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Original research article

Assessment of Ground Water Quality with Fluoride Contamination and Its Risk Quantification

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ABSTRACT

Freshwater demand has surged due to urbanization, population growth, and domestic use, while both natural and human activities have degraded water resources. Monitoring water quality and availability is therefore vital. Mathura, a well-known religious and tourist hub in Uttar Pradesh (UP), India, was selected to assess groundwater quality with respect to fluoride (F-). Groundwater samples were collected from seventeen tube wells and hand pumps across the district during 2021–22. Fluoride concentrations ranged from 0.7 to 3.3 mg/L. Notably, 12 out of 17 samples exceeded 1.5 mg/L, surpassing the Bureau of Indian Standards (BIS, 2012) permissible limit for drinking water. Health risk assessment indicated a heightened likelihood of non-carcinogenic hazards from oral fluoride intake, particularly at sampling site SS8. Among the exposed groups, children were the most vulnerable, followed by teenagers and adults. These findings highlight a pressing health concern in the region. The study provides valuable insights for policymakers, offering data to support targeted interventions and management strategies aimed at ensuring safe groundwater supply for domestic consumption and drinking in Mathura.

Keywords: Groundwater; Fluoride; Health risk; Mathura; Water quality

1. Introduction

Fluoride is a naturally occurring electronegative element which is very reactive. It can generate ionic or covalent fluorides with the majority of the elements by combining with them. Fluorides occur mainly in

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Table 1. Fluoride levels in groundwater are distributed across India.

S. No.	Ranges of concentration of Fluoride	Location (Study Area)	References
1	0.51 – 4.0	Tamil Nadu State	[3]
2	0.1– 40	Mehsana, Gujarat	[4]
3	0.2 – 6.4	Shivpuri, Madhya Pradesh	[5]
4	0.35–8.35	Vishakha-patnam, Andra Pradesh	[6]
5	1.0 – 7.4	Eastern and Southern Karnataka	[7]
6	0.16–10.10	Nayagarh , Orissa	[8]
7	0.56–5.80	Anantpur, Andra Pradesh	[9]
8	1.51– 5.75	Palghat, Kerala	[10]
9	0.48 -6.7	Sonbhadra, Uttar Pradesh	[11]
10	0.08 -11.3	Malpura, Rajasthan	[12]
11	0.1 -4.0	Mettur Taluk, Salem, amilnadu	[13]
12	0.28 – 11.5	Bassi, Jaipur, Rajasthan	[14]
13	0.3 – 9.0	Jhajjar, Haryana	[15]
14	0.4 – 5.8	Markapur, Andra Pradesh	[16]
15	0.01-18.55	Gharbar, Jharkhand	[17]

regions that have semi-arid environments, alkaline soils and crystalline igneous rock. A significant quantity of fluoride may also be contributed to the environment by human actions, in addition to the natural sources. Fluoride concentrations are mostly caused by the natural contamination of groundwater. Fluoride is one of the less important chemical constituents that are found in natural water and is one of the characteristics that determine water quality. There is a strong correlation between chemical environment and monitoring of water quality due to the fact that physical and chemical properties of water directly impact its quality. Groundwater is a major source of water for agriculture, domestic and drinking purposes. Globally, 33% of people use groundwater for drinking purposes [1].

In India, there is an issue with excessive fluoride consumption which affects the lives of 2.5 million people who live in over 8700 villages. The issue of too much fluoride levels in India's ground water was firstly brought to light in the year 1937 in Andhra Pradesh [2]. Table 1 depicts the findings of studies conducted by a variety of researchers on groundwater's quality with regard to fluoride in a number of different locations in India [3–17]. A significant fluoride presence in the water can lead to dental and skeletal fluorosis, despite the fact that it is proven to be good for human health in avoiding dental caries when consumed in controlled doses (less than 1 ppm) [18].

Fluorosis has been confirmed to be prevalent in seventeen distinct Indian states. Over sixty-two million inhabitants in India, along with six million children, have fluorosis because of drinking water with high fluoride contamination. Due to the high concentration of fluoride in drinking water, about 18 out of 19 districts in Gujarat are at risk of developing fluorosis [19].

Fluoride can enter the body in a variety of ways, including through water, medications, industrial exposure, food, cosmetics, fertilizers and other sources. However, drinking of water is the primary contributor (75–90%). Ali et al. (2024) performed a qualitative evaluation of groundwater in the rural district of Mathura and identified high fluoride concentrations in 27.27% of samples from the region. The computed average Hazard Quotient (HQ) values for fluoride in children, teenagers, and adults were 1.88, 0.98, and 0.90, re-

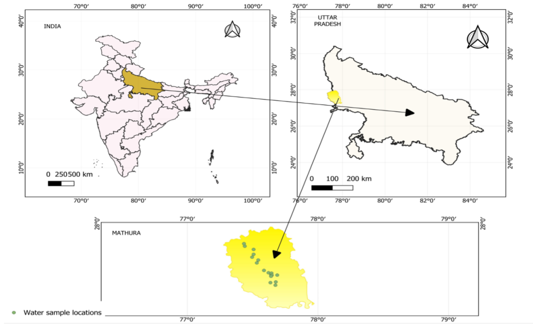

Fig. 1. Research area description.

spectively. The findings indicated that the highest hazard quotient (HQ) of fluoride exposure was recorded in a rural area of the Mathura region, measuring 2.67 and 10.75, respectively. The US Environmental Protection Agency (USEPA) advises against an HQ 1, since it results in significant non-carcinogenic diseases and cancer hazards associated with fluoride ingestion from groundwater [20].

The investigation conducted by S. Ahmed et al. (2020) of the collected samples in the study region indicates fluoride values ranging from 0.03 to 1.71 mg/L and 0.41 to 191 mg/L, respectively. The aforementioned analysis indicates that numerous villages failed to comply with the World Health Organization's guidelines [21].

2. Materials and Methods

2.1 Study area

The Mathura district is located in the west of Uttar Pradesh situated between the latitudes 27°15' & 27°56' N & the longitudes 77°16' & 78° 13' E. This area has a semi-arid and subtropical humid climate with mean value of 625 millimeters of rainfall per year. Temperatures range from 2 degrees Celsius to 46 degrees Celsius all year long. The research area experienced a rapid increase in urbanization and industrialization as a direct result of the attainability of crude materials & a perpetual water supply sourced from ground water. Groundwater exists under unconfined and semi-confined conditions. The region of study is depicted in Fig. 1.

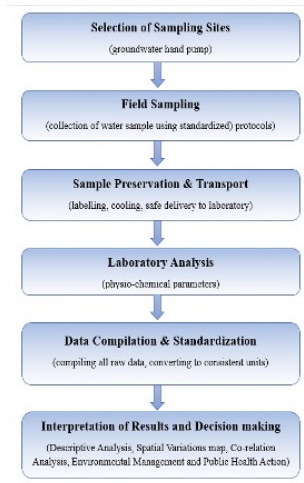

Fig. 2. Methodological flowchart.

2.2 Sample collection and analytical procedure

During March to June, 2021-22, water samples were collected from the different groundwater sampling sites in Mathura district. In order to prevent any kind of contamination, the samples were extracted by holding the vial upside down while doing so. Fig. 2 show the methodological framework. The concentration of Total dis-

Table 2. Groundwater quality characteristics with respective values.

S. No.	Sample No.	Temperature (^○C)	pH	Electric Conductivity (ms/cm)	TDS (mg/L)	Fluoride (mg/L)
1	SS1	26.9	6.70	1.100	870	0.7
2	SS2	27.5	7.02	1.230	900	0.9
3	SS3	27.3	6.95	1.251	890	1.0
4	SS4	26.5	6.66	3.350	2420	1.9
5	SS5	26.3	6.91	3.220	3190	2.1
6	SS6	27.1	7.66	3.302	3095	2.4
7	SS7	26.4	6.69	4.050	3380	2.6
8	SS8	27.6	7.02	4.210	3750	3.3
9	SS9	26.4	7.52	4.108	3490	3.1
10	SS10	26.1	7.45	3.800	3220	2.2
11	SS11	26.6	6.84	2.870	2220	1.7
12	SS12	26.1	7.4	3.870	3520	1.9
13	SS13	26.3	7.10	2.800	2450	1.6
14	SS14	27.2	6.32	2.890	2320	1.4
15	SS15	26.3	7.60	1.269	1240	1.2
16	SS16	26.5	7.10	1.510	1490	1.5
17	SS17	26.2	7.32	3.225	3350	2.1

Table 3. Summary of descriptive analysis of groundwater quality.

Parameters	Minimum	Maximum	Mean	Standard Deviation	Kurtosis	Skewness
Temp	26.100	27.600	26.665	0.497	-0.904	0.722
pH	6.320	7.660	7.074	0.376	-0.609	-0.121
EC	1.100	4.210	2.827	1.120	-1.269	-0.513
TDS	870.000	3750.000	2458.529	1028.822	-1.322	-0.479
F-	0.700	3.300	1.859	0.731	-0.304	0.365

solved solid (TDS) and Electrical Conductivity (EC) were measured at the sampling locations using digital TDS and digital electric meters. Fluoride was examined using the SPADNS (sodium 2- (para sulfophenylazo) -1,8-dihydroxy-3,6 naphthalene disulfonate) method of the American Physical Health Association (APHA 2012) [22].

The SPADNS method involves the reaction of fluoride with the dye lake, which results in the dissociation of a percentage to transform the dye into a colorless complex anion (ZrF62-). When there is a greater concentration of fluoride, the color that is created becomes lighter and lighter. A calibration standard containing fluoride at concentrations ranging from 0 to 1.4 mg/L was made by diluting a suitable volume of standard F- solution. The spectrophotometer was calibrated by using fluoride standards with varying concentrations, and the wavelength was established at 570 nanometers. The apparatus was prepared for the measurement of fluoride contamination when the graph showed a straight line for the first time. Both the detection & analytical limit for F- were set at 0.02 mg/L. The analytical range was from 0.0 to 2.0 mg/L. The SPADNS colorimetric method, while widely utilized for fluoride detection in water, is susceptible to numerous sources of analytical uncertainty. Instrumental limitations, such as the calibration precision of the spectrophotometer, wavelength accuracy (often at 570 nm), and baseline stability, may lead to fluctuations in absorbance measurements. Furthermore, the chemical characteristics such as the stability and purity of the SPADNS and zirconyl reagents may influence color development and, subsequently, the precision of the results. Matrix effects, such as interfering ions (e.g., phosphate, sulfate, or iron), sample turbidity, and intrinsic coloration, can alter absorbance measurements and reduce the selectivity of the approach. Procedural inconsistencies, including the timing of reagent addition, mixing, and temperature variations, may negatively impact reproducibility.

Furthermore, the method assumes linearity within a certain range of fluoride concentrations, and data at the detection or quantification limits demonstrate increased uncertainty. To mitigate these limitations, calibration was conducted with freshly created standards, blank corrections were applied, and samples were analyzed in duplicate. Quality control techniques, including traditional recovery evaluations, were established to ensure the correctness and precision of the analytical results.

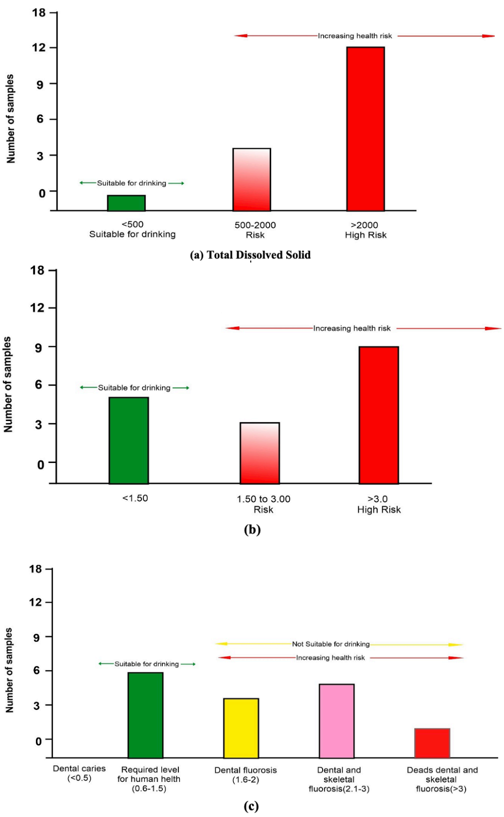

Fig. 3. (a) TDS and (b) Electrical conductivity (c) Fluoride, variations for risk evaluation for drinking purpose.

3. Results and Discussion

The point data fluoride variation map shows that the high fluoride ranges are broadly disseminated in groundwater throughout the research area (Fig. 3d). It is quite possible that this process is to blame for the increased fluoride levels seen in the groundwater in the research area, including SS9 and SS8, which suffer from quality issues such as high fluoride levels (3.1 and 3.3 mg/l) as illustrated in Tables 2-3. Communities need to be warned to stay away from these sources since exposure to fluoride from them could causes dental fluorosis. To avoid occurrences of fluorosis, fluoride can minimize the incidence of decay in the teeth when it is present in low amounts. High and long-term fluoride intake can cause skeletal fluorosis, a disorder that alters the bone, which brings on joint discomfort and is harmful to health (BIS, 2012) [23] as movement is restricted and some bone fractures may be more likely. Dental fluorosis is one of the conditions that can be caused by too much fluoride. The general public needs to be educated about the use of surface water, particularly river water supply systems, whenever it is feasible. As a long-term solution to the issue, the implementation of community-based defluorination systems needs to be given serious consideration. It is suggested that the brackish water, which is characterized by a salty flavor, should be combined with fresh water in order to enable previously unused water to be put to use for reasons related to potable consumption. In agricultural activities, it is important to avoid making higher use of phosphatic and potassium fertilizers. It is of the utmost importance that areas where ground water has a high fluoride content not be utilized for drinking purposes. Alternatively, an appropriate defluorination technique is recommended, such as the Nalgonda approach which combines blending with chemical treatment and low FÉ water ion exchange in order to maintain the fluoride range of water samples within acceptable limits.

The descriptive statistics analysis of the respective values for all physicochemical characteristics of samples of groundwater in the research region is shown in Table 4. The temperature of the collected water samples ranged from 26.1 to 27.6 °C. pH defines the state of equilibrium between the concentrations of ions of hydrogen and hydroxyl ions in aqueous solutions. The pH of all of the samples varied from 6.3 to 7.6 which indicate that pH was slightly acidic to alkaline in nature due to various anthropogenic process such as discharge of sewage and uses of fertilizers in study area and various natural activities such as the weathering process and brackish water intrusion. The all of the water samples pH lie 6.5 to 8.5 drinking water limit as per set by Bureau of Indian Standards (BIS), 2012. The investigation of this study reveals that the TDS and EC concentrations in 76.47% and 70.58% of water sampling sites surpassed BIS (2012) standards of drinking water (Figs. 3(a)-3(b), respectively. TDS represents the total concentration of inorganic and organic substances dissolved in the water. These may consist of minerals, salts, metals, ions, and other substances. Elevated TDS alter taste, smell, and general characteristics of water. Excessive levels suggest pollution or high salinity, which can have negative effects on both human health and the environment. The health effects of TDS in water is based on the particular composition of dissolved substances, their concentrations, and an individual's susceptibility. EC indicates the capacity of water to conduct an electrical current, mainly because of dissolved ions. Water becomes more conductive as it dissolves minerals and other solutes while percolating through the ground. Higher EC in groundwater is associated with elevated TDS levels, as both parameters reflect the number of dissolved ions present. Monitoring the TDS and EC in groundwater is crucial for evaluating water quality and determining its appropriateness for different uses such as drinking, agriculture, and industrial applications. High TDS and EC may suggest increased salinity, pollution from human activities or natural sources, and other potential concerns that could impact water quality and usability. A graphical analysis for risk assessment of TDS and EC for suitability for drinking purposes in study area is illustrated in Figs. 3(a)-3(b). Based on this illustration, none of the water samples are in the safe category and appropriate for drinking consumption, 29.41% and 17.64% groundwater samples are in high risk and 70.58% and 52.94% of groundwater samples under extremely high-risk zones, respectively, and are not safe for drinking. However, a spatial variation ground water samples map, shown in Figs. 4(a)-4(d), indicates various water quality parameters such as pH, TDS, EC and fluoride concentration in the study area. Fourteen out of the total collected samples of groundwater were found to have levels of impurities that were beyond the permitted limits as per guidelines of BIS, 2012 and WHO, 2017 [24] as shown in Tables 5 and 6. SS1 to SS17 represents sampling sites. Figs. 4(a)–4(e) illustrates the groundwater quality characteristics for different water sampling locations. The fluoride concentrations observed in this study range from 0.7 to 3.3 mg/L, comparable to or above amounts recorded in numerous other regions in India. The results of the current study are significantly superior to those recorded in

Table 4. Groundwater Samples surpassing the acceptable limit of fluoride range in research area.

Water Characteristic	BIS Standards, 2012		WHO Specifications, 2017		Number of samples surpassing acceptable limit	Total no. of samples in percentage (%) surpassing limit
	Acceptable limit (mg/L)	Max Allowable Limit (mg/L)	Acceptable limits (mg/L)	Max. Allowable limit (mg/l)		
Fluoride	1.0	1.5	1.0	1.5	14	82.35

Table 5. Guidelines for various chemical indicators of groundwater quality according to BIS standards, 2012 and WHO, 2017.

S. No.	Chemical Indicators	Unit	Acceptable level	Allowable level	% of water samples above permissible limit
1	pH	-	6.5 - 8.5	–	100
2	F-	mg/L	1.00	1.50	64.70
3	TDS	mg/L	500	2000	70.58
4	EC	ms/cm	1.5	–	76.47

Table 6. Summary of Correlation analysis.

	Temperature	pH	Electrical Conductivity	TDS	Fluoride
Temperature	1				
pH	-0.334	1			
Electrical Conductivity	-0.295	0.1252	1		
TDS	-0.349	0.2463	0.9688	1	
Fluoride	-0.141	0.2876	0.8915	0.896	1

Tamil Nadu (0.51–4.0 mg/L) [3] and Mettur Taluk, Salem Tamil Nadu (0.1 to 4.0 mg/L) [13], yet inferior to the maximum concentrations noted in Mehsana, Gujarat (up to 40 mg/L) [4] and Bassi, Jaipur (up to 11.5 mg/L [14]. Compared to studies in fluoride-endemic areas such as Nayagarh, Orissa (0.16–10.10 mg/L) [8] and Malpura, Rajasthan (0.08–11.3 mg/L), [12], the current findings suggest that while fluoride levels in our study area may not be among the highest nationally, they surpass the WHO permissible limit (1.5 mg/L) and constitute a significant public health concern. The regional heterogeneity in fluoride content throughout Indian regions, as shown in Table 1, highlights the impact of geogenic factors including fluoride-bearing minerals, hydrogeochemical conditions, and local hydrogeology. Nonetheless, human activities, such as agricultural runoff and excessive groundwater extraction, may also play a role, particularly in semi-arid regions.

A spatial variation ground water samples map (Figs. 4(a)-4(d)) indicates various water quality parameters such as pH, TDS, EC and Fluoride concentration in study area.

3.1 Correlation analysis

Table 3 illustrates the correlations among the recorded quality of water characteristics. The Pearson correlation coefficient, which quantifies the relationship that is linear between both variables, is among the most frequently utilized correlation coefficients. Each of the cells of the table illustrates the correlation between both of

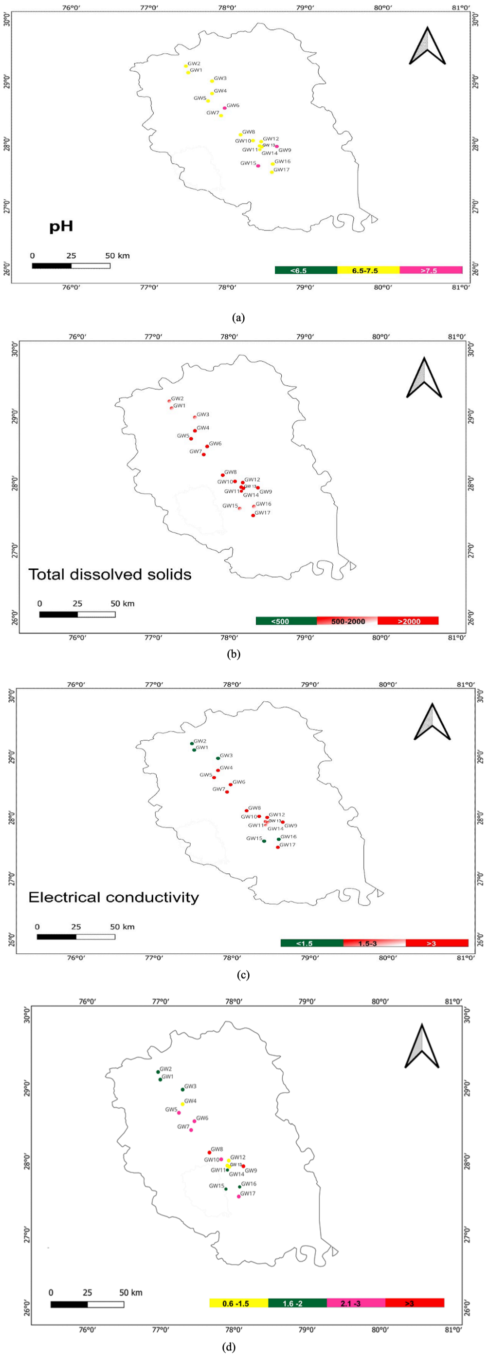

Fig. 4. Water sample spatial variation distribution maps over study area (a) pH, (b) TDS, (c) Electrical conductivity and (d) Fluoride.

the variables. Data were summarized utilizing correlation matrices, which serve as inputs for more advanced investigations and as tools for diagnosis for such evaluations. Correlation coefficients assess the pattern and strength of relationships among water quality parameters. There is a strong correlation between Total Dissolved Solids (TDS) and Electrical Conductivity (EC). The correlation between fluoride and pH, electrical conductivity, and total dissolved solids is significantly high. TDS is primarily affected by EC, while fluoride is significantly influenced by the concentrations of pH, EC, and TDS. A significant negative correlation was identified between pH and temperature.

3.2 Limitations, recommendation and future aspects

The investigation concentrated exclusively on a single season, while deviations from other seasons were excluded due to data limitations. The study relied on a limited quantity of groundwater samples, perhaps constraining the generalizability of the findings to larger spatial contexts. Therefore, additional research will be necessary to estimate the emerging contaminants that weren't investigated in this study. The study depended on a restricted dataset, and future expansions could include long-term monitoring for more substantial results. The study does not account for seasonal fluctuations in fluoride concentrations that may arise due to changes in rainfall, groundwater recharge, and anthropogenic activities. The study did not include measurements of well depth. A follow-up phase is planned to incorporate temporal and seasonal variations and well depth measurement for a more comprehensive analysis. A quantitative health risk assessment, such as Hazard Quotient

(HQ), using established exposure models (e.g., US EPA) to support claims about non-carcinogenic risk for children and adults were not applied. The Nalgonda technique, artificial recharge, and rainwater harvesting are recommended for diluting fluoridated water. The Nalgonda technology has been effectively implemented in various Indian cities for fluoride removal, demonstrating a reduction of fluoride levels to below 1.0 mg/L. It can eliminate up to 90% of fluoride from water. It is economical and appropriate for rural households and community-scale applications in India. Artificial recharge techniques, including percolation tanks, check dams, and recharge wells, facilitate the dilution of elevated fluoride levels by enhancing the entry of precipitation and surface water into aquifers. In Rajasthan, artificial recharge has resulted in a 30–60% decrease in fluoride levels over a 3–5-year timeframe (25) (CGWB Report, 2020). Regions characterized by semi-arid conditions and a diminishing water table, such as western Uttar Pradesh, including Mathura, can significantly benefit from the establishment of organized recharge zones to augment groundwater volume and diminish fluoride concentration. Rainfall harvesting is a sustainable method that collects and stores rainfall for immediate use or for aquifer replenishment. It can function both as a fluoride-free drinking water supply and as a method to dilute fluoride in the aquifer during recharging. Investigation conducted in Gujarat and Andhra Pradesh indicates that rainwater collecting systems resulted in a decrease in groundwater fluoride levels by as much as 1.5 mg/L within three years of deployment. With an average annual precipitation of approximately 650 mm, rooftop and community collection systems can substantially diminish dependence on fluoride-contaminated groundwater. Alternative and emerging community-based methods, such as activated alumina filters, exhibit excellent removal efficiency but necessitate regular regeneration and are susceptible to pH fluctuations. Charcoal adsorption is effective but constrained by cultural acceptability in vegetarian populations. The current study advances understanding or policy relevance:

- Addressing a geographic data deficiency regarding fluoride contamination in the Mathura region, which has been insufficiently represented in national groundwater quality assessments.
- Offering fluoride levels to establish a baseline for subsequent monitoring.
- Offering evidence-based recommendations for mitigation, such as the installation of community defluoridation units and rainwater harvesting, aligned with India's National Rural Drinking Water Programme (NRDWP).
- This study supports more informed water resource management and policy interventions in fluoride-affected regions.

4. Conclusion

The research findings of this investigation have shown that 64.70 percent of the study area was either highly contaminated (fluoride impacted) or severely polluted, with concentrations ranging from 1.6 to 3.3 mg/L (Table 2) and the majority of the groundwater samples did not fulfil the quality criterion for drinking water (IS: 10500 BIS 2012). According to the findings of the study, the interaction between rock and water is the primary source of fluoride in groundwater. This interaction is heavily impacted by the lithology of the surrounding area, and anthropogenic activities also play a crucial role in the prevalence of fluoride in ground water. It is recommended that only after adequate testing should pipe water be delivered to households for use in drinking and cooking, and other activities, such as washing and bathing, will no longer be permitted to take place at existing hand pumps and tube wells that have high fluoride sources. This study will provide useful information and appropriate management strategies to policymakers for safe supply of groundwater. The implementation of public awareness campaigns and rainwater conservation strategies are essential components in the achievement of water quality supervision for sustainable development. The Nalgonda technique, artificial recharge, and rainwater harvesting can be recommended for diluting fluoridated water.

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