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Evaluation of Groundwater Contamination for Fluoride in Pullamapatti Watershed, Northern Tamil Nadu, India: An Emphasis on Human Health Risk Assessment

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Abstract

The high fluoride concentration in groundwater, exceeding 1.5 mg/L in parts of Dharmapuri, Krishnagiri, Salem, and Vellore districts, poses significant public health concern. Notably, Dharmapuri district exhibits the highest fluoride levels in Tamil Nadu. Recent studies revealed that many groundwater samples from Dharmapuri district of Tamil Nadu are affected by fluoride contamination. To investigate the spatial distribution of fluoride in groundwater, hydrogeochemistry and the effect on human health, 66 water samples collected from Pullamapatti Watershed (PW) were analyzed. The samples collected from different groundwater extraction structures were analyzed for pH, EC, TDS, total hardness, alkalinity and major ions utilizing standard methods. In the case of cations, Na^+ is the most prevalent, followed by Mg^{2+}, Ca^{2+}, and K^+ and the major anions in the groundwater are ordered by proportional abundance as follows: Chloride > Bicarbonate > Sulfate > Nitrate > Fluoride. The investigation using Gibbs analysis revealed that the chief cause of groundwater contamination is rock-water interaction. Furthermore, Fluoride human health risk assessment for various age groups was performed by estimating the total hazard Index for all the collected samples. It is found that 75.7% of infants, 50% of children, and 34.28% of adults are affected by fluoride contaminated water.

Keywords: Hazard Quotient, Fluorosis, Groundwater contamination, Aquifer, Hydrochemistry

Introduction

Groundwater is a necessary natural material for mankind as it is renewable, low-cost cost and less polluted (Awadh *et al.,* 2021). Nevertheless, widespread withdrawal of groundwater is leading to over-exploitation of the resources in several areas of the globe, particularly in nations like India, where the demand for freshwater resources is rapidly increasing (Ashraf *et al.*, 2017). India is the largest global consumer of groundwater, utilizing approximately 250 billion cubic meters annually, which constitutes nearly a quarter of the worldwide groundwater consumption. Groundwater holds immense importance in India, as it supplies over 60% of the irrigation water for farmlands and supplies over 85% of the drinking water, making it an indispensable and essential resource for these regions (Saha and Ray, 2018). In India, injudicious management of water resources and environmental deterioration have sparked concerns regarding water scarcity and quality (Saha *et al*., 2017). The thresholds for various chemical constituents in groundwater to ensure its suitablility for potable use are followed after WHO, 2022 and BIS, 2022. Contamination of groundwater has direct implications for human health, particularly in arid and semi-arid terrains of India (Tiwari *et al.,* 2020).

Fluoride is the thirteenth most prevalent element in the lithosphere. Despite the vital role as a micronutrient essential for human well-being, fluoride has the capacity to pollute groundwater on a significant scale, considering it a global concern (Ghosh *et al*., 2013). Naturally raised levels of F^- found in groundwater present a worldwide health hazard, potentially affecting vast populations, especially in the developing countries (Sane *et al.,* 2025). The existence of fluoride in small concentrations (< 1.0 mg/L) in potable water generally contributes positively in reducing tooth decay, particularly among youngsters (Shomar *et al.*, 2004). Nonetheless, data from WHO (2022) highlights that the prolonged and excessive influence of fluoride (> 1.5 mg/L) can cause various harmful consequences. These include dental and skeletal fluorosis, an elevated likelihood of bone injury, diminished birth rates, enhanced susceptibility to kidney stones, compromised thyroid function, and a decline in children's cognitive development. Due to a variety of factors, like elevated pH, increased presence of Na^+ and HCO_3^- concentrations, and decreased Ca^{2+} ions, groundwater often contains elevated levels of fluoride (Jaunjalkar and Murkute, 2023). Human influence on fluoride mainly happens through the consumption of groundwater for drinking.

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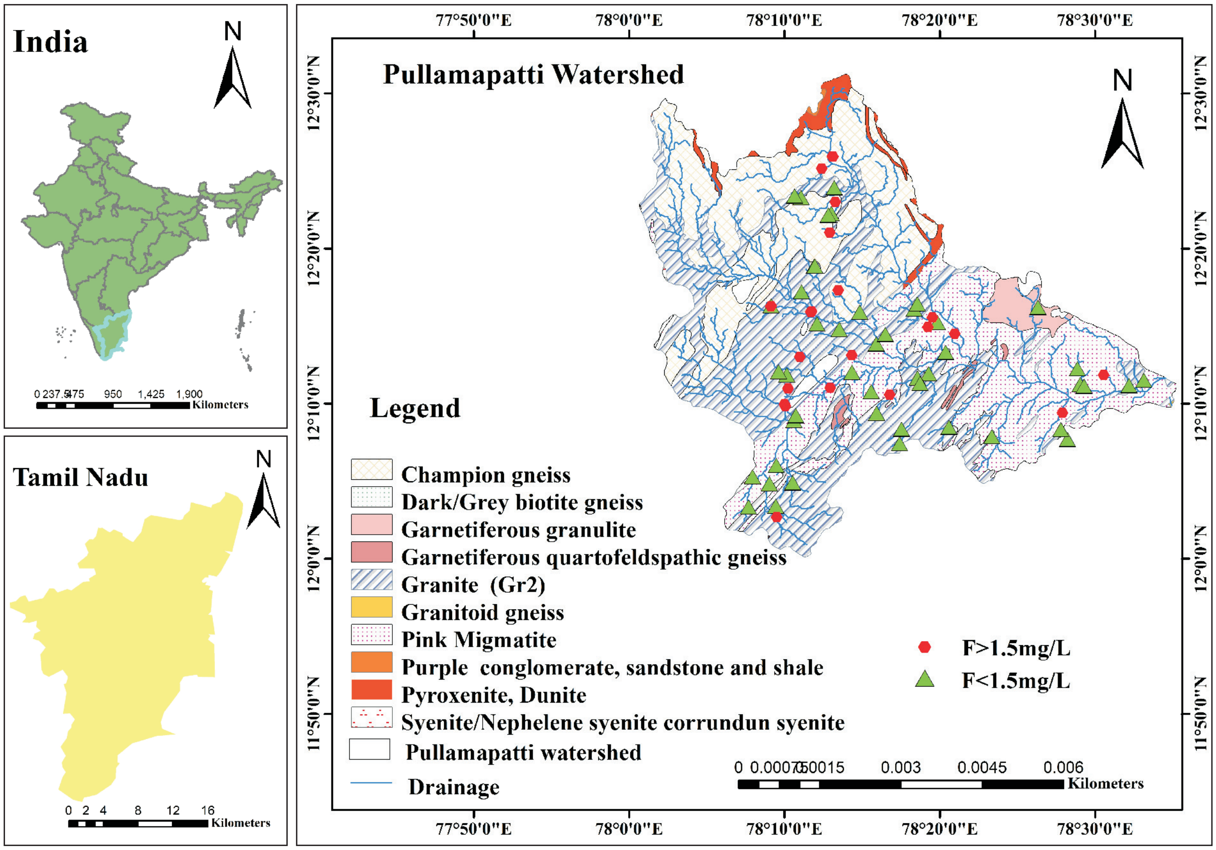

Fig.1. Geological map of the Pullamapatti watershed showing fluoride distribution (>1.5 mg/L and <1.5 mg/L) in relation to major lithological units

Board (TWAD, 2012) reported that approximately 30% of hand pump water samples in Dharmapuri and Krishnagiri districts are contaminated, significantly surpassing the state average of 6.9%. This alarming prevalence underscores the critical importance of comprehensive fluoride studies in Dharmapuri. These studies are essential to assess the extent of contamination, understand the sources, and implement effective mitigation strategies, ultimately, to maintain the health of the people. The population in Pullamapatti Watershed of Dharmapuri district primarily depends on groundwater for their drinking water requirements. This reliance leads to an increased susceptibility to health risks due to the prevalence of endemic fluorosis. The objective of the study was to analyze the mechanism of evolution of fluoride in sub-surface water and assess the related health hazards for the residents in the Pullamapatti watershed. Additionally, the study utilized the human health risk assessment framework planned by the United States Environmental Protection Agency (USEPA) to analyze the health risks posed to different age groups due to the use of fluoride-rich water. By integrating hydrochemical and geochemical analyses with advanced techniques and extensive field data, the research provides new insights into fluoride sources and contamination processes in aquifer systems. The findings enhance understanding of groundwater quality dynamics, support improved monitoring strategies, and offer valuable guidance for policymakers to identify vulnerable regions and implement targeted interventions.

Study Area

The Pullamapatti watershed of Dharmapuri district, Northern Tamil Nadu, India covers an area of 1701.69 km^2, located between 78°00' to 78°35'E long. and 12°00' to 12°31'N lat. falling in Survey of India toposheet Nos 57 L/2, L/3, L/4, L/7, L/8, and L/12 (Fig.1). The main drainage in the region is the Ponnaiyar River, which bifurcates into the Pullamapatti and Semmandakuppam rivers. The river originates in the northwest and empties into the southeast corner of the watershed. The terrain is primarily occupied by the Charnockites and gneissic rocks of the Archaean crystallines, marked by undulating plains and upland plateaus, and belong to a semi-arid region characterized by a dry and hot climate. Groundwater occurs under phreatic to semi-confined state in alluvium and crystalline rocks. The annual mean rainfall in the region amounts to 910 mm, with 82% occurring in the SW monsoon, while the remaining 12% and 6% in the NE monsoon and summer seasons, respectively.

Materials and Methods

Sampling and Analytical Procedure

As per the guidelines of APHA (2022), 66 groundwater samples from phreatic and deep aquifers and surface water bodies were sampled during May 2022 (pre-monsoon -PRM). Sampling locations were noted using a handheld GPS. A total of 1 liter of water was collected from each sampling site in clean polyethylene bottles after pumping the wells for 10 minutes. Before sampling, the bottles were rinsed with the groundwater to be collected, and all samples were stored and analyzed for the physicochemical parameters.

In situ measurements of EC, TDS, and pH were conducted using a Horiba LAQUA F-74 multi-parameter water quality analyzer. The titrimetric method involving standard EDTA was employed for the calculation of Ca^{2+} and Mg^{2+}. Chloride levels were estimated through $AgNO_3$ titration. The Na^+ and K^+ concentrations were measured using a flame photometer. The SO_4^{2-} was quantified by colorimetry using a UV-Visible Spectrophotometer. Furthermore, F^- concentrations were assessed electrochemically using the Fluoride Continuous Flow Analyzer Skalar san++.

The spatial analysis component within ArcGIS (ArcMap 10.3) was used to plot the spatial change of fluoride in the Pullamapatti Watershed. The resulting outcomes were then compared with the drinkable water guidelines of WHO (2022) and BIS (2012).

Health Risks Assessment, HRA

The HRA study was carried out by utilizing the United Nations Environmental Protection Agency (USEPA, 2004) recommended techniques and is widely employed to evaluate the potential negative impacts on health caused by groundwater exposure to various contaminants like heavy metals, fluoride, nitrate, *etc*. (Narsimha and Rajitha, 2018). Wu and Sun (2016) highlighted that the USEPA (2004) model has received widespread use and application all over the world, particularly in India, because human health risks have been raised in the last ten years owing to the use of polluted water in semi-arid and desert areas. Humans in the Pullamapatti watershed were exposed to fluoride through direct and indirect oral intake of groundwater.

For evaluating possible health threats associated with fluoride exposure through oral ingestion, specific parameters are considered for different age groups. Various parameters used for the HRA in the current study are compiled (Table 1).

To determine the average daily dosage (ADD) by groundwater consumption for adults, children, and infants individually, Equation (1) has been used.

$$\text{Oral Intake (ADD)} = \frac{Cw \times IR \times EF \times ED}{ABW \times AET} \quad \text{(USEPA, 2014)...1}$$

(IR-Ingestion Rate; EF-Exposure Frequency; ED- Exposure Duration; ABW- Average Body Weight; AET- Average Timing; Cw- Contaminant concentration in water)

The THI is the non-carcinogenic risk index associated with Fluoride (Equation-2) (USEPA 2004).

$$THI = \frac{ADD\,(Oral)}{RfD\,(Oral)} \quad ...2$$

RfD is the reference dose of fluoride, 0.06 mg/kg/day (USEPA, 2012; Craig *et al*., 2015).

According to USEPA HRA criteria, the non-carcinogenic risk THI level is 1(Zhang *et al*., 2018). The THI > 1 implies that the health risk caused by non-carcinogenic chemicals is unacceptable, and THI < 1 shows that the health risk caused by non-carcinogenic constituents is within a permissible level.

Results and Discussion

Drinking Water Quality

Various hydro-geochemical parameters of the water samples were analysed and compared with different guidelines (Table 2). In the Pullamapatti watershed, groundwater samples have pH values mostly exceeding 7, varying from 6.6 to 8.79, with a mean of 7.4. Only a few numbers of samples exceed 8, while the majority fall within the WHO (2022) maximum permissible limit of 7-8.5 for potable water. Groundwater salinity, varies significantly, from 781 to 6780 µS/cm at 25°C, with mean of 2268 µS/cm at 25°C. The EC categorization into three types, about 50% of samples fall into type II, indicating low salinity. Approximately 22% fall into type I, signifying minimal salt enrichment, and a few types III, suggesting high salt enrichment. Various factors, including weathering of aquifer materials and human activities, influence elevated EC levels. The TDS values range from 531 to 4746 mg/L (ava. 1547 mg/L), with a majority surpassing the WHO's maximum permissible limit (MPL) of 1000 mg/L. Less than 20% register TDS concentrations lower than the MPL of 1000 mg/L. The Total Hardness (TH) is less than the permissible level of 500 mg/L, with the majority falling within the hard and very hard water classes based on TH classification (Sawyer and McCarthy, 1967).

The Na^+ levels range from 33 to 990 mg/L, beyond the WHO's MPL of 200 mg/L. Mg^{2+} varies from 13 to 400 mg/L, exceeding the recommended limit of 100 mg/L. Ca^{2+} concentrations range from 10 to 236 mg/L, and K^+ concentrations range from 1 to 43 mg/L, with only 5% samples surpassing the MPL of WHO, 2022. Various minerals like amphiboles, pyroxenes, feldspars, fluorite, and apatite in granitic terrains contribute Ca^{2+}, Mg^{2+}, Na^+, and K^+ to the groundwater (Adimalla and Venkatayogi, 2017). Cl^- dominates as an anion, with concentrations varying from 92 to 1370 mg/L, notably exceeding 250 mg/L in 20% of the samples, pointing to significant human influence on groundwater. Sources of Cl^- may be weathering of Cl^- bearing minerals in rocks, domestic effluents, chemical fertilizers, and rainfall (Kumar *et al*., 2014). HCO_3^-

Table 1: Exposure frequency and obtained values of health risk parameters

Parameters	Infant	Children	Adult	Reference
IR, L/day	0.7L/day	0.78L/day	2.5L/day	USEPA 2004; USEPA 2014
EF, days/year	365days	365days	365days	USEAP, 2014
ED, years	1.5	15	50	USEPA 2004; Huang *et al*., 2018
ABW, Kg	6.9	18.7	57.5	Narsimha and Rajitha, 2018
AET, days	2190	5475	18,250,	Huang *et al*., 2018; USEPA 2014; WHO 2022
Cw, mg/L	Fluoride (0.19 to 3.60mg/L)			Current study

Table 2: Hydrochemistry of Groundwater samples of Pullamapatti Watershed (Premonsoon-2022)

Water quality parameter (IS 10500: 2012)		BIS Limit Acceptable	WHO Guideline Value (2022)	Concentration (Minimum-Maximum)	Average
pH	-	6.5-8.5	7.0 - 8.5	6.66 - 8.97	7.4
EC	µS/cm	-	- 1500	718 - 6780	2268
TDS	mg/L	500-2000	<1000	531 - 4746	1547
TH	mg/L as $CaCO_3$	200-600	Not specified	65 - 1865	638
Ca^{2+}		75-200	Not specified	10 -236	116
Mg^{2+}		30-100	Not specified	13 - 400	127
Na^+	mg/L	50 -200	- 200	33 -990	214
K^+		-	-	1-43	5
HCO_3^-		-	-	175 - 585	348
Cl^-		250-1000	250	92 -1370	445
SO_4^{2-}		200-600	250	74 -267	122
F^-		1.0 -1.5	1 - 1.5	0.19 -3.60	1.60

primarily arises from the disintegration of silicate minerals (Adimalla and Venkatayogi, 2018). SO_4^{2-} and HCO_3^- concentrations in the water samples vary between 175 and 585 mg/L: 74 and 267 mg/L, respectively.

Fluoride Variation

The occurrence of a high fluoride levels is of major concern in many countries, particularly in India, where the greater part of the population rely on groundwater for drinking purposes. Roughly 66 million individuals in India, with a significant portion being around 6 million children, experience the impacts of skeletal and dental fluorosis because of consistent ingestion of groundwater with elevated fluoride levels (Adimalla and Li, 2018).

Maintaining an optimal level of F^- in potable water is crucial for ensuring proper skeletal growth and the development of dental enamel (Uriah *et al*., 2014). In certain countries, fluoride is intentionally introduced to dietary salt or water used for potable water, typically at concentrations spanning from 0.5 to 1 mg/L. The limit of fluoride and the classification are compiled (Table 3).

Groundwater in the Pullamapatti watershed contains elevated fluoride levels. Measured fluoride concentrations ranged from about 0.19 to 3.6 mg/L (mean ≈ 1.6 mg/L) (Table 2), and roughly one-third of samples exceeded the WHO (2022) and BIS (2012) drinking-water guideline of 1.5 mg/L. The distribution of F^- in the groundwaters of Pullamapatti watershed (Fig.1) reveals that higher concentrations are clustered in specific parts of the watershed, suggesting links to local hydrogeology. The study indicates that fluoride levels exceeded the MPL of WHO (2022) of 1.5 mg/L for potable purposes in more than 39% (27 Nos.). Areas with notably high fluoride concentrations were reported in the villages of Dharmapuri (1.6-2.13 mg/L), Karimangalam (3.26 mg/L), Manicknoor (2.22mg/L), Mittanahalli (2.39mg/L), Timmanapuram (1.89mg/L), Nallampalli (1.92mg/L), Bandarahalli (1.81mg/L) and Kongarapatti (1.6 mg/L). The variation of fluoride along Pullamapatti watershed is depicted in the figure 2.

The dissolution of fluorine-containing minerals like fluorite, biotite, hornblende, and fluorapatite found in the underlying crystalline bedrock of gneisses and granites is the primary geogenic cause of the elevated fluoride levels in the Pullamapatti groundwater. The role of cation exchange and silicate weathering is indicated by the positive correlations with Na^+ and HCO_3^- and the inverse correlation with Ca^{2+}, which support the idea that rock–water interaction under alkaline conditions enhances fluoride mobilization (Su *et al*., 2013). This semi-arid aquifer high pH and bicarbonate levels further encourage fluoride desorption, as shown in areas where concentrations are higher than 1.5 mg/L. Other semi-arid regions, such as Tiruchirappalli, Tamil Nadu (0.22–4.5 mg/L; Vinothkanna *et al*., 2021), Rajasthan (0.1–25 mg/L; Ayoob and Gupta, 2006), and Nalgonda, Andhra Pradesh (0.1–8.8 mg/L; Brindha *et al*., 2011), have also reported similar geogenic controls.

Both shallow dug wells (3–25 m) and deeper bore wells (60–90 m) in the Pullamapatti watershed have high fluoride concentrations (>1.5 mg/L), suggesting contamination at all aquifer depths. Hotspots in Pullamapatti are concentrated in villages like Dharmapuri, Karimangalam, and Manicknoor, which is similar to Tiruchirappalli, where extended rock–water interaction increases fluoride in deeper aquifers. The weathering of charnockites and epidote-hornblende gneiss is the main geogenic source, and fluoride content in accessory minerals like biotite and hornblende is high (1.1% and 2.9%, respectively; Jagadeshan *et al*., 2015). Furthermore, fluoride contamination may be exacerbated by anthropogenic inputs from phosphate-based fertilizers, which contain fluorine as a contaminant, further seeping into groundwater through irrigation return flows (Su *et al*., 2013).

Table 3: Health risk effects associated with F- ingestion in individuals in the study area

Class	Limit of Fluoride (mg/L)	Impact on Human Well-being (Dissanayake 1991; WHO 2022)	Fluoride (%)
Class I	<0.5	Dental caries decay-promoting	14
Class II	0.5 -1.5	Supports the growth of healthy teeth and bones	47
Class III	1.5 - 4	Mottling of teeth	39
Class IV	4 - 10	Dental and skeletal fluorosis (back and neck bone pain)	0
Class V	>10	Severe fluorosis	0

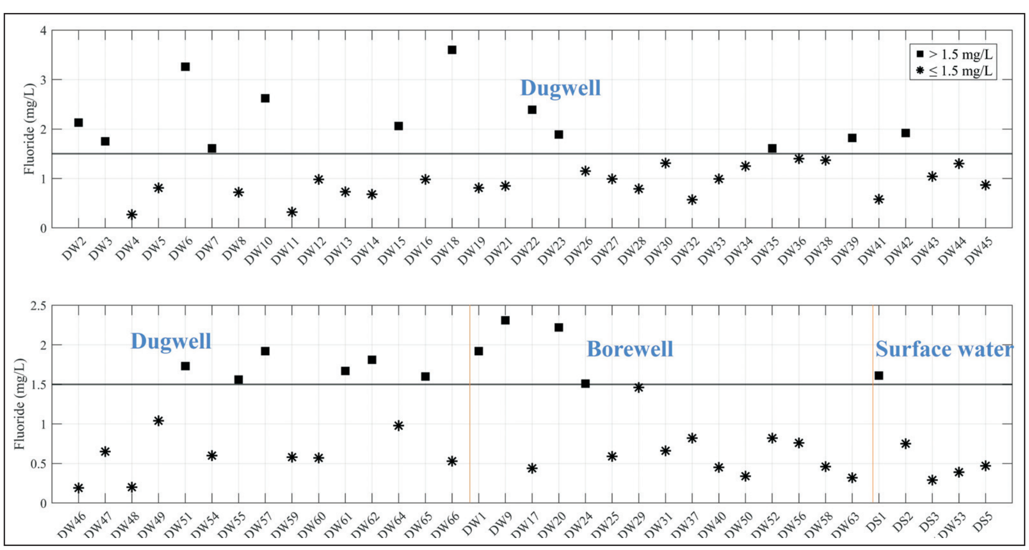

Fig.2. Fluoride concentration in the waters of dugwell, borewell, and surface water with respect to permissible limit (BIS, 2012; WHO, 2022)

Human Health Risk Assessment (HRA)

A combination of natural, geological, and anthropogenic factors shapes the composition of groundwater. In the hard rock terrains of Pullamapatti watershed, fluoride-bearing minerals like fluorite, biotite, and hornblende, etc. significantly contribute to the fluoride content through prolonged interaction with percolating groundwater. The extent of these geogenic contributions is further influenced by hydrochemical conditions like high pH, subtle calcium concentrations, and elevated sodium and bicarbonate levels, and these enhance fluoride mobility. Understanding these geochemical mechanisms is fundamental, but the ultimate concern revolves around the potential health risks arising from consuming fluoride-contaminated groundwater. As such, an evaluation of groundwater chemistry serves as the basis for conducting an HRA, which quantifies the non-carcinogenic risks associated with fluoride exposure among different age classes. By translating chemical data into health-related indices such as Total Hazard Index (THI), the HRA provides a practical framework to assess the severity of exposure, identify vulnerable populations, and inform mitigation strategies for safe water use in the region.

The groundwater within the Pullampatti watershed, contaminated with fluoride (F^-) known to pose non-cancer risks to human health, was evaluated for human HRA. The outcomes are indicated as Total Hazard Index (THI) and are calculated by comparing estimated daily ingestion and reference tolerable daily dose (RfD-0.06 mg/kg/day).

The HRA involved calculating potential oral exposure to groundwater for adults, children, and infants, considering risks that are not related to causing cancer. Consequently, increased levels of F^- in the potable water give rise to health dangers within the area. The average fluoride concentration, measured through the THI, remains below 1, indicating minimal potential risk. Spatial variation of THI along Pullamapatti watershed for infants, children, and adults affected by non-carcinogenic fluoride contamination is depicted in figure 3. The THI displays variations, ranging from 0.137 to 2.36 for adults, 0.19 to 3.27 for children, and 0.30 to 5.60 for infants. The mean THI values were recorded as 0.871014 for

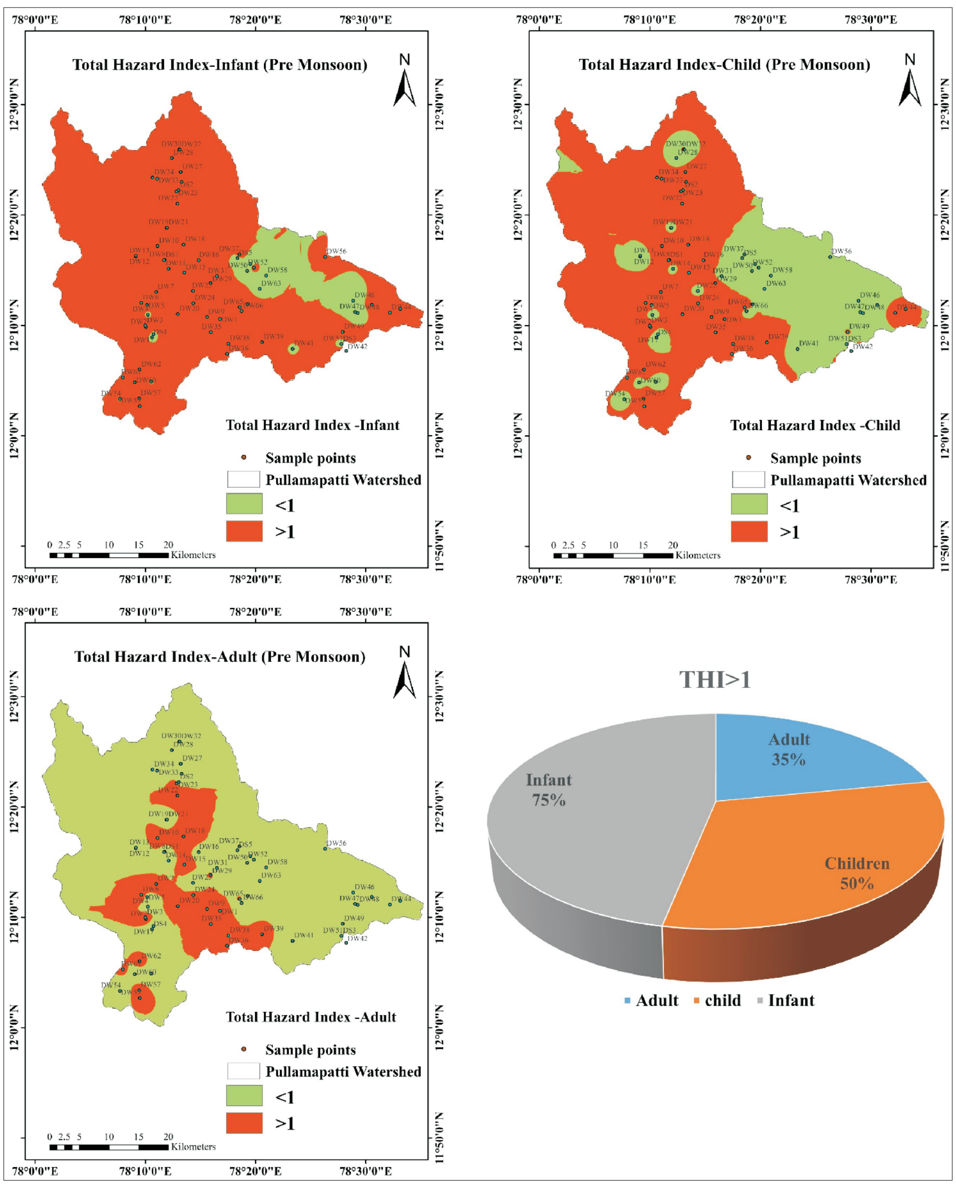

Fig.3. Spatial variation of THI along Pullamapatti Watershed for infants, children, and adults affected by non-carcinogenic fluoride contamination

Table 4: Human health risk assessment by Total Hazard Index of Fluoride in groundwater

##	THI. (Max.)	THI. Min.)	THI. (Average)
Infant	5.6	0.30	1.87
Children	3.27	0.190	0.8356
Adult	2.36	0.137	0.871014

adults, 0.8356 for children, and 1.87 for infants (Table 4). The THI mean strength of F^- is less than 1, signifying that these elements pose little hazard. Infants, children, and adults were vulnerable to fluoride contamination in potable water, with a high risk of 75.7%, 50%, and 34.28%, respectively (Fig. 3). The percentage values represent the proportion of groundwater samples with THI >1 for each age group, reflecting the fraction of samples indicating potential non-carcinogenic risk.

More than 34% of the samples go beyond the acceptable limit for non-carcinogenic risk, which is indicated by a THI > 1. According to the HRA, the Pullamapatti watershed showed harmful health issues in the order of severity: infants, > children, > adults, as depicted in Fig.3. These findings were reported by Narsimha and Rajitha (2018) and Adimalla and Li (2018) in different terrains of India.

Groundwater Chemistry Evolution Mechanism

Understanding the source and evolution of groundwater chemistry is vital for evaluating potential health risks related to raised fluoride concentrations. In order to explain the geochemical development of groundwater, Gibbs (1970) showed that the Gibbs plot is an effective tool for distinguishing between precipitation, evaporation, and rock dominance. The Gibbs plot in this study shows how fluoride-containing minerals have a significant lithological influence on groundwater chemistry in crystalline terrains. These revelations support the findings of the Health Risk Assessment and highlight the critical role that geogenic processes in particular, the interaction between rocks and water-play in fluoride enrichment and the associated health effects. Adimalla and Li (2018) also emphasized that slower percolation through interstices and longer residence times increase ion concentrations in crystalline regions with higher temperatures and less rainfall, which in turn shapes the hydrochemical character of groundwater. As depicted in Fig. 4a, groundwater samples with high fluoride content tend to fall within the rock predominance field and are slightly leaning towards the evaporation predominance field. This indicates that rock dominance is a substantial factor regulating groundwater chemistry and plays a major role in elevating the fluoride in the crystalline in the Pullamapatti watershed.

High fluoride concentrations in groundwater are largely governed by geogenic sources and hydrochemical processes, particularly under alkaline conditions. Aquifers characterized by high pH and intense evapotranspiration favour fluoride enrichment (Jacks *et al*., 2005), with elevated levels often occurring in fracture zones of fluoride-bearing rocks where prolonged water–rock interaction enhances mineral weathering and dissolution. The mechanism of fluoride mobilization into groundwater is illustrated in the evolution pathway (Fig. 4b), highlighting the role of fracture networks and water–rock interaction in fluoride release (Mukherjee and Singh, 2018). In semi-arid regions like the Pullamapatti watershed, the alkaline nature of groundwater, marked by higher Na^+

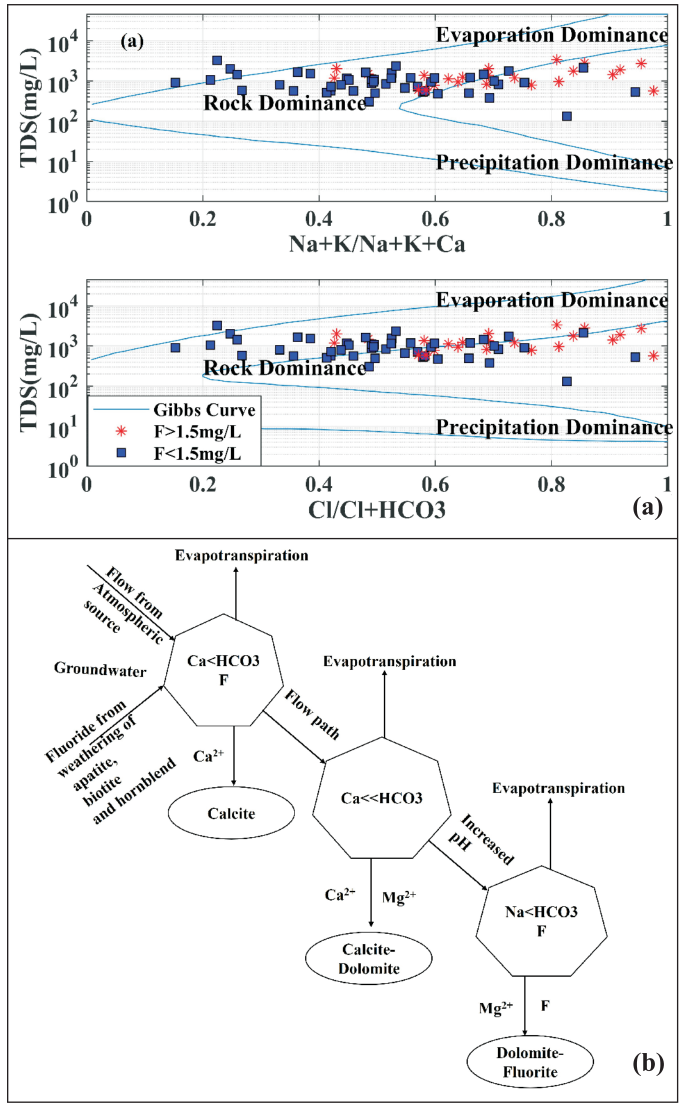

Fig.4. a) Gibbs plot showing hydrogeochemical processes controlling groundwater chemistry in Pullamapatti watershed; b) Flowchart illustrating the fundamental process of fluoride ion (F^-) evolution (*Modified after* Jacks *et al.,* 2005; Karthikeyan *et al.,* 2010)

and HCO_3^- and lower Ca^{2+} concentrations, further promotes fluoride mobilization (Fig. 5). Similar patterns of fluoride enrichment driven by geogenic controls have also been widely reported across semi-arid parts of India (Adimalla and Venkatayogi, 2018).

Conclusions

The present work investigates the geo-spatial variation and HRA of fluoride contamination in Pullamapatti watershed, Northern Tamil Nadu, India. The study revealed that 27 samples have fluoride concentration more than the BIS (2012), and WHO (2022) guidelines. Dharmapuri (1.6-2.13 mg/L), Karimangalam (3.26 mg/L), Manicknoor (2.22mg/L), Mittanahalli (2.39mg/L), Timmanapuram (1.89mg/L), Nallampalli (1.92mg/L), Bandarahalli (1.81mg/L), and Kongarapatti (1.6 mg/L) with high fluoride values in the Pullamapatti watershed. It is found that the major reason behind the fluoride enrichment in groundwater is owing to rock-water interaction. In the Pullamapatti watershed, the Health Risk Assessment (HRA) found that the non-carcinogenic risks from

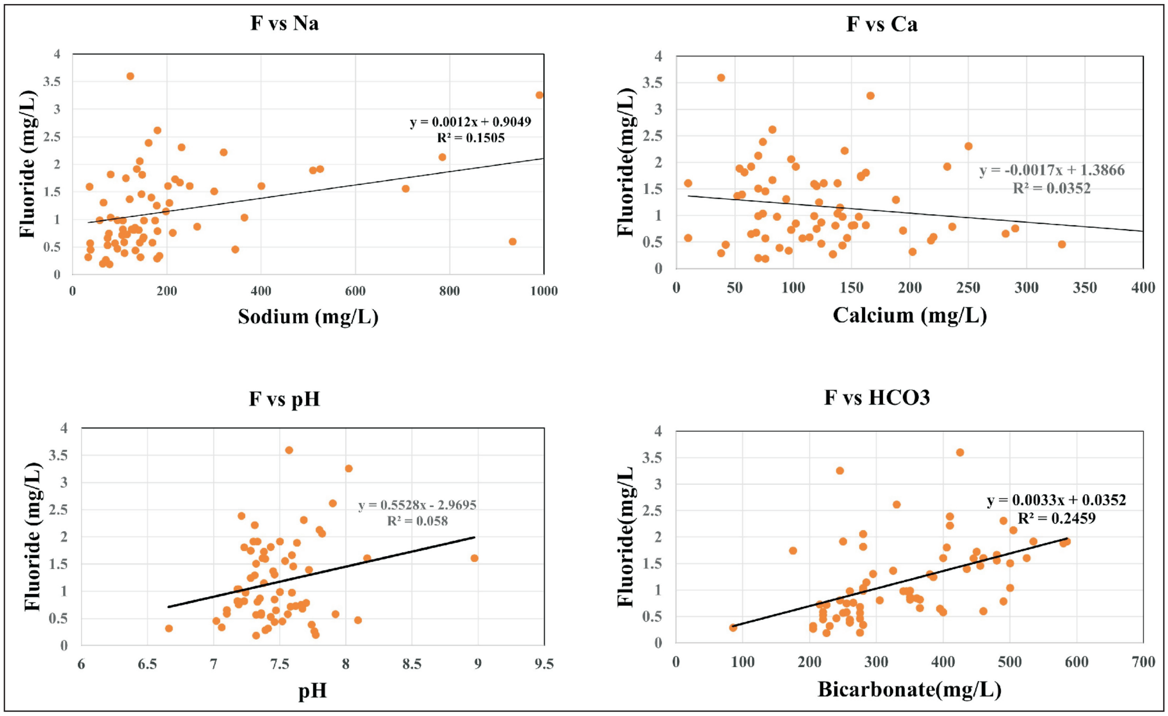

Fig.5. Cross plot showing relation between F *vs* Na, F *vs* Ca, F *vs* pH, F *vs* HCO_3

fluoride in potable water were higher than the permissible limit (THI > 1) for adults, children, and infants, suggesting possible negative health effects. The amount of water consumed per day and the fluoride concentration are directly related to serious these risks. Effective defluoridation techniques, community awareness, alternate safe water sources, and routine water quality monitoring is crucial for reducing this problem. For sustainable management and public health protection, it is essential for advance research on fluoride removal technologies, strengthen health surveillance, and encourage medical assistance. Strict regulatory enforcement and stakeholder collaboration are also essential to ensure effective implementation of groundwater protection and pollution control measures.

Authors' Contributions

DAS: Contributed to the conception, Field investigation, analysis, data processing, and drafting of the study. JVS: Provided conceptualization, supervision, data interpretation, and manuscript editing. DS: Review and editing of the manuscript

Conflict of Interests

The authors declare that they have no conflict of interests.

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