2006

Arsenic Contamination in Ground Water in South-Eastern Bangladesh

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University of Rajshahi

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Ph.D. THESIS

BY

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A thesis submitted in partial fulfillment of the requirement for the degree of Doctor of Philosophy (PhD)

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DEDICATED

TO

MY LATE FATHER

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DECLERATION

I, hereby declare that the entire work presented in this thesis as submitted to the University of Rajshahi, Bangladesh for the degree of Doctor of Philosophy, except for quotations and sources, which have been duly acknowledged.

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I, hereby certify that the entire work now submitted as a thesis for Doctor of Philosophy, University of Rajshahi, was done by the author himself. The materials included in this thesis are original and was not submitted before for any other degree.

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ACKNOWLEDGEMENTS

I would like to express my heartfelt respect, deepest sense of gratitude, immense indebtedness and profound appreciation to the supervisor, Dr. Md. Arifur Rahman, whose scholastic guidance, infinite enthusiasm, constant encouragement and valuable suggestions enabled me to complete this research work. I thank him for his invaluable patience and allowing me the freedom to explore during the course of my work. I also express my sincere appreciation and gratitude to Dr.A.J.M.S Karim, Coordinator, BARI-Arsenic Testing Laboratory, Bangladesh Agricultural Research Institute, Gazipur-1701, Bangladesh, for his encouragement, valuable suggestions during laboratory analysis and critical comments for the improvement of this thesis. My sincere appreciation is also extended to Dr. K.C Roy, CIMMYT-Bangladesh for his kind support provided during this study period and his constructive suggestions for the improvement of this dissertation.

I wish to express my gratitude to my wife Fatima Rukshana, Scientific Officer, River Research Institute, Faridpur-7800, Bangladesh, for her assistance in research and thesis writing. I am indebted to my father late Md. Mostafizur Rahman who inspired me to PhD study. I am thankful to my best friend Mr Abu Ahmed Mokammel Haque, PhD Student of AIT who gave me knowledge and suggestions about research and thesis writing. My sincere appreciation to Md. Helal Uddin, Senior Chemists, Regional Arsenic Testing Laboratory, Rajshahi, Bangladesh who helped me during laboratory analysis. My sincere acknowledgement is also due to my office staff and colleagues, Department of Agronomy and Agricultural Extension, Faculty of Agriculture, University of Rajshahi, Rajshahi-6205, Bangladesh.

The Author

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LIST OF ACRONYMS AND ABBREVIATIONS

ACA = Ammoniacal Copper Arsenate
ACZA = Ammoniacal Copper Zinc Arsenic
AAN = Asia Arsenic Network
BAMWSP = Bangladesh Arsenic Mitigation Water Supply Project
BARI = Bangladesh Agricultural Research Institute
BBS = Bangladesh Bureau of Statistics
BDWS = Bangladesh Drinking Water Standard
BGS = British Geological Survey
BCSIR = Bangladesh Council of Scientific and Industrial Research
BUET = Bangladesh University of Engineering and Technology
CCA = Chromated Copper Arsenate
CSIRO = Commonwealth Scientific and Industrial Research Organization
CIMMYT = International Maize and Wheat Improvement Centre
DANIDA = Danish International Development Agency
DPHE = Department of Public Health Engineering
DTWs = Deep Tube wells
GOB = Government of Bangladesh
HG-AAS = Hydride Generation Atomic Absorption Spectroscopy
HTWs = Hand Tube wells
INFS = Institute of Food and Nutrition Science
JICA = Japan International Cooperation Agency
LGED = Local Government Engineering Department
MSDS = Material Safety Data Sheet
MPs = Manual Pumps
NGO = Non-Government Organization
REB = Rural Electrification Board
RWH = Rainwater Harvester
STWs = Shallow Tube wells
SDDC = Silver diethyldithiocarbamate
NGO = Non-government Organization
PSF = Pond Sand Filter
ppm: Parts per million (equal to milligrams/kilogram, mg/kg)
ppb: Parts per billion (equal to micrograms/kilogram, μg/kg)
UNICEF = United Nations International Children Emergency Fund
USD = United States Dollar
USEPA = United States Environmental Protection Agency
WHO = World Health Organization

ABSTRACT

Groundwater with high arsenic concentrations from naturally occurring sources is the primary source of drinking water for millions of people in Bangladesh resulting in a major public health crisis of recent times. Arsenic concentrations can vary greatly from well to well, making the prediction of arsenic concentrations in a specific well very difficult. For that reason, arsenic concentration in various water sources at southeastern Bangladesh was studied.

People in Bangladesh, particularly in the rural areas, are accustomed to using groundwater from hand tube wells for long and unlike surface water, it is considered safe from bacteriological pollution. In view of the overwhelming dependence of the population on groundwater, development of existing arsenic mitigation technologies such as rainwater harvester method, four pitcher method, two bucket method, pond sand filter, removal of arsenic through aeration and sand filtration, removal of arsenic through aeration, gravel and sand filtration, arsenic and iron removal system, arsenic removal through different sealing technologies and arsenic mitigation in deep tube wells were studied in rural areas of south-eastern Bangladesh. But not any method was not succeeded due to reliable result, illiteracy of rural people, lacking of suitability and awareness of people.

Arsenic contaminated aquifers have no regular pattern, varies both horizontally and vertically within short distances and is commonly associated with fluctuating water tables. My research revealed about status of groundwater arsenic variations with respect to depth, seasons and age of tube wells. A total of 358 no. of tube well samples from the southeastern region were analyzed for arsenic contamination of ground water. MERCK Arsenic Test Kit no. 1,17926,0001 was used to determine arsenic contamination. Tube well water depth was measured with the help of avometer, measuring tap and plastic insulated wire. Also, depth and age of tube well were determined through asking question, secondary source of DPHE and Union Council. These data were particularly useful because arsenic testing in this study was done using standard testing method, which provide reliable results. It indicates that arsenic concentration level is about 41.31% under WHO guideline (0.01 mgl^{-1}), 29.69% is within permissible level (0.05 mgl^{-1}) and 29% exceeds the permissible level (> 0.05 mgl^{-1}). Arsenic concentrations in monitored wells were largely constant during the monitoring period. It demonstrates that there is no remarkable relationship between arsenic concentration and depth of water table. None of the shallow wells showed any seasonal response in As to rainfall. Among the research results, highest arsenic concentration (about 1310 microgram l^{-1}) was observed in 150 ft water table depth. There is no any significant relationship among

arsenic concentration, depth of water table and wet season. Less arsenic concentration was observed during dry season. Highest arsenic concentration (300 microgram/liter) was observed for the depth of 140, 150 and 165 ft respectively. From 120 to 160 ft arsenic concentration varied from 10 to 100 microgram/liter. Average concentrations for arsenic in tube wells grouped by age were calculated for all the tube wells and also for the depth segregated groups. It can be seen that for all the groups except >1000ft, the concentrations do not appreciably change with tube well age. The age of the tube wells had little or no bearing on the overall level of arsenic contamination.

Three-pitcher and water hyacinth plant was used to remove arsenic as a low cost technique. Hyacinth plant was used both for arsenic contaminated groundwater and artificial arsenic solution as a phytoaccumulator. A simple three-pitcher (locally known as '3-kalshi') filtration assembly made entirely from readily available local materials is tested for its efficacy in removing arsenic from the groundwater of Bangladesh. In a 3-kalshi assembly, the first kalshi has gravel, coal and coarse sand, the second kalshi has coarse and fine sand and the third kalshi is the collector for filtered water. Coal was used to absorb organic impurities that were present in arsenic contaminated groundwater. Silver diethyldithiocarbamate (SDDC) method was used to know the exact value of arsenic removed through three-pitcher filtration performed analytical measurements. The filtered water remained crystal clear for months and free from most toxic metal ions. From my results it was observed that level of arsenic was removed from about 0.8 mg/l to 0.02 mg/l (Below Bangladeshi standards). I suggest the use of this simple setup to make potable water. Water hyacinths (*Eichhornia crassipes*) plant remove arsenic remarkably both from artificial arsenic solution and arsenic contaminated groundwater. To test this, I set up two Chari. One with water hyacinths in distilled water as a control and one with water hyacinths in a 300 parts per billion (ppb) arsenic solution, a typical Bangladeshi drinking water (groundwater) concentration. I tested the arsenic concentration of the water over multiple trials using a hach colorimetric test kit. The water hyacinths reduced the arsenic level from 300 ppb to 0 ppb in the first trial (24 hours). To start each subsequent trial, I brought the arsenic level back up to 300 ppb. The same plants reduced the arsenic level to 50 ppb in the second trial and continued to reduce the arsenic level for three more trials, but only down to 70 ppb. After the fifth trial, the plants, though showing no significant deterioration in health, lost their ability to remove any arsenic. Again, I put water hyacinth in a Chari filled with arsenic contaminated water (0.8 mg/l) one month to reduce level of arsenic. Also, I determined where in the plants the arsenic was stored. I tested for arsenic in water extracts from the roots, stems and leaves of the plants. All laboratory method was performed by SDDC method to determine arsenic content in root, bladders leaves of hyacinth plant. About 40-50% arsenic was removed through water hyacinth from arsenic contaminated ground water. Most of the removed arsenic was absorbed by roots, then bladders and then leaves of hyacinth plant.

Since arsenic in groundwater was recognized as a potential threat to human life, much effort has been directed towards ensuring safe drinking water either through mitigation techniques or through finding alternative water sources. Even when alternate sources of water are used for potable purposes, continued cropping of long-term irrigated soils subjected to As contaminated ground water will pose significant risk to animal and human health through soil-crop transfer of As. In agriculture, there are also serious implications from the possible transfer of arsenic into the food chain through crops that are under irrigation with arsenic-contaminated water and then consumed by humans. Four Leafy vegetables like as Red amaranth, Stem amaranth, Spinach and Indian spinach were planted in tub/tob on same soil sample. About 0.8-mg/l arsenic contaminated ground water was used for irrigation. Due to effect of arsenic contaminated irrigation water, Indian spinach was not germinated well and not analyzed. After 8 weeks, rest of three vegetables plants were picked up and dried as well as chopped by knife and grinded after drying. The grinded samples were analyzed by atomic spectrophotometer (HG-AAS) following USEPA method 1632. In existing soil, 0.039-mg/l arsenic was accumulated in each four tub and leaves of Red amaranth, Stem amaranth and spinach were taken up 0.026, 0.022 and 0.017 mg/l arsenic respectively.

Chapter 1

GENERAL INTRODUCTION

1.1 RESEARCH BACKGROUND

Bangladesh has a network of hundreds of rivers and tributaries and thousands of ponds, wells and other bodies. Before the introduction of tube-wells, people did use surface water, the sources being rivers, canals, lakes and manmade water reservoirs, like dug-wells but the surface water in Bangladesh is mostly contaminated, not with arsenic but water borne diseases. Then the countrywide program of installing tube-wells came in. Tube-wells of course are free from water borne diseases and this did prevent diarrhoeal diseases. Now they are a source of causing the massive problem of arsenic contamination through out the country. Presence of high concentration of arsenic in tube well water has become a major concern in Bangladesh in recent years. The arsenic contamination in irrigation and drinking water is now one of the most vital problems in Bangladesh. Reports on arsenic contamination of groundwater in the west Bengal state of India bordering eastern Bangladesh was first published in 1983. Department of Public Health Engineering (DPHE) at Chapai Nawabgang district detected awareness about the presence of arsenic in Bangladesh at first in 1993. Arsenic has contaminated in the ground water of 61 districts out of 64 districts of Bangladesh. At present about 35 million of people of Bangladesh are at high risk due to consumption of arsenic contaminated water. Southeastern region of Bangladesh is one of the worst arsenic affected areas of Bangladesh. High concentration of arsenic in ground water has created serious problem for irrigation and drinking water in recent years. Specialists and Scientists suspect that the arsenic contamination in irrigation and drinking water in Bangladesh is probably the largest mass poisoning case in the world now. Some specialists opine that the arsenic problem in ground water particularly in southeastern region of Bangladesh is probably due to arsenic rich deltaic deposition and hydraulic connection with contaminated region of West Bengal province of India. There are many reports of death and serious infection relating arsenic poisoning. Recently it has been estimated that 30 to 40 percent of Bangladesh may have arsenic contaminated groundwater and as many as 29 million people could be at serious risk from arsenic poisoning (http:// phys4.Harvard.edu/~wilson/Feroze_Ahmed/Sec_2.htm). Water samples collected from the arsenic infected areas of the country showed that 28% of the affected people had more than 100 to 1500% arsenic in their urine (normal level: 0.005-0.04 mg/day), 47% had 8 to 20% in their nails (normal level: 0.043-1.08 mg/kg), and 98% had 100 to 15000% more than normal level of arsenic in their skin (normal level: 0.466-0.896

mg/kg)(Marimuthu 2001; Chowdhury 2000: Karim 2000). Therefore an urgent approach is needed to encounter the problem of arsenic contamination.

1.2 DESCRIPTION OF ARSENIC CONTAMINATION

1.2.1 What is arsenic?

Arsenic is a natural occurring element, usually geologically bound in sediments and rocks with a particular affinity to those bearing iron. In some conditions, such as for example oxidizing or reducing conditions, arsenic may be released from the parent compound in minute quantities. Arsenic cannot be seen, tasted or smelled when it is present in water. The maximum acceptable level in Bangladesh as per requirement of Bangladesh standards for drinking water is 0.05 mg/l (GOB 1997) and the provisional WHO guideline value for arsenic in drinking water is 0.01 mg/l (WHO 1999).

1.2.2 Occurrence of arsenic

Arsenic occurs naturally in rocks, soil, water, air, plants and animals. Natural activities such as volcanic action, erosion of rocks and forest fires can release arsenic into the environment. Industrial products containing arsenic include wood preservatives, paints, dyes, pharmaceuticals, herbicides and semiconductors. The man-made sources of arsenic in the environment include mining and smelting operations, agricultural operations, burning of fossil fuels and wastes, pulp and paper production, cement manufacturing and former agricultural uses of arsenic (U.S. EPA. 2000).

Arsenic is a naturally occurring poisonous chemical element and always present as compound. It is widely distributed in the soil profile as component of different minerals and found in nominal amounts in all organisms. Arsenic is a partially metallic substance and always present as compounds. It occurs naturally in rocks, soil, water, air, plants and animals. Natural activities such as volcanic action, erosion of rocks and forest fires can release arsenic into the environment. Industrial products containing arsenic include wood preservatives, paints, dyes, metals, pharmaceuticals, pesticides, herbicides, soaps and semiconductors. Man-made sources of arsenic in the environment including mining and smelting operations, agricultural applications and the use of industrial products and disposal of wastes containing arsenic (U.S. EPA. 2001).

Arsenic occurs widely in aquifers in deltaic sediments near mountain uplift zones and in deep sandy aquifer layers originating as riverine, lake or coastal deposits. Arsenic also occurs as a contaminant produced as a result of mining and manufacturing industries. Organic arsenic is generally less (about ten- fold) toxic than inorganic arsenic. The commonly existing inorganic As-species in groundwater are in the form of arsenate (As v) and arsenite (As III), the later being more mobile and toxic (40-60 times) for living organisms. Arsenic occurs in Bangladesh as geological deposits at a shallower depths

(usually at 40- 150 feet). Generally shallow tube well water is found heavily contaminated by arsenic ranging from 0.01-0.03 mg l^{-1}.
It is now generally agreed that the arsenic contamination of groundwater in Bangladesh is of natural origin, deriving from the geological strata underlying Bangladesh (Elizabeth 2000). It is also suggested that this arsenic is transported by rivers from the sedimentary rocks in the Himalayas (NWMP, 2000).
The arsenic is thought to be closely associated with iron oxides. Arsenic occurs in two oxidation states in water. In reduced (anaerobic) conditions, it is dominated by the reduced form: arsenite. In oxidizing conditions the oxidized form dominates: arsenate. The release mechanism of arsenic from the sediment is not yet clearly understood, but there are several theories relating to how arsenic has been mobilized in to the groundwater. According to the United Nations Foundation there are two leading theories (pyrite oxidation and oxyhydroxide reduction) but it is the oxyhydroxide reduction theory that is gaining credence. Both processes may also act in association.

Pyrite oxidation theory: Due to extraction of groundwater, air and water containing significant dissolved oxygen is drawn underground where it reacts with arsenopyrite minerals. This oxidation reaction releases arsenic into the groundwater. Thus the increased use of shallow tube wells is implicated as a cause of the current arsenic problem.

Oxyhydroxide reduction theory: Due to a high level of natural, arsenic-rich organic material buried in the sediments over time iron oxyhydroxides are being reduced and releasing the arsenic. This theory implies that the release of arsenic into groundwater has not been triggered by human action.
The second theory is thought to be more likely explanation. However, some other hypothesis like microbial activity and the effects of deposition via arsenic contaminated fertilizer have also been hypothesized.
Natural processes of groundwater flushing will eventually wash the arsenic away but this will take thousands or ten thousands of years. The flushing is particularly slow in the Bengal Basin in general because it is so large and flat.

1.2.3 Natural origin of arsenic

Groundwater pollution by arsenic is often a natural phenomenon attributed to subsurface sediments containing small amount of arsenic. The arsenic remains fixed in the sediments as long as the groundwater contains sufficient dissolved oxygen. However, arsenic is released from the sediments if these come into contact with oxygen-depleted groundwater. Oxygen depletion in groundwater is often caused by decomposition of organic material (e.g peat), which is highly abundant in soils of tropical river deltas. This natural process leads to arsenic contamination of groundwater in, for example, the Red River Delta (Vietnam) (Berg et al.2001) and Bengal Delta (Bangladesh and West Bengal)(Tran et. al.2003).

1.2.4 Theory of arsenic presence in groundwater

The most commonly accepted theory on the presence of arsenic in groundwater's postulates anoxic dissolution of iron (hydr) oxides and release of previously adsorbed arsenic (Nickson et al. 2000; Smedley and Kinniburgh 2002). The arsenic in the sediments and groundwater of the Red River Delta originates from the mountains in the catchments of the Red River, and has been deposited during thousands of years (Tong 2002). Mountain erosion leads to release of rock-forming minerals and arsenic into hydrosphere. Eroded iron turns to rust, iron (hydr) oxides and forms particles as well as coating and adsorbed arsenic are washed into rivers and transported downstream. River water with high loads of particles generally exhibits characteristics red to yellowish brown colour caused by the iron, a phenomena that gave the Red River its name. Arsenic is thus brought to the river deltas bound to sediment particles and deposited in the soil with the settling particles.

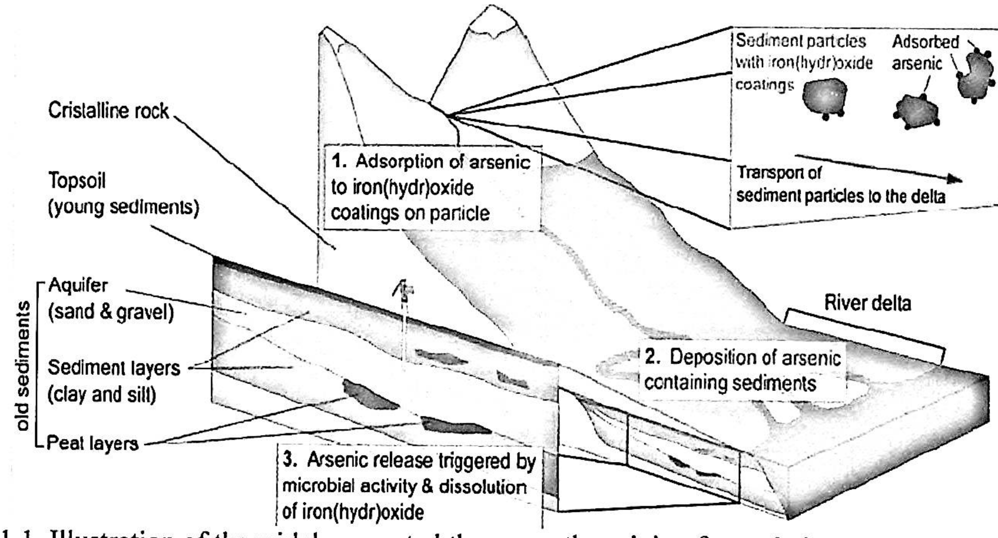


Figure 1.1. Illustration of the widely accepted theory on the origin of arsenic in groundwater.

In the flat lowlands of the river delta, suspended particles are deposited during floods. This was the case particularly in ancient times when the flow of the river water was not yet controlled by dykes. For thousands of years, deposits of river sediments have created the soil layers (sediments) that from the entire data, as it is known today. These sediments reach more than a hundred meters below the today's topsoil layer. Arsenic adsorbed on the surface of sediment particles is thus buried in the structure of the delta underground. The Red River Delta was formed by sediment layers deposited in the last ~ 10, 000 years (Figure 1.1).

1.2.5 Arsenic chemistry and the difficulty of measuring arsenic

Arsenic has the ability to switch between two valency forms, As^{3+} and As^{3-}. The three-valent form (As^{3+}) is more soluble and more likely to be absorbed than the five-valent form (As^{5+}). This switching property makes detection, measurement difficult and frequency unreliable. Measuring arsenic at low concentration is not simple and therefore needs to be undertaken with care by trained staff and with proper quality control measures in place. Any method designed to measure arsenic in only one form will commonly not detect its tool presence or concentration [usually expressed as micrograms per litre (µg/L) or parts per billion (ppb)].

Arsenic is a metalloid or inorganic semiconductor. It occurs with valence states of –3, 0, +3 (arsenite, As ([III]), and +5 (arsenate, As [V]). Because the valence states –3 and 0 occur rarely, this discussion of arsenic chemistry focuses on As (III) and As (V). Arsenic forms inorganic and organic compounds. Inorganic compounds of arsenic include hydrides (e.g., arsines), halides, oxides. acids, and sulfides. The most common arsenic mineral is arsenopyrite, which often occurs as impurity in arsenic-rich sulphide ores. Some common forms of arsenic are shown in Table 1.1.

Table 1.1 Common forms of arsenic

Name	Chemical formula	Form	Other names
Arsenic	As	Silvery metallic solid which quickly tamishes to dark grey or black	
Arsenate	AsO^{3-}_4	White soild as sodium and potassium salts	
Arsenite	AsO^{3-}_3	White soild as sodium and potassium salts	
Monomethyl arsonic acid	$H_2As(CH_3)O_3$	White solid	MMA
Dimethylarsinic acid	$HAs(CH_3)_2O_2$	White solid	DMA, cacodylic acid
Arsine	AsH_3	Colourless gas	
Trimethyl arsine	$As(CH_3)_3$	Colourless gas	Gosio's Gas
Arsenobetaine	$HAs(CH_3)_3CH_2CO_2$	White solid	Fish arsenic
Realgar	AsS	Red solid	Sandarach
Orpiment	As_2S_3	Yellow solid	Arsenikon
Arsenic trioxide	As_2O_3	White solid	White arsenic, Hatten-rauch, Hidri
Copper arsenite	$CuAsHO_3$	Green solid	Scheele's green
Copper acetoarsenite	$3CuO.As_2O_3.Cu[Ooc.CH_3]$	Green solid	Emerald green

1.2.6 Mechanism of arsenic contamination in groundwater

Presently, there are two well-known theories about the mechanism of arsenic contamination in groundwater. These are oxidation and oxyhydroxide reduction theory. The oxidation theory is so far the accepted theory. According to this theory, arsenic is released from the sulfide minerals (arseno-pyrite) in the shallow aquifer due to oxidation (Mandal et. al 2000). The lowering of water table owing to over exploitation of groundwater for irrigation has initiated the release of arsenic. The large-scale withdrawal of groundwater has caused rapid diffusion of oxygen within the pore spaces of sediments as well as an increase in dissolved oxygen in the upper part of the groundwater. The newly introduced oxygen oxidizes the arseno-pyrite and forms hydrated iron arsenate compound known as pitticitic in presence of water. This is very soft and water soluble compound. The light pressures of tube-well water break the pittictic layer into fine particles and make it readily soluble in water. Then it seeps like drops of tea from teabag and percolates from the subsoil into the water table. Hence, when the tube-well is in operation, it comes out with the extracted water. This mechanism is portrayed in Figure 1.2.

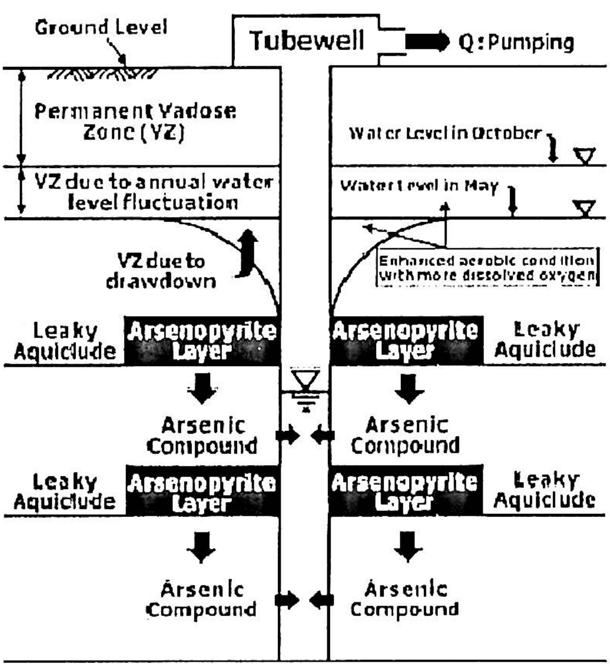


Figure 1.2 Aerobic Conditions in Groundwater around a Heavy-Duty Tube-Well

The alternative hypothesis on the arsenic contamination is the oxyhydroxide reduction theory proposed by Nickson et.al. (2000). Some scientists and researchers have accepted this theory recently as main the process for mobilization of arsenic in groundwater.

According to this theory, arsenic is derived by desorption from ferric hydroxide minerals under reducing conditions. Ferric hydroxide minerals are present as coatings in the aquifer sediments. In anaerobic groundwater, these sedimentary minerals release its scavenged arsenic.

1.2.7 Sources and/or causes of arsenic contaminations in Bangladesh

In different countries arsenic contamination in water supplies is known to have been caused by dissolving naturally occurring geological deposits from industrial discharges, and from application of pesticides. But, there are several speculations about the sources of arsenic contamination in Bangladesh, such as:

- Wooden electric poles of Rural Electrification Board (REB), which are, treated with chromated copper arsenate (CCA), ammoniacal copper arsenate (ACA) and ammoniacal copper zinc arsenic (ACZA).
- High use of fertilizers and pesticides, particularly phosphate fertilizers;
- Role of microbes in the aquifers;
- Reduction of iron oxyhydroxides; etc. Gradually, all these speculations were rejected based on the field observations and situation analyses.

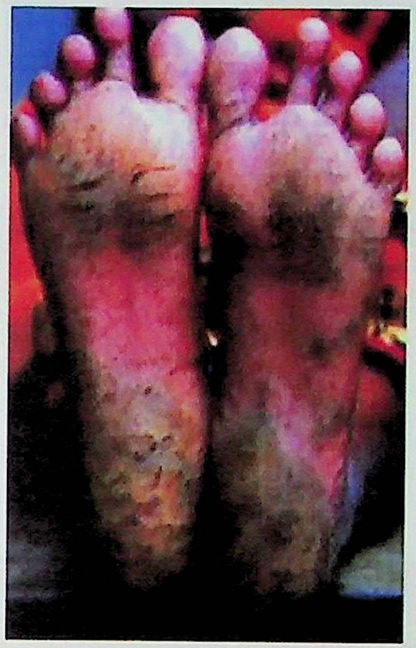
Finally it was recognized that only the shallow and hand tube well (STW & HTW) water contained high concentration of arsenic. A survey indicated that this high concentration is restricted in the upper alluvial sediments, usually at 40-150 feet depth. This indicates that the source of arsenic in Bangladesh is naturally occurring geological deposits.

1.2.8 Health hazard of arsenic

Groundwater extracted from arsenic contaminated aquifers in the worst affected areas of Bangladesh by shallow tubewells is considered no longer safe for drinking and cooking purposes. Arsenic in groundwater is predominantly in two inorganic forms- inorganic pentavalent arsenic (As^{5+}), and trivalent arsenic (As^{3+}). Of these As^{3+} is the most toxic to humans, near about 20 times more than the As^{5}. Generally arsenic III (As^{3+}) is found in anaerobic condition and arsenic V (As^{5+}) is in aerobic condition. Trivalent arsenic (As^{3+}) is also the most difficult to remove chemically from water. It is therefore also important to investigate the occurrence of these forms in water so as to determine the real toxicity and find ways of removing it.

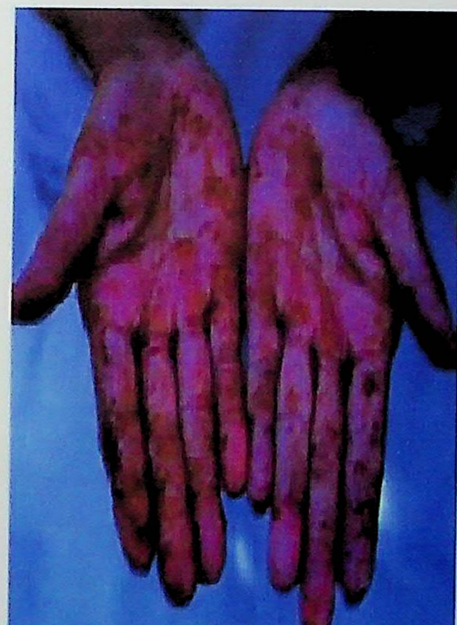
Arsenic tends not to accumulate in the body but is excreted naturally. If ingested arsenic is faster than it can be excreted, arsenic does accumulate in hair and fingernails. Early symptoms can range from the development of dark spots on the skin, to a hardening of the skin into nodules - often on the palms and soles. As a result different disease like as Melanosis, Keratosis, Uleer, Gangrene, Non-pitting oedema and Conjunctivitis occur (Figure 1.3). The World Health Organization (WHO) estimates that these symptoms can take 5 to 10 years of constant exposure to arsenic to develop. Over time, these symptoms can become more pronounced and in some cases, internal organs including the liver, kidneys and lungs can be affected. In the most severe cases, cancer can occur in the skin and internal organs, and limbs can be affected by gangrene. While evidence links arsenic with cancer, it is difficult to say how much exposure and for what period of time, will result in cancer.

Photograph 01: Melanosis.

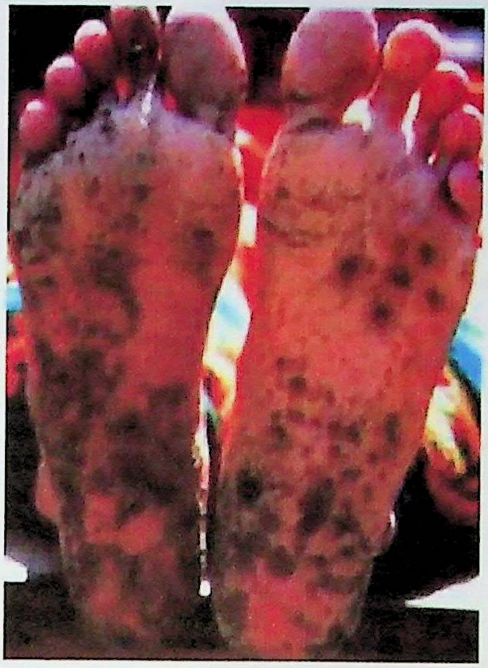


Photograph 02(a): Keratosis.

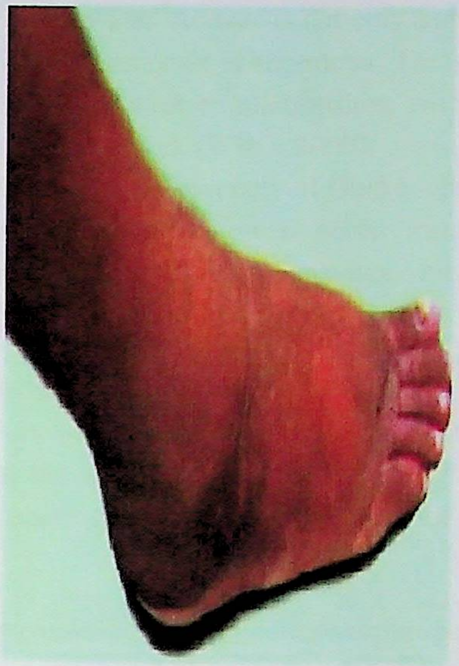
Photograph 02 (b): Keratosis.



Photograph 02 (c): Keratosis.



Photograph 03: Ulcer.



Photograph 04 : Gangrene.

Photograph 05: Non - pitting oedema.

Photograph 06: Conjunctivitis.

Figure 1.3: Photographic view of some arsenic affected disease

1.2.9 Social costs of arsenic contamination

While addressing the problem of arsenic contamination, emphasis is being put on the identification, mitigation and supply of safe drinking water. Arsenic is not only a physical but also a social phenomenon, the social fallout of arsenicosis is enormous. The arsenic hazard has a strong social dimension, affecting issues such as relationships within the family and the village, as well as the mental health of the sick (WHO, 2000).
Dr. Mahbuba Nasreen from the Department of Sociology, University of Dhaka, observed the social cost of arsenic contamination in the following forms: social instability, superstition, ostracism, marital problems, discrimination against women, increased poverty, diminished working ability and death (Mahbuba Nasreen, 2002).

Social Instability: Lack of proper knowledge about arsenic contamination and unavailability of arsenic-safe drinking water as well as proper treatment are creating instability in the social life of the people in the arsenic-prone areas of Bangladesh. Moreover, social conflict over contaminated water contributes to destruction of social harmony and network relationships.

Superstition: Superstitions and prejudices are constructed surrounding arsenic patients. For example, in the northeastern district of Kushtia, arsenic is considered as a "curse of Allah" or the work of evil spirits (The Bangladesh Observer 2000). People stay away from arsenic victims, neglect them, or become scared of them because of these superstitions.

Ostracism: Arsenic patients are often identified by the society as patients of leprosy and as a result they remain ostracized, at either the household or the village level. Children of arsenic patients are not allowed to attend social or religious functions. The patients as well as their close relatives are not allowed to use public tube wells and village ponds. Often family members, like households or wives, abandon the arsenicosis victims. The problem is more serious in the case of children (Bidyat et. al.2000). The entry of arsenic affected children into schools becomes restricted. Some may be denied the opportunity to go to school. They also are subject to social ostracism by their friends and classmates.

Diminished working ability: Arsenic is a silent killer. The black spots on a victim's body slowly become nodules and even grow if the victim remains exposed to arsenic contamination. Limbs and internal organs like the liver, kidney and lungs may be affected. Gangrene cripples the victim and makes him or her unable to do hard labor. As a result a person lose his working ability.

Marriage related problems: Arsenic has an adverse impact on marital relationships. People are reluctant to develop marital relationships with families whose members suffer from arsenicosis. This has caused serious anxiety for parents of unmarried adult children. Many women are divorced or abandoned by their husbands due to arsenicosis.

Increased poverty: Those in poverty are the main victims of arsenic contamination as they are compelled to drink contaminated tube well water. Researchers believe that the severity of arsenicosis is very much related to nutritional deficiency. Malnutrition makes them easy victims. Due to poverty, victims are deprived of proper treatment. When seeking treatment, the costs become a burden to them. As arsenicosis decreases the victim's ability to work, he or she often suffers from a reduced income. Due to ostracism, arsenic patients lose their jobs. Thus, arsenic negatively contributes to the poverty situation in Bangladesh.

Gender implications of arsenic contamination: In Bangladesh women perform the majority of the household work, but their work remains relatively invisible and unrecognized in society. Among many other tasks, collecting and carrying water for household use, particularly in the rural areas, is the responsibility of women and girls. Arsenic contamination in nearby drinking water often compels them to collect and carry water from a long distance, imposing an additional burden on them. Because of socio-cultural restrictions, women often do not receive opportunities to obtain information from outside sources. Thus, they are not properly aware of the danger of arsenic. This makes arsenic mitigation activities difficult. Women are frequent victims of ostracism due to arsenicosis. They are doubly vulnerable: from the disease itself and by being divorced, abandoned, or even forced out of the society. As gender discrimination exists in many forms in the patriarchal society of Bangladesh, women suffer more from these things than men.

1.2.10 Geographical distribution of arsenic concentration in Bangladesh

There is a distinct geographical distribution of arsenic with the greatest concentrations in the south and southeast and the smallest concentrations in the north and northwest of Bangladesh. This can be seen in a map of the point-source data (Figure 1. 4) but the regional trends are more clearly seen in the smoothed map (Figure 1.5). This smoothing was carried out by a statistical technique (disjunctive kriging). The high arsenic region in the south and east of Bangladesh is clear from this map. In the arsenic point source map, the lowest concentration class symbols have been plotted first, then the symbols for the next lowest class and so on. Therefore where there is some overlap of symbols, the higher concentration symbol will fail on top of the lower symbol and will tend to dominate the map.

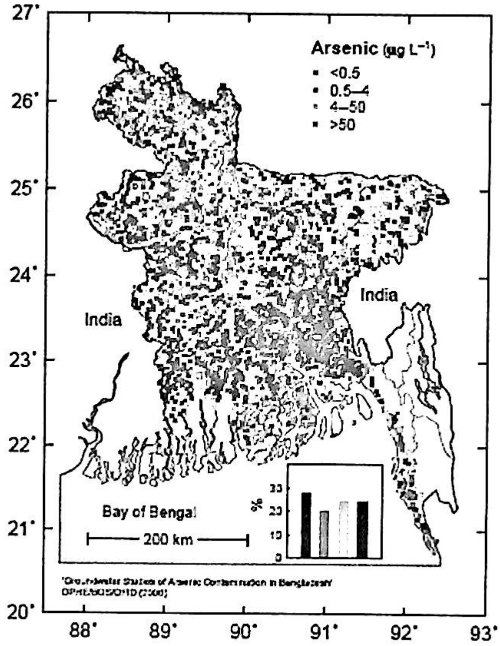


Figure 1. 4.Map of point source arsenic concentrations observed in groundwaters in the national Hydrochemical Survey. Inset shows the percentage frequency of each of the indicated class intervals (bar colours match those of the map symbols).

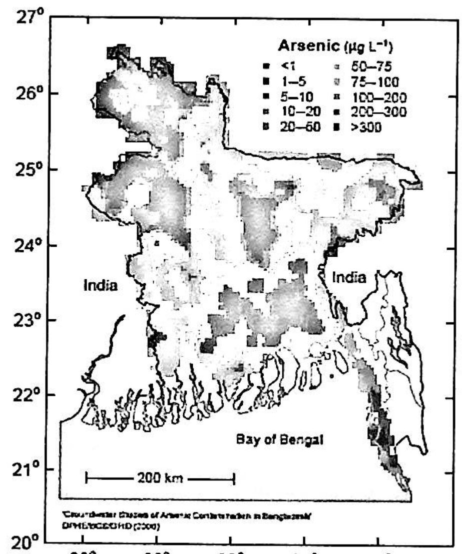


Figure 1.5. Map of smoothed groundwater arsenic concentrations from the National Hydro chemical survey. Smoothing was carried out by disjunctive kriging

From both Figure 1.4 and 1.5, it can be clearly visualized that there was a distinct regional pattern of arsenic contamination with the greatest contamination in the south and southeast of the country and the least contamination in the northwest and in the uplifted areas of north-central Bangladesh. However, there is occasional arsenic 'hot spots' in the generally low-arsenic regions of northern Bangladesh. In arsenic-contaminated areas, the large degree of well-to-well variation within a village means that it is difficult to predict whether a given well will be contaminated from tests carried out on neighboring wells. The problem is acute in tube wells abstracting ground water from 10 m to 100 m depths in the southeastern region of Bangladesh. It was estimated that more than 20 million people drink water exceeding the national standard for arsenic levels. The presence of arsenic is naturally occurring phenomenon, but prolonged use of the water can be very harmful when the levels cross the standard limit. Arsenic contamination has considerably serious implications for groundwater abstraction in different areas. This impinges on domestic water supply, since groundwater is preferred source because compared to surface water it is less likely to be fecally polluted.

The source of-and perhaps the solution to-Bangladesh's arsenic problem lies under the ground. The nation is largely a delta, formed by silt deposited over 250 million years by two great Himalayan Rivers, the Ganga and the Brahmaputra. In some areas, the sediment layer is as much as 20 kilometers deep. Most of the poisoned aquifers are shallow, however, from 10 to 70 meters deep, and lie to the south and southeast of the country. The BGS notes that around 18,000 years ago, when the sea level dropped by around 100 meters, the rivers cut deep channels into the existing sediment. In later years, these valleys filled up with gray clay that seems to hold the position. Older, brown alluvium, such as in the northwest or the hilly regions, is less contaminated.

1.2.11 Arsenic concentration in different water source in southeastern Bangladesh

The Table 1.2 displays arsenic content in surface water, irrigation tube well water (STW), drinking water (HTW) and surface water in some areas of southeastern Bangladesh. In Jhenaidah district, trend of arsenic contamination in different water source was

Table 1.2: Arsenic contamination in various water sources

District	Arsenic (microgram/liter) STW	Arsenic (microgram/liter) HTW	Arsenic (microgram/liter) Surface water
	157.54	131.61	2.55
Barisal	115.21	255.06	3.20
	102.48	256.00	4.34
	87.59	3.94	5.19
	54.64	105.15	1.86
	6.93	3.22	5.36
Faridpur	238.51	3.83	0.93
	151.14	1.65	1.44
	103.95	16.36	3.11
	147.77	0.88	0.80
	88.23	482.48	7.90
	76.54	551.93	2.91
Chuadanga	242.98	48.75	5.77
	1.68	102.42	4.63
	13.49	312.66	1.20
	7.84	1.09	5.11
	36.95	1.02	1.68
	215.29	33.50	2.97
	13.19	1.15	1.62
	14.94	0.47	3.24
	11.89	15.48	2.83
Jessore	20.77	96.43	2.83
	11.26	51.39	7.90
	81.44	38.37	3.11
	40.59	0.74	3.98
	93.26	0.52	7.00
Jhenaidah	13.30	0.50	2.07
	53.63	45.55	2.98
	5.20	14.64	3.14
	51.91	46.87	3.26
	39.01	183.29	5.63

Source: CIMMYT, Bangladesh

comparatively low as compared to other district. Data on water analysis from the arsenic affected areas were accomplished during Boro season. It can be observed that arsenic concentration varied widely within different district of STW and HTW. But in case of surface water arsenic concentration is always low in different southeastern district of Bangladesh. The arsenic content in surface waters (ponds, rivers, etc.) was found to be negligible in the arsenic problematic areas. In such areas, wherever possible, use of surface water for irrigation could be a tool to avoid arsenic contamination of the soils and crops.

1.3 RATIONALE OF THE STUDY

Giving the magnitude of the catastrophe, a resource constrained nation like Bangladesh is now struggling to cope with the new problem of arsenic contamination. The lack of awareness and dissemination of proper and rapid information has further deteriorated the problem of arsenic contamination. The arsenic concentrations variations with respect to depth and season, arsenic content in different water source, arsenic removal by water hyacinths, determination of arsenic content in various parts of plants and amount of arsenic contaminated soils that are irrigated with arsenic laden water and arsenic in the food chain can be an effective way for study about arsenic contamination.

1. 4 OBJECTIVE OF THE PRESENT RESEARCH

The objectives of this study were to evaluate arsenic contamination in groundwater in Bangladesh.

The specific objectives of this study are as follows:

1. To reviews the concepts of arsenic contamination in the context of disseminating information for arsenic contamination.
2. To evaluate arsenic content in different water source at southeastern Bangladesh.
3. To determine arsenic contamination variations with respect to depth, seasons and tube well age.
4. To develop techniques for removal of arsenic from contaminated groundwater.
5. Study about arsenic contamination in the food chain.

Chapter 2

REVIEW OF LITERATURE

The main aim of this chapter is to review of the results of some of the previous studies that are related to the present research work. Some of the pertinent literature is reviewed here.

Khan (1997) and Dhar (1998) reported that Bangladesh is grappling with the largest mass poisoning of a population in history because groundwater used for drinking has been contaminated with naturally occurring inorganic arsenic. It is estimated that of the 125 million inhabitants of Bangladesh between 35 million and 77 million are at risk of drinking contaminated water.

Sengupta (1997) stated that hyper accumulators are plants that accumulate metals within their biomass in higher concentrations than the concentrations in their resident soil.

UNICEF (1999) reported the crisis has its roots in another worthy effort to fight water-borne diseases that had impacted this tropical region for a long time. Acute health problems, such as gastrointestinal diseases and infant mortality, were attributed to drinking microbiologically contaminated surface water. It was widely believed that using groundwater would easily circumvent the problem because groundwater at certain depths is not exposed to microbiological contamination. It is now known that the alluvial aquifer that underlies the Ganges-Brahamputra river basin contains arsenic in mineral form. During the past two decades about four million wells have been installed to utilize the groundwater from shallow aquifer layers, typically less than 200m deep.

Anwar (2000) described in Bangladesh and India exist controversy on the mobility of arsenic concentration in ground water. But on the source of arsenic contamination in ground water all scientists home and abroad generally agree sources from Himalayan or mining region of India.

Smith et al. (2000) observed the groundwater pollution caused by arsenic in a number of Asian countries has led to a major environmental crisis. Some recent estimates indicate that more than 35 million people in West Bengal (India), Nepal and Bangladesh are potentially at risk from drinking arsenic-contaminated water.

Huq et al. (2000) stated that widespread exploitation of groundwater from the BDP aquifer system, which began in the 1970s, currently supplies drinking water for a population of more than 150 million. Moreover, groundwater is also used for extensive cultivation of rice and other crops that has led to creation of other pathways for As ingestion among the people living in the region.

Rahman et al. (2001) stated that exploitation of groundwater from these wells for drinking water and irrigation purposes has resulted in mobilizing the arsenic.

Welch and Stollenwerk (2003) observed that naturally occurring As in groundwater of sedimentary aquifers has emerged as a global problem. In the sedimentary aquifers of the Bengal Delta Plain (BDP) in Bangladesh and neighboring West Bengal, India, As is mobilized in groundwater by natural processes, which is an issue of major environmental health concern (Also BGS and DPHE, 2001; Bhattacharya et al., 2002).

Huq et. al. (2001) stated that adsorption-desorption of arsenic onto soil is key to the understanding of its fate in the environment. In irrigated agricultural land, water lost by evaporation and evapo-transpiration will leave arsenic along with other minerals in the topsoil. The arsenic is not likely to be dissolved or washed out by flood or rainwater in oxidized condition due to its affinity for iron, manganese, aluminum and other minerals in soil. As a result, a cumulative accumulation in surface soils is expected and some recent data suggest arsenic concentration as high as 83 mg/kg in topsoil, against a background concentration of 4-8 mg/kg (Also, Alam and Sattar, 2000).

Khan et. al. (2000) observed that a simple three-pitcher (locally known as '3-kalshi') filtration assembly made entirely from readily available local materials is tested for its efficacy in removing arsenic from the groundwater of Bangladesh. In a 3- kalshi assembly, the first kalshi has iron chips and coarse sand, the second kalshi has wood charcoal and fine sand, and the third kalshi is the collector for filtered water. The filtered water remained crystal clear for months and free from most toxic metal ions. The final water quality meets and exceeds the guideline values suggested by USEPA, World Health Organization and Bangladesh. Authors suggest the use of this simple setup to make potable water

UNICEF and INFS (2000) observed the results from BGS and DPHE indicated that nearly 27% of the shallow wells (<150 m) contain As concentrations above the BDWS, while 46% exceed the WHO provisional drinking water limit. Although the number of deep tube wells (>150 m deep) is relatively small, only 1 and 5% of the analyzed ground waters exceeded the BDWS and WHO limit, respectively. Analysis of some samples by UNICEF/INFS show that 23% of wells exceed BDWS and 42% samples exceed WHO limit.

Huq et al. (2001) observed a relatively higher concentration of arsenic has been found to be 20 mg/kg. Arum, a popular vegetable which is grown in wet soil near to tube wells, can have arsenic levels of nearly 150 parts per million. Arsenic in the agriculture environment can be fairly complex and a good understanding of the fate of arsenic pumped with groundwater is essential for the development of good management and remediation strategy.

Misbahuddin and Fariduddin (2002) found that just the roots of water hyacinths removed eighty-one percent from the four hundred ppb arsenic solution they were in. The entire water hyacinth plant (roots, leaves, stems, etc.) was reported in the same study to have removed one hundred percent of the arsenic, and to have done so in only three to six hours.

Amitech (2003) found the discovery of arsenic-contaminated ground water in Bangladesh has raised research questions regarding health effects and also about important sources of exposure. While drinking water has been the major focus of the research effort to date, ingestion of crops grown in soil with high arsenic content, or irrigated with arsenic contaminated water, has recently been raised as a potentially significant source of exposure to arsenic in Bangladesh.

Islam et al. (2004) stated to meet the growing demand for food, the farmers had to cultivate high yielding varieties of Boro rice, which require a large volume of irrigation water. Irrigation with arsenic contaminated groundwater increases its concentration in soil and eventually arsenic enters the food chain through crop uptake and poses long-term risk to human health.

Ingole and Bhole (2003) reported that the arsenic accumulates in the roots of the water hyacinth and not in the upper green part. If this is true, then the upper green part could be feed to cows without poisoning the cows or the people that may eat the cows.

Batcher (2004) stated that water hyacinths could tolerate extreme temperatures, pH and nutrient levels. They have been found to grow well in nutrient rich waters. Water hyacinths are common in Bangladesh, which is important for this study.

Chapter 3

MATERIALS AND METHODS

3.1 SELECTION OF THE STUDY AREA

The studies as well as visits were conducted in Achintyanagar in Jhenaidah District, Chandpur District, Rajnagar, Bankabarsi, Dourbadanga in Jessore district and Faridpur district. These districts were selected for the following reasons:

1. Different national and international organizations were implementing various arsenic mitigation technologies in these areas.
2. Arsenic contamination is higher in these districts.
3. Less research work about arsenic contamination was accomplished in these areas.
4. Due to aquifer characteristics arsenic contamination trend is increasing in this region recently.

3.2 RESEARCH METHODOLOGY

This study was conducted as a combination of exploratory and evaluation-type work (Fig. 3.1).**Error!**

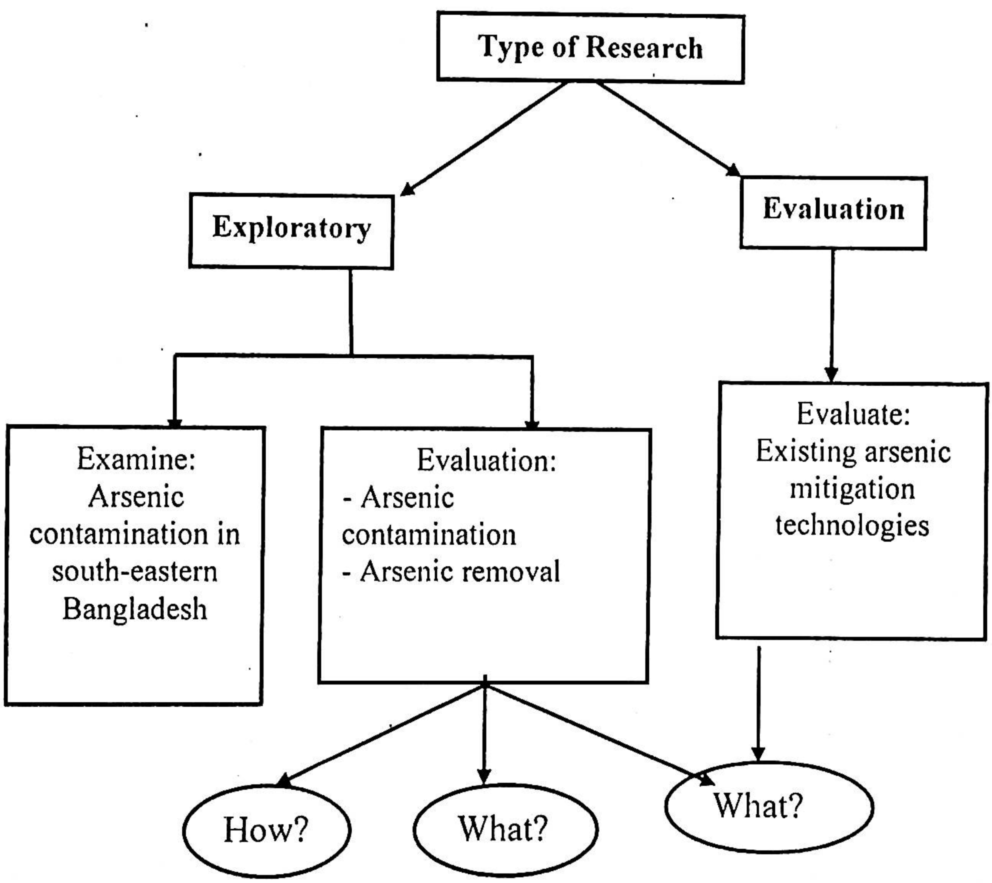


Fig. 3.1: Type of research

3.3 EXISTING MITIGATION METHODS OF ARSENIC CONTAMINATION

In highly arsenic-affected villages various projects have been gradually undertaken to supply safe drinking water by DPHE, BAMWSP, UNICEF, DANIDA, both national and local NGOs and others. Now I would like to summarize the current status and problems of mitigation measures to combat the arsenic contamination in rural areas in Bangladesh based on the findings of my own surveys and daily collection of information. During study period, I have observed the following methods as their mitigation measures.

3.3.1 Rainwater Harvester Method:

This is a round tank of about 1.8 meter high with a diameter of approximately 2 meter. The tank is made of concrete and connected with a gutter that collects rainwater from the tin roof of a house (Figure 3.2). A few neighboring families are using the rainwater

Figure 3.2: Photographic view of rainwater harvester that collects rainwater from tin roof

Harvester (RWH). UNICEF took the first initiative of the installation of the RWH without any cost sharing from the beneficiary families. Whereas, in Achintyanagar, Jhenaidah District and Chandpur District, a similar RWH (3,200 litre, Taka 9,000) was installed with 20% of the cost borne by community people and 80% financed by NGO

Forum. The weakness of this system is that people can use it only a few months after the rainy season. In Achintyanagar, two families are using this 3,200 litre RWH. If ten people of the two families drink five liter each per day, the water runs out after 64 days. After that, they have to look for another source of drinking water. Also, a plastic sheet, as shown in Figure 3.3, has been tried as a catchment for rainwater harvesting for people who do not have a roof suitable for rainwater collection. The use of land surface as a catchment area and underground gravel or sand-packed reservoirs as storage tanks can be an alternative system of rainwater collection and storage. In this case, the water has to be channeled towards the reservoir and allowed to pass through sand before entering underground reservoirs. This process is analogous to recharge of underground aquifers by

Figure 3.3: Photographic view of rainwater harvester that collects rainwater from without roof

rainwater during the rainy season for utilization in the dry season (Ahmed et.al.2002).

3.3.2 Four-pitcher method

Four Pitcher is used in this method. In the second pitcher there are charcoal and brick chips and in the third sand. Tube well water put into the first pitcher is oxidized while it flows down to the second pitcher and filtrated through charcoal, brick chips and sand. Thus arsenic- removed water is kept in the fourth pitcher (Figure 3.4). The system is based on the property that arsenic is adsorbed to iron by oxidization. Therefore, much effect cannot be expected unless groundwater is iron-rich.

Figure 3.4: Photographic view of four-pitcher method

The unit is a traditional system used for removing bacteria at a time when people used to drink pond water. After the source of drinking water was switched from a pond to a tube well, the system has been utilized to remove iron and reconsidered as an effective way to remove arsenic also since two and a half years ago.
The system does not require any chemicals. It is cheap (Taka 100) and simple. It has all the factors that can be accepted by villagers. DPHE is currently undertaking the test about the effectiveness of the system for arsenic removal.

3.3.3 Two-bucket method:

The "Two Bucket method" implemented by DANIDA. To the tube well water poured

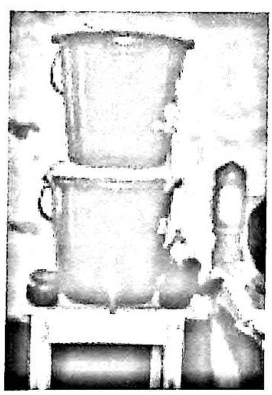


Figure 3.5: Photographic view of two-bucket method

into the red (top) bucket, potassium permanganate (oxidizing agent) and aluminum sulfate (coagulant) are added to remove arsenic from the water. The tube well water is then filtrated through the sand in the green (bottom) bucket (Figure 3.5). The arsenic concentration of 0.5 mgl^{-1} of the original tube well water was lowered to below 0.02 mgl^{-1} in this system by the measurement of a field arsenic testing kit.

3.3.4 Pond sand filter (No.1):

Figure 3.6: Photographic view of pond sand filter (No.1)

The pond sand filter (No.1) was installed by UNICEF with full financial support. Nearby there was also a deep tube well (DTW) sunk by DPHE. On visiting a household, I found that DTW water was kept for drinking and PSF water for cooking in separate kalshis in the kitchen. Here, water is taken from a relatively large pond (80 x 50 meter) for the PSF. However, villagers were using the pond for bathing and washing. It is my opinion that any pond for PSF should be kept hygiene for drinking purpose and not be used for any other purposes (Figure 3.6).

3.3.5 Pond sand filter (No.2):

The Pond Sand Filter (No.2) was constructed by the Asia Arsenic Network (AAN). Bamboo fencing protects the pond for the PSF and any use of the water other than drinking is prohibited (Figure 3.7). All the cares are taken to prevent bacterial contamination by removing a lavatory to a place more than 15 meter away and by

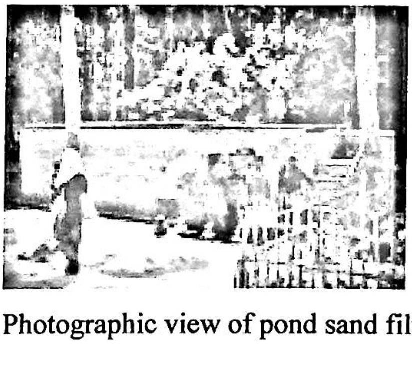


Figure 3.7 Photographic view of pond sand filter No.2

sterilizing the processed water with chlorine in the last chamber of the PSF (Figure 3.8).

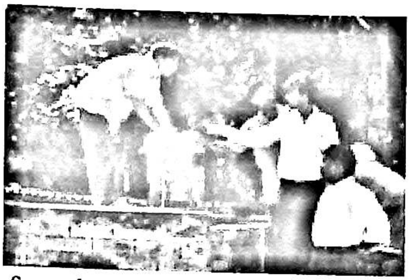


Figure 3.8 Photographic view of pond sand filter No.2 where water is processing with chlorine

A doctor of Samta Health Centre checks coliform and other bacilli once a month (Figure 3.9).

Figure 3.9: Photographic view of checking of coliform and other bacilli

The construction cost was Taka 1,50,000 for the PSF itself and Taka 2,00,000 in total inclusive of other expenses incurred for re-excavation of the pond, fencing and the removal of a lavatory. The cost was paid with the grant given by the Toyota Foundation. The PSF has the capacity for 150 households (750 people). The Samta Arsenic Prevention Committee is in charge of the maintenance and management of the pond and PSF. Since the pond water dries up toward the end of the dry season, they consider bringing water from the Betna River approximately 500 metre away to the pond. The collection of water rate is yet to be considered.

The medium-size PSF that AAN is currently promoting can supply water to 100 households (500 people) and can be constructed at a cost of Taka 1,00,000. The other expenses for re-excavation of a pond, fencing and maintenance are to be borne by the community people.

The condition for the installation of a PSF includes the replenishment of a pond of a suitable size free, the establishment of a solid group of community people for operation and maintenance and the availability of pond water throughout the year.

3.3.6 Removal of arsenic through aeration and sand filtration:

This is an arsenic removal system with aeration and sand filtration combined (Figure 3.10). The JICA Study Team on the Groundwater Development of Deep Aquifers is testing the effectiveness of the unit.

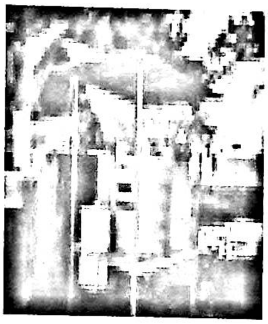


Figure 3.10 Photographic views of arsenic removals through aeration and sand filtration

3.3.7 Removal of arsenic through aeration, gravel and sand filtration:

This is an arsenic removal system with aeration, gravel and sand filtrations and adsorption by activated aluminium oxide combined (Figure 3.11). The JICA Study Team is testing the effectiveness of the unit.

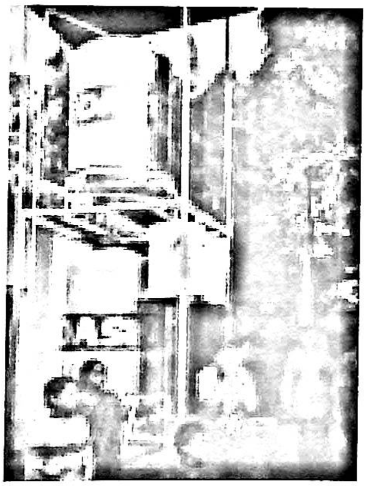


Figure 3.11 Photographic view of arsenic removals through aeration, gravel and sand filtration

3.3.8 Arsenic and iron removal system:

This is an iron removal system installed by NGO Forum at a cost of Taka 8,000 (Taka 1,600 was borne by community people). Since the system removes arsenic as well as iron at the same time, it is being introduced for arsenic removal in other affected areas. The arsenic of 0.5mg/L in the original groundwater was removed with iron and lowered to 0.02mg/L when tested with a field arsenic testing kit (Figure 3.12).

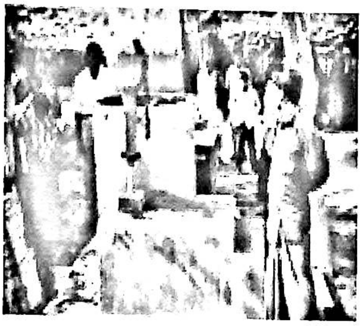


Figure 3.12 Photographic view of arsenic and iron removal system

The sand of the filtration tank is backwashed once a week, but the drained water is disposed on to the ground from the back of the unit (Figure 3.13). The arsenic of 0.4mg/L was detected from the drained water that was still on the ground. It is necessary to examine the disposal of drained water.

Figure 3.13 Photographic view of back portion of arsenic and iron removal system

3.3.9 Arsenic removal through different sealing technologies:

The JICA Study Team is conducting a monitoring research on the water quality of the three deep tubewells (222 metre deep) installed in Rajnagar, Bankabarsi, Jessore district with different types of sealing technologies; namely, cementing, nice-sealing and mechanical method. They plan to evaluate the cost, effectiveness of each sealing method, the feasibility of transferring the technologies, and other factors. Since no arsenic was detected in the pumped up water, villagers started collecting water from the deep tube well for drinking (Figure 3.14).

Figure 3.14 Photographic view of villagers are collecting water from arsenic free deep tube well

3.3.10 Arsenic mitigation in deep tube well

This is a deep tubewell (120 metre deep) installed in Gopinatpur village by DPHE. Measured by a field arsenic testing kit, it was found that the water contained arsenic of 0.3mg/L. Depending on the places, arsenic is detected at high concentrations in deep tubewell water. It cannot be said that a deep tubewell is always safe everywhere.

When I visited Dourbadanga village in the Jessore District, I saw a deep tubewell (220 metre deep) sunk by DPHE. The water sample was checked with a field arsenic testing kit and the arsenic concentration was only 0.01mg/L. However, due to the salinity, villagers were not drinking the water (Figure 3.15). A deep tubewell, as an alternative source for the mitigation of arsenic contamination, requires careful pre-installation assessment and post-installation monitoring.

Figure 3.15 Photographic view of villagers are not using deep tube well due to salinity

3.3.11 Arsenic analysis in the field:

To ensure quality of analytical work, CIMMYT was developed an analytical reference Laboratory for arsenic analysis in the Bangladesh Agricultural Research Institute (BARI-CIMMYT Arsenic Laboratory). This laboratory is playing a leading role in the development of analytical quality control protocols, standardizing analytical methods, identifying internal reference materials and assisting in routine internal quality control. The analytical reference laboratory took the leadership in creating a quality assured arsenic database that was used in understanding the nature and extent of the arsenic problem in Bangladesh and facilitating research for the development of arsenic management strategies.

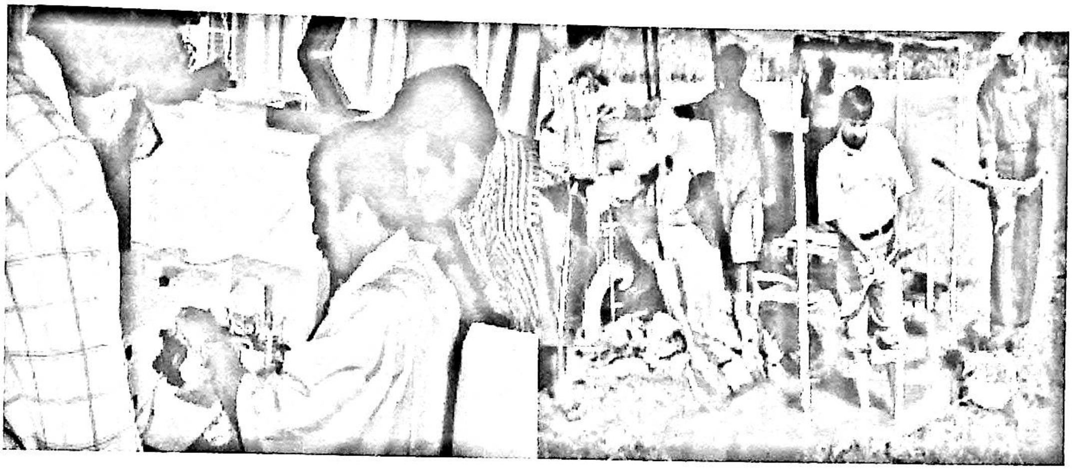


Figure 3.16: Instrumental set up and sampling (Source: CIMMYT)

3.4 CONCLUSIONS

People in Bangladesh, particularly in the rural areas, are accustomed to using groundwater from hand tube wells for long and, unlike surface water, it is considered safe from bacteriological pollution. In view of the overwhelming dependence of the population on groundwater, development of suitable treatment systems for arsenic removal from groundwater appears to a promising option for providing safe water to the rural population. Socio-economic conditions of Bangladesh demands low-cost as well as small-scale treatment systems that could be implemented in the rural areas at household or community levels. To save millions of people from arsenic poisoning it is important to detect the arsenic concentration in groundwater and also to provide a suitable, user friendly and cost effective arsenic removal process for the rural people of Bangladesh. Unfortunately, the very first step towards prevention and arsenic testing is in great chaos. The instruments are expensive and require skilled person to operate. There is no licensing authority to supervise the testing procedure in Bangladesh. At present, very few laboratories can provide reliable result. On the other hand removal technologies so far tried for the rural people have potential but not tested thoroughly for adoption. Most of the rural people are illiterate. They developed the habit of drinking hand tubewells water during the last 30 years. So any change in their behavioral needs more friendly approach and technology.

Chapter 4

SOUTHEASTERN GROUNDWATER ARSENIC VARIATIONS IN BANGLADESH: THE ROLE OF DEPTH, SEASONS AND AGE OF TUBE WELLS

4.1 INTRODUCTION

In Bangladesh, water supply, particularly the rural water supply, is almost entirely dependent on groundwater extracted from shallow tube wells. Presence of high concentration of arsenic in tube well water has become a major concern in Bangladesh in recent years. The arsenic contamination in irrigation and drinking water is now one of the most vital problems in Bangladesh. Reports on arsenic contamination of groundwater in the west Bengal state of India bordering western Bangladesh was first published in 1983. Department of Public Health Engineering at Chapai Nawabgang district detected awareness about the presence of arsenic in Bangladesh at first in 1993. Arsenic has contaminated in the ground water of 61 districts out of 64 districts of Bangladesh. At present about 35 million of people of Bangladesh are at high risk due to consumption of arsenic contaminated water. Southeastern region of Bangladesh is one of the worst arsenic affected areas of Bangladesh. High concentration of arsenic in ground water has created serious problem for irrigation and drinking water in recent years. Specialists and scientists suspect that the arsenic contamination in irrigation and drinking water in Bangladesh is probably the largest mass poisoning case in the world now. Some specialists opine that the arsenic problem in ground water particularly in southeastern region of Bangladesh is probably due to arsenic rich deltaic deposition and hydraulic connection with contaminated region of West Bengal province of India. There are many reports of death and serious infection relating arsenic poisoning. In Bangladesh most of the population does not know health effects of arsenic poisoning, even the doctors of the rural areas treat arsenic affected persons for skin disease. These range from skin lesions to cancers of the bladder, kidney, lung and skin, neurological effects, cardiovascular and pulmonary disease and diabetes. The disease may develop slowly over many years for continue intake of arsenic contaminated drinking water. This chapter presents results of arsenic contamination in the southeastern region of Bangladesh. The objective of the study are to know the distribution of arsenic level in groundwater, to detect the arsenic level in some tube wells for identifying the sources of high, medium and low arsenic content, depth wise distribution of arsenic in tube wells, amount of arsenic content in groundwater, arsenic concentration variations in different depth during dry and wet season and arsenic concentration variations with age of tube wells.

4.2 MATERIALS AND METHODS

4.2.1 Sampling procedure for drinking water

A random sampling technique was applied to select the arsenic contaminated tube wells. Each sample collected in plastic container was labeled separately with a unique identification number. It also included the information on the collection date, depth of the tube well and previously arsenic test was done or not. This was done to prevent possible contamination with other samples in the laboratory. The relationship between arsenic contamination and depths of tube well were also analyzed from collected data.

4.2.2 Procedure of tube well age determination

Tube well ages were determined in two ways. One was from the secondary source of Department of Public Health Engineering. Other was through asking question of tube well owners as well as surrounding neighbors when tube well was installed. Maximum people inform me based on war of independence (1971) as a basis of response. Such as after three years of independence and so on. At the end tube well age were verified through DPHE, Union Council and number of respondent opinion.

4.2.3 Procedure of tube well water depth measurement

Tube well water depths were measured with the help of Avometer, Measuring tap and Plastic insulated wire. A small heavy weight of rod piece was bonded tightly one end of the wire and other end of the wire was kept open to connect with Avometer. Before measurement of water depth, tube well was opened at the top of the pipe. During depth measurement, tube well was opened and then rod end of the wire entered in the pipe of the tube well up to when Avometer showed indication. It means rod end touch water level. After that with the help of measuring tap water depth was measured.

4.2.4 Role of test kits in measuring arsenic

Test kits are relatively inexpensive, portable and generally operate on measuring/observing an immediate chemical reaction. However, in practical terms, field test kits have inherent limitations to their use in isolated village situations. They require replenishment of chemical reagents so incur maintenance costs, and their results can be easily validated unless the chemical protocols that eliminate cross-contamination are adhered to strictly.

Test kits may be very good for demonstrating the presence of particular chemicals or pathogens, but currently they are not always sufficiently sensitive, or accurate for quantitative assessments. Test kit results should be regarded as initial indicators. Their main limitation is that in raw water samples many chemical reactions may be masked by others occurring in the same solution. The range of technical accuracy of test kits varies generally with heir price, but none currently on the market is sufficiently sensitive to provide the data needed to ensure particular quality standards are reached.

Arsenic's propensity to switch valency states means that As^{3+} is more likely to be indicated by test results, while the presence of As^{3+} may not be identified because its reacts more slowly. Test kits therefore commonly under-evaluate total arsenic presence. The most effective use for portable test kits is to indicate the presence of arsenic. As a general principle, these guidelines recommend that, if the test kit demonstrates the presence of arsenic, alternative safe drinking water sources need to be identified.

4.2.5 Arsenic testing procedures

4.2.5.1 Testing through MERCK Arsenic Kit

Some samples were tested using MERCK Arsenic Test Kit no.1.17926.0001. Zinc and sulfuric acid were added to compounds of arsenic (iii) and arsenic (v), arsenic hydride is liberated, which in turn reacts with mercury (ii) bromide contained in the reaction zone of the analytical test strip to form yellow- brown mixed arsenic mercury halogenides. The concentration of arsenic (iii) and arsenic (v) were measured by visual comparison of the reaction zone of the analytical test strip with the field of a color scale. Measuring range/color scale graduation were 0.00 - 0.01 - 0.025 - 0.05 - 0.1 - 0.5 mg / l $As^{3+/5+}$.

Removed 1 analytical test strip and immediately reclosed the tube. With the reaction zone first inserted the test strip about half way through the slot in the stopper of the reaction vessel. By means of the syringe, transferred 10 ml of the solution to be tested to the reaction vessel and added 2 measuring spoonfuls of reagent As–1(zinc). Rapidly added 10 drops of reagent As-2 (sulfuric acid) immediately closed the reaction vessel with the stopper and swirled gently. The sample solution was not come in to contact with the test strip. Leave to stand for 30 minute, gently swirled two or three times. Removed the test strip, briefly dip into water, shaked off excess liquid and determined with which colour field on the label the colour of the reaction zone coincides most exactly. Read off the corresponding concentration value in mg / l $As^{3+/5+}$. If an exact colour match could not be achieved estimated an intermediate value. If the colour of the reaction zone is equal to or more intense than the colour field for 0.5 mg / l $As^{3+/5+}$, use the Merckoquant Arsenic Test cat. No. 1.10026.0001 (measuring range 0.1-3 mg / l $As^{3+/5+}$). But in our observation any sample could not exceed 0.5 mg / l $As^{3+/5+}$.

The Merck kit, which is manufactured in Germany and has the major drawback that it can only measure down to 0.10 mg/l, that is, double the regional arsenic standard of 0.05 mg/l. However, it is widely acknowledged that none of the test kits are very accurate at low concentrations, and that Merck reagents are of very high quality.

The Merck field test kit is extremely simple, with emphasis on ensuring replicable and reliable results. The kit and all the individual reagents results. The kit and all the individual reagents (Zinc powder, hydrochloric acid, mercury bromide papers) carry expiry dates, and the well-packaged reagents ensure that users normally achieve about 100 tests per kit. Despite not having additional reducing reagents, or a method of removing sulfide interface, evaluations have found the Merck kit to be at least as accurate and reliable as other more complex field kits (Figure 4.1& 4.2).

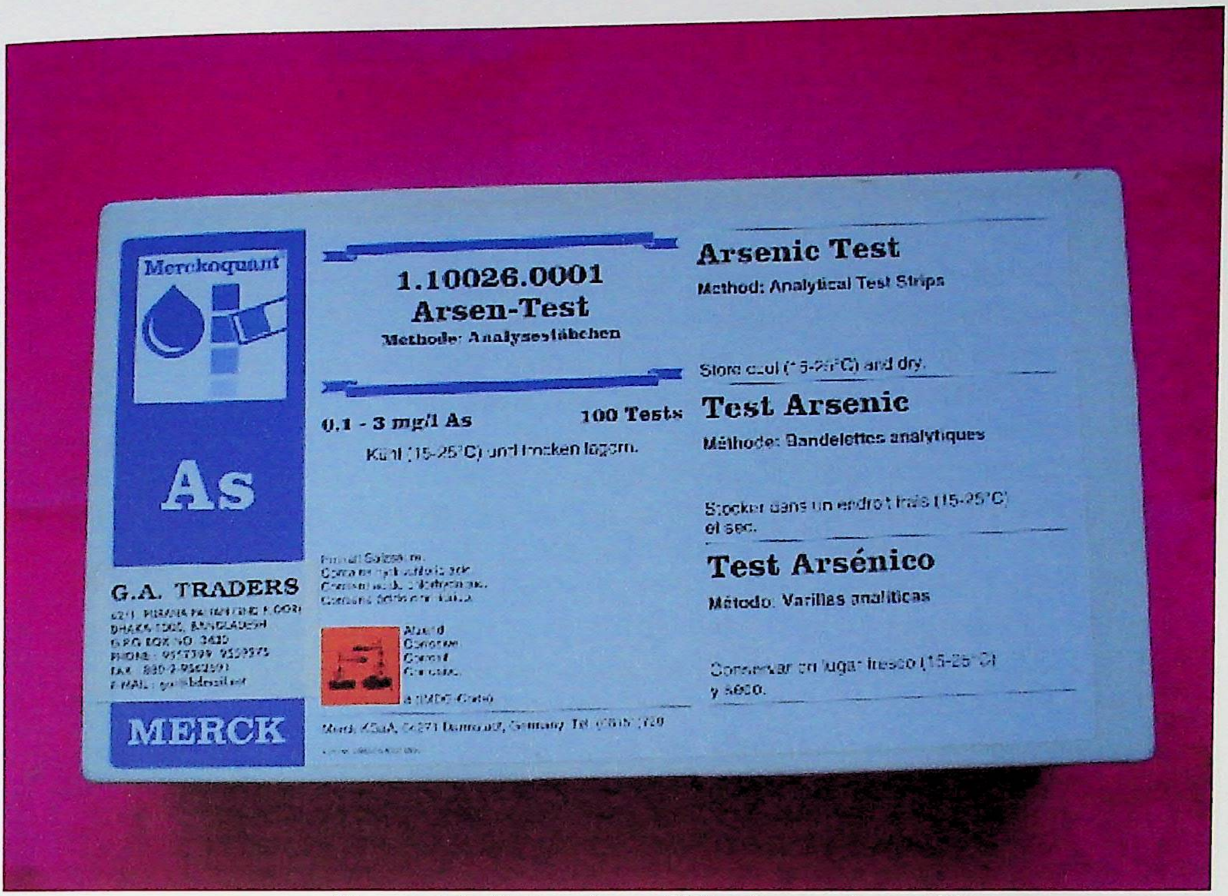


Figure 4.1: Outer view of Merck arsenic test kit

Figure 4.2: Inner view of Merck arsenic test kit

4.3 RESULTS AND DISCUSSION

4.3.1 Distribution of arsenic level in groundwater

Arsenic contaminated aquifers have no regular pattern, varies both horizontally and vertically within short distances. Data on arsenic concentration in the southeastern region of the Bangladesh were relatively scarce. The data on arsenic concentration of 305 groundwater provided a solid basis for evaluating arsenic contamination in the southeastern region of Bangladesh. These data were particularly useful because arsenic testing in this study was done using standard testing method, which provide reliable results. Figure 4.3 represents the distribution of arsenic in the tube wells located in the southeastern region of Bangladesh. It indicates that arsenic concentration level is about 41.31% under WHO guideline (0.01 mgl^{-1}), 29.69% is within permissible level (0.05 mgl^{-1}) and 29% exceeds the permissible level (> 0.05 mgl^{-1}). Out of the total 305 tube wells from southeastern region, arsenic concentrations in 124 were found to be below 0.01 mgl^{-1}, 90 were found in the range of 0.01 to 0.05 mgl^{-1} and 89 were found to be above 0.05 mgl^{-1}. More tube wells need to be tested for identifying the pattern of arsenic contamination in the southeastern region of Bangladesh.

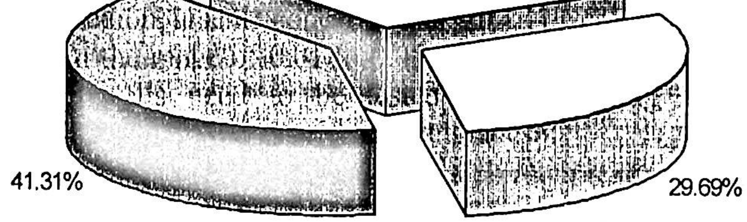


Figure 4.3: Distribution of arsenic level in ground water

A survey of well waters (n = 3534) from throughout Bangladesh, excluding the Chittagong Hill tracts, has shown that water from 27% of the 'shallow' tube wells, that is well less than 150 m deep, exceeded the Bangladesh standard for arsenic in drinking water (50 microgram/liter), 46% exceeded the WHO guideline value of 10 microgram/liter. Figures for 'deep' wells (greater than 150m deep) were 1% and 5%, respectively. Since it is believed that there are a total of some 6-11 million-tube wells in Bangladesh, mostly exploiting the depth range 10-50 m, some 1.5-2.5 million wells are estimated to be contaminated with arsenic according to the Bangladesh standard, 35 million people are believed to be exposed to an arsenic concentration in drinking water exceeding 50 microgram/liter and 57 million people exposed to a concentration exceeding 10 microgram/liter (Kinniburgh and Smedley 2001). However, according to Islam and Uddin (2002), the distribution of arsenic in the groundwater is related to the geology of the country rather than just the depth of the water table. According to them, the division of the aquifer system from the geological point of view – like the Upper Holocene aquifer, Middle Holocene aquifer, Upper Pleistocene-Early Holocene aquifer, Pilo-Pleistocene aquifer and older aquifers-is more logical when applied to the depth of the tube well pumping system as is customarily adopted in Bangladesh. They conclude that most of the arsenic-contaminated tube wells are drawing water from the Middle and Upper Holocene sediments.

4.3.2 Depth-wise distribution of arsenic in tube wells

Arsenic contamination is commonly associated with fluctuating water tables and flooding cycles particularly in acid sulfide/sulfate soils or where iron and/or manganese-enriched layers or saline-layered aquifers occur. Under these conditions the complex chemistry of arsenic will result in changes depending on exposure either to air or saturated soils. Levels of arsenic contamination in water supplies can vary through a year adding to the difficulties of identification and monitoring. A depth-wise variation of arsenic concentration in the wells of the southwestern region is shown in Figure 4.4. It indicates that the arsenic contamination increase with the increase of depth. Depth bellow 50 ft total 49 numbers of tube well samples were tested. Out of tested samples 92.68% of tube wells were arsenic level under WHO guideline (0.01 mg l^{-1}), 4.88% within permissible limit (0.05mgl^{-1}) and 2.44% exceeding permissible limit (>0.05 mg l^{-1}). Depth between 50 to 100 ft total 94 tube well samples was tested. Out of tested samples 40.43% of tube wells were arsenic level under WHO guideline (0.01 mgl^{-1}), 34.04% within permissible limit (0.05 mgl^{-1}) and 25.53% exceeding permissible limit (>0.05 mgl^{-1}). Depths between 101 to 150 ft total 51 nos of tube well samples were tested. Out of tested samples 25.49% of tube wells were arsenic levels under WHO guideline (0.01 mg l^{-1}), 39.22% within permissible limit (0.05 mg l^{-1}) and 35.29% exceeding permissible limit (>0.05 mgl^{-1}). Depth between 151 to 250 ft total 51 numbers of tube well samples was tested. Out of tested samples 0% of tube wells were arsenic level under WHO guideline (0.01 mgl^{-1}), 40% within permissible limit (0.05 mgl^{-1}) and 60% exceeding permissible limit (>0.05 mgl^{-1}).

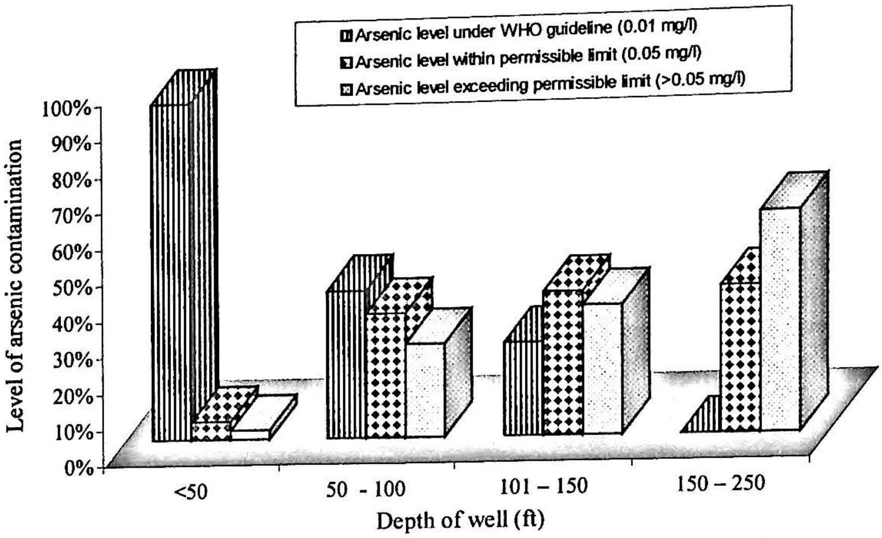


Figure 4.4 Depth-wise distribution of arsenic in tubewells

There were important differences between 'shallow' wells and 'deep' wells (defined here as greater than or equal to 150 m depth), as well as between samples from recent (Holocene) alluvium and older (Plio-Pleistocene) alluvium. Arsenic contamination was essentially confined to ground waters from the shallow aquifer (Figure 4.5). Of the wells sampled in the DPHE/BGS National Hydrochemical Survey, 9% were 'deep'. Most of these deep wells were collected from the southeastern part of Bangladesh. From the Figure 5.3, it can be clearly visualized that arsenic concentration is higher (600 microgram/liter) within 150 m tube well depth. In this arsenic contaminated areas, the arsenic concentration increases most rapidly between 10-60 m below ground level.

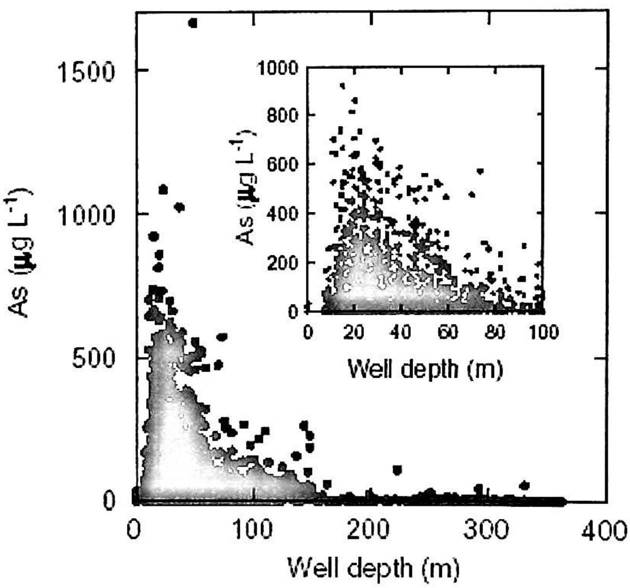


Figure 4.5: Arsenic concentration of groundwater in wells from the DPHE/BGS National Hydrochemical Survey plotted as a function of well depth

So, from this tested result no remarkable relation was found in arsenic concentration and depth of water table. Similar result was observed from other study. The majority of tests to date have been carried out on shallow tube wells used for drinking water. A significant number of tests have also been carried out on deep tube wells down to 100 meters used for agriculture. The tests show that at depths below 200 meters the incidence of contamination falls off and at 250 meters is, and will remain, arsenic-free provided that irrigation wells do not start using the same aquifer (Islam and Uddin 2002). Contamination of arsenic in ground water in Bangladesh has established significant variations with depth. Such variations may occur over very different timescales as a result of different depth. Arsenic concentrations in tube wells are lowest in high depth water table (CGWB, 1999). However, it was later discovered that many of the shallow aquifers in Bangladesh, mostly those less than seventy-five meters, contained high levels of arsenic (Burgrss et.al.2004).

4.3.3 Arsenic content in groundwater

Arsenic content in 59 contaminated tube wells varies from 0.02 to 0.5 mg/l. The tube-well depth ranges from 50 to 500 ft, the median depth being 100 ft. The mean and median of arsenic content are 0.12 and 0.08 mg/l respectively. The depth-wise arsenic distribution indicates highest average in 150-170 ft depth zone (Table 3.1). The sedimentary package in the southeastern area of Bangladesh is more or less continuous sand from the surface down to a depth of 120 ft. Maximum number of tube wells extract water from aquifers within this depth zone. The aquifers are semi-confined to unconfined and the average arsenic content is 0.11 mg/l. The other depth zones where tube wells are sunk represent multilevel coarser sand-gravel zones separated by finer sand and silt. The deeper aquifer, beyond 450 ft, is separated from those above by a clay layer. It is important to note that the deeper aquifer (450 to 500 ft) is also contaminated, though the average arsenic content is less (Table 4.1).

Table 4.1 Depth-wise arsenic distribution of tube wells

Depth of tube well (ft)	As (average)(mg/l)	No. of samples
50-120	0.11	34
150-170	0.23	4
230-280	0.09	3
350-400	0.13	6
450-500	0.09	12

In some areas arsenic contamination varies significantly and more predictably with depth. In Bangladesh, for example, almost all wells testing positive for arsenic are in aquifers shallower than 150 m. However, deeper wells may become contaminated over time (Alaerts et al. 2001).

4.3.4 Arsenic concentration variations in different depth during wet season

Arsenic concentrations in monitored wells were largely constant during the monitoring period. None of the shallow wells showed any seasonal response in As to rainfall. Concentrations were a little more variable during the early stages of sampling but this may due to temporary disturbances in the groundwater chemistry following drilling. Figure 4.6 shows the arsenic concentration of different depth of the wet season. It demonstrates that there is no remarkable relationship between arsenic concentration and depth of water table. Among the research results, highest arsenic concentration (about 1310 microgram l^{-1}) was observed in 150 ft water table depth. Some scientists speculate that arsenic concentration varied in depth of water table and wet season. I did not find any significant relationship among arsenic concentration, depth of water table and wet season. In the shallow piezometers, As concentrations show a progressive decrease with depth, from maximum values at 20 m (300 μgL^{-1}) to minimum at 50 m (160 μgL^{-1}). The profile may be explained in part at least by the occurrence of relatively high concentrations of oxalate-extractable As and Fe (denoting higher concentrations of liable As) at around 10 m depth. All are shallow wells and all exceed 50 μgL^{-1}. The highest As concentrations among these were found in tube well (24 m depth) which had concentrations in excess of 300 μgL^{-1}.

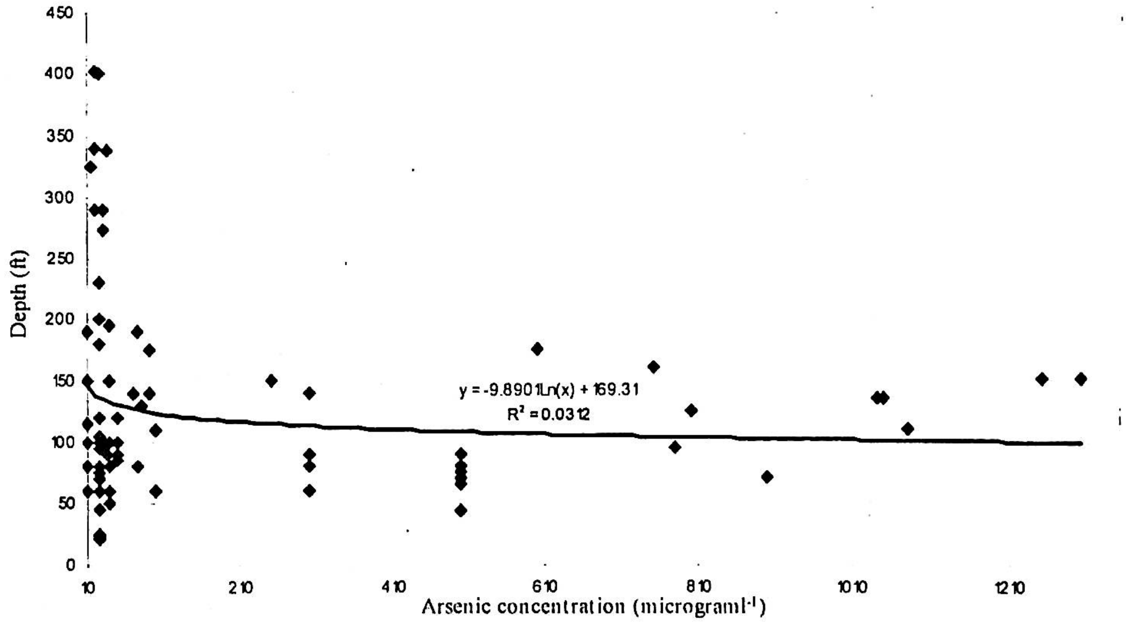


Figure 4.6 Arsenic concentration in different depth during wet season

During the course of the study, hand tube wells were monitored for arsenic, water levels and seasonal variations of arsenic contamination. No clear or consistent changes in arsenic were detected during monitoring period. However, longer-term monitoring of the wells is required to establish whether there will be significant seasonal and long-term trends in water chemistry. The relatively small variations in arsenic concentrations observed in many of the wells emphasis the need for very careful sampling and high precision analysis if seasonal or long-terms trends are to be detected reliably.

4.3.5 Arsenic concentration variations in different depth during dry season

Figure 4.7 illustrates arsenic concentration variations in different depth during dry season. Less arsenic concentration was observed during dry season. Highest arsenic concentration (300 microgram/liter) was observed for the depth of 140, 150 and 165 ft respectively. From 120 to 160 ft arsenic concentration varied from 10 to 100 microgram/liter. Research results are not shows any remarkable relationship among arsenic concentration, depth and dry seasonal variations. There is no evidence to support the proposition that the groundwater arsenic problem is caused by the dry season draw down of the water table due to a recent increase in irrigation abstraction. Monitoring groundwater at different depths has shown some variation with time but there is as yet no convincing evidence for seasonal changes. Dramatic changes in contamination are not expected within such a short time scale. A monitoring programme should be undertaken at a range of sites to monitor possible long-term changes.

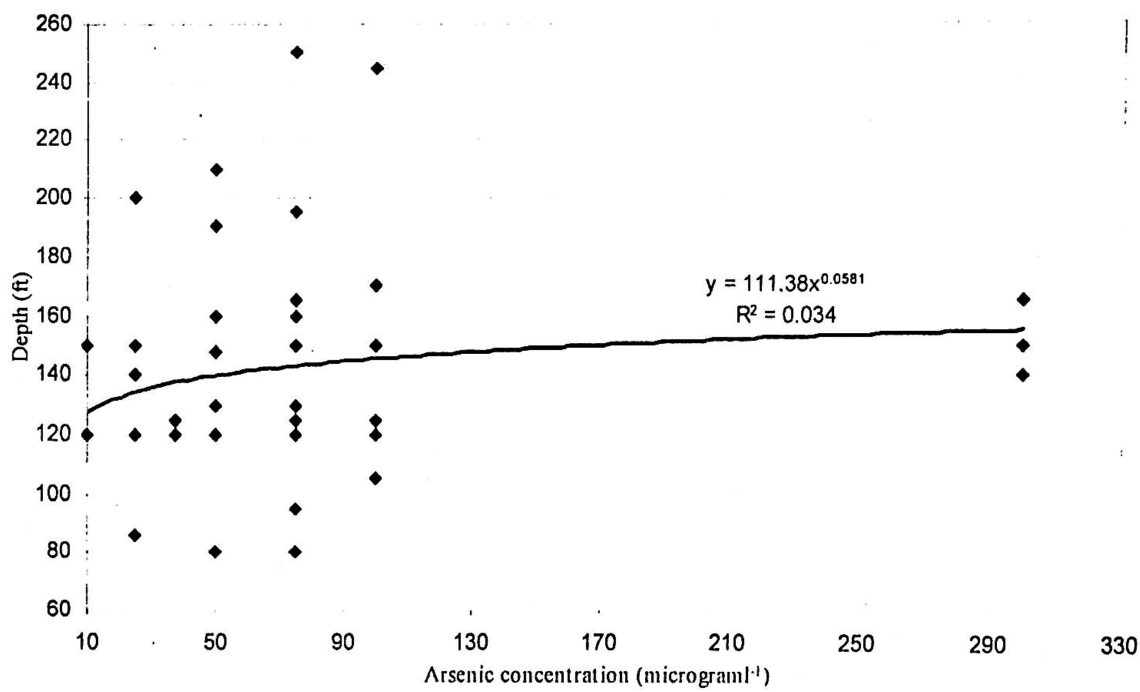


Figure 4.7 Arsenic concentration in different depth during dry season

Arsenic concentration is strongly dependent on depth. The highest concentrations of arsenic, and the highest probability of exceeding thresholds for arsenic, most often occur in wells screened between 20 and 60 m but the precise depth of the maximum varies between regions and the sharpness of the concentration peak differs from place to place (Karim et. al.1997). The depth distributions show to be fallacious the belief that drilling deeper than 100m provides arsenic free water; it will do so in some parts of Bangladesh, but not, for example, in the region of the Sylhet Basin. A sharp upper limit to high concentrations of arsenic appears to occur at about 10-15 m depth; few data are available for wells in the depth range 0-10 m, so this may be an artefact of data distribution. That it is not is suggested by the fact that dug wells, which are mostly much less than 10m deep, are rarely polluted with arsenic (Chakraborty 2001). Below 200 m, arsenic concentrations rarely exceed a few micrograms per liter (Frisbie et al. 1999).

4.3.6 Arsenic concentration variation with age of tube wells

One of the questions which was of concern to many was whether the tube wells currently safe will remain so in the future or the concentration of arsenic will change with time in the aquifers for worse due to stress imposed by abstractions. In order to obtain answer to such questions data have to be collected over extended period of time. Delay for such observation was not possible, as answers were needed immediately. I was found suitable proxy parameters that can reasonably replace the time parameter. One such proxy could be the age of the tube wells. As tube wells were sunk without specifically taking into consideration the aquifer properties, the average concentration of arsenic in tube wells sunk at any given period should be statistically the same as any other period. So, any change in the concentration of arsenic with tube well age should represent its abstraction-induced effect. Average concentrations for arsenic in tube wells grouped by age were calculated for all the tube wells and also for the depth segregated groups. The results were shown in Figure 4.8.

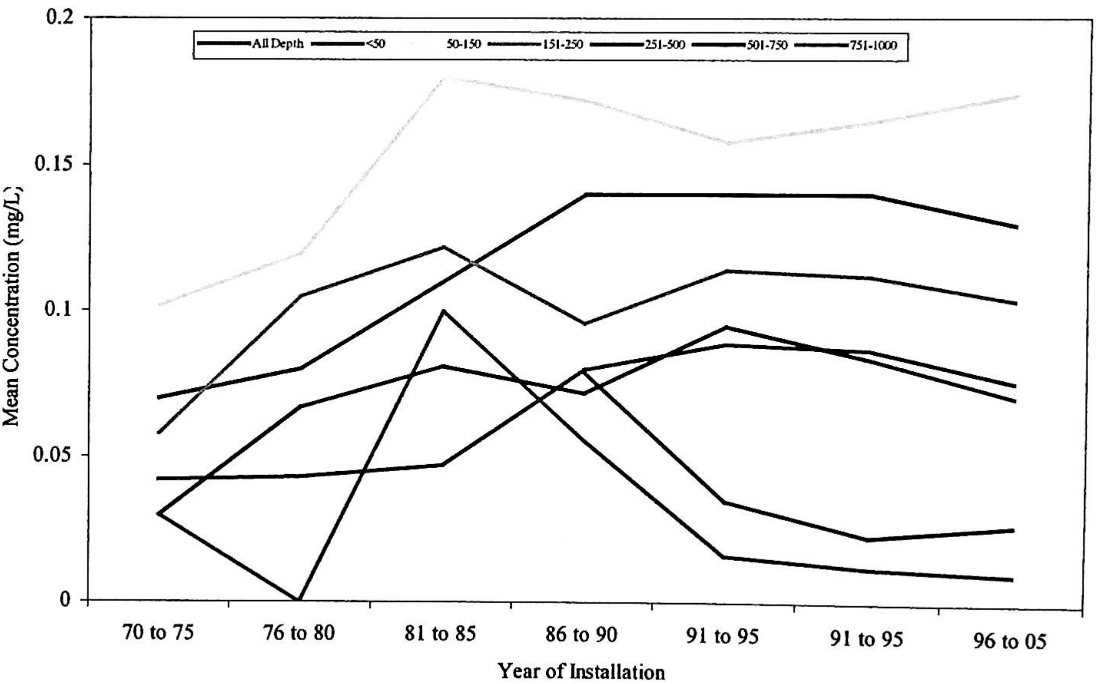


Figure 4.8 Arsenic concentration with age of tube well

It can be seen that for all the groups except >1000ft, the concentrations do not appreciably change with tube well age. There appears to a slight decrease with time but this may not be very significant as these mostly relate to tube wells dating back to seventies. As pointed out earlier data for these tube wells may not be all that accurate. In the case of tube wells of depth greater than 1000ft, there may be even greater problem with data as discussed earlier and this is probably reflected in the increase in the arsenic concentration for the tube wells between the periods 1970-80.Also, in eighteen decade trends of mean arsenic concentration was higher in all categories of depth.

4.4 CONCLUSION

Based on the experience of arsenic contamination of groundwater in West Bengal, India, it was initially thought that arsenic contamination of ground water would be concentrated in the northeastern and central region of the country. However as more data from other regions are becoming available, it appears that arsenic contamination in Bangladesh is much more widespread. This study clearly shows that the southeastern region of the country is severely affected by arsenic contamination of groundwater. Similar systemic study is needed to ascertain arsenic contamination in other region of the country. Ground water from the deep aquifer was sampled from the southeastern Bangladesh. These samples may or may not be representative of the deep aquifer in other areas. There is no reason to believe that the deep aquifer will become seriously contaminated, certainly in the short-term, providing that care is taken during borehole construction to isolate the upper and lower aquifers and so prevent direct leakage of contaminated water to the deep aquifer. From the above study, it can be concluded that water from the great majority of carefully constructed deep wells would not only pass the current Bangladesh standard for arsenic but would pass all other existing national and international standards and guidelines for arsenic. Most of the deep ground waters tested in my studies were from the southeastern region where the shallow ground water may not be typical of those from elsewhere in Bangladesh. The possible impact of the large-scale abstraction of irrigation water on the deep aquifer also needs to be considered.

Chapter 5

LOW COST TECHNIQUES FOR REMOVAL OF ARSENIC FROM CONTAMINATED GROUNDWATER

5.1 INTRODUCTION

Presence of elevated levels of arsenic in groundwater has become a major concern in Bangladesh. Although arsenic contamination of water sources has been reported for a number of countries, the contamination scenario in Bangladesh appears to be the worst detected so far worldwide, both in terms of area and population affected. Arsenic pollution of groundwater is particularly challenging in Bangladesh since tube well water extracted from shallow aquifers is the major source of drinking water for most of its population. Estimates of population exposed to arsenic concentration above the Bangladesh drinking water standard of 0.05 mg/L vary from about 20 million to over 36 million (EES/DCH, 2000). People in Bangladesh, particularly in the rural areas, are accustomed to using groundwater from hand tube wells for long and unlike surface water, it is considered safe from bacteriological pollution. In view of the overwhelming dependence of the population on groundwater, development of suitable treatment systems for arsenic removal from groundwater appears to a promising option for providing safe water to the rural population. Socio-economic conditions of Bangladesh demands low-cost as well as small-scale treatment systems that could be implemented in the rural areas at household or community levels. In the backdrop of the widespread arsenic contamination of groundwater in Bangladesh, I developed low-cost arsenic removal techniques. I am motivated by the need to developed simple, low cost technique for the removal of arsenic from the groundwater of Bangladesh by using locally available materials. In this chapter I have developed arsenic removal by three-pitcher method and removal of arsenic through water hyacinths both from natural arsenic contaminated ground water and artificial arsenic solution. As one of the oldest water purification methods known as the three pitcher (locally known as '3-kalshi') was developed in before. I modified the previous one by changing or adding materials for its efficacy in removing arsenic and other impurities. My specific objective of the present research is the possibility to bring the level of arsenic within tolerable range from arsenic contaminated ground water through three pitcher and water hyacinths plants.

5.2 Removal of arsenic through three-pitcher filter

5.2.1 Experimental set up

The basic 3-pitcher (hereafter referred as '3-kalshi', the local name) water purification setup is shown in Figure 5.1. It consists of three kalshis made of fired unglazed clay used as reservoir for drinking water by 60% of the people in Bangladesh. Local artisans make a variety of shapes of these kalshis. I have used the ones with narrow mouth, round bottom and have a volume of about 12 liter. The three kalshis are stacked on top of each other. The bottom of top two kalshis has a small hole plugged with 100% birth cloth or similar materials. The first kalshi contains 3 kg (approx. 1/6 kalshi volume) of gravels, 0.25 kg of coal, 0.2 kg of coarse sand and little bit fine sand that are surrounded by cloth. The second kalshi consists of 1-kg coarse sand and 2 kg of fine sand. Both coarse and fine sand was obtained from the local Padma River. Gravel was collected from road side in which road was constructing during seal coat. The river sand has about 0.05% iron oxide. The coal was collected from burned wood or khari for cooking. All precautions were taken to avoid the fine wood or khari ash, which may dissolve in water and produce a basic solution. The function of the coal is to adsorb organic impurities that may be present in groundwater. As the coal is lighter than water, it was placed in between layers of gravel and coarse sand. The fine sand was the final filter. The third kalshi was the collector. To prevent bacterial contamination from gravel, sand and coal, all was boiled and washed. All data obtained in this experiment were obtained after discarding first two kalshi volumes of water.

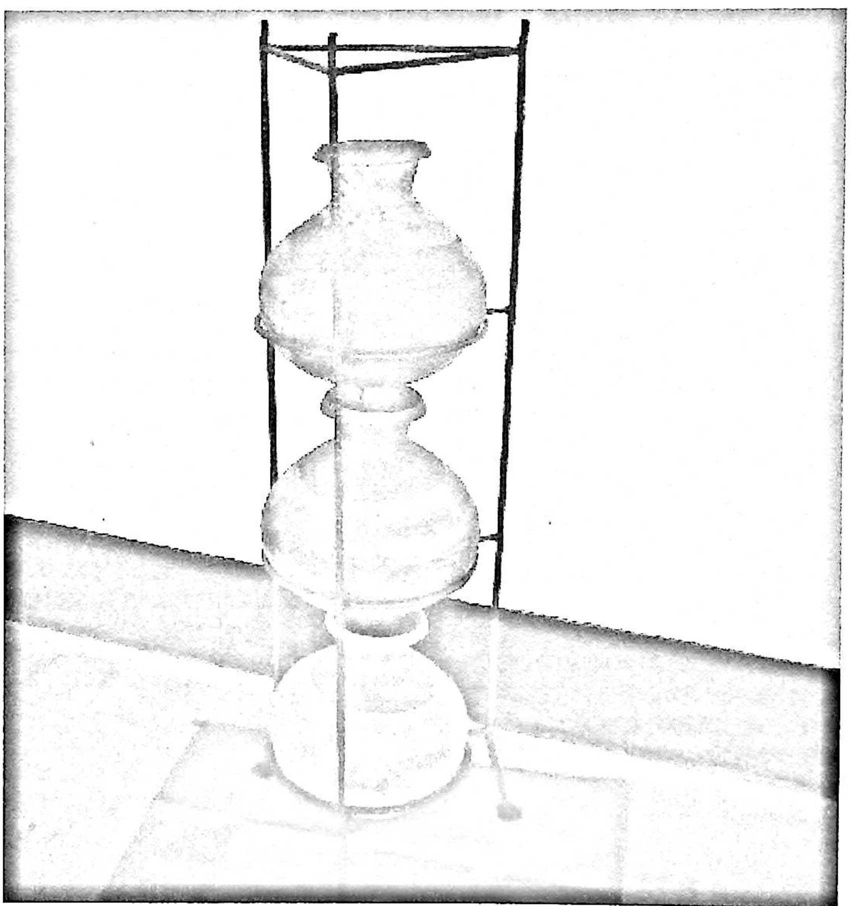


Figure 5.1: Experimental set up of three pitcher filter

5.2.2 Experimental procedure

Initially high arsenic contaminated water was used to remove arsenic. Using Merck arsenic test kit ensured high arsenic concentration for the treated water. The arsenic concentration was measured by Merck arsenic test kit as well as laboratory analysis during experiment. SDDC method was applied to know the exact the exact value of arsenic concentration before and after treatment of water. Five numbers of trails were done during experiment for the accuracy of the results.

5.2.3 Procedure for arsenic analysis by silver diethyldithiocarbamate (SDDC) method

5.2.3.A General discussion

a. *Principle:* Inorganic arsenic is reduced to arsine, AsH_3, by zinc in acid solution in a Gutzeit generator. The arsine is then passed through a scrubber containing glass wool impregnated with lead acetate solution and into a absorber tube containing silver diethyldithiocarbamate dissolved in pyridine or chloroform. In the absorber, arsenic reacts with the silver salt, forming a soluble red complex suitable for photometric measurement.
b. *Interference:* Although certain materials-chromium, cobalt, copper, mercury, molybdenum, nickel, platinum and silver- interference in the generation of arsine, the concentrations of these metals normally present in water do not interference significantly. Antimony salts in the sample form stibine, which interferes with color development by yielding a red color with maximum absorbance at 510 mm.
c. *Minimum detectable quantity:* 1µg As.

5.2.3.B Apparatus

a. *Arsenic generator and absorption tube*
b. *Photometric equipment:*
    1) *Spectrophotometer,* for use at 535 nm with 1-cm cells.
    2) *Filter photometer,* with green filter having a maximum transmittance in the range 530 to 540 nm, with 1-cm cells.

5.2.3.C Reagents

a. *Hydrochloric acid,* HCl, conc.
b. *Potassium iodide solution:* Dissolve 15g KI in 100 mL distilled water. Store in a brown bottle.
c. *Stannous chloride reagent:* Dissolve 40 g arsenic-free $Sncl_2.2H_2O$ in 100 mL conc. HCL.
d. *Lead acetate solution:* Dissolve 10 g $Pb(C_2H_2O_2)_2$, $3H_2O$ in 100 mL distilled water.
e. *Silver diethyldithiocarbamate reagent:* Dissolve 410 g 1-ephedrine in 200 mL chloroform ($CHCl_3$), add 625 mg $AgSCSN(C_2H_5)_2$ and adjust volume to 250 mL with additional $CHCl_3$. Filter and store in brown bottle.

f. Zinc, 20 to 30 mesh, arsenic free.

g. *Stock arsenic solution:* Dissolve 1.320 g arsenic trioxide, As_2O_3, in 10 mL distilled water containing 4 g NaOH, and dilute to 1000 mL with distilled water; 1.00 mL= 1.00mg As.

h. *Intermediate arsenic solution:* Dilute 5.00 mL stock solution to 500 mL with distilled water; 1.00 mL = 10.0μ As.

i. *Standard arsenic solution:* Dilute 10.00 mL intermediate solution to 100 mL with distilled water; 1.00 mL = 10.0μ As.

5.2.3.D Procedure

a. *Treatment of sample:* Pipet 35.0 mL sample into a clean generator bottle. Added successively, with through mixing after each addition, 5 mL conc HCL, 2 ml KI solution and 8 drops (0.40 mL) $Sncl_2$ reagent. Allow 15 min for reduction of arsenic to the trivalent stste.

b. *Preparation of scrubber and absorber:* Impregnate glass wool in the scrubber with lead acetate solution. Do not made too wet because water will be carried over into the reagent solution. Pipet 4.00 mL silver diethyldithicarbamate reagent into absorber tube.

c. *Arsenic generation and measurement:* Add 3 g zinc to generator and connect scrubber-absorber assembly immediately. Make certain that all connections are fitted tightly.
Allowed 30 min for complete evolution of arsine. Warm the generator slightly to insure that all arsine is released. Pour solution from absorber directly into a 1-cm cell and measure absorbance at 535 nm, using the reagent blank as the reference.

5.2.3.E Calculation

$$\text{mg As/L} = \frac{\mu\text{g As (in 4.00 mL final volume)}}{\text{mL sample}}$$

5.2.3 F Precision and Accuracy

A synthetic sample containing 40µg As/L, 250 µg B/L, 20 µg Se/L and 6µg V/L in distilled water was analyzed in 46 laboratories by the silver diethyldithiocarbamate method, with a relative standard deviation of 13.8% and relative error of 0%.

5.2.4 Results and Discussions

The Figure 5.2 unfolds clear idea about amount of arsenic removed through 3-pitcher filtration method in 5 nos. of different trail. In all the five trials arsenic removal through 3-pitcher filtration method was nearly same. Also, arsenic removal rate was higher in comparison with existing 3-pitcher filtration method. Because, in this experiment, coal was used as filtering material that extract more arsenic from arsenic contaminated water. Under this filtration process arsenic was reduced about twenty times from the contaminated water. Monitoring of the unit, it shows that arsenic removal is good, but

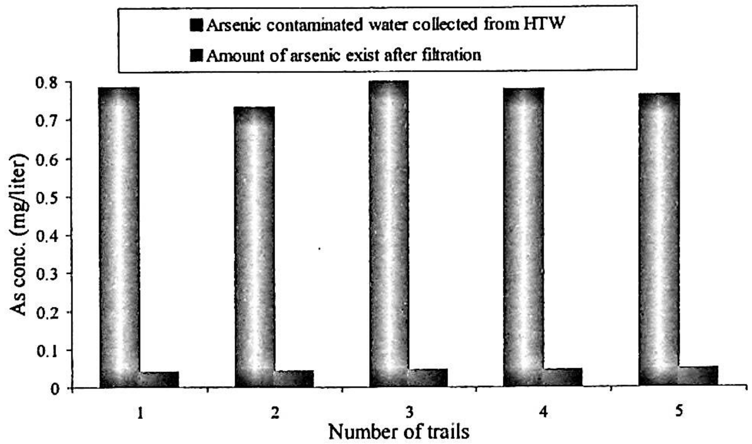


Figure 5.2: Amount of arsenic removal by 3-pitcher filtration method

one of the disadvantages for my developed three-pitcher method is as the pitchers are set one above another and not protected from the open air; there exists a risk for water to be contaminated with microorganisms in the atmosphere.

Previously, three-pitcher filter was developed and iron chip was used as a binder. Unfortunately, the iron filings cannot easily be cleaned or removed from the kalshi, as they rust into a solid lump after 10 days or so use. Therefore, it will be needed to provide a completely new kalshi, including fresh media, whenever monitoring reveals that arsenic removal is declining, or if the household complains that the flow rate is too low. Also, iron chips are not available in rural as well as remote areas of Bangladesh. For that reason, I have developed three-pitcher method that containing sand, gravel and coal for binding arsenic-so that safe water collects in the third pot. Operation is extremely simple as no reagents are involved. The exact mechanisms of the arsenic removal are unclear, but it seems likely that the major removal process is adsorption of arsenic by filtration on the coal. Also, most important things, all materials are available in any rural household of Bangladesh. Total cost of instillation is about 5 USD.

Existing three-pitcher method has lot of disadvantages such as potential problem of clogging with iron, the system produces sludge that contain iron, the flow rate of water through the system is low and hence amount of water produced is small. My developed three-pitcher method is so low cost, due to indigenous technology social acceptance is high and simple to construct with minimum skill.

5.3 Removal of Arsenic by Water Hyacinths (*Eichhornia crassipes*)

5.3.a Phytoremediation by water hyacinths

Phytoremediation is the process of using plants to remove pollutants from soil or water. Plants used to phytoremediate metals, like arsenic, are called hyper accumulators. Water hyacinths are free flooding aqueous weeds that multiply very quickly. They have very fibrous roots and get all of their nutrients from the water. They have pinkish purple flowers and grow in dense mats in tropical and subtropical freshwater rivers, lakes and reservoirs.

5.3.b The Potential Use of Water Hyacinths in Bangladesh

Several studies suggest that it may be possible to use water hyacinths effectively to remove the arsenic from the ground water that is poisoning the people of Bangladesh. Thus, water hyacinths may be a practical solution to the arsenic problem in Bangladesh because using them as a treatment method has very little cost. Water hyacinths grow naturally in the ponds in Bangladesh. The farmers in Bangladesh have containers, known as Chari, that they use for animal feed. These containers could be used to hold water hyacinths and arsenic contaminated water for treatment.

5.3.c The Focus of this study

Although the studies describe above have shown that water hyacinths remove different amounts of arsenic from water, they all show that water hyacinths do remove an appreciable amount of arsenic. The purpose of the study were: (1) to determine for how long the same water hyacinths plants can be used effectively to reduce arsenic concentrations and (2) to determine where in the plant structure the arsenic is stored. These are important questions regarding the use of water hyacinths to remove arsenic from the ground water in Bangladesh because, at some point, the water hyacinths will no longer reduce arsenic levels effectively. The people will need to remove the plants from the water before they lose their ability to remove arsenic effectively and replace them with fresh plants. At that point, if all of the arsenic is stored in the roots, then may be the stems and leaves could be used as animal feed.

5.3.1 Removal of arsenic from arsenic solution through water hyacinths

5.3.1.a Methodology to remove arsenic from arsenic solution by water hyacinths

5.3.1.a (i) Experimental design

One purpose of my study was to determine how much arsine water hyacinths can phytoaccumulate before losing their ability to phytoaccumulate arsenic efficiency. I defined effective phytoaccumulation as the ability of the plant to reduce arsenic levels from 300 ppb, a typical concentration in Bangladeshi well water, to 10 ppb, the US EPA drinking water standard, effective 2006, in an approximately twenty four hour period (Chowdhury, 2004). The control was water hyacinths living in the same conditions as the arsenic plants, but without the arsenic in their water.

5.3.1.a (ii) Safety when working with arsenic

While working with arsenic, safety was very important. I read all the Material Safety Data Sheets (MSDSs) before starting my experiment. I always wore protective gloves when necessary. Any waste solutions as well as anything that came in to contact with an arsenic solution above EPA's proposed drinking water standard were put in hazardous waste disposal. The arsenic solutions were kept in the locked chemical storeroom. Finally, to protect other students and animals, I prepared a placard adjacent to Chari containing the arsenic solution (Figure 5.3).

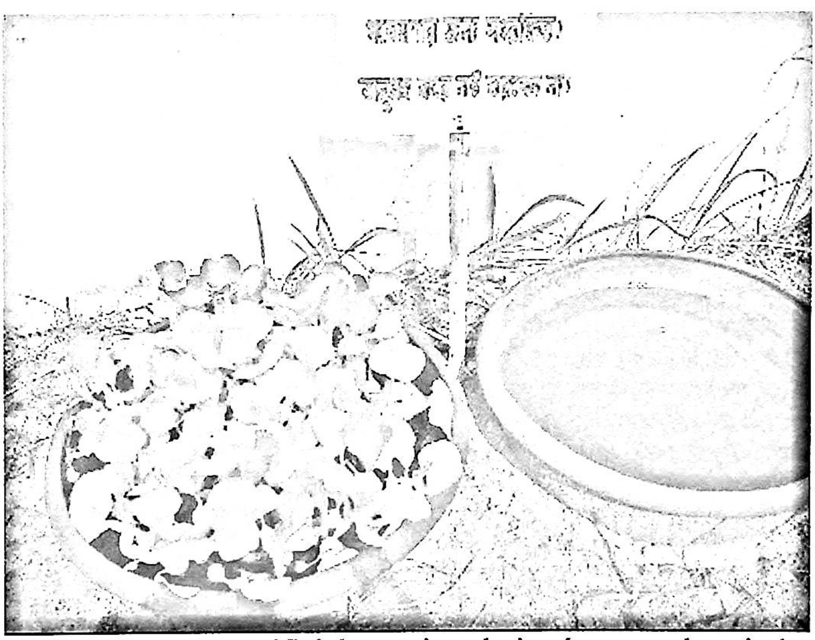


Figure 5.3: Experimental set up for artificial arsenic solution by water hyacinths

5.3.1.a (iii) Procedures of arsenic removal from artificial arsenic solution through water hyacinths

A. Creating initial arsenic solution and control

I set up my experimental system to mimic the treatment system of a Bangladeshi family might use. I filled a Chari with arsenic solution and put water hyacinth plants on that Chari. I maintained the water level one inch below from the top of the Chari. For the control, I filled a second Chari with distilled water (Figure 5.3) up to same level as before. Using sodium arsenate ($Na_2HasO_4*7H_2O$) and sodium arsenite ($NaAsO_2$) powder (Anstiss et. al.2001), I created a 300 ppm (as arsenic) arsenite solution and a 300 ppm (as arsenic) arsenate solution using distilled water. I had to add sodium hydroxide to make the arsenic powder dissolve and then I neutralized the solution with acetic acid. For the control, I created a solution containing the same amounts of sodium hydroxide and acetic acid that I had added to the arsenic solution. I added 10 ml of the arsenite solution and 10 ml of the arsenate solution and to the arsenic Chari to create 150 ppb arsenite (as arsenic) and 150 ppb of arsenate (as arsenic) for a total of 300 ppb (as arsenic) arsenic level. Then I stirred the water well. I added 10 ml of the sodium hydroxide-acetic acid mixture to the control Chari to match the sodium acetate concentration in the arsenic Chari and stirred well.

B. Testing arsenic levels in water

Tuesdays through Fridays, I started the sampling processes by stirring the arsenic from the Chari. I took a 50 ml sample from the Chari and tested the arsenic level using a Hach colorimetric test kit, following all of the instructions. In the Hach procedure the inorganic arsenic gets converted to arsine gas by sulfamic acid and zinc, and the arsine gas reacts with the mercuric bromide coating on the test strips to cause discoloration, which is compared to the colors and levels shown on the test kit bottle. Importantly, because this was a field test kit, it only indicated levels of 0, 10, 30, 50, 70, 300 and 500 ppb (Hach. Arsenic Test Strip, 100 Tests.(n.d.). Retrieved from Hach (Figure 5.4). From this test I determined the arsenic concentration in the Chari. I recorded the arsenic concentration of the Chari and then I added the amount of arsenite and arsenate solution needed to raise the arsenic level back up to 300 ppb, always using equal amounts of arsenite and arsenate solution. After adding the arsenic solutions, I measured the same amount of sodium hydroxide-acetic acid solution to the control Chari as the amount of arsenite solution I had added to the Chari. I added the solutions to the Chari and stirred well. I repeated these measurements and addition of more arsenic until the arsenic level was no longer reduced below 300 ppb after at least twenty-four hours. As needed, I re-filled the Chari with distilled water up to below one-inch top of the Chari. Once during the experiment, I tested the arsenic level of the control Chari to make sure that the solution and calculations were correct for restoring the arsenic level to 300 ppb. They were both at the corrected level.

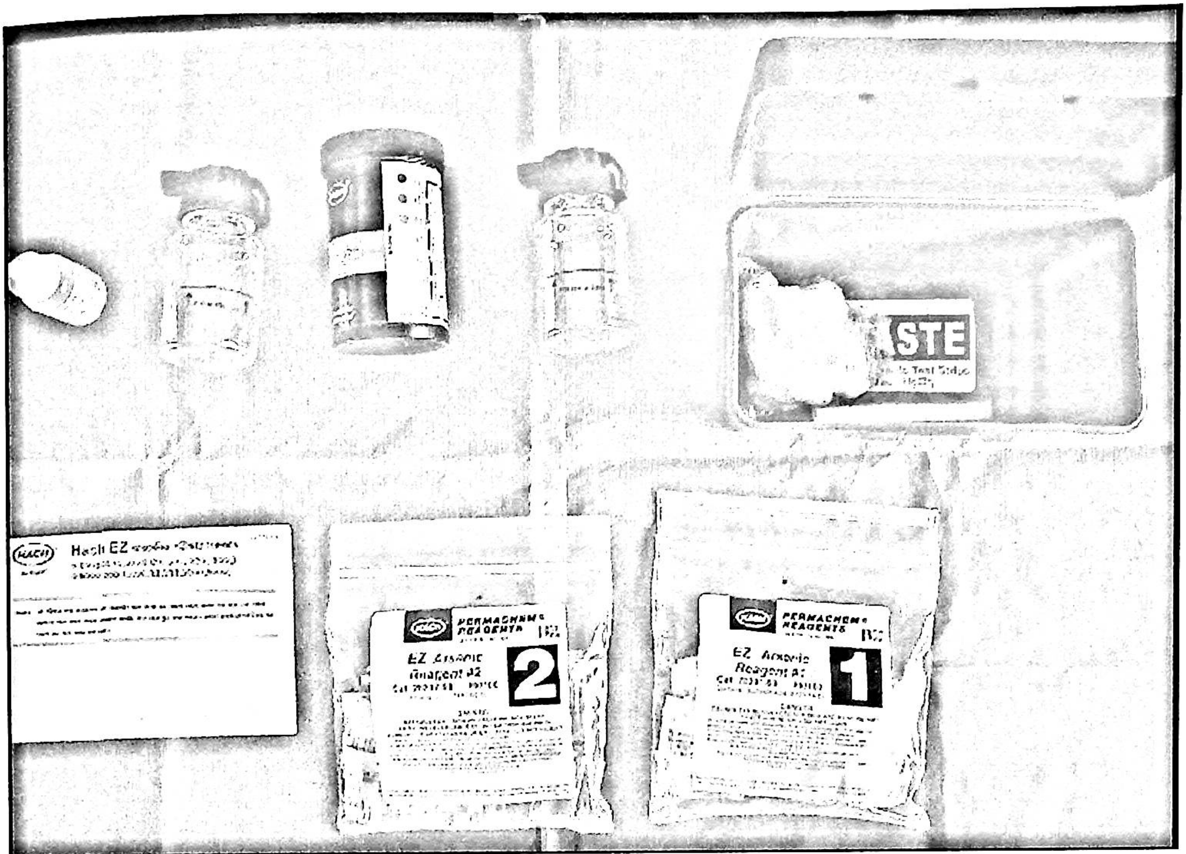


Figure 5.4: Hach arsenic test kit

C. Data Analysis

To answer my primary question, I counted the number of trials that the water hyacinths effectively phytoaccumulated arsenic by reducing a 300 ppb solution of total arsenic to less than 10 ppb. I also recorded the removal of arsenic in subsequent trails, even though the arsenic levels were not reduced to 10 ppb. To estimate the arsenic concentration in the roots, I first determined the total mass of arsenic extracted by multiplying the arsenic concentration I measured in the water extract (in ppb or μg/l) by 0.1 liter (because of the dilution of extract with 100 ml water). Next I divided the arsenic mass by the mass of the plant sample from which it had been extracted and converted the result to micrograms arsenic per kilograms of plant (ppb). I did the same for the stems/leaves/bladders. This calculation assumes 100 percent extraction into the water and negligible water loss in the extraction processes although this is not like because the plant tissues strongly binds the arsenic, a known property of metal accumulators.

5.3.1.a (iv) Results about arsenic remove from arsenic solution

A. Reduction of arsenic concentration from arsenic solution

Figure 5.5 Arsenic (ppb) left in each trail, starting with 300 ppb in each trail

The water hyacinths reduced the arsenic level from 300 ppb (typical Bangladeshi level) to below 10 ppb (EPA's proposed drinking water standard) for one trial. They continued to remove arsenic in the next four trails, but only down to 50 ppb (second trail) and 70 ppb (third through fifth trail), before ceasing to be effective at the removal of arsenic. I had thought water hyacinths might have been able to reduce the levels of arsenic in the water from 300 ppb (the arsenic concentration of water in typical wells in Bangladesh) to 10 ppb or less (US EPA's drinking water standard, effective 2006) for many trials, but they did this for only one trial. In the first twenty-four hours, trail one, they reduced the arsenic level from 300 ppb to 0 ppb. In the second twenty-four hour period, the water hyacinths reduced the arsenic level from 300 ppb to 50 ppb, which meets the Government of Bangladesh's drinking water standard, but not the US EPA's standard. During the third twenty-four hour period, the water hyacinths only reduced the arsenic level from 300 ppb to 70 ppb. During trial four the plants again reduced the arsenic level from 300 ppb to 70 ppb. I did not add any more arsenic, thinking that the water hyacinths might be able to remove the rest of the arsenic if they had more time. However, forty-eight hours later the arsenic level was still 70 ppb; the water hyacinths had not removed any more arsenic. To start trial five, I again increased the arsenic concentration to 300 ppb. In the next twenty-four hour period, the water hyacinths reduced the arsenic level from 300 ppb to 70 ppb. During the final trial, the water hyacinths did not reduce the arsenic level from 300 ppb at all. Three weeks later, when I tested the arsenic level of the water again, the arsenic level was still at 300 ppb.

B. Systematic error of the study

This method used for testing the arsenic levels was a field colorimetric test kit, which only indicated the following levels: 0, 10, 30, 50, 70, 300, and 500 ppb. This was probably not sufficiently precise for the experiment. For example, when I read a level of 300 ppb, the observed color was closer to 300 ppb than 70 ppb, but could have been somewhere in between.

5.3.1.a (v) Discussions about arsenic remove from arsenic solution

Under the conditions of my experiment, the water hyacinths reduced the arsenic level from a typical Bangladeshi well water concentration (300 ppb) to EPA's proposed drinking water standard (10 ppb) for one trial, to the Bangladeshi drinking water standard (50 ppb) for two trials and lost all ability to remove arsenic after five trials. The fact that most of the arsenic was not extracted by water is very good. That means the arsenic is tightly bound at a molecular level so that, even when finely chopped, the arsenic did not leach out of the plants. Because of this, when the plants are removed from treatment, they would not leach large amounts of arsenic into the environment and could be stored safely for disposal in an area away from drinking water, at least until they started to decompose.

5.3.2 *Removal of arsenic from ground water through water hyacinths*

5.3.2.a Experimental procedure

Initially high arsenic contaminated tube wells were selected through MERCK test kit in which arsenic concentration was more than 0.5 mg/liter. After that water samples were sent to arsenic testing laboratory to know the exact value of arsenic concentration. Before placement of water hyacinths into chari, all hyacinths plant was stored two weeks in a bucket that was filled with distilled water. Stored hyacinths plant washed by distilled water and separated all spoiled root, stem and leaves just before placement of chari that was filled with naturally arsenic contaminated ground water (Figure 5.6). I took out water hyacinths plant four week after placement from Chari. Then I immediate sent water that

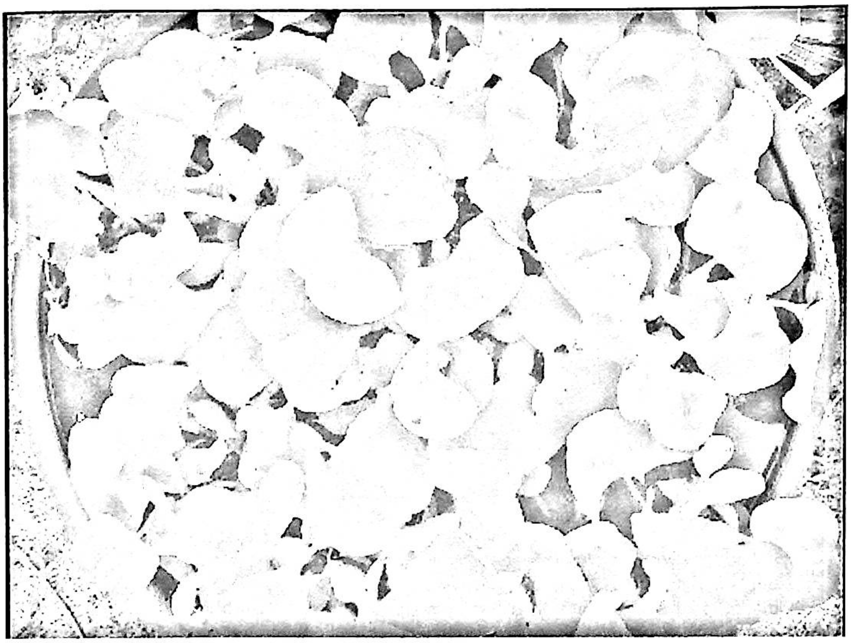


Figure 5.6: Water hyacinths in a Chari filled with natural As contaminated groundwater

was extracted by water hyacinth to the laboratory to know the exact value of arsenic exist in that sample. After receiving testing result, I have observed about fifty percent arsenic was removed by hyacinths plant. Then, I conducted a water extraction of select parts of the plant to see whether in the plant arsenic was stored. To create the extract, I took three plants that had been in the arsenic Chari and washed them off well, first with tap water and then with distilled water. I dried the plants very well and I separated the roots from the stems and leaves. I weighed a root sample and then cut it up into small pieces. I put the root sections with 100 ml distilled water into a blender. I "chopped" and "liquefied" the mixture for about seven minutes, stopping occasionally to scrape down the side of the container, until it was all pureed. I put a funnel over the mouth of a beaker and lined the funnel with a piece of filter paper. I poured the puree into the funnel and allowed all of the water to drip through over night. After about twenty-four hours, I took a sample of the extract and sent for laboratory analysis. I repeated the same procedure for the stems and leaves for the same plant. All samples were acidified before laboratory analysis and five numbers of sampling were conducted during experiment. The Silver Diethyldithiocarbamate (SDDC) method (Described in section 5.2.3) was applied for determining exact arsenic concentration in water sample before and extracted by water hyacinths plant after, roots, stem and leaf of hyacinths plant.

5.3.2.b Results and discussions about arsenic remove from arsenic contaminated water

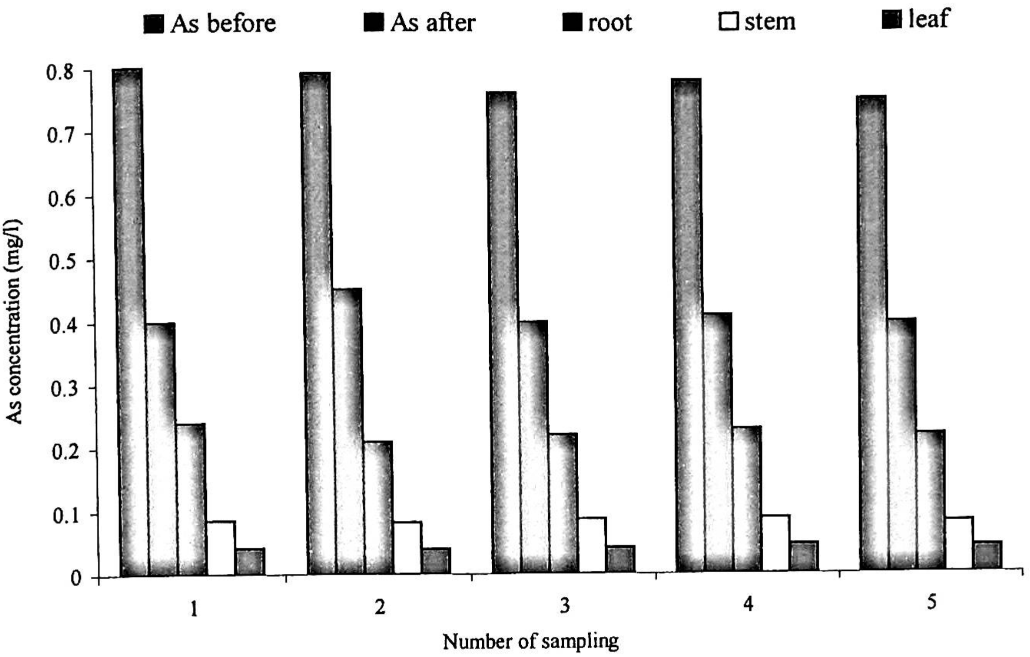


Figure 5.7 Arsenic absorption pattern by water hyacinths

Figure 5.7 clearly visualize pattern of arsenic removal by water hyacinths plant. In my experimental result, it can be clearly observed that arsenic can be removed 50% to 60% by water hyacinths plant from arsenic contaminated groundwater. From my preliminary plant tissue extraction data, there appears to be more arsenic being stored in the roots than in the stems and leaves. These are important questions to be resolved if this system is to be used in Bangladesh because, at some point, the water hyacinths is available in everywhere in Bangladesh. Numerous studies have investigated the possibility of using a variety of plant types to remove arsenic and other metals from water with different results. Ahsan (2002) found that most aquatic plants could have an arsenic level up to 3,000,000 ppb even though the water they are in has an arsenic level of less than 1,000 ppb. In particular, water hyacinths (*Eichhornia crassipes*) have been found to be accumulators of chromium and copper and hyperaccumulators of cadmium, mercury, lead and arsenic (Zhu et al. 2004). Several scientists have looked at water hyacinths' ability to remove arsenic from water, with somewhat differing results. Some studies have reported water hyacinths to be very effective at removing arsenic from contaminated water. Misbahuddin and Fariduddin (2002) found that just the roots of water hyacinths removed eighty-one percent from the 400 ppb arsenic solution they were in. (Note: this would still leave an arsenic concentration of 76 ppb, which is above both Bangladesh and US EPA drinking water standards.) The entire water hyacinth plant (roots, leaves, stems, etc.) was reported in the same study to have removed one hundred percent of the arsenic, and to have done so in only three to six hours. Other scientists have reported that water hyacinths do not have very high arsenic removal capabilities. Zhu, Zayed, Qian, Souza, and Terry (1999) reported that water hyacinths do not accumulate arsenic well and that most of the arsenic they take up is stored in their roots. Saha, Dikshit, and

Bandyopadhyay (2004) reported that water hyacinths in water with an arsenic level of 10,000,000 ppb remove forty-five percent of the arsenite and seventy percent of the arsenate. Zhu, Lytle, and Terry (2004) reported that water hyacinths convert a large portion of the arsenate they remove to the more toxic form of arsenic, arsenite, within the plant itself.

5.4 CONCLUSIONS

No perfect technology exists for providing safe water to poor communities plagued by the arsenic problem in Bangladesh. The pond sand filter reduces the pathogens in pond or river water by two orders of magnitude-which is not enough. Existing three-pitcher method is simple and cheap. It removes arsenic from tube well water by passing it through iron saving. Unfortunately, disposing of the contaminated sludge is a problem for arsenic filters, including larger, commercial varieties. My developed three-pitcher method shows that arsenic in groundwater can be removed by a simple filtration procedure using gravel, sand, coal and without adding chemicals. Clearly, the water quality obtained from 3-kalshi meets within Bangladeshi permissible standard and made potable water from contaminated water of near wastewater quality. The present system can be further optimized to increase the flow rate, the efficiency, and the aesthetics for a wider acceptance and use. In this respect local innovation and local participation are essential. Because groundwater is assumed to be free from pathogenic bacteria, the water quality parameters shown do not include such information. So, I urge immediate use of the 3-kalshi methods to mitigate part of the present arsenic crisis. To further this work and to understand the speciation of groundwater by chemical equilibrium models require complete analysis of cations, anions, organic humic materials, and mineral precipitates. To save millions of people from arsenic poisoning it is important to detect the arsenic concentration in groundwater and also to provide a suitable, user friendly and cost effective arsenic removal process for the rural people of Bangladesh. Unfortunately, the very first step towards prevention and arsenic testing is in great chaos. The instruments are expensive and require skilled person to operate. There is no licensing authority to supervise the testing procedure in Bangladesh. At present, very few laboratories can provide reliable result. On the other hand removal technologies so far tried for the rural people have potential but not tested thoroughly for adoption. Most of the rural people are illiterate. They developed the habit of drinking hand tube wells water during the last 30 years. So any change in their behavioral needs more friendly approach and technology.
Under the conditions of my experiment, the water hyacinths reduced the arsenic level from a Bangladeshi well water concentration (300 ppb) to EPA's proposed drinking water standard (10 ppb) for one trail, to the Bangladeshi drinking water standard (50 ppb) for two trails, and lost all ability to remove arsenic after five trails. From my water hyacinths results, there appears to be more arsenic being stored in the roots than in the stems, leaves and bladders. The fact that most of the arsenic was not extracted by water is very good. That means the arsenic is tightly bound at a molecular level so that, even when finely chopped, the arsenic did not leach out of the plants. Further research is required to retest the arsenic content of the plant using a method that does not depend on water extraction in order to determine accurately the amounts accumulated in the stems, leaves and bladders as compared to roots.

Chapter 6

ARSENIC IN GROUNDWATER OF BANGLADESH: CONTAMINATION IN THE FOOD CHAIN

6.1 Introduction

Until recently, research and remediation focused almost exclusively on drinking water. Key links between arsenic and agriculture, specifically irrigated land and crops, had been largely overlooked. In agriculture, there are also serious implications from the possible transfer of arsenic into the food chain through crops that are under irrigation with arsenic-contaminated water and then consumed by humans. The arsenic contaminations are a result of either natural geologic processes or the release of arsenic wastes by man for mining, industrial and agricultural activities. One of the key research issues is to assess reactions, movement, transformations, environmental fate and bioavailability of arsenic in landscape-soil-water-plant systems. This helps fill a knowledge gap- that is, what happens to arsenic once it is withdrawn from the aquifer and used most frequently for agricultural purposes. The problem as it relates to the food chain and human health is multifaceted (Figure 6.1). The crops most likely to absorb arsenic from irrigation are leafy vegetables. These crops can pass arsenic into food chain. Both soil and underground water of a vast area of the north and southern Bangladesh has already been threatened with arsenic contamination affecting health of millions of people. Arsenic content in soil and underground water has identified higher contamination. The experts at Bangladesh Council for Scientific and Industrial Research (BCSIR) found the highest contamination 14 mg/l of shallow tube well water in Pabna, a northern district and 220 mg/kg of soil in Sylhet area of Bangladesh. The World Health Organization (WHO) standard for arsenic is in between 0.01 and 0.05 mg/l. It is still unresolved the reason of such high contamination of arsenic in the subsurface of Bangladesh. As, water is the essential input for crop production. If water is polluted, it may be dangerous for plants, animals as well as for human being. About 40 percent of total arable land of our country is now under irrigation facilities. Most of the above-mentioned lands are irrigated with groundwater that comes from deep tube well, shallow tube well and hand tube well. Most groundwater used for irrigation in Bangladesh is contaminated with arsenic. If arsenic contaminated water is used for irrigation, it may create hazard both in soil environment and crop quality. Irrigation with arsenic contaminated groundwater increases its concentration in soil and eventually arsenic enters the food chain through crop uptake and poses long-term risk to human health. But study on the levels and distribution of arsenic in irrigated crops and vegetables are still lacking in Bangladesh. There is a growing national concern about arsenic contamination of groundwater, especially because millions of people in Bangladesh face a two-way risk of exposure to arsenic, directly through drinking water and indirectly through food crops grown arsenic contamination of soils from contaminated irrigation water. There is an urgent need for an understanding of the nature and extent of arsenic pollution of the irrigation waters, soils and crops of Bangladesh and developing water-soil-crop management practices to mitigate the arsenic problem in Bangladesh agriculture and food. Very limited work has been done on the effects of using arsenic contaminated water on crop production and it's carried over effect on food chain.

With this view in mind, the study was undertaken to find out the level of arsenic transmission from irrigation water into crops. This chapter represents review of arsenic in food chain with total exposure of human beings and arsenic content in food and determination effects of vegetable production due to arsenic contaminated irrigated water.

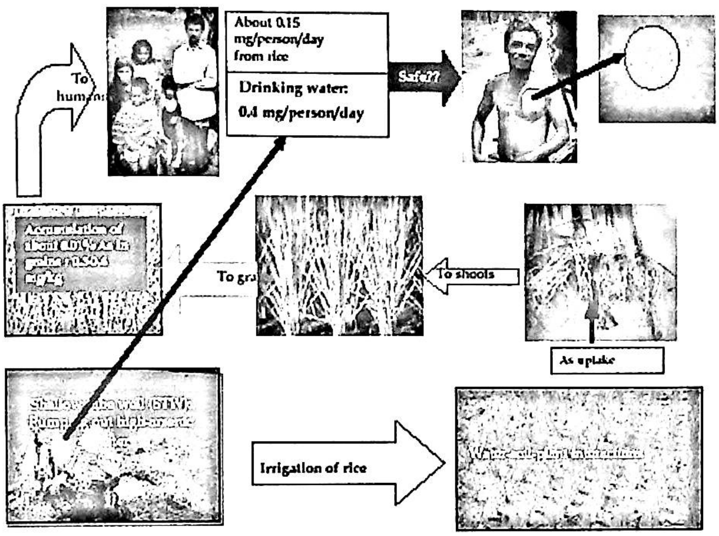


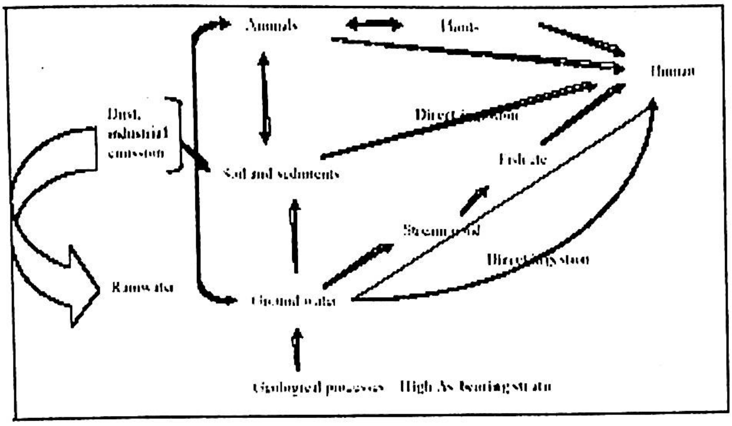
Figure 6.1: Arsenic contamination in Bangladesh Agriculture

6.2 Possibility of arsenic ingestion through consumption of different food materials

By now, the dangers of drinking arsenic-contaminated water have been well recognized. Consequently, research and studies are focusing on ensuring safe drinking water, either through mitigation techniques or through finding alternative sources of arsenic-safe drinking water. However, these studies do not discuss all potential arsenic exposure pathways that are important to animal and human systems. Even if an arsenic-safe drinking water is ensured, contaminated groundwater will continue to be used for irrigation purposes. Its use creates a risk of soil accumulation by the toxic element and eventual human exposure to it through the food chain via plant uptake and animal consumption. Between thirty and forty percent of the net cultivable area of the country is under irrigation and more than sixty percent of irrigation needs are met from groundwater, either through deep tube wells or through shallow tube wells.

The observation that arsenic poisoning amongst the population is not consistent with the level of arsenic in water has raised questions on potential pathways of arsenic ingestion. This necessitates an in-depth study on the bio-magnification of arsenic toxicity through the food chain.

Figure 6.2 Total Exposures of Human Beings to Arsenic in Nature



Reproduced from S M Imamul Huq and Naidu, Ravi, "Arsenic in Groundwater of Bangladesh: Contamination in the Food Chain," in *Arsenic Contamination in Bangladesh* (Dhaka: ITN Bangladesh 2002)

A recent study conducted by S. M. Imamul Huq of the Department of Soil, Water & Environment, Dhaka University, and Ravi Naidu of Commonwealth Scientific and Industrial Research Organization (CSIRO), Land and Water, Australia, demonstrates that apart from direct ingestion through drinking water, the major possible pathways of arsenic contamination are Soil-Crop-Food, as well as cooking water. The transfer could be schematized as Groundwater-Land-Crop-Human Beings. The study by Huq and Naidu analyzed water, soil, and vegetables/crops growing on arsenic contaminated lands in fourteen districts (out of sixty-four in the country). Fish, cooked food, and grasses were also analyzed. The study concluded that there is a possibility of arsenic ingestion through consumption of different food materials.

Research is continuing into the impacts that irrigation with arsenic-contaminated water might have on food safety. According to the National Water Management Plan (December 2001), no firm conclusion can be drawn as yet. If it is shown to be unsafe, the implications will depend on whether the health hazards are applicable to some or all crops and whether treatment is a viable option. However, if there were a need to ban irrigation from groundwater in these areas, the impacts would be moderated by the fact that most shallow tube well irrigation is not in areas of high arsenic contamination.

6.3 Arsenic in different food item

Arsenic has not only infiltrated our drinking water sources, but also started impregnating our food chain with a vengeance. Soils in Bangladesh conspicuous with a large number of irrigation pumps supplying arsenic-tainted water contained high levels of arsenic. Concentration of arsenic in the soil, particularly in the top layer, increases in direct proportion to the increase of arsenic in irrigation water. Food crops grown with arsenic laden soil imbibe arsenic in dangerous proportion (Figure 6.3). Rice samples tested in the contaminated regions have been reported to contain arsenic at unsafe levels. The

Figure 6.3: Photographic view of arsenic affected vegetables

recommended daily safe levels of dietary intake of arsenic (milligramme) for adults range between 0.171 and 0.189 and for children between 0.092 and 0.101. It is not only the raw food items that are spiked with high concentration of arsenic. Studies show that cooking doesn't rid arsenic from our food, as cooked food has also been reported to be high arsenic (Table 6.1). (Andrew Meharg 2004).

Table 6.1: Arsenic level in food items

Food items	**Arsenic level (mg/kg)**
Tuber	150
Tomato	20.1
Cabbage	7.2
Bean	5.1
Leafy vegetables	4.5
Cauliflower	2.7
Papaya	1.1
Vegetable curry	0.81
Rice	0.35
Spinach	0.33
Fish curry	0.27

Note: Maximum permissible limit – 0.2 mg/kg

Source: Andrew Meharg 2004, Arsenic in rice – understanding new disaster for South East Asia, in Trends in Plant Science, Vol. 20, No 20, pp 1-3.

6.4 Effects of vegetable production due to arsenic contaminated irrigated water

6.4.1 Experimental design

Tub experiment with vegetables (Red amaranth, Stem amaranth, Spinach and Indian spinach) was carried out. Four tubs were arranged as arsenic contaminated irrigated water with red colour mark and four types of vegetables were planted. Also, four tubs were arranged as distilled water irrigated with green colour mark for the same four types of vegetables planted parallel.

6.4.2 Sampling and experimental procedure

Soil sample was collected from barren land in which no crops were grown. Same soil sample was placed in all tubs to produce vegetables. After those four types of vegetables seed was broadcasted in each two tubs. Both distilled (for control) and arsenic contaminated water was used in tub for irrigation purpose. Samples of arsenic contaminated groundwater were taken from an agriculturally used area of shallow tube well in Faridpur district, southeastern Bangladesh, in which arsenic concentration was 0.8 mg/liter. Arsenic contaminated irrigated water was collected each week duration during experiment and stored in a container at normal temperature. All tubs were placed on farm shed to avoid addition of rainwater. After eight weeks' vegetables plants was picked up and chopped by knife to separate root, stem and leaves. Chopped plants parts were kept in the sun for one to two days for removing moisture and were labeled. Sundried samples were taken in labeled brown paper and kept in oven at 80°C for 72 hours. Precautions were taken so that arsenic could not transfer from one sample to another through knife. The dried plant samples were grinded by an electric grinder. After grinding one sample, the grinder was cleaned so that arsenic could not transfer from one sample to another. The grinded samples were immediately sent to analytical laboratory where arsenic was determined with atomic absorption spectrophotometer (HG-AAS) following USEPA method 1632.

6.4.3 laboratory analysis

Analysis of soil and plant samples were conducted in the following ways:

Briefly, the soil digestion procedure for soil was consisted of the following steps:

(i) Oven dried each segment of soil sample at 110-degree centigrade for 24 hours.
(ii) 5-gram oven-dried sample was taken in a volumetric flask, added 2.5 ml of concentrated nitric acid and 7.5 ml of concentrated hydrochloric acid to the soil sample and kept it overnight.
(iii) Heated the sample for 2 to 3 hours to boiling, then allowed it to cool and adjust the volume to 500 ml by adding deionised water
(iv) Filtered the sample and stored it for analysis.
(v) Arsenic analysis was carried out with an AAS (Shimadzu, AA6800) attached with a graphite furnace.

Briefly, the digestion procedure for vegetables were consisted of the following steps:

(i) Washed vegetable samples with distilled water
(ii) Divide the vegetable sample into parts like as root, stem and leaves
(iii) Weight of each part of the sample was taken

(iv) Oven-dried the sample at 65 degree centigrade for 24 hours and taken weight of the oven-dried sample

(v) 2 grams of dry vegetable sample in a volumetric flask was added and made it moist by adding a few millilitres of deionised water then added 25 ml nitric acid to the flask and kept it overnight

(vi) Heated the flask for two hours to boiling, then after cooling added 10 ml of perchloric acid to the flask and heated again (to boiling) for one hour,

(vii) Digestion was assumed to be complete, if colour of the sample turned yellow

(viii) When colour of the sample turned in to dark, added 2 to 3 ml of nitric acid to the flask and apply heat, repeat the process until the colour turned to yellow.

Arsenic analysis of vegetable samples was carried out with hydride generation atomic absorption spectrophotometer using an AAS (Shimadzu, AA6800).

6.4.4 Analysis

Dried soil and plant samples were digested with concentrated HNO_3 and H_2O_2. Total As in the digest and water samples were determined by flow-injection hydride generation atomic absorption spectroscopy (HG-AAS). Analytical work on arsenic was conducted BARI arsenic testing laboratory, Gazipur, Dhaka.

6.4.5 Results and Discussions

Leafy vegetables are generally grown for their tender, edible foliage. The importance of green leafy vegetables in the diet has long been recognized because they are very rich in minerals, vitamins A and C. They also supply needed roughage in the daily diet. Nutritionists recommend daily consumption of at least 116 gm of leafy vegetables. The leafy vegetables are grown throughout the year and their culture is relatively simple.

Figure 6.4: Effects of leafy vegetables due to arsenic contaminated irrigated water

Others like Stem amaranth can be grown during the summer. For that reason, leafy vegetables were selected in this investigation. My experiment showed that vegetables production affected due to arsenic contaminated irrigated water. From Figure 6.4, it can be clearly observed that plant health and growth rate was so high for distilled water irrigated as compared to arsenic contaminated irrigated water. Even Indian spinach was not germinated well in the arsenic contaminated irrigated tub. For that reason no plant analysis was conducted for the case of Indian spinach. So, it can be clearly observed that vegetable production decreases due to arsenic contaminated irrigated water.

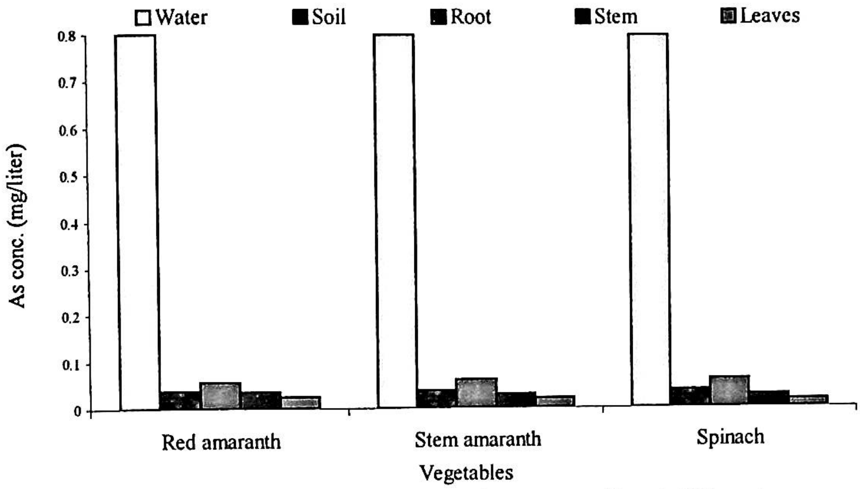


Figure 6.5 Interaction of As conc. in water, soil and different parts of vegetable plants

Figure 6.5 visualized interactions of arsenic concentration in water, soil and different parts of vegetables plants. Same amount of arsenic contaminated water 0.8 mg/l was applied in each tub. In all kind of vegetables, arsenic extractions were higher in root then stem and then leaves respectively. Similar opinion was found from other research. A study by Ali et al. (2003) revealed that high arsenic in irrigation water and soil appears to result in higher concentration of arsenic in root, stem and leaf of vegetables plants. Arsenic accumulation (0.039 mg/l) in soil at each tub remains nearly same after picked up of vegetables. Level of arsenic in red amaranth, stem amaranth and spinach leaves were 0.026, 0.022 and 0.017 mg/l respectively that was within Bangladeshi standards but exceeding WHO permissible limit. It was satisfied from other studies. Huq et al. (2003) have shown that arsenic may accumulate in the edible parts of some leafy vegetables. Among the three vegetables, stem amaranth and spinach plant roots accumulated the highest amount of As (0.062 mg/l) followed by red amaranth root (0.062 mg/l). My experiments could be verified from other studies. Ali et al. (2003) reported that the content of arsenic, in general, was higher in the vegetables grown with arsenic contaminated irrigation water than in those grown with arsenic free water. So, leafy vegetables absorb arsenic from arsenic contaminated irrigated water and arsenic pass in to the food chain. Similar opinion was found from other scientists. Groundwater is also used in the irrigation of household vegetables grown during the dry winter season. The As in the irrigation water therefore has the potential to enter the human food chain via plant uptake of As contained in the irrigation water (Imamul Huq and Naidu 2003). A relatively higher concentration of arsenic has been found in leafy vegetables, the concentration of arsenic in arum, a vegetable/food in Bangladesh has been found to be 20 mg/kg (Huq et al., 2001). Vegetables can become a path by which As may enter the food chain, because they can reflect the levels of As that exist in the environment in which they are cultivated (soil and irrigation water). Recent studies by Meharg *et al.* (2002) and Huq *et al.* (2001) demonstrate significant uptake of As by a range of vegetable crops commonly grown in Bangladesh. Numerous greenhouse studies by a number of researchers have revealed that an increase in As in cultivated soils leads to an increase in the levels of As in edible vegetables (Burló *et al.*, 1999; Carbonell-Barrachina *et al.*, 1999; Helgensen and Larsen, 1999) with many complex factors affecting bioavailability, uptake and phytotoxicity of As (Carbonell-Barrachina *et al.*, 1999).

6.5 CONCLUSIONS

Much effort has been directed towards ensuring supply of arsenic free drinking water with varying successes. Even if arsenic-safe drinking water is assured, the question of irrigating soils with arsenic-laden groundwater will continue for years to come. The possibility of arsenic accumulation in soils through irrigation water and its subsequent entry into the food chain through various food materials cannot be overlooked. It should be noted that very little is known about the chemical forms of arsenic (e.g., inorganic and organic) in vegetable, which in turn is needed for estimating its toxicity. It is apparent from the findings of the present work that there is no direct relationship with As in irrigation water and corresponding As in soil and plant. The release of As in the vegetables plants has no bearing on the total As content of irrigated water. From my study, it may be concluded that arsenic contaminated irrigated water effects production and yield of vegetables. The leafy vegetables had higher arsenic accumulation. Therefore, the use of leafy vegetables in arsenic contaminated areas would greatly increase health concern. My limited available data revealed extent and severity of arsenic contamination among different varieties of vegetable and also among different parts of the same plant. However, there is a tendency of soil build-up of As in some cases where As-contaminated ground water is used for irrigation. Therefore, it is assumed that, a large portion of arsenic is accumulating in the food that will definitely accumulate in humans via the food chain. More studies are needed to develop a better picture of arsenic accumulation in the food chain. Also, further study is needed about how much arsenic is actually absorbed by plant and then how much of that is taken under diverse conditions and farm management systems.

Chapter 7

GENERAL SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

7.1 Summary

Arsenic contamination of groundwater in Bangladesh is an important issue, while thirty percent of the water wells have an arsenic level above the drinking water standard. Review about arsenic, occurrence of arsenic, natural origin of arsenic, theory of arsenic presence in groundwater, arsenic chemistry and the difficulty of measuring arsenic, mechanism of arsenic contamination in groundwater, sources and/or causes of arsenic contamination in Bangladesh, health hazard of arsenic and social cost of arsenic contamination were studied. It's a difficult contaminant to control since it occurs naturally in most cases and cannot be detected by taste, sight or smell. Arsenic mobilization in nature depends on the source of arsenic, arsenic speciation, interactions of arsenic with soils, sediments and geologic materials and transformation and translocations by soil microbes and higher plants. Arsenic contamination does not occur in a regular, consistent pattern. It may be at various levels depending on the geographical and socio-economic environment of the locality. In the case of southeastern Bangladesh, a high degree of short-range spatial variability in the levels of arsenic has been observed. Based on the experience of arsenic contamination of groundwater in West Bengal, India, it was initially thought that arsenic contamination of ground water would be concentrated in the northeastern and central region of Bangladesh. However as more data from other regions are becoming available, it appears that arsenic contamination in Bangladesh is much more widespread. This study clearly shows that the southeastern region of the country is severely affected by arsenic contamination in groundwater. One of the major difficulties with groundwater contamination is that it occurs underground, out of sight. The pollution sources are not easily observed nor are their effects often seen until damage has occurred. The tangible effects of groundwater contamination usually come to light long after the incident causing the contamination has occurred. Using the data gathered from CIMMYT, Bangladesh, so far on various fractions of arsenic in the soil as accumulated from irrigation water, I tried to determine arsenic content in various water sources. Existing arsenic mitigation study from southern Bangladesh reveals that different national and international organizations were trying to implement various arsenic mitigation technology in scattered way. But people of these areas are not too much adapted this technology. In my study, I have found a large number of ideas for existing arsenic mitigation through filters and chemical treatment. But there is no proven affordable arsenic removal technology is developed. Different existing arsenic mitigation technologies were observed in southeastern Bangladesh that was implemented by various national and international organizations. But rural people adapted not any of the mitigation technologies. Because, peoples are not too much interested to use new technique. Arsenic concentration in groundwater in southeastern Bangladesh has shown that 41.31% under WHO guideline (0.01 mg l^{-1}), 29.69% within permissible level (0.05 mg l^{-1}) and 29% exceeds the permissible limit (>0.05 mg l^{-1}). From my results, it can be concluded that arsenic concentration did not vary with depth of water table. Also, in my research results I have not found arsenic concentration variation between dry and wet season. Data on age of tube well indicate arsenic concentration increases from eighty decade in which tube wells were installed. Groundwater sampling from the study area may or may not be representative of the shallow and deep aquifer in other areas. My experiment showed that 3-pitcher removed arsenic from arsenic contaminated groundwater filtration procedure remarkably using locally available material and without adding chemicals. Clearly, the water quality obtained from 3-kalshi setup meets and exceeds the WHO and Bangladesh standard and made potable water from contaminated water of near wastewater quality. The developed system can be further optimized to increase the flow rate, the efficiency and the aesthetics for a wider acceptance and use. I have found that water hyacinths are capable of substantially reducing arsenic levels, even to the Bangladeshi drinking water standard (50 ppb) from artificially created arsenic solution. Even also water hyacinths plant removed 50-60% arsenic from arsenic contaminated ground water. Although there was more arsenic extracted from the roots of the arsenic plants than from the upper part of the plants, arsenic was present both in the roots and upper plant. All removed arsenic stored in roots, stems and leaves of water hyacinths plants. Out of them maximum amount of arsenic was extracted by roots then stems and leaves of water hyacinths plants. From different study, it was observed that arsenic contamination in the Bangladesh agriculture via food chain was proved. Arsenic in irrigation water poses a potential threat to soils and crops especially in the leafy vegetables that is contaminating in the food chain. Most common leafy vegetables consumed by all Bangladeshi people's are red amaranth, amaranth and spinach. Edible parts of these three vegetables contaminated with arsenic level of 0.026,0.022 and 0.017 mg/l respectively in which about 0.8 mg/l arsenic contaminated water used for irrigation was observed from my experiment.

7.2 Conclusions

The results shows that the water sample analyzed of arsenic have high values above the maximum permissible limit for domestic uses as per Bangladesh and WHO standards. The studies carried out during the last decade indicate that the groundwater in the study area is being contaminated slowly and continues. there is a possibility of further contamination of groundwater and its will be hazardous to human population. There is no direct relationship with As in groundwater and corresponding As in water table depth. No clear or consistent changes in arsenic were detected in the aquifers during the short monitoring interval. However, longer-term monitoring of the wells is required to establish whether there will be significant seasonal and long-term trends in water chemistry. The relatively small variation in arsenic concentrations observed in many of the wells emphasis the need for very careful sampling and high precision analysis if seasonal or long-term trends are to be detected reliably. Arsenic from groundwater can be removed by a simple filtration procedure using locally available material and without adding chemicals. Water quality obtained from 3-kalshi set up meets and exceeds the WHO and Bangladesh standard and made potable water from contaminated water of near wastewater quality. Phytoremediation by water hyacinths (*Eichhornia crassipes*) represents a potential solution to the arsenic problem. My research work proved that water hyacinths are capable of substantially reducing arsenic levels, even to the Bangladeshi drinking water standard (50 ppb) from artificial arsenic solution and reduced arsenic about 60% from highly arsenic contaminated groundwater. Out of this around 65-70%, 20-25% and 5-10% arsenic extracted by roots, stems and leaves of water hyacinth plants. In agriculture, there are also serious implications from the possible transfer of arsenic into the food chain through crops that are under irrigation with arsenic contaminated water. The crops most likely to absorb arsenic from irrigation that is leafy vegetables and pass arsenic into the food chain. My research on arsenic contamination in food chain solve the questions about how much arsenic is absorbed by leafy vegetables due to irrigated with arsenic contaminated water.

7.3 Recommendations

During the last decades, scientists gathered information that helped greatly improve the understanding the nature and extent of the arsenic contamination problem in Bangladesh waters, soils and crops. However, there are still critical questions that need to be answered. While continuing the present activities, new research, education and technology development initiatives need to be taken. Some of these issues are pointed out below:

- Since arsenic is a cumulative poison, people living in areas of arsenic contaminated groundwater should be encouraged to eat food irrigated with surface or rain water, and/or to dilute their arsenic intake by drinking and cooking water from arsenic-free alternative sources where possible.

- There is no reason to believe that the deep aquifers will become seriously contaminated, certainly in the short term, providing that care is taken during borehole construction to isolate the upper and lower aquifers and so prevent direct leakage of contaminated water to the deep aquifer. Therefore deep wells provide a possible option for the long-term supply of safe drinking water. Ideally, the development of deep wells should be contaminated with a basic water distribution network that makes it economically feasible as well as being attractive and convenient to use. Existing wells could still be used for non-potable uses.

- A comprehensive water distribution system should be implemented and an efficient monitoring system should be established to provide potable water and to prevent future arsenic contamination in drinking water.

- Development of effective, affordable and environment friendly arsenic removal technologies for use in rural areas of Bangladesh.

- Development of a nationwide database on "background arsenic" in soils and the contribution of this arsenic in addition to irrigation water arsenic to contamination of agriculture and food.

- Take an initiative on mass awareness of the problem of arsenic in agriculture and food.

- What should be the safe levels of arsenic in irrigation water, soils and crops under the prevailing cropping systems of Bangladesh in terms of yield and crop quality?

- Guidelines on the disposal of arsenical wastes should be established to minimize the contamination in soil and water.

- A package of water-soil-crop management and cropping systems practices to avoid/manage the problem of arsenic contamination.

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PUBLICATIONS FROM THIS THESIS

i. **Iqbal, M.T** (2006) "*Arsenic Contamination in Groundwater in South-eastern Bangladesh*" AMA, Japan, Vol. 37, No.1, pp90-91.

ii. **Iqbal, M.T** (2006) "*Groundwater Arsenic Variations in Bangladesh: the Role of Depth, Seasons and Age of Tube wells*" accepted for oral presentation as a full paper at Third International Groundwater Conference (IGC-2007) to be held at Tamil Nadu Agricultural University, Coimbatore, India during February 7-10, 2007.