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Research Article

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Posted Date: April 6th, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1512920/v1>

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Assessment of groundwater quality and human health risk impact on fluoride concentration: A case study of Namakkal district, South India

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Abstract

This study evaluates the health hazards of fluoride contamination of groundwater in the Namakkal district of south India based on the water quality index. The samples were collected during the NEM season from bore wells and analysed for major anion and cation. There is generally no smell or colour to the groundwater in the study area. The pH of the groundwater samples is acidic to alkaline in nature. Higher EC was noted at 7650 $\mu\text{S}/\text{cm}$ during the NEM season. Na^+ is the most abundant ion in groundwater, followed by Ca^{2+} , Mg^{2+} , K^+ , then Cl^- , $\text{SO}_4^{2-} > \text{HCO}_3^-$, and then F^- . The majority of ions like Sodium, Potassium, Calcium, Magnesium, Cl^- , SO_4^- , HCO_3 , and F^- recorded higher concentrations during NEM seasons, representing the action of leaching, dissolution, and weathering anthropogenic influences on the groundwater quality. Hill-Piper plot showed the majority of the groundwater samples fall in CaHCO_3 , NaCl , and Mixed CaNaHCO_3 , CaMgCl , with minor representations in CaCl and NaHCO_3 being noted during the NEM. The Gibbs plot shows all the water samples belonging to rock dominance. The WQI value varies between 22.08 and 211.64 throughout the NEM seasons, with a mean of 78.90. According to the Water Quality Index (WQI), 26%, 33%, and 14% of water types have good, poor, and extremely poor quality, respectively. The average F^- level is 1.12 mg/l and ranges from 0.27 to 0.39 mg/l. There is excessive fluoride concentration in 76 % of the groundwater samples (WHO 2011). Children, women, and men should undergo health risk assessments based on non-carcinogenic risk estimates for fluoride oral and dermal pathways. The health risk assessment for this study is that children are the most susceptible to fluoride intake. Consequently, more treatments must be initiated to appraise the water as good quality, and government organizations need to select and provide appropriate groundwater for the inhabitants.

Keywords: water quality index (WQI), fluoride risk, health hazards.

1. Introduction

The groundwater is essential for food security and drinking water, rendering a good ecosystem and human health (Raj and Shaji, 2016). The groundwater is a significant causal factor in climate change because of its proportional steadiness in quantity and quality (Saha et al., 2018). The groundwater is a freshwater reserve vulnerable to unmaintainable abstraction (Wada et al., 2010; Taylor et al., 2013). Groundwater reduction is a universal matter whose scale is poorly known today (Konikow and Kendy, 2005). The groundwater abstraction rate is about 1500 km³/year for agricultural and drinking water use (Döll et al., 2012; UNESCO, 2015). The over-exploitation of groundwater led to the rapid decline in groundwater levels in many parts of the aquifer system globally (Wada et al., 2010). In India, the groundwater abstraction is about 243 km³ and abstraction meets more than 50% of urban water demand, about 62% of irrigation requirements, and about 85% of rural drinking supply (Saha et al., 2018). The number of wells, such as diesel and electric pumps, has increased considerably since the 1970s, after introducing the green revolution (CGWB, 2015; Mishra et al., 2018; Sinha et al., 2019). As a result, groundwater resources are decreasing rapidly in many parts of India (Rodell et al., 2009; Tiwari et al., 2009; Chen et al., 2014; Bhanja et al., 2017; Van Dijk et al., 2020; Joshi et al., 2020 & 2021) due to irrigation and change in monsoon precipitation pattern (Sinha et al., 2019; Asoka and Mishra, 2020).

Water is one of the essential natural resources for all living beings. As reported by the United Nations, about 780 million people across the globe do not have access to uncontaminated water (WHO, 2017). Population growth has increased water consumption and stress on groundwater supplies. Moreover, global climate and land-use changes have reduced the quality and quantity of drinking water (Hunter, 2003; Duran et al., 2017). In both wealthy and developing countries, changes in water quality represent a danger to crop production, population health, ecology, and many other natural ecosystems as a result of climate change (Mishra, 2014). Furthermore, because of the difficulties involved in distinguishing between geogenic and anthropogenic causes, it is difficult to identify the precise mechanisms responsible for the degradation in groundwater quality (Jampani et al., 2020).

Groundwater scarcity is becoming a significant issue in some parts of India, predominantly in hard rock terrain, due to the country's increasing population and growing demand. Furthermore, regional differences in water availability have developed due to the geographic and systemic distribution of available water resources. Pollutants impact the water quality of both surface and underground water, resulting in the following changes: (Mahadev and Gholami 2010). In this day and age, water quality decline is an important alarm, predominantly considering the limited availability of water (Subramanian 2000; Singh et al. 2011, 2013). Contamination of water and the health risks that come with it are major study topics. Fluoride-contaminated groundwater has affected nearly 300 million people, both directly and indirectly (Kumar et al., 2016; Singh et al., 2021). Groundwater contains the most electronegative and sensitive element because of geogenic practices involving the dissolution of F-containing minerals (Maity et al., 2018). Long-term fluoride exposure of more than 1.5 mg/l is harmful to the development of children (Yelidji et al., 2017). Excessive fertilizer use in industry and agricultural pollution are the main anthropogenic reasons for F (Brindha et al., 2017; Kumar et al., 2018; Alcaine et al., 2020).

The human health risk assessment defines the hazard quotient for fluoride exposure in groundwater (Aravinthasamy et al. 2019a, b). The fluoride exposure was assessed using EPA standards (USEPA 2006). However, there have been no comprehensive studies on the health effects of high fluoride concentrations in south Indian hard rock terrains. The current study region is characterized by rapid population increase, industrialization, and agricultural activity. The groundwater in this area is over-extracted to fulfil everyday demands, resulting in significant water quality issues. The unregulated use of pesticides and fertilizers has also harmed groundwater quality. F concentrations in the groundwater in this location are high. The current study tries to quantify the potential health risk associated with fluoride and nitrate pollution in groundwater. The aim of the present study is (1) to characterize groundwater using the water quality index (WQI) and (2) to assess the hazards of excessive fluoride in drinking water to human health. Eventually, a proposal to improve the current situation was proposed.

2. Study Area

2.1. The geographical position of the study area

The splitting of Salem district formed the research location Namakkal district in Tamilnadu on January 1, 1997. It is bordered by Salem District in the north and east, Tiruchirapalli District in the south and east, Erode District in the west, and Karur District in the south. It is between the latitudes of N 11° 00' and N 11° 36' and the longitudes of E 77° 28' and E 78° 30', with a total geographical area of 3363.35 km² (Fig.1). The Kollimalai hills, Bodamalai hills, Naraikinaru hills, and Pachamalai hills are the district's most notable hill ranges. The Kollimalai hilltop, with a height of 1293 m above sea level, is the district's highest point. Other prominent peaks in the Bodamalai range are Kedda Malai (1284 m) and Melur hill.

2.2. Drainage

The district's western and southern borders are bordered by the Cauvery River, which is perennial. The Tirumanimuttar river, the district's most major tributary of the Cauvery, rises in the Manjavadi region and winds its way across the district until joining the Cauvery near Nanjai Edayar hamlet in the Paramathi taluk. A small area in the northeast district is drained mainly through the Nadi and Sweta Nadi rivers, perennial sources of the Cauvery river, which flows along the district's western and southern borders. The Tirumanimuttar river, Salem most significant hill, runs through the district before joining the Cauvery at Nanjai Edayar settlement in Paramathi taluk. Vasista southern bounds of the district mostly drain a small region in the northeastern section. The Tirumanimuttar river, the districts most major tributary of the Cauvery, rises in the Manjavadi part of Salem district Shevroy hills and winds its way across the district until joining the Cauvery near Nanjai Edayar hamlet in Paramathi taluk. Rivers, tributaries of the Vellar River, drain a tiny region in the city's northeastern section.

2.3. Climate Condition and Rainfall Data

The climate in the region is often hot and dry. Warm weather begins in March and peaks in April, October, and December. The average annual rainfall ranges from 58 cm to 70 cm, most of it falling during the summer. The

climate in the region is often hot and dry. Hot weather begins in March and peaks in April and June. Between October and December, the northeast monsoon brings rain to the region. The average annual rainfall ranges from 58 cm to 70 cm, most of it falling during the summer. The research region has a hot and dry climate, with the hottest months being March and June. Between October and December, the northeast monsoon brings rain to the area. The average annual rainfall ranges from 58 cm to 70 cm, with November being the rainy season.

2.4. Geology of the study area

The geology of the studied area is mainly based on the Archaean crystalline and metamorphic complex. The region's geology is complicated by the pre-Cambrian epoch's frequent tectonic and magmatic activity. This region has the well-known Sithampoondi complex, known for its complex geology. Gneisses are the oldest rocks in the area, and they are found all across the plains. The gneisses rock formations have a lot of wear on them. The Charnockites are coarse-grained and have a blue, dark to grey colouration. They are the second most common rock type in the area. They are larger than gneisses and less weathered. In some regions, iron ore deposits are related to felspathic quartz gneisses and garnetiferous quartz gneisses. These rocks have many folds and joints and are weathered. Along with Cauvery and Thirumanimuthar, a thin veneer of alluvium may be discovered. Around the confluence of the rivers Thirumanimuthar, however, there is alluvium of a few meters in thickness. There are many faults and shears, especially along with the northeast-southwest trend. They are predicted to impact the district's groundwater circulation, storage, and development potential.

2.5. Hydrogeology

The studied region features a hard-rock topography made up of Archaean, granites, gneisses, and dykes, suggesting geology (Chakrapani and Manickyan 1988). Pink migmatite rocks dominate the research area, followed by hornblende-biotite gneiss and Charnockite (Fig.1). Garnet, fluvial (sand, silt, gravel and clay), ultrabasic, calc-gneiss/limestone, and carbonatites were identified among these minimum minerals (Krishnan 1982). Foliation generally follows an NNE–SSE trend, with dips in the NW and SE directions. The water depth in the area ranges from less than 1m to 25m. There are varying heights between 5 and 10 meters in the south and northeast of the region (Kumar and Ramasesha 2002). Groundwater is most commonly found in weathered and semi-contained to restricted rock formations such as jointed and fractured rocks beneath water-table settings. Dug wells, dug-cum-bore wells, and bore wells produce groundwater. There is the potential for finding water tables as deep as 3 to 12 m below ground level, depending on the weathered zone thickness, the degree of weathering, and the withdrawal rate. Most of the geomorphological properties of the research region, including pediments and structural hills, are covered by weathered pediplains, structural hills, and pediments.

3. Material and Methodology

3.1 Sampling and data analysis

During the North-East Monsoon (NEM) of 2015, groundwater samples were collected from 58 bore well locations throughout the research region (Fig.1). The precise coordinates of the sample locations were determined using a GARMIN GPS. For sampling, acid-washed clear polythene bottles were utilised. Before sampling, standing water in wells was removed by pumping the wells briefly. While filling the vials with samples, extra care was taken to avoid air bubbles. The samples were then sealed and delivered to the laboratory. Precautions were made by keeping the samples at 4°C and filtering them using 0.45-µm paper for further analysis. The measurement of various cations and anions is done following APHA, 1998. Hydrogen ion concentration activity is one of the factors being studied. The temperature, EC, TDS, and pH were measured on-site using the pH and TDS meter. The titrimetric technique helps detect Calcium(Ca²⁺) by combining it with a standard acid such as Ethylene Diamine Tetra Acetic (EDTA). The content of bicarbonate (HCO₃⁻), magnesium (Mg²⁺), and potassium (K⁺) were all determined using the conventional Silver Nitrate (AgNO₃) titration method. A flame photometer was used to determine the concentration of sodium (Na²⁺) (Elico SL 198). A spectrophotometer also measured levels of Nitrate (NO₃⁻), Sulphate (SO₄²⁻), and phosphate (PO₄⁻) (Elico SL 207 Mini). Calibration standards were applied to equipment and instruments, and the results were compared to the specifications of the standard water. The USSL and Gibbs diagrams were generated using the WATCLAST programme in C (Chidambaram et al., 2003). The total components equivalents per million concentrations are derived by multiplying the ppm concentrations by the weight common. The concentrations of major ions were converted to meq/L, and the analytical accuracy for major ion measurement ranges from 5 to 10%. Anion balancing (Freeze and Cherry, 1979). The geological and water sample location maps were prepared in ArcGIS version 10.2.1.

3.2 The calculation of the water quality index (WQI)

The most significant factor in determining drinking water quality is groundwater chemistry (Kalaivanan et al., 2018; Ramya Priya and Elango, 2017; Verma et al., 2018; Gopinath et al., 2018; Karunanidhi et al., 2020). The WQI is useful for calculating groundwater quality for drinking and irrigation. It was determined by weighing (wi) the physical and chemical parameters (pH, EC, TDS, Ca²⁺, Mg²⁺, Na⁺, K⁺, HCO₃⁻, Cl⁻, SO₄²⁻ and F⁻). Table 1 shows the assigned weight (wi) and relative weights (Wi) for all chemical characteristics according to WHO (2017).

Weights vary from 1 to 5. Due to its importance in assessing water quality, TDS, NO₃⁻, Cl⁻, and SO₄²⁻ have been given a maximum weight of 5. (Karunanidhi et al., 2020; Liu et al., 2020). Bicarbonate and phosphate were assigned a weightage of 1 since they are minor water quality components. The water quality was assessed by assigning weights to Ca²⁺, Mg²⁺, and other factors (Ketata-Rokbani et al., 2011). Equation 1 below calculates relative weight.

$$W_i = \frac{w_i}{\sum_{i=1}^n w_i} \quad (\text{Eq.1})$$

This equation has ‘n’ parameters, and ‘Wi’ is the relative weightage. Eq. 2 is used to construct the quality rating scale for each parameter (WHO 2017)

$$q_i = \frac{C_i}{S_i} \times 100 \quad (\text{Eq.2})$$

In this equation, q_i represents the quality rating, C_i represents each chemical parameter's absorption (mg/l), and S_i represents the WHO (2017) expected value. Using Eq. 3 to calculate WQI, SI is first calculated for each chemical parameter. This information is then used by Eq. 4 to calculate WQI according to the SI.

$$SI_i = W_i \times q_i \quad (\text{Eq.3})$$

$$WQI = \sum SI_i \quad (\text{Eq.4})$$

The equal to a sub-index of the parameter, where 'SI' With 'n' parameters, 'qi' gives an overall rating based on how concentrated each one is in the environment. The calculated WQI map is classified into five stages (< 50 excellent categories, 50 - 100 good categories, 100 - 200 poor categories, 200 - 300 very poor categories, and > 300 water unsuitable for drinking purposes).

3.3 Health Risk Assessment

Non-carcinogenic health risks are assessed based on the consumption, skin contact, and inhalation of a substance to determine the likelihood of a risk for people of all ages. (Li and Wu 2019 a, b). Drinking water consumption, rather than the other two channels previously reported, appears to be the primary exposure pathway in this study. For this reason, F- was selected as pollutants to assess health issues. USAEPA (1991) indicates that F- is the most frequently evaluated non-carcinogenic pollutant. Determine the oral exposure; the mean daily dosage of F- consumed from drinking water was estimated using Eq. 5.

$$DD = \frac{CP \times IR \times ED \times FE}{BW \times ET} \quad (\text{Eq. 5})$$

Where,

DD - Daily mean dosage of pollutant (mg/kg/day); *CP* - Pollutant concentrations of F- in the groundwater (mg/l); *IR* - Ingestion rate per unit time (L/day), 0.78 L/day for children and 2.5 L/day for women and men; *ED* - Exposure duration (years), 12 years for children, 64 years for women, and 67 years for men; *FE* - Frequency of exposure (days/year), which is 365 days/year for children, women, and men; *BW* - Known mean body weight of a person (kg), 15 kg for children and 65 kg for women and men; *ET* - Mean exposure time of age (years), 4380 (children), 24,455 (women) and 23,360 (men).

The hazard quotient increases when the pollutant exposure dosage exceeds the permissible level (Eq. 11) when the pollutant exposure dosage exceeds the permissible level (Eq. 6).

$$HQ = \frac{DD}{PD} \quad (\text{Eq. 6})$$

Where,

HQ - hazard quotient; *PD* - permissible dose for chronic oral exposure, 0.60 mg/kg/day for F-

The total chronic hazard index (TCHI) for non-carcinogenic risk was computed. TCHI limit is 1.0. (USEPA 2014). When the TCHI is less than 1.0, the non-carcinogenic risk health hazard is within the standard limit. A TCHI of 1.0 or above indicates a health concern.

4. Results and Discussion

4.1 Hydrochemistry

The physicochemical characteristics of groundwater samples collected from the Namakkal district during the NEM seasons are presented in Table 2. Along with descriptive statistical methods such as minimum, maximum, mean, and standard deviation values of water quality parameters studied following the World Health Organization standards (WHO 2017). Water used for drinking should be chemically soft, low in dissolved salts, and free of contaminants (Ramesh and Elango 2011). In a process in equilibrium, the pH represents the condition in which water precipitates (Hem 1985). When measured during the NEM seasons, the pH value varies from 8 (Sowdapuram) to 8.80 (Ayilpatti). On average, pH ranges from 8.48, indicating that monsoon effects change the groundwater's pH from acidic to basic. The high pH readings indicate the presence of carbonate due to temperature fluctuations and water movement from the Charnokite highlands to the gneissic rock formation. It is alkalinity that is produced by the bicarbonate and carbonate ions. When the pH is high, hydrogen oxide, iron, and silicate may affect alkalinity. Groundwater pH concentrations in suitable drinking water regions during the seasons assist in limiting the availability of groundwater for drinking purposes in such areas. The pH of groundwater decreased little from the seasons, indicating the rock-water interaction (Srinivasamoorthy et al., 2014; Kalaivanan et al., 2018).

The electrical conductivity (EC) concentrations ranged from 570 (Solaiyudaiyampatti) to 7650 $\mu\text{S}/\text{cm}$ (Malayalapatti), respectively, with an average electrical conductivity of 2002 $\mu\text{S}/\text{cm}$ for the groundwater samples (Table.2). As per the WHO (2017) standard, the EC of groundwater samples falls between the desired limit categories of 45% of the desirable limit category. 55 % of the samples were considered unsuitable for drinking during NEM. The high concentrations of Na^+ and Cl^- in these samples are mainly due to the high EC values. In humans, it has been shown that high EC in groundwater may cause gastrointestinal awkwardness after long exposure (Ramesh and Elango 2011). Total dissolved solids (TDS) of the Namakkal district vary from 313 (Solaiyudaiyampatti) to 3504 (Unjanai), averaging 1148.47 mg/l. TDS are an important factor in determining water suitability for various purposes. 10% of samples fall in the freshwater, and the remaining 90% of the water samples are unsuitable categories for drinking purposes. These changes indicate that rainfall has a significant impact on reducing TDS in groundwater. TDS has also been added to rainfall due to its interactions with minerals and rocks.

The groundwater sample in the study area shows cation domination in ascending order of $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ due to the dissolution of aquifer minerals in rainfall in the study area. In the NEM seasons, rock dominance and anthropogenic contribute to $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+$ values being higher. The Ca^{2+} content varies from 6 (Erayamangalam) to 152 (Muthugapatti) with an average of 42.59 mg/l for NEM seasons, respectively. The desirable

limit of Ca^{2+} for drinking water quality is less than 75 mg/l classified by the WHO, 2017. 88 % of groundwater samples fall under the most desirable limit. The rest of the 12% samples occur in the not permissible limit during the NEM season respectively (Table 2). The increasing Ca^{2+} content in groundwater is a result of might be due to silicate weathering minerals.

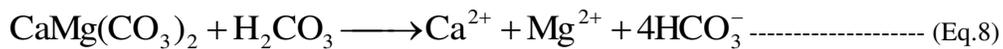
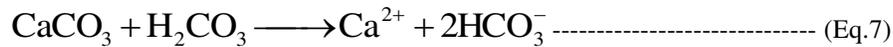
Magnesium is an alkaline earth metal with a 2+ oxidation state, Mg^{2+} , and Ca^{2+} . It has similar effects on water chemistry, as their contribution leads to the permanent hardness of the water. It is a common element and is essential for plant and animal nutrition. The geochemical behaviour of magnesium is relatively different from that of calcium. Comparatively, the size of magnesium ion is smaller than sodium and calcium and can be engaged in the space at the center of six octahedral coordinate water molecules; hence the effects of hydration are much greater for magnesium than for the larger ions of calcium and sodium (Vasanthavigar et al. 2010). Magnesium in groundwater ranges between 28 mg/l to 306.00 mg/l, with an average of 85.22 mg/l during the season. A higher concentration of 306 mg/l is noted during NEM, which might have been derived from rock water interaction of magnesium-bearing silicate minerals. Besides, the lower range of 28.00 mg/l noted during the season is mainly due to the ionic exchange process by Ca. During the NEM season, 71 % of samples exceed the not permissible limit. The rest of the 29% of groundwater samples are most desirable for drinking purposes.

Sodium is the most abundant member of the alkali metal group of the periodic table. In igneous rocks, sodium is slightly more abundant than potassium. According to Clarke's (1996) estimate, about 60 % of the igneous rock in the Earth's outer crust consists of feldspar minerals. The feldspars are tectosilicates, with some aluminium substituted for silica and other cations making up for the positive charge deficiency. The common feldspars are orthoclase and microcline, which have the plagioclase series ranging in composition from albite to anorthite. Some sodium may be present, substituting for potassium in orthoclase and microcline. Sodium concentrations vary from 14.0 to 667 mg/l with an average of 209.79. Higher concentration was noted during the seasons (667 mg/l). Most of the samples record higher Na, indicating contribution from the weathering process of Na plagioclase feldspar and the dissolution of Kankar (Srinivasamoorthy et al. 2008; Gopinath et al. 2019; Saravanan et al. 2018; Vinnarasi et al. 2020). Human activities can significantly influence the concentrations of sodium in groundwater (Hem, 1985). The use of salt for deicing highways in winter flowing from wells has had direct, noticeable regional effects. Somewhat less directly, water reuse for irrigation commonly leaves a much higher residual sodium concentration than the original water. The minimum concentration of 14.0 mg/l noted during the NEM season signifies adding fresh recharge water into the aquifer and eliminating pre-existing mineral components in the water.

Potassium is an important element for both plants and animals. The major potassium minerals of silicate rocks are the feldspars (Orthoclase and Microcline), micas, Feldspathoid, and Leucite. Potassium Feldspars are resistant to chemical weathering. Presumably, they are altered to silica, clay, and potassium ions by the same process as other feldspars but more slowly. In general, natural water contains sodium, less than one-tenth because of potassium-bearing aluminosilicate minerals' high degree of stability (Davis & DeWiest 1970). Potassium in groundwater samples of the study area ranges from 3.0 to 47 mg/l with an average of 24.67 mg/l during NEM seasons, respectively. A higher concentration of K was noted during the NEM (47.0 mg/l), which may be due to soil leaching

by runoff. 84 % of samples exceed the not permissible limit and are unsuitable for drinking purposes in the study area. The remaining 16 % of groundwater samples are suitable for drinking utility.

The alkalinity of water is expressed as the total measure of the substances in water that can neutralize the acid character of the water. In other words, the solution power to react with acid and buffer pH and keep pH from changing. The higher the alkalinity, the higher the buffering capacity against pH changes. The following equation can be mathematically determined alkalinity (Eq.7 & 8).



According to Drever and Stillings (1996), HCO_3^- can be produced by the dissolution of silicate and calcic minerals and the carbonic acid produced by organic materials. HCO_3^- can also be produced by the atmosphere (Srinivasamoorthy et al., 2008). NEM concentrations HCO_3^- vary from 128 to 671 mg/l, with an average of 337.14 mg/l. HCO_3^{2-} concentrations in water are directly proportional to its alkalinity, and higher HCO_3^- concentrations indicate that the groundwater is alkaline. Weathering silicate minerals such as anorthite, sodium, and potassium feldspar results in a rise in the HCO_3^- the content of groundwater samples in the study area. The comparatively high HCO_3^{2-} content is due to the direct mixing of municipal sewage and industrial water with streams from the Northeastern part of the study area.

Chloride, the anion of the halogen group, is a lithophile element with five oxidation states (-1, +1, 3, 5, and 7), of which the -1 state is most common in groundwater (Belkhiri and Mouni 2012). Sources for chloride in groundwater are mainly anthropogenic, like halide usages, industrial sewages, and road salts. The natural chloride-bearing rocks minerals are sodalite, phosphate, and apatite but have lower concentrations. The chloride behaviour in groundwater is considered conservative, which implies its circulation through the hydrological cycle, which is controlled by physical rather than chemical processes. Cl^- concentrations in the study region vary between 39 to 2127 mg/l with an average of 349.31 mg/l. The Cl^- content in water is used as a measure of pollution and is regarded as the cause of groundwater contamination (Loizidou and Kapetanios 1993). Appetite, sodalite, connate water, and hot springs are all significant geological sources of chloride (Anithamary et al., 2012; Freeze and Cherry, 1979). A high Cl^- concentration was observed in the Southwestern region of the study area, owing primarily to the influence of municipal sewage and municipal wastes, as well as surface runoff from agricultural land, and Shin et al. (2011) reported effluents from dyeing and bleaching industries, respectively. In the study area, during the NEM season, 52 % of samples exceed the not permissible category, and the rest of the 48 % of groundwater samples are suitable for drinking purposes.

Sulfate is a significant anion found in natural water, most likely formed through the weathering of sulfate and gypsum-bearing sedimentary rocks (Elango et al., 2003). The sulfide minerals introduce soluble sulfate into the

groundwater (Ramesh and Vennila 2012). The groundwater's highest sulfate (SO_4^{2-}) limit is less than 200 mg/l (Table). The maximum permissible limit is 200 - 400 mg/l (WHO 2011). The SO_4^{2-} concentration value ranges from 15 mg/l to 768 mg/l, with an average of 160.05 mg/l during NEM seasons. In the study area, during the NEM season, 9% of samples exceed the not permissible category, and the rest of the 91% of groundwater samples are suitable for drinking purposes. The contribution of dissolved sulfate ions varies in minerals like gypsum and marcasite (Anandhan, 2005). The other sources of this ion may also be due to bacterial fixation, the impact of fertilizer, tannery, and anthropogenic sources (Chidambaram et al. 2012).

Fluoride (F) ions play an important role in affecting groundwater quality. However, fluoride causes severe health risks to humans (Kale and Pawar 2017; Das and Nag 2017; Panaskar et al. 2017). The high F content groundwater source is the parent rocks with F-bearing minerals (Kale et al. 2010). The concentration of F- above 1.5 mg/l in drinking water affects the teeth and bones of humans. The local geological setting and climatic conditions determine the concentration of F- ions in groundwater. The subsurface ion exchange results in favourable conditions for leaching fluoride ions from host rocks into groundwater. Fluoride leaching increases silica in groundwater. Natural fluoride leaching occurs when hornblende material dissociates in Na^+ and HCO_3^- rich groundwater. Due to semi-arid conditions and evaporation, the study region's alkaline water is more suitable for fluorite mineral breakdown (Chen et al., 2017; Kadam et al., 2021a). In addition, fluoride-rich minerals like fluorite and muscovite found in the area's host rocks may leach fluoride into the groundwater (Adimalla et al., 2020). The F- content in the study area ranges from 0.27 to 2.24 mg/l with an average of 1.12 mg/l, respectively. In the NEM season, 24 % of samples exceed the not permissible category.

The rest of the 76 % of groundwater samples are suitable for drinking purposes. In groundwater, the excess fluoride ion is considered toxic to human health when present in excess or deficit amounts. Consumption of higher fluoride (<1.50 mg/l) can affect bones and soft tissues like erythrocytes, skeletal muscles, gastrointestinal tissues, and ligaments (Brindha et al. 2010). The effect of fluoride on teeth is called dental fluorosis, which causes a mottled appearance in teeth. The impact of fluoride in bone is called skeletal fluorosis, where the rigidity of bone increases and the curvature of bone along with severe pain. Paralysis of the lower part of the body with legs and quadriplegia is paralysis of all limbs. A lower fluoride concentration in groundwater increases the risk of tooth decay.

The spatial distributions map of Fluoride concentration differentiates into three groups such as < 0.5 indicates low risk, 0.5 to 1.5 indicates moderate risk, > 1.5 means high risk. The fluoride ranges from 1.5 to 2.24, which indicates very high risk, while more than 2 indicates extremely high risk (Aravinthasamy et al. 2019). The spatial distribution map represents 460.56 km² area covers high risk categories (Sample.no.1, 3, 17,18, 24, 25, 27, 31, 34, 40, 42, 44, 47, 55, 57). Medium-risk categories occupied 2919.62 km² area, and the remaining 26.18 km² area covers low-risk categories (Fig.2). 14 % of the samples fall under the high-risk category in this study region. 86 % and 1 % of the samples fall under medium and low risk.

4.2 Hydro-geochemical facies

The hydrochemical approach can help understand groundwater systems' movement and transportation principles and provide access to a database of paleoenvironmental data (Pierre et al., 2005; Ophori & Toth, 1989; Hem, 1992). A Hill Piper plot (Piper 1953) is adopted to determine the geochemical assessment of groundwater. Triangular-shaped fields and a diamond-shaped centre field make up the diagram. The % epm values of main cations (Ca, Mg) alkali earth, (Na + K) alkali, ($\text{HCO}_3 + \text{CO}_3$) weak acid, and ($\text{SO}_4 + \text{Cl}$) strong acid are displayed individually in the two triangle fields before being projected onto the centre field to indicate overall water properties. Applications of the diagram pointed out by Piper include testing groups of water analyses to determine whether particular water may be a simple mixture of others for which analyses are available or whether it is affected by the solution or precipitation of a single salt. It can be shown easily that the analysis of any mixture of waters.

The Piper plot reveals the properties and relationships for large sample groups. The highlighted sample points indicate samples selected in the database and highlighted on all other open graphical displays. Several authors have applied this plot to understand the hydrogeochemical facies (Manavalan Satyanarayanan et al., 2007; Reddy et al., 2010; Fantong et al., 2008; Walton, 1970). This AQUACHEM based major-ion trilinear diagram (Fig.3) indicates that the groundwaters have different chemical compositions. During the NEM, the majority of the groundwater samples fall in CaHCO_3 , NaCl, and Mixed CaNaHCO_3 , CaMgCl , with minor representations in CaCl and NaHCO_3 being noted.

Groundwater samples cluster CaHCO_3 , NaCl, Mixed CaNaHCO_3 , CaMgCl , with moderate CaCl and NaHCO_3 type representations, during NEM. In NEM, Na decreases with Ca and Mg increases, signifying ion-exchange weathering. The CaHCO_3 water is predominantly the consequence of carbonate minerals dissolution, and its origin is primarily due to rainfall-derived recharge over decades to centuries. Surface water saturated with environmental and biological CO_2 penetrates the subsurface, resulting in mixed Ca-Mg-Cl water. Due to the elimination of Na from the solution in return for bound Ca at higher salinities, the process of reverse cation exchange may produce CaCl_2 waters. CaCl_2 type water might also develop by combining younger, fresher water with older, more salty water (Adams et al. 2001). The recharge of rains that delivered saline water mixing is more likely to produce calcium-sodium bicarbonate water. The figure shows that an alkali (Na + K) is more powerful than alkaline earth (Ca + Mg), and strong acids (Cl + SO_4) are more powerful than weak acids (HCO_3^-). The water chemistry in the research region is mostly controlled by silicate weathering, secondary leaching, ion exchange, and human activities.

4.3 Gibb's Diagram

Gibbs (1970) proposed the Boomerang envelope model to represent groundwater sources geochemical influences and divided them into the following types: rock control, evaporation/crystallization, and precipitation. Precipitation events water has the highest concentration of Na^+ , whereas weather-dominated water has a high concentration of Ca^{2+} , Cl^- , and HCO_3^- , and evaporation/crystallisation-dominated water has a high concentration of Na^+ . By graphing total dissolved salt concentrations against relative proportions of the principal anions for a variety of surface water drinks across the world, he was able to create a curve with two arms. As the lower arm moves to the

right, the salt percentage of the water decreases, and the water takes on a rainwater-like composition (indicated by a high Cl to HCO₃ ratio) (Fig.4).

The diagram illustrates the ratios of Na: (Na+K) and Cl: (Cl+HCO₃) as a function of TDS that are widely used to assess the available sources of dissolved chemical constituents, such as precipitation, rock, and evapotranspiration dominance. Various processes and mechanisms regulate the water chemistry of the study area. Besides, the study area is chiefly composed of silicate rocks. Its impact on water chemistry is inevitable. During NEM, the plot for most samples of cations and anions falls within the rock dominance zone with minor indications in the evaporation zone (Fig.4). However, Major representations are also noted in the rock dominance zone, indicating effective interaction between rock and groundwater. During the NEM sample is moved towards the evaporation increases salinity (Na) and also increases the TDS.

4.4 Spatial distribution of Water Quality Index (WQI)

The geochemistry of water may certify the drinking water quality (Subba Rao 2006; Vasanthavigar et al. 2010; Kumar et al. 2015; Kalaivanan et al. 2018). The Water Quality Index (WQI) for 58 samples was derived using the values of TDS, Ca²⁺, Mg²⁺, Cl⁻, HCO₃⁻, SO₄²⁻, F⁻, EC, and pH at various locations. The WQI value varies between 22.08 and 211.64 throughout the NEM seasons, with a mean of 78.90. According to the WQI, 3 excellent samples and 15 locations fell under good water type. In the poor and very poor groups, 19 and 8 samples, respectively. The acceptable category for drinking water consists of 18 groundwater sampling locations. As a result, if very poor (14% of samples), poor (33% of samples), or not acceptable (22% of samples) water is utilised for drinking, health risks may arise. Furthermore, the WQI findings revealed that 31% of samples fell into the "good" category, meaning they may be taken without causing harm to one's health (Fig.5). It might be due to the gypsum-bearing and rock salt deposits being effectively leached and dissolved. High absorption of EC, chloride, and sodium, followed by calcium, are the main drivers of water quality degradation in the research region, suggesting that the rock-water interaction mechanism is at work. The results listed the appropriate and insufficient water quality for drinking purposes (Table 3 & 4).

4.5 Human health risk assessment of fluoride

Fluoride concentrations in several rural water wells are significant. It also is critical to assess health hazards based on fluoride exposure in the research region. The impact of fluoride on human health has been investigated using the hazard quotient (HQ), a useful technique for expressing non-carcinogenic dangers in the human system. HQ was calculated for men, women, and children in the research area. For the estimation of non-carcinogenic hazards among children, teenagers, and adults, a variety of characteristics were employed, including ingestion rate (IR), average time (AT), exposure frequency (EF), exposure duration (ED), body weight (BW), and reference intake of fluoride (RfD). Higher consumption of fluoride can create several health issues for people. Fluoride ingestion is linked to a variety of health problems. Table 4 shows the calculated HQ values for the research region. For males, women, and children, they ranged from 0.00E+00 to 1.43E+00, 0.00E+00 to 1.36E+00, and 0.00E+00 to 1.94E+00. The findings show that 48 men, 46 women, and 30 children constitute possible health hazards, accordingly (Fig. 6). According to the survey, people are particularly vulnerable to health risks than men and women. Various researchers in various parts of the

globe have reached the same conclusion (Kadam et al. 2021; Karunanidhi et al. 2021 a & b; Pant et al. 2021; Selvam et al. 2021; Subba Rao 2021).

5. Conclusions

In this study area, 58 groundwater samples were taken and analyzed for both physicochemical parameters to evaluate the groundwater quality using the water quality index (WQI). This study mostly shows fluoride analysis and its influence on the groundwater. It also states the human health risk using human health risk evaluation. The groundwater is slightly alkaline and maintains an average TDS value of 1148.47 mg/l. The ascendancy pattern of the cations and anions are the majority of ions like Sodium, Potassium, Calcium, Magnesium, Chloride, Sulphate, bicarbonate, and fluoride were recorded at higher concentrations during NEM seasons, representing the action of leaching, dissolution, and weathering along with anthropogenic influences on the groundwater quality. The fluoride concentration varies from 0.27 to 2.24 mg/l with an average of 1.12 mg/l. In this region, 24 % of the groundwater samples are not the permissible limit of WHO guidelines (> 1.5 mg/l). The major mechanism for the increase in the groundwater parameter is the weathering of rocks and leaching. The piper trilinear diagram exposes that the groundwater samples fall under the mixed type indicating the impact of weathering of ion exchange. The CaHCO_3 water is mainly a result of the dissolution of carbonate minerals, and the origin of the water is primarily due to rainfall derived recharge over decades to centuries. The mechanism of groundwater chemistry Gibbs plots reveals that all the samples fall under the rock water interface. The appraisal of non-carcinogenic risk was done to stress the health issues that succeed due to the intake and dermal contact of drinking water in the Namakkal district. The percentage of risk $\text{HQ} > 1$ shows that 48 men groundwater samples, followed by 46 groundwater samples women and 30 groundwater samples children, constitute possible health hazards, accordingly. According to the survey, people are particularly vulnerable to health risks than men and women. Overall, health risk estimation results showed that all the groundwater samples have surpassed the permissible limit of $\text{HQ} < 1$ for children. As a result, the current study determines that distinct sensitive zones at a single site have varying spatial intensities, allowing for effective management strategies to protect groundwater resources from contamination and, as a result, to improve human health.

Credit authorship contribution statement

K. Sankar: Writing – original draft, methodology, investigation. **D. Shanthi:** Data analysis and Review & editing. **K. Kalaivanan:** Software conceptualization and writing review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We would also like to thank the editors and anonymous reviewers for providing valuable suggestions on the manuscript.

Supplementary data

All data are provided as tables and figures.

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Figures

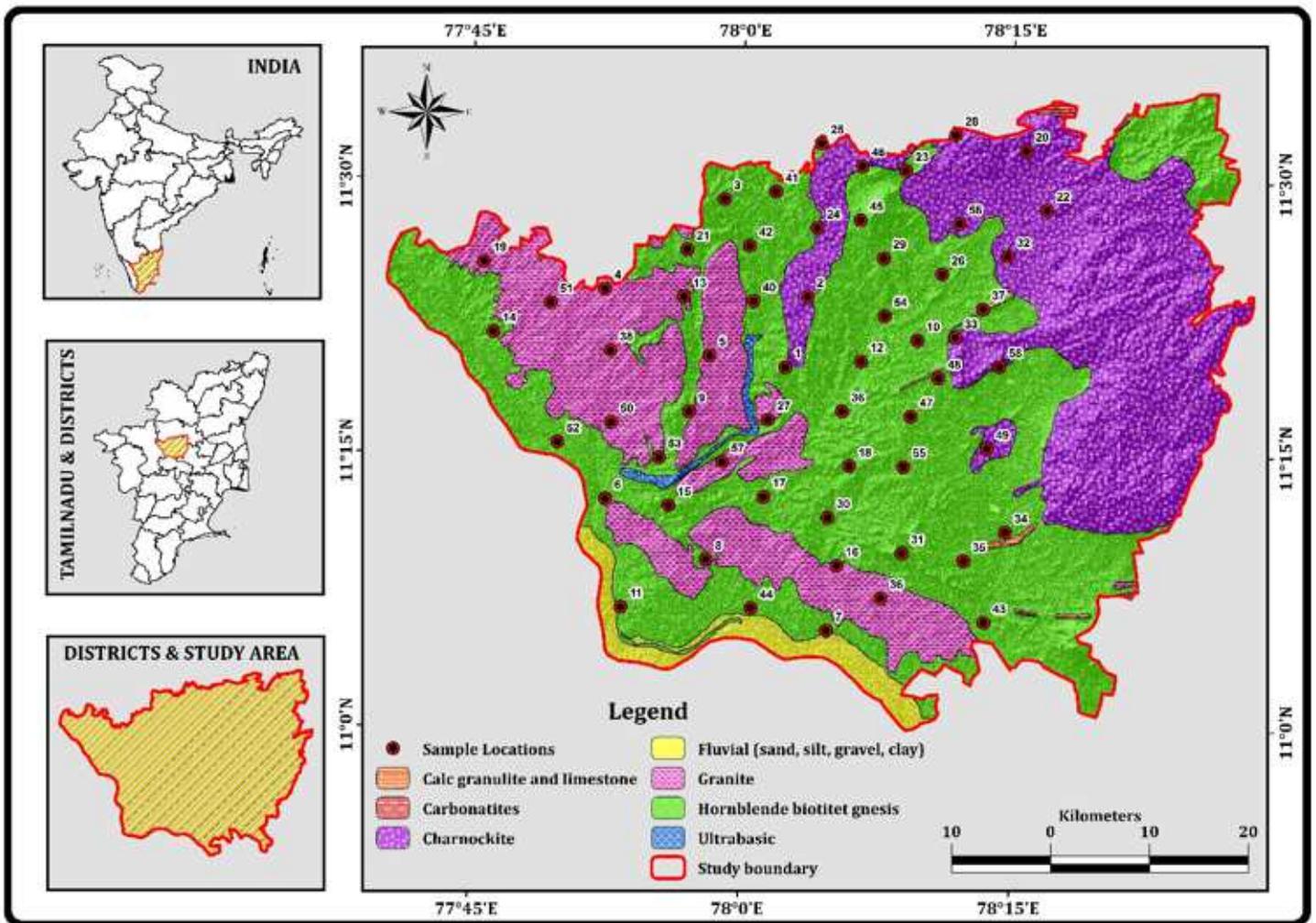


Figure 1

Location, Geology and groundwater sampling points of the study area.

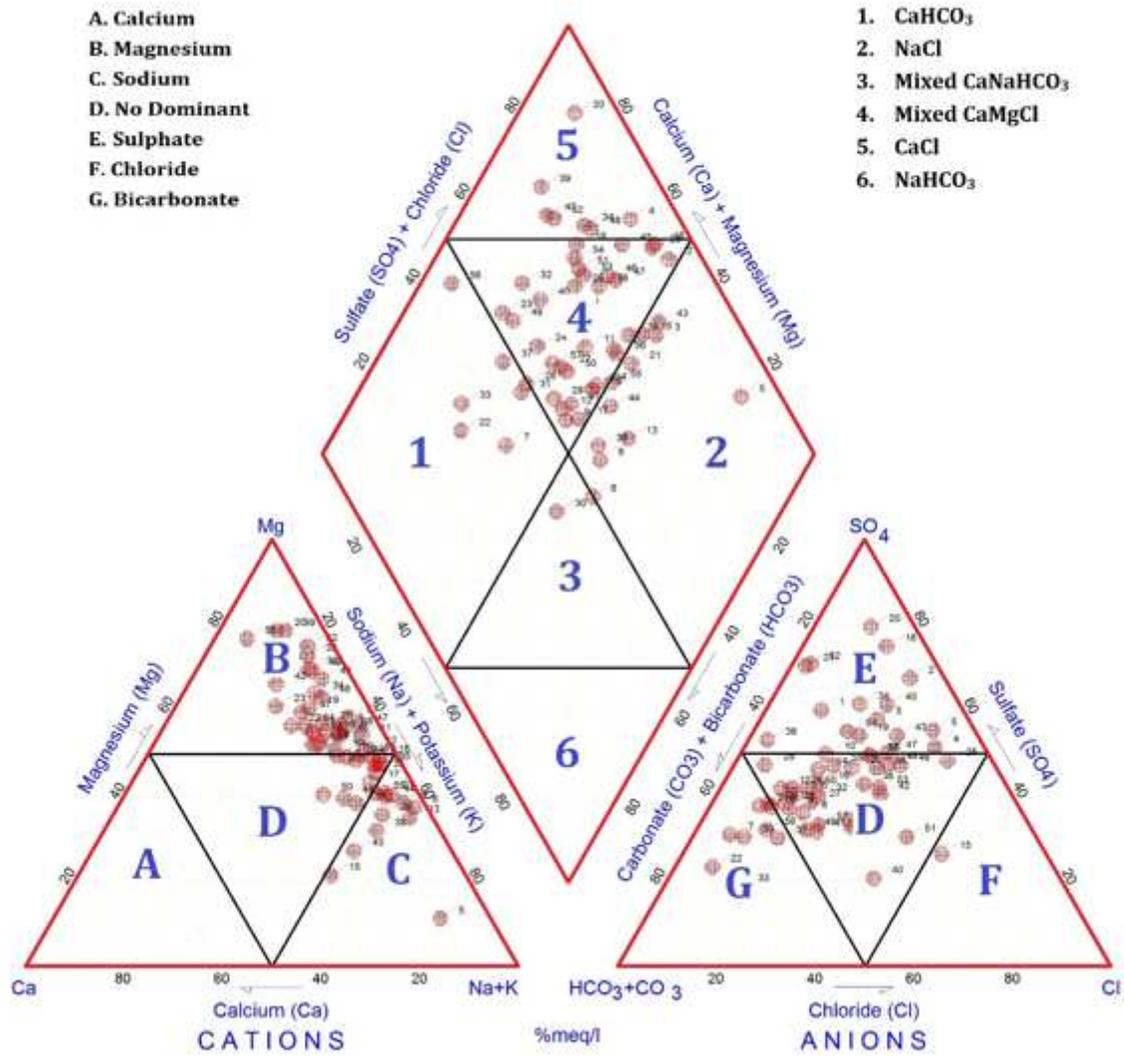


Figure 2

Hill piper plot for groundwater samples

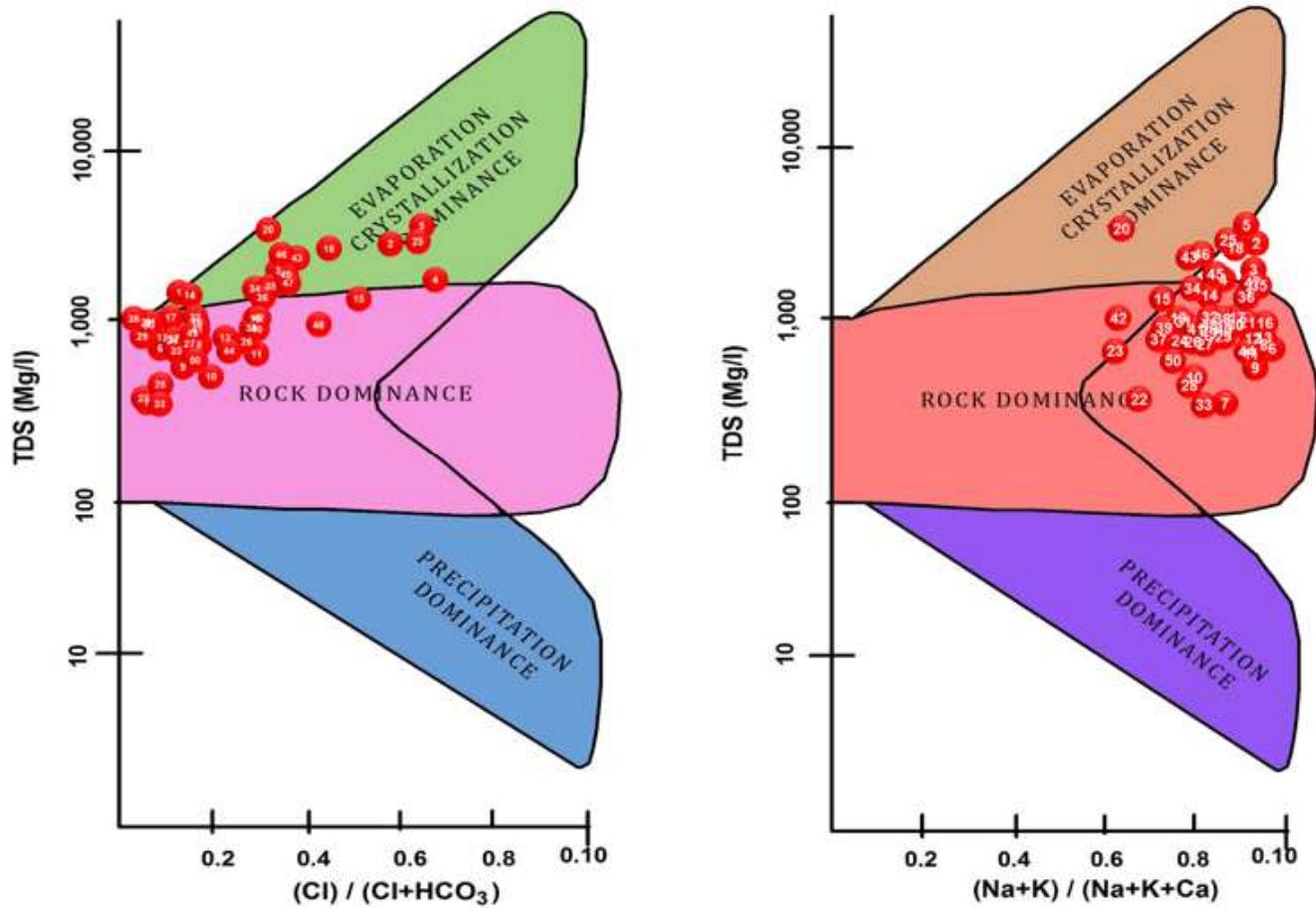


Figure 3

Gibbs plot for groundwater samples (Gibbs 1970)

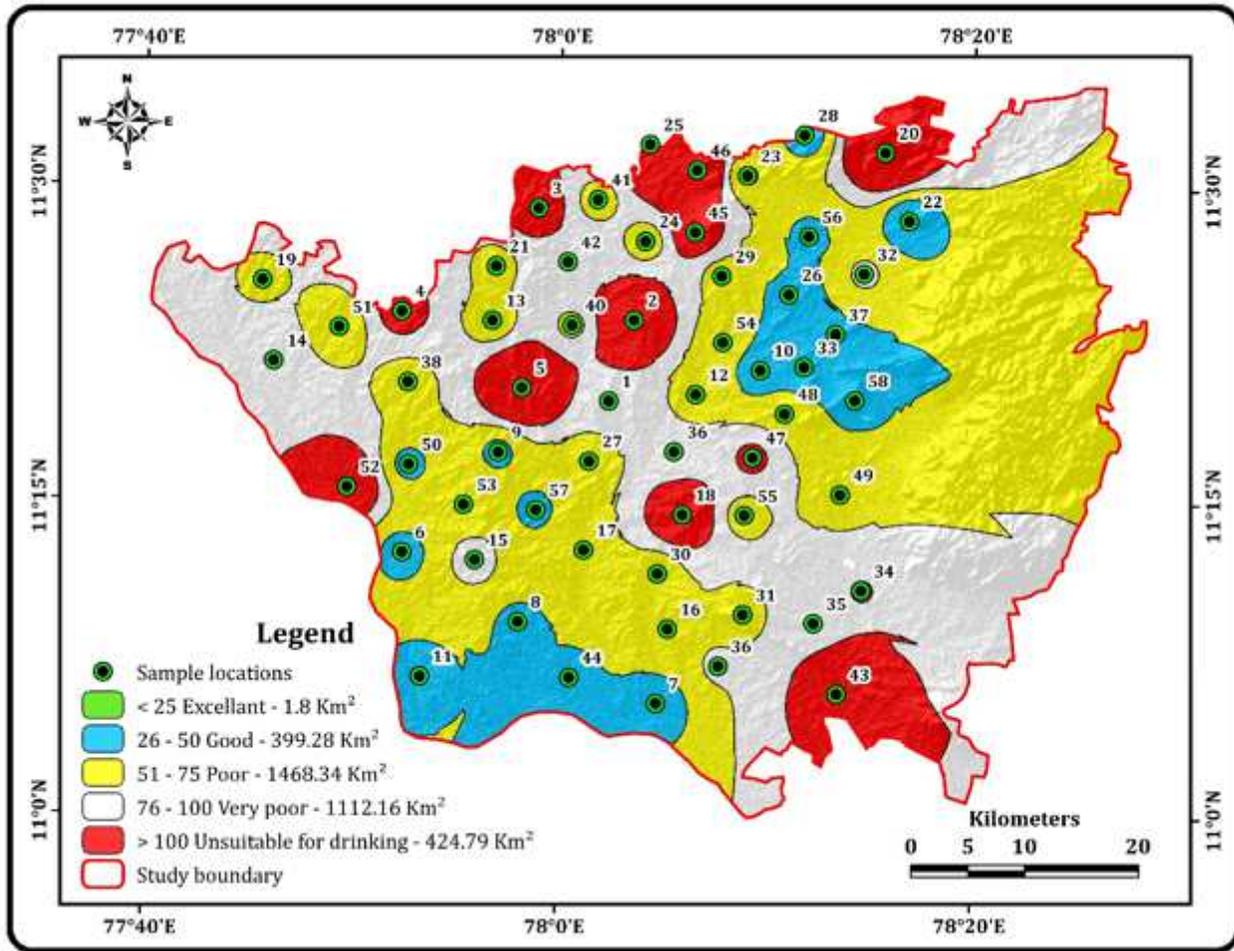


Figure 4

Spatial distribution map of Water Quality Index in the study area

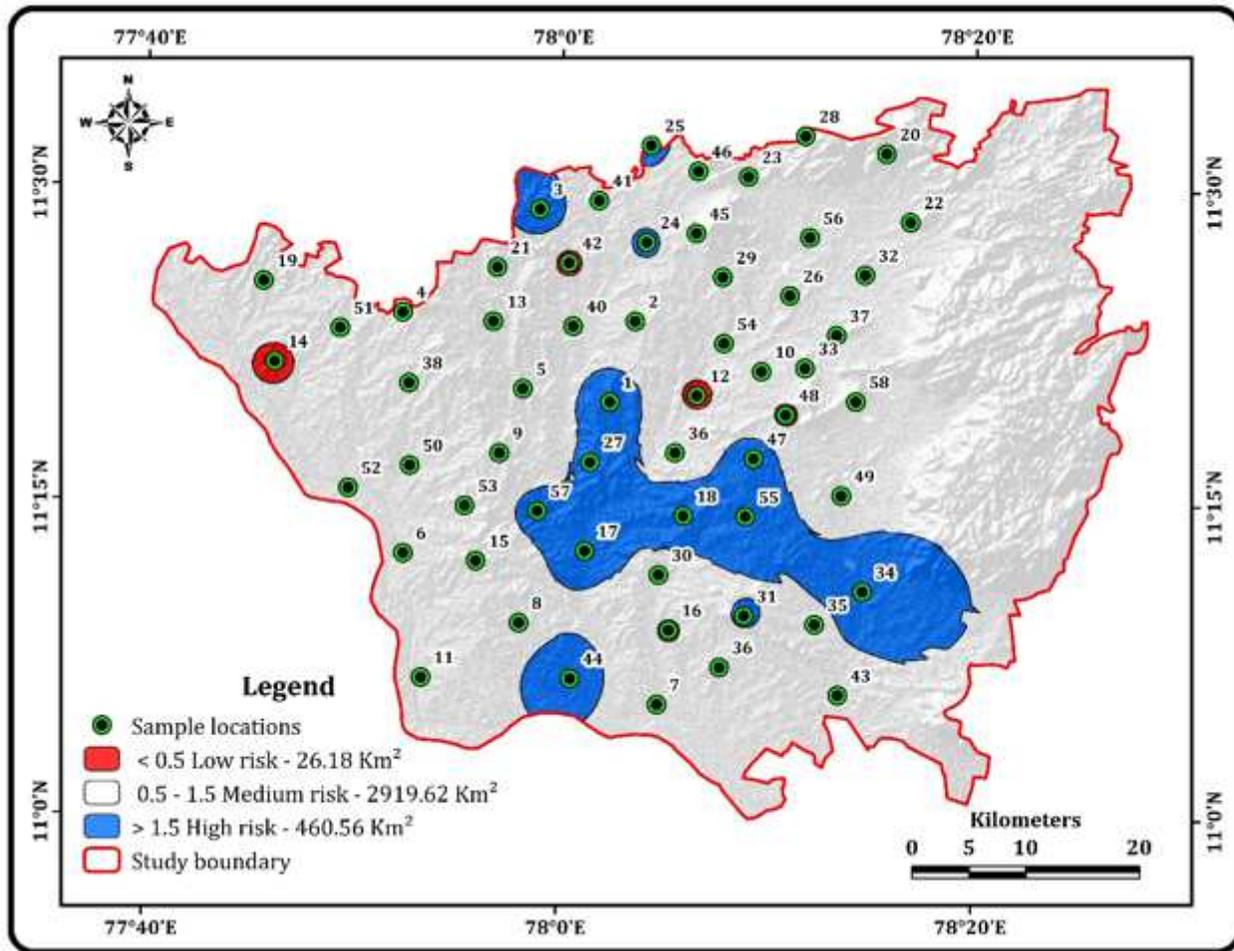


Figure 5

Spatial distribution of health risk based on fluoride concentration in groundwater of the Namakkal district

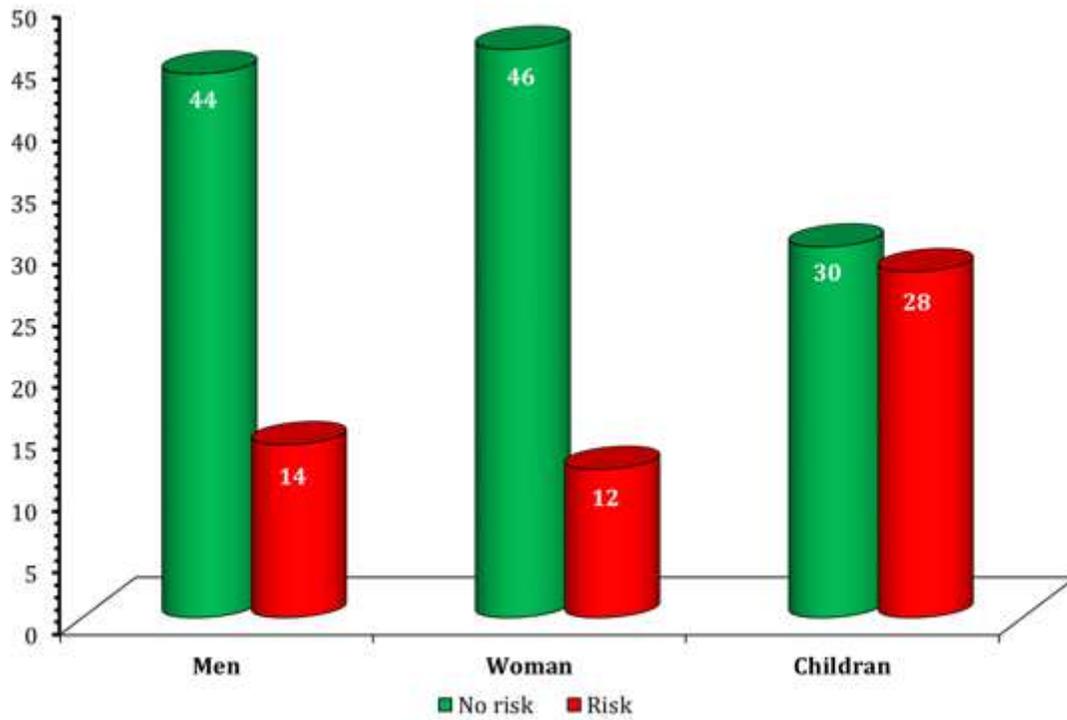


Figure 6

Health risk (Total Hazard Index) in groundwater samples of the Namakkal district. (a) Men (b) Women and (c) Children