Arsenic contamination in groundwater of the Majha belt of Punjab and its probable carcinogenic and non-carcinogenic health hazards

Hardev Singh Virk*

Sri Guru Granth Sahib World University, Fatehgarh Sahib 140 406, India

Arsenic is widespread in groundwater in India. High levels of arsenic in the three districts of Amritsar, Gurdaspur and Tarn Taran (also called Majha belt) have caused a crisis in Punjab. According to the Indian Council of Agriculture Research report, 13 districts of Punjab have arsenic content beyond the safety limit. This study aims to estimate probable health hazards due to ingestion of water with high levels of arsenic in the groundwater of Majha belt. Analysis of groundwater samples done using inductively coupled plasma mass spectrometry were collected from the Department of Water Supply and Sanitation, Government of Punjab, Mohali. The highest arsenic contamination of 111 ppb was found in the groundwater of Amritsar district, followed by Gurdaspur and Tarn Taran districts. The average value of hazard quotient (HQ) for children and adults in Amritsar district is estimated to be 11.13 and 8.0 respectively. HQ values for all the 650 habitations surveyed in the Majha belt of Punjab are greater than 1, which is a matter of concern because of high-risk potential for developing adverse carcinogenic and non-carcinogenic health hazards. The predicted values for cancer induction in children and adults of Amritsar district are 500 and 360 per million respectively. Mitigation of arsenic in groundwater is an urgent need in the Majha belt of Punjab.

Keywords: Arsenic, cancer risk, groundwater, health hazards, mitigation.

ARSENIC occurrence and distribution in groundwater of different states of India (Figure 1) have been of wide interest since the 1980s (refs 1–3). We have previously studied groundwater contamination caused by uranium and other heavy metals[4–8]. Health risk analysis due to fluoride and arsenic in the groundwater of Patiala and Roopnagar districts respectively, has been reported recently[9,10]. This study focuses on arsenic (As) contamination of groundwater and its probable carcinogenic and non-carcinogenic health hazards in the three districts of Majha belt of Punjab, India (Figure 2).

An extensive study of arsenic contamination in groundwater of Indian and Pakistani Punjab was undertaken by the scientists of both countries in a collaborative research project funded by Columbia University, New York, USA[11]. However, there is no epidemiological study on health hazards of arsenic in the Majha belt of Punjab. To date, the source of arsenic contamination in Punjab groundwater is uncertain. We[5,7] along with other researchers[12–14] have reported high levels of arsenic beyond the permissible limit fixed by WHO (10 ppb) in groundwater of Punjab.

Arsenic is widely distributed as a metalloid in the environment (soil, water, air and rocks). It possesses characteristics of both a metal and a non-metal. Inorganic arsenic is the most prevalent in nature, and its high toxicity is the cause of major health hazards. It can react with oxygen or other molecules in the air, water or soil. It forms various

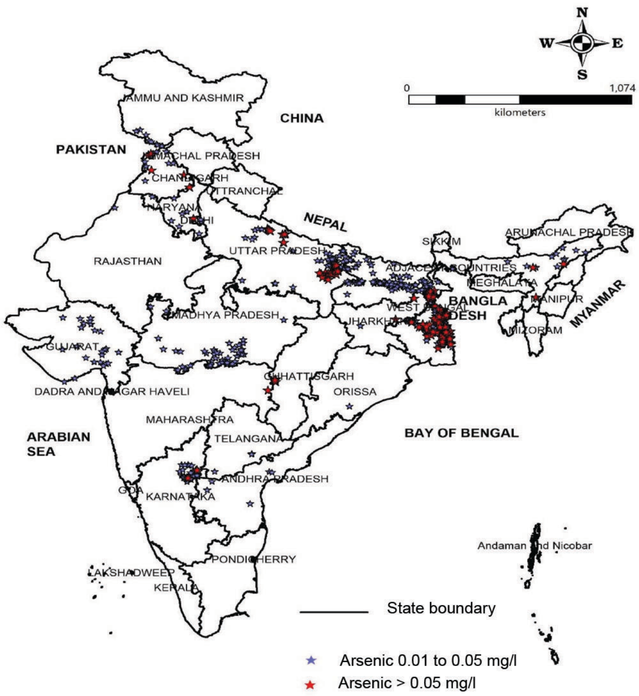

Figure 1. Map showing locations of arsenic contaminated groundwater in different states of India[1].

*e-mail: hardevsingh.virk@gmail.com

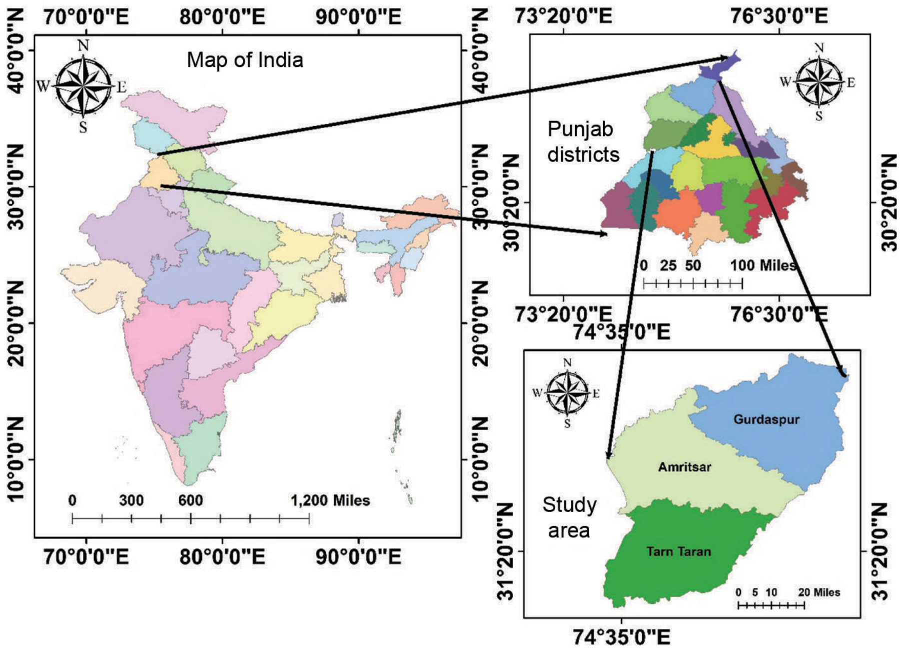

Figure 2. Map showing districts of Amritsar, Gurdaspur and Tarn Taran in the Majha belt of Punjab, India.

compounds[15] by reaction with oxygen in the environment; trivalent arsenite and pentavalent arsenate are most predominant among these.

In several countries of the world, viz. Argentina, Australia, Bangladesh, Cambodia, Chile, China, Hungary, India, Lao People's Democratic Republic, Mexico, Myanmar, Nepal, Pakistan, Peru, Thailand, United States and Vietnam[16], arsenic contamination higher than the WHO limit (30 ppb) has been detected in the groundwater. A limit of 10 ppb (1 ppb = 1 μg/l)[17] was fixed in 2016 by the US Environmental Protection Agency (EPA) and the WHO for arsenic in drinking water. Exposure to arsenic in drinking water over long periods poses a great risk to human health[18]. Arsenic caused mass poisoning of a population in Bangladesh due to groundwater contamination with naturally occurring inorganic As[19] in groundwater. An estimated 160 million people are living in regions where arsenic levels in drinking water[20] are known to be higher than WHO limit. The physical characteristics of arsenic make it difficult to be observed in environment, especially when ingested through drinking water.

Reviews of carcinogenic and non-carcinogenic health effects of arsenic have been published earlier[20–22]. Arsenic ingestion has been associated with several types of cancers, including skin and several other organs, including bladder, kidney, liver, prostate and lung[23–27]. In addition to several types of cancers, arsenic-related diseases include several other diseases and neuropathies[28–31]. A daily consumption of 1.6 litres of water with inorganic arsenic contamination of 50 μg/l has been estimated to cause cancer with a probability of 21/1000 (ref. 32).

Evidence of carcinogenicity of arsenic in humans has been confirmed. The International Agency for Research on Cancer (IARC) has classified arsenic as the number one human carcinogen. Animal models fail to replicate the observed effects of arsenic-related carcinogenicity[33], hampering the progress in the field of cancer research.

Materials and methods

Geographical location

Punjab is situated in the northern part of India between lat. 29.30°N to 32.32°N and long. 73.55°E to 76.50°E. Malwa Majha, and Doaba constitute three geographical entities in Punjab, with natural boundaries created by rivers Sutlej and Beas. These rivers are the primary freshwater sources in Punjab. Himachal Pradesh and Haryana states are on the north and east respectively, and Rajasthan in the south of Punjab. Pakistan forms an international boundary on the western side.

The groundwater conditions of recharge are influenced by soil characteristics of the region. Surface contaminants affect the quality of water at a higher rate if recharge rate is higher. The groundwater level in various districts of Punjab is falling rapidly due to the overexploitation and is recorded beyond 60 mbgl (metre below ground level)[34].

Geomorphology and soil types of Majha belt

The geographical location, geomorphology, and nature of soil of three districts of Amritsar, Gurdaspur and Tarn Taran of Majha belt of Punjab have been described by us in a recent study[5]. The major portion of the Majha belt constitutes the Indo-Gangetic alluvial plains, which are deposited by the rivers Ravi and Beas and their tributaries. Soils in the Majha belt vary from coarse loamy to calcareous soils in the western part, but in the central part, they are fine loamy, calcareous and well drained. Gurdaspur district can be divided into three geomorphological regions, i.e. hilly area, piedmont zone and alluvial plain. Approximately 86% of the study area is predominantly used for intensive agriculture[35].

Sample collection and analysis

Standard protocol was used for sample collection and analysis as described by the author[36]. Arsenic data used for our investigation is derived from Annual Water Quality Report of DWSS[37] for 2021–22. The process used for collection, processing, and analysing the samples in the laboratories of the DWSS, based in SAS Nagar (Mohali), Punjab, has been accredited by the National Board for Accreditation of Testing and Calibration Laboratories (NABL). The accreditation is renewed every two years. The calibration of the Agilent 7700 series inductively coupled plasma mass spectrometry used for sample analysis was done using certified reference material provided by the National Institute of Standards and Technology (NIST), USA. The correlation coefficient of the linear calibration curve (R^2) is ≥0.999. The relative standard deviation (RSD) was found to be <4%. We have been using the DWSS facility for heavy metal analysis since 2016 and have published more than two dozen papers.

Carcinogenic and non-carcinogenic health hazards of arsenic

Arsenic contamination in groundwater has added significance in the Majha belt of Punjab due to its probable health hazards. Arsenic investigation in groundwater and canal waters in Majha belt of Punjab was undertaken by Punjab Agriculture University (PAU) scientists[12] before anyone else. The Indian Council of Agriculture Research (ICAR) reported high arsenic beyond WHO limit (30 ppb) in groundwater of 13 districts of Punjab[38]. There is an urgent need to undertake epidemiological investigations to study the health hazards of arsenic in groundwater on the population of Punjab.

Arsenic dissolved in water is toxic and can lead to several health problems. Tseng Multistage Model[39] based on an epidemiological study has been used to estimate the risk of cancer from arsenic in drinking water. Long-term exposure to arsenic in drinking water, beyond 0.01 mg/l, causes increased risks of cancer of skin, lungs, bladder and kidney[21].

There are two routes, respiratory exposure and gastrointestinal exposure[40], open for Arsenic to cause cancer. The International Agency for Research on Cancer (IARC)[41] officially recognized arsenic as a carcinogenic agent during the 1980s. Further studies[42] were conducted in the United States, Taiwan, Bangladesh, India, Argentina and Chile to examine the association of arsenic with carcinogenicity.

Repeated epidemiological investigations have confirmed that arsenic induces numerous diseases, which include dermal, cardiovascular, respiratory, gastrointestinal, endocrinological (diabetes mellitus), neurological, reproductive and developmental, cancer and other effects[22]. American Cancer Society[43] has also enumerated long- and short-term exposure health effects of arsenic on human health. Bangladesh residents exposed to drinking water with arsenic content of 10 ppb or less suffered from melanosis and keratosis. This study[44] proved that 36 out of 167 residents exposed (13.9%) were affected by arsenic. In another study by Lee *et al.*[45], it was reported that arsenic may be the cause of some cardiovascular diseases as it affects thrombocytes.

Arsenic health-risk assessment

Risk assessment[46] is a probability of occurrence of any given magnitude of adverse health effects over a specified time-period. It is a function of two variables, the hazard caused and the exposure time. Health risks consist of two types: carcinogenic and non-carcinogenic.

The two principal toxicity risk factors are the slope factor (SF) and the reference dose (RefD). They are used for calculating carcinogenic and non-carcinogen risks[47,48] respectively. Average daily dose (ADD in mg/kg/day) due to ingestion of the arsenic-rich water is a useful parameter used to calculate the health-risk assessment. ADD is calculated using the formula[49,50] given below

$$\text{ADD} = C \times \text{IR} \times \text{ED} \times \text{EF/BW} \times \text{AT},$$

where C is the arsenic concentration (mg/l = ppm) in groundwater, IR is the average daily intake rate which is assumed to be 2 l/day and 3.45 l/day for children and adults respectively[51]; ED, exposure duration which is assumed to be 10 years for children, and 70 years for adults; EF, exposure frequency which is 365 days/year; BW is the average body weight which is assumed to be 25 kg and 60 kg for children and adults respectively; AT, average time which is 25,550 days, i.e. (70 × 365) days for adults, and 3650 days, i.e. (10 × 365) days for children[49,51].

Hazard quotient (HQ) is a parameter used for calculating the non-carcinogenic risk for the consumers of arsenic-rich groundwater in the study area. It is defined as the ratio of ADD and RefD, where RefD for arsenic is 3×10^{-4} mg/kg/day[52].

There are two limits of hazard quotient: HQ > 1 indicates potential health risk to the consumers of arsenic-rich water due to non-carcinogenic type hazard, whereas HQ < 1 is considered safe for consumers of drinking water[47].

The carcinogenic risk or cancer risk (CR) for the consumers of arsenic-rich groundwater in the study area is

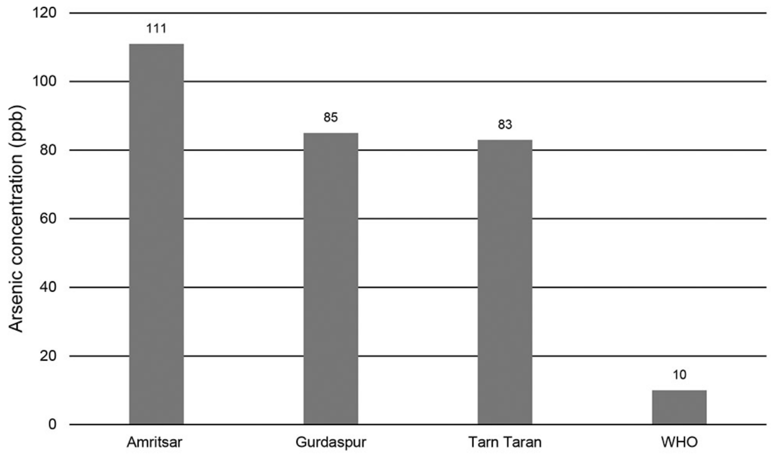

Figure 3. Arsenic concentration in groundwater of Majha belt vis-á-vis WHO value.

Table 1. The values of average daily dose (ADD), hazard quotient (HQ) and cancer risk (CR) of arsenic in groundwater of Majha belt districts of Punjab

	Amritsar			Gurdaspur			Tarn-Taran		
Parameter	Minimum	Maximum	Average	Minimum	Maximum	Average	Minimum	Maximum	Average
ADD children (mg/kg/day)	9.00E-04	8.90E-03	3.30E-03	8.80E-04	6.80E-03	2.60E-03	9.60E-04	6.60E-03	2.50E-03
ADD adult (mg/kg/day)	6.00E-04	6.40E-03	2.40E-03	6.30E-04	4.90E-03	1.90E-03	7.00E-04	4.80E-03	1.80E-03
HQ children	2.93	29.6	11.13	2.93	22.67	8.67	3.2	22.13	8.21
HQ adult	2.11	21.28	8	2.11	16.29	6.23	2.3	15.91	5.9
CR children	1.32E-03	1.33E-02	5.00E-03	1.30E-03	1.02E-02	3.90E-03	1.44E-03	9.96E-03	3.69E-03
CR adult	9.50E-04	9.57E-03	3.60E-03	9.50E-04	7.30E-03	2.80E-03	1.04E-03	7.16E-03	2.65E-03

calculated as the product of ADD and SF where the value of SF for oral carcinogenic arsenic is 1.5 mg/kg/day[49].

Results and discussion

Health risk assessment due to groundwater arsenic contamination has been reported in case of children and adults by some workers[10,53]. The purpose of our study is to evaluate both carcinogenic and non-carcinogenic health hazards for the population of Majha belt of Punjab. The arsenic content of groundwater in the three districts of Majha belt, namely, Amritsar, Gurdaspur and Tarn Taran are recorded in Supplementary Tables 1–3. Amritsar district has 325 habitats with groundwater arsenic contamination varying from 11 to 111 ppb, Gurdaspur has 218 habitats with arsenic content variation from 11 to 85 ppb, and Tarn Taran has 107 habitats with arsenic variation 12–83 ppb. All these habitats record arsenic higher than the safe limit of 10 ppb recommended by WHO. A histogram (Figure 3) represents the peak values of arsenic in these districts vis-á-vis the threshold recommended by WHO.

The average daily dose (ADD), hazard quotient (HQ), and cancer risk (CR) for the consumers of arsenic-rich groundwater for all the three districts of Majha belt are listed in Table 1. ADD depends upon the toxicity of heavy metals, and it is a function of magnitude, frequency and duration of human exposure to the potentially toxic metals in the environment[52]. Non-carcinogenic risk is measured in terms of HQ and carcinogenic or cancer risk in terms of the product of ADD and SF, as defined in the last section.

Depending upon variation of arsenic concentration, these parameters also vary from minimum to maximum. The average HQ for children and adult consumers in Amritsar district is 11.13 and 8.0 respectively. For Gurdaspur and Tarn Taran districts, HQ values are lower than in Amritsar district. However, the HQ values calculated for all the 650 habitations surveyed in the Majha belt are greater than 1, which shows that all of the consumers of the arsenic-rich groundwater in the study area are at high potential risk for developing non-carcinogenic adverse health effects. The health risk is more pronounced in children compared to adults. The average CR values for children and adults in Amritsar district are 5.00E-3 and 3.60E-3 respectively. It translates into 500 and 360 cancer cases per million for children and adults respectively. These values for other two districts are lower than in Amritsar district. Rapant and Krčmová[54] reported a high value of the cancer risk caused by arsenic in groundwater in Slovakia. It is estimated to be 10^{-4}, or 100 people per million population, which is lower than the risk calculated for the Majha belt. Carlson-Lynch *et al.*[55] have published their commentary on the approaches adopted for arsenic risk assessment challenging the CR criteria, which is based on a linear dose–response relationship for cancer even for low doses of arsenic.

What is the source of arsenic in the groundwaters of Majha belt? To eliminate health hazard effects of arsenic, a detailed investigation needs to be undertaken of groundwater resources of the Majha belt. High levels of arsenic are attributed to both geogenic and anthropogenic sources. Anthropogenic sources include intensive agricultural practices based on use of fertilizers, herbicides and weedicides[10]. It is revealed that heavy metal contamination is a significant problem in the water and soil[56] of agricultural communities because of excessive use of agrochemicals.

Mitigation of effects of arsenic in the Majha belt is on top priority of Punjab government. Report of mitigation measures adopted has been published by the author[7], which include a nanotechnology-based technique called AMRIT (arsenic and metal removal by Indian technology) and ion-exchange technology. Considering high costs of technologies being proposed for mitigation, DWSS[37] has opted to use surface water available from irrigation channels/ canals for drinking purposes as a cost-effective measure. Our investigation[57] of heavy metals content in surface water of Sirhind canal has confirmed that arsenic content is either below the detection limit (1 ppb) or much lower than the recommended limit of WHO (10 ppb). There is an urgent need for undertaking an epidemiological survey of the villages/habitations showing arsenic anomalies in groundwater being used for drinking.

Conclusion

The 650 habitations covered in this study of the groundwater contamination of three districts of Majha belt exhibit arsenic contamination beyond the safe limit of 10 ppb fixed by the World Health Organisation. HQ values for children and adults in all habitations are >1; the maximum values for Amritsar district are 29.6 and 21.6 respectively. Hence, both these population groups are exposed to non-carcinogenic health hazards.

CR factors are estimated for both adult and child population groups and predicted values are 500 and 360 per million respectively, which is extremely high compared with national and global values. Epidemiological survey is a must to determine health hazards due to arsenic in groundwater in the Majha belt. Mitigation measures must be prioritized to eliminate the risk of health hazards.

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