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Research article

Evaluating arsenic contamination in northwestern Bangladesh: A GIS-Based assessment of groundwater vulnerability and human health impacts

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A R T I C L E I N F O

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A B S T R A C T

One of the biggest environmental worries in the world today is the risk of arsenic (As) contamination in groundwater. The Atomic Absorption Spectrometer (AAS) was used in this work to assess the As content in groundwater samples from 38 shallow (27 m) tubewells in northwest Bangladesh to determine the existing situation, potential source(s), and likely health risk of As and other important water quality parameters. The range of arsenic concentrations (μgL^{-1}) was troublesome and greater than the WHO recommended level for drinking water, ranging from 0.50 to 164 (mean ± SD: 20.22 ± 36.46). In groundwater, the concentrations of Fe, and Mn vary from 0.04 to 52.75 mgL^{-1} (mean ± SD: 4.23 ± 9.68), and 0.23 to 3.27 mgL^{-1} (mean ± SD: 1.10 ± 0.67). The obtained groundwater samples have pH values ranging from 5.9 to 7.1, which indicates a somewhat acidic to neutral character. Major cations have an average abundance that is as follows: $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+}$, while major anions have an average abundance that is as follows: $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$; Ca^{2+} and HCO_3^- are the main cation and anion, respectively. The groundwater in the Rajarampur village was deemed unfit for drinking or irrigation based on analyses of water quality performed using the entropy water quality index. The Ca–HCO_3 type of water, in which Ca^{2+} and HCO_3^- are the main positive ions and negative ions, is suggested by the Piper tri-linear diagram. It was discovered that silicate weathering regulates the hydrogeochemical activities in groundwater using a bi-variate examination of several hydro-chemical variables. Four major clusters were observed for the water sample. According to reductive dissolution processes and principal component analysis, the arsenic in groundwater is geogenic in origin. Arsenic is discharged from sediment to groundwater by reductive dissolution of FeOOH and MnOOH, as shown by the modest connection between As, Fe, and Mn. The United Nations Environmental Protection Agency's (USEPA) suggested value for probable cancer risk assessment was 10^{-6}, however the probable cancer risk assessment found a higher value, indicating that the population in the study region was at high risk for cancer. Remedial measures for arsenic mitigation include removing arsenic from groundwater after it is extracted, searching for alternative

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aquifers, and implementing various water-supply technologies such as dugwells, deep tubewells, pond-sand filters, and rainwater harvesting systems.

1. Introduction

Both urban and rural areas rely on groundwater for drinking purposes, but they face challenges such as increasing pressure and contamination from inorganic pollutants. In recent decades, water contaminants from dye materials, pesticides, and **other** organic pollution, have difficult for human, and living organisms health [1]. Human life has become difficult due to groundwater pollution. The two main factors that contributed to groundwater pollution and the subsequent decline in quality to unsuitable for consumption are rapid industry and urbanization [2]. Nowadays, the environmental pollution is increasing due to industrial activities [3]. Research on the presence of chemicals in groundwater is crucial because their accumulative effects on people's health throughout the food chain. To reduce health concerns, it is essential to measure the concentrations of trace materials in groundwater and assess their potential health effects. Both natural processes and human activity can introduce toxic trace metals into water supplies. Because of the weathering of rocks and the decomposition of biological organisms, natural supplies of trace elements are anticipated. Hazardous compounds may be transferred to soil and subsequently to water sources through the interaction between water and rocks. Processes may cause trace elements, which are persistent in the environment, to build up in soils and streams. However, distinct geochemical properties of elements can result in differences in the water chemistry during processes involving water and rock, for instance, oxidation, reduction, ion exchange, competitive adsorption, weathering, and dissolution. Heavy metals are mostly introduced to groundwater from anthropogenic sources such as mining, industrial waste, disposal of solid waste, and agricultural waste [4,5]. Due to both natural and man-made processes, water quality has quickly decreased around the world, especially in developing nations. Bangladesh is currently suffering significant contamination problems of heavy metals, especially arsenic poisoning in groundwater and soil, as a result of its increasing industrialization and urbanization. Worldwide, high levels of arsenic have been found in groundwater, including Bangladesh, Cambodia, Vietnam, Argentina, the United States, India, Chile, China, and Pakistan. Bangladesh, India, and China are the three countries with the highest rates of local waterborne arsenic as a poison. Arsenic can accumulate in groundwater under two specific geoenvironmental conditions: confined basins with semi-arid climates as well as highly decreasing alluvial aquifers. They both are related to geologically recent sediments in low-lying, flat locations where the flows of groundwater are slow [6,7]. As a result, areas close to sources of surface water, such as rivers and lakes, frequently contain arsenic concentrations. It is still unclear what causes the amounts of arsenic to vary and how they build up in the soil, surface water, and groundwater in different parts of the Bangladesh Plain.

The groundwater's geographical variation as concentrations at ranges of 10^1–10^4 m for 10 thousand years old Holocene fluvial-deltaic aquifers in the Bengal Basin have been widely recorded [8]. According to mounting evidence [9–12], at the minimum some of this variability is perhaps attributable to differences in the native geology and their effect on shallow sub surficial water flow. Given this regional variability and the likelihood that subsurface flow may be impacted by sizable water withdrawals for agriculture in some regions of Bangladesh, there is a legitimate fear that shallow groundwater Arsenic concentration perhaps also shift over time. Predicting changes in groundwater As concentration over time or space is especially challenging due to ongoing knowledge gaps regarding the mechanisms that trigger As mobilization. These gaps are compounded in the water levels by distinct seasonal variations in shallow as well as deep aquifers connected to the monsoon. However, this awareness is required urgently because a sizable portion of those shallow wells that currently fulfil the drinking water standard of 50 μg/L in Bangladesh are shared, at the minimum for the time being, by Bangladeshi villagers in order to lessen their disclosure to As and, consequently, lessen the likelihood that they will contract a number of life-threatening diseases.

The first as patient in Bangladesh was discovered in the Chapai-Nawabgonj area in 1993, and as time went on, the situation deteriorated. A larger concentration of as patients was discovered in Bangladesh's northwest, in the Ganges flood plain. Many Bangladeshis (97%) rely on groundwater for domestic, agricultural, and drinking needs, and tens of thousands of wells have been dug by them to get to fresh water aquifers. However, drinking groundwater that has been polluted with As due to natural processes and excessive groundwater removal as a result of population growth presents a serious risk to individuals who consume it. According to Anawar et al. [13], 27% of wells in Bangladesh are significantly polluted with immense amount (>50 μg/L) of As.

There are three factors that contribute to the increase, in concentrations of As in groundwater. To begin with, the saturation of water plays a role by limiting the diffusion of oxygen. In addition, there needs to be a supply of sulfur. Finally, there must be a carbon containing source to facilitate the dissolution of Iron oxides. Arsenic release from sediment and its quantity is based on the existence of carbon from organic materials and the amount of As is available within the sediment. The prediction is that Arsenic release is to occur when sediment contains either recalcitrant (resistant) organic carbon or, Iron oxides bearing As or, both. On the other hand sediments with active carbon from organic materials as well as labile (easily released) confined Arsenic will result in a more pronounced discharge. Field studies conducted in Bangladesh, West Bengal, Nepal, Cambodia and Vietnam provide evidence for both shallow releases as well as gradual release at greater depths [14–20]. In Bangladesh specifically groundwater pumping for irrigation occurs at a rate compared to integrated flow from hand pumps. This irrigation pumping along with return flow through fields leads to changes in recharge areas, discharge areas, recharge rates as alterations in regional and local flow patterns. For irrigation the highest use of groundwater is observed along Nepal border in the Bengal Basin and Terai Basin while less usage is seen in the Red River Basin and less so, in the Mekong River Basin.

According to several studies, groundwater As in aquifers from river deltas (such as the Red River delta, the Mekong delta, the

Ganges delta, the Yellow River delta and the Pearl River delta) is very variable at both local and regional scales. Hetao basins, Datong basins, Yinchuan basins, West Jilin basins, and Zhunger basins, among others and inland basins. We need to understand the distribution pattern and factors that lead to low As levels in groundwater aquifers in order to find the location of water wells there which might be drinkable. Groundwater pollution in aquifers due to arsenic (As), a hazardous element that has been categorized as a Group A human carcinogen, is a problem for world health. Over 105 nations have detected this poisonous contamination of arsenic in groundwater (up to 5000 μg/L), and and the prediction is that over 200 million people globally face a danger of As poisoning from drinking as much as they should. The most severe poisoning influenced by Arsenic of the century has occurred in Bangladesh, where 59 districts have been examined out of 64 districts which show a higher level of As which exceed WHO's safe limit of 10 μgL^{-1}. People in these areas depend on groundwater contaminated with As for drinking, cooking, and even though irrigation of food crops like rice or, vegetables. The number of individuals in Bangladesh who have been vulnerable to health concerns as a consequence of drinking water that contains As is estimated to be over 80 million. Chronic vulnerability to elevated levels of arsenic is connected to a higher chance of

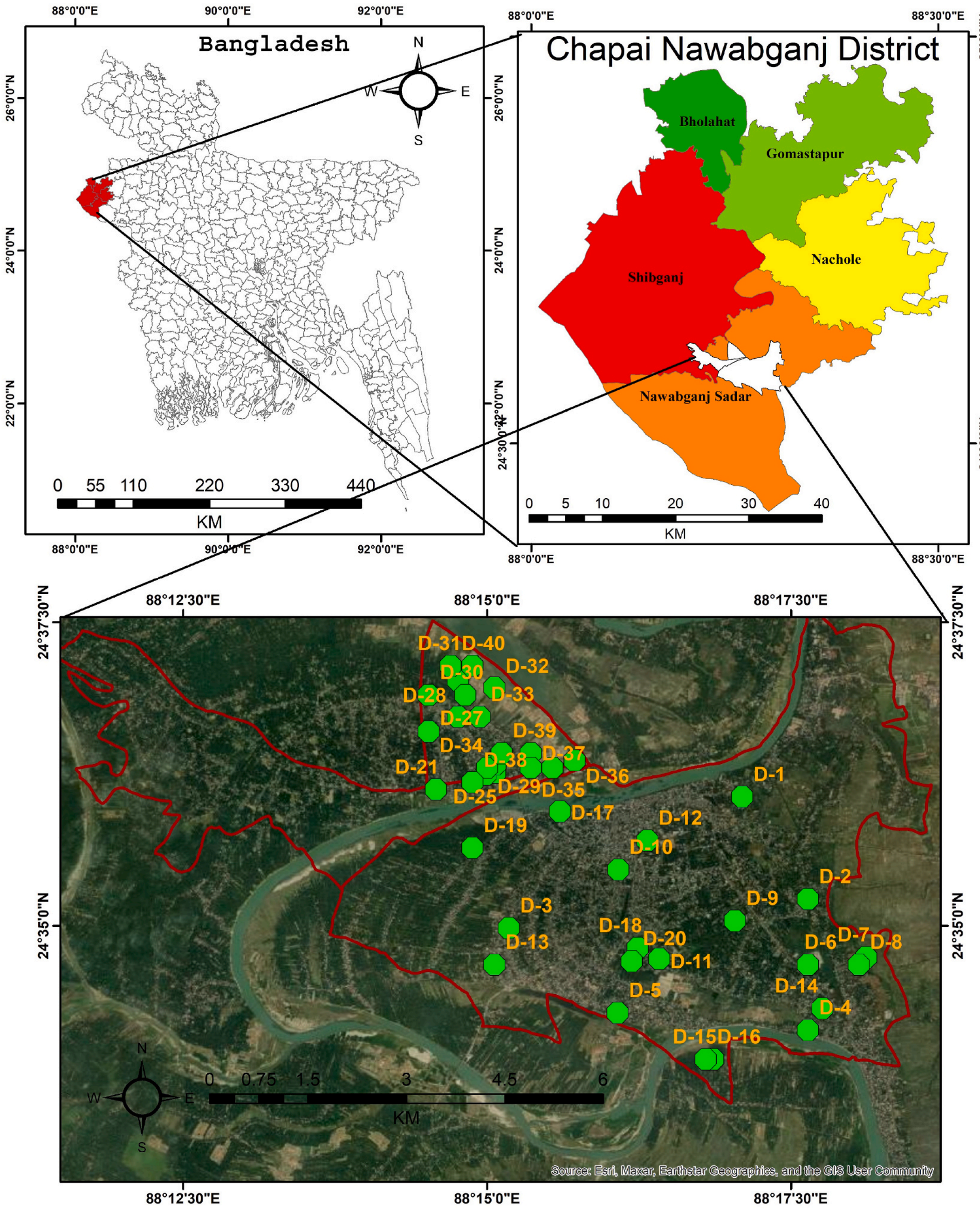


Fig. 1. Location map of study area.

developing carcinogen illness such as lung, bladder, liver, skin, and kidney cancers [21]. People who are susceptible to long-term arsenic pollution of their waters for drinking purpose may develop keratosis (dry, rough, popular skin lesions) and melanosis (change in pigmentation). According to Shakoor et al. [22], arsenic poisoning can also affect a person's neurological, reproductive, cardiovascular, hepatic, hematological, respiratory, and diabetic systems. Shallow aquifers in Bangladesh have been examined for as pollution by a number of agencies. In order to look at the state of As and its important properties, NRECA [23] inspected over 570 tube wells across the nation.

The health risk of arsenic can be measured from a pair of major vulnerability routes of As in humans which are As-contaminated water and food [24]. By evaluating the exposure to As resulting from the consumption of cultured food crops, the harm to health from food may be quantified in terms of estimated diurnal input (EDI). The direct, violent, and continuous lethal exposure to As, on the other hand, makes a health danger evaluation based on drinking As-contaminated water quite significant. As seen in our examinations, the main research's objectives are to: (1) determine the position of the As impurity in groundwater; (2) investigate the origins and likely mechanisms of As release in groundwater; (3) raise awareness of the terrifying effects of arsenic and encourage people to drink and use water from arsenic-free zones; and (4) calculate the implicit benefits to mortal health in the area of interest from drinking groundwater.

The novelty of the research work lies in several aspects: (1) The study integrates the assessment of arsenic (As) contamination in groundwater with the analysis of various other water quality parameters such as pH, EC, TDS, K^+, Na^+, Cl^-, Mg^{2+}, Ca^{2+}, SO_4^{2-}, NO_3^-, HCO_3^-, Mn, and Fe. This comprehensive approach provides a holistic understanding of the water quality and the potential health risks associated with arsenic contamination; (2) The research employs a hydro-geochemical analysis to understand the underlying processes regulating the presence of arsenic in groundwater. This includes the identification of the geogenic origin of arsenic, its release from sediment to groundwater, and the influence of factors such as silicate weathering and reductive dissolution processes. These findings contribute to a deeper understanding of the mechanisms driving arsenic contamination in the specific research region; (3) The study goes beyond the traditional water quality analysis by conducting a health risk assessment associated with the consumption of arsenic-contaminated groundwater. This assessment, which revealed a higher probable cancer risk than the suggested value by the United Nations Environmental Protection Agency (USEPA), highlights the significant health implications for the population in the study region. In summary, the novelty of the research work stems from its comprehensive and integrated approach to assessing arsenic contamination in groundwater, its focus on understanding the underlying hydro-geochemical processes, and its emphasis on conducting a health risk assessment for the affected population. The main data offered in this paper might be useful for further research into the subject. Groundwater geochemistry have been studied to interpret the spatial distribution of As in aquifer and its goods on mortal health.

The limitations of this research include: (1) The research may be limited by the availability and quality of data on groundwater arsenic contamination, human health impacts, and environmental conditions; (2) GIS-based assessments often rely on various assumptions and simplifications, which can introduce uncertainties and limitations to the findings; (3) The study may be limited in its spatial and temporal scope, potentially not capturing the full extent of groundwater vulnerability and human health impacts; (4) Integrating GIS, hydrogeology, and human health assessments can be challenging, and the research may be limited by the interdisciplinary nature of the study and (5) The research may not address the practical implications of the findings, such as policy recommendations and implementation strategies.

2. Materials and methods

2.1. Description of the study area

The research is being carried out in the Chapai Nawabganj Sadar Upazila, which encompasses the Chapai Nawabganj Municipality. It is divided into two rural areas, Rajarampur and Baragharia Unions (Fig. 1), and is bordered to the west by the Rajmahal trap, the south by the Bogra shelf, and the north by the Himalayan foredeep. It lies between latitude 24°33′54″N to 24°37′8.4″N and longitude 88°14′31.2″E to 88°18′7.2″E. The region within the Ganges-Mahananda flood plain is distinguished by its moderate slope and covers an area of roughly 50 km^2. It is elevated between 20 and 25 m above mean sea level. The primary channel in the research region is the Mahananda. The Ganges River also follows a certain course from west direction to southeast direction in the southwest portion of the study area.

The area under study is a portion of the flood plain that, at its northernmost point, reaches into Bangladesh. It is situated on the Bogra slope of the Rangpur Saddle of the Bengal Basin. The whole surface geology is composed of sedimentary layers made up mostly of alluvial sand, silt, and clay. The intracratonic structure of Gaibandha [25], located within the Nawabganj is representing a depression, developed as a result of the eastern margin's uplifting of the Barind Tract. The quaternary layers, which range in thickness from 65 to 111 m, covered the Tura sandstone, which dates back to the Eocene. The Ganges and Mahananda Rivers' meandering paths put down alluvial deposits that make up a large portion of the Nawabganj area. A fault trending north to south with a downthrown direction to the west, separates the high Barind Tract to the east from the present Mahananda River channel.

The Chapai-Nawabganj region's sediments may be split into four units. Gray silts, extremely fine-grained sand overbank floodplain deposits, and fine to medium channels sands with thicknesses of 0–45 m make up the initial unit at the bottom of the series. The Barind Tract's orange gray hard clay beds and siltstone beds are covered by fine, medium, and coarse sands which are gray in color stays in the second unit, which has a thickness of 45–80 m. The third unit is made up of silty overbank sediments and gray brown micaceous, fine to medium channel sands that are 35–45 m deep. Along the faulted intersection with the Barind Tract to the east, clay deposits can be found. Clay layers with orange gray weathering and lithification of the Barind Tract Formation, silts, and very thin sands form the subsurface of the alluvial deposits. The hills to the east are formed by the fourth unit, the Barind Tract Formation, which is composed of

Table 1
Summery Results of physicochemical analysis of the groundwater.

Sample ID	pH	EC	TDS	Na^{+}	K^{+}	Mg^{2+}	Ca^{2+}	Cl^{-}	HCO_3^{-}	NO_2^{-}	NO_3^{-}	SO_4^{2-}	P	Cd	Hg	Pb
		(μS/cm)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(μg/L)	(mg/L)	(μg/L)
Average	6.5	970.368	599.631	29.496	6.672	30.696	92.796	49.807	381.94	3.123	2.984	7.65	0.415	1.855714	1.175	8.12
Maximun	7.1	1616	1034	80	12.6	54.13	164.25	305.2	580	9.61	8.8	74.56	2.18	9.28	25.85	21.34
Minimum	5.9	520	235	11.25	2.48	3.96	17	6.84	122.9	0	0	0	0.03	0	0	0.0016
Range	1.2	1096	799	68.75	10.12	50.17	147.25	298.36	457.1	9.61	8.8	74.56	2.15	9.28	25.85	21.338
SD	0.346	281.192	202.825	20.646	2.839	11.324	29.054	52.109	84.022	3.412	3.316	15.173	0.523	2.546116	4.785	11.547

silty, very fine sandstone with interbedded fine to coarse sandstone [26]. A series of long, marshy depressions distinguished the line as a fault line which seem to be sustained by water rising along the fault from the sandstone aquifer inside the Barind Tract Formation.

2.2. Groundwater sampling, processing and analysis

Thirty-eight ground water samples were obtained from the research area for major ionic compounds and trace elements research studies (Table 1). The village of Rajarampur provided twenty samples (D-1 to D-20), and the village of Baragharia provided eighteen samples (D-21 to D-38) from the Chapai Nawabganj Paurasava. Groundwater samples (composite) were gathered at depths ranging from 20 to 50 m in Chapai Nawabganj, northwest Bangladesh, using manual and electric pumps. Plastic bottles of 500 mL have been previously cleaned and were used to collect Groundwater samples. To obtain fresh water, each well was pumped manually for two to 3 min to get fresh water prior to sampling. The samples were taken in triplicate and placed in three different plastic bottles with airtight closures. In order to determine trace elements, during sampling acidification of water samples were made with the help of concentrate HNO_3^- (2 mL/L) to obtain pH < 2, whereas fresh groundwater (non-acidified) was accumulated for negative ions analysis. In accordance with the recommendations of researchers [27–30], all samples were tagged, promptly corked to preserve them airtight, transferred to the laboratory as soon as possible, and kept at 4 °C.

A portable bench meter with a microprocessor (Model: HI 9813-6, Hanna Instrument, Portugal) was used to test the water samples pH, temperature, electrical conductivity (EC), and total dissolved solids (TDS) in-situ. The Institute of National Analytical Research and Service (INARS), an ISO/IEC 17025:2017 accredited laboratory, of the Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka, Bangladesh, assessed the concentrations of chemical elements and anion. Groundwater was tested for the chemical elements As, Fe, Mn, Mg, and Ca using atomic absorption spectrophotometers (AAS, Models: AA240FS and SpectrAA220, Varian, Australia), whereas Na^+ and K^+ were tested using flame photometers (Model: PFP7, Jenway, UK). Cl^-, NO_3^-, and SO_4^{2-} ion concentrations were measured using an ion chromatograph (IC, Model: SIC10AVP, Shimadzu, Japan). The titration technique was then used to calculate the HCO_3^- concentration [27].

In the laboratory analysis, the Atomic Absorption Spectrometer (AAS) is utilized. This involves sample preparation, where groundwater samples undergo digestion to ensure the elements are in a suitable form for analysis, followed by atomization and absorption measurement in the AAS. Calibration standards and quality control measures, such as blanks and standards, are used to ensure the accuracy and precision of the AAS results. Other tools and techniques for assessing water quality parameters include pH meters, conductivity meters, dissolved oxygen (DO) meters, turbidity meters, ion chromatography (IC), gas chromatography (GC), mass spectrometry (MS), and microbiological analysis. Quality control measures, including the use of blanks, duplicates, and certified reference materials, are implemented to ensure the accuracy and precision of the results. Finally, data interpretation and reporting involve interpreting the results in the context of relevant water quality standards and guidelines and generating a comprehensive report outlining the findings and implications for groundwater quality. It's important to note that the specific methods and tools used can vary depending on the study's goals, contaminants of interest, and local regulatory requirements.

2.3. Study of water usage patterns in the research area

The average rates of drinking water intake were 3.2 L and 2.8 L for males and females who are adult in the research region respectively (data not shown). Our study area's average daily water consumption is virtually identical to the results of 3.2 L and 2.7 L day^{-1} for adult men and females, respectively [31,32] in Bangladesh and in West Bengal, India (3.1 and 2.6 L day^{-1} for adult males and females, respectively).

2.4. Mapping and data analysis

The research area was mapped using Arc-GIS software (version 10.8), and the measured characteristics of the water samples were distributed spatially. For the statistical analysis of the observed water quality parameters and the presentation of the water chemistry-related diagrams, AquaChem, PHREEQC, and SPSS (version 20, IBM Corporation, Armonk, New York, USA) were used. We used Microsoft Excel 2013 to do our standard analytical calculations.

2.5. Drinking water quality and health risk assessment

The entropy water quality index was used to figure out the quality of groundwater for drinking. In order to calculate the health hazards, the United States Environmental Protection Agency's [33] average daily dose (ADD), hazard ratio (HQ), and cancer risk (CR) were calculated.

2.6. Exposure assessment

2.6.1. Groundwater quality assessment

The physical and chemical characteristics of the groundwater were compared to the normative pattern advised by the different organization recommended standard. The Entropy water quality index (EQWI) was also calculated.

2.6.2. Human health risk assessment

The harmful effects of As contamination in drinking water on people's health were assessed using the human health risk assessment model developed by US-EPA Eqs (1)–(3) in two research locations [22,34]. With the help of the available data, health risk assessment may be evaluated critically and simply without the need for in-situ toxicological dosage assessment. By employing the average daily dose (ADD) and the hazard quotient (HQ) values according to the provided Eqs. (1) and (2), health risk from toxic metals that are not carcinogenic is assessed. The health risk assessment was done to estimate the probability of individuals being exposed to As poisoning from drinking water. For this purpose, average daily dose (ADD) of As due to the intake of As-contaminated drinking water was calculated by Eq (1):

$$ADD = \frac{C \times IR \times ED \times EF}{BW \times AT} \dots\dots\dots \quad (1)$$

where:

C = Concentration of As in water (mgL^{-1})

Table 2
Results of trace elements analysis of the groundwater with their descriptive statistics, recommended values and entropy water quality index (EWQI).

Sample ID	As (μg/L)	Fe (mg/L)	Mn (mg/L)	EWQI
D-1	21.00	7.95	2.05	160.82
D-2	8.50	2.94	3.27	37.86
D-3	36.75	0.10	1.70	37.18
D-4	85.50	13.00	1.59	184.98
D-5	8.75	3.15	0.23	150.54
D-6	2.25	0.87	1.43	35.93
D-7	1.00	0.23	1.17	31.85
D-8	66.75	4.05	1.83	46.03
D-9	18.25	27.35	1.53	150.99
D-10	133.75	10.25	1.94	181.42
D-11	9.50	12.25	2.25	12.47
D-12	19.25	52.75	1.93	260.58
D-13	34.50	1.03	1.03	28.40
D-14	164.00	5.57	1.21	159.19
D-15	33.40	6.24	0.89	156.90
D-16	41.25	3.53	1.03	41.23
D-17	1.50	0.29	1.65	43.48
D-18	34.00	3.17	1.43	42.10
D-19	14.50	0.05	1.36	32.48
D-20	2.25	0.12	1.63	35.25
D-21	1.75	0.16	0.28	29.23
D-22	3.25	0.94	0.54	44.55
D-23	1.75	0.33	0.63	32.19
D-24	2.00	0.22	0.38	30.25
D-25	2.50	0.44	0.72	40.60
D-26	1.75	0.06	0.68	49.44
D-27	1.75	0.14	0.37	28.35
D-28	1.75	0.31	0.63	32.84
D-29	1.00	0.30	0.73	29.62
D-30	1.25	0.27	0.39	27.30
D-31	1.25	0.91	0.35	29.74
D-32	1.00	0.23	0.46	26.89
D-33	4.00	0.24	0.51	24.18
D-34	1.75	0.53	0.72	40.55
D-35	2.50	0.15	0.77	38.46
D-36	1.00	0.04	0.68	34.31
D-37	1.25	0.23	0.76	37.11
D-38	0.50	0.54	1.12	35.56
Average	20.227	4.235	1.101	64.23
Maximun	164.00	52.75	3.27	260.58
Minimum	0.50	0.04	0.23	12.47
Range	163.50	52.71	3.04	248.11
SD	36.463	9.682	0.670	60.83
Drinking Water Standard				
WHO (2007,1997)	10	0.3	0.1–0.50	
ECR (1997)	50	0.3–1.0	1	
USEPA (2005, 2004, 1998)	10	2	0.5	
EU (1998)	10	0.2	0.05	
BIS (1991)	50	0.3	1	
DoE (1997)	50	0.3–1.0	0.1	

Table 3
Comparison of groundwater quality parameters with relevant literature data.

Sample area	Na^+ (mg L^{-1})	K^+ (mg L^{-1})	Ca^{2+} (mg L^{-1})	Mg^{2+} (mg L^{-1})	Cl^- (mg L^{-1})	HCO_3^- (mg L^{-1})	SO_4^{2-} (mg L^{-1})	As (μg L^{-1})	Fe (mg L^{-1})	Mn (mg L^{-1})	EWQI	References
Chapai Nawabganj	29.50	6.68	92.80	30.70	49.81	381.95	7.65	20.23	4.02	1.08	64.24	This Study
Meherpur	31.80	8.90	64.80	34.20	31.40	388.70	8.80	156.90	2.32	0.72	302.002	Shazzad et al. (2023)
Jessore,	68.31	1.39	67.74	27.40	43.55	405	0.90	15.97	0.20	0.48	–	Kabir et al. (2021)
Kushtia	26.87	4.73	88.81	33.54	64.09	388.50	3.19	0.37	0.97	0.33	–	Hossain et al. (2013)
Faridpur	35.34	5	103.80	32.92	23.95	542.40	5.27	118.60	5.95	0.001	–	Bodrud-Doza et al. (2016)
Khulna	647.20	17.05	101.50	78.28	1776.70	510.10	4.97	–	–	–	–	Islam et al. (2017)
Gopalganj	547.30	8.14	106.50	80.47	847.40	263.50	–	50.31	5.13	0.20	–	Islam et al. (2018)
Barguna&Patuakhali	863	16.20	136	155	3513	254	19.50	–	–	–	–	Islam et al. (2016)
Noakhali	146.30	12	23.20	40.76	112.60	76.53	11.40	3.93	1.95	0.13	111.80	Islam et al. (2021)
Chottogram	9.58	0.77	6	3.67	4.44	27.06	0.03	0.12	1.14	22.20	–	Rifat et al. (2021)
Laksam, Cumilla	174	–	30.90	52.90	180	388	1.68	199	4.68	0.37	171	Rahman et al. (2022)
Sylhet	39.48	2.33	7.62	4.83	21.23	123.70	1.40	39.30	8.73	0.21	–	Ahmed et al. (2019)
Rangpur	39.10	6.51	12.63	7.55	78.64	95.50	7.93	–	–	–	–	Saha et al. (2019)
Rajshahi	23.12	5.66	78.35	27.02	55.32	245.30	38.80	17	3.10	1.47	–	Mostafa et al. (2017)

IR = Water ingestion rate (Lday^{-1})
ED = Exposure duration (to compare with earlier research from Pakistan and other nations, we used the 67-year-old assumption.)
EF = Exposure frequency (365 days year^{-1})

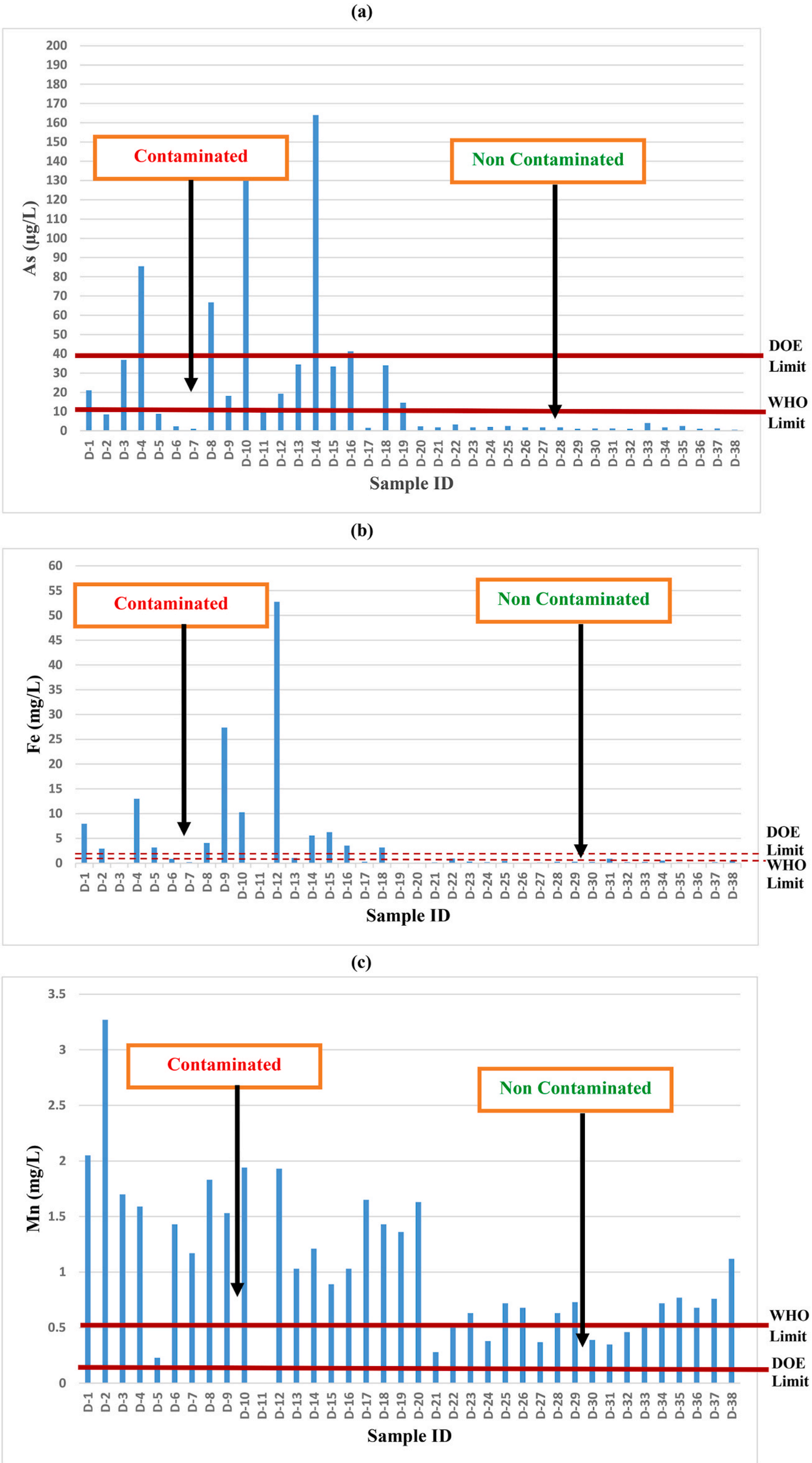


Fig. 2. Contamination levels in the groundwater samples, 2(a) As, 2(b) Fe and 2(c) Mn collected from the study area.

BW = Body weight (72 kg, [35])
AT = Average life time (24,455 days)
Degrees of chronic and carcinogenic risk were also assessed in study participants. The formula (2) was used to calculate the hazard quotient (HQ), ([34]):

$$HQ = \frac{ADD}{RfD} \dots\dots\dots\dots \tag{2}$$

where RfD represents oral reference dose (0.0003 $mgkg^{-1}day^{-1}$) for As calculated by US-EPA (2005) [36].
Cancer risk (CR) was calculated by using Eq (3):

$$CR = \frac{ADD}{CSF} \dots\dots\dots \tag{3}$$

where CSF is the cancer slope factor for As which is 1.5 $mgkg^{-1}day^{-1}$ [36]

3. Results

3.1. Groundwater chemistry

In Table 1, the physicochemical variables, cations, and anions of the examined groundwater samples are detailed. The pH ranged

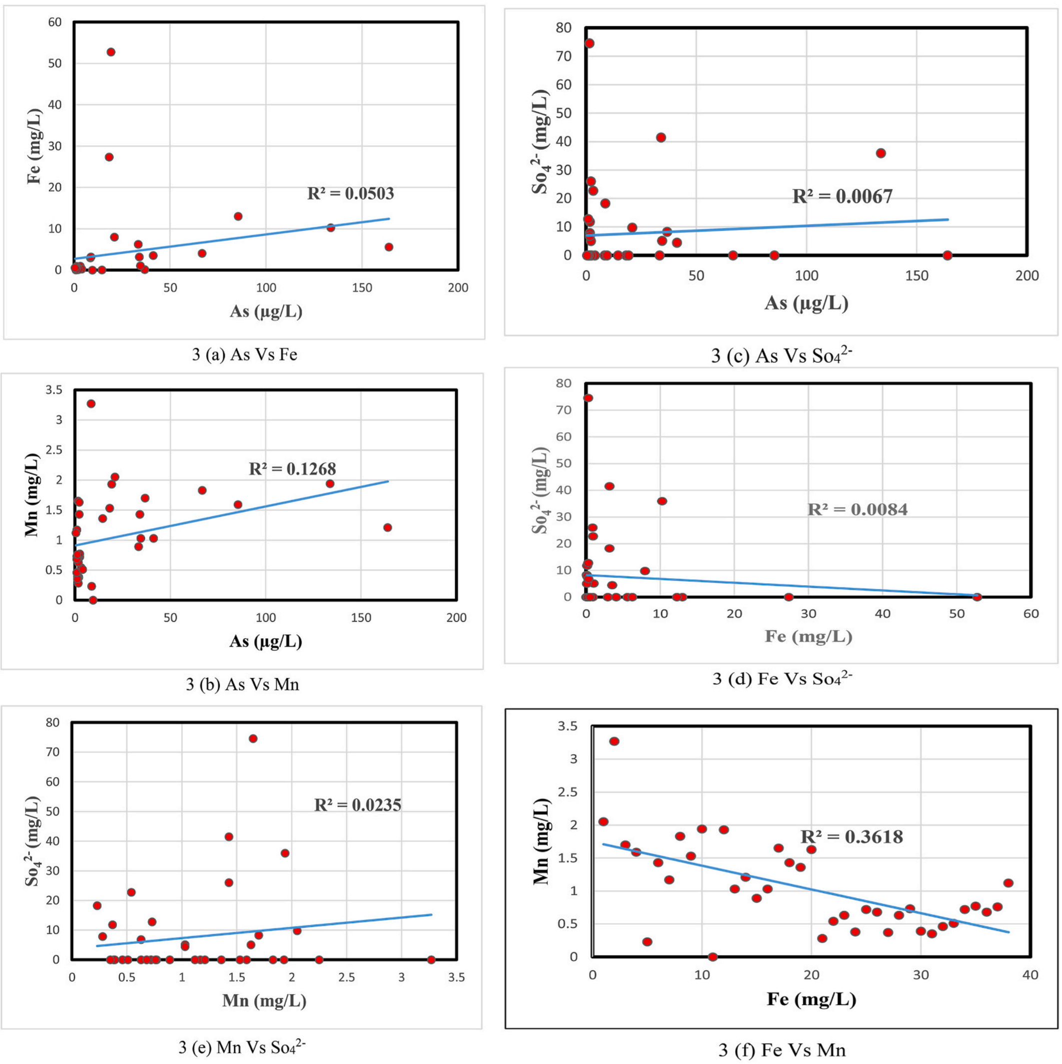


Fig. 3. Correlations for 3(a) As vs. Fe, 3(b) As vs. Mn, 3(c) As vs. SO_4^{2-}, 3(d) Fe vs. SO_4^{2-}, 3(e) Mn vs. SO_4^{2-}, and 3(f) Fe vs. Mn.

from 5.9 to 7.1 (mean ± SD: 6.55 ± 0.346), indicating slightly acidic to neutral conditions within an acceptable range. The electrical conductivity (EC) values ranged from 520 to 1616 μS/cm (mean ± SD: 970 ± 281.19), suggesting slightly salty water. Major cations (Na^{+}, K^{+}, Ca^{2+}, Mg^{2+}) and anions (HCO_3^-, Cl^-, NO_3^-, SO_4^{2-}) concentrations were generally within permissible limits, with exceptions for Mg^{2+} (in sample D-17) and Ca^{2+} (in 31.57% samples). Some samples surpassed hydrogen carbonate (HCO_3^-) and sulfate (SO_4^{2-}) recommendations.

Trace mineral analysis in Table 2 revealed concentrations of As, Fe, and Mn in groundwater samples. Arsenic concentrations ranged from 0.5 to 164 μg/L (mean ± SD: 20.22 ± 36.46), with 34.21% of samples exceeding the WHO guideline of 10 μg/L. Groundwater quality summary in Table 3 compared results with various guidelines, indicating notable contamination in some samples.

The study region, particularly the south, showed significantly high levels of As contamination in groundwater. Trace elements, including As, Mn, and Fe, were identified as geogenic in origin. The average As concentration of 20.22 μg/L in the study region was higher than the national average. A graphical representation (Fig. 2a) indicated that 34.21% of samples exceeded WHO's recommended value for arsenic. Additionally, 42.10% of samples surpassed the WHO limit for Fe (0.3 mg/L), and 81.57% exceeded the WHO standard for Mn (0.5 mg/L) (Fig. 2b–c).

Correlation studies (Fig. 3a–f) revealed poor correlations between As, Fe, and SO_4^{2-}. Positive correlations among soluble As, Fe, and Mn, as well as sequential extraction studies, suggested the importance of reductive dissolution of MnOOH and FeOOH in the presence of anaerobic bacteria as a mechanism for As discharge from sediment to groundwater (see Fig. 4).

The shallow aquifers in the Bengal delta plain were identified as being polluted by As, originating from eroding Himalayan sediments. The spatial distribution of As, Fe, and Mn showed higher concentrations in the southern region, particularly in Rajarampur village of Chapai Nawabganj Paurosava compared to Baragharia village. Vertical profiling indicated higher arsenic concentrations at depths between 18 and 27 m, suggesting contamination in the shallow and intermediate units deposited between 10,000 and 5500 years ago. The ongoing oxidation-reduction cycle in the subsurface was attributed to variations in the water table.

3.2. Spatial distribution of hydro-chemical parameters in groundwater

Spatial distribution maps [Fig. 5(a-j)] revealed variations in Total Dissolved Solids (TDS), EC, pH, arsenic, iron, manganese, ADD, HQ, cancer risk (CR) and EWQI across the study area. The southern part exhibited higher mineralization, elevated concentrations of arsenic, iron, and manganese, and increased cancer risk.

3.3. Understanding the source and analysis of studied groundwater

Principal Components Analysis (PCA) results (Table 4, Fig. 6) identified four main components explaining 74.271% of variance. Positive loadings of Na^{+}, K^{+}, Mg^{2+}, As, Fe, Mn, and NO_3^- in different components indicated the diverse sources and interactions of chemical elements. Hierarchical Cluster Analysis (CA) (Fig. 7) grouped water samples into four clusters based on similarities, highlighting influences of septic tank effluents, salt dissolution, calcite dissolution, and arsenic enrichment factors.

The Piper diagram (Fig. 8) classified groundwater samples as Ca–HCO3 type, providing further insights into hydro-chemical facies and aquifer regulation.

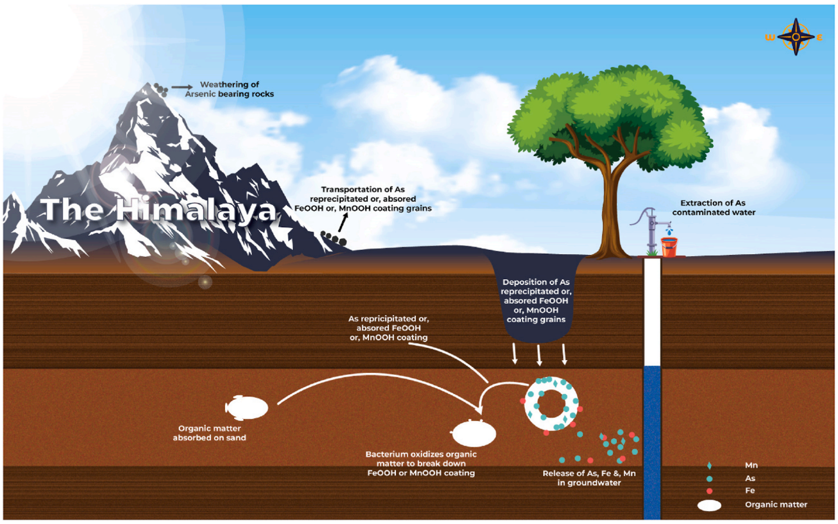


Fig. 4. Hypothetical model showing release of arsenic from sediment into groundwater (Source: Reza et al., 2010).

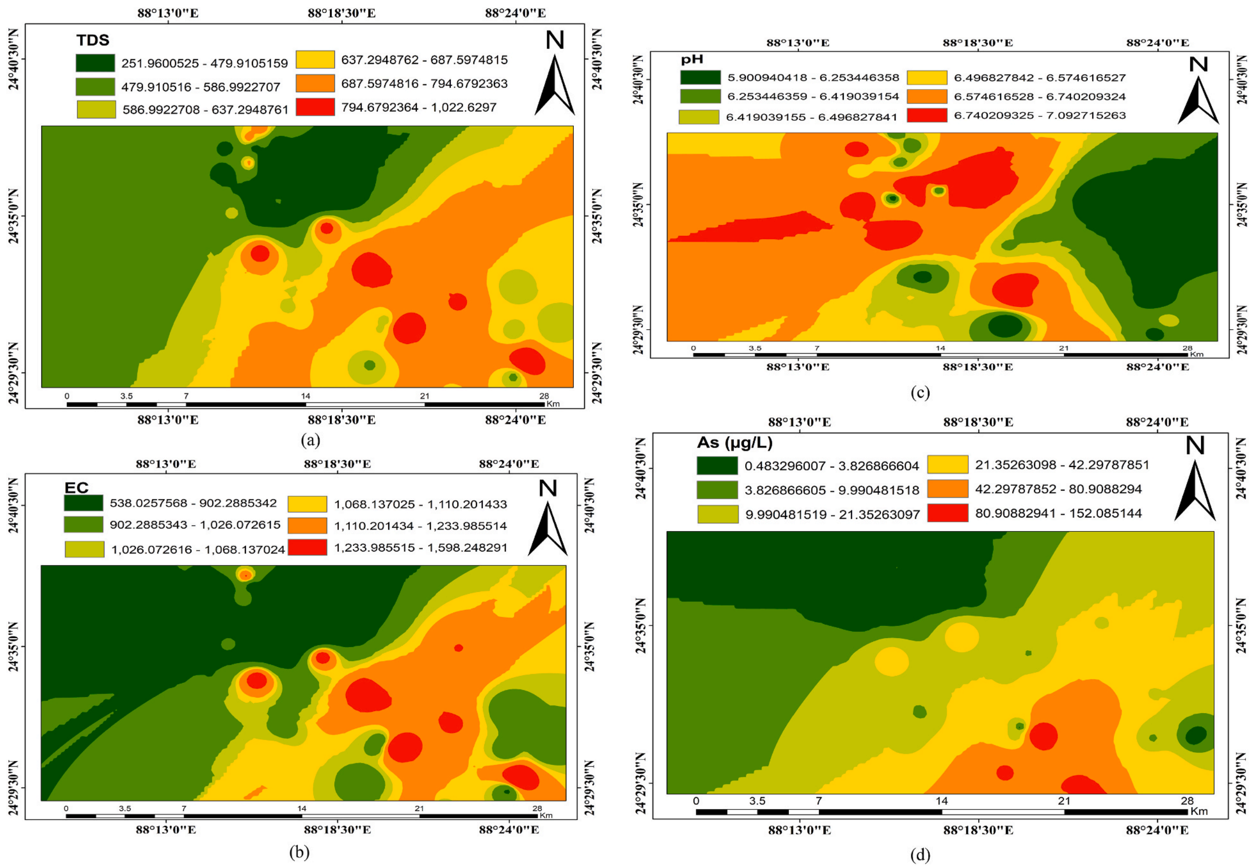


Fig. 5. Spatial distribution of 5(a) TDS, 5(b) EC, 5(c) pH, 5(d) As, 5(e) Fe, 5(f) Mn, 5(g) ADD, 5(h) HQ, 5(i) CR, 5(j) EWQI for groundwater of northwestern Bangladesh.

3.4. Classification of groundwater and EWQI assessment

Researchers often categorize groundwater using various criteria. In this study, physical and chemical characteristics were compared to recommended standards to determine drinking water quality. The Entropy Water Quality Index (EWQI) ranged from 12.47 to 260.58 (mean ± SD: 64.23 ± 60.83), with 21.05% of water samples deemed unfit for human consumption (EWQI >150) (Table 5).

3.5. Health risk assessment

Table 6 presented the health risk assessment of arsenic exposure, including Average Daily Dose (ADD), Hazard Quotient (HQ), and Cancer Risk (CR). ADD values were comparable to other regions, indicating inorganic arsenic exposure through drinking water. HQ values exceeding the hazardous risk index of 1.00 suggested potential health risks. The calculated CR values exceeding the US-EPA limit ($10^{\hat{}-6}$) indicated a high risk of cancer, emphasizing the need for continuous monitoring of arsenic levels in groundwater.

3.6. Comparison of this study with other related research

Supplementary Table 1 presents a comparative analysis of groundwater chemistry from various locations in Bangladesh, with a specific focus on the newly studied area in Chapai Nawabganj. The data provides insights into the variability of water quality parameters across different regions. The Meherpur region exhibits moderate levels of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-}, with a relatively high arsenic concentration of 156.90 μg/L. Compared to Chapai Nawabganj, Meherpur has higher Na^+, Ca^{2+}, and Mg^{2+} levels, while Chapai Nawabganj has a slightly higher concentration of Cl^- and HCO_3^-. Jessore's groundwater shows lower levels of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, but both areas have elevated arsenic levels, with Jessore having a higher concentration. Kushtia demonstrates lower Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and arsenic levels in Kushtia are negligible. Faridpur has higher concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, but arsenic levels are lower in Faridpur compared to Chapai Nawabganj. Khulna exhibits

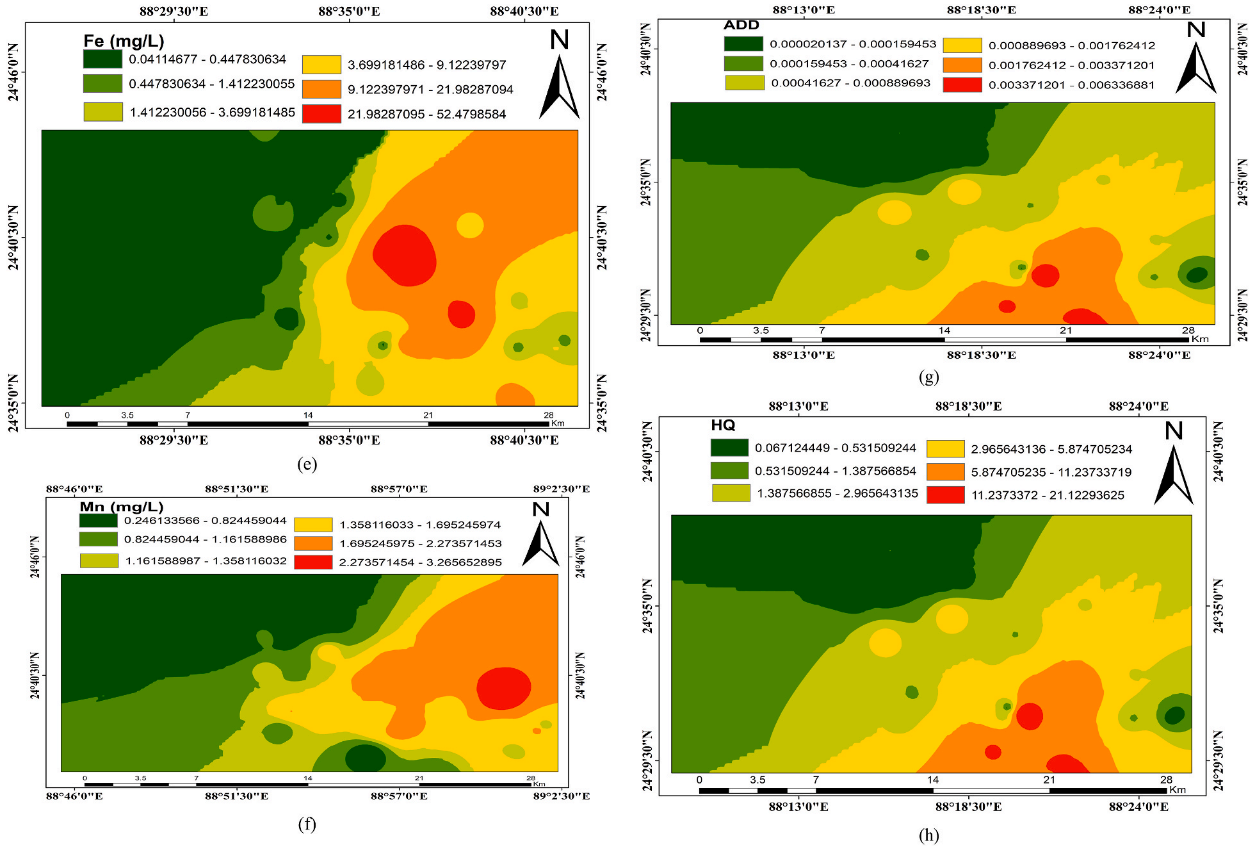


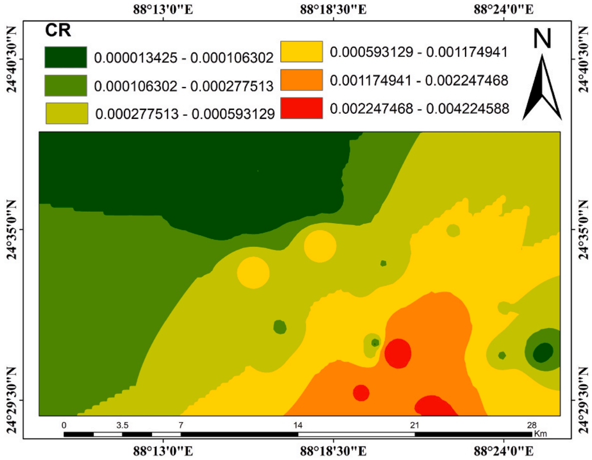
Fig. 5. (*continued*).

significantly higher concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and the arsenic concentration in Chapai Nawabganj is lower than that in Khulna. Gopalganj's groundwater has higher levels of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and arsenic levels in Gopalganj are lower than in Chapai Nawabganj. Barguna & Patuakhali show significantly higher concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and arsenic levels in Chapai Nawabganj are lower compared to Barguna & Patuakhali. Noakhali has higher concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and both areas show similar levels of arsenic. Chottogram exhibits lower concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and arsenic levels are lower in Chottogram than in Chapai Nawabganj. Laksam, Cumilla, demonstrates higher Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} levels compared to Chapai Nawabganj, and arsenic levels in both areas are comparable. Sylhet has lower concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} compared to Chapai Nawabganj, and arsenic levels are lower in Sylhet compared to Chapai Nawabganj. Rangpur exhibits lower Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-} concentrations compared to Chapai Nawabganj, and arsenic levels are lower in Rangpur than in Chapai Nawabganj. The newly studied area in Chapai Nawabganj has moderate levels of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, HCO_3^-, and SO_4^{2-}, with a relatively lower arsenic concentration of 20.23 μg/L. The Entropy Water Quality Index (EWQI) of 64.24 suggests a moderate overall water quality.

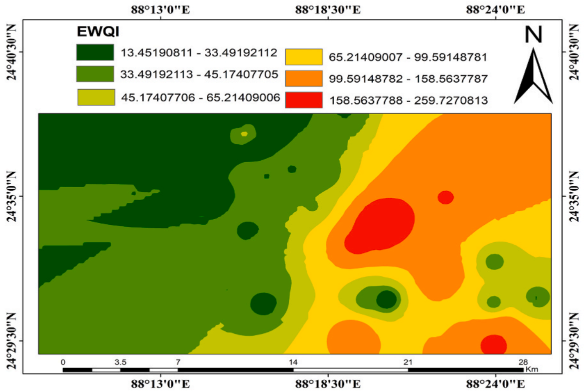
A comparison highlights the spatial variability in groundwater chemistry across different regions of Bangladesh. While Chapai Nawabganj shares similarities with some areas, unique characteristics such as arsenic levels distinguish it from others. The comprehensive understanding of regional water quality is crucial for informed water resource management and public health initiatives.

4. Discussion

The study found that the groundwater samples from Rajarampur and Baragharia village in northwest Bangladesh have almost neutral and declining aquifer conditions. The groundwater is typically acidic to neutral, with electrical conductivities between 520 and 1616 μS/cm, making it a great choice for irrigation and taste-wise categorized as non-saline. The range of the TDS measurement, from 234 to 1035 mgL^{-1}, shows that the water is palatable. The Piper tri-linear diagram revealed Ca–HCO_3 kinds of groundwater, with Ca^{2+} and HCO_3^- as the predominate cation and anion, respectively. Fe, Mn, and As are the major trace elements in the study area. While Fe concentrations of most of the groundwater samples are highly exceeded the WHO standard for drinking water quality in Rajarampur village, only a few samples slightly exceed this safe limit in Baragharia village. Mn concentrations are also higher than the WHO safe



(i)



(j)

Fig. 5. (*continued*).

Table 4
Varimax rotated principal component analysis.

Parameters	PCA-1	PCA-2	PCA-3	PCA-4
Na+	**0.850**	0.021	0.160	0.210
K+	**0.520**	−0.371	0.009	−0.078
Mg2+	**0.865**	0.097	0.236	−0.125
Ca2+	−0.186	0.204	0.250	**0.815**
Cl-	0.349	0.041	**0.889**	−0.004
HCO3-	0.426	0.112	−0.378	**0.781**
As	−0.390	**0.619**	0.049	0.326
Fe	0.152	**0.726**	−0.224	−0.272
Mn	0.174	**0.734**	0.129	0.271
NO3-	−0.214	**0.691**	0.215	0.196
SO42-	0.065	0.075	**0.933**	0.058
Total variance %	21.230	19.421	18.752	14.868
Cumulative % of variance	21.230	40.650	59.402	74.271

limit in almost all the groundwater samples in Rajarampur village, whereas Mn concentrations of most of the samples in Baragharia village are moderately higher than the WHO standard. Most of the groundwater samples in Rajarampur village run over the safe limit of As concentration set by WHO but As concentrations are less than the safe limit in all samples of Baragharia village. Positive correlations between dissolved As, Fe, and Mn concentrations imply that anaerobic bacteria play a key role in the reductive breakdown of MnOOH and FeOOH, where As is released from sediment into the groundwater. Poor relationships between the amounts of dissolved As, Fe, and SO_4^{2-} in groundwater show that sulfide/pyrite is not the main pathway for arsenic to be transferred from sediment to groundwater. Groundwater As comes from geogenic sources, according to principal component and cluster analyses. To determine the likelihood that people would become ill from As poisoning from drinking water, the ADD, HQ, and CR are analyzed. Groundwater

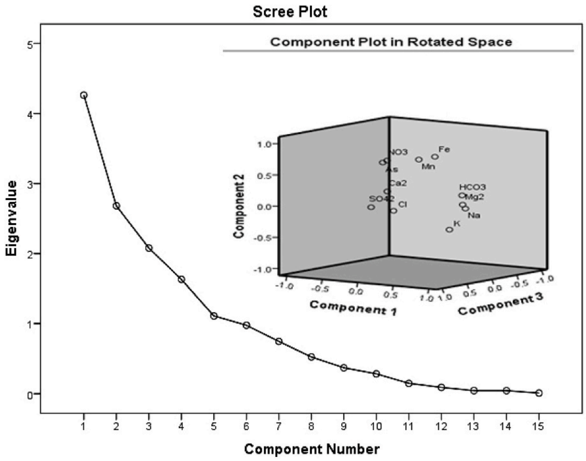


Fig. 6. PCA analysis with component plot in rotated space and scree plot of element.

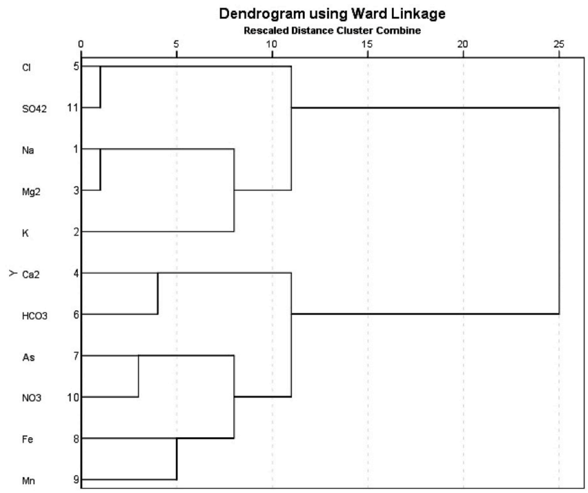


Fig. 7. Cluster analysis of elements in the groundwater samples.

samples from Rajarampur village consistently have greater ADD, HQ, and CR values than those from Baragharia village. The findings of HQ values show that the majority of Rajarampur village's drinking water samples were over the average hazardous risk index of 1.00. The CR values above the US-EPA standard (10^{-6}), indicating that the inhabitants of the research region were at a high risk of developing cancer. The Rajarampur village area's groundwater is not acceptable for irrigation or consumption, according to the Entropy Water Quality Index.

Overall, the results and discussion highlight the complexity of groundwater chemistry, the spatial distribution of contaminants, and the associated health risks in the study region. Continuous efforts are crucial to manage and mitigate groundwater contamination for the well-being of the local population.

5. Conclusion

In conclusion, the study found that the groundwater in Rajarampur and Baragharia village in northwest Bangladesh is contaminated with arsenic, iron, and manganese, with Rajarampur village having higher concentrations than Baragharia village. The groundwater in Rajarampur village is not acceptable for irrigation or consumption, and the inhabitants of the research region are at a high risk of developing cancer due to the high levels of arsenic in the water. To provide residents with clean drinking water, integrated water resource management is crucial, and zoning mapping should be included to maintain optimal public health. Regular monitoring of the amounts of arsenic in the groundwater is also necessary. The study proposed several sustainable strategies for mitigating arsenic contamination in drinking water, including substituting high-arsenic sources, such as groundwater, with low-arsenic, microbiologically safe sources such as rainwater and treated surface water for drinking, cooking, and irrigation purposes, and undertaking arsenic-mitigation actions within a framework of health-based targets and independent surveillance to ensure that exposure to arsenic through

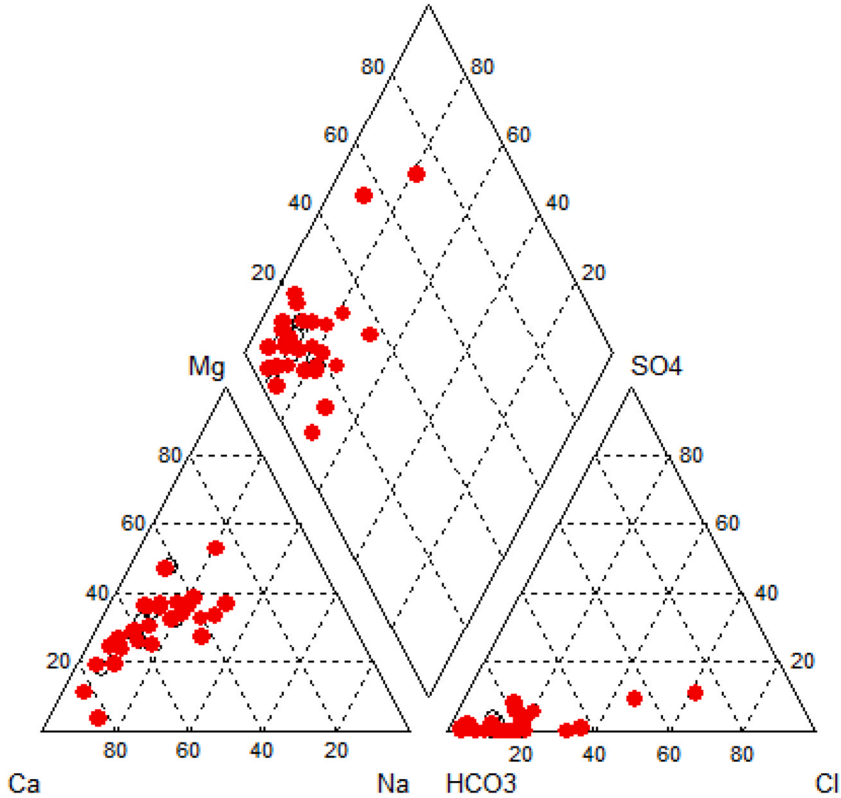


Fig. 8. Piper diagram illustrating the main hydrochemical features of the groundwater.

Table 5
Water quality classification criteria based on EWQI (Su et al., 2018).

EWQI	Rank	Water quality
<25	1	Excellent
25–50	2	Good
50–100	3	Medium
100–150	4	Poor
>150	5	Extremely poor

Table 6
Summary of Average Daily Dose, Hazard Quotient and Carcinogenic Risk of Asin groundwater samples collected from Rajarampur and Baragharia village, Chapai Nawabganj district, Bangladesh.

Parameter		Rajarampur village (D-1 to D-20	Baragharia village (D-21 to D-38)
As (μg/L)	Mean	36.208	1.778
	Range	1–164	0.5–4
Average Daily Dose ($mg^{-1}kg^{-1}day^{-1}$)	Mean	1.5×10^{-3}	7.42×10^{-5}
	Range	4.17×10^{-5}– 6.83×10^{-3}	2.08×10^{-5} – 1.67×10^{-4}
Hazard Quotient	Mean	5	0.247
	Range	0.139–22.778	0.069–0.556
Carcinogenic Risk	Mean	0.001	4.95×10^{-5}
	Range	2.78×10^{-5} - 4.56×10^{-3}	1.39×10^{-5} – 1.11×10^{-4}

all possible routes is sufficiently mitigated and the potential substitution of arsenic with other risks is adequately managed. The limitations of the research include data limitations, model assumptions, spatial and temporal scope, interdisciplinary factors, and policy and implementation implications. The limitations of the research can be discussed as follows: 1. Data Limitations: The research may be limited by the availability and quality of data on groundwater arsenic contamination, human health impacts, and environmental conditions. The study relies on localized studies, which may not provide a comprehensive understanding of the entire region. 2. Model Assumptions: GIS-based assessments often rely on various assumptions and simplifications, which can introduce uncertainties and limitations to the findings. 3. Spatial and Temporal Scope: The study may be limited in its spatial and temporal scope, potentially not capturing the full extent of groundwater vulnerability and human health impacts. The research may not account for the variability in arsenic concentration and the different toxicity and removability of arsenic species. 4. Interdisciplinary Factors: Integrating GIS, hydrogeology, and human health assessments can be challenging, and the research may be limited by the interdisciplinary nature of the study. The study needs to consider the different aspects of arsenic contamination, such as localized studies, global overviews, and the toxicity and removability of arsenic. 5. Policy and Implementation Implications: The research may not address the practical implications of the findings, such as policy recommendations and implementation strategies. The study should focus on strategies for mitigation, including the removal of arsenic from water sources and the prevention of arsenic exposure.

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Data availability

Data will be made available on request.

Ethical approval

Not applicable.

Consent to participate

Not applicable.

Consent to publish

Not applicable.

CRediT authorship contribution statement

Md Ahsan Habib: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **A.H.M. Selim Reza:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Md Irfanul Hasan:** Visualization, Validation, Software, Resources, Formal analysis, Data curation. **Md Aminul Ahsan:** Validation, Supervision, Resources, Methodology, Formal analysis, Data curation. **Md Moniruzzaman:** Validation, Resources, Formal analysis, Data curation. **Asma Binta Hasan:** Validation, Resources, Formal analysis, Data curation. **Shofiul Islam Shofi:** Validation, Formal analysis, Data curation. **Kayesh Mahmud Hridoy:** Visualization, Software.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e27917.

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