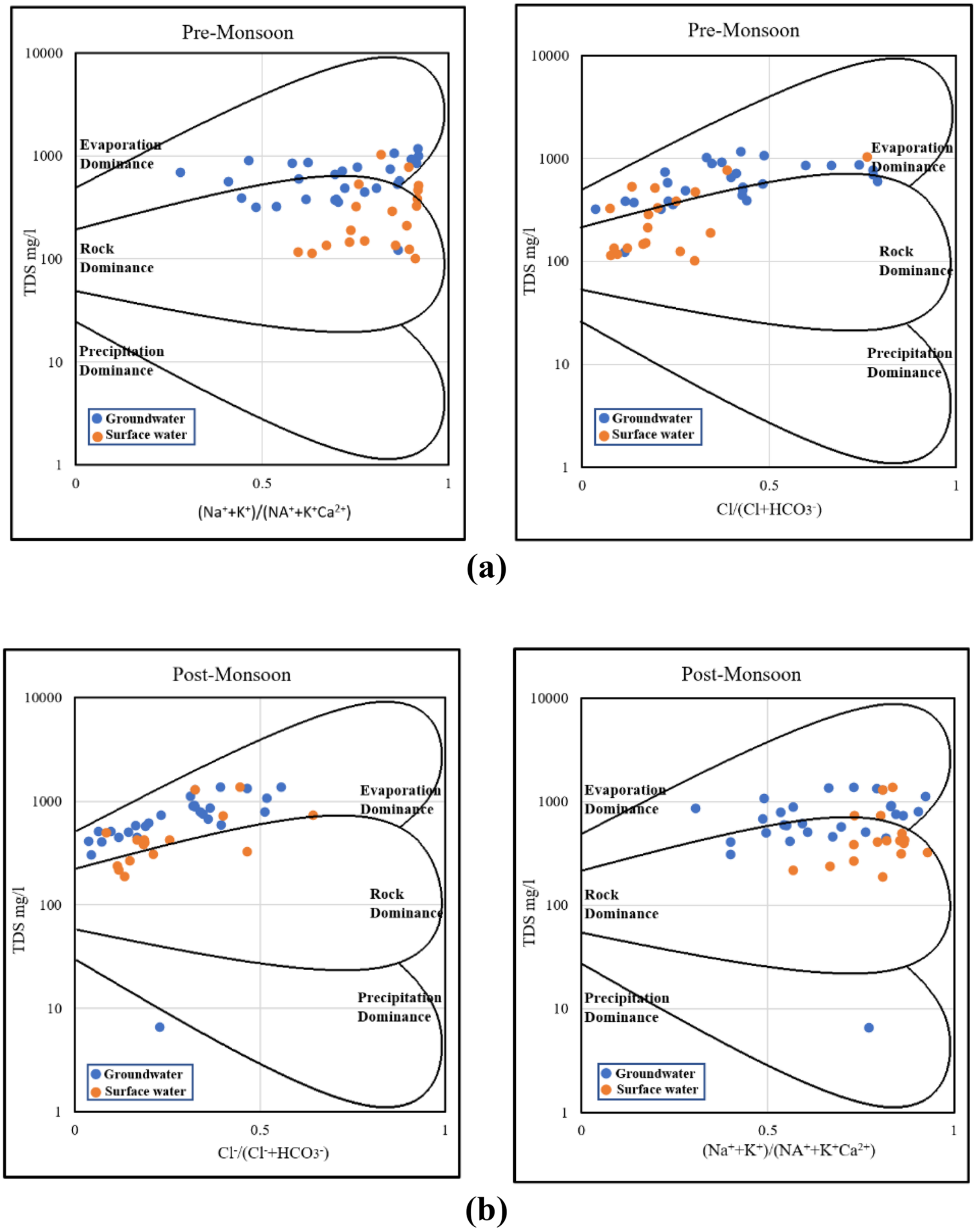
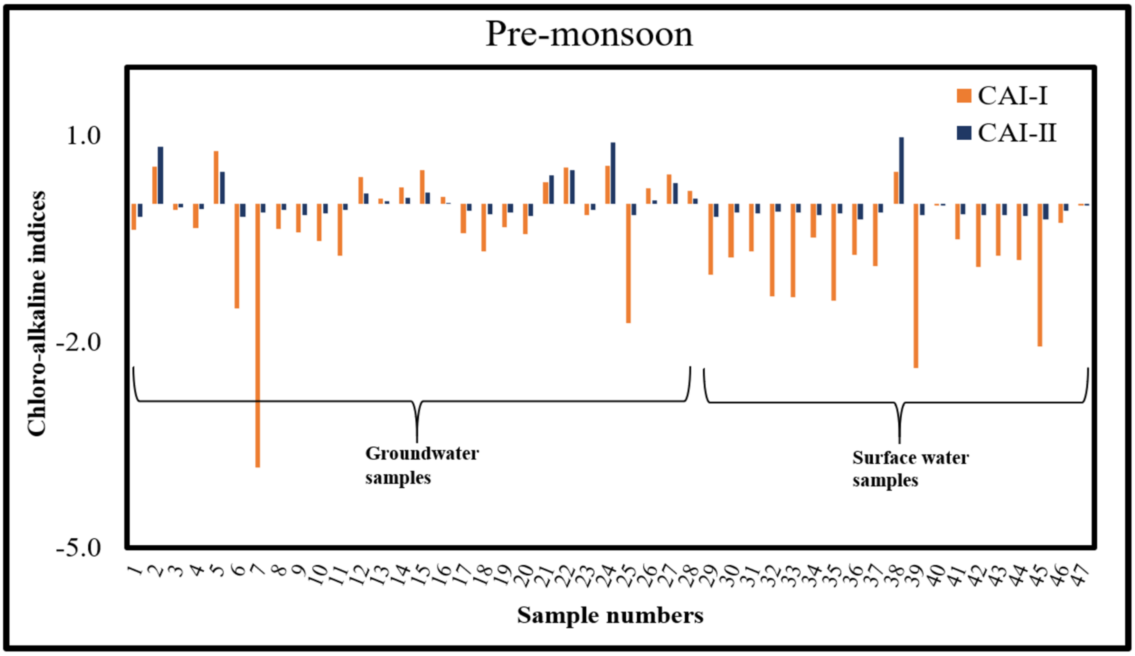
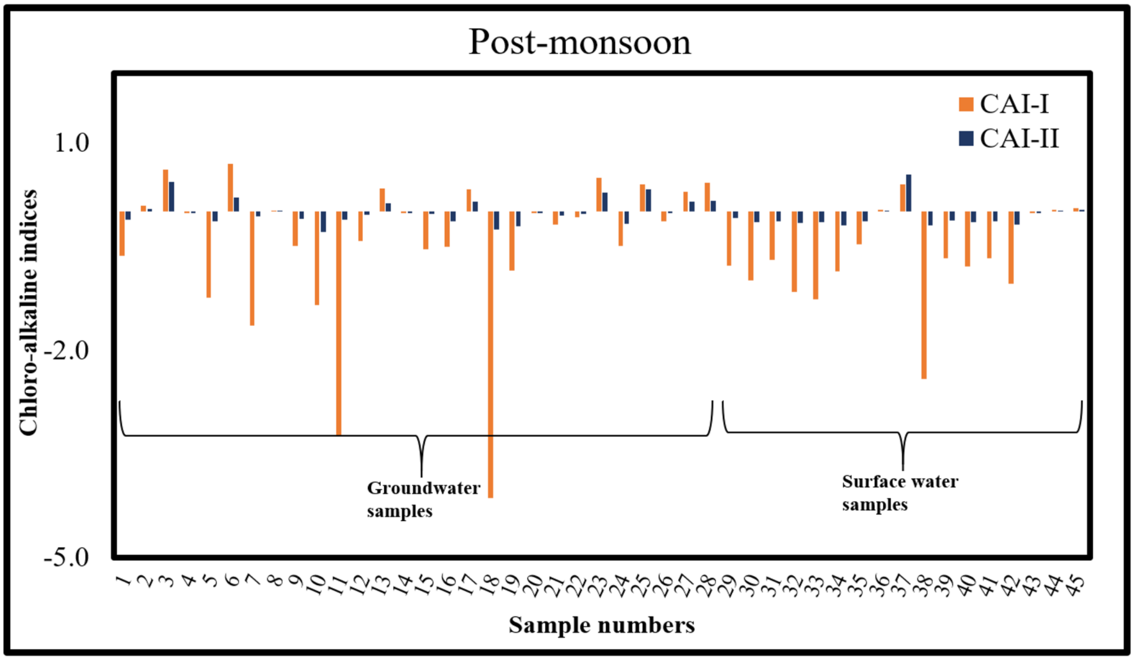


Fig. 5 Mechanism controlling chemistry of water (Gibbs Plot) for groundwater and surface water samples **a** pre-monsoon season, **b** post-monsoon season

During ion exchange, when there are Ca^{2+} and Mg^{2+} ions present in water are exchanged with Na^+ and K^+ ions in aquifer material, the indices value is negative, representing cation–anion exchange reaction or chloro-alkaline disequilibrium and the concentration of Na^+ and K^+ will increase in water. If Na^+ and K^+ ions in water exchange with Ca^{2+} and Mg^{2+} ions in aquifer material, then the indices value will be positive, representing direct base-exchange reaction or chloro-alkaline equilibrium and the concentration of Ca^{2+} and Mg^{2+} will increase in water. The results plotted in the



(a)



(b)

Fig. 6 Chloro-alkaline indices (CAI-I and CAI-II) for groundwater and surface water samples **a** pre-monsoon season, **b** post-monsoon season

bar diagram are nearly 70% of pre-monsoon and 69% of post-monsoon samples (CAI-I and CAI-II) shows negative results with the cation–anion exchange reaction (Fig. 6a and b).

Principal component analysis (PCA)

Principal component analysis is obtained by initial Eigen values and percentage of the variance for the groundwater and surface water chemical data of pre and post-monsoon season. The dimensionality is reduced from the 13 original physical and chemical parameters to two, three, and four factors by using Statistical Package for Social Sciences (SPSS 23) software (Table 3a and b). Factor analysis was applied to the varimax normalized data to relate the compositional patterns between the analysed water samples and to identify the factors that impact each one. The factors loading are classified as strong (high), moderate and weak corresponding to absolute loading values of > 0.75, 0.75–0.50, and 0.50–0.30, respectively. PCA and multivariate statistical approaches are very useful tools for understanding fluoride and nitrate concentration in water and groundwater quality assessment

Table 3 (a) Principal component analysis on groundwater and surface water data in pre-monsoon season, (b) Principal component analysis on groundwater and surface water data in post-monsoon season

(a)

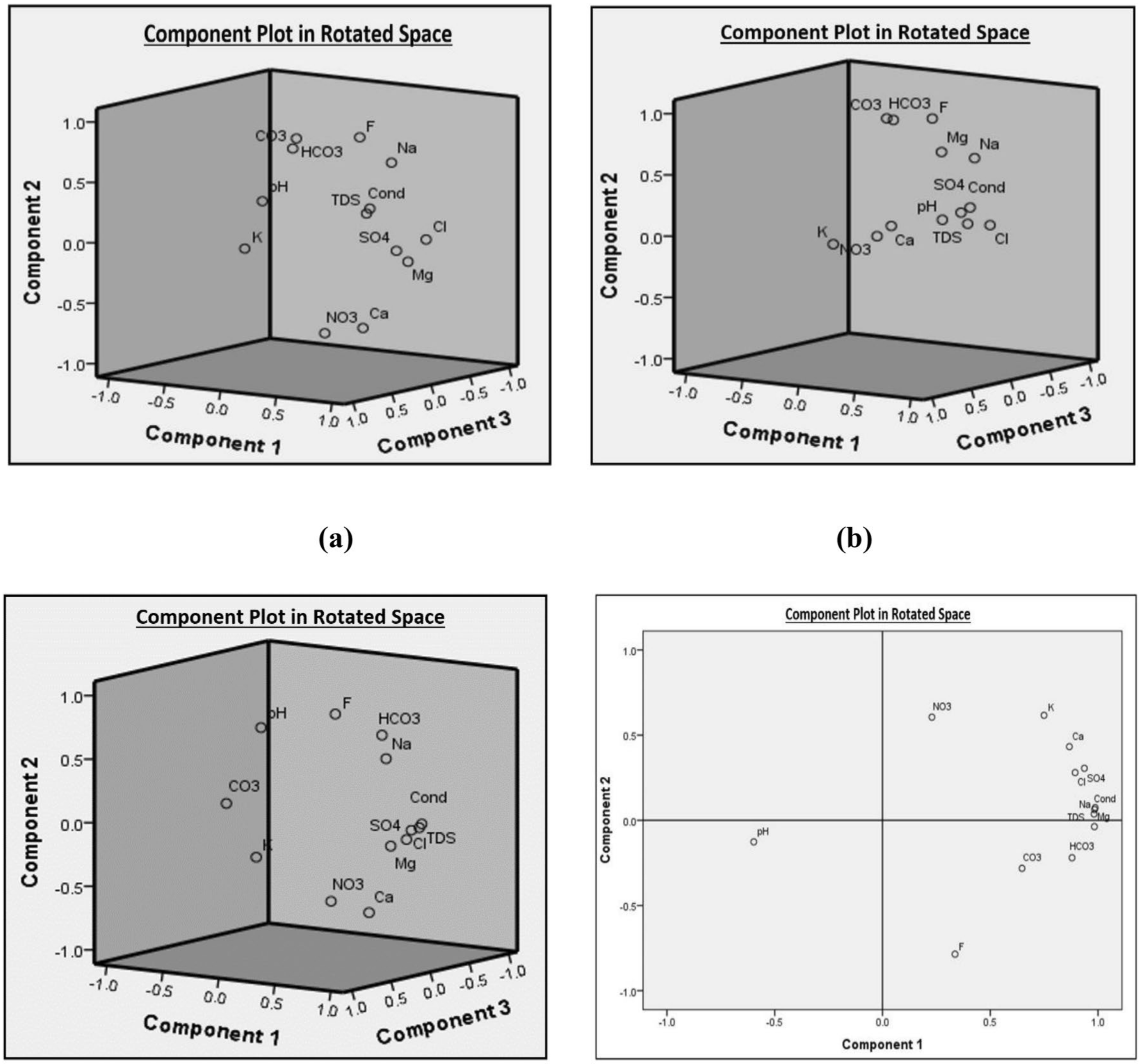
Variables	Component						
	Groundwater			Surface water			
	1	2	3	1	2	3	4
pH	−0.521	0.219	−0.170	−0.071	−0.040	−0.819	0.021
EC	0.767	0.355	0.294	0.855	0.269	0.266	0.274
TDS	0.762	0.319	0.328	0.872	0.171	0.205	−0.270
Na^+	0.762	0.319	0.328	0.691	0.638	− 0.141	0.244
K^+	0.057	0.037	0.881	0.064	0.003	0.762	0.190
Mg^{2+}	0.839	−0.133	−0.093	0.609	0.724	0.163	0.160
Ca^{2+}	0.513	−0.699	0.018	0.466	0.169	0.598	0.558
Cl^-	0.887	0.032	−0.257	0.956	0.144	0.041	0.197
SO_4^-	0.896	−0.004	0.136	0.678	0.240	− 0.103	0.596
HCO_3^-	0.030	0.845	0.181	0.168	0.938	0.145	0.187
CO_3^{2-}	0.084	0.785	0.304	0.138	0.954	0.188	0.028
NO_3^-	0.429	−0.697	0.388	0.072	−0.010	0.217	0.951
F^-	0.280	0.813	−0.274	0.207	0.890	−0.295	−0.209
Eigen value	4.95	3.56	1.49	6.56	2.56	1.50	1.28
% of variance	38.05	27.49	10.76	50.48	18.89	11.53	9.04
Cumulative % of variance	38.05	65.44	76.09	50.48	69.40	80.90	89.93

(b)

Variables	Component				
	Groundwater			Surface water	
	1	2	3	1	2
pH	−0.145	0.738	0.366	−0.597	−0.127
EC	0.989	0.002	0.067	0.984	0.064
TDS	0.964	0.025	− 0.102	0.987	0.074
Na^+	0.775	0.546	0.084	0.982	0.037
K^+	0.152	−0.181	0.850	0.749	0.617
Mg^{2+}	0.804	−0.142	0.065	0.983	− 0.037
Ca^{2+}	0.479	−0.725	−0.122	0.867	0.433
Cl^-	0.914	−0.015	−0.142	0.894	0.280
SO_4^-	0.961	−0.071	0.088	0.937	0.305
HCO_3^-	0.446	0.637	−0.334	0.879	−0.220
CO_3^{2-}	−0.117	0.214	0.846	0.647	−0.282
NO_3^-	0.527	−0.551	0.430	0.229	0.606
F^-	0.168	0.804	−0.135	0.337	−0.786
Eigen value	5.71	2.85	1.93	8.75	1.71
% of variance	43.93	21.90	14.88	67.28	13.14
Cumulative % of variance	43.93	65.83	80.71	68.28	80.43

in mining areas (Sudheer Kumar et al. 2017; Sakram et al. 2019; Dhakate et al. 2013).

The obtained results of the final rotated loading matrix for groundwater data of pre-monsoon season (Table 3a and Fig. 7a) procures that the three-factor components representing 76.09% of the total variance. Factor-1 represents 38.05% of the variance with negative loading for pH and high positive loading for SO_4^-, Cl^-, Mg^{2+}, EC, TDS, Na^+, Ca^{2+} because of cations and anions resulted due to the contribution from the weathering of mineral and interaction of water with the rock for a long period in the aquifer. Factor-2 represents 27.49% of the total variance and high loadings on HCO_3^-, F^-, CO_3^{2-}, Na^+, and negative loadings on Ca^{2+} and NO_3^- which specifies carbonate weathering, ion-exchange, and leaching from fluoride bearing minerals such as apatite, mica, hornblende, etc. Factor-3 represents 10.76% of the total variance and shows high loading for K^+ which specifies weathering reactions including silicate minerals, K-feldspar is pre-dominant, contamination of groundwater. The surface water samples in the pre-monsoon season resulted in four components account for 89.93% of the total variance in the hydrochemical data (Table 3a and Fig. 7b) Factor-1 represents 50.48% had strong positive loadings on Cl^-, TDS, EC, Na^+, SO_4^- and Mg^{2+}. Factor 2 represents 18.89% had strong positive loadings on CO_3^{2-}, HCO_3^-, F^-, Mg^{2+} and Na^+. Factor-3 represents 11.53% had strong positive loadings on K^+, Ca^{2+}, and negative loading on pH. Factor-4 represents



(a)

(b)

(c)

(d)

Fig. 7 Principal component plot in rotated space groundwater and surface water samples **a** groundwater sample for pre-monsoon season, **b** surface water sample for pre-monsoon season, **c** groundwater sample for post-monsoon season, **d** surface water sample for post-monsoon season

9.04% had strong positive loadings on NO_3^-, SO_4^- and Ca^{2+} (Kanade and Gaikwad 2011; Shyu et al. 2011).

In the post-monsoon season, three-factor analysis represents 80.71% of the total variance. The components 1, 2, and 3 were found to be responsible for the variations in groundwater quality and accounts for 43.93%, 21.90%, and 14.88% of the total variance in the groundwater data respectively (Table 3b and Fig. 7c). Factor-1 shows high positive loadings on EC, SO_4^-, TDS, Cl^-, Mg^{2+}, Na^+, and NO_3^-. The Factor-2 has strong positive loadings on F^-, pH, HCO_3^-, Na^+ and negative loading on NO_3^-, which indicates the predominance of ion-exchange and carbonate weathering, leaching from fluoride bearing minerals and high loading of nitrate represents an enormous quantity of fertilizers are being used in this region (Elumalai et al. 2017). Factor-3 had strong positive loadings for K^+ and CO_3^{2-}. The surface water samples of the post-monsoon season resulted in only two components account for 80.43% of the total variance in the hydrochemical data (Table 3b and Fig. 7d). Factor 1 represents 67.28% had strong positive loadings on TDS, EC, Mg^{2+}, Na^+, SO_4^-, Cl^-, HCO_3^-, Ca^{2+}, K^+, CO_3^{2-} and pH is negative loading. Factor 2 represents 13.14% had strong positive loadings on K^+, NO_3, and negative loading on F^-.

Health Risk Assessment (HRA)

Hazard quotient (HQ) via ingestion (HQ_{ing}) and dermal (HQ_{der}) associated with fluoride concentration for different age group was calculated by computing the values obtained from the United States Environmental Protection Agency's Exposure Factor Handbook (US-EPA 2011) with the fluoride concentration values of the groundwater and surface water samples of pre and post-monsoon seasons from the Chinneru River basin of Yadadri Bhuvanagiri district, Telangana, India.

Chronic daily dose and hazard quotient of fluoride via Ingestion (HQ_{ing}) and Dermal (HQ_{der}) pathway are calculated using the equations given below:

$$\mathrm{CDD_{ing}} = \frac{C_{\mathrm{fw}} \times \mathrm{IR_w} \times E_{\mathrm{fr}} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \tag{3}$$

$$\mathrm{CDD_{der}} = \frac{C_{\mathrm{fw}} \times \mathrm{ESA} \times K \times E_{\mathrm{fr}} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}}, \tag{4}$$

$$\mathrm{HQ_{ing}} = \frac{\mathrm{CDD_{ing}}}{\mathrm{RfD}}, \tag{5}$$

$$\mathrm{HQ_{der}} = \frac{\mathrm{CDD_{der}}}{\mathrm{RfD}}, \tag{6}$$

where CDD_{ing} is chronic daily dose through ingestion pathway (mg/kg day), CDD_{der} is chronic daily dose through dermal exposure pathway (mg/kg day), C_{fw} is fluoride content in water (mg/L), E_{fr} is exposure frequency (days/years), ED is duration to the exposure (years), BW is the weight of the body (kg), AT is average time for effect (days/years), ESA is exposed skin area (cm^2), K is skin adherence factor, CF is conversion factor (L/cm^3), RfD is reference amount of fluoride (0.06 mg/kg day). Key parameter values for calculating the exposure risk of fluoride via ingestion and dermal pathways are presented in Table 4.

Table 4 Key parameters for computing the exposure risk of fluoride through ingestion and dermal pathways

Parameters	Unit	6–12 months	6–11 years	11–16 years	16–18 years	18–21 years	≥ 21 years	> 65 years
Ingestion pathway								
Ingestion rate (IR_w)	L/day	1	1.32	1.82	1.78	2.34	2.94	2.73
Exposure frequency (E_{fr})	Days/year	365	365	365	365	365	365	365
Exposure duration (ED)	Year	6	6	6	6	30	30	30
Body weight (BW)	kg	9.1	29.3	54.2	67.6	67.6	78.8	80
Average time (AT_r)	Days	2190	2190	2190	2190	10,950	10,950	10,950
Concentration of element (C_{fw})	Mg/L	Present study						
Dermal pathway								
Skin surface area (SA)	cm^2	4500	10,500	15,700	18,000	19,550	19,800	19,400
Exposure time (ET)	h/event	0.54	0.54	0.54	0.54	0.71	0.71	0.71
Exposure frequency (E_{fr})	Days/year	350	350	350	350	350	350	350
Exposure duration (ED)	Year	6	6	6	6	30	30	30
Conversion factor (CF)	L/cm^3	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Body weight (BW)	kg	9.1	29.3	54.2	67.6	67.6	78.8	80
Average time (AT_r)	Days	2190	2190	2190	2190	2190	2190	2190
Skin adherence factor (K_p)	cm/h	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Concentration of element (C_{fw})	mg/L	Present study						

Ingestion pathway

Ingestion is considered to be the main pathway of exposure to fluoride from water. Hazard quotient associated with non-carcinogenic risk for the age group of 6–12 months, 6–11 years, 11–16 years, 16–18 years, 18–21 years, ≥ 21 years, > 65 years caused due to the ingestion of fluoride are evaluated (Table 5a and b). It is important to observe from the tables that the influence of F^- on the age group 6–12 months showed the greatest exposure as compared to other age groups, while age group 16–18 years exposed the least. This may be due to the lower bodyweight of the age group 6–12 months than the age group 16–18 years. The HQ_{ing} values of groundwater and surface water for the age group 6–12 months ranged from 0.79–5.36 and 0.69–6.37 for pre-monsoon and 1.10–6.53 and 1.66–5.89 for post-monsoon season respectively. Few samples of pre-monsoon which were within the permissible limit are greater than HQ = 1 in post-monsoon season samples. Samples of post-monsoon have a higher concentration of fluoride than pre-monsoon samples, due to which there is an increase in health risk. Almost all samples of the age group 6–12 months are exciding the permissible limit of HQ = 1 (Table 6). The health risk due to ingestion of F^- contaminated water is in the order of (6–12 months) > (6–11 years) > (≥ 21 years) > (65 years) > (18–21 years) > (11–16 years) > (16–18 years).

Dermal pathway

The non-carcinogenic risk due to fluoride via dermal (HQ_{der}) pathway is found to be less than acceptable limit for the age group of 6–12 months, 6–11 years, and 11–16 years indicating that there is no health risk. The HQ_{der} results evaluated are presented in (Table 7a and b) and it is clear that the few samples of age group 18–21 years, ≥ 21 years, > 65 years have exceeded the limit of HQ = 1. This suggests that there may be a non-carcinogenic adverse health effect. Pre-monsoon HQ_{der} percentage values greater than the acceptable limit for groundwater and surface water of age group 18–21 years, ≥ 21 years, > 65 years are 25 and 36.84, 21.42 and 36.84, 17.85 and 36.84, while post-monsoon percentage values are 35.71 and 29.41, 28.57 and 11.76, 28.57 and 11.76 respectively (Table 6). The health risk via dermal of F^- contaminated water is in the order of (18–21 years) > (≥ 21 years) > (> 65 years). Classification of health risk based on HQ_{ing} values for different age groups of groundwater and surface water sample during pre and post-monsoon season are shown in (Table 8a and b). Hence from the values of ingestion and dermal, it is clear that the ingestion pathway is more likely to cause non-carcinogenic risk, especially in children.

Table 5 (a) Hazard quotient (HQ_{ing}) of fluoride via ingestion of groundwater and surface water samples for pre-monsoon season; (b) Hazard quotient (HQ_{ing}) of fluoride via ingestion of groundwater and surface water samples for post-monsoon season

(a)

Age group	Pre-monsoon (groundwater samples)				Pre-monsoon (surface water samples)			
	Min	Max	Ave	SD	Min	Max	Ave	SD
6–12 months	0.79	5.36	2.80	1.41	0.69	6.37	3.54	1.59
6–11 years	0.32	2.20	1.14	0.58	0.28	2.61	1.45	0.65
11–16 years	0.24	1.63	0.85	0.43	0.21	1.94	1.08	0.48
16–18 years	0.18	1.28	0.67	0.33	0.16	1.52	0.84	0.38
18–21 years	0.24	1.69	0.88	0.44	0.21	2.00	1.11	0.50
≥ 21 years	0.26	1.82	0.95	0.48	0.23	2.16	1.20	0.54
> 65 years	0.24	1.66	0.87	0.43	0.21	1.97	1.10	0.49

(b)

Age group	Post-monsoon (groundwater samples)				Post-monsoon (surface water samples)			
	Min	Max	Ave	SD	Min	Max	Ave	SD
6–12 months	1.10	6.53	3.16	1.67	1.66	5.89	3.25	1.14
6–11 years	0.45	2.67	1.29	0.68	0.68	2.41	1.33	0.46
11–16 years	0.33	1.99	0.96	0.51	0.50	1.80	0.99	0.34
16–18 years	0.26	1.56	0.75	0.40	0.39	1.41	0.78	0.27
18–21 years	0.34	2.05	0.99	0.52	0.52	1.85	1.02	0.35
≥ 21 years	0.37	2.21	1.07	0.56	0.56	2.00	1.10	0.38
> 65 years	0.34	2.02	0.98	0.52	0.51	1.83	1.01	0.35

Table 6 Percentage of samples having hazard quotient > 1 based on ingestion and dermal contact of fluoride in groundwater and surface water for pre-monsoon and post-monsoon season

Age group	Pre-monsoon				Post-monsoon			
	GW		SW		GW		SW	
	ingestion (%)	Dermal (%)	Ingestion (%)	Dermal (%)	ingestion (%)	Dermal (%)	Ingestion (%)	Dermal (%)
6–12 months	92.85	–	94.73	–	100	–	100	–
6–11 years	50	–	73.68	–	50	–	76.47	–
11–16 years	32.14	–	47.36	–	39.28	–	52.94	–
16–18 years	21.42	–	36.84	–	35.71	–	23.52	–
18–21 years	32.14	25	52.63	36.84	39.28	35.71	52.94	29.41
≥ 21 years	35.71	21.42	52.63	36.84	42.85	28.57	58.82	11.76
> 65 years	32.14	17.85	52.63	36.84	39.28	28.57	52.94	11.76

GW groundwater sample, *SW* surface water sample

Table 7 (a) Hazard Quotient (HQ_{der}) of Fluoride via Dermal of groundwater and surface water samples for pre-monsoon season; (b) Hazard Quotient (HQ_{der}) of Fluoride via Dermal of groundwater and surface water samples for post-monsoon season

(a)

Age group	Pre-monsoon (groundwater samples)				Pre-monsoon (surface water samples)			
	Min	Max	Ave	SD	Min	Max	Ave	SD
6–12 months	0.003	0.02	0.01	0.006	0.002	0.02	0.01	0.006
6–11 years	0.002	0.01	0.008	0.004	0.002	0.01	0.01	0.005
11–16 years	0.001	0.01	0.00	0.003	0.001	0.01	0.008	0.004
16–18 years	0.001	0.012	0.00	0.003	0.001	0.01	0.008	0.003
18–21 years	0.19	1.35	0.70	0.35	0.17	1.60	0.89	0.40
≥ 21 years	0.17	1.17	0.61	0.31	0.15	1.39	0.77	0.35
> 65 years	0.16	1.13	0.59	0.29	0.14	1.34	0.75	0.33

(b)

Age group	Post-monsoon (groundwater samples)				Post-monsoon (surface water samples)			
	Min	Max	Ave	SD	Min	Max	Ave	SD
6–12 months	0.004	0.02	0.01	0.007	0.007	0.02	0.01	0.004
6–11 years	0.003	0.02	0.009	0.005	0.005	0.01	0.01	0.003
11–16 years	0.002	0.01	0.007	0.004	0.004	0.01	0.008	0.002
16–18 years	0.002	0.01	0.007	0.003	0.003	0.01	0.007	0.002
18–21 years	0.27	1.64	0.79	0.42	0.41	1.48	0.82	0.28
≥ 21 years	0.24	1.43	0.69	0.36	0.36	1.29	0.71	0.25
> 65 years	0.23	1.38	0.66	0.35	0.35	1.24	0.68	0.24

Conclusions

The present study is an attempt to delineate high fluoride contaminated zones and its non-carcinogenic effects on the human health of different age groups via ingestion and dermal in the Chinneru river basin of Yadadri Bhuvanagiri district, Telangana, India. Groundwater and surface water samples of pre and post-monsoon seasons were evaluated for various physicochemical parameters. The groundwater and surface water are alkaline in nature. The dominance cation in groundwater and surface water is $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ for both seasons. The anion dominance in groundwater for both the seasons is $HCO_3^- > Cl^- > NO_3^- > SO_4^{2-} > CO_3^{2-} > F^-$, whereas surface water anion concentration is $HCO_3^- > Cl^- > CO_3^{2-} > SO_4^{2-} > NO_3^- > F^-$ and $HCO_3^- > Cl^- > SO_4^{2-} > CO_3^{2-} > NO_3^- > F$ for pre and post-monsoon seasons respectively. The fluoride concentration in surface water ranged from 0.3–3.4 to 0.9–3.2 mg/L while in groundwater is from 0.4–2.9 to 0.6–3.6 mg/L for pre and post-monsoon seasons respectively.

Table 8 (a) Classification of health risk as per HQ_{ing} values (%) for different age groups of groundwater and surface water samples for pre-monsoon season; (b) Classification of health risk as per HQ_{ing} values (%) for different age groups of groundwater and surface water samples for post-monsoon season

(a)

Water type	HQ_{ing}	Class	Age group						
			6–12 months	6–11 years	11–16 years	16–18 years	18–21 years	≥ 21 years	> 65 years
Groundwater	< 1	No risk	7.14	50.00	67.86	78.57	67.86	64.29	67.86
	1–1.5	Slight risk	14.29	21.43	14.29	21.43	14.29	14.29	14.29
	1.5–2.5	Moderate risk	32.14	28.57	17.86	0.00	17.86	21.43	17.86
	2.5–3.5	High risk	14.29	0.00	0.00	0.00	0.00	0.00	0.00
	> 3.5	Very high risk	32.14	0.00	0.00	0.00	0.00	0.00	0.00
Surface water	< 1	No risk	5.26	26.32	52.63	63.16	47.37	47.37	47.37
	1–1.5	Slight risk	0.00	31.58	21.05	31.58	15.79	15.79	21.05
	1.5–2.5	Moderate risk	21.05	36.84	26.32	5.26	36.84	36.84	31.58
	2.5–3.5	High risk	31.58	5.26	0.00	0.00	0.00	0.00	0.00
	> 3.5	Very high risk	42.11	0.00	0.00	0.00	0.00	0.00	0.00

(b)

Water type	HQ_{ing}	Class	Age group						
			6–12 months	6–11 years	11–16 years	16–18 years	18–21 years	≥ 21 years	> 65 years
Groundwater	< 1	No risk	0.00	50.00	60.71	64.29	60.71	57.14	60.71
	1–1.5	Slight risk	17.86	14.29	14.29	32.14	10.71	14.29	14.29
	1.5–2.5	Moderate risk	32.14	28.57	25.00	3.57	28.57	28.57	25.00
	2.5–3.5	High risk	14.29	7.14	0.00	0.00	0.00	0.00	0.00
	> 3.5	Very high risk	35.71	0.00	0.00	0.00	0.00	0.00	0.00
Surface water	< 1	No risk	0.00	23.53	47.06	76.47	47.06	41.18	47.06
	1–1.5	Slight risk	0.00	41.18	47.06	23.53	47.06	47.06	47.06
	1.5–2.5	Moderate risk	23.53	35.29	5.88	0.00	5.88	11.76	5.88
	2.5–3.5	High risk	35.29	11.76	0.00	0.00	0.00	0.00	0.00
	> 3.5	Very high risk	47.06	0.00	0.00	0.00	0.00	0.00	0.00

The piper trilinear diagram revealed pre-monsoon groundwater samples are of Mixed CaMgCl-type, $CaCO_3$-type, and NaCl-type, while surface water samples are dominant in Mixed $CaNaHCO_3$-type and $CaCO_3$-type. Post-monsoon samples of surface water are dominant in the Mixed $CaNaHCO_3$-type and few in NaCl-type, but groundwater is in dominance of Mixed CaMgCl-type, $CaCO_3$-type and Mixed $CaNaHCO_3$-type. The surface water and groundwater chemistry are controlled by the mechanism of rock-water interaction dominance process. CAI-I and CAI-II of pre and post-monsoon show nearly 70% and 69% negative results with the cation–anion exchange reaction. Principle component analysis (PCA) results show that water chemistry is controlled by geogenic activities. The PCA results obtained by the final rotated loading matrix of the groundwater data from the pre and post-monsoon season resulted in three-factor components account for 76.09% and three-factor component account for 80.71% of the total variance in hydrochemical data, respectively. The surface water samples of pre and post-monsoon season resulted in four components account for 89.93% and two components account for 80.43% of the total variance in the hydrochemical data, respectively.

The non-carcinogenic risk associated with fluoride contaminated water via ingestion has more risk as compared to the dermal pathway. Children of age group 6–12 months and 6–11 years are found to be more susceptible to non-carcinogenic health risk compared to that of other age groups. Hence, a study of water contamination associated with human health is important. This study will help in making a strategy for the alleviation of fluoride contamination in water. Remedial measures like isolating fluoride contaminated sources and adopting alternative drinking water sources is an advisable measure and also proper treatment is mandatory, adopting rainwater harvesting and artificial recharge techniques to reduce the fluoride content.

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Declarations

Conflict of interest No conflict of interest to declare.

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