J. Bangladesh Acad. Sci. 49(2); 157-177: December 2025 DOI: https://doi.org/10.3329/jbas.v49i2.85456

Journal of Bangladesh Academy of Sciences

Journal homepage: http://www.bas.org.bd/publications/jbas.html

Review Article

Arsenic contamination of groundwater and its effects on drinking water, irrigation and public health in Bangladesh

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ARTICLE INFO

Article History

Received: 04 November 2025

Revised: 07 December 2025

Accepted: 09 December 2025

Keywords: Arsenic in drinking water, Arsenic in Agri-environment, Prevalence of arsenicosis, Dose-response relationship and Translocation of Arsenic.

ABSTRACT

Bangladesh is the worst affected country in the world by arsenic contamination of groundwater. The widespread installation of low-cost shallow tubewells, initially intended to control waterborne diseases, has exposed millions to high arsenic concentrations through drinking water. Arsenic (As) in groundwater in the absence of dissolved oxygen is predominantly present as As(III) which is more toxic than As(V). Food, particularly rice, represents another significant pathway of arsenic exposure. Approximately75% of dry-season irrigation relies on groundwater, arsenic accumulation in cropsespecially ricemay substantially contribute to the total body burden in affected areas. The cause, magnitude, and health impacts of arsenic contamination of groundwater have been investigated in many studies. This paper provides an updated synthesis based on available research. The paper also presents a comprehensive analysis of the cause and extent of contamination, andits impacts on drinking water, agro-environment and prevalence of arsenic-related diseases. Arsenic risk management measures implemented in Bangladesh are discussed. The analysis includes estimates of population exposed to varying arsenic concentrations, an assessment of the effects of contaminated irrigation water on soil, paddy plants, and rice grains, and an exploration of the correlation between average drinking water arsenic contents at the Upazila (sub-district) level and the prevalence of arsenic-induced skin lesions in that Upazila..

Introduction

Arsenic is a ubiquitous element, naturally present in the Earth's crust, and biosphere. It is the 20th most abundant element in the Earth’s crust and the 12th in the biosphere, and its environmental cycling is governed by both natural processes and anthropogenic activities (Ahmed, 2003). The Department of Public Health Engineering (DPHE), with international support, undertook a massive campaign in the late 1970s and early 1980s to control waterborne diseases such as cholera, typhoid, and dysentery by providing pathogen-free drinking water. Installation of shallow tube wells (STWs) to withdraw groundwater was promoted under this campaign, which is naturally filtered and free of microbial pathogens. The program successfully achieved its immediate goal. An estimated 10 million tube wells were installed in rural Bangladesh (Zahid, 2018), providing what was believed to be safe water to approximately 97% of the rural population, the highest coverage in the region (Ahmed, 2002). However, the potential for elevated arsenic concentrations in groundwater was not anticipated during this expansion of the drinking water supply.

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Subsequent detection of arsenic contamination in many shallow aquifers rendered shallow tube well water unsafe for drinking purposes. The problem is further exacerbated by considerable spatial variability in arsenic concentrations, which vary significantly even within small geographic areas. Bangladesh, situated in the Bengal Basin, became the most severely affected country in terms of population exposure to arsenic.

The problem began to unfold when arsenic contamination was first detected in 1983, but it took nearly a decade to be recognized as a "large-scale public health crisis" (Nordstrom, 2000). The World Health Organization (WHO) declared it the "largest mass poisoning of a population in history" in the late 1990s. This widespread arsenic exposure directly challenged the public health success story of the STW program (Caldwell et al., 2003). Within a few years, the arsenic crisis overshadowed the earlier achievements, thrusting the nation into a desperate nationwide mitigation effort, documented extensively in the national and international literature (Ahmed, 2002; Milon et al., 2012; Sakamoto, 2021).

The nationwide testing revealed that about one-fourth of all shallow tube wells in the country exceeded the national standard of 50 µg/L for arsenic. The test result indicated that the arsenic content of tube wells varied with depth and short horizontal distances. Bangladesh, in terms of population exposure, becomes the most severely affected country in the world.

Arsenic in groundwater is of geological origin, and it has been accepted that arsenic is being dissolute in water from sediment under certain conditions. Arsenic is found in the soils of Bangladesh in concentrations like those in many countries. The average concentration in soils in arsenic affected areas has been found to be approximately 100 times higher than the acceptable concentration in water. The transfer of arsenic from soil to groundwater and vice-versa is dependent on soil-water interaction in the subsoil environment. A clear understanding of origin and mechanism of dissolution of arsenic from soil to water is necessary for mitigation.

The importance of groundwater irrigation increased with the introduction of HYV seeds in the late 1960s to meet the food demand of a growing population (Zahid, 2018). Arsenic contaminated water in the shallow aquifer can be easily abstracted for irrigation by installing of low-cost shallow tube wells. As a result, most irrigation tube wells are shallow and contain higher arsenic concentrations in arsenic-affected areas. In the absence of surface water during the dry season, the future expansion of irrigation depends even more on groundwater. Arsenic withdrawn with groundwater can build up in soil and translocate into irrigated crops. Arsenic intake through food is equally important as arsenic ingestion through drinking water, except that a part of the arsenic intake through food is organic in nature. Arsenic ingestion through both food and water increases the body burden to cause arsenic-related diseases. Phytotoxicity from high concentrations of arsenic in soil and irrigation water, and its long-term impact on crop yield is another primary concern for food security in Bangladesh.

Mechanism of Arsenic Contamination of Groundwater

Many hypotheses have been initially proposed to explain the possible causes of arsenic contamination in Bangladesh. Still, most scientists have settled on oxidation and reduction hypotheses in the absence of adequate evidence for other hypotheses. The most important ores of arsenic are arsenic pyrites, realgar, and orpiment (Yan-Chu, 2004). Mok and Wai (1994) reported arsenic release from these minerals in groundwater by oxidation as shown in the following equations:

$$4\,FeAsS+13O_2+6H_2O \rightarrow 4FeSO_4+4H_3AsO_4 \qquad 1$$

$$As_2S_3 + 7\,O_2 + 6\,H_2O = 3H_2SO_4 + 2H_3AsO_4 \qquad 2$$

$$4AsS + 11O_2 + 10H_2O = 4H_2SO_4 + 4H_3AsO_4 \qquad 3$$

In this process, seasonal water-level fluctuations or water-table lowering due to large-scale groundwater withdrawal may expose the aquifer to aeration-induced oxidation. Soils may also be oxidized by infiltration of water saturated with dissolved oxygen.

The intensity of the arsenic problem has not been found to be related to groundwater fluctuations. Similarly, the hot spots in Bangladesh are not located in areas with high groundwater withdrawal for irrigation (Ahmed 2007a and 2007b). A very low sulfate concentration in groundwater is also inconsistent with the pyrite oxidation hypothesis (BGS, DPHE, and MML, 1999; Bhattacharya et al., 1999). The traces of arsenopyrite or arsenic sulfides found in sediments might have formed under conditions of enhanced reduction. Hence, the hypothesis of arsenic release from the oxidation of the top soil layer is not considered a primary mechanism of groundwater contamination in Bangladesh.

According to the reduction hypothesis, arsenic can be mobilized from soil in a reducing environment. In the reducing zone at low redox potential, insoluble ferric iron is partially reduced to soluble ferrous iron, and similarly, manganese is partially reduced to the soluble manganous state. Adsorbed arsenic on dissolution of the minerals is released into pore water. Thus, the reduction and subsequent dissolution of iron, manganese, and other minerals that contribute to the sorption and retention of arsenic can provide a mechanism for arsenic contamination of groundwater. The reduction process also converts precipitated and adsorbed As(V) into more soluble As(III), as shown in the following equation:

$$H_2AsO_4^- + 3\,H^+ + 2\,e^- \rightarrow H_3AsO_3 + H_2O \qquad 4$$

In reducing soil environments, arsenic will predominate in pore water, as As(III). Further reduction of As(III) in the presence of sulfides as reported by Mok and Wai (1994), will immobilize arsenic in soils with the formation of arsenic sulfide precipitates:

$$2\,H_3AsO_3 + 6\,H^+ + 3\,S^{2-} \rightarrow As_2S_3 + 6\,H_2O \qquad 5$$

$$2\,As_2S_3 + 4\,e^- \rightarrow 4\,AsS + 2\,S^{2-} \qquad 6$$

Arsenic from orpiment (As_2S_3) and realgar (AsS) can also be released in water by oxidation, as shown in Equations [2] and [3].

It is generally accepted that arsenic in groundwater is of natural origin and is believed to be released under conditions conducive to the dissolution of arsenic from the solid phase on soil grains into the liquid phase of groundwater. Arsenic occurs in soils at an average concentration of about 5 to 6 mg/l (Bhumbla and Keefer, 1994), but mean arsenic contents in soils as high as 20 ppm in Italy, 14 ppm in Mexico, 11.2 ppm in China, and 11 ppm in Japan have been reported (Yan-Chu, 2004). The average arsenic concentrations in alluvial sand and mud/clay have been reported as 2.9 mg/kg and 6.5 mg/kg, respectively, in Bangladesh (BGS and DPHE, 2001). The presence of high arsenic concentrations in groundwater is not generally dependent on soil arsenic levels. The geochemical and environmental conditions of the soil have a greater influence on arsenic speciation, solubility, and mobility.

The solubility of arsenic in water is usually controlled by redox conditions, pH, biological activity, and reductive dissolution reactions. As(V) is the major arsenic species under oxidative conditions at high Eh values and under reducing conditions at low Eh values, it converts arsenic into a more mobile As(III) form. Arsenic in soil is relatively stable at neutral pH, but exhibits mobility at both higher and lower pH values. Metal ions solubilize at lower pH values from the sediments with concurrent release of arsenic species. The increased hydroxide concentrations at high pH levels displace arsenic species from their binding sites (Mok and Wai, 1994). Desorption of arsenic can also be promoted in the presence of more competing anions, such as phosphate.

The most severely flooded areas are mainly arsenic-contaminated areas of Bangladesh (Ahmed 2000). Reducing the soil environment in most severely flooded areas appears to promote the release of arsenic into groundwater. In these areas, the soils are characterized by paludal deposits of clay, silt and peat and alluvial deposits of silt, and silty clay. The luxuriant vegetation in nutrient-rich floodplains enriches fine-grained soils with organic debris. The anaerobic condition in soils in deeply flooded wetlands is characterized by a gray to blackish color and the release of methane gas. The dissolved oxygen available in infiltrated floodwater is exhausted in the topsoil, which in rich in biodegradable organic matter. Pore water devoid of dissolved oxygen creates a reducing environment that favors the dissolution of both iron and arsenic (Ahmed 2000).

Rahman and Rahman (1998) collected sediment samples from different arsenic-prone areas, mainly lying in the lower Gangetic plain, and found arsenic-rich iron oxide coatings of varying thickness on sand grains. They concluded that adsorption of arsenic on iron oxide might have occurred during transportation of sand and arsenic-bearing mineral grains by flowing water in open channels. British Geological Survey (BGS), Department of Public Health Engineering (DPHE), Bangladesh, and Mott MacDonald Limited (MML), considered the reductive desorption and dissolution of arsenic adsorbed onto iron oxyhydroxides in recent sediments to be the most probable mechanism of arsenic mobilization in groundwater (BGS, DPHE, and MML, 1999). Moreover, mineralogical examination suggested that the small amount of pyrite present in the sediments had been precipitated since burial.

Bhattacharya et al. (1999) stated that during groundwater development, the flow of reducing groundwater through the aquifers resulted in the dissociation of ferric hydroxides, released the bulk of the arsenic due to the reductive dissolution of ferric oxyhydroxides and the arsenic previously adsorbed onto these minerals. The traces of arsenopyrite or arsenic sulfides found in sediments might have been formed under an enhanced reducing environment, as shown in Equations 5 and 6. Ravenscroft et al (2000) concluded that neither pyrite oxidation nor competitive exchange of fertilizer-phosphate for sorbed arsenic caused arsenic pollution of groundwater in the Bengal basin. Indeed, pyrite in Bangladesh's aquifers is a sink, not a source, of arsenic. Arsenic pollution occurs when FeOOH is microbially reduced, releasing the arsenic it sorbs to groundwater.

Reductive dissolution of iron oxyhydroxides by anaerobic microorganisms has been recognized as a key process governing arsenic mobilization under reducing environments (Ahmann et al., 1997; McCreadie et al., 2000). The arsenic present in these aquifers is thought to have originated from multiple source areas in the upper catchments of the Ganges, Brahmaputra, and Meghna rivers and to have been transported downstream, adsorbed onto colloidal iron oxyhydroxides (Ravenscroft et al., 2005).

In the Bengal Basin of Bangladesh and West Bengal (India), the primary mechanism of groundwater arsenic pollution is the reductive dissolution of iron oxyhydroxides (FeOOH), which releases sorbed arsenic into groundwater (Ahmed, 2007; Nickson et al., 1998; McArthur, 1999; Ravenscroft et al., 2001; Ravenscroft and Ahmed, 2005). Arsenic mobilization occurs under iron-reducing conditions in shallow aquifers (<35 m depth), that are predominantly Holocene in age, where microbial processes drive reduction reactions. The groundwater in these aquifers typically has high concentrations of both arsenic and iron. The sediments of these aquifers are characteristically dark in color, reflecting strongly reducing conditions. The microbial biodegradation of organic matter, mediated by anaerobic iron-reducing bacteria (FeRB) such as *Geobacter* species, facilitates the dissolution of FeOOH and the concomitant release of arsenic (Anawar et al., 2011).

In contrast, oxic to sub-oxic aquifers, which generally correspond to older Pleistocene deposits, produce groundwater with low arsenic concentrations (Ravenscroft and Ahmed, 2005).Arsenic remains mobile under sulfate-reducing conditions, suggesting that authigenic sulfide precipitation is not a significant sink for arsenic in these groundwaters (Zheng et al., 2004). The combined geochemical and microbial evidence supportsthat microbially mediated reductive dissolution of iron oxyhydroxides is the dominant process governing arsenic release in the Bengal Basin aquifers. Chowdhury et al. (2003) confirmed that arsenic was initially transported to the Bengal Basin with sediments from the Ganges, Brahmaputra, and Meghna (GBM) river systems and subsequently deposited in the basin.

A joint study by the Bangladesh University of Engineering and Technology (BUET), the Massachusetts Institute of Technology (MIT), and the University of Cincinnati (UC) exhibited that arsenic release in aquifers is triggered by the introduction of organic carbon, which serves as a food source for bacteria (Harvey et al, 2002). Evidence of this process includes the presence of methane in groundwater, indicating that anaerobic bacteria are metabolizing organic matter. The resulting anaerobic conditions create a reducing environment that favors arsenic mobilization. This anoxic state, characterized by the absence of dissolved oxygen and very low redox potential, has been confirmed through careful groundwater sampling and field measurement.

The BUET-MIT-UC study observed a positive correlation between arsenic and ammonia and a negative correlation between arsenic and sulfate. These observations are consistent with the mobilization of arsenic through the anoxic degradation of organic matter, rather than the oxidation of sulfide minerals (Harvey et al, 2002). They also indicated that irrigation pumping may facilitate the rapid transport of dissolved organic carbon in the aquifers, thereby triggering anaerobic biochemical reactions. The concept of microbial reductive dissolution of arsenicis further supported by the works of Akai et al. (2001). Their culture experiments using sediments from Bangladesh and Japanese lakes (Sagata and Matarese) showed arsenic elution following a rapid drop in Eh values. This drop was induced by increased bacterial activity after the addition of nutrients (glucose and polypeptone). These experimental results provide findings under reducing conditions created by microbial breakdown of organic matter in aquifers.

Analysis of hydrology, hydrogeology, and soil characteristics of Bangladesh, combined with the distribution and intensity of arsenic contamination, supports the following conceptual model for the origin, transport, deposition, and mobilization of arsenic in Bangladesh:

Origin of Arsenic –The origin of arsenic is arsenic-rich minerals in the upstream basins of the Ganges, Brahmaputra, and Meghna Rivers. Weathering and oxidative processes release arsenic into the river water. The high-energy induced turbulent flow in the upstream reaches saturates the water with oxygen, promoting the oxidation of both arsenic species and suspended sediments. Consequently, arsenic is adsorbed onto suspended particles that are rich in oxidized iron, aluminum, manganese, and other ions.

Transportation - In the middle reaches of the rivers, the arsenic-contaminated suspended sediments are transported downstream. The river velocity at this stage remains too high for significant sediment deposition, though occasional deposition of coarse sand may occur. Due to its relatively small surface area and fewer active sites per unit volume, this sand does not adsorb significant amounts of arsenic. Sediments transported by the Ganges-Brahmaputra-Meghna river system carry arsenic concentrations ranging from 1.002–2.983 mg/kg in sand, 1.858–3.943 mg/kg in silt, and 3.525–6.476 mg/kg in clay, leaving the river water almost free of dissolved arsenic (Chowdhury et al., 2003).

Deposition in Bangladesh –The slope of the rivers is flatter in the lower reaches of Bangladesh. Seasonal floods submerge one-third to one-half of the country each year, depending on their intensity. As water spreads into low-lying floodplains, the water becomes nearly stagnant. This allows arsenic-rich fine-grained silt and clay particles to settle out of suspension. These deposited fine-grained arsenic-rich sediments constitute the primary source of arsenic in Bangladesh.

Mobilization - The aquatic weeds and agricultural residues in nutrient-rich floodplains, mixed with or submerged in sediment, finally decompose an aerobically. A large quantity of organic matter submerged under floodplain sediments forms peat. Anaerobic decay of organic matter in submerged soils creates reducing and low redox conditions, mobilizing arsenic from sediments into groundwater.

Magnitude of the Arsenic Problem in Bangladesh

The World Health Organization Guideline Value (WHO GV) for non-threshold chemicals, such as toxic and carcinogenic substances like arsenic, represents the concentration corresponding to an upper-bound estimate of an excess lifetime cancer risk of 10^{-5}, i.e., one additional cancer case per 100,000 people. The WHO GV is based on 60 kg adult person, drinking 2 liters of water per day, for a lifetime of 70 years. The WHO provisional Guideline Value (WHO GV) of 0.01 mg/L for arsenic in drinking water has been adopted as the national standard in many countries. In comparison, many other countries, including Bangladesh, have retained the earlier WHO Guideline Value of 0.05 mg/L as the national standard or interim target, with the intention of lowering the arsenic standard for drinking water in the future (Ahmed, 2007c).

The disease burdens for arsenic exposure from drinking water, even complying with the WHO GV and Bangladesh Standard (BDS), are comparatively higher (Ahmed et al. 2006b) than the WHO reference disease burden of 1µ DALY per person per year (WHO, 2011). The joint FAO/WHO experts committee on food additives (JECFA) in 1983 derived a value of 0.013 mg/L assuming a 20% allocation to drinking water based on provisional maximum tolerable daily intake (PMTDI) of inorganic arsenic of 0.002 mg/kg of body weight and confirmed as a provisional tolerable weekly intake (PTWI) of 0.015 mg/kg of body weigh in 1988 (FAO/WHO, 1989)

An extensive study on arsenic contamination in Bangladesh was conducted by the British Geological Survey (BGS), the Department of Public Health Engineering (DPHE), and Mott MacDonald Limited (MML) in two phases. The project examined 3534 distributed water samples from 61 districts (excluding 3 hill districts) in an approximate 6x6 km grid. (DPHE, BGS, and MML, 1999; BGS and DPHE, 2001). On average, 58 samples per district and 8 samples per upazila were analyzed. Although the sample size is small, considering the variation in arsenic content over short distances, the study provided a reasonable distribution of arsenic contamination in Bangladesh. The study showed that when shallow tubewells are considered, arsenic concentrations of 46% and 27% exceeded the WHO guideline value of 10µg/L and the Bangladesh Standard of 50µg/L, respectively. In case of deep tubewell samples (>150m depth), arsenic content of only 5% exceeded 10 µg/L and 1% exceeded 50µg/L, indicating that deeper tubewells provide safer drinking water.

Since the variation in arsenic levels in tubewells over short distances is unpredictable, the Government of Bangladesh, in collaboration with partner organizations, implemented a National Screening Program. This program aimed to screen all tubewells in the affected Upazilas identified by the BGS–DPHE study, to delineate contaminated tubewells and identify individuals with arsenic-related skin lesions.

Table 1. Level of Arsenic Contamination in Bangladesh (BAMWSP, 2001).

Percent TW>50 µg/L	Category	Districts	Upazilas	Union	Villages
< = 5 µg/L	Low Risk	7	35	668	22,544
> 5 – 40 µg/L	Moderate Risk	31	145	1,176	14,788
> 40 – 80 µg/L	High Risk	15	65	621	8,331
> 80 – 100 µg/L	Very High Risk	1	23	416	8,378
Total Screened		54	268	2,881	54,041

The National Arsenic Mitigation Information Centre (NAMIC), established under the Bangladesh Arsenic Mitigation Water Supply Project (BAMWSP), compiled the screening results and made the data available to support targeted mitigation efforts. A total of 4.95 million tubewells in arsenic-affected areas were screened, and 1.44 million (29%) were found to be contaminated with arsenic greater than 50 μg/L. The levels of arsenic contamination reported by the National Screening Program are summarized in Table 1(BAMWSP, 2001).

Population Exposure

The population exposed to arsenic concentrations exceeding specific thresholds was estimated by considering the area-wise distribution of shallow tubewells, the percentage of those tubewells exceeding the given concentration, and the population density. Data were compiled from multiple sources, including BGS and DPHE (2001), BAMWSP (2001), DPHE (2000), and BBS (2001). The total exposed population was calculated as the sum of the products of the population in each area and the fraction of contaminated tubewells within that area. Assuming that both contaminated and uncontaminated tubewells were used by an equal number of people within a given geographical unit. The estimated population exposed to various levels of arsenic in drinking water from tubewells is presented in Fig. 1. This estimate showed that 29 million people in Bangladesh in 2001 were exposed to drinking water containing arsenic concentrations exceeding the national standard of 50 μg/L, and 46 million exceed the WHO provisional guideline value of 10 μg/L.

The BGS-DPHE studies finally gave two estimates of population exposure based on a projected population of 125.5 million in 1999 (BGS and DPHE, 2001). The estimates of total population exposed to arsenic concentrations above 50 and 10 μg/L using the kriging method were 35.2 million and 56.7 million, respectively. Based on upazila statistics, the exposure levels to arsenic exceeding 50 and 10 μg/L were 28.1 million and 46.4 million, respectively. School of Environmental Studies, Jadavpur University (SOES, JU), Calcutta, and Dhaka Community Hospital (DCH), Dhaka, estimated that the populations exposed to above 10 μg/L and 50 μg/L in 43 districts of Bangladesh were 51 million and 25 million, respectively (SOES and DCH, 2000).

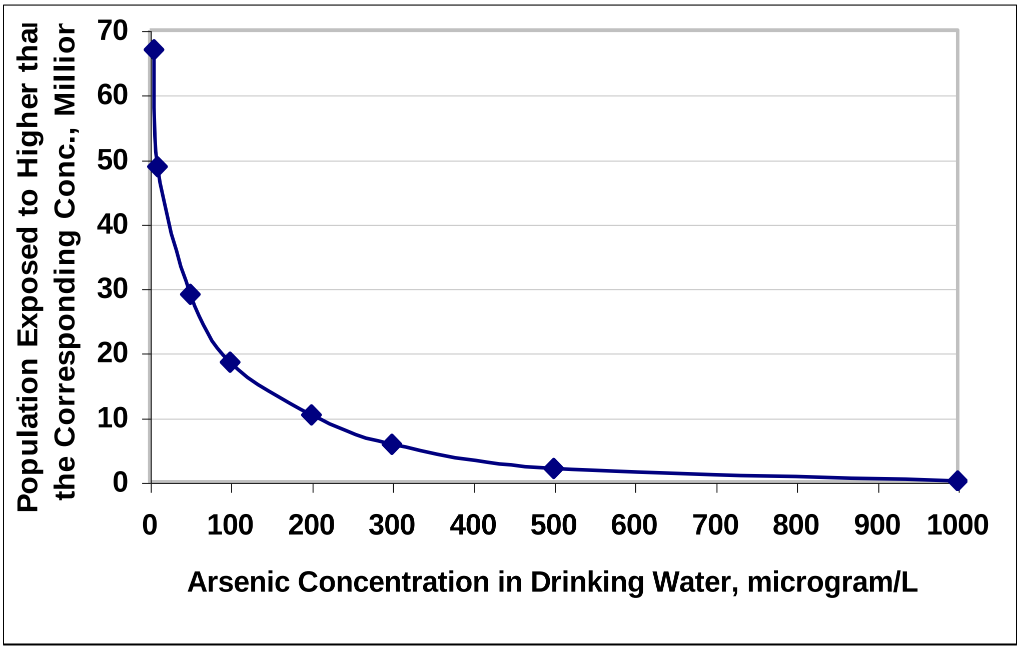

Fig. 1. Population exposed to different levels of arsenic in drinking water.

Arsenic in Agro-environment

Arsenic in Irrigation Water

Arsenic-contaminated water from shallow aquifers is extensively used for irrigation in the dry season in Bangladesh. Dry-season irrigation is needed to increase cropping intensity and produce more food. The percentages of shallow tube well (STW) producing water with arsenic content exceeding the Bangladesh standard of 50 µg/L and the WHO Guideline value of 10 µg/L, and intensity of STW-based irrigation in 8 hydrological regions of Bangladesh are shown in Fig. 2 (WARPO,2001).

The intensity of irrigation using STW is highest (50%) in the northwest region, and fortunately, very few tube wells are contaminated with arsenic there. But groundwater used for irrigation in the southeast regions is significantly elevated, and it is highly contaminated with arsenic. Groundwater-based irrigation in Southcentral, Southeast, and hilly regions is very low. However, the use of groundwater for irrigation in arsenic-contaminated regions will increase in the future for growing more crops to meet the growing demand for food.

Irrigation tube wells operate seasonally for 3-4 months' and a considerable amount of arsenic is withdrawn with groundwater and spread over irrigated land. The highest concentration of arsenic is found around water distribution channels on irrigated lands. Rice crops require about 1000 mm of water, and a concentration of 100 µg/L of arsenic in irrigation water can contribute about 1 kg of arsenic per hectare of irrigated land in each season. If all irrigation wells operate at full capacity, over 900 metric tons of arsenic could recycle each year through irrigation water (Ali et al., 2003a).

Arsenic builds up in the topsoil when irrigated with arsenic-contaminated water, and studies show that the concentration may reach a critical level to affect crop productivity. The results of limited experiments on rice grown with arsenic-contaminated soil and water in pots have demonstrated a decline in yield and accumulation of arsenic in rice grains, as well as high concentrations of arsenic in soils and water (Heikens, 2006).

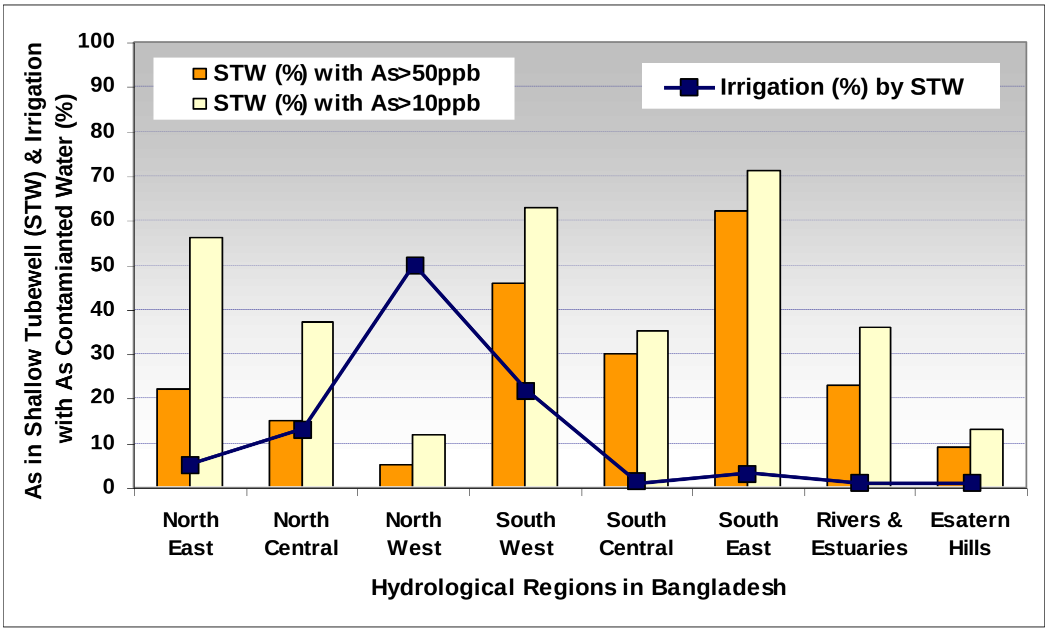

Fig. 2. Irrigation with arsenic contaminated water in Bangladesh.

Arsenic in Soil

Arsenic is a naturally occurring element in soil worldwide. The global average soil arsenic concentration ranges from 5 to 6 mg/kg. In Bangladesh, concentrations vary by soil type, averaging 2.9 mg/kg in alluvial sand and 6.5 mg/kg in mud/clay (DPHE and BGS, 2001). Irrigation dynamics significantly influence soil arsenic levels. As irrigation water evaporates, transpires, or percolates, the arsenic it carries is primarily adsorbed by soil grains containing iron, manganese, and aluminum oxides. Consequently, soil arsenic levels correlate strongly with the arsenic content of irrigation water. However, under certain conditions, this process can reverse, with arsenic dissolving from the solid phase back into the groundwater. Meharg and Rahman (2003) recorded concentrations up to 46 mg/kg in affected areas, compared to less than 10 mg/kg in areas with low-arsenic water. Localized studies have reported even higher levels, such as 51 mg/kg in Faridpur and 83 mg/kg in Comilla (Ullah, 1998). The critical threshold for arsenic in soil is variable, ranging from 21 mg/kg to 51 mg/kg depending on soil type. However, a general acceptable level is considered to be 20 mg/kg (BARI, 2007, citing Yan-Chu, 1994; Wauchope, 1983).

In Bangladesh, irrigation with arsenic-contaminated water results in significant accumulation of arsenic in the soil profile, with the highest concentrations in the top layer. Research indicates a sharply declining gradient with depth. An estimated 90% of the arsenic introduced via irrigation accumulates in the upper 450 mm of soil, with the top 75 mm layer alone retaining approximately 71% of the total (Saha, 2006). At the end of the irrigation season, the average arsenic concentration in this top layer (0-75 mm) was 14.8 mg/kg in arsenic-affected areas, compared to only 1.5 to 3.1 mg/kg in unaffected regions.

The arsenic content of soil during the post-irrigation period decreases significantly, likely due to leaching by monsoon rains, flood water, and microbial methylation processes. Roberts et al. (2010) estimated that between 52 and 250 mg m^{-2} of soil arsenic is released into floodwater during the monsoon season, corresponding to a loss of 13-62% of arsenic added to soil through irrigation each year. The potential for arsenic loss through volatilization—conversion to gaseous forms by arsenic-methylating bacteria—has been quantified. Natural biological gasification rates across Bangladesh range from 0.0003 to 0.014 µg As/kg/day, which can increase to 0.017- 0.679 µg As/kg/day under optimal conditions (Islam et al., 2007). However, in soils with poor drainage and low leaching potential, arsenic can accumulate over time, reaching critical levels that threaten crop productivity.

Excessive soil arsenic accumulation significantly affects rice production, a major concern for food security in Bangladesh. As illustrated in Fig. 3, data from Faridpur shows a negative correlation between soil arsenic and productivity (Panaullah et al., 2009).

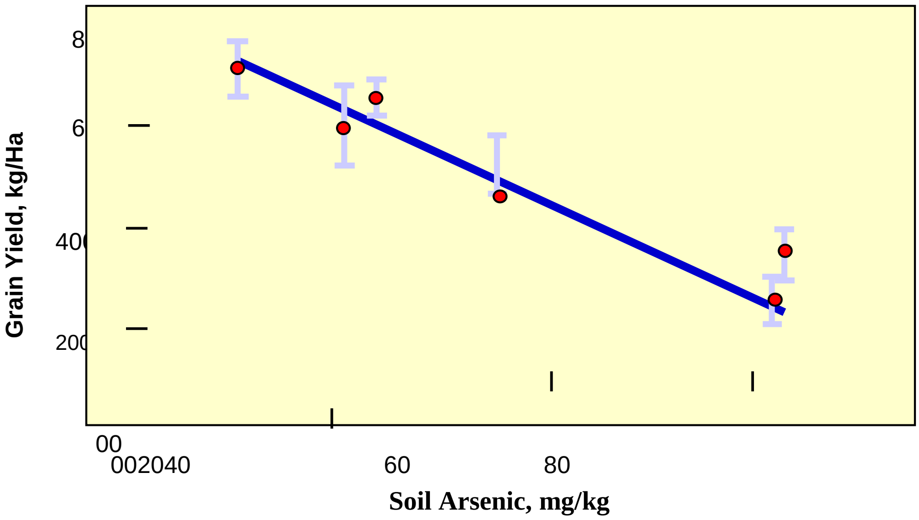

Fig. 3. Loss of Rice Productivity in Arsenic Contaminated Soil.

Arsenic in Paddy Plants

Ali et al. (2003b) found that high arsenic in water resulted in higher concentrations of arsenic in root, stem, and leaf of rice plants, and arsenic in rice grain positively correlated with arsenic in different parts of the rice plant. Saha (2006) established correlations between arsenic concentrations in the top 75 mm of agricultural soil and various parts of the paddy plant, based on approximately 85 samples from across Bangladesh. The study quantified the translocation of arsenic from soil to the root, stem, leaf, and grain, as illustrated in Figs.4 to 8.

The results indicate a strong gradient within the plant. Arsenic concentrations were highest in the roots and decreased progressively through the stem, leaves, and husk. Translocation into the edible rice grain was minimal. Furthermore, arsenic levels in the roots, stems, and leaves demonstrated a strong to moderate correlation with topsoil arsenic concentrations. In contrast, the correlation between arsenic in rice grains and the topsoil was poor, as shown in Fig.7. van Geen et al. (2006), by comparing several rice paddies from Bangladesh, including a control site, have shown that arsenic supplied with irrigation water accumulates in soil and soil-water but much less in rice grain. The observations suggest that exposure of the Bangladesh population to arsenic contained in rice is less of an immediate concern than the continued use of groundwater containing elevated arsenic levels for drinking and cooking.

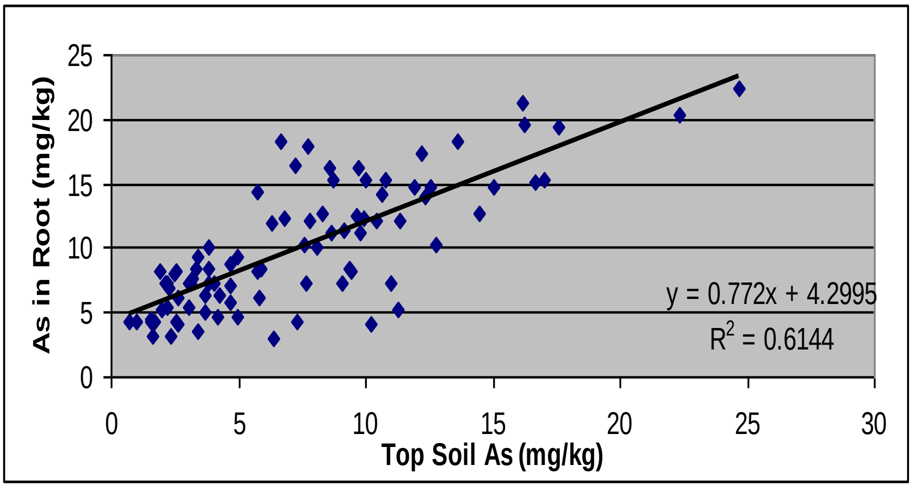

Fig. 4. Relation between As in topsoil & root.

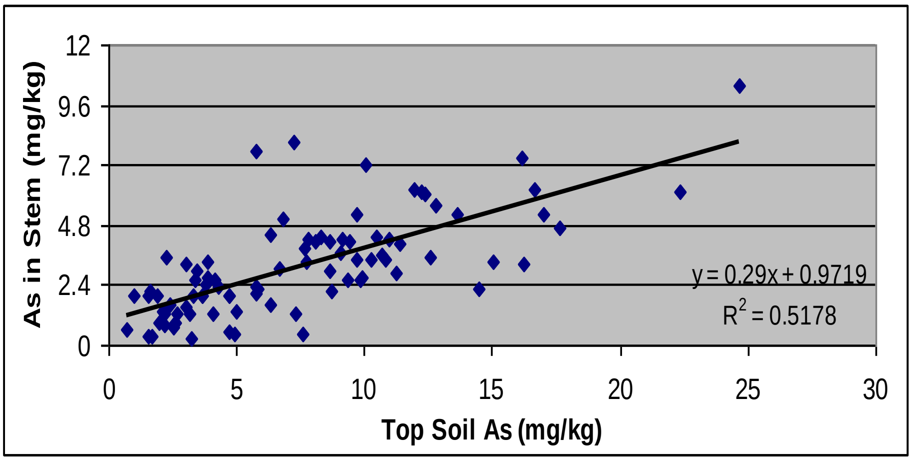

Fig. 5. Relation between As in topsoil & stem.

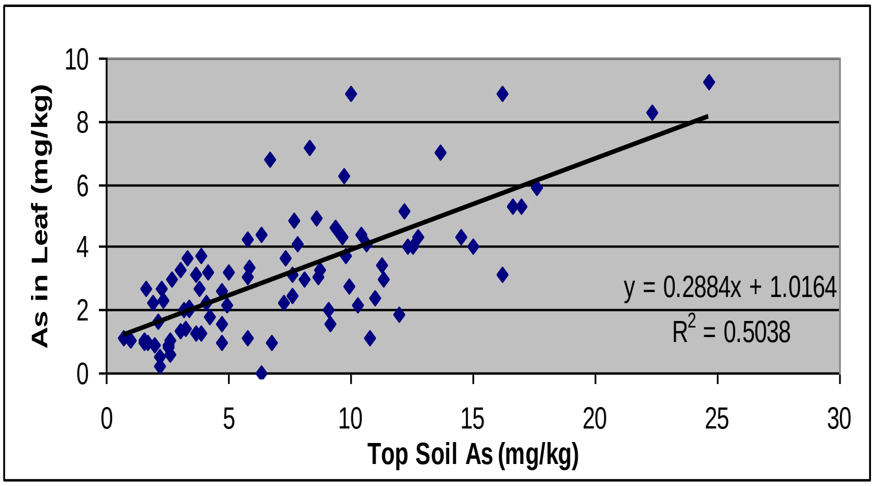

Fig. 6. Relation between As in topsoil & leaf.

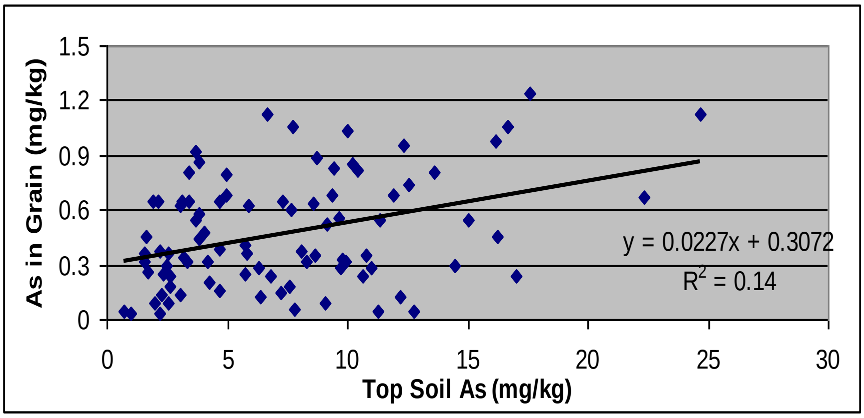

Fig. 7. Relation between As in topsoil & grains.

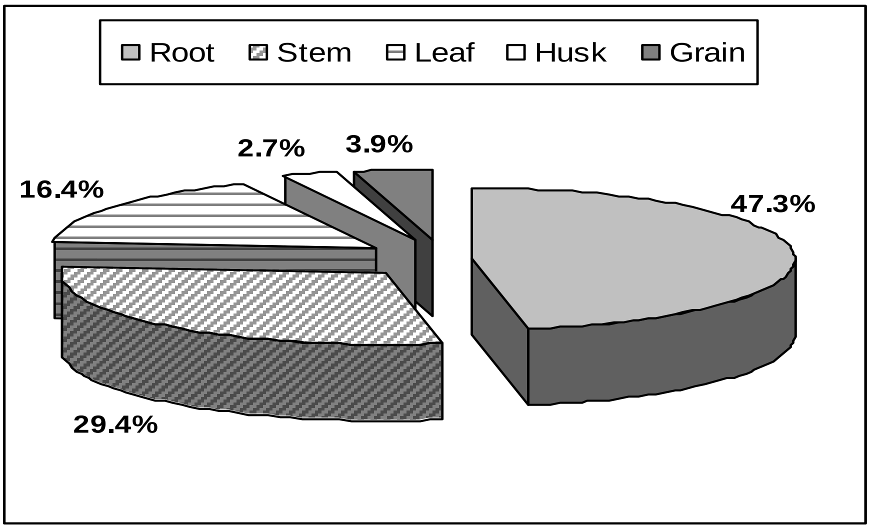

Fig. 8. Arsenic accumulation in different parts of paddy plants in Bangladesh.

Saha (2006) also determined the distribution of arsenic within paddy plants. In a HYV short-stem rice plant, 93.4% of the total arsenic accumulated in the roots, stems, and leaves, leaving only 6.6% in the husk and grains combined (Fig. 7).

A study comparing rice from arsenic-contaminated and uncontaminated regions of Bangladesh found median arsenic concentration of 0.25 ppm and 0.1 ppm, respectively (Hironaka and Ahmad, 2003). Despite levels in contaminated areas being 2.5 times higher, the average arsenic content of Bangladeshi rice was comparable to that of Japanese rice. Subsequent research has established clear links between environmental arsenic and rice grain content. Williams et al. (2006) collected 330 rice samples nationwide and found the highest arsenic levels in the southwestern region. The authors also found a positive correlation between arsenic in irrigation water and rice, noting a stronger relationship for Boro rice grown in the dry season with intensive irrigation than for Aman rice. A similar positive correlation between arsenic in rice and soil was observed by Meharg and Rahman (2003). Empirical data from Duxbury et al. (2003) recorded a wide range of arsenic in Bangladeshi rice, from 10 to 420 µg/kg. The mean concentration in Boro rice was 1.5 times higher than in Aman rice. However, the variation in rice arsenic concentrations was only partially consistent with the spatial pattern of arsenic in drinking water tube wells.

Arsenic Speciation in Rice and Food Safety

Arsenic in foods is present in both inorganic and organic forms. Inorganic arsenic is more toxic than organic arsenic present in food. The chemical form, or speciation, of arsenic in Bangladeshi rice is a critical determinant of its toxicity. Misbahuddin et al. (2007) analyzed rice samples and found the mean concentrations of inorganic arsenic, monomethyl arsenic acid (MMA), and dimethyl arsenic acid (DMA) to be 296.3 µg/kg (33.6%), 222.5 µg/kg (25.2%), and 363.4 µg/kg (41.2%), respectively. Williams et al. (2005) found that inorganic arsenic accounted for about 80% of the total arsenic in Bangladeshi rice, a proportion nearly double that found in U.S. rice (42%). It is a great concern because rice is the staple food in Bangladesh.

The high proportion of inorganic arsenic in Bangladeshi rice may pose a significant food safety risk. Australia's Maximum Permissible Concentration (MPC) for total arsenic in food is 1 mg/kg, but seafood, where arsenic is predominantly organic, the limit is 5 mg/kg. China's food safety standard specifically limits inorganic arsenic in rice to 0.15 mg/kg (Heikens, 2006). Given the high daily rice consumption in Bangladesh, dietary intake of inorganic arsenic is consequently elevated. Therefore, while the total arsenic content in Bangladeshi rice may not appear exceptionally high, its contribution to the total body burden is significant given the prevalence of more toxic inorganic species. Poor translocation of arsenic in rice grain generally leads to a belief that the productivity of rice will be affected before reaching a high level of arsenic in rice grain. If the productivity is affected by high levels of arsenic in irrigation water, it will be equally disastrous for Bangladesh, which depends mostly on rice for food security.

Estimation of Risk and Prevalence of Arsenicosis

Arsenic is naturally found in the atmosphere (0.4–30 ng/m^3), food (0.4–120 µg/kg), and water (from undetectable to 12,000 µg/L). Consequently, the global population is routinely exposed to low background levels. While arsenic is considered essential for some animal species, it is non-essential, toxic and carcinogenic to humans. As a known carcinogen and toxin, the ingestion of any amount of arsenic constitutes a potential health risk (Ahmed, 2007b).

Dose-Response Models and Health Effects

Chronic arsenic exposure is associated with a range of symptoms, including melanosis (hyperpigmentation, depigmentation), keratosis, gangrene, peripheral vascular disorders, skin cancer, and various internal cancers. In Bangladesh, skin lesions are the most commonly manifested symptom. Quantifying the relationship between arsenic ingestion and health effects remains challenging. The U.S. Environmental Protection Agency (USEPA) used a multistsge model in its 1988 assessment, based on epidemiological data from Taiwan. This model estimated that arsenic concentrations in drinking water associated with excess lifetime skin cancer risks of 10^{-4}, 10^{-5}, and 10^{-6} are 1.7 µg/L, 0.17 µg/L, and 0.017 µg/L, respectively. This indicates an approximately linear dose-response relationship at low doses.

Using this linear relationship, the number of additional skin cancer cases (N) can be estimated for a population (P) exposed to a given arsenic concentration (C) with the following equation:

$$N = 5.882 \times 10^{-5} CP \qquad 7$$

It is important to note that these estimates are conservative and may overestimate the actual incidence of skin cancer. At the time of the assessment, data were insufficient to quantitatively define an exposure-response relationship for internal cancers (USEPA, 1988).

Yu et al. (2003) developed dose-response functions for arsenic-induced non-carcinogenic skin lesions in Bangladesh. Using age-adjusted data from a survey by Mazumder et al. (1998) in West Bengal, India, they established quadratic-exponential models of the form:

$$p\,(c)_{(male\,/\,female\,)} = 1 - exp(-(q_1 c + q_2 c^2)) \qquad 8$$

where $p(c)$ is the prevalence of a specific type of non-carcinogenic arsenicosis within a gender group, c is the arsenic concentration in drinking water (µg/L), q_1 and q_2 are non-negative parameters derived from the survey data. Based on this approach, Ahmed (2007d) developed similar dose-response functions for skin lesions using patient data from 14 Upazilas in Bangladesh. This dataset was compiled by Dhaka Community Hospital through collaborative studies with organizations including BAMWSP, SOES, and UNICEF.

Ahmed et al. (2006b) developed an integrated model for the quantitative health risk assessment of drinking water contaminants. This model takes two primary inputs: Arsenic concentration (µg/L) and Microbial concentration (as TTC or *E. coli* per 100 ml). The output is the total disease burden, quantified in Disability-Adjusted Life Years (DALYs), attributable to arsenic-induced cancers of the skin, lung, and bladder.

The practical difficulty of reliably measuring arsenic at the 0.17 µg/L level (corresponding to a 10^{-5} risk, or ~1 µDALY per person per year) led the World Health Organization (WHO) in 1993 to set a provisional guideline value of 10 µg/L. This concentration is associated with a lifetime excess skin cancer risk of approximately 6 in 10,000 people (WHO, 1993). In contrast, the Bangladesh standard of 50 µg/L, when applied to the same linear model (Equation [7]), is associated with a significantly higher lifetime skin cancer risk of about 29 in 10,000 people.

Empirical Model Based on National Screening Data in Bangladesh

The empirical model is developed based on the relationship between the average arsenic content of tubewells in an Upazila and the number of cases having arsenic-related diseases in that Upazila. A national screening program was conducted from 2001 to 2002 across 268 arsenic-affected Upazilas (sub-districts) to identify contaminated tubewells and arsenicosis cases. Diagnosis was primarily based on visible dermatological manifestations, including melanosis (hyperpigmentation, leukomelanosis), keratosis, hyperkeratosis, gangrene, and skin cancer, following a protocol developed in Bangladesh and later modified by the WHO. This methodology had inherent limitations. The identified cases, while exhibiting arsenic-related skin lesions, were not confirmed through biological sample analysis (e.g., urine, hair, or nail samples). Consequently, the screening may have included non-arsenic-related skin conditions. Furthermore, the survey could not account for internal cancers and other systemic health effects of arsenic ingestion.

The program tested 4.9 million tube wells and found that 29.12% exceeded the Bangladesh Standard (BDS) limit of 50 µg/L for arsenic. By screening 66 million people in these areas, 38,430 cases of arsenicosis were identified (BAMWSP, 2001). This aligns with the earlier National Hydrochemical Survey (BGS and DPHE, 2001), which found 25% of tube wells nationwide exceeded the BDS. The prevalence of identified arsenicosis cases was significantly lower than expert predictions based on exposure levels. The leading explanation is that the duration of exposure to contaminated water was not sufficient for the full spectrum of health effects to manifest in the exposed population.

Establishing a dose-response relationship

The distribution of patients across Upazilas did not perfectly correlate with the local intensity of tubewell contamination. To analyze this relationship, the average arsenic concentration for each Upazila was computed using BGS and DPHE data. While the sample size is limited, given the high spatial variability, it provides a reasonable estimate. A key methodological consideration was handling samples below the detection limit; following the BGS/DPHE report, these were assigned a value of half the detection limit. Fig.9 plots the number of arsenicosis cases in each Upazila against the average arsenic content of its tubewell water. The result demonstrates an observable dose-response relationship, despite the reliance on clinical diagnosis of skin lesions.

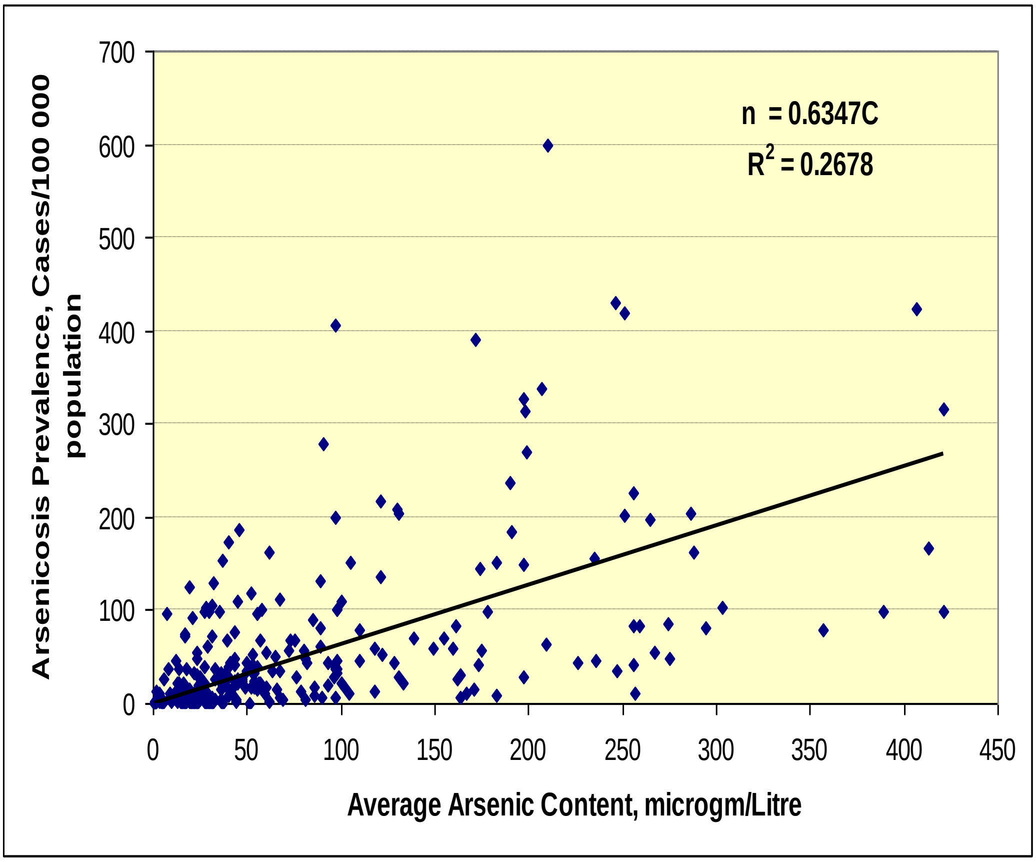

Fig. 9. Relationship between arsenical skin lesion and average arsenic content in drinking water.

A linear fit forced through the origin yields a correlation coefficient (R) of 0.2678, indicating a very weak and poorly defined dose-response relationship, as evidenced by the erratic and scattered data points.

$$n = 0.6347\, C \qquad 8$$

Where n is the total (male + female) cases of arsenic-related skin lesions per 100,000 population, and 'C' is the average arsenic content in µg/L of tubewell water. So Eqn. 8 can be written as:

$$N = 0.6347 \times 10^{-5} CP \qquad 9$$

A comparison of the WHO dose-response [Eqn. 7] and empirical dose-response [Eqn. 9] models reveal a significant discrepancy. The current average prevalence rate of clinical arsenical skin lesions [Eqn. 9] is substantially lower than the excess skin cancer risk predicted by the WHO linear no-threshold model [Eqn. 7]. This is consistent with the age-adjusted skin cancer estimate by Yu et al. (2003), which also falls far below the linear model's prediction.

Applying the empirical model [Eqn. 9] to the national context—using an average arsenic concentration of 55 µg/L (BGS & DPHE, 2001) and a 2001 population of 129 million (BBS, 2001)—yields an estimated 45,032 arsenicosis patients. This figure aligns closely with the 38,430 cases identified in the National Screening Program, validating the empirical model's relevance for the observed clinical presentation.

The Director General of Health Services (DGHS) recorded 38,320 arsenicosis patients in 2009 among those who approached health centers for treatment with skin lesions. These two studies, conducted about 8 years apart, show similar numbers of patients, indicating a stagnation in the prevalence of arsenicosis cases. However, these two sets of data are not comparable because the DGHS recorded cases were medically treated for arsenicosis by qualified medical professionals, but many cases might not have been approached for medical treatment. On the other hand, the national screening compiled by NAMIC and BAMWSP was identified by non-professionals observing skin lesions.

Two key interpretations emerge from this data.

First, the prominent regional variability and the lower-than-predicted case numbers suggest that the skin cancer risk of 10^{-5} at 0.17 µg/L, as derived from the linear model, may be a significant overestimate for the Bangladeshi population.

Second, it is equally plausible that the health effects are in a preliminary stage, and the full burden of disease, particularly internal cancers, corresponding to the present contamination level, has yet to manifest, which can only be confirmed by a second round of National screening of arsenicosis cases and levels of contamination of drinking water sources.

Arsenic Risk Mitigation

The Government of Bangladesh embarked on an arsenic mitigation program by developing an arsenic mitigation strategy. In the first phase, Bangladesh has undertaken a program to screen all the tubewells and population in 268 potentially arsenic-affected upazilas to identify contaminated wells and arsenicosis cases. Screening of all tube wells was necessary due to unpredictable variations in arsenic levels in tube well water, even over short distances and at different depths. The contaminated and uncontaminated tube wells are marked red and green respectively. A protocol was developed for diagnosing cases of arsenicosis. The arsenicosis cases are referred to Upazila health centers or specialized hospitals depending on the severity of the cases. An arsenicosis case management protocol was developed to provide health care for affected patients in health centers and hospitals.

Arsenic toxicity has no known effective treatment. Chelating agents used for acute arsenic poisoning have been tested for chronic cases, but the results have generally been unsatisfactory. However,long-term drinking of arsenic-free water has been shown to reverse symptoms in patients at early stages of arsenicosis. Hence, provision of arsenic-safe water to about 30 million people exposed to high levels of arsenic was given priority in arsenic risk mitigation in Bangladesh. The strategy for providing arsenic-safe water was guided by the principles stated below:

• Alternative technologies are area dependent and cannot be generalized uniformly across the country.

• No single option can serve the needs of people with different social and economic conditions.

• Choice of the community shall be given priority in the selection of technological options.

• Alternative water supplies must comply with the Bangladesh Standard for arsenic in drinking water.

The treatment of arsenic-contaminated groundwater or switching over to arsenic-safe sources like surface water and rainwater are possible alternatives. In some areas, deeper aquifers are found to yield water with low arsenic content and can be used as a source of arsenic-safe water supply. The National Policy and Implementation Plan for Arsenic Mitigation (IPAM), 2004, emphasized that while research to devise appropriate options was ongoing, arsenic mitigation programs shall promote Improved Dug Well, Pond Sand Filters, Deep Tube well, Rainwater Harvesting, Arsenic Treatment Technologies, and Piped Water Supply System for arsenic safe water supply (GoB, 2004).

The possibility of cross-contamination from shallow aquifers due to over pumping of deep aquifers cannot be excluded. Treatment of surface water having low arsenic content by small treatment units, rainwater harvesting and use of dug well water were promoted extensively in arsenic-affected areas of both Bangladesh and India. A risk assessment of arsenic mitigation options revealed that although surface water and rainwater provided arsenic-safe water, consumers in some cases were exposed to a higher microbial health risk from these sources (Ahmed et.al., 2006b).

The approaches outlined in the Bangladesh National Arsenic Mitigation Policy and Implementation Plan were pursued but achieved only partial success. The progress in arsenic mitigation was very slow, and as of 2006, only about 14 percent of the exposed people had access to arsenic-safe water (Ahmed et al., 2006a).

Many units developed for the treatment of arsenic-contaminated water at household and community levels and installed for experimental use in different parts of Bangladesh have shown good potential for arsenic-safe water supply. The Bangladesh Council of Scientific and Industrial Research (BCSIR) conducted an evaluation of prospective arsenic removal technologies in collaboration with the Ontario Centre for Environmental Technology Advancement (OCETA), Canada under the Environmental Technology Verification- Arsenic Mitigation (ETV-AM) program. The performance of the technologies was found to be greatly influenced by the presence of phosphate, silica, pH, and dissolved organic matter, and in some areas with adverse water quality, no technology worked satisfactorily. Only four household and community-based technologies were accepted for deployment in Bangladesh after extensive laboratory and field verification (Ahmed and Ahmed, 2014). All arsenic treatment technologies have their merits and demerits, and require refinements to make them suitable for rural conditions.

BBS and UNICEF (2019) Multiple Indicator Cluster Survey (MICS) study showed that 11.8% water was contaminated with arsenic exceeding Bangladesh standard of 50 μg/L at source, which reduces to 10.6% at the point of consumption. Arsenic contamination, compared with the WHO Guideline value, was higher: 18.6% at the source and 16.7% at the point of contamination. Arsenic reduction over time is due to its coagulation with iron in water and sedimentation in storage containers. The UN Joint Monitoring Program (JMP) estimated that Bangladesh achieved 59 percent coverage of a safe managed water supply, whereas coverage of basic water is 99 percent (WHO/UNICEF, 2000-2024). This wide gap in coverage of drinking water supply was due to microbial and arsenic contamination. Consequently, Bangladesh remains off track in achieving Sustainable Development Goal (SDG) for a safe and managed drinking water supply.

Conclusions

Arsenic contamination of groundwater in Bangladesh is a natural phenomenon triggered by reductive dissolution of sorbed arsenic on oxidized iron, alumina, manganese, and other minerals carried by the fine-grained sediments of the rivers Ganges, Brahmaputra, and Meghna River system. These fine-grained sediments carry arsenic released by the weathering of arsenic-rich minerals in upstream basins and deposit in floodplains, particularly in depressed areas with relatively stagnant water, where reducing conditions with low redox potential are created by the anaerobic decomposition of organic matter. Dissolution and desorption of arsenic from sediments, particularly from arsenic-rich iron oxyhydroxide present on soil grains, and reduction of As(V) to more mobile As(III) appear to be the main mechanisms of groundwater contamination in Bangladesh.

At present, 75% of the areas under irrigation in the dry season use groundwater from shallow aquifers, and fortunately, the levels of arsenic contamination in the areas of intensive irrigation, except the south-west region, are comparatively low. Arsenic content in rice is generally higher in arsenic-contaminated topsoil, but the relationship between arsenic content in rice and arsenic in topsoil is not strong. Although total arsenic content in rice in Bangladesh is not very high, the fraction of inorganic arsenic particularly in Boro rice, appears to be high. High rice consumption and a comparatively higher proportion of inorganic arsenic in rice need to be considered in the estimation of arsenic body burden and revision of national standard for arsenic in Bangladesh.

The National Screening of arsenic-contaminated tubewells and arsenicosis cases in 2001 revealed that the prevalence rate of skin lesions was several times lower than the estimated excess skin cancer risk attributable to arsenic-contaminated drinking water in Bangladesh. Limited data suggest that the skin cancer risk of 10^{-5} for drinking water arsenic content of 0.17 μg/L may be an overestimate. On the other hand, the health effects in Bangladesh might be in the early stages of manifestation; another National Screening is needed to better understand the situation.

The progress in arsenic mitigation in Bangladesh by providing access to arsenic-safe water has been very slow. The UN Joint Monitoring Program (WHO/UNICEF) report 2025 indicated that Bangladesh was out of track in achieving universal access to safely managed water. If the current rate of progress continues, Bangladesh will not meet the Sustainable Development Goal (SDG) for drinking water.

Authors contribution

The corresponding author, M Feroze Ahmed, declares that this paper has been prepared by compiling and analyzing data from relevant sources by me. The co-author, Tanvir Ahmed, has edited the paper and given his consent for the article to be considered by the Editorial Board for publication in the Journal of Bangladesh Academy of Sciences.

Conflict of interest

Regarding publication of this paper, the authors have no conflict of interest.

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