

Recent Status of Arsenic Contamination in Groundwater of Northeastern India – A Review

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Abstract: High concentration of arsenic (As) in groundwater in the northeastern states of India has become a major cause of concern in recent years. Arsenic in groundwater has been detected in some parts of Assam, Tripura, Manipur, Nagaland and Arunachal Pradesh. The ground water in the past was considered to be safe for drinking purpose but now it came to be known that many shallow tube wells contain arsenic at concentrations higher than the safe limit set for drinking purpose by WHO 1993. It is becoming an emerging issue in the water supply and health sectors of Northeastern India. This paper reviews the recent status of arsenic in ground water of northeastern states of India and recommends proper research methods to be adopted in mitigating the effects of it to human health. [Report and Opinion. 2009;1(3):22-32]. (ISSN: 1553-9873).

Keywords: Arsenic, Arsenic in ground water, contaminated tube wells, drinking water in northeastern states of India.

Introduction

Arsenic contamination of drinking water is a worldwide problem due to its detrimental effects on health. These effects range from skin ailments through to serious diseases such as Cancer and to death. Arsenic contamination of ground water has been reported from many countries including Bangladesh, Vietnam, Argentina, China and parts of USA and now in India. The provisional limit of Arsenic in drinking water as recommended by WHO is 10 microgram per liter. Arsenic is a chemical that is widely distributed in nature and principally occurs in the form of inorganic or organic compounds. Inorganic compounds consist of arsenite, the most toxic forms and arsenate the less toxic forms. Exposure to inorganic compounds may occur in a variety of ways through certain industrial effluents, chemical alloys, pesticides, wood preservative agents, combustion of fossil fuels, occupational hazards in mining and dissolution in drinking water. The most commonly found arsenic compounds in drinking water are trivalent arsenite or pentavalent arsenate. However, groundwater is notoriously prone to chemical and other types of contamination from natural sources or by anthropogenic activities. Reliable data on exposure and health effects are rarely available, but it is clear that there are many countries in the world where arsenic in drinking water has been detected at concentrations greater than the WHO Guideline Value, 0.01 mg/l or the prevailing national standards. The major issue of arsenic contaminated water is to find out the level of contamination of arsenic and it is not so easy because of no color, no odor and no taste even in the highly contaminated water. Arsenite is more toxic (about ten times) than the arsenate due to former ability to react with sulfhydryl groups there by increasing the residence time (Nagarnaik et al., 2002). Although there is no widely accepted mechanism of the release of arsenic in ground water, but it has been accepted that most of all including in northeastern states of India is of natural, geological origin. The arsenic is thought to be closely associated with oxidation-reduction process of iron oxides and pyrite.

A study has revealed that thousands of underground water sources in India's northeast are unfit for consumption due to highly toxic contamination of arsenic (Sujit Chakraborty, 2007). 28,181 water sources located in Assam have been contaminated with arsenic, iron and fluoride inorganic materials, followed by 2,931 in Tripura, 566 in Arunachal Pradesh, 136 in Nagaland, 124 in Meghalaya, 76 in Sikkim, 37 in Manipur and 26 in Mizoram. Due to the withdrawal of excessive amounts of groundwater, problems of increased iron, fluoride and arsenic (As) contamination have been reported in different parts of India (Khan, H. R., 1994, Singh, A. K., 2004). A study on cancer risks from As in drinking water indicates that it could cause liver, lung and kidney/bladder cancer besides skin cancer (Smith, A. H. *et al.*, 1992). The most

important remedial measure is prevention of further exposure by providing safe drinking water. From this point of view, several ideas have been highlighted in this article to control and remove As from groundwater used for household activities as well as irrigation.

Distribution of arsenic in groundwater in northeastern states

The presence of As in groundwater in West Bengal is the most serious health hazard India has ever faced. According to the North Eastern Regional Institute of Water and Land Management (NERIWALM), a staggering 32,077 water sources have been contaminated with naturally occurring inorganic materials like arsenic, iron and fluoride (Sujit Chakraborty, 2007). A recent study shows that out of three blocks of Gplaghat district of Assam, Podumoni shows maximum 67.57% groundwater contamination by Arsenic followed by Kathalguri block 44.45% (Mridul Chetia, 2008).

Many areas within the northeastern states with As concentration greater than 0.05 mg/l, implying that millions of people are at serious risk of As poisoning (Singh, 2004). According to a report, the concentration of As generally varies from 0.02 to 0.9 mg/l (exceeding the WHO standard of 0.01 mg/l and Bureau of Indian Standard (BIS) of 0.05 mg/l). While As and iron pose individual problems in the aqueous environment, their association in groundwater have the potential of providing a simple means of removing As by co-precipitation and adsorption. About 95% of the area of the northeastern region contains dissolved iron in excess of 2 mg/l, and the iron concentration is as high as 15 mg/l in many areas (Singh, 2004). The North Eastern Regional Institute of Water and Land Management (NERIWALM) report 2007 said arsenic levels in Assam, Manipur, Tripura and Arunachal Pradesh were above 300 parts per billion (ppb). According to the World Health Organisation (WHO), consumption of water contaminated with arsenic levels of over 50 ppb can cause skin lesions and even cancer.

Maximum arsenic concentration content of 490 µg/l was observed in Jorhat (Titabor, Dhakgorah, Selenghat and Moriani blocks), Dhemaji (Sissiborgoan and Dhemaji blocks), Golaghat district (Podumani block) and Lakhimpur (Boginodi and Lakhimpur blocks), Assam (Chakraborti, D. et al, 2004). In Thoubal district of Manipur, about 25% of the collected samples had As in the range 798–986 µg/l. The sediment in the northern region contains a high percentage of clay and organic compounds (Acharyya, 1999), which may retain and release As in the groundwater aquifers. A study (Baruah, 2003) on As in carbonaceous matter in Arunachal Pradesh, Assam, Nagaland and Meghalaya, indicated that the mean As concentration of samples from the four regions was 95.1 mg/kg. It is believed that the weathering of sulphide associated with carbonaceous matter may have produced As-rich iron oxyhydroxides, which in turn released As (after reduction) to the existing sedimentary environment (Khan 1994, Nickson 1998). Results indicated increased As enrichment from east to west in Northeast India. The distribution of arsenic in northeastern India is presented in Figure 1.

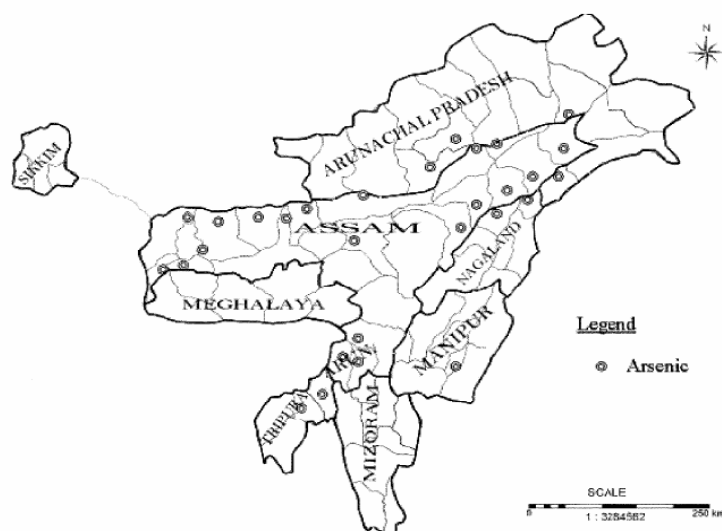
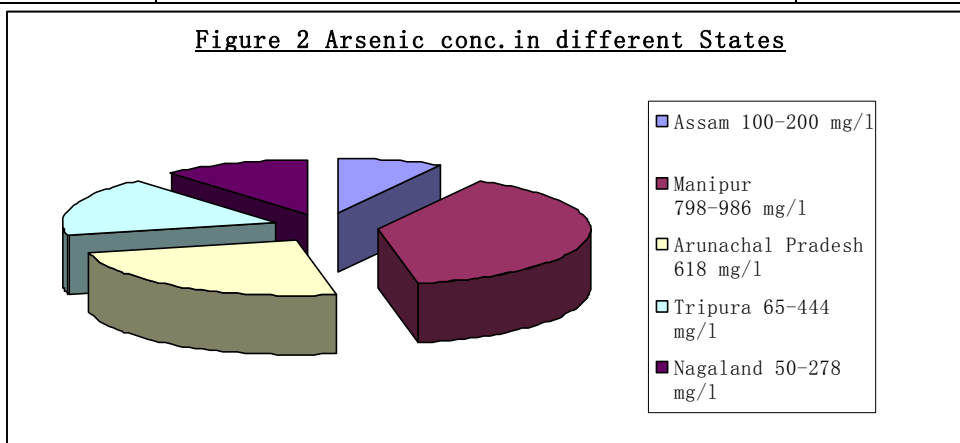


Figure 1. Distribution of arsenic in groundwater of northeastern India.

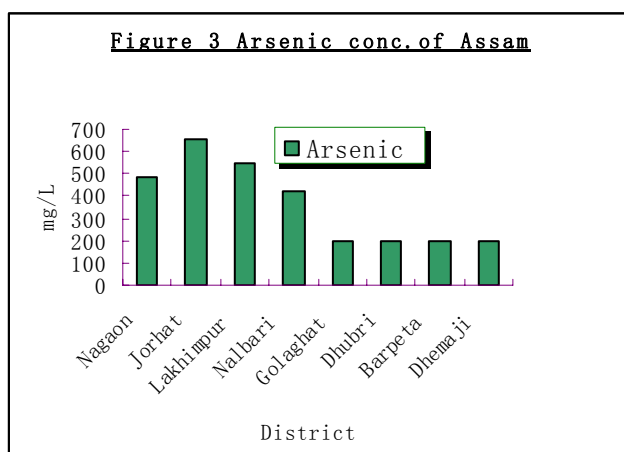
Table 1: Arsenic contaminated areas of different northeast states in India *Source: (Singh, 2004)*

States	District	Arsenic range
Assam	Barpeta, Dhemaji, Dhubari, Darrang, and Golaghat,	100-200 ($\mu\text{g/l}$)
Manipur	Thoubal	798-986 ($\mu\text{g/l}$)
Arunachal Pradesh	Papum Pare, West Kameng, East Kameng, Lower Subansiri, Dibang Valley, Tirap	618 ($\mu\text{g/l}$)
Tripura	West Tripura, North Tripura and Dhalai	65-444 ($\mu\text{g/l}$)
Nagaland	Mokok chong, Mon	50-278 ($\mu\text{g/l}$)

Figure 2 Arsenic conc. in different States



According to the Agartala news reports 2007, arsenic was found in Jirania and Bishalgarh in West Tripura district, Salema, Halahali, Kamalpur, Joynagar in Dhalai district, Sanitala, Rajbari, Dharmanagar, Kailishahar, Kanchanpur and Jampui in North Tripura district. After observing from table 1 and recent publications, news we can find out that the distribution of arsenic in northeast regions is increasing rapidly. The NERIWALM report said arsenic levels in Assam, Manipur, Tripura and Arunachal Pradesh were above 300 parts per billion (ppb). According to the World Health Organisation (WHO), consumption of water contaminated with arsenic levels of over 50 ppb can cause skin lesions and even cancer (Sujit Chakraborty, 2007). Most of the district and blocks of Assam were affected by arsenic and its spreading. The concentration of arsenic in affected districts as shown fig 3.



Arsenic sources

Arsenic is a natural constituent of the earth's crust and is the 20th most abundant element. The average concentration (NAS 1977, Taylor 1985) of As in the continental crust is 1–2 mg/kg. Arsenic is released in the environment through natural processes such as weathering and volcanic eruptions, and may be transported over long distances as suspended particulates and aerosols through water or air. Arsenic emission from industrial activity also accounts for widespread contamination of soil and groundwater environment (Jacks, G. 1998, Juillot, F. 1999), Many authors have reported As emissions to the atmosphere on global, regional and local scales (Nriagu, J. O. 1988, Pacyna, J. M. 2001). Once introduced into the atmosphere, As may circulate in natural ecosystems for a long time depending on the prevailing geochemical environments (Boyle, R. W. 1973, Yan Chu, H., 1994)

Toxicity and health effects of arsenic:

The United States Environment Protection Agency (USEPA) and International Agency for Research on Cancer (IARC) have specified arsenic as known human carcinogen. According to EPA weight of evidence classification for carcinogens, arsenic is categorized as Group A Carcinogen. Its classification indicates that there is sufficient evidence from epidemiological studies to support a case effects relationship between the substance and cancer. As is virulent poison on acute ingestion, 76 mg As (3+) is considered to be lethal to adults and extremely toxic on long-term exposure to a very low concentration (Azcue and Narjagu, 1994 in Sharma 1999). Extremely toxic arsenic oxide former use as a poison for vermin has been vastly reduced on account of concerns over its accumulation in the food chain. Arsenic enters the human body through ingestion, inhalation, or skin absorption. Significant route of As ingestion is drinking water. The amount of As species in urine is often considered an indication of chronic As exposure. The species are the result of the biotransformation of inorganic As (Table 2)

Table 2: Arsenic speciation in human urine: (Smith,T.J. 1977)%

Species	Average values	Mg/L
As(III)	1.3	1.9
As(V)	1.3	3.9
Methylarsonic acid	3.4	1.8
Dimethyl arsenic acid	11.5	15.0
Total As	21.2±2.04	22.5±8.5

The discovery of arsenic in drinking water in many areas of the world has caused widespread public health concern. Close to 100 million people in the world, including about 13 million in the United States, are chronically exposed to inorganic As [International Agency for Research on Cancer (IARC) 2004]. Inorganic As is an established potent human carcinogen (IARC 2004). In addition, ingestion of As through drinking water has been implicated in several noncancer diseases, for example, peripheral vascular disease; hypertension; respiratory, neurologic, and liver disorders; and diabetes mellitus [IARC 2004; National Research Council (NRC) 2001; WHO/IPCS (International Programme on Chemical Safety) 2001]. Early effects of exposure to As in drinking water include pigmentation changes and hyperkeratosis (IARC 2004; Smith et al. 2000), which reportedly appear after 5–10 years of exposure (Guha Mazumder et al. 1998). There may be some degree of skin absorption in contact of trivalent oxide, because of its rapid solubility. Most ingested and inhaled arsenic is well absorbed through the gastrointestinal tract and lungs into blood stream. It is distributed in large number of organs including the liver, lung, kidney and skin. About 70 % of the arsenic is excreted mainly through the urine. Most arsenic absorbed into the body is converted by liver to less toxic methylated arsenic then efficiently excreted in the urine (Saha, 1999, in Nagarnaik 2002). In the initial stage of chronic exposure of arsenic for more than five years, skin colour becomes black (melanosis), rough and tough (keratosis), eyes become red (conjunctivitis). Also in some cases there will be pain in inhaling (Bronchitis) and Vomiting and Diarrhea (Gastroenteritis). The manifestations of arsenicosis are clearer in the second stage with black and white spots on the skin (leukomelanosis), palms and soles are affected by hard nodules (Hyperkeratosis), swelling of legs, Peripheral Neuropathy and complications of kidney and liver. Finally it also turns to cancer in skin, lungs, kidneys, liver and other organs.

Guideline value for arsenic in drinking water

The first version of International standards for drinking water included arsenic in the category of toxic substances and established 200 ppb as the allowable concentration in drinking water (WHO, 1958). In updated standards of 1963, WHO lowered the allowable concentration to 50 ppb (WHO, 1963). The WHO continued its review work to lower the guideline value for arsenic in drinking water by establishing a guideline value (provisional) of 10 ppb in 1993 (WHO 1993). This provisional Guideline Value (GV) of 10 ppb has been adopted as the national standards for drinking water by a number of countries. However many developing countries have retained the previous WHO GV of 50 ppb as their national standards.

Table 3: Currently accepted national standards for arsenic in drinking water in some selected countries.

S.No.	Countries	National Standards for As in drinking water (ppb)	Year since when it has been accepted
1	Australia	7	1996
2	Jordan	10	1991
3	Japan	10	1993
4	EC	10	1998
5	Mongolia	10	1998
6	Laos	10	1999
7	USA	10	2001
8	Canada	25	1999
9	Philippines	50	1978
10	Sri Lanka	50	1983
11	Vietnam	50	1989
12	India	50	NA
13	Bangladesh	50	NA
14	China	50	NA
15	Nepal	50	2003

Testing for arsenic

Field methods

The most important characteristic of the field-testing method is that the testing can be carried out in field, where the sample is taken. In this method relatively very simple testing field-kits are used. When any metal arsenide reacts with strong acids, arsine gas is formed. Most arsenic test kits rely on the reduction of inorganic arsenic to arsine gas (AsH_3) using zinc metal and hydrochloric acid. This gas is allowed to pass through the mercury bromide (HgBr_2) indicator paper and the intensity of colour indicates the concentration of arsenic.

Many field kits, including only one India kit are available in listed as follows;

- AAN Kit (Japan)
- E-Merck Kit (Germany)
- NIPSOM Kit (Bangladesh)
- AIHHPH Kit (India)
- ENPHO Kit (Nepal)
- Modified AAN Kit (Nepal)
- Hach EZ (USA)
- Wagtech Arsenator (UK)

Analytical Methods

Numerous methods are described in the literature, for the analysis of total arsenic in water. Many analytical methods essentially employ the same principles, but apply different reagents or concentrations. The possibilities for total arsenic determination include:

1. Atomic absorption spectrometry (AAS)
 - a. Hydride Generation System (AAS-HG)
 - b. Graphite furnace (AAS-GF) for atomization,
2. Inductively coupled plasma
 - a. With atomic emission spectrometry (ICP-AES)
 - b. With mass spectrometry (ICP-MS)
3. Atomic fluorescence spectrometry (AFS)

4. Anodic stripping voltammetry (ASV) or
5. Spectrophotometry.

Both atomic absorption spectrometry and atomic fluorescence spectrometry, a relatively new and sensitive technique, are single element specific techniques with known and controllable interferences. The inductively coupled plasma techniques offer the possibility of examining a number of contaminants, as they are multi-element techniques, again with known and controllable interferences. Anodic stripping voltammetry is a useful technique for samples containing only free dissolved arsenic, while the spectrophotometric method, which is also a single element technique, has the advantage of being relatively inexpensive in terms of equipment. The mostly used in Indian laboratories AAS-HG technique is based on the atomic absorption measurement of arsenic generated by thermal decomposition of arsenic (3+) hydride. As (3+) is reduced to arsine gas by reaction with sodium tetrahydroborate in a hydrochloric acid medium (ISO 11969:1996; SM 3114:1999). Arsenate and Arsenite have different sensitivities using this technique so any arsenate must be reduced to trivalent arsenic prior to the determination and done by using a solution of hydrochloric acid, potassium iodide and ascorbic acid. In India the silver diethyldithiocarbamate spectrometric method (SDDC) and spectrophotometry have been also used as alternate to AAS-HG. The key points of each of the above techniques are summarized below in Table 4

Table 4: Summary of analytical methods for Arsenic test:

Method	Detection Limit (ppb)	Sample Size (ml)	Equipment Cost (US\$ in '000)	Daily Through put	Remarks	Selected References
AAS-HG	0.05	50	20-100	30-60	Single element	ISO,11969 (1996); SM 3114(1999)
AAS-GF	1-5	1-2	40-100	50-100	Single element	ISO/CD 15586 (2000); SM 3113 (1999)
ICP-AES	35-50	10-20	60-100	50-100	Multi element	ISO/CD 11885 (1996); SM 3120 (1999)
ICP-MS	0.02-1	10-20	150-400	20-100	Multi element	SM 3125 (1999)
HGAFS	0.01	40-50	20-25	30-60	Single element	CEN/TC/230/WGI/TG, 12 N3 (1999)
ASV	0.1	25-50	10-20	25-50	Only free Dissolved arsenic	USEPA 7063
SDDC	1-10	100	2-10	20-30	Limited to water sample	SM 3500 (1999) ISO 6595 (1982)

Global situation of arsenic in groundwater

Active groundwater contributes about 0.274 percent of total water budget and is the major source as fresh water for drinking water proposes. Arsenic contamination in groundwater has been reported in 20 different countries of the world. Four major calamities in order of magnitude are in Asia and these are Bangladesh, India (West-Bengal), China (including Inner Mongolia) and Taiwan. In terms of population exposed, arsenic problems in ground water from the alluvial deltaic aquifers of Bangladesh and West Bengal represent the most serious occurrences identified globally. The southwest coastal zone of Taiwan was perhaps the first area to be identified as a problem area for health effects arising from chronic arsenic exposure. Awareness of the arsenic problem began during the 1960s (Smedly et al., 2003) and

arsenic-related health problems were documented. Well-known black-foot diseases in Taiwan had been identified since then. The Chaco-Pampean Plain of central Argentina perhaps is the largest region of high-arsenic groundwaters known, covering around one million km².

Table 5. Naturally-occurring As problems in world ground waters: (In Smedley et al., 2003)

S.No.	Country/Region	Population exposed (million)	Area (Km ²)	Max Conc. Range (ppb)
1	Bangladesh	30	150000	2500
2	India/West Bengal	6	23000	3200
3	China	5.6	NA	NA
4	Argentina	2	1000000	5300(7800 in some pore waters)
5	Nepal	0.46-0.75	30000	600(2620 in one case)
6	Chile(North)	0.5	125000	1000
7	Mexico	0.4	32000	620
8	USA(South west)	0.35	206300	2600
9	Taiwan	0.1	4000	
10	Mongolia (Huhhot Basin)	0.1	4300	2400

Discussion:

Despite limited studies and information, a significant segment of the rural Indian population residing in the northeastern has been exposed to an elevated level of arsenic. An accurate figure of the exposed population may be much higher than presently believed, and requires the majority of the tube wells to be examined. The North Eastern Regional Institute of Water and Land Management (NERIWALM) report said people in the northeastern region use water from tube wells for drinking, cooking and agricultural purposes and this way arsenic enters the food chain. This could cause chronic arsenic toxicity in the course of time, resulting in arsenical skin lesions and dermatitis in the initial stages and cancer and death if patients are exposed to high concentration over prolonged periods. According to another study, more than 15 million people face arsenic contamination in the five states bordering Bangladesh - West Bengal, Tripura, Assam, Mizoram, and Meghalaya (Sujit Chakraborty, 2007). In recent study, Assam deep well arsenic concentration is very high, peoples not yet identified the some places which are affected but in future the problem of arsenicosis may arise (Mridul Chetia, 2008). So long term environmental planning is required to get rid of the ensuring danger of this pollution of groundwater.

One of the ways to mitigate this arsenic problem is to change the source (uncontaminated groundwater), which brings up other issues of privacy and ownership. Some field tests suggested that water from deeper tube wells (>30 m) contains a low level of arsenic (World Bank, 2005); therefore pumping water from deep tube wells for supply is another option. However, this requires a thorough investigation of the aquifers and a detailed cost analysis. And again, it may need some sort of treatment depending on the arsenic level. The rural people should aware about the arsenic pollution and uses of field kids, does at least rural people can mitigate the arsenic pollution and less arsenicosis. Therefore, government should take seriously of these rural places and need to do further monitoring.

Many technologies have been developed for the removal of As. All the technologies for As removal rely on a few basic chemical processes. Oxidation/reduction reactions reduce (add electrons to) or oxidize (remove electrons from) chemicals, altering their chemical form. These reactions do not remove As from solution, but are often used to optimize other processes. Precipitation causing dissolved As to form a low-solubility solid mineral, such as calcium arsenate. This solid can then be removed through sedimentation and filtration. When coagulants are added and form flocs, other dissolved compounds such as As can become insoluble and form solids. This is known as co-precipitation. The solids formed may remain suspended, and require removal through solid/liquid separation processes, typically coagulation and filtration. In adsorption and ion exchange, various solid materials, including iron and aluminum hydroxide flocs have a strong affinity for dissolved As. As is strongly attracted to sorption sites on the surfaces of these solids, and is effectively removed from solution. Ion exchange can be considered as a special form of adsorption. From several studies, it is already known that coagulation precipitation is an effective and most frequently applied technique in As removal (Edwards, M., 1994, Jekel, M. R., 1994, Hering, J. G., 1996). In this technique, addition of coagulant facilitates the conversion of soluble As species into insoluble

products through co-precipitation and adsorption. Adsorption of As on preformed $\text{Fe}(\text{OH})_3$ has also been shown to be an effective method for removing As (Driehaus, W., 1998). However As removal during coagulation with FeCl_3 is more efficient than As adsorption onto preformed hydrous ferric oxide (Hering, J. G., 1997). It is suggested that where As removal depends on iron precipitation, the settling time must exert a major influence on the removal process. Some simple household As-removal systems have also been developed based on traditional-sand filtration water purification along with wood charcoal or rusted iron nails as an adsorption medium. These As-removal filters have been used in As-affected areas of Bangladesh and Nepal.

Therefore, Arsenic could be removed by co-precipitation as well as by adsorption on ferric hydroxide surfaces if the groundwater contains an appreciable concentration of dissolved iron. Oxidation of iron (II) chemically or biologically to ferric hydroxide is essential. About 95% of the area of the northeastern region contains dissolved iron in excess of 2 mg/l, and the iron concentration is as high as 15 mg/l in many areas (Singh, 2004); therefore, a simple iron assisted biological sand filter may be applicable to remove arsenic to an acceptable level. The household filters currently distributed in the arsenic affected districts contain rusted iron nails (ferric hydroxides) as the sink for arsenic adsorption by a surface complexation mechanism. The soluble iron (Fe^{2+}) already present in the groundwater oxidizes in the form of ferric hydroxide, and the elemental iron (iron nails) oxidizes to ferric hydroxide chemically (air) and biologically, especially by air and iron bacteria, respectively.

The arsenic loaded iron particulates are retained on the sand filter by mechanical straining. The advantages of this kind of filter are that the iron bacteria, which thrive on iron and organic matter, develop on the filter. The outer cell of these bacteria provides a large surface area for arsenic loaded iron oxide adsorption (Katsoyiannis and Zouboulis, 2004). The pitcher filter unit can remove 90–98% As-bearing water when As bearing groundwater was passed through wood charcoal and sand. This technology is effective at household level in removing As, but the system may be quickly clogged as groundwater of the northeastern region contains excessive iron. Risk of bacterial contamination may also be checked by sterilizing the medium before filter-construction. (Murcott, Lukacs 2002) conducted a technological evaluation of the biosand filter and the possible mechanism of arsenic, iron, and pathogen removal. A recent study by Jin and Chiu (2007) at the University of Delaware stated that zero-valent iron based filters were effective in removing pathogenic bacteria and viruses (99.999%) from drinking water. The mechanism of virus removal was not clear, but the researchers believe that viruses might have been inactivated chemically by iron oxide or due to the adsorption of viruses on the iron oxide surfaces; E. coli, Hepatitis A, Norovirus and Rotavirus were also removed by zero-valent iron assisted filters (Jin and Chiu, 2007). The filters currently introduced in the northeast region are a good arsenic mitigation alternative. A long-term plan should be formulated by the government for the treatment and sustainable use of northeast groundwater. Every field kits should be checked for their performances. For this purpose establishment of a reference laboratory is urgent. Every new tube well should be constructed only after the test for arsenic.

Conclusion:

This review has attempted to summarize the incidents of arsenic contamination in groundwater in the northeastern states of India. It poses a significant risk to public health in the northeast region of India. Therefore, the first priority to remediate the crisis should be early identification of the affected sources, and the next hurdle is to provide arsenic-safe water to the affected masses. It is necessary to seal the highly contaminated tube wells to protect the non-contaminated aquifers and provide the good biosand filters to the rural northeast regions.

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