

Review of Arsenic Contamination in the Shallow Groundwater of the Bengal Basin, Bangladesh

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1. Abstract

Arsenic is present in a variety of organic and inorganic forms in the atmosphere, soils, minerals, rocks, natural waters and organisms. The Holocene alluvial aquifers of the Bengal Basin have excess levels of dissolved arsenic, with most of the sediment being derived from Himalayan erosion. Around 20% of the country's tube wells have been supplied with water for an estimated 20 million people, or 14% of the national population with arsenic exceeding 50 µg/l. Arsenic concentrations tends to increase with depth in the shallowest part of the aquifer system but then decrease at depths below 100 meters. High contamination takes place in shallow alluvial aquifers, which has an alarming effect on the health of millions in Bangladesh and in many other countries. Oxidation of arsenic bearing minerals into the alluvial sediment, and anoxic conditions enable the iron Oxy-hydroxide (FeOOH) to reduce arsenic concentrations are two most common hypotheses in arsenic release mechanisms in groundwater of Bengal Basin. The current scientific understanding is that arsenic is released into groundwater from sediments by reduction dissolution. This article provides an overview of the current status of contamination, distribution, origin and causes of arsenic in shallow groundwater of Bengal Basin, Bangladesh and recommends probable measures to mitigate the situation.

2. Introduction

Since last few decades, presence of high concentration of arsenic in groundwater has been recognized as a major public-health concern in several parts of the world. Arsenic contamination has been detected in many parts of the countries worldwide. The South Asian countries such as India and Bangladesh, East Asian countries such as China, Cambodia and Vietnam, South American Countries such as Chile and Argentina, and North American country such as USA have been affected by arsenic in large scale [1, 2, 3]. Bangladesh faces many health issues due to arsenic contamination in shallow groundwater. The level of arsenic is much greater than established allowable limits for human health. About 43 thousand population of Bangladesh die from arsenic related disease every year [4]. Unfortunately, the country has been facing this problem since many years.

In Bangladesh, arsenic occurs naturally in groundwater. But the deadly contamination of the drinking water of millions of rural population in Bangladesh by arsenic is a disaster which has been caused and perpetuated by people. Arsenic does not affect the drinking water of the capital city of Dhaka, or other large towns where drinking water comes from deep aquifers of higher quality water or from treated surface water, which is then distributed

through a pipeline network. Instead, it affects hand-pumped shallow tube wells, across large areas of the rural Bangladesh. Since arsenic contaminated shallow groundwater is pervasive from outline areas due to large-scale drawdown causing huge pumping in the Dhaka city, it has potential to migrate downward [5]. There is likely to get deep groundwater locally vulnerable to early ingress of arsenic due to discontinuity of aquitard layering. The occurrence of saline groundwater at shallow and intermediate levels in the aquifer system secure in deep pumping strategy at Southeast region of Bangladesh [6]. Therefore, the caution and further study are needed about arsenic contamination.

In the early 1990s, it was not known that much of the shallow groundwater in Bangladesh, especially in central and southern Bangladesh, contains arsenic. Bangladesh has a standard of 50 $\mu\text{g/l}$ of water arsenic. The significant death and illness in Bangladesh for the region of Arsenic contaminated water increases 10 and 50 $\mu\text{g/l}$ [4]. Human Rights Watch reviewed arsenic-contaminated tubewells throughout the country at more than 50 $\mu\text{g/l}$ of water in 2016. National tubewell screening from 2000 to 2006 found that approximately 20 percent of tubewells in the country delivered water for an estimated 20 million people or 14 percent of the national population with arsenic above 50 $\mu\text{g/l}$ (Figure 1a). Most tubewells in Bangladesh were installed at a shallow depth of less than 150 meters and often in the 20 to 50-meter range. The main exception to this is in the coastal areas of Bangladesh, where salinity in the shallow aquifer has meant that tubewells have historically been installed at a depth of between 150 and 200 meters to reach fresh water. Multi-parameter groundwater hazard maps (Figure 1b) is shown that a 5-24% land area are under extremely high to high risks in Bangladesh. This area has risk of arsenic and salinity contamination, and groundwater storage depletion [7]. Near about 6.5 million (2.2 million poor) to 24.4 million (8.6 million poor) people are affected in high risk of arsenic, salinity and groundwater storage depletion.

2.1. Occurrence of Arsenic in the GBM Basins

Geologically, Bengal basin is a young and largest deltaic basin in the world. It comprises primarily of a large alluvial basin floored with Quaternary sediments deposited by the Ganges and Brahmaputra rivers and their numerous associated streams and distributaries. The country occupies most of the Bengal delta. The Ganges-Brahmaputra delta basin or the Bengal basin is part of the Indian state of West Bengal in the west and Tripura in the east. Bangladesh's geological development is essentially linked to the elevation of the Himalayan Mountains and the outbuilding of deltaic landmass by major river systems originating in the elevated Himalayas. This geology is mainly characterized by the rapid subsidence and filling of a basin in which a large thickness of deltaic sediments was deposited as an out built mega- delta and advanced towards the

south. The delta building continues into the present Bay of Bengal and gradually follows a wide fluvial front of the Ganges-Brahmaputra-Meghna river system from behind. Only the eastern part of Bangladesh has been elevated to a hilly landscape that is part of the front belt of the Indo- Burman region to the east.

In recent years, the presence of arsenic in groundwater of Bangladesh has disturbed the whole scenario of its use. It has been reported that out of 64 districts 61 are affected more or less (Figure 1). In 1993, the Department of Public Health and Engineering (DPHE) first detected the presence of arsenic in tube-well water at Chapais Nawabganj, a northwestern district of Bangladesh. In 1996, Bangladesh Water Development Board (BWDB) confirmed arsenic contamination in groundwater of the western border belt. Since the identification of arsenic contamination in Bangladesh, the scientists and several agencies have been engaged themselves to carry out local, regional or national surveys to detect and mitigate the problem. Besides scientific study and investigation, wise management and governance of water resources in the arsenic affected areas has become an important issue in Bangladesh.

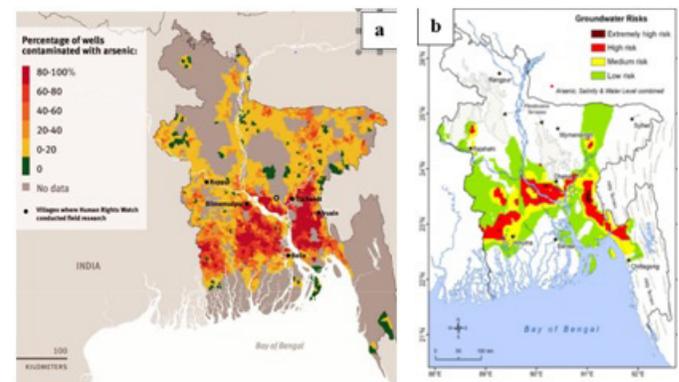


Figure 1: a) Map showing the proportion of wells at more than 50 $\mu\text{g/l}$ of arsenic in water per administrative union (Human Rights Watch, 2016); b) Groundwater risk map based on arsenic, salinity and dry-season groundwater level (Shamsudduha et al., 2019).

Concentrations of arsenic up to 4,400 $\mu\text{g/l}$ have been detected in groundwater from Chinese Provinces of Xinjiang and Shanxi and have also been identified in parts of north-central and north-east China [8]. Arsenic concentration of between 40 and 750 $\mu\text{g/l}$ is found in deep artesian groundwater, increasing concentration with increasing depth (upto 600m), while, in shallow non-artesian groundwater it was observed between <10 and 68 $\mu\text{g/l}$ (Wang and Huang, 1994). Shallow (<50 m depth) groundwater in the Quaternary alluvial sediments of the lowland Terai region of southern Nepal is containing high arsenic concentration (up to 120 $\mu\text{g/l}$) and occur in anaerobic groundwater, often associated with high concentrations of dissolved iron [9]. The West Bengal, India was the first Southeast region to be recognized with the endemic arsenic problems in 1983 by Chakraborty and Saha (1987) and was internationally well known in the mid-1990s. The Holocene alluvial and

deltaic sediments of the region of 8 districts of eastern West Bengal is similar to the large parts of Bangladesh where the arsenic in groundwater have been identified in the range of $10-3200\ \mu\text{g/l}$ [10]. High arsenic concentration is restricted to groundwater between 10 and 80 m depth. The sediments of West Bengal appear to contain arsenic has similar concentrations to those found in Bangladesh [8].

In Bengal Basin, Occurrence of toxic level arsenic predominantly found in the middle Holocene sediments deposited in low-lying delta and floodplain areas. Highly arsenic contamination occurs in shallow alluvial aquifers, which dangerously affects to the health of millions of people and has become an alarming health and environmental issue in Bangladesh and neighboring countries [2].

The main objective of this paper is to review the current status of arsenic contamination considering its origin, distribution and causes in shallow groundwater of Bangladesh.

3. Methodology

This paper is based on information and data collected from a number of well-recognized international and national journals and related documents to provide a complete picture of the recent status of arsenic contamination in shallow groundwater and sources as well as distribution of arsenic contamination. Analyzed data are reviewed and have tried to explain in technical aspects. Though information and data are not sufficient, a great deal of effort has been taken to conclude an appropriate explanation in this article.

3.1. Geochemistry and Speciation of Arsenic

Of the more than 320 known arsenic minerals, less than 10 are commonly identified in the environment and most common arsenic minerals are arsenic oxides or arsenic sulfides [11]. Arsenic is an element with the atomic weight of 74.92159 and atomic number of 33 and found in the atmosphere, soils and rocks, natural waters and organisms, in a number of organic and inorganic forms. Arsenic occurs as a major constituent in more than 200 minerals in association with the transition metals as well as Cd, Pd, Ag, Au, Sb, P, W and Mo [12]. The Clinical effects of chronic arsenic poisoning tend to have a long term exposure of arsenic drinking water ranging from skin ailments, through damage to internal organs to gangrene and cancer. Unfortunately, some of the Ganges and Yellow river basin countries are highly affected and exposed for its population due to arsenic poisoning. It is known as the worst cases of arsenic related health hazard high-arsenic groundwater that is constrained to a restricted range of geological, hydrogeological and geochemical condition in aquifer [8]. Usually, groundwater has full of arsenic minerals such as arsenopyrite (FeAsS), realgar (AsS), orpiment (As_2S_3), scorodite ($\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$), and arsenolite (As_2O_3) and the most toxic form is arsine gas, arsenite (As III) and arsenate (As V). The interactions between groundwater and aquifer sediments are derived from natural or geogenic as the primary source

of arsenic. Mining activity, combustion of fossil fuels, pesticides, herbicides, fertilizers, and crop desiccants etc. are the most common occurrences of arsenic release to the groundwater.

The absorption and precipitation reactions are the processes controlling the partitioning of arsenic and arsenic species which have the result of arsenic retention by the solid phases. Similarly, the reverse, desorption and dissolution reactions have the result of arsenic release from the solid phase. However, on a scenario of particular chemistry of reactants, products and the chemical environment of the aquifer groundwater, the oxidation/reduction reactions are seen as its retention or release of arsenic [13, 14]. The overall mass of arsenic transferred from solid phases to dissolved phase or dissolved phase to solid phase will act as a net source or sink for arsenic to the aquifer [13]. Elevated concentrations of dissolved iron, bicarbonate, ammonium, and phosphate under reducing condition are the characterization of arsenic rich aquifer [15]. Adsorption/desorption processes are very important for controlling the arsenic. Moreover, high acidity (low pH), and presence of iron and sulfate in the oxic horizon are the characterization for arsenic rich aquifer [16]. Oxidized mineral phases of iron, manganese and aluminum, iron-sulfides (pyrite), organic matter, redox-reactions, hydrolysis and diffusive transport are the controlling methods for the partitioning of arsenic sediments [17, 18]. The organic matter can have an important role in controlling arsenic release and transport for the correlation between organic carbon content and arsenic distribution [19]. Unlike the young sedimentary aquifers, the local mineralized areas such as mining areas are known to be hazardous for groundwater arsenic contamination. Even though it is known to be less regional significant occurrence, it can be seen as the result of increasing concentrations of groundwater in arsenic for some geothermal areas [20].

Anoxic conditions develop resulting burial of sediments as the Arsenic species in form of arsenic (V) and arsenic (III) are strongly adsorbed on and/or co-precipitated with iron and manganese oxide/hydroxides in the oxic environment and release in the interstitial water [21, 22, 23]. The significant amounts of arsenic-species are deposited by hydrous manganese oxides and hydrous aluminum oxide [13]. Arsenic (V) and arsenic (III) adsorbed to hydrous ferric oxides and arsenic (III) adsorbed to hydrous aluminum oxides are the Preliminary XANES (X-ray absorption near edge spectroscopy) evaluation [13] that predominant species in nature. From this phase, Hydrous manganese oxides and hydrous aluminum oxides are capable to reductive dissolution by microbes specializing in respiration of arsenic (V) to arsenic (III) can produce the selective release of arsenic [24]. Pyrite, arsenopyrite and/or unspecified sulfide minerals are known to be the primary source of arsenic in groundwater [25, 26].

3.2. Arsenic Contamination in Bangladesh Groundwater: Origin, Distribution and Causes

Excess concentrations of dissolved arsenic exist in Holocene alluvial aquifers of the Bengal Basin and most of the sediment originated from Himalayan erosion. On deltaic floodplains, like major part of Bangladesh, the soil conditions range from permanent reducing conditions controlled by high groundwater to conditions of alternating reduction and oxidation because of irrigation submergence and groundwater fluctuations. For the cultivation of paddy, the soil is usually kept submerged or at least saturated with fresh water throughout the growing period of the crop and reduction of the top soil starts [27]. Conversion of original soils into paddy soils can change many properties. An iron enriched subsurface horizon can form rapidly which is a diagnostic feature of this type of paddy soil. A layer rich in iron hydroxides and manganese oxides occurs under the ploughpan that is, generally, rich in arsenic [28].

The contamination of arsenic in Bangladesh groundwater is generally found within 5 to 50 m depths, little is found at lower depths. Arsenic contamination is not uniform in all areas and the broad surface geological divisions have a good correlation with the arsenic distribution (Figure 2). The Holocene floodplain and deltaic sediments are severely affected. There is a distinct regional pattern with the greatest contamination found in the south and southeast and the least contamination in the northern most and in the Pleistocene uplifted terraces of north and central regions of the country. Variability of arsenic concentration within short distance makes it difficult to predict the level of concentration of a given well even when the concentration of the adjacent wells are known. Salient observations indicate that the shallower and deeper aquifers of the country also have different isotopic signatures [29]. However, study [30] indicates that high arsenic was also detected in deep (150 - 240 m) groundwater in the central-SW Bengal Basin, Bangladesh and this high-arsenic water has radiocarbon ages younger than the low-arsenic deep water. This study opined that stratigraphic feature i.e. thick sand aquifer without aquitard together with local topography creates a natural deep flow system in some places and allows shallow arsenic-rich water to migrate towards deep of the aquifer.

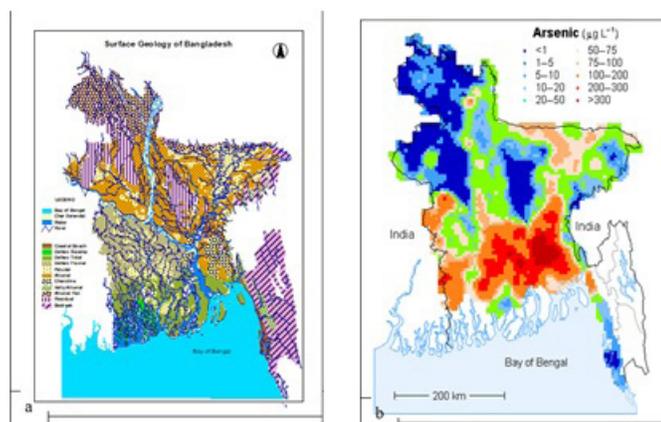


Figure 2: Correlation of (a) surface geology (Alam et al, 1990) and (b) groundwater arsenic concentrations (DPHE-BGS, 2001).

35% of all samples examined exceeded the limit of Bangladesh standard of 0.50 µg/l as stated in the Summary analysis results compiled under the project of DPHE-BGS (2001). There are 252 Upazilas (sub districts) have been covered as the Systematic Regional Arsenic Survey project in the country. 51% of total samples exceeded WHO guidelines and 35% of total samples exceeded Bangladesh standard for arsenic concentration in phase I study. About 433 sub districts have done the hydro-chemical survey for the Phase II of DPHE-BGS (2001) study. 25% of the tubewells sampled exceeded 50 µg/l, 9% exceeded 200 µg/l, 1.8% exceeded 500 µg/l and 0.1% exceeded 1000 µg/l according to the survey. World Bank has compiled information on Arsenic, Salinity, and Water Storage at the multi-hazard map with the national scale of Bangladesh [7]. According to the Map, about 6.5 to 24.4 million people have been exposed to the combined risk; 5-24% of Bangladeshi population is under extreme high to high of arsenic and salinity contamination, and groundwater storage depletion.

It is known fact that the sediments of delta have originated from the Himalayan erosion. The Predominantly silt, clay and fine sand with occasionally medium to coarse sand are depositional history of the deltaic sedimentation. Alluvial aquifers originating from Himalayan erosion supplies immature sediments with low surface loading of iron-oxy-hydroxides (FeOOH) on mineral grains to a depositional environment that is rich in organic matter where complete reduction of FeOOH is common (McArthur et al., 2004). During the cultivation age, the ration to total iron and crystalline iron oxides increases by the process of better soil aeration [31]. Bangladeshi Soil Ferrollysis (Soil Forming Process) in the wet season produces ferrous iron under reducing environment. It is known that the displaced part of exchangeable basic cations and aluminum causing leaching of bases and part on the aluminum; and interlayer formation by the remaining aluminum while some exchangeable ferrous iron is trapped in the interlayers [32]. When major clay minerals of illite and kaolinite occur in almost equal proportion in bed sediments, the Quartz and feldspars are the dominant of mineral assemblage [33]. The aquifer has close correlation with arsenic in the content of significantly high iron, manganese and aluminum. Biotites, chloride, etc. iron containing minerals are very common in Holocene alluvial sediments and above the water table, the weather forms FeOOH/FeOH_x coatings which contain as much as 800 ppm of arsenic [34]. Weathering of arsenic rich minerals release finely divided iron-oxyhydroxides that strongly adsorb co-weathered arsenic [35]. This process would supply arsenic containing iron-oxyhydroxides to Ganges sediments since the late Pleistocene [25]. Hydrous iron-oxide has a very high adsorption capacity [36]. It has a very high specific surface area of 600m²/g to adsorb the bulk of arsenic [19].

The mobilization and release of arsenic to groundwater may occur by biomediated reduction of dissolution of hydrated iron-oxide/

oxyhydroxides which is the main occurrences of coatings on sediment grains [37, 38, 39, 40, 41]. The process of FeOOH is common in nature for its reduction. It has been invoked to explain the existence of arsenic in groundwater. It is supported by high content of dissolved iron in aquifers. It is a microbial process driven by microbial metabolism of organic matter. The Reduction of FeOOH is commonly proposed because of the high arsenic concentrations accompanied by microbial reduction of arsenic (V) to arsenic (III) [42]. The FeOOH enriched sediments function as a sink for arsenic in the shallow aquifer. The local hot spot of the arsenic contamination in shallow groundwater in the Bengal Basin has the process of reductive dissolution of the FeOOH. This phenomenon has become an alarming health and environmental hazard [43].

Release of arsenic may also occur in groundwater with a high pH (> 8) in oxidizing (aerobic) conditions. These tend to occur in arid and semiarid settings with pH increases resulting from extensive mineral reaction and evaporation. High-arsenic groundwaters with this type of association have not been reported in Quaternary aquifers in Bengal Basin and surroundings [20]. In Bengal Basin, there is mainly reducing environments (low pH) are common for arsenic occurrences. The current scientific consensus is that arsenic is released by a natural chemical process called “reductive dissolution” into groundwater from these sediments [4].

Flow paths and travel time of groundwater are controlled primarily by the hydrogeologic characteristics such as anisotropy and pattern of pumping which is shown by the Model study [44]. The recharge zone for different aquifer units are about same under different development stresses in different geologic conditions. Deeper aquifers have been recharged from the vertical percolation of water from long distance travel from mainly highly elevated eastern hilly areas. The first shallow and the upper part of the second main aquifer units retards vertical percolation of water in deeper aquifers resulting the increase of travel time in the lower part of second and upper part of third deep aquifer units. The average travel time such as the age of water for the upper and the lower parts of the first and the second and the upper part of the third aquifers at different geologic conditions under the current trend of groundwater developments are estimated between 37 and 234, 133 and 317, 832 and 2485, 1009 and 3027 and 1065 and 3543 years respectively. While the water travels through the time in vertical percolation in order to recharge, the water will decrease in the first aquifer with increased irrigation pumping in future from lower part of the first aquifer. As the huge pumping continues from the first aquifer, the average travel time and length of flow lines will increase for the second and the third aquifers.

In the moment of irrigation abstraction, generally the first aquifer of water has a higher head than the second aquifer of water. Through the aquitard windows, water from the first aquifer may move downward into the deeper fresh water zones. The third aquifer

of the groundwater generally flows laterally. The third aquifer of the water level is higher than that of the second aquifer. The upper aquifers are not likely to be drawn into the deep aquifer under conditions of moderate use of the deep aquifer water for arsenic or chloride-rich groundwater. The possibility of advective transport of arsenic in the lower part of the first aquifer below the arsenic contaminated zone will increase in future under the current trend of groundwater abstraction. If only domestic wells are shifted to the second aquifer from the first aquifer, for the maintenance of current trend of irrigation abstraction, would provide better results and lower part of the second aquifer and the third aquifer will remain safe for a longer period of time. According to the Study, even under the domestic demand projected for 2050, the deep groundwater will remain secure against the invasion of arsenic across the entire region if it is restricted to domestic use only [6]. The excessive withdrawal may lead to widespread pollution of the deep groundwater after 1000 years or longer. However, for a considerable time, at least for the next 100 years, the properly monitored deep groundwater pumping could support regional development. Nevertheless, the impact of the arsenic contaminated irrigation water on food chain need to be monitored and studied carefully. In contrast, where the sea water intrusion is a problem in coastal basins, the possibility of sea water encroachment will accelerate with increased stress on the groundwater and will anticipate impact of climate change.

In arsenic affected areas, Deep tubewell (DTW), Dug Wells (DW), Pond Sand Filter (PSF), Rain Water Harvesting Systems (RWHS) and groundwater based Arsenic Iron Removal Plants (AIRPs) are effective options at different circumstances for safe water supply [45]. 15-30 families are getting benefits from each dug wells and AIRPs. RWHSs are useful for a family or few families with limited use of preserved water in dry season. Water quality needs to be monitored regularly for DWs, RWHSs and AIRPs. DTW (>300 m deep) can be installed for community water supply in the arsenic and iron affected areas. Dual platform DTWs are useful option in the flood-prone areas and usable during flood by moving pumps on higher platforms. PSFs and DTWs can be installed for community water supply too.

4. Summary

Bangladesh is under threat of severe arsenic related health problems since many years. Oxidation of arsenic bearing minerals into the alluvial sediment, and anoxic conditions enable the iron Oxyhydroxide (FeOOH) to reduce arsenic concentrations are two most common hypotheses in arsenic release mechanisms in groundwater of Bengal Basin. The current scientific understanding is that arsenic is released into groundwater from sediments by reduction dissolution. Provision of arsenic safe water in the affected areas has been the greatest challenge. To manage and govern the situation, ‘National Policy for Arsenic Mitigation 2004 has been enacted by

the Government and safe water supply has been promoted but giving priority to surface water over groundwater source. The policy allows the further development of safe groundwater in the affected areas only if other surface water options are not available. More activities and effort are needed to ensure fresh and safe water facilities for all including in the arsenic affected areas. Still expansion and improvement of the water supply services is required to satisfy the basic needs in these areas. The need is greater for under privileged groups and geophysically vulnerable regions. Proper maintenance and monitoring of context specific existing technologies are also important for long-term sustainable use. Deep groundwater, generally encountered at depths of $\pm 275\text{m}$, is a suitable option for safe drinking water in the arsenic affected areas. Large-scale withdrawal of deep groundwater, mainly for irrigation and industrial use, must not be encouraged because of the long travel-time of recharged water as well as due to possibility of arsenic leakage from upper aquifer or saline water intrusion. Maintaining current trend of irrigation abstraction, if only domestic wells are shifted to the second aquifer from the first aquifer, would provide better results and lower part of the second aquifer and the third deep aquifer will remain safe for a longer period of time.

There can be strategic and economic value, and ethical justification, in time-limited development of the deep groundwater resource, even though it may be unsustainable in the very long term. Detailed investigation and characterization of aquifers, assessment of groundwater quality and quantity is required for each groundwater zone before exploration. Regional modeling of groundwater system has to be developed for effective management plan. Model can be developed considering effects of increased abstraction stresses from aquifers, analysis of recharge and water balance. To augment the fresh water infiltration within the brackish and above the arsenic prone shallow groundwater simple recharge technology, like small diameter recharge basin/pile, can be constructed. This can be a low cost household level solution to provide safe water in hand tubewells. Great emphasis should be placed on obtaining good-quality analytical data during testing and monitoring programs. Such programs need to take account of local laboratory arsenic analytical capability and build in capability development where necessary.

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