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ORIGINAL PAPER

Evaluation and health risk assessment of arsenic and potentially toxic elements pollution in groundwater of Majha Belt, Punjab, India

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Abstract Concentrations of potentially toxic elements (PTEs) like arsenic, uranium, iron, and nitrate in the groundwater of the Majha Belt (including Tarn Taran, Amritsar, Gurdaspur, and Pathankot districts) in Punjab, India were measured to evaluate the health risks associated with its consumption and daily use. The average concentrations of these elements in some locations exceeded the WHO-recommended values. Arsenic and iron toxicity levels were found to be higher in the Amritsar district, while uranium toxicity was more prevalent in Tarn Taran. The Trace Element Evaluation Index suggests that Amritsar is one of the districts most affected by toxic elements. According to the US Environmental Protection Agency's (USEPA) guidelines, the HQ values of U, Fe, and nitrate were less than one, indicating that there is no non-carcinogenic health risk for adults and children. However, the hazard quotient (HQ) value for arsenic was greater than one, indicating a higher possibility of health risk due to arsenic in the study area. The total hazard index values of 44.10% of samples were greater than four for arsenic, indicating that people in the Majha Belt are at a very high health risk due to the usage of water for drinking and domestic purposes. The cancer risk assessment values for arsenic in children (5.69E + 0) and adults (4.07E + 0) were higher than the accepted limit of USEPA (10^{-4} to 10^{-6}) in the Majha Belt. The average radiological cancer risk values of U for children and adults were 8.68E-07 and 9.45E-06, respectively, which are well below the permissible limit of 1.67×10^{-4} suggested by the Atomic Energy Regulatory Board of DAE, India. The results of this study confirm that the residents of the Majha Belt who use contaminated groundwater are at a serious risk of exposure to arsenic in the Amritsar district and uranium in Tarn Taran district.

Keywords Groundwater · Arsenic · Uranium · Nitrate · Iron · Majha belt

Introduction

Water is one of the most underrated but valuable natural resources that occurs and covers about 71% of the earth's surface. Out of it, 3% are fresh, and the remaining 97% are salty. The presence of water on the earth makes the planet suitable for the occurrence and existence of life. Based on its occurrence, whether at the surface or subsurface of the earth, water can be classified as surface water (ocean, sea, rivers, ponds, and lakes) and subsurface water (groundwater). In many countries, groundwater plays a major role where there is a lack of freshwater resources such as

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coastal regions, arid regions, and metropolitan areas (Dehghani et al., 2019 and Sajjadi et al., 2022). In India, 85% of drinking water and 89% of agricultural and industrial needs are supported by groundwater (CGWB 2019; Mukherjee et al., 2020; Rani et al., 2023). However, such a valuable asset is now under intense stress due to uncontrolled and express growth of population, irresponsible administration, and industrial-agricultural actions (Mohammadpour et al., 2022; RadFard et al., 2019; Saleh et al., 2018). The quality of water is degraded mainly due to activities such as discharge of sewage and wastewater in aqueous systems, radioactive waste, atmospheric deposition, industrial waste, oil pollution, leakage from underground pipes and storage containers, global warming, rock water interaction, over-pumping, landfills and eutrophication (Gambhir et al., 2012; Li et al., 2018; Şener, 2023). Such intensive stress on water causes degradation of water quality and quantity. The stress on groundwater resources in India, i.e., water deficit and pollution, directly affects human health, crop growth, and yield. It in turn retards socioeconomic development and threatens ecosystems (Adimalla, 2020; Hua et al., 2020; Ma et al., 2020; Turan et al., 2019).

There are several contaminants of groundwater among which certain compounds, heavy metals and metalloids have attracted greater attention, due to their high toxicity, persistence, non-biodegradability characteristics, and potential for bioaccumulation in food chains (Sajjadi et al., 2022; Kulkarni et al., 2018 and Sadeghi et al., 2020). Such contaminants are collectively called Potential Toxic Elements (PTEs). It includes As, Pb, Sb, Cu, Cd, Co, Zn, Ni, Fe, Hg, Mn, Mo, Se, nitrate, fluoride, and radioactive elements like U (Bounar et al., 2020; Heshmati et al., 2020; WHO, 2017). These PTEs can enter the groundwater through geogenic processes (weathering, erosion, soil formation and the rock cycle transport and redistribution of heavy metals in the soil, water and air) and anthropogenic processes (mining, industries, agriculture, landfills, over-pumping, sewage, etc.) (Sajjadi et al., 2022). Also, oil shale in situ exploitation processes such as fracturing and pyrolysis can increase the permeability of rocks and have a profound effect on the water–rock interaction process, thereby leading to the diffusion of contaminants into groundwater (Li et al., 2022). The red mud fly ash material which is composed of toxic metals like Pb, As, Hg, Cr, etc. is used for constructional projects due to its high compressive strength under an alkaline environment. However, the same material under acidic conditions can liberate those toxic elements into the environment (Bai et al., 2024). In general, the elements with atomic masses in the range of 63.54–200.59 g/mol and densities above 4.0 g/cm^3 are referred to as heavy metals (Jiang et al., 2021). Heavy metals and metalloids are the natural elements that exist in the earth's crust. Some of these elements, like Zn, Ni, Cu, Se, and Co, are essential for all living things in very low concentrations. But other elements like As, Pb, Sb, Fluoride, and U are highly toxic even at very low concentrations (Sharafi et al., 2022; Soleimani et al., 2020). These PTEs can bioaccumulate for decades and years in the organs of the human body such as the liver, brain, kidneys, and bones. It can cause several health disorders like cancer (skin, lung, and liver), hyperpigmentation, thyroid problems, infertility, diabetes, heart disorders, gastrointestinal issues, etc. Hence, these PTEs are regularly monitored in groundwater by public agencies. Apart from that, the disastrous effects caused by those elements on human health are systematically studied by a method called Human Health Risk Assessment (HHRA), which was introduced by the United States Environmental Protection Agency (USEPA, 1980). This technique can assess the possibility of exposure to those PTEs when their concentration crosses the permissible limit prescribed by the World Health Organization (WHO, 2017) and the Bureau of Indian Standards (BIS, 2012) in the case of India. It accounts for the actual concentration range of each PTE in the groundwater of a given area in time and space (Mohammadpour et al., 2022). So, it can aid us in evaluating the risk associated with respect to the exposure of an individual to contaminated water both by oral intake and dermal contact and developing sustainable risk prevention measures.

According to the reports of the Central Ground Water Board (CGWB 2022), Punjab is one of the states in India that has over-exploited groundwater resources mainly for agricultural activities. The Punjab's economy is completely based on agriculture. It contributes paddy, wheat, and cotton to the people of India and other countries (Baweja et al., 2017; Gupta, 2009). The rotational pattern of cultivation has led to a decline in water levels of 33, 50, and 36 cm/year in the northeast zone, central part, and southwest part of Punjab, respectively (Baweja et al., 2017). Apart from the geology of the Punjab (granites and sediments derived from Himalaya), the usage of agrochemicals, including plant nutrients and fertilizers, and coal fly ash from thermal power stations had led to the release of high amounts of PTEs in groundwater (Virk, 2019a). Punjab tops the list of states where groundwater is highly polluted by PTEs, followed by West Bengal and Assam (Virk, 2019a; CGWB 2022). Several studies have also reported the contamination of groundwater in districts of Punjab by arsenic (As) (Hundal et al., 2007; Sharma et al., 2018; Kumar & Singh, 2020; Krishan et al., 2021, 2023; Kumar et al., 2020); uranium (U) (CGWB 2014; Sharma & Singh, 2016; Virk, 2016, 2017, 2018, 2019b; Narang et al., 2018; Archana and Singh 2021; Sahoo et al., 2022); iron (Fe); and nitrate (NO3-) (Krishan et al., 2015; Singh et al., 2022). These studies reported that PTEs in groundwater are sourced from primary and secondary minerals bearing U, As, and Fe, coupled with a high evaporation rate, semi-arid and semi-humid climatic conditions, higher Total Dissolved Solids (TDS), redox conditions, and the depth of the aquifer. In addition to that, groundwater pollution in Punjab is triggered by intense pumping of groundwater, canal irrigation, and the usage of nitrate and phosphate-based fertilizers.

The studies related to negative health impacts caused by PTEs for the Majha Belt of Punjab are negligible, rather non-existent. Hence, it would be appropriate to evaluate the probability of health effects on locals using the health risk assessment technique. The objectives of the present study are: i) to determine the concentration of PTEs, viz., As, U, Fe, and Nitrate, in the groundwater of the Majha Belt, Punjab; ii) to assess the synergistic effect of those contaminants in groundwater quality; iii) to evaluate health risks associated with the exposure of these contaminants via groundwater using the Human Health Risk Assessment tool proposed by USEPA (2014). Our investigations provide a comprehensive picture of arsenic and other PTE exposure and associated health hazards for the entire population of the Majha area of Punjab. Additionally, from a geographical point of view, the present study will aid in making appropriate decisions for the prevention and mitigation of diseases caused by those contaminants.

Study area

Location

Punjab is a north-western state of India located between the latitudes 29° 32′ and 32° 28′ N and the longitudes 73° 50′ and 77° 00′ E. It covers a total geographical area of 50,362 km^2, with an average elevation of 200 m above mean sea level. The Majha Belt extends from the northern to the north-eastern part of Punjab. It includes four districts of Punjab, namely, Tarn Taran, Amritsar, Gurdaspur, and Pathankot (Fig. 1).

Geography

The entire state is geographically classified into three major regions, namely Malwa, Majha, and Doaba. Among them, Malwa is the largest, followed by Majha and Doaba. Approximately 86% of the study area is predominantly used for agriculture (Krishan et al., 2021; Sahoo et al., 2022). This state contributes 40.38% of agricultural products to satisfy the needs of the nation. Wheat, paddy, and cotton are the main crops cultivated in this region. The Majha Belt is surrounded by Jammu and Kashmir in the north, the Doaba belt in the southeast, and the Malwa belt in the south and south-west directions. In the northwest and western directions, the study area is bound by Pakistan. These districts, except Gurdaspur, were divided into three geographic zones: alluvial plain, flood plain, and sand bars. The major portion constitutes the alluvial plains, which are deposited by the river Ravi and its tributaries (Sharma et al., 2019). Gurdaspur district is divided into three regions, i.e., hilly area, piedmont zone, and alluvial plain. Pathankot district is located at the foothills of the Siwaliks.

Drainage basin

There are three major perennial rivers originating from the north-western Himalayas that drain the study area. They are the rivers Sutlej, Beas, and Ravi (CGWB 2021). The rivers Beas and Ravi flow from the north-eastern to the south-western parts of the study area. Whereas the river Sutlej flows from eastern to western and from western to south-western parts of the study

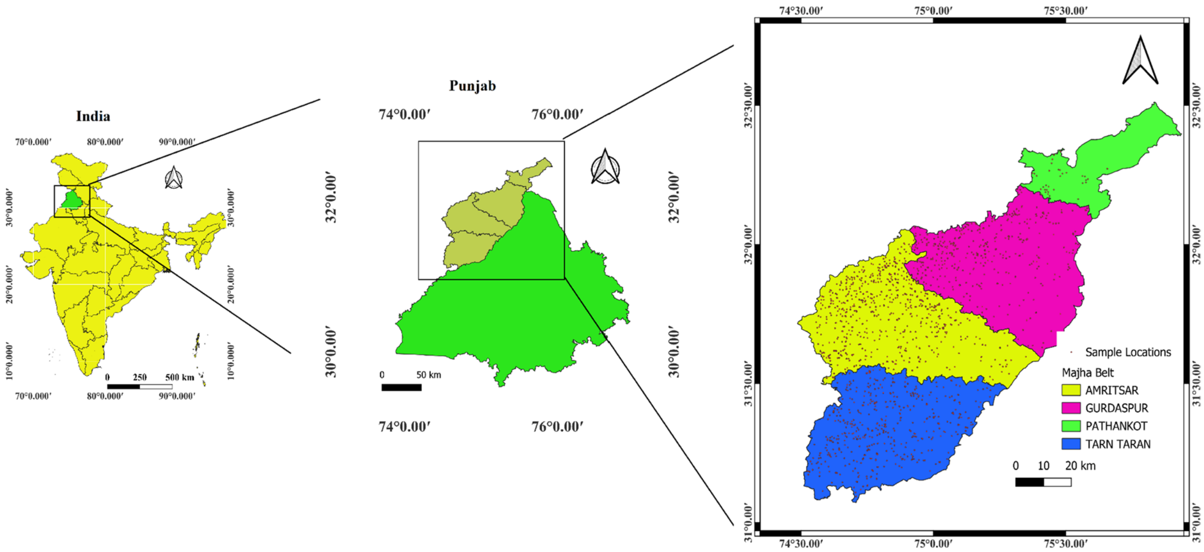

Fig. 1 Study area map with sample locations

area. The study area lies between the rivers Ravi and Beas.

Geology of the basin

Geologically, Punjab lies within the Indo-Gangetic plains, covered by alluvial sediments derived from the Himalayan mountains. The rock formation of the study area ranges in age from the middle Miocene to the Recent denoting Siwalik and alluvial deposits (Fig. 2). The thickness of unconsolidated sediments is higher near the foothills of the Himalaya and decreases towards the extreme south and south-west parts of the study area (CGWB). The study area belonging to Siwaliks is composed of clay and boulders. Piedmonts of the study area were derived from Siwaliks, which consist of pebbles and cobbles in addition to sand of medium- to coarse-grained gravel. The alluvial plains of the study area are intercalated by clays deposited by dry rivers Ravi and Beas. The lithological description of the study area is explained briefly in Table 1 (GSI 2011).

Groundwater system

Older Alluvium and Upper Siwaliks are the potential aquifers, followed by Newer Alluvium in the study area (Fig. 2). The water is held within those aquifers under unconfined (shallow) and semi-confined (deeper) conditions (CGWB ; Krishan et al., 2021). The thickness of sediments (granular and sandy in nature) in the north-western and northern parts of the study area is high (10–450 m), where high-yielding aquifers are present (Sahoo et al., 2022). It is the zone where the recharge of the aquifer takes place. In the south-western areas, the thickness of sediments is less, due to which aquifers are thinner (CGWB 2021). Hence, the groundwater flows and becomes saline in the northeast to southwest part of the state. Due to a lack of groundwater resources in the south-western part of the state (viz., Bathinda, Muktsar, Fazilka, and Ferozepur), water needs for agriculture are met by canal irrigation. Due to canal irrigation, there is seepage of water, leading to waterlogging and soil salinity (Krishan et al., 2021).

Climate

Punjab experiences semi-humid to arid climatic conditions. The north and northeast parts of the state are proximal to the Himalayan ranges, i.e., the Majha Belt experiences semi-humid to semi-arid climatic conditions. In the south and southwest, the climate is arid in nature due to the warm wind from the Thar

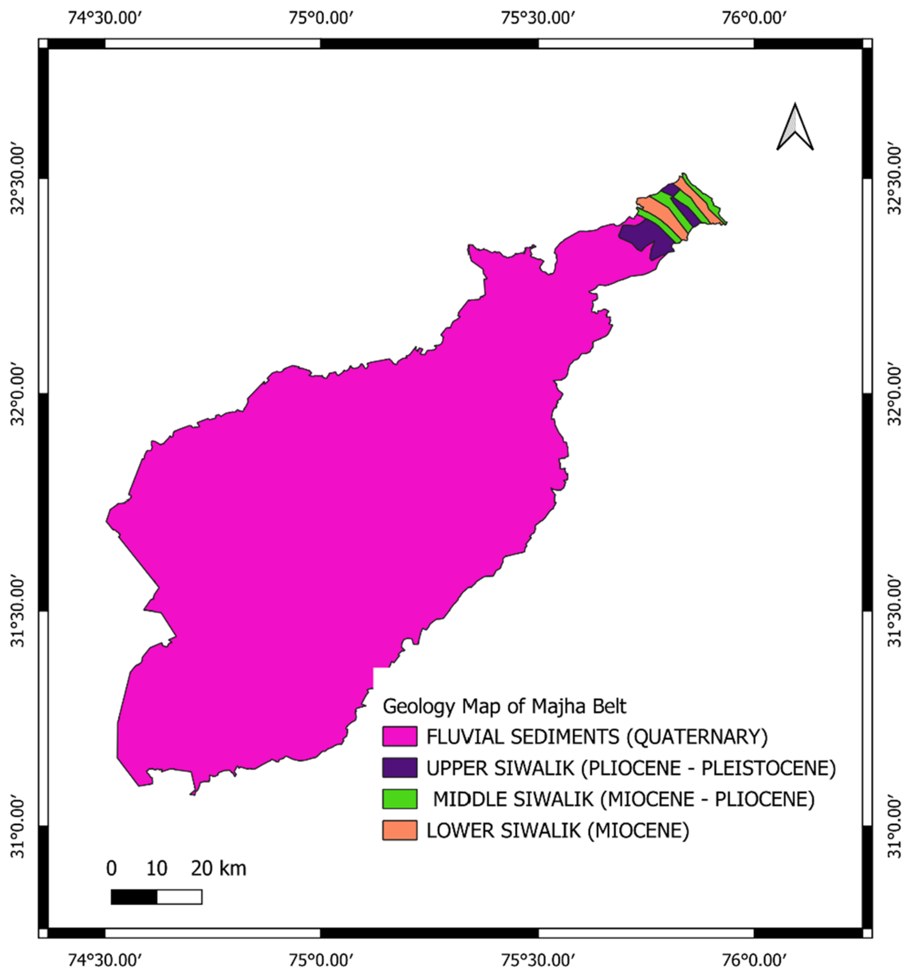

Fig. 2 Geology map of Majha Belt, Punjab, India

Desert. The climate of the remaining part of the state is semi-arid. The study area experiences very high temperatures (45 °C) during the summer (May and June) and low temperatures (3–9 °C) in the winter (December and January). The highest rainfall is received during the southwest monsoon (June to September). The mean annual rainfall received by the study area is 648 mm. The northeastern part of the study area, closer to the Himalayan range, receives an average rainfall of 1020 mm.

Methodology

Sample collection and data generation

The data for As, U, Fe, and Nitrate concentrations in groundwater samples (N = 2709) of the study area were obtained from the Department of Water Supply and Sanitation (DWSS), Government of Punjab compiled in April 2021(DWSS, 2021). In the bulk database, only the samples with data on the well depth and concentration of As, U, Fe, and NO^{-}_{3} were sorted out. As per the report, water samples were collected, filtered, and pre-treated with nitric acid (to pH < 2) to analyze As, U, and Fe using Agilent 7700 Series Inductively Coupled Plasma Mass Spectrometer (ICP MS) whose detection limit for heavy metals ranges from < 100 ppb to < 1ppt (DWSS, 2021). The calibration of the instrument was done using certified reference material (CRM) 2A (Agilent Technologies—Part Number 8500–6940). The correlation coefficient of the linear calibration curve (R^2) is ≥ 0.999. For every ten samples, 20 and 50 ppb standards were analyzed to ensure the reliability of data. The relative standard deviation (RSD) was found to be < 4%.

Table 1 Stratigraphic succession of Punjab

Period	Formation	Lithological description
Recent	Aeolian Deposits	Medium to fine grained and buff colored rounded sand composed of quartz, flakes of mica and ferromagnesian minerals in the form of undifferentiated aeolian flats/dunes/sand sheet and newer dunes
Upper Pleistocene to Recent	Newer Alluvium	Blue to white–grey micaceous sand with alluvium interbands of purple and red clay
Middle to Late Pleistocene	Older Alluvium	Reddish clay, silt, sand with kankar, grey medium to coarse calcareous sand with kankar and sub-rounded to sub-angular unsorted pebble, gravel and cobble beds
Late Pliocene to Early Pleistocene	Upper Siwaliks	Soft grey medium to coarse-grained poorly lithified sandstones with mica flakes and concretions of clay, yellowish brown and brown clays. Boulders, cobbles and pebbles of quartzite, granite, limestone, sandstone and lumps of claystones are also present; conglomerates occur as wedge shaped or as lenticular bands
Mio-Pliocene	Middle Siwaliks	Grey micaceous, medium grained soft sandstones interbedded with red, orange and yellowish (buff colored) clays. Sandstones occasionally contain pebbles of calcareous clay, shale and quartzite
Miocene	Lower Siwaliks	Massive grey to light grey, micaceous sandstones interbedded with dark red to maroon clays grading upward in to micaceous sandstone with thick beds of red clays

The samples collected for determining NO^{-}_{3} concentration were filtered and analyzed using the Ion Chromatographic System (ICS) which was calibrated using multiple standards of concentration of 0.1, 1, 5, 10 and 20 ppm prepared from bulk standard solution 1000 ppm of National Institute of Standards and Technology (NIST). The sample was analyzed three times and the mean value of it was considered as the sample value. The standard solution of 5 ppm was analyzed once for every ten samples and the average value obtained was 5.22 ± 0.26. The overall accuracy and precision of the standards analyzed were $< 5\%$ and $\leq 3\%$ respectively. The process used for collection, processing, and analyzing the samples has been reported by Virk (2022) is a standard procedure adopted in the laboratories of the DWSS, based in SAS Nagar (Mohai), Punjab, and accredited by the National Board for Accreditation of Testing and Calibration Laboratories (NABL). The maps essential for the study, such as geology and spatial distribution were prepared using ArcGIS (10.3).

Trace element evaluation index (TEI)

Trace elements are the elements that exist in groundwater at very low concentrations. An increase in the concentration of such elements may affect biodiversity to a greater extent. The water quality of a location with respect to trace elements can be evaluated using the following equation proposed by Edet and Offiong (2002):

$$\mathrm{TEI} = \sum_{i=n}^{n} \left(\frac{T_c}{T_{mac}} \right) \tag{1}$$

where Tc is the concentration of individual trace elements measured in each location, and T_{mac} is the maximum admissible concentration for ith parameter suggested by WHO. The maximum admissible or permissible concentrations of As, Fe, and U are 0.05, 0.3, and 0.03 mg/L, respectively (WHO, 2017). The calculated TEI values can be classified into 3 categories, namely, low (< 10), medium (10–20), and highly (> 20) polluted.

Single and comprehensive factor pollution index

Each PTE poses its toxic effects and is designated as a single-factor pollution index method (PI). It is calculated by the following equation (Luo et al., 2021):

$$\mathrm{PI} = \frac{\mathrm{Ci}}{\mathrm{Qi}} \tag{2}$$

where Ci and Qi are the measured and standard values for each element, respectively. Based on the values of PI, the quality of water is classified as follows: Class 1: very pure (PI < 0.30); Class 2: pure (PI = 0.31–1.0); Class 3: slightly affected (1.1–2.0); Class 4: moderately affected (PI = 2.1–3.0); Class 5: highly affected (4.1–6.0); and Class 6: tremendously affected (PI > 6). (Mitra et al., 2010). The combined effect of two or more toxic elements that are present in groundwater would be higher than the impact caused by individual toxic elements. It can be called the comprehensive pollution index (WQI). The comprehensive pollution index can be evaluated by the following equation (Luo et al., 2021):

$$WQI = \frac{1}{n}\sum_{1}^{n} PI \tag{3}$$

The values obtained from the above expression can be classified as not polluted (if WQI < 1); slightly polluted (if WQI = 1–2), moderately polluted (if WQI = 2–3), and heavily polluted (if WQI = ≥ 3).

Human health risk assessment (HHRA)

The human health risk associated with the contaminant can be estimated only by tracing its exposure route to the human body (Qasemi et al., 2023). Oral intake and dermal contact are the major pathways responsible for the entry of PTEs into the human body. The oral intake can be either direct consumption of contaminated water or the agricultural products cultivated by using polluted water. However oral intake is more important than others due to its large-scale detrimental health effects. The probability of health issues caused by such contaminated water is evaluated using a technique called Human Health Risk Assessment (HHRA), developed by USEPA in 1989. The concentrations of PTEs are used to evaluate chronic daily intake (CDI) (mg/kg/day) via the drinking of contaminated water.

$$\mathrm{CDI} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{EDXEF}}{\mathrm{ABW} \times \mathrm{AET}} \tag{4}$$

$$\mathrm{DAD} = \frac{\mathrm{C} \times \mathrm{Ki} \times \mathrm{EV} \times \mathrm{ED} \times \mathrm{EF} \times \mathrm{SSA} \times \mathrm{CF}}{\mathrm{ABW} \times \mathrm{AET}} \tag{5}$$

where CDI is the chronic daily intake (mg/kg/day), DAD is the dermally absorbed dose (mg/kg/day), C represents contaminant concentration (μg/L), and all the other parameters in the expressions (4, 5) are explained in detail in Tables 2 and 3. Followed by it, the non-carcinogenic risk of PTEs exposure in children and adults was determined using hazard quotient (HQ).

$$\mathrm{HQ\ Ingestion} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{6}$$

$$\mathrm{HQ\ Dermal} = \frac{DAD}{RfD} \tag{7}$$

Table 2 List of parameters and their values used in common for calculating health risks associated with toxic elements in drinking water with respect to age (USEPA, 2020a and b)

Sr. no.	Parameters	Adult	Child	Unit
1	Average Body weight (ABW)	70	15	kg
2	Exposure duration (ED)	26	6	years
3	Exposure frequency (EF)	350	350	days/year
4	Exposure time (ET)	0.71	0.54	hr/events
5	Resident events (EV)	1	1	per day
6	Skin surface area (SA)	0.71	0.54	cm^2
7	Resident water intake ratio (IRW)	2.5	0.78	L/day
8	Resident averaging time (AT) (non-carcinogenic)	365 × ED		days
9	Resident water ingestion rate (IRW)	Age adjusted		L/kg
10	Resident water dermal contact-factor (DFW)			Cm^2-event/kg
11	Resident water exposure time (ET)			hr/event
12	Life-time (LT)	70	70	years
13	Averaging Exposure Time (AET) (carcinogenic)	365 × LT		days

Table 3 Permissible limit potentially toxic elements and parameters used for health risk assessment (WHO, 2017) and (USEPA, 2020a and b)

Parameter	As	Fe	U	NO^-_3
Permissible Limit (WHO) mg/L	0.05	–	0.03	50
Permissible Limit (BIS) mg/L	0.05	0.3	0.03	45
Dermal permeability constant (Kp) cm/hr	0.001	0.001	0.001	0.001
Gastrointestinal absorption GIABS no unit	1	1	Not defined	1
Oral reference dose (RfD) mg/kg/day	0.0003	0.7	0.00453	1.6
Slope Factor (CSF) mg/kg/day	1.5	Not defined	Not defined	Not defined
Risk Coefficient (r) (Bq^{-1})	–	–	1.13×10^{-9}	–

In the equation, RfD is the oral reference dose. The value of HQ < 1 implies that the contaminant does not cause any non-carcinogenic health effects (USEPA, 1989 and 2014). On the other hand, PTEs can cause possible non-carcinogenic health effects on the exposed population when HQ is > 1 (USEPA, 1989 and 2014). The sum of the hazard quotient of ingestion and dermal contact is designated as the total hazard index (HI) (USEPA, 1989 and 2014). The total hazard index is calculated by using Eq. (8).

$$HI = HQ_{Ingestion} + HQ_{Dermal} \tag{8}$$

The HI values are classified into four classes namely, negligible (HI < 0.1), low significant health effect (0.1 < HI < 1), medium significant health effect (1 < HI < 4), and very high risk (HI > 4) (USEPA, 1989 and 2014; Qasemi et al., 2020).

Some of the PTEs, such as As and U, are likely to cause a risk of cancer in human beings. It is evaluated by an index called carcinogenic risk (CR). It is evaluated using the following equation:

$$CR = CDI \times SF \tag{9}$$

where SF is a cancer slope factor. Slope factor (SF) is applied to estimate the extent of carcinogenic risk due to exposure to contaminants. The SF always exhibits a linear relationship between contaminant concentrations, exposure, and the risk of cancer (Mohammadpour et al., 2022). According to USEPA (2014), the standard range for the CR is 10^{-6} and 10^{-4}. The value of CR below 10^{-6} does not pose any cancerous effect on human beings (Nkpaa et al., 2015). The CR value > 10^{-4} implies that the contaminant has the potential to cause cancer in an individual consuming contaminated water (Jiang et al., 2021).

Uranium, being a radioactive pollutant in groundwater, can cause a radiological cancer risk during the daily intake of radionuclides. The carcinogenic risk caused by the ingestion of U can be evaluated using the following equation proposed by Sharma et al. (2019).

$$ECR = U_a \times R \tag{10}$$

where U_a is uranium activity in water (Bq L^{-1}) and R is Risk Factor (L Bq $^{-1}$) which is calculated using Eq. (11):

$$R = r \times IR \times ED \tag{11}$$

where r is uranium risk coefficient, IR is ingestion rate and ED is exposure duration (Tables 2 and 3).

Results and discussion

The measured concentration of As and other PTEs in the groundwater of the Majha Belt of Punjab, India, has been displayed in the Table 4. The analytical results showed that the mean concentration of As in the Majha Belt was 0.03 ppm, with maximum and minimum values of 18.7 ppm and BDL (below the detection limit), respectively. The mean, maximum, and minimum concentrations of Fe in the study area are 0.31 ppm, 10.15 ppm, and BDL, respectively. For U, the mean, maximum, and minimum concentrations are 0.007 ppm, 0.30 ppm, and BDL. For nitrate, the mean, maximum, and minimum concentrations are 9.20 ppm, 314 ppm, and BDL.

Table 4 Analytical results of As and other PTEs (Fe, U and Nitrate) in groundwater of the study area

S.no.	Study area	As (ppb)			Fe (ppm)			U (ppb)			NO^-_3 (ppm)		
		Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
1	Entire Majha Belt	187	BDL	31	10.15	BDL	0.31	303	BDL	7.73	314	BDL	9.20
2	Tarn Taran	100	BDL	23	4.47	BDL	0.28	303	BDL	15.60	182	BDL	8.18
3	Amritsar	187	BDL	37	10.15	BDL	0.36	59.93	BDL	5.95	314	BDL	11.53
4	Gurudaspur	168	BDL	31	2.30	BDL	0.32	15.57	BDL	2.33	122	BDL	4.52
5	Pathankot	12	BDL	11	0.71	BDL	0.12	5.50	BDL	1.01	78	BDL	13.54

Spatial analysis of PTEs

Arsenic

In the study area, 16% (N = 334) of the locations have As concentrations above the maximum permissible limit of 0.05 ppm. Among them, most of the locations belong to the district of Amritsar. Pathankot is the only district that does not have As concentrations above the maximum permissible limit in the Majha Belt. Among the four districts of the Majha Belt, Amritsar is found to have the highest concentration of As (mean-0.03 ppm, Max-0.18 ppm, and Min-BDL), followed by Gurdaspur (mean-0.03 ppm, Max-0.16 ppm, and Min-BDL), and Tarn Taran (mean-0.02 ppm, Max-0.10 ppm, and Min-BDL) (Table 4). The spatial distribution map of As shows that the central part of the study area has As concentrations greater than the permissible limit (Fig. 3a). Studies on As pollution in groundwater have gained a lot of importance due to its carcinogenic risk to human health and its high stability characteristics (Murtaza et al., 2020). As far as India is concerned, studies on arsenic pollution are higher in number in the Ganga–Brahmaputra basin (Ghosh & Singh, 2009). Though arsenic pollution in groundwater was first reported in Chandigarh, the capital city of Punjab (Datta, 1976), only a few studies have been conducted in the Indus River basin, especially in the Majha Belt of Punjab. The arsenic contamination of groundwater can be of natural or manmade origin.

In the study area, excess arsenic in groundwater is due to anthropogenic activities, namely, agricultural activities. For agricultural purposes, due to a shortage of surface water resources and the failure of monsoons, farmers rely on groundwater. To satisfy food needs, farmers aim for higher production and yield by applying chemical fertilizers. The use of excessive pesticides in the form of calcium arsenate, lead arsenate, sodium arsenate, and arsenic acid on crops may leach down from the soil and reach shallow aquifers (Rasool et al., 2018). Apart from that, due to a lack of rainfall, farmers rely on groundwater from deeper aquifers. These deeper aquifers usually experience reducing conditions favorable for the mobility of toxic metals and metalloids such as As and Fe. And they are sourced from As- and Fe-bearing primary and secondary minerals present in the aquifer matrix of the region. It is one of the major causes of geogenic As in the groundwater of the study area. These minerals, when subjected to reductive (iron-oxyhydroxide) or oxidative (pyrite) dissolution, lead to the release of As into groundwater (McArthur et al., 2001; Nickson et al., 2005). In the study area, groundwater sampled from tube wells of deeper depth has an As concentration > 0.01 ppm, and only very few samples collected from hand pumps have a high As concentration. The majority of samples collected from hand pumps have As concentrations < 0.01 ppm. Groundwater from deeper depths exists under reducing conditions favouring the release of As by the reductive dissolution process. Hand pumps delivering water with higher As may be due to agricultural activities or the competitive exchange of As with nitrate, phosphate, or bicarbonate (Nickson et al., 2005). A similar scenario was observed in agricultural lands and urban areas of Rupnagar district of Punjab, India lying in Malwa region where the As concentration is maximum (i.e. 0.91 ppm) in shallow wells (Krishan et al., 2023).

Iron

The Fe concentration in the groundwater of the Majha Belt is in the range of BDL to 10.15 ppm. In

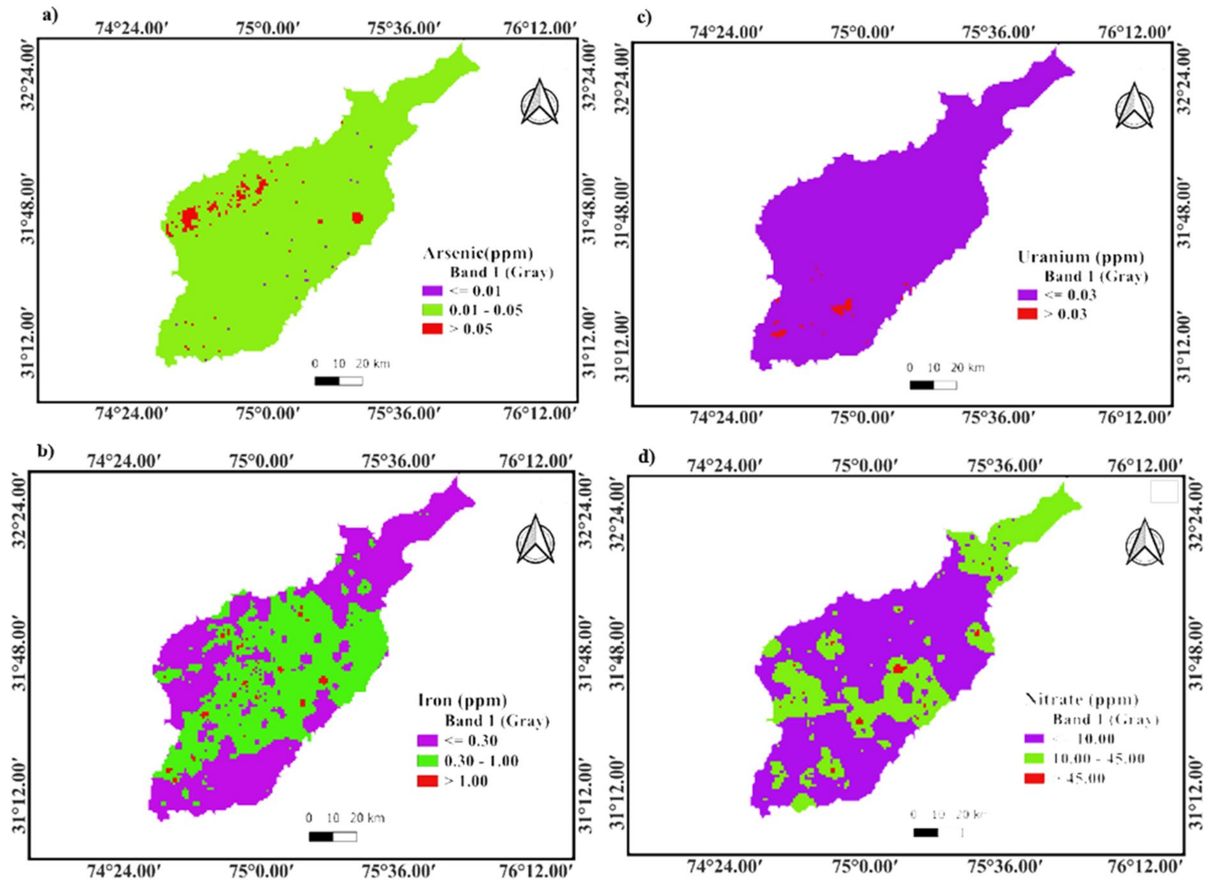

Fig. 3 Spatial distribution maps of **a** Arsenic, **b** Iron, **c** Uranium and **d** Nitrate

all, 12.4% of the samples in the study site were found to have higher Fe than the WHO standard (0.3 ppm), with the maximum number of samples crossing the standard limit in Amritsar, followed by Gurdaspur, Tarn Taran, and Pathankot (Fig. 3b and Table 4). The distribution of Fe and As in the groundwater of the Majha Belt follows a similar pattern. The concentration of Fe in groundwater can be attributed to rainfall, intensity of recharge of groundwater, geology, water depth, groundwater flow pattern, residence time, redox condition, etc. (Hossain et al., 2015; Islam & Mostafa, 2023). As the study does not have data on rainfall, water flow patterns, or recharge rates, inferences on that basis cannot be drawn.

Geologically, the study area is found to have an aquifer matrix intercalated with several clay layers and clay concretions. These fine-grained sediments are usually known for their organic matter, which can impose a reducing environment in the aquifer, leading to the mobility of Fe from sediments and subsequent enrichment in the groundwater. Apart from that, the study area is also known for river flow and intensive agricultural practices. The river flow carries a huge amount of debris (sediment and plants), which can impart reducing condition of the aquifer. Similarly, during irrigation, huge amounts of water are flooded over the land, which is responsible for the waterlogged condition of the soil. It can inhibit the entry of oxygen into shallow aquifers, too. The enrichment of arsenic can also happen when Fe-bearing minerals (pyrite, siderite, limonite, etc.) undergo weathering in the absence of dissolved oxygen and by microbial activities (Marion 2008).

Uranium

The uranium concentration in the districts of the Majha Belt, namely, Tarn Taran, Amritsar, Gurdaspur, and Pathankot districts, is found to vary from BDL to 0.30 ppm, BDL to 0.06 ppm, BDL to 0.02 ppm, and BDL to 0.005 ppm, with mean values of 0.02, 0.005, 0.002, and 0.001 ppm, respectively (Table 4). The mean value of uranium content in the entire Majha Belt is 0.007 ppm, with the highest mean value in the Tarn Taran district (the southernmost part of the study area), followed by Amritsar, Gurdaspur, and Pathankot (Fig. 3c). Such a drastic variation in U concentration in all four districts is due to the different sources and depths of groundwater occurrence. It has been observed that only 3.3% and 0.01% of samples of the Tarn Taran and Amritsar districts have uranium content higher than the WHO limit of 0.03 ppm in pre-monsoon. Similarly, the U concentration in the present study of Amritsar and Gurdaspur was higher than the values reported earlier in the study area by other researchers (Tarn Taran: 0.16 ppm (Sahoo et al., 2022); Amritsar: 0.19 ppm (Sahoo et al., 2022); 0.06 ppm (Sharma et al., 2019) and 0.04 ppm (Rani et al., 2013); and Gurdaspur: 0.032 ppm (Sahoo et al., 2022) and 0.04 ppm (Sharma et al., 2019); and Pathankot: 0.04 ppm (Sahoo et al., 2022).

Sahoo et al. (2022) also reported that in Punjab as a whole, the groundwater exists mostly under oxic and alkaline conditions. Apart from that, the weathering of granitic rocks from the Himalayan region and an aquifer matrix composed of sandy fragments contribute to U. All the above-said conditions prevailing in the study area reduce the adsorption capacity of U onto the aquifer sediments, favoring the release of U in groundwater. In the case of the Majha Belt, the higher concentration of U in Tarn Taran district is mostly from tube wells whose maximum depth is approximately 150–350 m. These deeper confined aquifers, with their recharge zones at the Himalayan and Siwalik hills and granitic and calcareous rocks, can contribute U into groundwater (Archana and Singh, 2021). Apart from that, the usage of phosphate fertilizers with trace amounts of U and Th for more than a decade can also contribute U to groundwater (Schnug & Lottermoser, 2013).

Nitrate

Nitrate in the groundwater of the Majha Belt varies from BDL to 182 ppm, BDL to 314 ppm, BDL to 122 ppm, and BDL to 78 ppm, with mean values of 8.18, 11.53, 4.52, and 13.54 ppm in Tarn Taran, Amritsar, Gurdaspur, and Pathankot districts, respectively (Table 4). Overall, in the Majha Belt, the mean value of nitrate is 9.20 ppm. 6.7% of samples in the Majha Belt (comprising 0.03% of Tarn Taran, 0.02% of Amritsar, 2.7% of Gurdaspur, and 0.09% of Pathankot) have nitrate concentrations higher than the WHO (50 ppm) and BIS permissible limits (45 ppm) (Fig. 3d). Karanveer et al. (2022) has reported maximum nitrate concentration of 259 ppm in shallow and 112 ppm in deeper groundwater of alluvial plains in Punjab. Excessive and unmonitored usage of NPK fertilizers such as urea, diammonium phosphate (DAP), mono ammonium phosphate (MAP), and nitro phosphate combined with potash is found to be the major contributor of nitrate in groundwater of alluvial plains of Punjab, India (Karanveer et al., 2022).

The spatial distribution map of nitrate depicts that the southern portion of the study area has nitrate higher than the permissible limit. Nitrate in groundwater is mainly attributed to anthropogenic activities such as nitrogen-based fertilizers in agricultural land, landfill leachates, and disposal of sewage and livestock wastes (Ramalingam et al., 2022; Sunitha et al., 2022). In addition to the above activities, geological factors (Feng et al., 2022) like porosity, permeability, water depth, etc. also contribute to nitrate enhancement. The major aquifers of the study area are Older Alluvium, Upper Siwaliks, and Younger Alluvium, composed of sand, poorly sorted gravel, pebbles, cobbles, and conglomerates with larger values of porosity. It aids in transporting leached nitrogen-based contaminants, such as nitrate, from surface to subsurface.

Trace element evaluation index (TEI)

Trace elements in groundwater can be of natural geochemical and/or anthropogenic origin. They have a peculiar characteristic of bioaccumulation, which thereby imparts toxicity to the life forms. The maximum, minimum, and mean values of TEI in districts of the Majha Belt, Punjab, India, are reported in Table 5. The maximum value of TEI (48.70) is found to occur in the village of Bagrian in Amritsar district. Among the four districts comprising the Majha Belt, Amritsar district has higher values of TEI, indicating intensive pollution of groundwater by trace elements, followed by Tarn Taran, Gurdaspur, and Pathankot. Higher values of TEI are associated with higher loadings of arsenic, iron and nitrate in Amritsar and uranium in the Tarn Taran district. It is also noted that groundwater pumped from shallow depths (< 100 m) using hand pumps has high TEI values. It suggests that unconfined aquifers comprising sandy and gravelly beds of Older Alluvium and Upper Siwaliks are highly contaminated by potentially toxic trace elements in the Majha Belt.

Single-factor (PI) and comprehensive pollution index (WQI)

The maximum, minimum, and mean values of PI for all the PTEs considered for the study area have been mentioned in Table 6. Based on the ranking of PI, all four districts of the Majha Belt are found to be slightly to moderately affected by As, U, and Fe. Among the PTEs used in the present study, Fe has the highest PI value, followed by U, As, and nitrate. Whereas, based on the mean values of PI, the order of PTEs in the study area varies in the following order: Tarn Taran: U > As > Fe > NO_3^-; Amritsar: Fe > As > NO_3^- > U; Gurdaspur: As > NO_3^- > Fe > U; and in Pathankot: Fe > NO_3^- > U > As.

WQIs values range from a minimum of 0.14 to a maximum of 12.23, with mean value of 0.15 in the Majha Belt (Table 5). Among the 2709 groundwater samples collected from the study area, 94.4% are unpolluted, 3.3% are slightly polluted, 0.8% are moderately polluted, and 1.4% of the samples are heavily polluted. From the observations, it can be inferred that the selected PTEs in the study area have a synergistic effect on the quality of the groundwater. Among 1.4% (N = 37) of heavily polluted samples in Majha Belt, 27 samples represent Amritsar district.

Health risk assessment of PTEs present in water samples

The harmful health risks associated with the consumption of PTEs via drinking water on human health are assessed using various indices, viz., Hazard Quotient (HQ), Total Hazard Index (HI), and Carcinogenic Risk Index (CR). In the present study, the adverse effects of As, Fe, U, and NO^-_3 via ingestion and dermal contact were evaluated. The health risk assessment was done on two groups of population, namely, adults (20–60 years) and children (5–10 years), and is depicted in Fig. 4a, b, c, and d. The results depict that the HQ value of Fe, U, and

Table 5 Trace element evaluation index (TEI) and Comprehensive pollution index (WQI) values of As and other PTEs (Fe, U and Nitrate) in groundwater of the study area

Sr. no.	Study area	Trace element evaluation index (TEI)			Comprehensive pollution index (WQI)		
		Max	Min	Mean	Max	Min	Mean
1	Entire Majha Belt	48.70	0	0.52	12.23	0	0.14
2	Tarn Taran	29.67	0	1.24	7.49	0	0.33
3	Amritsar	48.70	0	1.94	12.22	12.22	12.22
4	Gurudaspur	17.80	0	1.12	4.51	0	0.31
5	Pathankot	2.58	0.01	0.41	1.05	0.01	0.16

Table 6 Single-factor pollution index (PI) of As and other PTEs (Fe, U and Nitrate) in groundwater of the study area

Sr.no.	Study area	As			Fe			U			NO^-_3		
		Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
1	Entire Majha Belt	2.00	BDL	0.41	29.67	BDL	0.31	10.1	BDL	0.42	6.07	BDL	0.58
2	Tarn Taran	3.34	BDL	0.63	48.67	BDL	0.01	1.99	BDL	0.09	10.47	BDL	0.15
3	Amritsar	3.74	BDL	0.44	17.80	BDL	11.11	1.07	BDL	0.15	7.05	BDL	0.38
4	Gurudaspur	0.24	BDL	0.01	2.37	BDL	0.22	0.18	BDL	0.01	3.67	BDL	0.05
5	Pathankot	2.00	BDL	0.41	29.67	BDL	0.19	10.10	BDL	0.42	6.07	BDL	0.46

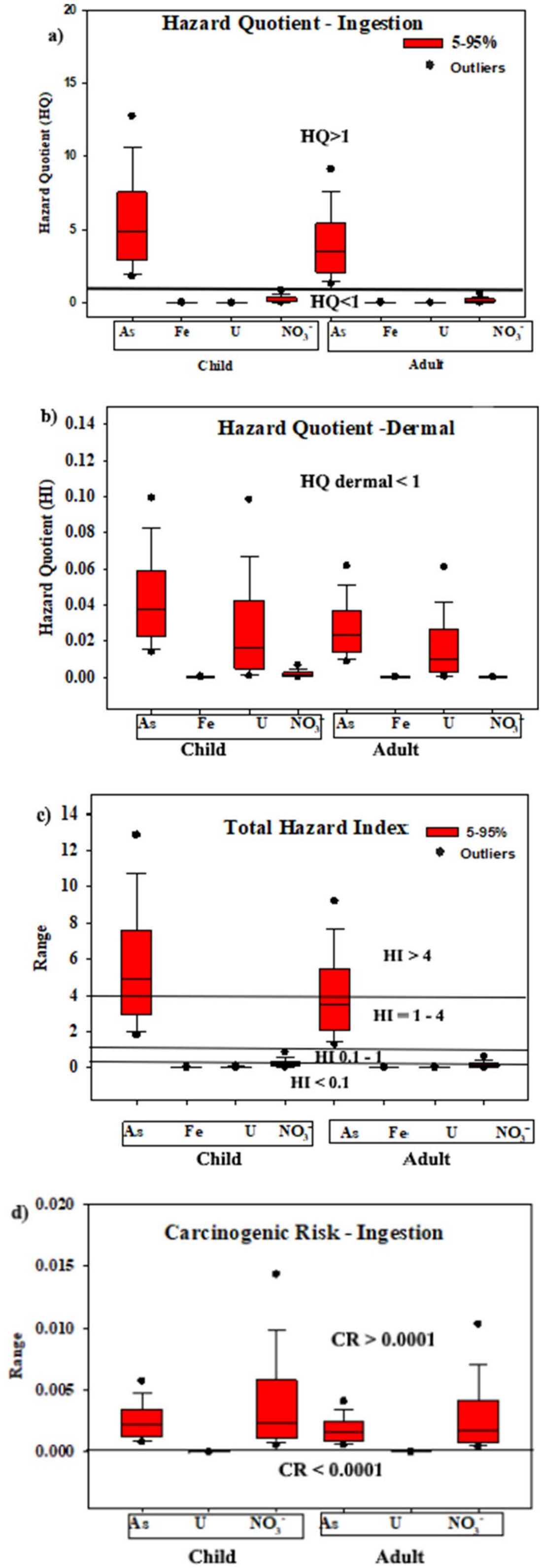

Fig. 4 Box plot of **a** hazard quotient-ingestion, **b** hazard ▸ quotient-dermal, **c** total hazard index and **d** carcinogenic risk-ingestion

nitrate falls below 1 for both exposure routes for adults and children (Table 7 and Fig. 4a, b, and c). It indicates that there won't be any adverse health implications due to the measured PTEs for both the population groups in the study area. However, arsenic (As) was found to show an HQ value > 1 with respect to the ingestion pathway, irrespective of age, in almost all the samples (Fig. 4a, b, and c). A high HQ value can impose severe non-carcinogenic risk for adults and children via oral exposure. This clearly indicates that the probability of health hazards due to As pollution in the Majha Belt districts is high, which may lead to dermal effects, cardiovascular effects, respiratory effects, gastrointestinal effects, endocrinological effects (diabetes mellitus), neurological effects, reproductive and developmental effects, cancer effects, and other health effects (Ghosh & Singh, 2009; Mart´ınez-Castillo et al., 2021). Krishan et al. (2023) came to the same conclusion for As in the Rupnagar district of Punjab, India. As polluted water not only has health impacts but also affects the yield of plants (Sridharan & Nathan, 2018). When As-rich groundwater is used for irrigation, As can accumulate in edible parts of the plants. It, in turn, enters the food chain and can intensify the disease. Less body weight and skin surface area in children makes them more susceptible to arsenic toxicity (Eslami et al., 2022).

Hazard index

The present study evaluated hazard index values due to intake of As, Fe, U, and nitrate via ingestion and dermal contact paths for the two population groups (adults and children). Based on USEPA (1989), 44.10% of samples in the Majha Belt have a hazard index > 4, indicating very high health risks due to oral intake and dermal contact in both age groups of people (Table 8 and Fig. 4c). Among all the districts of the Majha Belt, Amritsar is found to be largely affected by As poisoning. About 65% and 51% of samples in Amritsar are in threatening condition for children and adults, respectively. It implies that drinking groundwater and using it for household work and recreational purposes are not safe with

Table 7 Hazard quotient mean values of As and other PTEs (Fe, U and Nitrate) in groundwater of the study area

Sr.No.	Study area	As				Fe				U				NO^-_3			
		HQ-C-I	HQ-C-D	HQ-A-I	HQ-A-D	HQ-C-I	HQ-C-D	HQ-A-I	HQ-A-D	HQ-C-I	HQ-C-D	HQ-A-I	HQ-A-D	HQ-C-I	HQ-C-D	HQ-A-I	HQ-A-D
1	Entire Majha Belt	5.64	4.04	0.04	0.02	2.42E-02	1.73E-02	1.88E-04	1.17E-04	4.16E-03	2.98E-03	3.24E-02	2.01E-02	3.09E-01	2.22E-01	2.41E-03	5.04E-05
2	Tarn Taran	4.17	2.94	0.03	0.02	2.20E-02	1.58E-02	1.71E-04	1.06E-04	8.40E-03	6.02E-03	6.53E-02	4.05E-02	2.74E-01	1.97E-01	2.14E-03	4.46E-05
3	Amritsar	6.57	4.71	0.05	0.03	2.75E-02	1.97E-02	2.14E-04	1.33E-04	3.22E-03	2.30E-03	2.50E-02	1.55E-02	3.88E-01	2.79E-01	3.03E-03	6.32E-05
4	Guru-daspur	5.61	4.02	0.04	0.02	2.50E-02	1.79E-02	1.94E-04	1.20E-04	1.26E-03	9.01E-04	9.78E-03	6.07E-03	1.52E-01	1.09E-01	1.19E-03	2.48E-05
5	Pathankot	1.89	1.36	0.01	0.01	9.50E-03	6.81E-03	7.39E-05	4.58E-05	5.50E-04	3.94E-04	4.28E-03	2.65E-03	4.56E-01	3.27E-01	3.55E-03	7.42E-05

* *HQ-C-I*: Hazard quotient-child-ingestion and *HQ-A-D*: Hazard index-adult-dermal

respect to arsenic concentration. The HI-Fe in the entire Majha Belt is less than 1, indicating negligible health risks (for both adults and children). The HI-U suggests that 15.5% of locations in the Majha Belt, especially Tarn Taran district, fall under the medium-significant health effect (HI values lie between 1 and 4). In the case of nitrate, 69.20% of samples in the study area have an HI value between 0.1 and 1, indicating a low significant health effect.

Carcinogenic health risk

Among the four PTEs chosen for the study, As, U and nitrate-rich groundwater can cause cancerous effects in human beings. Hence, an estimation of the carcinogenic health risk index can predict the possibility of cancer for the entire adult and child population of Majha Belt. Figure 4d shows the estimated CR values for adults and children. The CR mean value of arsenic (As) for child and adult (via ingestion and dermal pathway) is 2.54E-03 and 1.82E-03, respectively, while for U, the CR values for child and adult were 8.68E-07 and 9.45E-06, respectively (Table 9 and Fig. 4d). For nitrate, estimated CR values are 5.15E-03 and 3.70E-03 for children and adults, respectively. The accepted TCR values for PTEs in groundwater range between 10^{-4} and 10^{-6} as per USEPA (2014). The TCR values of As and Nitrate in the entire Majha Belt were greater than the USEPA standard value, indicating the chances of carcinogenic risk for both children and adults. Such a higher TCR value for As was reported by Proshad et al. (2020) for all the age groups in the Rupsha river basin of Bangladesh. The radiological risk assessment for adults and children due to uranium intake was performed by calculating the carcinogenic risk. The mean value of it lies within the permissible limit of 1.67×10^{-4} (AERB, 2004). Based on the maximum uranium concentration and CR value observed in the groundwater of the Majha Belt, Tarn Taran district is found to have a high cancerous risk, especially in adults. With continuous exposure to uranium through drinking water, there is a higher probability of the occurrence of cancer cases in Tarn Taran district.

Table 8 Hazard index mean values of As and other PTEs (Fe, U and Nitrate) in groundwater of the study area

Sr.no.	Study area	As		Fe		U		NO^-_3	
		HI-C	HI-A	HI-C	HI-A	HI-C	HI-A	HI-C	HI-A
1	Entire Majha Belt	5.69E+00	4.07E+00	0.024	0.017	3.65E-02	2.30E-02	3.12E-01	2.22E-01
2	Tarn Taran	4.21E+00	3.01E+00	0.022	0.016	7.37E-02	4.65E-02	2.76E-01	1.97E-01
3	Amritsar	6.63E+00	4.74E+00	0.028	0.020	2.82E-02	1.78E-02	3.91E-01	2.79E-01
4	Gurudaspur	5.66E+00	4.05E+00	0.025	0.018	1.10E-02	6.97E-03	1.53E-01	1.09E-01
5	Pathankot	1.91E+00	1.37E+00	0.010	0.007	4.83E-03	3.05E-03	4.59E-01	3.27E-01

**HI-C*: Hazard index child; *HI-A*: Hazard index adult

Table 9 Carcinogenic risk assessment mean values of As and other PTEs (U and Nitrate) in groundwater of the study area

S.no.	Study area	As		U		NO^-_3	
		CR-C	CR-A	CR-C	CR-A	CR-C	CR-A
1	Entire Majha Belt	2.54E-03	1.82E-03	8.68E-07	9.45E-06	5.15E-03	3.70E-03
2	Tarn Taran	1.88E-03	1.35E-03	1.75E-06	1.91E-05	4.56E-03	3.28E-03
3	Amritsar	2.96E-03	2.12E-03	6.71E-07	7.30E-06	6.45E-03	4.64E-03
4	Gurudaspur	2.53E-03	1.81E-03	2.62E-07	2.86E-06	2.53E-03	1.82E-03
5	Pathankot	8.54E-04	6.12E-04	1.15E-07	1.25E-06	7.58E-03	5.45E-03

**CR-C*: Carcinogenic risk child and *CR-A*: Carcinogenic risk adult

Conclusion

In this study, the concentration and spatial distribution of PTEs (As, Fe, U, and Nitrate) and the non-carcinogenic and carcinogenic health risk assessment of As, U, and Nitrate in the groundwater resources of the Majha Belt, which includes Tarn Taran, Amritsar, Gurdaspur, and Pathankot districts of Punjab, India, were studied. Based on the results, it was found that the mean concentrations of As, Fe, U, and Nitrate are 0.03 ppm 0.31 ppm, 0.007 ppm and 9.20 ppm, respectively. The maximum concentrations of As (0.18 ppm), Fe (10.15 ppm), and NO^-_3 (314 ppm) have been observed in Amritsar district, while the maximum concentration of U (0.30 ppm) is found in Tarn Taran district. Altogether, the Amritsar district of the Majha Belt is found to be contaminated by higher concentrations of As, followed by Fe and NO_3^-. At the same time, Tarn Taran district is highly polluted by U in groundwater. The synergistic effect of toxic elements was evaluated by the WQI index, which suggests that Amritsar district of the Majha Belt is severely affected by toxic constituents in groundwater. The non-carcinogenic risk of HQ for As in groundwater was greater than 1 for both adults and children. Hence, people in the Majha Belt are more susceptible to As health hazards via groundwater consumption. Whereas, with respect to other PTEs chosen for study areas, HQ is less than 1. It suggests that non-carcinogenic health effects due to those elements are negligible. The HI-As (combined effect of oral and dermal exposure to As) for 44.10% of samples in Majha Belt is > 4 indicating a very high risk for both adults and children. The CR assessment reports that the As and nitrate values are greater than the USEPA standard value (10^{-6} to 10^{-4}), indicating a greater probability of carcinogenic risk in the study area. Finally, it is concluded that exposure to PTEs, especially As and Nitrate, through groundwater via drinking can pose significant health hazards for people belonging to the Amritsar district of the Majha Belt due to their highly toxic and carcinogenic characteristics. Though the Tarn Taran district has a high U concentration, health risk assessment shows that the chances for carcinogenic risks are quite low.

Limitations and future scope of the study

To mitigate As ingestion and external exposure, the groundwater in the study area has to be treated with appropriate techniques, such as a combination of oxidation, precipitation, and adsorption methods. Along with As, proper care should be taken for other co-contaminants also. Moreover, the source and mechanism of the release of As and its relationship with other PTEs should also be evaluated in detail with respect to geology, water depth, climate (seasonal sample collection), major ion chemistry of groundwater, etc. which is found to be the major limitation of the present study.

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Author contributions Hardev Singh Virk is responsible for collection and curation of data used in this paper and its final revision. M. Sridharan focused on writing this paper and analysis of the data using statistical and modelling techniques. D. Senthil Nathan helped in conceptualization. All authors have worked as a team in planning its execution.

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Declarations

Conflict of interest The authors declare no competing interests.

Ethical approval Not required.

Consent to participate All the authors give their consent to participate equally in this endeavour.

Consent for publication All of the authors give their consent to publish this work.

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