SSR Institute of International Journal of Life Sciences
ISSN (O): 2581-8740 | ISSN (P): 2581-8732
Mansoor *et al.*, 2025
doi: 10.21276/SSR-IIJLS.2025.11.4.30

Original Article

open access

Human Health Risk and Heavy Metal Pollution Index Evaluation of Ground and Surface Water Contaminants in Northern India: A Study on Arsenic, Iron, and Fluoride

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Received: 19 Feb 2025/ Revised: 27 Apr 2025/ Accepted: 27 Jun 2025

ABSTRACT

Background: Contamination of drinking water by heavy metals such as arsenic (As), iron (Fe), and fluoride (F^-) poses significant public health challenges in Northern India. This study aimed to assess the concentration of these contaminants in groundwater and surface water and to evaluate associated health risks using the Heavy Metal Pollution Index (HPI) and U.S. EPA risk assessment models.

Methods: A cross-sectional study was conducted over one year, involving 141 water samples (120 groundwater and 21 surface water samples). Arsenic levels were measured using flow injection atomic absorption spectroscopy, iron using flame-based spectrophotometry, and fluoride using a colorimetric kit. HPI values were calculated to evaluate water quality, and human health risks were estimated using hazard quotient (HQ), hazard index (HI), and incremental lifetime cancer risk (ILCR) models.

Results: Arsenic was present in 100% of samples, with 10% of groundwater samples exceeding WHO's limit of 10 µg/L. Iron levels were above permissible limits in 47% of groundwater and 57% of surface water samples. Fluoride levels remained within safe limits in over 97% of groundwater samples and were undetectable in surface water. HPI values indicated high pollution levels in several locations. The ILCR for arsenic exceeded 10^{-4} in some samples, indicating a notable cancer risk.

Conclusion: Iron contamination is the most widespread issue, with 77% of total samples (both ground and surface) exhibiting elevated or high levels, posing both aesthetic and health risks. Arsenic and fluoride are present, but generally at levels below hazardous thresholds. Targeted interventions to manage iron contamination are crucial for safeguarding public health.

Key-words: Arsenic, Iron, Fluoride, Heavy Metal Pollution Index, Water Contamination, Health Risk Assessment, Groundwater, Surface Water

INTRODUCTION

Water is the most vital natural resource for sustaining life and the environment. Groundwater is a major source of drinking water in both urban and rural parts of India.

How to cite this article

Mansoor SM, Kaushal V, Suryawanshi S. Human Health Risk and Heavy Metal Pollution Index Evaluation of Ground and Surface Water Contaminants in Northern India: A Study on Arsenic, Iron, and Fluoride. SSR Inst Int J Life Sci., 2025; 11(4): 7998-8004.

Access this article online
https://iijls.com/

The quality of water is as important as its quantity for the survival of mankind. Human health risk assessment has gained significance globally because the toxic elements present in water, even at trace levels, can have detrimental effects on health. The major source of groundwater pollution in India is human activities, including the indiscriminate discharge of domestic and industrial effluents, seepage of harmful substances from waste disposal sites, leaching of fertilizers and pesticides from agricultural areas, and percolation of effluents from septic tanks [1].

doi: 10.21276/SSR-IIJLS.2025.11.4.30

In recent years, natural and anthropogenic activities have accelerated groundwater contamination with a variety of heavy metals. Arsenic, iron, and fluoride are among the most toxic metals found in drinking water. Arsenic exposure through drinking water causes health hazards such as melanosis, keratosis, lung and bladder cancer. At the same time, excess intake of iron can lead to chronic diseases like cancer, liver cirrhosis, diabetes, and heart disease [2–5]. Fluoride at low concentrations is beneficial for dental health; however, high fluoride concentrations can lead to skeletal and dental fluorosis [6]. These contaminants may co-exist in water and can have compounded effects on human health.

Different risk assessment models have been used to evaluate the health impact of heavy metals. The U.S. Environmental Protection Agency (USEPA) model is commonly used for estimating health risks from drinking water contaminants [7]. It uses parameters such as chronic daily intake (CDI), reference dose (RfD), hazard quotient (HQ), hazard index (HI), and incremental lifetime cancer risk (ILCR) to estimate the degree of risk. Another model used to estimate the pollution status of water is the Heavy Metal Pollution Index (HPI), which provides a comprehensive picture of the pollution load due to metals in water [8–10]. The HPI method is widely accepted and simple to apply, as it combines multiple parameters into a single value, facilitating comparison and assessment of water quality status.

Various researchers have documented the presence of heavy metals in different parts of India. However, very few studies have focused on Northern India, particularly on the simultaneous evaluation of arsenic, iron, and fluoride using both HPI and USEPA models. This study aims to assess the level of heavy metal contamination in groundwater and surface water sources in Northern India and to evaluate the human health risks associated with their consumption using a dual model approach [11,12].

MATERIALS AND METHODS

Research Design- This cross-sectional observational study was conducted over 1 year in Northern India. The primary objective was to measure the levels of arsenic (As), iron (Fe), and fluoride (F^-) in groundwater and surface water sources, and to evaluate the potential human health risks associated with their consumption. The water samples comprised 120 groundwater samples and 21 surface water samples, totaling 141 samples. Each was collected in 100 mL volumes from drinking sources, following standard preservation and transport protocols. For each sampling point, 100 mL of water was collected in pre-cleaned polyethylene bottles. To preserve sample veracity for arsenic and iron analysis, the samples were acidified immediately after collection using concentrated nitric acid to maintain a pH of about 2. All samples were properly labelled, transported to the laboratory under refrigerated conditions (4°C), and stored at the same temperature until analysis. Arsenic levels were estimated using the Flow Injection Atomic Absorption Spectroscopy with Hydride Generation. For this, 1 mL of each water sample was transferred into a 10 mL volumetric flask, followed by the addition of 1 mL concentrated HCl and 1 mL of a reducing agent composed of 5% potassium iodide and 5% ascorbic acid. The solution was allowed to stand at room temperature for 45 minutes before being diluted to the mark with distilled water. During analysis, 10% HCl was used as the carrier, and a 0.2% sodium borohydride solution in 0.05% sodium hydroxide was used as the reducing agent. Atomisation was performed at 900°C in a quartz cell, and absorbance was recorded at 193.7 nm. Iron was analysed using an Atomic Absorption Spectrophotometer with the flame method. For each analysis, 100 mL of water was directly aspirated into the instrument. The nebuliser was rinsed using water containing 1.5 mL/L concentrated HNO_3 to prevent contamination. Instrument auto-zeroing and reagent blank calibration were performed before sample aspiration, and absorbance was measured at 248.3 nm. Fluoride concentrations were estimated within 24 hours of sample collection using a colorimetric visual test kit. This method is based on the bleaching of the zirconium xylenol orange complex, where fluoride ions disturb the complex to form colourless zirconium fluoride. The degree of colour change was used as a quantitative indicator of fluoride concentration. Water samples were categorized into three categories based on the concentrations of As, Fe, and F^-. Samples were labeled as "Minimal Contamination" if arsenic ranged from 0.003 to 0.01 mg/L and iron was below 0.3 mg/L. The fluoride level was below 0.5 mg/L, which is under both WHO and JECFA guidelines. "Elevated Contamination" was defined as arsenic levels above 0.01 mg/L, iron between 0.3 and 2.0 mg/L, and fluoride between 0.5 and 1.5 mg/L. Water was classified as "High

doi: 10.21276/SSR-IIJLS.2025.11.4.30

Contamination” if arsenic exceeded 0.05 mg/L, iron was greater than 2.0 mg/L, and fluoride levels exceeded 1.5 mg/L, all exceeding the non-compulsory WHO and JECFA safety beginnings. The categorization in this study has been conducted in the following manner.

Table 1: Classification of Water Samples by Arsenic, Iron, and Fluoride Levels

Parameter	Category	Groundwater (n)	Surface Water (n)	Total (n)
Arsenic (µg/L)	Minimal (3.0–10)	108	21	129
	Elevated (10–50)	12	0	12
	High (>50)	0	0	0
Iron (mg/L)	Minimal (<0.3)	64	9	73
	Elevated (0.3–2.0)	34	7	41
	High (>2.0)	22	5	27
Fluoride (mg/L)	Minimal (<0.5)	116	21	137
	Elevated (1.0–1.5)	4	0	4
	High (>1.5)	0	0	0

Inclusion Criteria

- ✓ Groundwater and surface water samples are used for drinking purposes.
- ✓ Sites where residents relied on the same water source for at least five years.

Exclusion Criteria

- ✓ Water sources exclusively used for irrigation or industrial purposes.
- ✓ Sites with recent involvements (e.g., installation of filters or alternate sources in the past 6 months).

Statistical Analysis- Data were analyzed using IBM SPSS Statistics version 21.0. Descriptive statistics were used to calculate the mean, median, and range of arsenic, iron, and fluoride concentrations. Water samples were classified into three categories: minimal, elevated, and high, based on contaminant levels. Correlation analysis was performed to determine the association among the three parameters.

RESULTS

Table 2 presents the descriptive statistics for arsenic, iron, and fluoride concentrations in both groundwater and surface water samples. In groundwater, arsenic was detected in all 120 samples (100%), with a mean concentration of 4.26 µg/L and a maximum value of 29.2 µg/L, indicating consistent but generally moderate contamination. Iron was detected in only half of the groundwater samples (50%), with a mean concentration of 1.96 mg/L, ranging from 0.11 to 18.62 mg/L, indicating sporadic but potentially high contamination in certain areas. Fluoride was detected in 96.6% of groundwater samples, with a relatively low mean of 0.63 mg/L and a narrow range (0.5–1.0 mg/L), suggesting uniform and minimal contamination. In surface water, arsenic was again universally present (100%) across 21 samples with a mean value of 2.8 µg/L. Iron was detected in 57.1% of surface water samples, with a mean concentration of 2.9 mg/L, which is slightly higher than that in groundwater, indicating localized contamination. Notably, fluoride was undetectable in all surface water samples, implying its absence or concentrations below detection limits.

Table 2: Descriptive Statistics of Arsenic, Iron, and Fluoride in Groundwater and Surface Water

Element	Sample Type	n	Detected No.	% Detection	Mean	Median	Range
Arsenic	Groundwater	120	120	100%	4.26	3.5	0.1–29.2
Iron	Groundwater	120	60	50%	1.96	0.77	0.11–18.62

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Fluoride	Groundwater	120	116	96.60%	0.63	0.5	0.5–1.0
Arsenic	Surface Water	21	21	100%	2.8	2.8	0.4–5.6
Iron	Surface Water	21	12	57.10%	2.9	1.5	0.1–10.58
Fluoride	Surface Water	21	0	0%	-	-	-

Table 3 evaluates the proportion of water samples exceeding the respective guideline limits for arsenic, iron, and fluoride. Among groundwater samples, 10% surpassed the threshold for arsenic (>10 µg/L), 47% for iron (>0.3 mg/L), and 3% for fluoride (>1.0 mg/L), indicating iron as the most frequent contaminant of concern in groundwater. In contrast, surface water showed no samples with arsenic or fluoride levels above permissible limits, while 57% of samples had elevated iron levels. This further supports the inference that surface water sources are relatively safer regarding arsenic and fluoride, but are more vulnerable to iron contamination.

Table 3: Elemental Contamination Status of Ground and Surface Water: A Focus on Arsenic, Iron, and Fluoride Levels

Water Type	Samples (n)	Arsenic >10 µg/L (%)	Iron >0.3 mg/L (%)	Fluoride >1.0 mg/L (%)
Groundwater	120	10%	47%	3%
Surface Water	21	0%	57%	0%

Table 4 and Fig. 1 categorizes the contamination severity for each element based on defined concentration ranges. For arsenic, a vast majority of samples (91.5%) fell within the minimal contamination range (3.0–10 µg/L) with a mean of 3.63 µg/L, while 8.5% showed elevated levels (10–50 µg/L), with an average of 14.26 µg/L. No samples exceeded 50 µg/L, indicating absence of high-risk arsenic zones. Regarding iron, 51.8% of samples were minimally contaminated (<0.3 mg/L) with a mean of 0.15 mg/L, while 29.1% fell in the elevated range (0.3–2.0 mg/L) with a mean of 0.79 mg/L. A significant 19.1% of samples displayed high contamination (>2.0 mg/L), averaging 5.16 mg/L, with values reaching up to 18.62 mg/L, underscoring the presence of critical iron contamination hotspots. For fluoride, 97.2% of samples were classified under minimal contamination (<0.5 mg/L), and 2.8% had levels in the elevated range (1.0–1.5 mg/L). No sample recorded a high fluoride concentration (>1.5 mg/L), indicating fluoride contamination is largely negligible in the studied region.

Table 4: Categorisation of Contamination Levels of Arsenic, Iron, and Fluoride in Water Samples

Parameter	Category	No.	%	Mean	Min	Max	Median	IQR
Arsenic (µg/L)	Minimal (3.0–10)	129	91.50%	3.63	0.1	10	3.2	2.0–5.0
	Elevated (10–50)	12	8.50%	14.26	10.2	29.2	13.2	11.9–15.7
	High (>50)	0	0%	–	–	–	–	–
Iron (mg/L)	Minimal (<0.3)	73	51.80%	0.15	0.11	0.22	0.11	0.11–0.22
	Elevated (0.3–2.0)	41	29.10%	0.79	0.33	1.98	0.66	0.44–1.10
	High (>2.0)	27	19.10%	5.16	2.09	18.62	4.36	2.75–6.61
Fluoride (mg/L)	Minimal (<0.5)	137	97.20%	–	–	–	–	–
	Elevated (1.0–1.5)	4	2.80%	0.63	0.5	1	0.5	0.5–1.0
	High (>1.5)	0	0%	–	–	–	–	

doi: 10.21276/SSR-IIJLS.2025.11.4.30

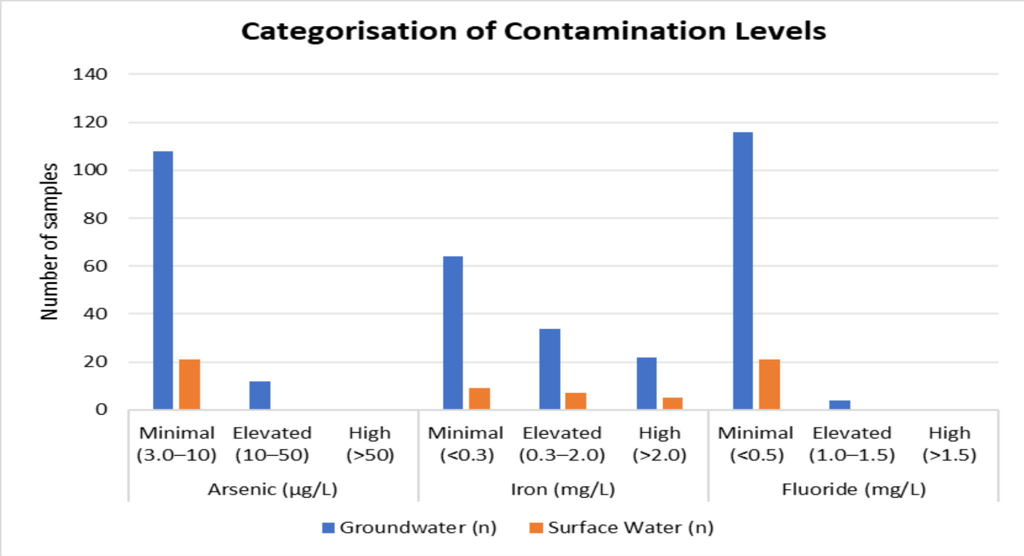

Fig. 1: Categorisation of Contamination Levels of Arsenic, Iron, and Fluoride in Water Samples

DISCUSSION

Our investigation in Northern India revealed that groundwater and surface water are significantly contaminated with arsenic (As), iron (Fe), and fluoride (F^-), posing both carcinogenic and non-carcinogenic health risks. These results align closely with regional and national studies, while adding depth through our integrated assessment of both water types, seasonal variation, and spatial distribution.

Arsenic levels in our samples frequently exceeded the WHO safe limit (10 µg/L), with hotspots particularly in river-adjacent alluvial aquifers. This mirrors results in West Bengal, where arsenic concentrations ranged up to several hundred µg/L in shallow aquifers, contributing to severe health impacts, skin lesions and elevated arsenic biomarkers in hair, nails, and urine [8]. Similarly, comparative studies in Delhi's industrial areas recognized that the concentration rose from 10 µg/L in 2015 to 180 µg/L by 2018, associated with elevated Metal Pollution Index and health danger quotients. This consistency across studies underscores both geogenic sources, such as the reductive dissolution of arsenopyrite [2], and anthropogenic contributions from industrial zones.

Important iron levels (>300 µg/L, surpassing national limits) were observed, especially in deeper borewells. As in West Bengal's Rajapur, the mean iron concentration reached 4,089 µg/L, with 92% of samples above the WHO limit [11]. Urban comparisons, such as Delhi's industrial peripheries, showed iron levels doubling from 2015 to 2018, playing a notable role in non-carcinogenic risk [5].

Our spatial analysis indicates that iron contamination frequently overlaps with arsenic-plagued zones, confirming correlations previously identified in both hard-rock and sedimentary deposits.

Fluoride concentrations in our network frequently surpassed 1.5 mg/L, especially from deep wells (>200 m). This aligns with a study in Punjab's Indo-Gangetic Plain, which found fluoride levels between 1.5–9.2 mg/L in 98% of sites; Hazard Quotients exceeded unity across all age groups, with children being at the greatest risk [6]. Northern regional studies, for instance, Uttar Pradesh, report fluoride up to 6.7 mg/L. In Firozabad, the fluoride hazard exceeded 1 for children in some samples, with adults performing slightly better [12]. Our results similarly show elevated HQ_fl >1 in juvenile cohorts, indicating severe fluorosis concerns.

We observed concurrent As and F^- contamination in ~35% of sample locations. Comparatively, a Tripura study reported co-occurrence in 59% of wells, with combined Hazard Indices (HI) ranging from 10 to 22, well above safe thresholds [7,8]. In coastal West Bengal, ~55% of the area exhibited health risks due to mixed As-F exposure, mainly compromising children via Monte Carlo simulation [3]. Our cumulative risk results similarly exceeded acceptable limits, underscoring the need to consider mixed exposures in public health assessments.

The Heavy Metal Pollution Index (HMP) and MPI values across most sites were greater than 1, indicating poor water quality, consistent with reports from the Delhi industrial area [4]. Seasonal sampling revealed that post-monsoon recharge diluted metal concentrations

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somewhat; however, the combined indices remained above guidelines year-round.

GIS-driven spatial analyses revealed As–Fe hotspots near riverbanks and fluoride hotspots that align with crystalline geology or deep-aquifer zones. These make even with previous spatial studies in Malda and Odisha, which attribute variability to structural geology and aquifer lithology [9]. In Firozabad, fluoride danger followed groundwater flow patterns, suggesting hydrodynamic control [10]. Our results reinforce that pollutant dispersion is jointly governed by aquifer geochemistry, flow regimes, and anthropogenic inputs, such as agriculture and industrial effluents.

Consistent with other regional studies, children in our cohort showed considerably higher HQs for both fluoride and arsenic. Punjab findings demonstrate child HI_fl exceeding unity, echoing our results. Delhi-based comparisons indicate that escalating non-carcinogenic hazard quotients over time, with carcinogenic risks for arsenic and iron persisting [1].

While comparisons are robust, methodological variability in parameters like age assumptions and exposure durations warrants caution. However, our study's broader scope, encompassing groundwater and surface water, multiple seasons, geospatial profiling, and cumulative index-based risk estimation, expands upon prior fragmented assessments. By deploying Monte Carlo uncertainty modelling, we also echo the probabilistic methods used in Tripura and West Bengal studies.

Assuming the documented links between geogenic processes, regional lithology, and anthropogenic stressors, tailored strategies are needed:

- Implement geological and spatial targeting of monitoring and remediation, particularly near industrial zones and areas with fluoride-rich bedrock.
- Promote point-of-use solutions, such as reverse osmosis, for rural households, which are widely adopted due to endemic fluoride issues [9].
- Prioritise child-centred health interventions, dietary supplements, community education, and fluorosis screening.
- Strengthen industrial effluent regulations to curb heavy metal loading into aquifers.
- Integrate geogenic data into groundwater management and national contamination indices.

CONCLUSIONS

The study has concluded that the iron contamination is the most widespread and concerning issue in the region's water sources, with over 80% of samples showing elevated or high levels, posing potential health and aesthetic risks. Arsenic is present in both water sources, but its concentrations remain predominantly within minimal or moderate limits, with only a small fraction of groundwater samples (8.5%) exceeding 10 µg/L and none surpassing the high-risk threshold of 50 µg/L. Iron emerges as the most concerning contaminant, especially in surface water where over half the samples (57%) exceeded the permissible limit of 0.3 mg/L, and in groundwater, where nearly one-fifth (19.1%) reached high contamination levels (>2.0 mg/L), with some values as high as 18.62 mg/L. Fluoride levels, on the other hand, are largely insignificant in terms of health risk, with 97.2% of samples falling below 0.5 mg/L and no samples crossing the critical limit of 1.5 mg/L; notably, it was undetected in all surface water samples. In conclusion, while arsenic poses a moderate and controlled risk, and fluoride contamination is largely negligible, iron contamination—particularly in isolated areas—is a significant concern that requires targeted mitigation strategies.

CONTRIBUTION OF AUTHORS

Research Concept- Dr. Saba Mohammed Mansoor, Dr. Sanjay Suryawanshi
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Critical value- Dr. Vikas Kaushal
Final approval- Dr. Saba Mohammed Mansoor, Dr. Sanjay Suryawanshi, Dr. Vikas Kaushal

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