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Analysis of heavy metal contamination in groundwater and associated probabilistic human health risk assessment using Monte Carlo simulation: A case study in Gaya, Bihar

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ABSTRACT

The occurrence of heavy metal contamination in groundwater poses significant health risks through ingestion and dermal exposure, with potential links to cancer and other diseases. This study evaluated groundwater samples for 10 heavy metals (Al, As, Cr, Cu, Cd, Fe, Mn, Ni, Pb, and Zn) using ICP-OES. While cadmium and chromium levels were within acceptable limits prescribed by the Bureau of Indian Standards, aluminum and iron exceeded these limits in 56 and 58% of samples, respectively. Other metals surpassed limits in 2–20% of cases. Health risk analysis revealed non-carcinogenic risks for 28% of adults and 44% of children, alongside carcinogenic risks from arsenic (36% of samples) and nickel (46% of samples), especially affecting children. Sensitivity analysis highlighted heavy metal concentration as the key variable influencing risk, and principal component analysis suggested geogenic sources, like rock weathering, as major contributors to contamination. Despite these risks, the heavy metal pollution index remained within acceptable limits for all samples. The study emphasizes the necessity for continuous monitoring and targeted mitigation strategies to address heavy metal contamination and protect public health.

Key words: groundwater, health risk assessment, heavy metal contamination, pollution indices, principal component analysis, sensitivity analysis

HIGHLIGHTS

- This type of study has not been done before in this study area.
- The health risk for children in the study area was higher than that for adults.
- Based on the probabilistic risk assessment, the most relevant parameters identified were the concentrations of heavy metals and the daily ingestion rate.
- The findings of this study will offer valuable insights into the health risks associated with heavy metals in groundwater.

INTRODUCTION

The availability of potable water is essential for safeguarding public health and ensuring the well-being of all living organisms. Due to its easy access, groundwater is considered a readily available source of fresh water, thus making it essential for several aspects of human life, including the economy, industry, and food production. Regrettably, the rapid expansion of urban areas and industrial sectors, along with the increased discharge of hazardous materials, has led to a decline in groundwater quality (Kumar & Singh 2024a). Approximately one-third of the global population depends on groundwater (Xing *et al.* 2013). Eighty-eight per cent of the rural Indian population rely upon groundwater (Ravikumar *et al.* 2011).

The contamination of groundwater poses a significant risk to both human health and the environment. Declining groundwater quality is caused by both biological and chemical contaminants (Egbueri & Mgbenu 2020). Excessive quantities of heavy metals in drinking water may lead to physiological complications in human. Arsenic (As), cadmium (Cd), chromium (Cr), nickel (Ni), and lead (Pb) are highly toxic to humans even at low quantities and cause both immediate and long-term health issues (Elwakeel *et al.* 2018; Kumar *et al.* 2021). These include trachoma, diarrhea, nausea, vomiting, stomach pain, renal failure, cognitive impairment, muscular weakness, thyroid abnormalities, and cancer (Chowdhury *et al.* 2016; Mehmood *et al.* 2019). Many metals such as copper, manganese, iron, and zinc are essential nutrients for proper functioning of the human body (Jomova *et al.* 2022; Ayejoto & Egbueri 2024). Heavy metals are appearing in groundwater through various natural and anthropogenic processes (Wagh *et al.* 2018; Barzegar *et al.* 2019; Egbueri & Mgbenu 2020). The natural processes include rock and soil weathering, organic matter decomposition, atmospheric deposition etc., whereas anthropogenic activities are mining, overutilization of fertilizers and pesticides, mineral processing industries, disposal of domestic and industrial waste etc. (Belkhiri *et al.* 2017; Singh & Vaishya 2017; Ukah *et al.* 2019).

Several investigations have previously shown that arsenic, along with other heavy metals, is the primary pollutant found in the groundwater aquifer of the Gangetic Plain in India (Acharyya 2005; Nayak *et al.* 2008; Ravindra & Mor 2019; Yadav *et al.* 2020). In 2002, a small village, Semaria Ojha Patti in the Bhojpur district of Bihar, reported the first instance of arsenic pollution in groundwater. The maximum concentration was observed at 1,654 ppb. Subsequently, it has spread to 16 districts, affecting a total of 10 million individuals within the state (Kumar *et al.* 2016; Kumari *et al.* 2018; Suman *et al.* 2020; Kumari & Maurya 2023). A study conducted by Jha *et al.* (2023) in the Samastipur district, Bihar, found a maximum arsenic concentration of 91 ppb in groundwater, while Maity *et al.* (2020) reported a higher heavy metal (arsenic 168 ppb) concentration in the groundwater of Bhojpur district, Bihar. A study conducted by Ranjan *et al.* (2012) in Gaya district identified the presence of heavy metals, namely arsenic, zinc, copper, and iron, in groundwater without mentioning concentration. Gaya is one of the rapidly growing cities of Bihar, having around 4.4 million population and thus withdrawing a large quantity of groundwater for agricultural, industrial, and domestic uses. All the previously discussed causes may collectively impact groundwater quality and introduce heavy metals as contaminants. Furthermore, Gaya is a famous historical and religious site in Hinduism and Buddhism. The residents of Gaya depend entirely on groundwater for their daily needs. In view of the above, the study of heavy metals in groundwater is necessary to protect human health. Evaluating drinking water quality parameters will definitely help to identify potential health risks and thus ensure safe consumption. This may also be essential to prevent long-term health hazards and safeguard the community's well-being.

The groundwater in the study region (Bodhgaya, India) has not been analyzed for heavy metal contamination and its associated human health risks. Since heavy metals adversely impact on human health, a health risk assessment (HRA) using Monte Carlo simulation (MCS) may be evaluated. The MCS approach is often used to assess probabilistic risk and evaluate the variability and uncertainty of the data (Jafarzadeh *et al.* 2022). The probabilistic risk assessment (PRA) approach utilizes the MCS technique to evaluate the health risk considering all input parameters (namely heavy metal concentration in drinking water, exposure duration (ED), exposure frequency, ingestion rate, and body weight) and determines all probable effects (Jafarzadeh *et al.* 2022). Hence, it is important to know the concentration of heavy metals and identify their possible sources of contamination to effectively manage and mitigate health hazards. The HRA may also estimate long-term health impacts.

Based on the literature survey, we identified a significant research gap in understanding groundwater heavy metal contamination and its health impacts in the Bodhgaya region. In order to fill the identified research gap, this study aims to investigate the concentrations of 10 heavy metals in groundwater and evaluate the associated potential non-carcinogenic and carcinogenic health risks. To achieve this, we will employ deterministic and probabilistic HRA approaches, specifically utilizing the MCS to quantify the variability and uncertainty associated with the risk estimates. Furthermore, the study also analyses the contamination indices of heavy metals to provide a comprehensive understanding of groundwater quality.

MATERIALS AND METHODS

Study area

The Gaya district is situated at a distance of 115 km from Patna, the capital of Bihar. The region has a total area of 4,986 square kilometres. According to the 2011 census, the total population of the Gaya district is 4,391,418. Out of this, 581,601 (86.76%) of the population live in urban areas, and the remaining 3,809,817 (13.24%) live in rural areas (CGWB 2022). The drainage pattern in the study area is controlled by four major rivers/streams: the Morhar, the Phalgu, the Paimar, and the Dhadhar. These rivers originate from the southern plateau of Jharkhand and flow in northerly and north-easterly directions. The whole study area is impacted by a continental monsoon characterized by a challenging environment condition. Summer months are marked by scorching westerly breezes, reaching temperatures as high as 46 °C, and sometimes dropping as low as 4 °C in winter. The geological composition of the Gaya district can be classified into two primary categories: (i) Quaternary sediments, encompassing both younger and older alluvium and (ii) pre-cambrian rock formations comprising the Chhotanagpur granite gneiss complex and meta-sedimentary layers (CGWB 2022).

Sample collection and laboratory analysis

A total of 50 groundwater samples were collected using a systematic grab sampling approach from a selected area of Gaya district, Bihar (Figure 1). Samples were collected from hand pumps and bore wells. The depth of the hand pumps and bore wells varied between 12 and 35 m. The samples were collected in clean and dry polypropylene bottles and stored in a cold and dry container until they were analyzed in the laboratory (APHA 2017). Prior to sample collection, hand pumps and borewells were purged for a duration of 5–10 min to eliminate any stagnant water and ensure the collection of representative samples. Prior to filling the bottles with the samples, each bottle underwent a rinsing process consisting of 2–3 repetitions using the same groundwater samples. A set of water samples, each having a volume of 125 mL, were filtered using a 0.4 μ size Millipore membrane filter. The filtered samples were then treated with HNO_3 to lower the pH to 2. The pH was measured with the help of a portable pH meter (PCS Testr 35). The presence of heavy metals was analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent 5110 VDV). The operating conditions were maintained in accordance with the manufacturer's specifications to ensure the most accurate determination. To ensure the accuracy of the analytical data, the same samples were tested three times. All the reagents used were of analytical grade. Blanks and standard solutions were prepared and regularly analyzed after every tenth sample to ensure the precision of the results. The Arc GIS software version 10.5 was used to create a map employing the Inverse Distance Weighted (IDW) interpolation method. This method expressly assumes that objects in close proximity to each other are more similar than those that are farther away.

Statistical analysis

To understand the characteristics of the hydro-chemical parameters in the study area, descriptive statistical analysis, including the correlation coefficient and Principal Component Analysis (PCA), was applied. Correlation and PCA were conducted using IBM SPSS Statistics V26. The correlation coefficient was calculated to examine the strength of relationships between

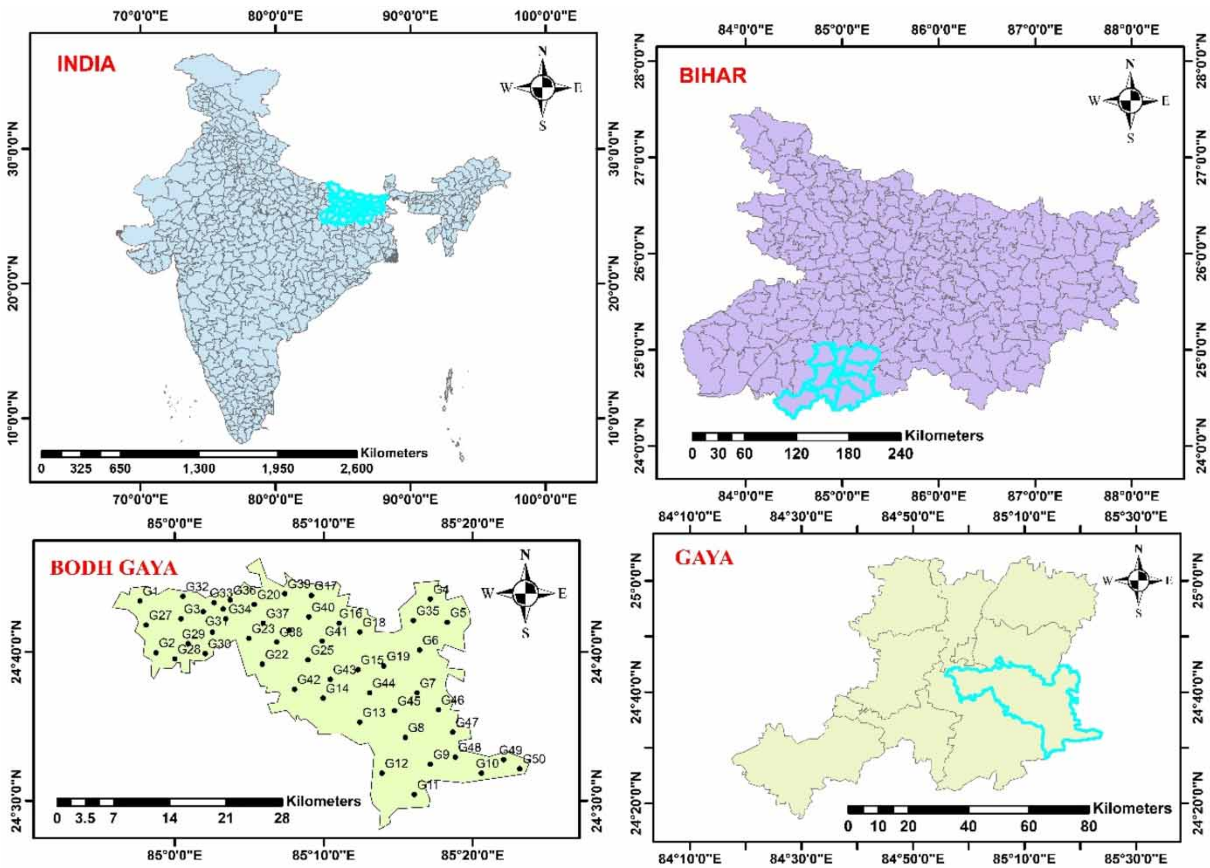

Figure 1 | Map showing the study area with sampling locations.

various groundwater quality parameters. PCA is a powerful statistical technique that was used to reduce the complexity of a large dataset by transforming it into a smaller, more interpretable dataset while preserving most of the original variability. By applying PCA, researchers can uncover and identify the most significant factors driving the patterns and trends within a complex dataset.

The study employed PCA to examine the primary chemical characteristics that influence groundwater quality and to identify their origins. The most influential chemical constituents of groundwater were identified, providing valuable insights into the factors controlling water quality and their sources. The datasets were evaluated for their suitability for PCA using two statistical tests. First, the Kaiser–Meyer–Olkin (KMO) test was applied, with a threshold value of 0.5 or higher indicating adequate data quality. Additionally, Bartlett's test of sphericity was performed, with a significance level set at 0.05 to confirm the validity of the results (Mukherjee & Singh 2022; Kumar & Singh 2024b).

Non-carcinogenic and carcinogenic HRA

The method as described in Equations (1)–(4) is employed for assessing health risks (USEPA 1989). It involves calculating CDI (chronic daily intake) mg/kg. day, using input parameters such as C_w (concentration of heavy metal) mg/L, IR (rate of ingestion) L/day (2.5 for adults and 0.78 for children) (Dehghani *et al.* 2019), EF (exposure frequency) 365 days per year, ED years (70 for adults and 6 for children), BW (body weight) kg (70 for adults and 15 for children), and AT (average time of exposure) days (365 multiplied by ED) (Barzegar *et al.* 2019; Ayejoto & Egbueri 2024):

$$\frac{\mathrm{CDI} = \mathrm{C_w} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

HQ (Hazard Quotient), HI (Hazard Index), and CR (Carcinogenic Risk) are calculated using Equations (2)–(4), respectively. Rfd stands for the reference dosage of a given element, measured in mg/kg/d. The RfD (Reference Dose) values for various heavy metals are as follows: 7 for Aluminum (Al), 0.0003 for Arsenic (As), 0.0005 for Cadmium (Cd), 0.003 for Chromium (Cr), 0.005 for Copper (Cu), 0.7 for Iron (Fe), 0.02 for Nickel (Ni), 0.0036 for Lead (Pb), and 0.3 for Zinc (Zn) (Abdelhalim *et al.* 2023):

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{2}$$

The HI is calculated by adding the HQ values of the many heavy metals present in drinking water in order to measure the overall non-carcinogenic hazard:

$$\mathrm{HI} = \sum_{i=1}^{n} \mathrm{HQ} \tag{3}$$

$$(= \mathrm{HQ_{Al}} + \mathrm{HQ_{As}} + \mathrm{HQ_{Cd}} + \mathrm{HQ_{Cr}} + \mathrm{HQ_{Cu}} + \mathrm{HQ_{Fe}} + \mathrm{HQ_{Mn}} + \mathrm{HQ_{Ni}} + \mathrm{HQ_{Pb}} + \mathrm{HQ_{Zn}})$$

CR refers to the potential of a substance or exposure to cause cancer in humans. The CR is calculated as the product of CDI multiplied by the slope factor (SF). According to other studies (USEPA 2011; Egbueri & Mgbenu 2020), 1×10^{-4} to 1×10^{-6} is considered the permissible limit for CR:

$$\mathrm{CR} = \mathrm{CDI} \times \mathrm{SF} \tag{4}$$

MCS and sensitivity analysis

MCS is a widely used method for PRA modeling, enabling the evaluation of variability and uncertainty in multiple parameters of the human HRA model (Huang *et al.* 2010). This approach allowed us to quantify the variability and uncertainty associated with model inputs, such as contaminant concentrations, exposure durations, and individual susceptibility, as well as the model's predictions, and to identify the most critical parameters influencing the risk assessment outcomes (Bhat & Kumar 2008; Parsons *et al.* 2012).

Sensitivity analysis (SA) was performed using the MCS technique, with 10,000 iterations, in Oracle Crystal Ball (version 11.1.34190) (Soleimani *et al.* 2022). This approach involves selecting parameter values from their fitted distributions, based on input data, and subsequently calculating both point estimates and distributions of exposure and risk (Abbasnia *et al.* 2019). The parameters employed in the MSC are presented in Supplementary material, Table S1. The data in this table, along with the distribution functions utilized, are sourced from various literature works (Smith 1994; USEPA 2011; Fallahzadeh *et al.* 2019). The SA aims to identify the model inputs that have the greatest impact on health outcomes and to quantify the effect of uncertainty in model inputs on health outcomes, such as the impact of varying contaminant concentrations or exposure durations (Pianosi *et al.* 2016).

Pollution index estimation of heavy metals

Pollution indices assist in identifying and mapping pollution levels, allowing for an understanding of both the current and potential future impacts on human health. The metal index (MI) was computed based on the ratio of the measured value to the acceptable value of the metal ions (Tamasi & Cini 2004). The heavy metal pollution index (HPI) evaluates the combined effect of each heavy metal on water quality and is developed through two main steps. First, a rating scale is established for each selected parameter, assigning weight to each. Second, specific pollution parameters are chosen as the basis for the index. The rating system assigns an arbitrary value ranging from zero to one, with the selection of these values reflecting the relative importance of each quality consideration. Alternatively, the values can be determined by making them inversely proportional to the recommended standards for the corresponding parameters (Prasanna *et al.* 2012; Sirajudeen *et al.* 2014).

Metal index

The MI, proposed by Tamasi & Cini (2004) was employed to evaluate the overall quality of groundwater and the potential additive effects of 10 heavy metals on human health, (Abdullah 2013; Rezaei *et al.* 2019). It can be calculated using the following formula:

$$\mathrm{MI} = \sum_{i=1}^{n} \frac{C_i}{S_i} \tag{5}$$

where C_i and S_i represent the obtained concentration (mg/L) and the standard concentration (mg/L) of heavy metals, accordingly.

Heavy Metal Pollution Index

The HPI is a mathematical model that uses the weighted arithmetic mean approach to assess the groundwater quality (Mohan *et al.* 1996). The equations for the determination of HPI are presented in Equations (6)–(8):

$$\mathrm{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \tag{6}$$

where W_i is the weight of heavy metals (mg/L), Q_i is the sub-index of the element, and n is the number of heavy metals being analyzed.

$$W_i = \frac{K}{S_i} \tag{7}$$

where K is a proportional constant ($K = 1$) and S_i is the standard value (acceptable limit) for each heavy metals as per BIS 10500 (2012).

$$Q_i = \sum_{i=1}^{n} \frac{|(M_i - I_i)|}{(S_i - I_i)} \times 100 \tag{8}$$

where M_i is the measured value in the water sample, I_i is the ideal value for the heavy metal and S_i is the standard value for the heavy metal set by BIS. An HPI value exceeding 100 indicates that water contamination with heavy metals is at a high level and poses a significant threat to human health. If the value is <100, the level of water pollution from heavy metal is deemed minimal (Kazemi *et al.* 2023).

RESULTS AND DISCUSSION

Evaluating the occurrence of heavy metals in groundwater and their uptake via several exposure pathways may heighten the potential dangers to human well-being. Table 1 presents statistical measurements (maximum, minimum, standard deviation, and third quartile) of pH and heavy metal concentrations, along with their acceptable limits as described in BIS 10500:2012. The analysis shows that the abundance of the heavy metals, according to the 3rd quartile, is in the following order: Fe > Zn > Mn > Al > Cu > As > Cr > Ni > Pb > Cd. The occurrence of heavy metals in groundwater may be from anthropogenic sources like industrial and domestic effluent, mining, and agriculture activities, and can also have natural origins like higher concentrations of CO_3^{2-} and SO_4^{2-} minerals and other types of minerals (Liu *et al.* 2019; Kozisek 2020).

The investigation revealed that the aluminum content ranged from 12.01 to 445.26 μg/L, with 56% of the samples exceeding the acceptable limit. Arsenic was found at levels ranging from not detectable (ND) to 15.48 μg/L, with 10% of the samples exceeding the acceptable limit. The concentration of cadmium (Cd) ranges from ND to 1.21 μg/L, and no samples exceeded the acceptable limit. The primary sources of cadmium (Cd) in the environment include the combustion of fossil fuels, the melting and incineration of municipal waste, the disposal of Ni–Cd batteries, and other industrial operations (ATSDR 2017). The concentration of chromium ranges from 0.94 to 46.54 μg/L. Iron (Fe) is essential as a micronutrient for a wide range of species and can be found in igneous rocks, ferromanganate soils, and other combinations that result in iron oxides, magnetite, sulfides, and iron clay minerals, and others (Ranjan *et al.* 2012). The concentration of iron (Fe) varies from 56 to 12,350 μg/L, with 58% of the samples exceeding the acceptable limit. Manganese (Mn) is a crucial micronutrient for humans. Manganese pollution in groundwater may arise from both natural geological processes and human activities. The manganese content ranges from 1.42 to 1,430 μg/L, with 20% of the samples exceeding the acceptable level. Nickel (Ni) is a known carcinogenic element and is responsible for inducing serious lung and renal diseases (Multhaup 2005). Common sources of nickel include industrial operations, power plants, and the glass, ceramic, battery and color-producing industries, as well as urban sewage and sludge (Govil *et al.* 2008). The concentration of Ni in groundwater ranges from ND to 33.96 μg/L, with 4% of the samples exceeding the acceptable limit in the research region. Lead (Pb) is primarily classified as an anthropogenic contaminant due to the small amount emitted by natural sources in contrast to the significant contributions from human activities (Buragohain *et al.* 2010). The sources of lead (Pb) pollution are synthetic pesticides, fossil fuels, improper disposal of batteries, emissions from paint industries, and use of lead in gasoline as anti-knocking agent

Table 1 | Statistical summary of heavy metal concentration compared with BIS (10500 2012)

				BIS Standard (10500 2012)			
Elements[a]	**Range**	**Mean ± std. deviation**	**Third quartile**	**Acceptable limit**	**Permissible limit**	**WHO (2017)**	**% of the sample exceeding the acceptable limit (BIS10500)**
Al	12.01–445.26	49.92 ± 66.71	46.375	30	200	–	56
As	0–15.48	2.98 ± 4.38	6.5425	10	50	10	10
Cd	0–1.21	0.09 ± 0.26	0	3	NR	3	0
Cr	0.94–46.54	3.94 ± 6.51	3.6525	50	NR	50	0
Cu	0–151.59	13.65 ± 30.42	6.895	50	1,500	2,000	8
Fe	56 – 12,350	1,254.8 ± 1,946.32	1,560	300	NR	300	58
Mn	1.42 – 1,430.69	88.64 ± 211.65	60.0625	100	300	100	20
Ni	0–33.96	2.94 ± 6.42	2.5225	20	NR	70	4
Pb	0–22.99	1.71 ± 5.32	0	10	NR	10	8
Zn	12.18–6,280.55	556.83 ± 1,183.67	302.6425	5,000	15,000	3,000	2

NR, no relaxation.
[a]Heavy metals are measured in μg/L.

(Jumbe & Natural 2009). The lead values in the sample region ranged from ND to 22.99 μg/L. Eight per cent of the samples in the studied region exceeded the acceptable level. Zinc concentrations in the samples range between 12.18 and 6,280.55 μg/L, with 2% of the samples found above the acceptable limit.

Correlation analysis

Correlation analysis is a useful tool in the study of water quality, as it helps in the identification of patterns and relationships among various parameters. For example, it can be used to determine the relationship between the concentrations of heavy metals and their likely sources. A significant positive correlation between parameters suggests a common source, whether natural or man-made. The correlation analysis of heavy metals in the study area revealed significant relationships between various elements. A strong positive correlation was observed between Cd and Fe (0.750), Cu and Pb (0.733), and Ni and Cr (0.667), suggesting possible common sources or geochemical associations. Moderate correlations were found between Zn and Pb (0.600), Cu and Zn (0.517), and Fe and Pb (0.515), indicating similar environmental behavior. Conversely, arsenic (As) showed negative correlations with Cu (−0.205), Fe (−0.164), Pb (−0.165), Zn (−0.267), Cr (−0.132), and Ni (−0.201), suggesting a distinct source or different mobility in the environment (Table 2). The weak correlation between Mn and most metals suggests that Mn may have a different origin or lower interaction with other elements. Overall, the correlation trends suggest that heavy metals such as Cu, Pb, Zn, and Fe may originate from industrial or anthropogenic activities, while elements like As may be from natural sources. According to Egbueri (2018) and Mgbenu & Egbueri (2019), chromium, lead, zinc and iron in water may be extracted from waste sources related to metallurgy, whereas zinc, nickel, and lead may be extracted from discarded automotive batteries, tires, paints, pipes, and electronic trash found in dumpsites.

Principal component analysis

A dataset of 10 heavy metals, namely Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn, from 50 groundwater samples was used to conduct PCA. The KMO value of 0.581 and Bartlett's test of sphericity, with a significance level of 0.0001, indicate that the selected dataset is suitable for statistical analysis. Furthermore, PCA was conducted using Kaiser categorization, and four principal components (PCs) with eigenvalues greater than 1 were extracted (Mukherjee & Singh 2022; Alam & Singh 2023). PCA reduced from 10 parameters to four significant components, which cumulatively contributed to 73.97% of the total variance in the dataset. The rotated component matrix for these PCs is shown in Table 3. Figure 2 displays the scree plot, illustrating the relationship between the four significant components and their corresponding eigenvalues, all of which are greater than or equal to 1. Additionally, Figure 2 includes the rotated component plot, where the four components are projected onto a rotated space, providing a visual representation of their relationships and underlying patterns.

PC 1 having an eigenvalue of 3.41, contributed 25.53% to the total variance and showed positive loading for Cu and Pb. This component is highly influenced by Cu and Pb, suggesting that these two elements tend to vary together and dominate the overall variability in the dataset. The sources of these substances can be both natural and anthropogenic. Natural sources include geogenic processes such as weathering and leaching of minerals from rocks, while anthropogenic sources refer to urban runoff, especially from sources such as vehicle emissions and improper disposal of electronic waste. PC 2 having an

Table 2 | Correlation analysis of heavy metals in the study area

	Al	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Al	1.000									
As	−0.117	1.000								
Cd	0.057	−0.044	1.000							
Cr	0.170	−0.132	0.121	1.000						
Cu	0.344	−0.205	0.380	0.020	1.000					
Fe	0.064	−0.164	0.750	0.097	0.711	1.000				
Mn	0.016	0.223	0.073	0.035	0.026	0.078	1.000			
Ni	0.387	−0.201	0.261	0.667	0.155	0.245	0.036	1.000		
Pb	0.009	−0.165	0.162	0.020	0.733	0.515	0.107	0.119	1.000	
Zn	0.149	−0.267	0.309	0.168	0.517	0.416	0.008	0.408	0.600	1.000

Table 3 | Principal component analysis with four extracted components

	PC1	PC2	PC3	PC4
Al	0.310	0.554	− 0.255	0.036
As	− 0.273	− 0.197	0.024	0.702
Cd	0.161	0.127	0.913	0.037
Cr	− 0.105	0.829	0.149	− 0.037
Cu	0.874	0.051	0.264	− 0.030
Fe	0.536	0.054	0.778	− 0.006
Mn	0.135	0.105	0.005	0.842
Ni	0.117	0.892	0.183	− 0.063
Pb	0.883	− 0.051	0.101	0.024
Zn	0.696	0.281	0.172	− 0.145
Eigenvalues	3.412	1.698	1.251	1.036
% Variance	25.533	19.417	16.687	12.332
Cumulative %	25.533	44.950	61.637	73.969

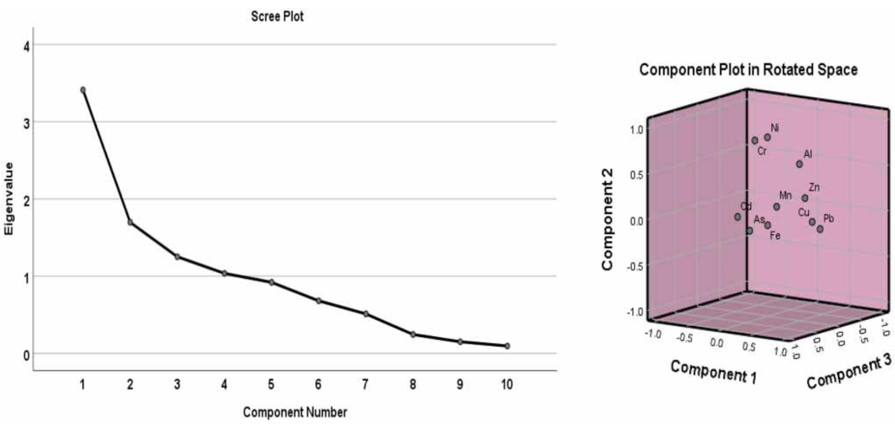

Figure 2 | Scree plot of the eigenvalues and PCA component loading in rotated space.

eigenvalue of 1.69, contributed to 19.42% of total variance and showed positive loading of Al, Cr, and Ni. The occurrence of these elements in groundwater is through geogenic processes such as the weathering of minerals like garnierite, chromite, and feldspars present in the earth's crust. PC 3, having an eigenvalue of 1.25, contributed to 16.69 of the total variances and showed positive loadings for Cd and Fe. PC 4 had an eigenvalue of 1.04 and showed positive loading for As and Mn. In the study area, the source of these elements are geogenic processes such as the weathering of rocks like pyrolusite, manganite, and mineral deposits. The results of PCA in the present study show an agreement with the result obtained by Ranjan *et al.* (2012), Abdelhalim *et al.* (2023), and Hamidu *et al.* (2021) in Gaya Bihar, Minia area Egypt and Kano City Nigeria region, respectively. The component loading pattern of the above said studies demonstrated that geogenic processes, along with anthropogenic sources, impacted the groundwater quality and contributed to the elevated heavy metal concentration in groundwater, which is also similar to the results obtained in the present study.

Health risk assessment

Understanding the composition of heavy metals in groundwater and their potential impact on health is essential for effectively overseeing, controlling, and maintaining the long-term viability of water resources. HRA involves the evaluation and quantification of the potential adverse effects on human health resulting from groundwater pollution (Zhu *et al.* 2017; Egbueri 2020). Risk assessment aims to mitigate the health hazards linked to the use of polluted drinking water using a variety of ways (USEPA 1999, 2011; Zhu *et al.* 2017). Heavy metal toxicity in humans is proportional to the amount absorbed daily; therefore, chronic daily intake (CDI) is used to assess non-carcinogenic and carcinogenic health risks (USEPA 1989). HRA can be classified into non-carcinogenic, such as CDI, HQ, and HI, and CR. This study considers only oral intake for assessing health risks for adults and children. Table 4 presents a statistical overview of the HQ and CR (Cancer Risk) values obtained in the study. These values are used to assess the potential health hazards, including both non-carcinogenic and CR.

Non-carcinogenic risk

Non-CR is evaluated by comparing the estimated exposure to a contaminant with a regulatory limit or reference dose (RfD) (Abdelhalim *et al.* 2023; Kazemi *et al.* 2023). For both adults and children, the mean HQ values followed the order: As > Cu > Zn > Fe > Cr > Mn > Pb > Cd > Ni > Al. The HRA reveals varying levels of non-carcinogenic risks posed by heavy metals in groundwater. Most heavy metals analyzed have average HQ values below 1, suggesting an acceptable risk level of non-CR. However, when HQ exceeds 1, it signals a potential adverse health impact. For arsenic (As), 12% of groundwater samples for adults and 20% for children exceeded an HQ > 1, making arsenic a primary contaminant of concern, with children being more vulnerable due to higher exposure rates relative to body weight. These findings align with previous studies in Bihar (Singh & Ghosh 2012; Singh *et al.* 2014; Chakraborti *et al.* 2016), which reported HQ values above 1 for adults and children, emphasizing the widespread risk posed by arsenic in the region. Copper (Cu) also shows elevated HQ values in 2% of the samples for adults and 4% for children. Similarly, zinc (Zn) exceeds HQ > 1 in 2% of the samples intended for children, further reinforcing their heightened susceptibility to certain metals. These findings underscore the importance of targeted mitigation strategies, particularly for children, to reduce exposure to heavy metals like arsenic, copper, and zinc. Improved water treatment and regular monitoring of groundwater quality are essential to safeguard public health. A study conducted by Jha *et al.* (2023) in the Samastipur district 35% of the population, especially children, is at non-CR (HQ > 1). Similarly, Devi & Yadav (2018) showed that the estimated HQ values for all metals were higher in children than in adults, indicating that children are more susceptible to non-carcinogenic risks.

To measure the combined impact of many heavy metals on non-CR, the hazard quotients (HQs) calculated for individual heavy metals are summed together and presented as a hazard index (HI). If the HI is <1, there is no obvious danger to the health of the exposed individual. If the value of HI is more than 1, it indicates a non-carcinogenic health risk (USEPA 1989;

Table 4 | Statistical summary of non-carcinogenic and carcinogenic health risk assessment for heavy metals

Heavy metals	Hazard quotient (HQ) range	HQ mean	Range carcinogenic health (CR) risk for adult and children	
Al (adult)	0.000–0.002	0.0002	As (adult)	$0.000–7.73 \times 10^{-4}$
Al (children)	0.000–0.003	0.0003	As (children)	$0.000–1.13 \times 10^{-3}$
As (adult)	0.000– 1.717	0.3550	Cd (adult)	$0.00–1.64 \times 10^{-5}$
As (children)	0.000–2.501	0.5169	Cd (children)	$0.00–2.39 \times 10^{-5}$
Cd (adult)	0.000–0.086	0.0058	Cr (adult)	1.68×10^{-5}–8.08×10^{-4}
Cd (children)	0.000–0.125	0.0084	Cr (children)	2.44×10^{-5}–1.18×10^{-3}
Cr (adult)	0.011–0.538	0.0456	Ni (adult)	$0.000–1.94 \times 10^{-3}$
Cr (children)	0.016–0.783	0.0664	Ni (children)	$0.000–2.83 \times 10^{-3}$
Cu (adult)	0.000–1.089	0.0868	Pb (adult)	$0.00–6.98 \times 10^{-6}$
Cu (children)	0.000–1.586	0.1264	Pb (children)	$0.00–1.02 \times 10^{-5}$
Fe (adult)	0.002–0.575	0.0614		
Fe (children)	0.003–0.837	0.0894		
Mn (adult)	0.000–0.339	0.0222		
Mn (children)	0.000–0.494	0.0323		
Ni (adult)	0.000–0.057	0.0052		
Ni (children)	0.000–0.083	0.0075		
Pb (adult)	0.000–0.228	0.0159		
Pb (children)	0.000–0.332	0.0232		
Zn (adult)	0.001–0.747	0.0668		
Zn (children)	0.002–1.088	0.0973		

Su *et al.* 2018). The HI values calculated range from 0.021 to 2.19 and 0.031 to 3.199 for adults and children, respectively. Around 28% of adults and 44% of children have non-carcinogenic risks due to groundwater consumption (Figure 3). Abdelhalim *et al.* (2023) also reported a similar observation that adults are less likely to be exposed to carcinogenic hazards compared to newborns and children. The spatial distribution map of non-CR (Figure 4) has shown the high value of HI for adults and children in the central region and western part of the study area.

Carcinogenic risk

CR is a type of risk associated with the potential for adverse health effects related to cancer. The assessment of CR involves determining the likelihood of a person acquiring cancer over their lifetime due to exposure to a harmful substance. Prolonged exposure to trace amounts of heavy metals, including As, Cd, Cr, and Ni in groundwater, may significantly increase the likelihood of cancer in humans (Abdelhalim *et al.* 2023; Ayejoto & Egbueri 2024). Therefore, these harmful metals are classified as carcinogens in the present study for both adults and children. For As, Cd, Cr, and Ni the average value of CR for adults are 1.6×10^{-4}, 1.1×10^{-6}, 6.8×10^{-5} and 1.7×10^{-4}, respectively (Table 4). According to CR results, As shows 36% of the sample exceeds the limit for adults and children, Cr shows 2% for adults and 22% of the sample for children exceed the limit and Ni shows 28% of the sample for adults and 46% of the sample for children exceed the permissible limit. As a result, As and Ni pose a high cancer risk in the study area. In this study area, Pb had the lowest cancer risk of all the heavy metals studied. The CR distribution map (Figure 4) for As in adults and children shows high-value CR in the central region, western region, and some patches in the southeast area. For Ni, the distribution map has shown some high values of CR are found in the southwest of the study area. Jha *et al.* (2023) also reported that in the Samastipur district, 100% of the population (children, females, and males) is at CR (CR $> 10^{-6}$). Children were identified as the most vulnerable group, followed by females and males, due to lower body weight and higher exposure per unit body weight.

MCS and SA

In addition to calculating point estimates of HQ using Equations (1) and (2), this study employed the MCS technique to estimate HQ variances. Specifically, 10,000 iterations of the MCS were run using Oracle Crystal Ball software (version 11.1.34190) to quantify the uncertainty associated with HQ and CR estimates. To account for the probabilistic nature of the exposure parameters, the study incorporated the appropriate distributions of contaminant concentration, ingestion rate, body weight, and exposure frequency for each exposed group. The simulation results for heavy metals such as (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) revealed that the 95th percentile of HQ values for the children and adults were 0.0005, 1.09, 0.01, 0.07, 0.19, 0.19, 0.09, 0.01, 0.04, 0.23 and 0.0002, 0.52, 0.008, 0.03, 0.09, 0.10, 0.04, 0.006, 0.02, 0.13, respectively (Supplementary material, Table S2). The simulation results revealed that the highest 95th percentile of calculated HQ values among the study areas was 1.09 for children. This indicates a higher non-CR for children exposed to the contaminant, suggesting a potential health concern for this vulnerable population.

For CR, the simulation results of heavy metals such as As, Cd, Cr, and Ni revealed that the 95th percentile of CR values for children and adults were 4.95×10^{-4}, 3.2×10^{-6}, 1.10×10^{-4}, 4.22×10^{-4} and 2.37×10^{-4}, 4.9×10^{-8}, 5.4×10^{-5}, 7.7×10^{-7}, respectively. For children, As, Cr and Ni indicate a CR, whereas for adults, only As indicates a significant CR. These results

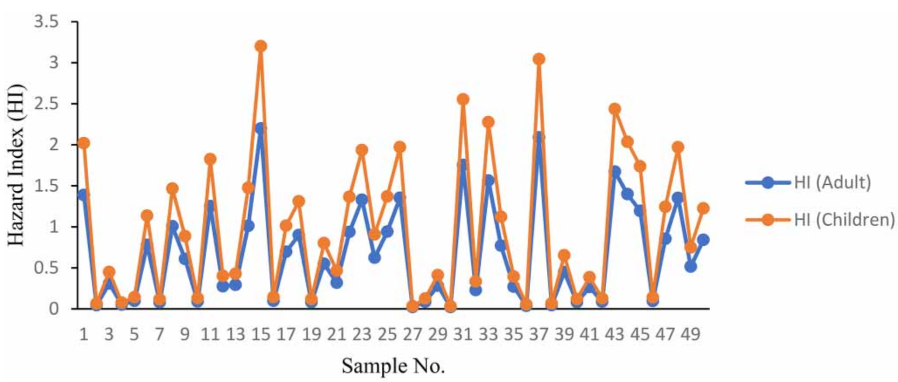

Figure 3 | Statistical graph representing heavy metal-based non-carcinogenic health risk for adults and children.

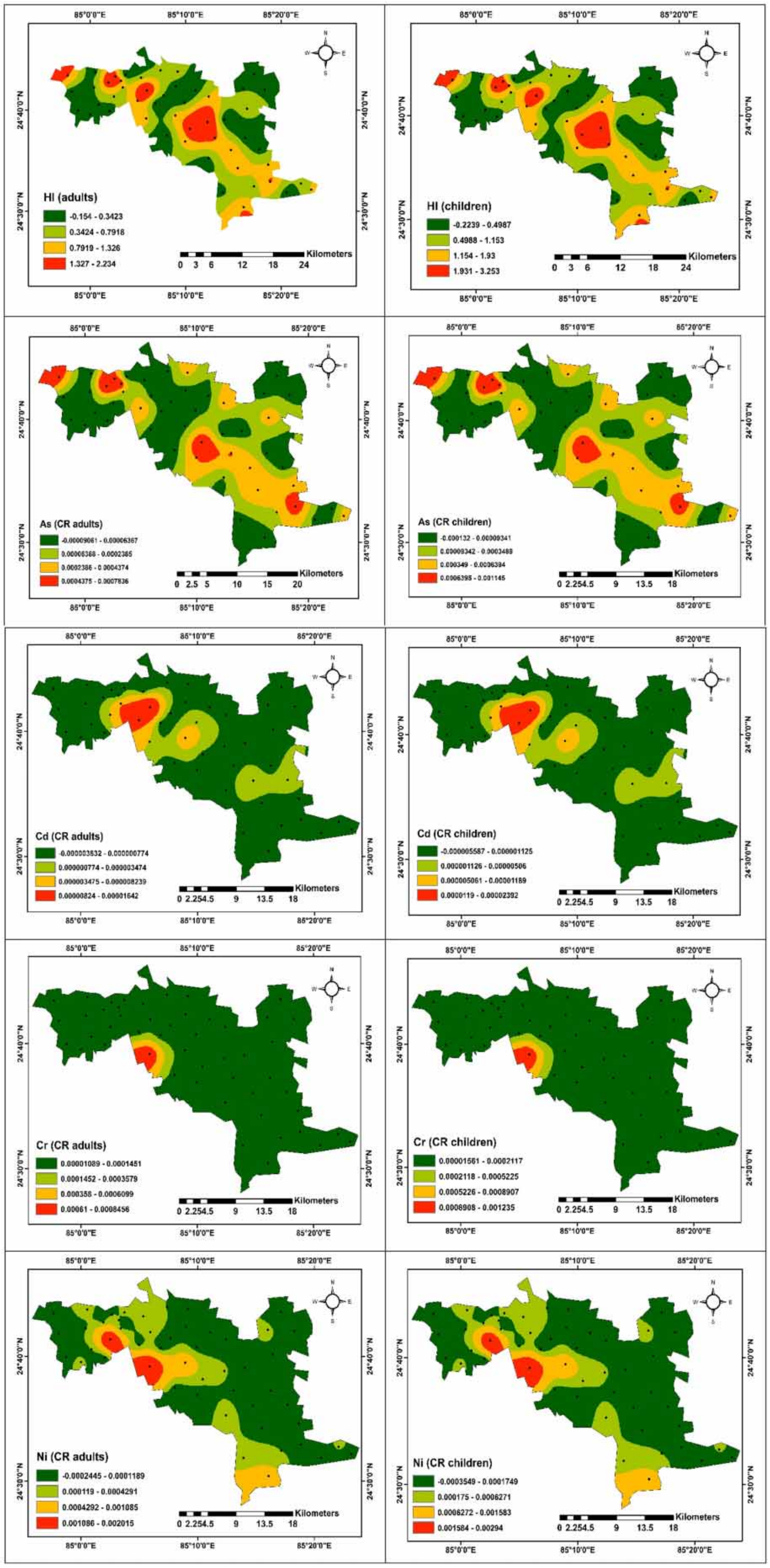

Figure 4 | Spatial distribution of non-carcinogenic (HI) and carcinogenic (CR) health risks for adults and children in the study area.

showed that most of the child population were affected. The elevated risk levels observed in children may be attributed to their relatively low body weight compared to other age groups (Fallahzadeh *et al.* 2019). A similar result has been reported in a study conducted by Kumari & Maurya (2023) in Maner, Bihar.

SA is a powerful tool used to uncover the relative importance of each input parameter in predicting the outcome of risk assessment. Applying this technique, we can identify the most influential input parameter that drives the results of the assessment, providing valuable insight into the key drivers of risk (Ma *et al.* 2000). A comprehensive SA was performed to identify the impact of various parameters on both non-carcinogenic and CR simulations. The results of the analysis are graphically represented in tornado plots (Figure 5), which illustrate the % contribution to variance on a percentage scale. These plots provide a clear and concise representation of the relationships between the input parameters and the simulated risk outcomes, allowing for the identification of the most influential factors. The analysis shows that the influencing input parameters are in the following order: C > IR > BW > EF (Figure 5). Further, the concentration of heavy metals (C) in the groundwater is the most influential variable for both groups of the population, with 95.3, 94.1, and 74, 66.2% of the contribution to the output variance for adult and children population, respectively. Inversely, BW has a negative influence for children (−1.9, −11.2%) and adults (−1.2, −7.7%). This suggests that individuals with higher body weights are associated with lower sensitivity, implying that they may be less responsive to changes in the input parameters. Similar results have also been reported by Kumari & Maurya (2023) and Giri *et al.* (2020) for Maner Bihar and Singhbhum Jharkhand, respectively, which identified the metal concentration (C), as the most significant variable affecting all population groups. The MCS model provides a range of possible outcomes that also incorporates uncertainty and variability, instead of a single estimate of the traditional deterministic method. Thus, MCS enhances risk assessment, highlights extreme scenarios, and improves decision-making and communication of health risks compared to the traditional deterministic method.

Index estimation of heavy metals

Various heavy metal indexing methods have been developed and widely applied by researchers worldwide to evaluate pollution levels and water quality concerning heavy metal concentrations in both surface and groundwater. These methods provide a standardized approach to effectively assess contamination and its potential risks (Dash *et al.* 2019; Bhardwaj *et al.* 2020; Egbueri 2020; Rahman *et al.* 2020). Mohan *et al.* (1996) demonstrated that the indexing method provides a

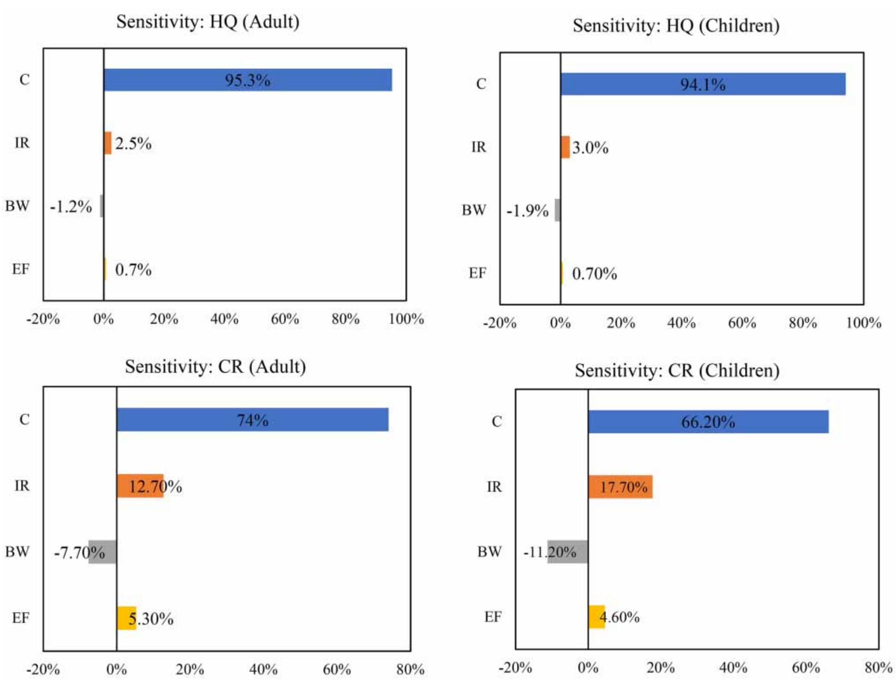

Figure 5 | SA of non-carcinogenic and CR for adults and children. C, concentration of heavy metal; IR, ingestion rate; BW, body weight; EF, exposure frequency.

comprehensive view of the combined influence of various heavy metals on overall water quality. In this study we have applied two indices (MI and HPI). These indices are valuable tools utilized by executives, environmental managers, stakeholders, and decision-makers to assess water quality. The statistical summary of the MI and HPI is presented in Table 5.

Metal Index

The MI is computed to assess the cumulative impact of various quality factors that may negatively affect the quality of drinking water. The analysis results indicated that the range and average value of MI are 0.005 to 1.793 and 0.156, respectively (Table 6). According to MI classification, 92% of samples are classified as very pure, 4% as pure, and 4% as slightly affected in the study area. The spatial distribution map (Figure 6) of the MI shows a slightly higher value of MI in the central region, along with some small patches in the northwest and southeast parts of the study area.

Table 5 | Statistical summary of water quality pollution indexes for heavy metals

	Metal index (MI)	Heavy metal pollution index (HPI)
Minimum	0.006	0.872
Maximum	1.793	75.601
Mean	0.157	10.067
Std. dev.	0.337	15.745

Table 6 | Classification of water quality based on the MI (Kazemi *et al.* 2023)

MI	Characteristics	Class	% of sample
<0.3	Very pure	I	92
0.3–1.0	Pure	II	4
1.0–2.0	Slightly affected	III	4
2.0–4.0	Moderately affected	IV	–
4.0–6.0	Strongly affected	V	–
>6.0	Seriously affected	VI	–

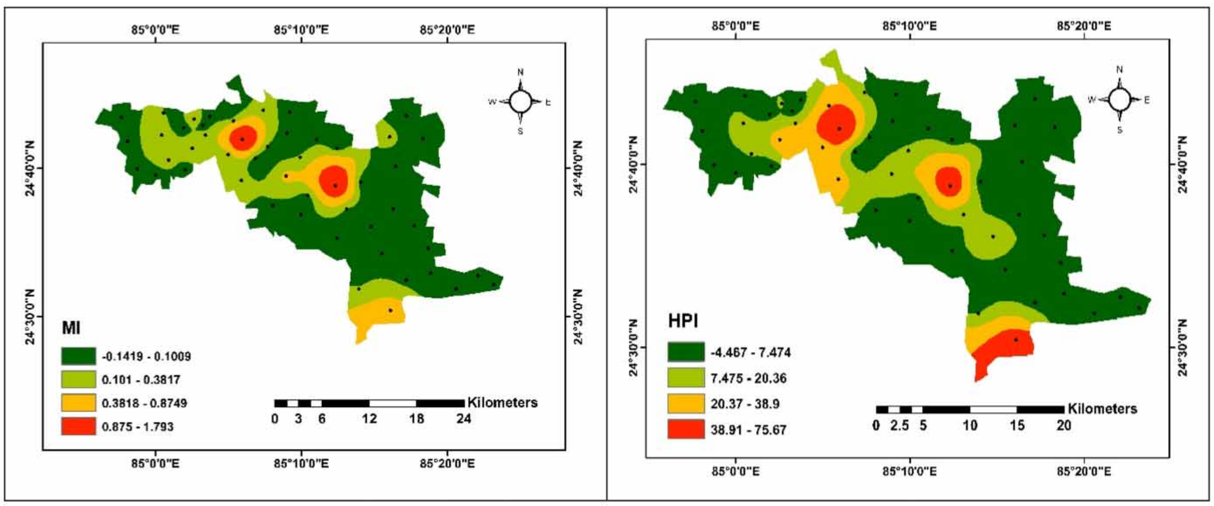

Figure 6 | Spatial distribution map of metal index (MI) and heavy metal pollution index (HPI) values in the study area.

Heavy Metal Pollution Index

The HPI is used to assess the risk associated with the presence of heavy metals in drinking water (Brown *et al.* 1970; Wagh *et al.* 2018; Egbueri 2020). The results indicated that the range of HPI values is from 0.872 to 75.601, with an average value of 10.067. The HPI value in all samples was below 100, indicating a low concentration of heavy metals in the groundwater samples of the study area (Figure 6). The MI also suggests that only 4% of the samples are slightly affected, while the remaining 96% fall into the pure or very pure category. Ghaderpoori *et al.* (2018) also has similar results where all samples had an HPI value <100, indicating water is free from heavy metal pollution.

Limitations and future scope

The limitations of the present study stem from its focus on a single-time sampling in the post-monsoon season only, which hinders the ability to generalize findings across different seasons. Furthermore, this research primarily addressed only heavy metal contamination while overlooking other potential pollutants. These factors highlight the need for further studies that cover a wider geographic area, account for seasonal variations, and examine a broader range of contaminants.

CONCLUSIONS

This study has assessed the heavy metal concentration in groundwater, potential sources of pollution, and associated human health risk due to consumption of water. Both non-carcinogenic and carcinogenic risks have been examined. The conclusions of the study are briefly below:

- The study reveals the presence of heavy metals in the groundwater of Bodhgaya, Gaya. In a significant portion of the samples, 56, 58, 20, and 10% exceeded the acceptable concentration limits for aluminum, iron, manganese, and arsenic, respectively. The elevated levels of these elements raise serious health concerns, as prolonged exposure can lead to adverse effects.
- Correlation analysis indicated a significant positive relationship between Fe–Cd, Fe–Cu, Pb–Cu, Zn–Cu, Pb–Fe, and Zn-Pb. The primary causes of groundwater contamination by heavy metals are mainly attributed to the overuse of fertilizers, the discharge of industrial waste, and the weathering and dissolution of minerals/rocks in groundwater.
- The PCA analysis reveals that geogenic factors are responsible for the deterioration of groundwater quality.
- The investigation of non-CR shows that the average HQ values are in the following order: As > Cu > Zn > Fe > Cr > Mn > Pb > Cd > Ni > Al. In groundwater samples, the HQ values for As and Cu are significantly higher for both adults and children, while the HQ value for Zn is higher only for children. The HI revealed that 28% of adults and 44% of children face non-CR due to groundwater consumption. Regarding CR, only As and Ni have mean CR values that exceed the acceptable limit for both adults and children.
- Results of the MCS showed that the children residing in the study area exhibit a higher vulnerability to non-carcinogenic health hazards resulting from exposure to heavy metals. The SA revealed that the presence of heavy metal concentrations significantly heightened sensitivity in both adults and children.
- According to the MI classification, 92% of the samples show very pure water quality, while 4% of the samples show slightly affected in the study area.

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AUTHORS CONTRIBUTION

S.K. contributed to literature review, conceptualization, sample collection, laboratory analysis, results analysis and interpretation, writing the original manuscript. N.S.M. contributed to conceptualization, methodology development, results interpretation, supervision/guidance and reviewed original manuscript

STATEMENTS & DECLARATIONS

All authors have read, understood, and have complied as applicable with the statement on 'Ethical responsibilities of Authors' as found in the Instructions for Authors.

DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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