DEVELOPMENT AND FIELD TESTING OF A LOW-COST HOUSEHOLD ARSENIC REMOVAL UNIT

BY
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A thesis approved as to style and content
for the degree of M.Sc.in Civil and Environmental Engineering

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DECLARATION

I hereby certify that the research work reported in this thesis has been performed by me and this work has not been submitted elsewhere for any other purpose (except for publication).

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ABSTRACT

In this study, efficiency of arsenic removal from groundwater by alum and iron coagulation was evaluated. Experimental results showed that vigorous mixing for 10-15 seconds followed by 90 slow turns yielded optimum floc formation for the coagulants. This method of mixing was adopted in all coagulation experiments conducted in this study. Results of coagulation experiments suggest that ferric chloride is much more efficient than alum in removing arsenic from groundwater. As expected, removal of arsenate was much more efficient than arsenite. Potassium permanganate was used as an oxidizing agent for oxidation of arsenite to arsenate for effective removal of arsenic. Experimental results suggest that a dose of potassium permanganate twice the stoichiometric requirement is sufficient for oxidation of arsenite to arsenate. However, use of potassium permanganate produced slight pink color in the treated water, which would be objectionable to the users. This color was removed using a sand filter. Laboratory test results suggest that a sand filter 20-cm deep was sufficient for removal of color. In this study, effect of phosphate on arsenic removal efficiency was evaluated in batch sorption experiments. Results show that presence of high level of phosphate can reduce the efficiency of arsenic removal by ferric chloride to some extent.

From the results of batch experiments, a household arsenic removal unit (ARU) based on ferric chloride coagulation was designed and the ARU was tested in an arsenic-affected village in Comilla district. The unit is similar in design to the two-bucket treatment unit developed by DPHE-Danida. About 25 liters of arsenic-affected groundwater is treated in the unit in one batch, in which a chemical packet containing 2.5 gm of ferric chloride and 35 mg of potassium permanganate are added to the groundwater in the upper bucket. The cost of the unit is Tk. 520/- and chemical cost for groundwater treatment is about Tk. 0.10 per liter.

Field testing of 15 units at Adda village in Barura thana of Comilla district showed very good arsenic removal efficiency. Arsenic concentrations in the treated water were found to be mostly below 20 ppb; while maximum arsenic concentration in the tubewell water was about 400 ppb. For some of these units, presence of fecal coliform was detected in the treated water. In order to eliminate this problem, 1 mg of bleaching powder was added to the chemical packet. Continued use of bleaching powder (along with ferric chloride and potassium permanganate for a period of about 15 days) eliminated fecal coliform. Analysis of the performance of the different parts of the unit suggest that over 70 percent of total arsenic is removed the upper bucket, while the rest is removed in the sand filter in the lower bucket. The cloth strainer does not appear to contribute in arsenic removal, and hence should no longer be used in the ARU system. The developed ARU appears to be widely accepted and in great demand at the village.

List of Abbreviations:

AAS Atomic Absorption Spectrophotometer

ARP Arsenic Removal Plant

ARU Arsenic Removal Unit

BGS British Geological Survey

BTU Bucket Treatment Unit

BUET Bangladesh University of Engineering and Technology, Dhaka, Bangladesh.

DPHE Department of Public Health Engineering

IRP Iron Removal Plant

NGO Non Government Organisation

PVC Polly Vinyl Chloride

SHTW Shallow Hand Tube Well

TW Tube Well

UNU United Nations University, Tokyo, Japan

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CHAPTER 1 : INTRODUCTION

1.1 Background

Arsenic contamination of groundwater in the alluvial aquifer underlying Bangladesh and India has been recognized as a major problem of catastrophic proportions. In the early 1990s high arsenic concentrations were first reported in the groundwater of Nawabgonj, western Bangladesh. Since then, high levels of arsenic, exceeding the Bangladesh drinking water standard of 0.05 mg/l, have been detected in groundwater of many regions of the country. An estimated 268 upazillas out of 465 have so far been affected with significantly high concentrations of arsenic. Figure 1.1 shows distribution of arsenic in shallow aquifer in Bangladesh. Tubewell water extracted from shallow aquifers is the primary source of drinking/cooking water for most of its population in Bangladesh. An estimated 7.5 to 8.0 million hand-tubewells constitute the backbone of the rural water supply in Bangladesh. The urban water supply is also heavily dependent on groundwater. Thus, the detection of high levels of arsenic in groundwater in Bangladesh has put the entire water supply, especially the rural water supply, at risk.

In Bangladesh, an estimated 29 million people are exposed to arsenic concentrations in tubewell water above the national drinking water standard of 50 ppb (Ahmed et al., 2002). This has reduced the percentage of people having access to safe water from around 97% to below 80%. So far, about 8500 arsenic patients have been detected across the country (Badruzzaman, 2003). The actual number of people suffering from arsenic-related diseases is thought to be much higher. In a study by the National Institute of Preventive and Social Medicine (NIPSOM), arsenic related diseases (arsenicosis) have been identified in 37 districts (Ahmad *et. al.*, 1998). Arsenic toxicity has no known effective cure, but drinking of arsenic free water can help the arsenic affected people to get rid of the symptoms of arsenic toxicity. Provision of arsenic free drinking water is urgently needed for immediate protection of public health and well being of the people living in arsenic affected areas.

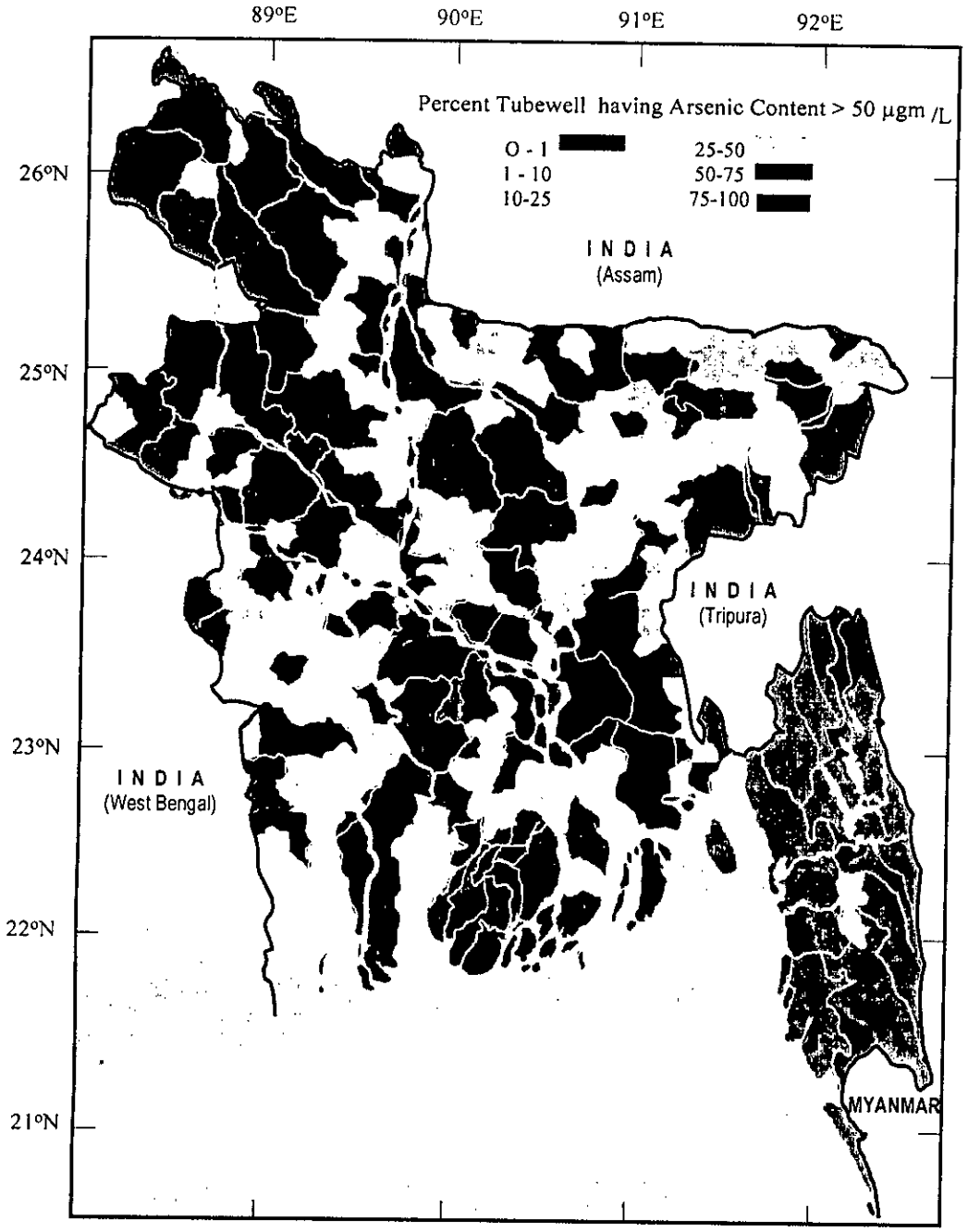

Figure 1.1: Intensity of Arsenic Contamination in Relation to Bangladesh Standard of 50 µg/L (Source : Ahmed et. al. 2002)

Broadly, supply of arsenic-free safe water can be accomplished: (i) by avoiding arsenic contaminated water by using alternate groundwater source or surface water source, and (ii) by treating the arsenic contaminated groundwater. Rainwater harvesting is another possible option. Arsenic-free groundwater development options include VSST, shallow tubewell at greater depth, deep tubewell and dug well. Surface water development options include Pond Sand Filter (PSF), infiltration gallery and household filters. All these techniques have certain advantages and disadvantages and they are not equally applicable in all areas of Bangladesh. The principal problem with surface water is bacteriological contamination. In addition, availability of surface water is not uniform throughout the year. Rainwater harvesting can be a probable alternative, but seasonal variation in rainfall pattern, proper storage of rainwater, rainwater characteristics and public acceptance are some of the issues that need to be addressed.

While arsenic-free aquifers (primarily deep aquifers) have been identified in some places; this option requires high initial investment. Groundwater treated for arsenic removal is a promising option for providing arsenic-free water to the rural population. The most commonly used technologies for arsenic removal include oxidation processes (e.g., passive sedimentation), coagulation-adsorption-coprecipitation (using alum or iron salts); adsorptive filtration (e.g., using activated alumina); ion exchange; and membrane process such as reverse osmosis.

A number of household and community based arsenic removal units (based primarily on coagulation and sorptive filtration) have been developed for use in arsenic affected areas. Available information (Ali and Nibedita, 2000; Ahmed et al., 1999; Ali et al., 1998; Hering et al., 1997; Hering et al., 1996) suggest that arsenic removal system based on ferric chloride coagulation could be a promising technique for removing arsenic from tubewell water. Success and sustainability of any arsenic removal unit depend on a number of factors including: (i) arsenic removal efficiency, (ii) ease of operation and maintenance under field condition, (iii) cost, and (iv) user acceptability. Careful evaluation of these parameters and issues is needed for ensuring safe and sustainable use of any arsenic removal unit in rural Bangladesh.

1.2 Objectives

The major objectives of this research work are:

(1) To evaluate effectiveness of alum and ferric chloride coagulation in removing arsenate and arsenite from groundwater.

(2) To evaluate the effect of various parameters (e.g. pH, PO_4^{3-}, oxidizing agent) on arsenic removal efficiency by ferric chloride coagulation.

(3) To develop a household arsenic removal unit for use in rural Bangladesh, and

(4) To conduct a long-term field-testing of the arsenic removal unit. The principal objectives of the field-testing would be to evaluate efficiency (in removing arsenic and other relevant water quality parameters) of the unit under field condition and user acceptability (e.g., in terms of water quality, cost, operation and maintenance, etc.) of the unit.

Results of this study would contribute to the understanding of the arsenic removal efficiency by ferric chloride under varying conditions and development of a low-cost arsenic removal unit for use in rural areas of Bangladesh. Efforts would be made to ensure that the developed arsenic removal unit is low-cost, easy to construct, and very simple in operation and maintenance. Results of the field-testing would be evaluated to make necessary modifications needed in the design of the arsenic removal unit.

1.3 Scope of Research

There are places in Bangladesh where all the tube wells are contaminated with high arsenic concentrations and no other safe drinking water source (e.g., a surface water source) is available in nearby localities. In such areas arsenic removal unit can be a very promising option. Elsewhere, where alternate safe water sources are available (e.g., a surface water source), it can be used in the dry seasons as a component of conjunctive water supply system. This research was limited to the optimization of an arsenic removal process, which was thought to be most suitable and easily replicable in the countryside of Bangladesh. The target users are the rural arsenic affected population who are unable to afford costly measures for arsenic free water. In the development of an optimum process for arsenic removal (by ferric chloride), typical

water requirements for drinking and cooking have been considered. During the process development, efforts were made to simulate field conditions as much as possible. No attempt was made to adjust pH of groundwater used in batch experiments; in coagulation experiments, mixing was done by manual stirring; commercial grade chemicals and locally available raw materials were used in all cases. Considering the time and resource constraints, field-testing was conducted in only one arsenic affected village (Adda village of Barura Thana) in the Comilla district.

1.4 Methodology

At the beginning of this research work, literature review was made to assess ongoing and completed work on arsenic removal processes. Literature on arsenic removal systems already in use in Bangladesh has also been reviewed. Batch studies/experiments were conducted in the laboratory to assess arsenic removal from natural groundwater for different initial arsenic concentrations and coagulant (alum or ferric chloride) doses. Batch experiments were also conducted to assess effects of various parameters (e.g., mixing energy as well as different water quality parameters) on arsenic removal efficiency.

Based on the laboratory batch test results, a ferric chloride coagulation based arsenic removal unit (ARU) has been developed and tested in the laboratory with natural as well as synthetic groundwater. Field-testing of the ARU was then done next to assess its performance in the actual conditions so that necessary adjustments could be made. The field testing was conducted in Adda village of Barura Thana of Comilla district. 15 units were set in different households in that village and being monitored for more than 7 months for arsenic in the treated water.

1.5 Organization of the Thesis

There are five chapters in this thesis including this introductory chapter. Chapter 2 presents a literature review on occurrence of arsenic, sources and uses of arsenic, and chemistry of arsenic. Chapter 2 also provides an overview of the principles of arsenic

removal from groundwater and factors and parameters affecting arsenic removal efficiency. This Chapter also provides a brief assessment of the different types of arsenic removal units that have already been developed for arsenic removal in rural Bangladesh.

Chapter 3 describes the laboratory experiments conducted to assess the removal efficiency of arsenic from groundwater under a wide range of conditions. Major laboratory works include: (i) batch experiments to determine arsenic removal efficiency for different initial arsenic (both arsenite and arsenate) concentrations as a function of coagulant (both alum and ferric chloride) dose; (ii) batch experiments to determine optimum dose of potassium permanganate for transformation of arsenite to arsenate; (iii) batch experiments to assess effect of different water quality parameters on arsenic removal efficiency. This Chapter provides a detailed analysis of the laboratory test results, in an effort to identify the coagulant and other relevant parameters (e.g., optimum coagulant and oxidant doses for a particular arsenic concentration) to be used for the development of a household arsenic removal unit.

Chapter 4 describes the actual design and construction of the household arsenic removal unit (ARU) based on ferric chloride coagulation. Chapter 4 also describes the results of the field testing and monitoring of the arsenic removal units carried out at Adda village of Barura Thana of Comilla district. Performance of the arsenic removal units was evaluated in terms of arsenic removal efficiency, effluent quality (with respect to other parameters), user acceptance of the unit, and operation and maintenance issues.

Finally, chapter 5 presents major conclusions of the study and also provides recommendations for future study.

CHAPTER 2: LITERATURE REVIEW

2.1 Introduction

Through the processes of earth materialization, the mantle was formed to contain the toxic metals in small quantities and in a way that does not affect our biosphere to any large extent. The toxic metals occur only spot-wise and mainly immobilized in geological formations. This allows groundwater to migrate through the deep soil without dissolving toxic metals in significant quantities. Similarly rainwater passes through multiple geological layers of varied chemical compositions often containing potentially toxic compounds, yet it most often comes out in springs as fresh, appropriately enriched with tasty and healthy minerals, and free of toxic substances. There is no doubt that this immobilization of toxic components in the geosphere has been of significant importance for the development and survival of mankind, as we know it today.

Like most of the toxic metals, arsenic occurs mostly immobilized in the geosphere. However, arsenic is not a typical metal. It is a so-called metalloid, exhibiting metallic as well as non-metallic characters and corresponding chemical processes. It is due to this arsenic chemistry that mankind has developed the multiple uses of arsenic, which in turn contributed to anthropogenic arsenic pollution. It is also due to this chemistry that arsenic is mobilized naturally in the biosphere. Thus arsenic is extremely toxic, even carcinogenic, and yet estimated to be the 20^{th} and 12^{th} most abundant element in earth-crust and biosphere, respectively, (Katrinen and Martin, 1995).

A good understanding of arsenic chemistry is vital for assessing the fate of arsenic in the environment and also for developing appropriate remedial measures against widespread arsenic contamination now threatening Bangladesh, India and many other countries. For example in order to understand the mobility of arsenic in the subsurface environment, it is essential to understand the geochemistry of arsenic. In this regard the important aspects of arsenic chemistry include redox reactions, adsorption-desorption reactions and precipitation-dissolution reactions. Similarly, a good

understanding of arsenic chemistry is also needed for developing efficient arsenic removal systems and disposal of arsenic-rich wastes. Toxicity of arsenic is also dependent on its chemical form. This Chapter provides a brief overview of arsenic chemistry and discusses the important chemical processes that controls its removal by different processes and governs its fate in water and soil environments.

2.2 Sources and uses of arsenic

2.2.1 Natural Sources

Arsenic bearing minerals are the primary natural sources of arsenic. There are more than 245 such minerals, mostly ores containing sulfide, along with copper, nickel, lead, cobalt and other metals, as well as some oxides. Table 2.1 provides a list of some important arsenic bearing minerals. The most important ores of arsenic are arsenopyrite or mispickel (FeAsS), realgar (As_4S_4), orpiment (As_2S_3), cludite, lollingite ($FeAs_2$), nicolite (NiAs), cobalt-glance (CoAsS), nickel-glance (NiAsS), smaltite ($CoAs_2$), and arsenolite (As_2O_3). Among these, arsenopyrite is probably the most common mineral. Weathering of rock coverts arsenic-rich metal sulfides to arsenic trioxide, which eventually finds its way into surface water, groundwater and sediments. Arsenic is often found to be associated with iron oxyhydroxides in sediments because of its affinity for iron oxyhydroxides. The oxidized forms of arsenic may be converted back to sulfides by anaerobic processes occurring on land and in sediments. Volatile forms of arsenic, e.g., arsine (AsH_3) and trimethyl arsine [$(CH_3)_3As$] enter the atmosphere from land and water and are returned by rain and atmospheric fallout. Arsenic occurs in uncontaminated soil at an average concentration of about 5 to 6 mg/kg, but this varies among geographic regions.

2.2.2 Anthropogenic Sources

Arsenic may accumulate in soil from anthropogenic activities. Arsenic is used in a variety of products. The principal arsenical compounds are herbicides, cotton desiccants, and wood preservatives and in 1990 their production rates were 8000, 12000, and 10000 tons As per year, respectively (Allaway, 1990). Arsenic may accumulate in soil through use of arsenical pesticides, dust from burning fuels (e.g,

coal with high arsenic content), and disposal of industrial and animal wastes. It is a natural contaminant in lead, zinc, gold and copper ores and can be released during the smelting process. The stack dust and flue gases from smelters often contaminate soils with arsenic, downstream from the operation. Some important arsenic-bearing products are listed in Table 2.2.

Table 2.1 : **Naturally Occurring Minerals Containing Arsenic (NRCC, 1978)**

Mineral	Formula	Mineral	Formula
Arsenite	As	Arsenolite	As_2O_3
Antimony arsenide	AsSb	Mutite	$Pb_5(PO_4,AsO_4)_3Cl$
Realger	AsS	Adamite	$Zn_2AsO_4(OH)$
Orpiment	As_2S_3	Erythrite	$Co_3AsO_4.8H_2O$
Arsenopyrite	FeAsS	Annabergite	$N_{13}(AsO_4)_2.8H_2O$
Nicolite	NiAsS	Scorodite	$(Fe.Al)AsO_4.2H_2O$
Gersdorffite	CoAsS	Pharmacosiderite	$Fe_3(AsO_4)_2OH_3$
Cobaltite	CoAsS	Olivenite	$Cu_2(AsO_4)OH$
Smaltite	(Co,Ni) As_x	Beaudanite	$PbFe_3(AsO_4)SO_4$
Skutteridite	(Co,Ni)As_x		
Loellingite	($FeAs_2$)		
Tennantite	($Cu_{12}As_4S_{13}$)		
Jordanite	$(Pb,Ti)_{13}As_7S_{23}$		
Pearcite	$Ag_{16}As_2S_{12}$		
Proustite	Ag_2AsS_3		
Energite	Cu_3AsS_4		
Rathite	$Pb_3As_5S_{10}$		

Table 2.2 : **Some Arsenic-bearing Products (Dahi, 1997)**

Name	Chemical Formula	Use
Arsenic trioxide (white arsenic)	As_2O_3	Most common chemical
Lead arsenate	$Pb_3(AsO_4)_2$	Pesticide
Lead hydrogen arsenate	$PbHAsO_4$	Pesticide
Scheele's green	$CuHAsO_3$	Paint pigment
Paris green	$3Cu(AsO_2.Cu(C_2H_3O_2)_2$	Paint pigment
Phenylarsenoic acid	-	Feed additive
Potassium arsenite	KH_2AsO_3	Fowler's solution
Arsphenamine	-	Antisyphilitic

2.2.3 *Principal compounds of arsenic and their uses*

Because arsenic has a range of oxidation states from -3 to +5, it can form a variety of different kinds of compounds. Among the most important commercial compounds are the oxides, the principal forms of which are arsenious oxide (As_2O_3) and arsenic pentoxide (As_2O_5). Arsenious oxide, commonly known as white oxide, is the material most widely used for the synthesis of arsenic compounds. It is produced as a by-product of the nonferrous metal industry, primarily from the smelting of copper ores. Naturally occurring metal arsenides, realgar and orpiment also convert to the trivalent oxide when roasted in air. The formation of the trioxide by the roasting of a sulfidic ore is illustrated in Eq. (1).

$$2\ FeAsS + 5\ O_2 \rightarrow Fe_2O_3 + As_2O_3 + 2\ SO_2 \quad (1)$$

Elemental arsenic undergoes reaction with oxygen to yield the trioxide as follows:

$$4\ As + 3\ O_2 \rightarrow 2\ As_2O_3 \quad (2)$$

The trioxide is moderately soluble in water, but dissolves easily in aqueous alkali to produce a solution of arsenic, AsO^{2-}. It is slightly soluble in polar organic solvents such as alcohols and ethers and insoluble in benzene. The most useful reagent for the synthesis of pentoxide (As_2O_5) is concentrated nitric acid. The reaction between elemental arsenic and nitric acid gives H_3AsO_4. The controlled dehydration of this acid (Eq. 3) gives the pentoxide.

$$4\ H_3AsO_4 \rightarrow 6\ H_2O + As_4O_{10} \quad (3)$$

Hypochlorous, hydrochloric and perchloric acids also oxidize the metal or As_2O_3 to the pentavalent state. Arsenic pentoxide dissolves readily in water to produce arsenic acid, H_3AsO_4.

Arsine (AsH_3) is the best known of the hydrides of arsenic. It is a colorless poisonous gas composed of arsenic and hydrogen. The gas, also called arsenic hydride, is produced by the hydrolysis of metal arsenides and by the reduction by metals (e.g., zinc) of arsenic compounds in acidic solutions. Other hydrides of arsenic are diarsine (As_2H_4), diarsine dihydride (As_2H_2), and polymeric diarsine monohydride $(As_2H)_x$.

Arsenic pentoxide, the anhydride of arsenic acid (H_3AsO_4) is very soluble in cold water and dissolves to form a solution of arsenic acid. The free acid can be obtained as a hydrate, $H_3AsO_4.0.5\ H_2O$, by the evaporation of a cold aqueous solution. Arsenic trioxide is the anhydride of arsenious acid. The solubility of arsenic trioxide in water at 25°C is 21.6 g L^{-1}. The rate of dissolution of trioxide in water is painstakingly slow, sometimes requiring up to 50 h of continuous agitation. The solubility of arsenic trioxide increases greatly and occurs much more rapidly in both acid and alkaline media. Metal salts containing orthoarsenate, AsO_4^{3-}, monohydroarsenate, $HAsO_4^{2-}$, and dihydrogen arsenate, $H_2AsO_4^-$ are known. Diarsenic disulfide, As_2S_2, but more properly written as As_4S_4, exists in nature as mineral realgar. As_4S_4 is normally prepared as an impure material and must be purified by sublimation under an atmosphere of CO_2. Diarsenic trisulfide (As_2S_3), found in nature as orpiment, has been referred to as yellow arsenic sulfide. Diarsenic pentasulfide (As_2S_5), has been described as brownish-yellow, glassy, amorphous, and highly refractive. When suspended in water and heated, it decomposes into the thermodynamically more stable As_2S_3 and free sulfur. Two binary As-P compounds have been reported. They are As_2P and AsP. Diarsenic phosphide is black and lustrous and turns brown on exposure to air. AsP is described as a lustrous and red brown powder.

Arsenic also forms numerous organic compounds, as for example, tetramethyldiarsine, $(CH_3)_2As\text{-}As\ CH_3)_2$, used in preparing the common desiccant cacodylic acid. Several complex organic compounds of arsenic have been employed in the treatment of certain diseases, such as amebic dysentery, caused by microorganism. Some of the most important compounds and species of arsenic are shown in Table 4. Figure 1 (Dahi, 1997) shows a qualitative scale indicating that the toxicity of arsenic compounds varies to a large extent depending upon their chemical form.

2.3 The occurrence of Arsenic in ground water

Some major incidents of this global phenomenon have been listed in Table 2.3.

Table 2.3 : **Major incidents of Groundwater Arsenic Contamination in different countries (De A.K., 2000)**

Location	Year	No. of People exposed	% of people with arsenical skin lesions
Taiwan, China	1961-68	103,154	19
Antofagista, Chile	1958-70	130,000	16
Lagunera, Mexico	1963-83	200,000	21
Mante Quemado, Argentina	1938-81	10,000	-
Ranpibool, Thailand	1987-88	2,800	21.6
W. Bengal, India	1983-95	1,000,000	20

Arsenic pollution of groundwater is particularly challenging in Bangladesh since tubewell water extracted from shallow aquifers is the major source of drinking water for most of its population. Estimates for 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 (DPHE/BGS/MMI., 1999; EES/DCH, 2000). In a recent survey conducted in 270 villages of Bangladesh, more than 7000 arsinicosis patients have so far been identified (Rahman et al., 2000). Arsenic toxicity has no known effective treatment, but drinking of arsenic free water can help arsenic affected people at early stage of ailment to get rid of the symptoms of arsenic toxicity. Therefore, the most important measure needed is to prevent further exposure of population by providing them with arsenic-free safe drinking water.

In the context of very high prevalence of diarrhoeal diseases in Bangladesh, bacteriological quality received priority as a criterion for drinking water supply. Groundwater is free from pathogenic micro-organisms and available in adequate

quantities in shallow aquifers, permitting development of cost effective water supply systems for scattered rural population. Groundwater abstracted by shallow tubewells was found to be the best option for rural water supply and Bangladesh achieved remarkable success by providing 97% of the rural population with tubewell water. Unfortunately, when rural people have developed the habit of drinking tubewell water, being aware of its importance to avoid diarrhoeal diseases, arsenic in excess of acceptable limits has been found in tubewell water in many parts of Bangladesh. Thousands of people are reported to have already shown symptoms of bgeing poisoned by arsenic and several millions are at risk of arsenic contamination from drinking tubewell water. The people in arsenic affected areas are likely to use unprotected surface waters to avoid arsenic poisoning and get sick by water borne/related diseases (Ahmed et al., 2000).

Most of the reported arsenic problems in water supplies are found in ground waters containing geogenic arsenic in elevated concentrations. They are caused, too, by a nonexistent or inefficient treatment for arsenic removal. The arsenic in deep well waters is often the only pollutant, while the water resource is otherwise of good to very good quality. Groundwaters in general are a preferred resource in rural areas because treatment, including a disinfection, is often not required and the groundwater extraction can be placed near consumers.

Arsenic may be also an anthropogenic pollutant of ground and surface waters, derived from chemical wastes and wastewaters. It may not be retarded effectively in groundwater movement by adsorption, but can penetrate far into the saturated zone, in a manner similar to conservative tracer.

If groundwaters with arsenic are treated for iron and manganese removal, the oxidation and filtration steps reduce the arsenic concentrations to low levels. The arsenic is not detected in the supplied water but in the water-work sludges, causing a problem with disposal of the residual solids. In the Netherlands, where arsenic is present in low levels in the groundwater, about 50% of all water-work sludges are qualified as a chemical and toxic waste: the criterion is 50 mg As/kg dry solids (Lee et at., 1991). the arsenic is, however, firmly bound to sludge (Lee et al., 1991) and the

leaching is negligible if the ferric hydroxide sludge is not dissolved by reduction or acidification.

2.4 Chemistry of Arsenic

To master the arsenic removal techniques it is necessary to know its chemistry. The common valencies of arsenic in raw water sources are +3 (arsenite) and +5 (arsenate) as shown in the inorganic hydrolysis species such as H_3AsO_3, $H_2AsO_3^-$, $HAsO_3^{2-}$, and AsO_3^{2-} and H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$, and AsO_4^{3-} (Jekel, 1994).

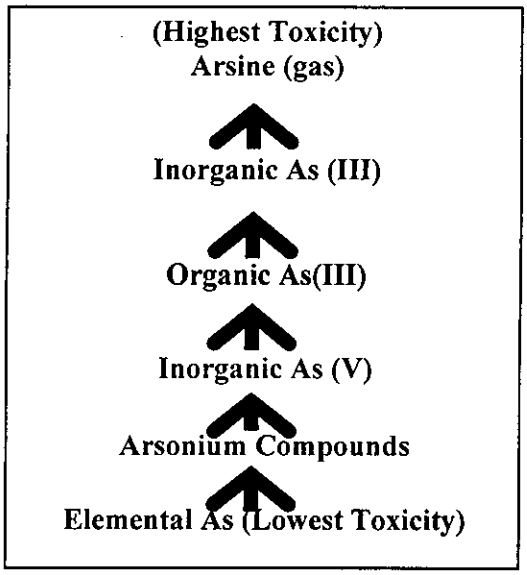

Figure 2.1: Arsenic Toxicity Scale (Dahi, 1997)

The dissociation of As(III) and As(V) acids is quite different and can be quantified by the equilibrium constants of dissociation ($pK_{a,i}$):

H_3AsO_4: $pK_{a,1} = 2.2$; $pK_{a,2} = 6.97$; $pK_{a,3} = 11.53$;

H_3AsO_3: $pK_{a,1} = 9.22$; $pK_{a,2} = 12.13$; $pK_{a,3} = 13.40$;

In typical pH ranges of natural waters, the species $H_2AsO_4^-$, $HAsO_4^{2-}$, H_3AsO_3, and to a minor degree $H_2AsO_3^-$, are dominant and determine the reactions in the removal techniques. Oxidized arsenic shows close similarity to the orthophosphate ions in terms of dissociation, precipitation, adsorption and ion exchange. The redox reaction of the As(III)/As(V) system can be described by the following equation:

$H_3AsO_4 + 2H^+ + 2e^- \rightarrow H_3AsO_3 + H_2O$ $E_0 = +0.56$ V

Reduced arsenic is found in reduced, oxygen-free groundwaters, but not in waters with dissolved oxygen. In cases there the water is extracted from different strata, the pumped water can contain both redox forms simultaneously, even though the dissolved oxygen content of mixed water is above zero (Jekel, 1994).

2.4.1 Acid-Base Chemistry

Apart from the elementary arsenic with oxidation state of 0, arsenic is stable in the oxidation states of +5, +3 and –3 (see Table 4), but generally found in water only in the trivalent and pentavalent states. The oxides of both arsenic (III) and arsenic (V) are soluble in water. The dissolution implies direct reaction with the water, hydration, where the oxides behave like non-metals and exhibit acidic character. Arsenic (III) forms arsenious acid also called arsonic acid. Arsenic (V) forms the arsenic acid, also called arsinic acid. The two acids dissociate to form respectively arsenite and arsenate ions as shown in the following reactions.

Dissociation of Arsenious Acid:

$H_3AsO_3 = H^+ + H_2AsO_3^-$ pKa = 9.22 (4)

$H_2AsO_3^- = H^+ + HAsO_3^{2-}$ pKa = 12.13 (5)

$HAsO_3^{2-} = H^+ + AsO_3^{3-}$ pKa = 13.40 (6)

Dissociation of Arsenic Acid:

$H_3AsO_4 = H^+ + H_2AsO_4^-$ pKa = 2.20 (7)

$H_2AsO_4^- = H^+ + HAsO_4^{2-}$ pKa = 6.97 (8)

$HAsO_4^{2-} = H^+ + AsO_4^{3-}$ pKa = 11.53 (9)

Figure 2.2 shows the predominance diagram of arsenic species as a function of pH. From Fig. 2.2 it is seen that arsenic acid is stronger than arsenious acid. Within the range of natural waters (particularly groundwater), where pH is usually between 6 and 9, the trivalent inorganic arsenic is found as non-dissociated arsenious acid (H_3AsO_3); while the pentavalent arsenic is primarily found as the ionized di-hydrogen arsenate ($H_2AsO_4^-$) and mono-hydrogen arsenate ($HAsO_4^{2-}$). The relatively more mobile monomethylated and dimethylated forms are observed in ocean and lake waters, but seldom in groundwater.

mol/L) in a system including oxygen, H_2O and sulfur (total concentration 10^{-3} mol/L) is shown in Fig. 2.3. The diagram represents equilibrium conditions of arsenic under various redox potentials. Well-aerated surface waters would tend to induce high *Eh* values, therefore, any arsenic present should be in the arsenate [As(V)] form. Mildly reducing conditions, such as can be found in groundwater, should produce arsenite [As(III)]. By determining the *p*H and *Eh* of water, it is possible to determine which species of arsenic will be prevalent.

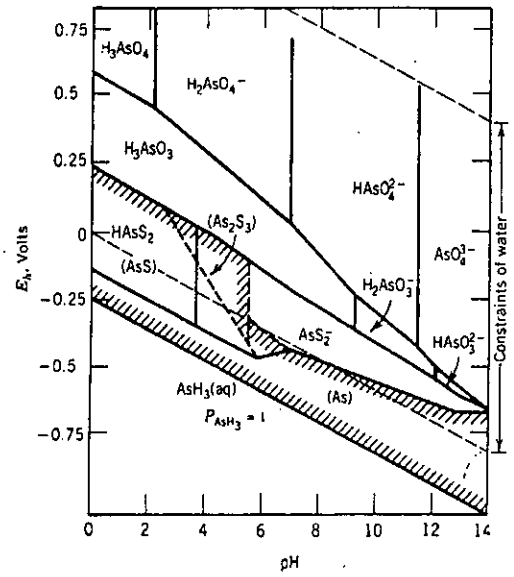

Figure 2.3: The Eh-pH diagram for As at 25°C and 1 atm with total arsenic 10^{-5} M and total sulfur 10^{-3} M. Solid species are enclosed in parenthesis in cross-hatched area, which indicates solubility less than $10^{-5.3}$ M (Montgomery, 1985)

2.4.3 Oxidation Reactions

As stated earlier, arsenate is dominant in oxygenated water while arsenite is dominant in non-oxygenated water. Although thermodynamics can provide an accurate prediction of possible changes in a given non-equilibrium conditions, they give no insight to the rate at which those changes will occur. While As(III) and As(V) acid-base reactions can be assumed to occur instantaneously, changes between oxidation states require indeterminate time periods in natural waters. For instance, the conversion of As(III) to As(V) in oxygenated water is thermodynamically favored, yet the transformation takes days, weeks or months depending on the specific conditions. The reduction of As(V) to As(III) is similarly kinetically constrained. This is the

reason why As(V) can be found in some anoxic waters (Dahi, 1997). This process is however also known to be facilitated through catalysis and require bacterial mediation.

In strongly acidic or alkaline solutions, the presence of copper salts, carbon, certain catalysts and higher temperatures can increase the arsenic oxidation rate (Ferguson and Davis, 1972). Catalytic oxidation of arsenic can be achieved by powered active carbon and dissolved oxygen in stirred reactors. The rate of oxidation can be described by a first-order equation.

The effective removal of arsenic from water requires the complete oxidation of As(III), especially if the drinking water standard is low. There are various means of oxidation available, but in drinking water treatment there are important considerations such as the limited list of safe chemicals, the residuals of oxidants, oxidation by-products and the oxidation of other inorganic and organic compounds. In the oxidation processes with dosing of chemicals, effective oxidants are free chlorine, hypochlorite, ozone, permanganate, and hydrogen per oxide/Fe^{2+} (Fenton's reagent), but not the chloramines (Frank and Clifford, 1986). These oxidants can directly transform As(III) to As(V) in the absence of oxygen. Chlorine is widely used for oxidation purpose, but may lead to chlorinated by-products, namely trihalomethenes (THMs), from reactions with natural organic matter. Ozone, widely used in surface water treatment for oxidation and disinfection, is quite effective but is not feasible for a specific application with As(III) oxidation. The most feasible oxidants are potassium permanganate and Fenton's reagent (H_2O_2/Fe^{2+}). Permanganate oxidizes As(III), ferrous and manganese ions specifically and quickly. Chlorine and permanganate are able to oxidize arsenic (III) to (V) within a very short time, e.g., half an hour or even few minutes (Dahi, 1997).

Arsenious acid oxidation by most common oxidants are shown in the following reactions (Dahi, 1997):

$$H_3AsO_3 + \tfrac{1}{2} O_2 = H_2AsO_4^- + 2H^+ \quad (10)$$

$$H_3AsO_3 + HOCl = H_2AsO_4^- + 2H^+ + Cl^- \quad (11)$$

$$H_3AsO_3 + 2/3MnO_4^- = H_2AsO_4^- + 2/3MnO_2 + 1/3H^+ + 1/3H_2O \quad (12)$$

2.4.4 *Photochemical Oxidation of Arsenic*

Besides oxidationwith chemical dosing, photo-oxidation using either UV or solar light has been used for converting arsenite to arsenate. Khoe et al. (1997) developed and patented an As-removal procedure using addition of Fe(II, III) followed by exposure to UV or solar light to accelerate oxidation of As(III). Here iron is used both as photo-absorber and co-precipitator. This procedure was initially developed to treat acidic mining effluents. Efforts to enhance As(III) photo-oxidation at higher pH values are being made, e.g., by adding S(VI) (Khoe et al., 1999).

Another arsenic removal technology – SORAS (Solar Oxidation and Removal of Arsenic), is also based on photochemical oxidation of As(III) followed by precipitation or filtration of As(V) adsorbed on Fe(III) oxides. In this method water in transparent bottles are irradiated with sunlight for oxidation of As(III) to As(V). Lemon juice was found to be most effective in enhancing the photochemical oxidation of As(III). A small amount of lemon juice is used in this process so that the pH of water (which is buffered by the presence of bicarbonate) is not changed (Wegelin et al., 1999).

UV irradiation for As(III) oxidation requires high-pressure mercury lamps with an emission spectrum between 190 and 254 nm; low -pressure mercury lamps, with their main line at 254 nm, are ineffective. The rate of oxidation can also be described by a first order rate equation, but the rate constants are considerably higher compared to the activated carbon catalysis. Nearly complete oxidation can be achieved within 30 to 60 seconds but with a high-energy input of 3 to 4 kWh/m^3 treated water (Jekel, 1994).

2.4.5 *Analysis Reactions*

Determination of arsenic by the "hydride generation" methods involve reduction of arsenic, present in water either as As(III) or As(V), into arsenic hydride or arsine (AsH_3). Arsine is insoluble in water, making it easy to purge arsenic from the water phase. It is quantitatively captured by organic solvents (e.g., silver diethyldithiocarbamate, mercuric bromide), forming colored complexes. These two

properties of arsine make it unique in the arsenic analytical chemistry and enables its detection in small quantities by the so-called Marsh's test.

In acidic solutions, arsine generation can be carried out by metallic zinc according the following reactions:

$Zn + 2\ H^+ = Zn^{2+} + H$ (*in statu nascendi*) (13)

$H_3AsO_3 + 6\ H = AsH_3 + 3H_2O$ (14)

$H_2AsO_4^- + 8\ H + H^+ = AsH_3 + 4H_2O$ (15)

AsH_3 + diethyldithiocarbamate = Coloured complex (16)

$AsH_3 + HgBr_2$ = Coloured complex (17)

Alternatively, as suggested in the latest Standard Methods (AWWA, 2000), the arsine development can be carried out using sodium borohydride, according to the following reactions:

$H_3AsO_3 + 3BH_4^- + 6H_2O + 3\ H^+ = AsH_3 + 3B(OH)_3 + 9H_2$ (18)

$H_2AsO_4^- + 5BH_4^- + 11H_2O + 6H^+ = AsH_3 + 5B(OH)_3 + 16H_2$ (19)

Reaction (18) can be performed at pH = 6, whereas reaction (19) demands strong acidification. This very important detail allows for quantitative differentiation between arsenate and arsenite. It should be noted that the methylated arsenic compounds do not take part in this arsine generation. They therefore escape the standard analytical procedures based on arsine generation. Sulfide may interfere in coloration of the reagents. It is therefore scrubbed off by gas flow through lead acetate.

2.4.6 Adsorption-Desorption

Adsorption-desorption reactions are very important in determining the mobility of arsenic in nature as well as its removal in many treatment systems. Attachment of arsenic to an iron oxide surface is an example of an adsorption reaction. The reverse of this reaction, arsenic becoming detached from such a surface is an example of desorption. Both arsenate and arsenite adsorb to surfaces of a wide range of solids including iron, aluminum and manganese oxides (e.g., iron oxyhydroxides), and clay minerals.

The strong adsorption characteristics of arsenic has been utilized in its removal from water by coagulation using alum, lime or ferric salts, where arsenic is removed primarily by adsorption onto solid flocs (e.g., aluminum hydroxide or ferric hydroxide) and subsequent precipitation. Arsenate is much more strongly adsorbed and removed than arsenite. Ferric salts have been found to be more effective in removing arsenic than alum on a weight basis and effective over a wider pH range. The strong adsorption of arsenic onto hydrous iron, aluminum and other solids has also been utilized in removing arsenic using a wide range of solid sorption media. These include activated alumina, iron coated sand, granular ferric hydroxide, and a wide range of other materials. Besides arsenic, a number of other ions present in natural water (e.g., phosphate, silicate, sulfate) also have strong affinity for solid surfaces and presence of high concentrations of these ions can reduce removal efficiency of arsenic in adsorption-based treatment systems.

Adsorption-desorption of arsenic onto iron oxide surfaces are important controlling reactions in the subsurface because iron oxides are widespread in the hydro-geologic environment as coatings on other solids, and because arsenate adsorbs strongly to iron oxide surfaces in acidic and near-neutral pH conditions. Desorption of arsenate is favored at higher (i.e., alkaline) pH values. The pH dependence of arsenate adsorption-desorption appears to be related to the change in net charge on iron-oxide surface with pH. The net charge on iron oxide surface changes from positive to negative as pH increases above the "zero-point-of-charge" (pH at which net surface charge is zero). The "zero-point-of-charge" is about 7.7 for goethite (crystalline iron oxide) and about 8.0 for ferrihydrite (amorphous iron oxide). Thus as pH increases above about 8, the net negative surface charge on iron oxides can repel the negatively charged ions such as arsenate. Compared to arsenate, arsenite is less strongly adsorbed by iron oxides. Arsenate and arsenite adsorption-desorption reactions onto other common surfaces are less well characterized.

In Bangladesh arsenic-rich iron oxyhydroxides present in aquifer materials appear to be the primary source of arsenic in groundwater. In the subsurface environment, adsorption-desorption of arsenic onto iron oxyhydroxides is an important mechanism controlling its mobility. As noted earlier, presence of ligands, which may compete

with arsenic for adsorption sites on iron oxyhydroxides, e.g., phosphate, silicate and sulfate can also influence the mobility of arsenic in the subsurface, if present in large enough concentrations. Besides oxyanions of molybdenum, selenium and vanadium can also compete with arsenic for adsorption sites.

As a result of pH dependence of arsenic adsorption, changes in groundwater pH can promote adsorption or desorption of arsenic. Similarly, redox reactions can control aqueous arsenic concentration by their effect on arsenic speciation and hence on adsorption-desorption reactions. For example reduction of arsenate to arsenite can promote arsenic mobility because arsenite is less strongly adsorbed than arsenate. It should be noted that in nature bacteria often mediate oxidation-reduction reactions.

Finally, structural changes in solid phases at the atomic level can also affect arsenic adsorption-desorption (USGS, 1999). For example, conversion of amorphous ferrihydrite to crystalline goethite may occur gradually over time (Dzombak and Morel, 1990) and this can be accompanied by a decrease in adsorption site density. This reduction in site density may result in desorption of adsorbed arsenic. Structural changes in other solid phases may also affect arsenic mobility (USGS, 1999).

2.4.7 Modeling Adsorption-Desorption Reactions

Adsorption-desorption reactions on solid surfaces are usually modeled using surface complexation approach. The fundamental concepts upon which all surface complexation models are based are as follows (Dzombak and Morel, 1990):

- Sorption on oxides takes place at specific coordination sites.
- Sorption reactions on oxides can be described quantitatively via mass law equation.
- Surface charge (on oxides) results from the sorption reactions themselves.

The effect of surface charge on sorption can be taken into account by applying a correction factor derived from the EDL theory to mass law constants for surface reactions.

A number of surface complexation models (SCMs) are available which basically differ in their description of the electrostatic component of sorption. Some important surface complexation models include: (i) constant capacitance model, (ii) diffuse layer model, (iii) triple-layer model, and (iv) generalized two-layer model.

According to the two-layer model, surface ionization reactions resulting in development of surface charge on iron oxide surfaces can be described by:

$$\equiv FeOH_2^+ = \equiv FeOH^0 + H^+ \quad ; K_{a1}^{app} \tag{20}$$

$$\equiv FeOH^0 = \equiv FeO^- + H^+ \quad ; K_{a2}^{app} \tag{21}$$

Here, K_{a1}^{app} and K_{a2}^{app} are "apparent" equilibrium constants, because they include surface charge effect and hence are dependent on extent of surface ionization. The mass law equations for the above reactions in terms of apparent equilibrium constants can be written as follows:

$$K_{a1}^{app} = (\equiv FeOH^0)\,(H^+) / (\equiv FeOH_2^+) \tag{22}$$

$$K_{a2}^{app} = (\equiv FeO^-)\,(H^+) / (\equiv FeOH^0) \tag{23}$$

Although it is impossible to separate experimentally the chemical and electrical contributions to total sorption energy (Dzombak and Morel, 1990), in SCMs these two are separated theoretically in order to obtain a specific (i.e., chemical) interaction term that does not vary with surface charge. A variable electrostatic interaction term is then added, resulting in a model that accounts for observed variations in effective mass law constants for sorption reactions. For the two layer model (Dzombak and Morel, 1990), we can write:

$$K^{app} = K^{int} \exp(-\Delta Z F \Psi / RT) \tag{24}$$

where, K^{int} is the intrinsic equilibrium constant that does not depend on surface charege, and K^{app} is the apparent equilibrium constant. Here Ψ is the surface potential, ΔZ is the change in charge of surface species due to sorption reaction, and the exponential term [exp $(-\Delta Z F \Psi/RT)$] is commonly referred to as electrostatic or coulombic correction factor. Thus the mass law equations given by Eqs. 22 and 23 can be written as:

$$K_{a1}^{int} \exp(F\Psi/RT) = (\equiv FeOH^0)\,(H^+) / (\equiv FeOH_2^+) \tag{25}$$

$$K_{a2}^{int} \exp(F\Psi/RT) = (\equiv FeO^-)\,(H^+) / (\equiv FeOH^0) \tag{26}$$

Fitting of the model to experimental data enables determination of intrinsic equilibrium constants for surface reactions.

Adsorption-desorption reactions of arsenate and arsenite on hydrous ferric oxide modeled using the generalized two-layer model (Dzombak and Morel, 1990) are shown by the following reactions:

Arsenate Adsorption:

$$\equiv FeOH^0 + AsO_4^{3-} + 3\ H^+ = \equiv FeH_2AsO_4^0 + H_2O \quad (27)$$

$$\equiv FeOH^0 + AsO_4^{3-} + 2\ H^+ = \equiv FeHAsO_4^- + H_2O \quad (28)$$

$$\equiv FeOH^0 + AsO_4^{3-} + H^+ = \equiv FeAsO_4^{2-} + H_2O \quad (29)$$

$$\equiv FeOH^0 + AsO_4^{3-} = \equiv FeOHAsO_4^{3-} + H_2O \quad (30)$$

Arsenite Adsorption:

$$\equiv FeOH^0 + H_3AsO_3 = \equiv FeH_2AsO_3^0 + H_2O \quad (31)$$

Possible desorption of arsenate in the presence of phosphate ions are shown by the following reactions:

$$\equiv FeH_2AsO_4^0 + PO_4^{3-} = \equiv FeH_2PO_4^0 + AsO_4^{3-} \quad (32)$$

$$\equiv FeHAsO_4^- + PO_4^{3-} = \equiv FeHPO_4^- + AsO_4^{3-} \quad (33)$$

$$\equiv FeAsO_4^{2-} + PO_4^{3-} = \equiv FePO_4^{2-} + AsO_4^{3-} \quad (34)$$

2.4.8 Precipitation and Dissolution

Precipitation-dissolution reactions are important mechanisms controlling mobility of arsenic in the subsurface. Arsenic contained within solid phases, either as a primary structural component of arsenic bearing minerals (e.g., arsenopyrite) or an impurity in any of a variety of solid phases (e.g., pyrite), is released to groundwater when these solid phases dissolve. Similarly, arsenic is removed from groundwater when solid phases containing arsenic precipitate from aqueous phase. As an example, because arsenic often coprecipitates with iron oxide, iron oxides may act as an arsenic source (case of dissolution) or a sink (case of precipitation) for groundwater (USGS, 1999). Besides, solid phase dissolution will contribute not only arsenic contained within that phase, but also any arsenic adsorbed to the solid-phase surface. In Bangladesh,

reductive dissolution iron oxyhydroxides and consequent release of adsorbed arsenic could be an important mechanism of arsenic mobilization in the subsurface.

Oxidative dissolution reactions (Bhumba and Keefer, 1994) of arsenopyrite (FeAsS) (Eq. 35-36), Orpiment (As_2S_3), (Eq. 37) and Reagler (AsS) (Eq. 38) are shown below. Oxidative dissolution of pyrite (FeS_2) is shown in Eq. 39-41 (Chowdhury et al., 1998).

$$4FeAsS + 11O_2 + 6H_2O = 4FeSO_4 + 4H_2AsO_3^- + 4H^+ \quad (35)$$

$$4FeAsS + 13O_2 + 6H_2O = 4FeSO_4 + 4H_2AsO_4^- + 4H^+ \quad (36)$$

$$As_2S_3 + 7O_2 + 6H_2O = 2H_3AsO_4 + 6H^+ + 3SO_4^{2-} \quad (37)$$

$$4AsS + 11O_2 + 10H_2O = 4H_3AsO_4 + 8H^+ + 4SO_4^{2-} \quad (38)$$

$$2FeS_2 + 7O_2 + 2H_2O = 2Fe^{2+} + 4HSO_4^- \quad (39)$$

$$4\,Fe^{2+} + O_2 + 4H^+ = 4Fe^{3+} + 2H_2O \quad (40)$$

$$FeS_2 + 14Fe^{3+} + 8H_2O = 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \quad (41)$$

The interplay of redox reactions and solid phase precipitation and dissolution may be particularly important with regard to aqueous arsenic and solid-phase iron oxides and sulfide minerals (USGS, 1999). High concentrations of arsenic often are associated with iron oxides and sulfide minerals. Iron oxides frequently dissolve under reducing conditions (e.g., in the presence of organic matter, as shown in Eq. 42), but often precipitate under oxidizing conditions. Sulfide minerals generally are unstable under oxidizing conditions, but may precipitate under reducing conditions (e.g., precipitation of As_2S_3, as shown in Eq. 43). Thus, as a result of redox sensitive nature of iron oxides and sulfide minerals, transfer of large amounts of arsenic between these solid phases and neighboring water may result from redox-facilitated precipitation and dissolution reactions (USGS, 1999).

$$Fe(OH)_3(s) + ¼\,CH_2O + 2H^+ = Fe^{2+} + ¼\,CO_2 + 11/4\,H_2O \quad (42)$$

$$2H_3AsO_3 + 6H^+ + 3S^{2-} = As_2S_3 + 6H_2O \quad (43)$$

Precipitation of arsenic has been utilized in the removal of arsenic from water. The insolubility of certain inorganic arsenic (V) compounds is the basis of many hydrometallurgical arsenic removal processes (Robins et al., 2001). The most common methods of removing arsenic from aqueous systems are by precipitation as arsenic (III) sulfide, calcium arsenate, or ferric arsenate. The sulfide As_2S_3 has its

lowest solubility as pH 4, but this solubility is significantly higher than has been generally accepted (Robins et al., 2001). A number of calcium arsenates [e.g., $Ca_3(AsO_4)_2$] can be precipitated from As(V) solutions by lime addition to high pH. Arsenic (V) can also be precipitated from process solutions below about pH 2 with Fe(III) to form ferric arsenate, $FeAsO_4.2H_2O$. Other solids of interest include ferrous arsenate [$Fe_3(AsO_4)_2.xH_2O$], calcium-arsenate-phosphate [$Ca_{10}(AsO_4,PO_4)_6(OH)_2$], and ferric sulfide [$Fe_2S_2$]. Some other metal arsenates, such as those of Fe(II), Zn(II), Cu(II) and Pb(II) are less soluble and more stable in the neutral pH region than calcium arsenates and ferric arsenate, but these have not been seriously considered as disposal forms (Robins et al., 2001). Barium (II) arsenate was proposed to as being an extremely insoluble arsenate, but this was shown to be incorrect. More complex compounds, such as the apatite structured calcium phosphate-arsenate have recently been demonstrated to be of low solubility and of appropriate stability for disposal considerations. Ferric arsenite sulfate is also of recent interest and may prove to be useful in stabilizing arsenic (III) (Robins et al., 2001). A number of mixed oxidation state materials [both Fe(II)-Fe(III) and As(III)-As(V)] are currently being studied (Robins et al., 2001).

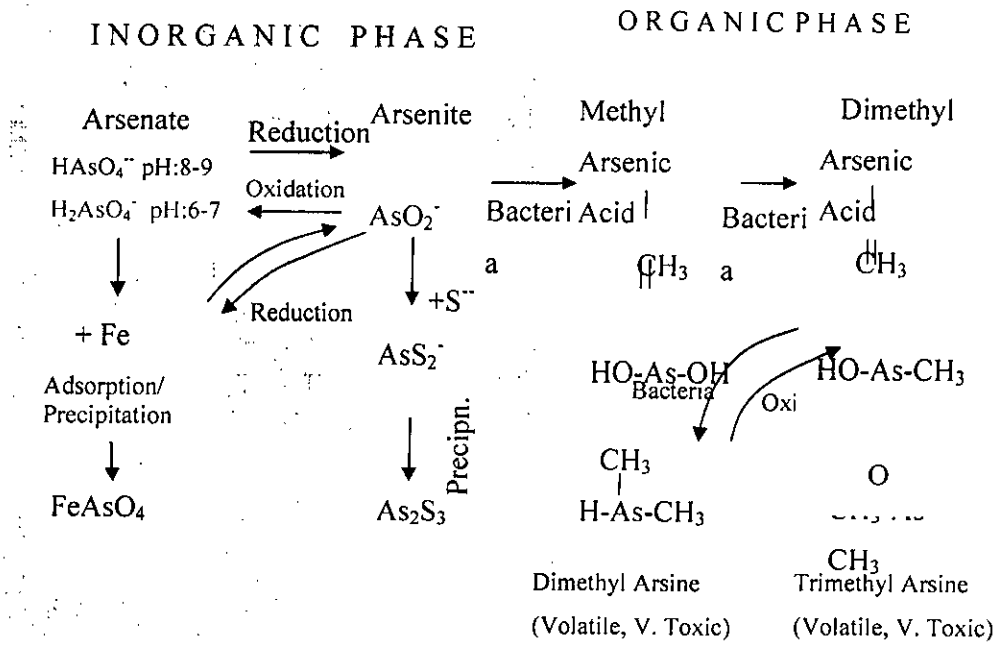

Figure 2.4 : Chemical forms of arsenic and their transformations in soils (Bhumba and Keefer, 1994)

2.4.9 Arsenic in Soil

In uncontaminated soils, average arsenic concentration varies from about 5 to 6 mg/kg, but this varies among geographic regions. However, significantly high arsenic concentrations have been found in agricultural soil irrigated with arsenic contaminated groundwater. Concentrations as high as 51 mg/kg and 83 mg/kg have been reported in soils of Faridpur and Comilla districts, respectively of Bangladesh (Ullah, 1998). A concentration varying from 1.5 to 19 mg/kg showing higher concentration in the top layers of soil has been found in Samta village in Jessore (Kubota, 1998).

2.4.10 Chemical Forms of Arsenic in Soils

The chemical forms of arsenic in soil have been illustrated in Fig. 4 (Bhumba and Keefer, 1994). The reactions and processes involved in the transformation of different forms of arsenic include oxidation, reduction, adsorption dissolution, precipitation and volatilization. Bacteria and fungi play important roles in chemical transformation of arsenic to volatile arsine gases, which are extremely toxic. Limited data suggest presence of both As(III) and As(V) as well as organic arsenic in agricultural soil and the processes of transformation/bio-transformation of arsenic are not clearly understood. Soil components that contribute to arsenic sorption and retention include oxides of Al, Fe and Mn, soil mineralogy, and organic matter.

2.4.11 Methylation Reactions

Arsenic taken by mammals is subject to either direct excretion, direct accumulation in some parts of the body (e.g., nails, hair and skin tissue), or to biotransformation in the form of methylation. Besides microbial processes has also been utilized in the bioremediation of arsenic contaminated soils.

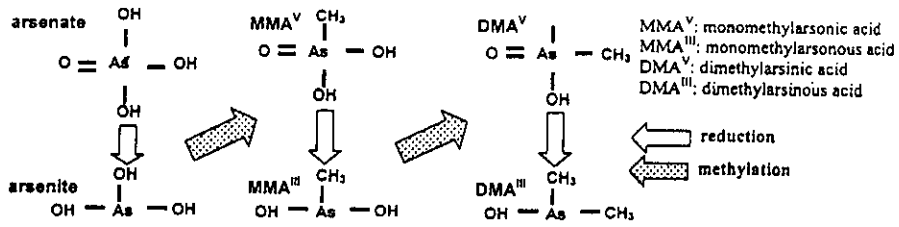

Figure 2.5: Reduction and methylation reactions in the metabolism of arsenic (Suzuki, 2002)

Humans are exposed to arsenic mostly in the forms of arsenate/arsenite and organic arsenosugars/arsenobetaines and marine products. The inorganic forms are more toxic than organic forms. Methylation seems to be most important pathway of biotransformation of inorganic arsenic. The inorganic forms are metabolized by consecutive reduction and methylation reactions in humans and mammals to dimethylated arsenic (DMA) (see Fig. 5), which is excreted into urine (Suzuki, 2002). The toxicity of arsenite is highly dependent on animal species, which in turn depends on the differences in the metabolism shown in Fig. 2.5. The methylation process leading to DMA was believed to be the detoxification pathway, but recent studies document it as toxification pathway (Suzuki, 2002). Research works are being carried out to better understand these processes.

Several fungi and bacterial species have been demonstrated to methylate inorganic arsenic by an initially reducing arsenate fraction to arsenite, which then is methylated and released to the environment (Kartinen and Martin 1995). However, the concentration of methylated arsenic in the natural waters, whether ground or surface, is normally low. This is because the methylated arsenic is taken up by the biota where it undergoes metabolic conversion into organic arsenical. Compounds like arsenobetaine and arsenocholine, can thus be found in fish and crustaceans. These compounds do not have any toxicological significance. Upon consumption by man they are directly excreted through urine without any biotransformation (Vahter, 1994).

2.5 Arsenic Removal Technologies: Principles

This section provides a brief description of the important principles used for arsenic removal in different household and community based arsenic removal plants, based on the detailed review of the technologies by Ahmed (2001).

2.5.1 Removal techniques based on oxidation of trivalent arsenic

Arsenic is present in groundwater in As(III) and As(V) forms in different proportions. Most treatment methods are effective in removing arsenic in pentavalent form and hence include an oxidation step as preteatment to convert arsenite to arsenate. Arsenite can be oxidized by oxygen, ozone, free chlorine, hypochlorite, permanganate, hydrogen peroxide and fulton's reagent but Atmospheric oxygen, hypochloride and permanganate are commonly used for oxidation in developing countries. Oxidation reactions have been described in section 2.4.3.

Passive Sedimentation

Passive sedimentation received considerable attention because of rural people's habit of drinking stored water from pitchers. Oxidation of water during collection and subsequent storage in houses may cause a reduction in arsenic concentration in stored water (*Bashi Pani*). Experiments conducted in Bangladesh showed zero to high reduction in arsenic content by passive sedimentation. Arsenic reduction by plain sedimentation appears to be dependent on water quality particularly the presence of precipitating iron in water. Ahmed et al.(2000) showed that more than 50% reduction in arsenic content is possible by sedimentation of tubewell water containing 380-480 mg/L of alkalinity as $CaCO_3$ and 8-12 mg/L of iron but cannot be relied to reduce arsenic to desired level. Most studies showed a reduction of zero to 25% of the initial concentration of arsenic in groundwater. In rapid assessment of technologies passive sedimentation failed to reduce arsenic to the desired level of 50 μg/L in any well (BAMWSP, DFID, WaterAid , 2001).

In-situ Oxidation

In-situ oxidation of arsenic and iron in the aquifer has been tried under DPHE-Danida Arsenic Mitigation Pilot Project. The aerated tubewell water is stored in a tank and

released back into the aquifers through the tubewell by opening a valve in a pipe connecting the water tank to the tubewell pipe under the pump head. The dissolved oxygen in water oxidizes arsenite to less mobile arsenate and also the ferrous iron in the aquifer to ferric iron, resulting a reduction in arsenic content in tubewell water. The possible reactions of arsenate to ferric hydroxide are shown in Equations (27) to (34) of section 2.4.7. Experimental results show that arsenic in the tubewell water following in-situ oxidation is reduced to about half due to underground precipitation and adsorption on ferric iron.

Solar Oxidation

SORAS is a simple method of solar oxidation of arsenic in transparent bottles to reduce arsenic content of drinking water (Wegelin et al., 2000). Ultraviolet radiation can catalyze the process of oxidation of arsenite in presence of other oxidants like oxygen (Young, 1996). Experiments in Bangladesh show that the process on average can reduce arsenic content of water to about one-third. Energy requirements for this type of oxidation have been described in section 2.4.4.

2.5.2 Removal techniques based on co-precipitation and adsorption process

Water treatment with coagulants such as aluminum alum, $Al_2(SO_4)_3.18H_2O$, ferric chloride , $FeCl_3$ and ferric sulfate $Fe_2(SO_4)_3.7H_2O$ are effective in removing arsenic from water. Ferric salts have been found to be more effective in removing arsenic than alum on a weight basis and effective over a wider range of pH. In both cases pentavalent arsenic can be more effectively removed than trivalent arsenic.

In the coagulation-flocculation process aluminum sulfate, or ferric chloride, or ferric sulfate is added and dissolved in water under efficient stirring for one to few minutes. Aluminum or ferric hydroxide micro-flocs are formed rapidly. The water is then gently stirred for few minutes for agglomeration of micro-flocs into larger easily settable flocs. During this flocculation process all kinds of micro-particles and negatively charged ions are attached to the flocs by electrostatic attachment. Arsenic is also adsorbed onto coagulated flocs. As trivalent arsenic occurs in non-ionized form, it is not subject to significant removal. Oxidation of As(III) to As(V) is thus required as a pretreatment for efficient removal. This can be achieved by addition of

bleaching powder (chlorine) or potassium permanganate as shown in Equations 11 and 12 of section 2.4.3. The possible chemical equations of alum coagulation are as follows:

Alum dissolution:

$$Al_2(SO_4)_3.18H_2O = 2Al^{3+} + 3SO_4^{2-} + 18H_2O \quad (44)$$

Aluminium precipitation(acidic):

$$2Al^{+++} + 6H_2O = 2Al(OH)_3 + 6H^+ \quad (45)$$

Co-precipitation (Non-stoichiometric, non-defined product):

$$H_2AsO_4^- + Al(OH)_3 = \text{Al-As (complex)} + \text{Other Products} \quad (46)$$

Arsenic adsorbed on aluminiun hydroxide focs as Al-As complex is removed by sedimentation. Filtration may be required to ensure complete removal of all flocs. Similar reactions take place in case of ferric chloride and ferric sulfate with the formation of Fe-As complex as end product, which is removed by the process of sedimentation and filtration.

The possible reactions of arsenate with hydrous iron oxide have been described in section 2.4.7. Arsenic removal is dependent on pH. In alum coagulation, the removal is most effective in the pH range 7.2 ~ 7.5 and in iron coagulation, efficient removal is achieved in a wider pH range usually between 6.0 and 8.5 (Ahmed and Rahaman, 2000).

Naturally Occurring Iron

The use of naturally occurring iron precipitates in ground water in Bangladesh is a promising method of removing arsenic by adsorption. It has been found that hand tubewell water in 65% of the area in Bangladesh contains iron in excess of 2 mg/L and in many acute iron problem areas, the concentration of dissolved iron is higher than 15 mg/L. Although no good correlation between concentrations of iron and arsenic has been derived, iron and arsenic have been found to co-exist in ground water. Most of the tubewell water samples satisfying Bangladesh Drinking Water Standard for Iron (1 mg/L) also satisfy the standard for Arsenic (50 μg/L). Only about 50% of the samples having iron content 1 - 5 mg/L satisfy the standard for

arsenic while 75% of the samples having iron content > 5 mg/L are unsafe for having high concentration of arsenic.

The iron precipitates [$Fe(OH)_3$] formed by oxidation of dissolved iron [$Fe(OH)_2$] present in groundwater, as discussed above, have the affinity for the adsorption of arsenic. Only aeration and sedimentation of tubewell water rich in dissolved iron has been found to remove arsenic. The Iron Removal Plants (IRPs) in Bangladesh constructed on the principles of aeration, sedimentation and filtration in a small units have been found to remove arsenic without any added chemicals. The conventional community type IRPs, depending on the operating principles, more or less work as Arsenic Removal Plants (ARPs) as well. A study suggests that As(III) is oxidized to As(V) in the IRPs to facilitate higher efficiency in arsenic removal in IRPs constructed in Noakhali (Dahi and Liang, 1998). The Fe-As removal relationship with good correlation in some operating IRPs has been plotted in Figure 4. Results shows that most IRPs can lower arsenic content of tubewell water to half to one-fifth of the original concentrations. The efficiency of these community type Fe-As removal plants can be increased by increasing the contact time between arsenic species and iron flocs. Community participation in operation and maintenance in the local level is absolutely essential for effective use of these plants.

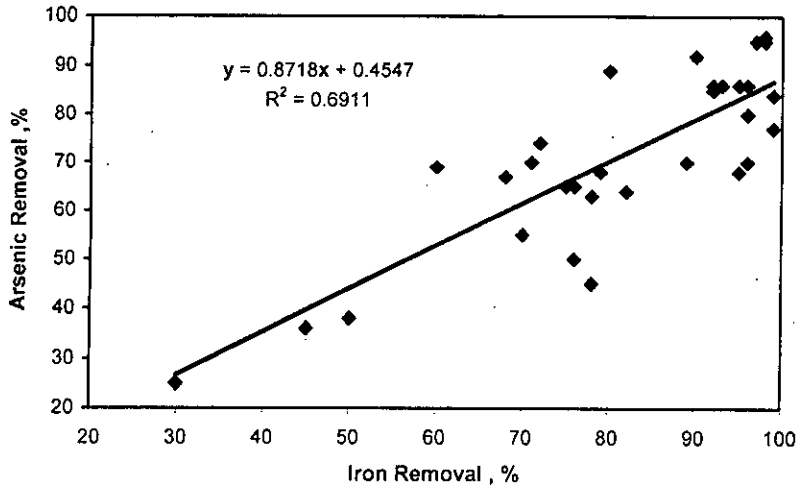

Figure 2.6: Correlation between Fe and As Removal in Treatment Plants (Source : Ahmed, 2001)

2.5.3 *Removal techniques using lime treatment*

Water treatment by addition of quick lime, CaO, or hydrated lime, $Ca(OH)_2$ also removes arsenic. The precipitated calcium hydroxide, $Ca(OH)_2$ acts as a sorbing flocculent for arsenic. Excess lime will not dissolve, but remains as a thickener and coagulant aid, which has to be removed along with precipetates through a sedimentation-filtration process. It has been observed that the arsenic removal by lime is relatively low, usually between 40 and 70% (Ahmed et el., 2000). The highest removal is achieved at pH 10.6 to 11.4. Obviously the water treated by lime would require secondary treatment in order to adjust the pH to an acceptable level. Lime softening may be used as a pre-treatment to be followed by alum or iron coagulation.

2.5.4 *Removal by sorptive media*

Several sorptive media have been reported to remove arsenic from water. These are activated alumina, activated carbon, iron and manganese coated sand, kaolinite clay, hydrated ferric oxide, activated bauxite, titanium oxide, silicium oxide and many natural and synthetic media. The efficiency of all some sorptive media depend on the use of oxidizing agent as aids to sorption of arsenic. Saturation of media by different contaminants and components of water takes place at different times of operation depending on the specific sorption affinity of the medium to the given component. Saturation means that the efficiency in removing the desired impurities becomes zero.

2.5.5 *Removal techniques based on ion exchange*

This process is similar to that of activated alumina, but the medium is a synthetic resin of more well-defined ion exchange capacity. The process is normally used for removal of specific undesirable cations or anions from water. As the resin becomes exhausted, it needs to be regenerated. The arsenic exchange and regeneration equations with a common salt solution as regeneration agent are as follows:

Arsenic exchange: $2R\text{-}Cl + HAsO_4^{2-} = R_2HasO_4 + 2Cl^-$

Regeneration : $R_2HasO_4 + 2Na^+ + 2Cl^- = 2R\text{-}Cl + HAsO_4^{2-} + 2Na^+$

Where R stands for exchange resin.

The arsenic removal capacity is dependent on sulfate and nitrate contents of raw water as sulfate and nitrate are exchanged before arsenic. The ion exchange process is less dependent on pH of water. The efficiency of ion exchange process is radically improved by pre-oxidation of As(III) to As(V) but the excess of oxidant often needs to be removed before the ion exchange in order to avoid the damage of sensitive resins. Development of ion specific resin for exclusive removal of arsenic can make the process very attractive.

2.5.6 Removal by membrane techniques

Membrane techniques like reverse osmosis, nanofiltration and electrodialysis are capable of removing all kinds of dissolved solids including arsenic from water. In this process water is allowed to pass through special filter media which physically retain the impurities present in water.

Demineralization of water can be accomplished using micro-porous membrane. There are two basic modes of operation in use. One system uses pressure to drive water through the membrane against the force of osmotic pressure and is called reverse osmosis, even though the pressure applied is several orders of magnitude in excess of the natural osmotic pressure. The other process, called electro-dialysis, uses electrical forces to drive ions through ion-selective method.

Reverse osmosis or electro-dialysis can be effective process for arsenic removal, but may be applied only if partial or total desalting is necessary in addition to arsenic separation (Jekel, 1994). Clifford (1986) pointed out that in reverse osmosis, only As(V) is effectively removed (98-99%; initial concentration up to 2 ppm), while As(III) is only partially separated (46-75%) due to neutral form of As(III). It is a recondition that the water does not contain any suspended solids and that arsenic is in its pentavalent state (Dahi, 19978). Most membranes, however, cannot withstand oxidizing agents. Moreover, these methods are already of no interest in developing countries, because of their nature as high technology and high cost (Dahi, 1997)

2.5.7 Microbial processes (biomethylation) of arsenic removal

Microbial removal of arsenic is based on two important metal-microbe interactions: (i) microbial oxidation of As(III) to As(V) to facilitate its removal by conventional arsenic removal processes and (ii) bioaccumulation of arsenic in bacterial biomass from the surrounding water environment. There are a number of microorganisms capable of oxidizing arsenite at neutral pH. The common iron bacteria, which oxidizes ferrous iron to ferric iron can oxidize as well as absorb arsenic. Removal of trace metal from water through accumulation in algae is well recognized. Several form of algae are known to assimilate arsenic from water in a biological process. Arsenic can conventionally be oxidized from As(III) to As(V), adsorbed or assimilated through microbial growth in a simple reactor in nutritionally balanced condition at appropriate temperature and pH and subsequently removed by precipitation/filtration. Microbial growth on fixed media or suspended growth should be equally effective for arsenic removal.

2.6 Household and Community-based Arsenic Removal Units

Several household and community based arsenic removal systems have been developed over the last several years in Bangladesh and many of these units have been testes either at bench scale or at field level. This section provides a brief description of the important household and community based arsenic removal units, based on the detailed review of the technologies by Ahmed (2001).

The most commonly used technologies include oxidation, co-precipitation and adsorption onto coagulated flocs, lime treatment, adsorption onto sorptive media, ion exchange resin and membrane techniques (Cheng et al., 1994; hering et al.,1996, 1997; kartinen and Martin, 1995; Shen, 1973; Joshi and Chaudhuri, 1996). Sorg and Logsdon (1978) present a detailed review of arsenic removal technologies. Jackel (1994) has documented several advances in arsenic removal technologies. In view of the drinking water standards by USEPA, a review of arsenic removal technologies was made to consider the economic factors involved in implementing lower drinking water standards for arsenic (Chen et al., 1999). Many of the arsenic removal technologies have been discussed in details in AWWA reference book (Pontious,

1990). A comprehensive review of low-cost, well-water treatment technologies for arsenic removal with the list of companies and organizations involved in arsenic removal technologies has been compiled by Murcott (2000). Some of the units, used in Bangladesh, have been described in the following section.

2.6.1 Bucket Treatment Unit

The Bucket Treatment Unit (BTU), developed by DPHE-Danida Project is based on the principles of coagulation, co-precipitation and adsorption processes. It consists of two buckets, each 20 liter capacity, placed one above the other. Chemicals are mixed manually with arsenic contaminated water in the upper red bucket by vigorous stirring with a wooden stick for 30 to 60 seconds and then flocculated by gentle stirring for about 90 second. The mixed water is then allowed to settle for 1- 2 hours. The water from the top red bucket is then allowed to flow into the lower green bucket via plastic pipe and a sand filter installed in the lower bucket. The flow is initiated by opening a valve fitted slightly above the bottom of the red bucket to avoid inflow of settled sludge in the upper bucket. The lower green bucket is practically a treated water container.

The DPHE-Danida project in Bangladesh distributed several thousands BTU units in rural areas of Bangladesh. These units are based on chemical doses of 200 mg/L aluminum sulfate and 2 mg/L of potassium permanganate supplied in crushed powder form. The units were reported to have very good performance in arsenic removal in both field and laboratory conditions (Sarkar et al., 2000 and Kohnhorst and Paul, 2000). Extensive study of DPHE-Danida BTU under BAMWSP, DFID, WaterAid (2001) rapid assessment program showed mixed results. In many cases, the units under rural operating conditions fails to remove arsenic to the desired level of 0.05 mg/L in Bangladesh. Poor mixing and variable water quality particularly pH of groundwater in different locations of Bangladesh appeared to be the cause of poor performance in rapid assessment.

2.6.2 Stevens Institute Technology

This technology also uses two buckets, one to mix chemicals (reported to be iron sulphate and calcium hypochloride) supplied in packets and the other to separate flocs

by the processes of sedimentation and filtration. The second bucket has a second inner bucket with slits on the sides as shown in Figure 2.7 to help sedimentation and keeping the filter sand bed in place. The chemicals form visible large flocs on mixing by stirring with stick. Rapid assessment showed that the technology was effective in reducing arsenic levels to less than 0.05 mg/L in case of 80 to 95% of the samples tested(BAMWSP, DFID, WaterAid , 2001). The sand bed used for filtration is quickly clogged by flocs and requires washing atleast twice a week.

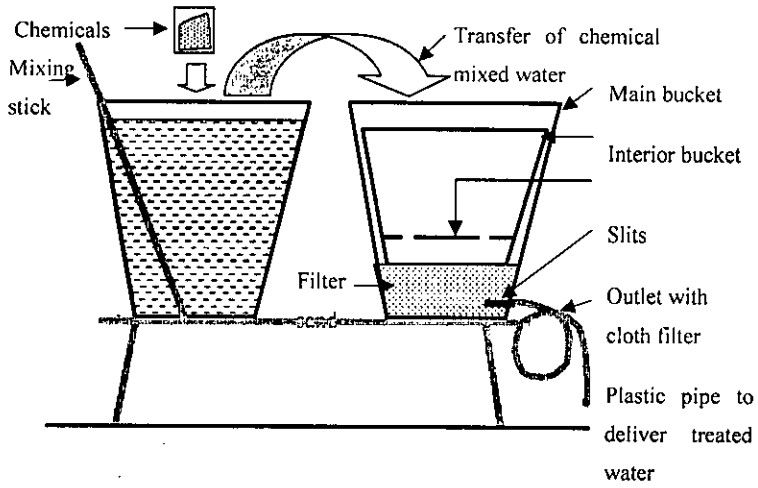

Figure 2.7 : Stevens Institute Technology (Source: Ahmed, 2001)

2.6.3 BCSIR Filter Unit

Bangladesh Council of Scientific and Industrial Research (BCSIR) has developed an arsenic removal system, which uses the process of coagulation/co-precipitation with an iron based chemical followed by sand filtration. The unit did not take part in a comprehensive evaluation process.

2.6.4 Fill and Draw Units

It is a community type treatment unit designed and installed under DPHE-Danida Arsenic Mitigation Pilot Project. It is 600 L capacity (effective) tank with slightly tapered bottom for collection and withdraw of settled sludge. The tank is fitted with a manually operated mixer with flat-blade impellers. The tank is filled with arsenic contaminated water and required quantity of oxidant and coagulant are added to the

water. The water is then mixed for 30 seconds by rotating the mixing device at the rate of 60 rpm and left overnight for sedimentation. The water takes some times to become completely still which helps flocculation. The floc formation is caused by the hydraulic gradient of the rotating water in the tank. The settled water is then drawn through a pipe fitted at a level, few inches above the bottom of the tank and passed through a sand bed and finally collected through a tap for drinking purpose as shown in Figure 2.8. The mixing and flocculation processes in this unit are better controlled to effect higher removal of arsenic. The experimental units installed by DPHE-Danida project are serving the clusters of families and educational institutions.

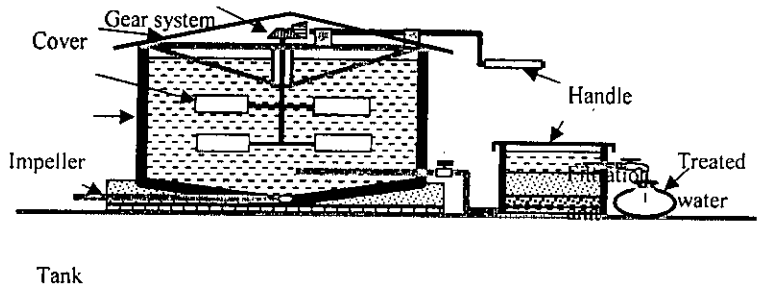

Figure 2.8 : DPHE-Danida Fill and Draw arsenic removal unit (Source: Ahmed, 2001)

2.6.5 Arsenic Removal Unit Attached to Tubewell

The principles of arsenic removal by alum coagulation, sedimentation and filtration have been employed in a compact unit for water treatment in the village level in West Bengal, India. The arsenic removal plant attached to hand tubewell as shown in Figure 2.9 has been found effective in removing 90 percent arsenic from tubewell water having initial arsenic concentration of 300μg/L. The treatment process involves addition of sodium hypochloride (Cl_2), and aluminum alum in diluted form, mixing, flocculation, sedimentation and up flow filtration in a compact unit.

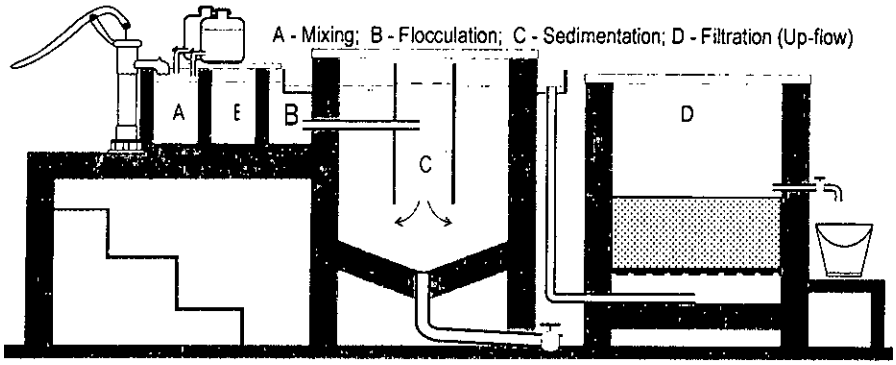

Figure 2.9 : Arsenic removal plants attached to tubewell (designed and constructed in India) (Source: Ahmed, 2001)

Some medium scale Fe-As removal plants of capacities 2000-3000 m^3/d have been constructed for water supplies in district towns based on the same principle. The treatment processes involved in these plants include aeration, sedimentation and rapid sand filtration with provision for addition of chemical, if required. These plants are working well except that treated water requirement for washing the filter beds is very high. Operations of small and medium size IRP-cum-ARPs in Bangladesh suggest that arsenic removal by co-precipitation and adsorption on natural iron flocs has good potential.

2.6.6 Chemical Packages

In Bangladesh, different types of chemical packages have been distributed in the form of tea bags, small packets and powder or tablet form for the removal of arsenic from drinking water. The principles involved in arsenic removal by these chemicals involve oxidation, sorption and co-precipitation. Application methodology and efficiency of any of these chemicals have not been fully optimized by long experimentation. Quality assurance and dose control in rural condition are extremely difficult. The residuals of added chemicals in water after treatment can do equal harm. The use of unknown chemicals and patented process without adequate information should be totally discouraged.

2.6.7 Activated Alumina

Activated alumina, Al_2O_3, having good sorptive surface is an effective medium for arsenic removal. When water passes through a packed column of activated alumina,

the impurities including arsenic present in water are adsorbed on the surfaces of activated alumina grains. Eventually the column becomes saturated, first at its upstream zone and later the saturated zone moves downstream towards the bottom end and finally the column get totally saturated.

Regeneration of saturated alumina is carried out by exposing the medium to 4% caustic soda, NaOH, either in batch or by flow through the column resulting in a high arsenic contaminated caustic waste water. The residual caustic soda is then washed out and the medium is neutralized with a 2% solution of sulfuric acid rinse. During the process about 5-10% alumina is lost and the capacity of the regenerated medium is reduced by 30-40%. The activated alumina needs replacement after 3-4 regeneration. Like coagulation process, pre-chlorination improves the column capacity dramatically. Some of the activated alumina based sorptive media used in Bangladesh include:

- BUET Activated Alumina
- Alcan Enhanced Activated Alumina
- ARU of Project Earth Industries Inc., USA
- Apyron Arsenic Treatment Unit

The BUET and Alcan activated alumina have been extensively tested in field condition in different areas of Bangladesh under rapid assessment and found very effective in arsenic removal (BAMWSP, DFID, WaterAid , 2001). The Arsenic Removal Units (ARUs) of Project Earth Industries Inc. (USA) used hybrid aluminas and composite metal oxides as adsorption media and were able to treat 200-500 Bed Volume (BV) of water containing 550 g/L of arsenic and 14 mg/L of iron (Ahmed et al., 2000). The Apyron Technologies Inc. (ATI) also uses inorganic granular metal oxide based media that can selectively remove As(III) and As(V) from water. The Aqua-Bind^{TM} arsenic media used by ATI consist of non-hazardous aluminum oxide and manganese oxide for cost-effective removal of arsenic. The proponents claimed that the units installed in India and Bangladesh consistently reduced arsenic to less than 10μg/L.

2.6.8 *Granular Ferric Hydroxide*

M/S Pal Trockner(P) Ltd, India and Sidko Limited, Bangladesh installed several Granular Ferric Hydroxide based arsenic removal units in India and Bangladesh. The Granular Ferric Hydroxide (AdsorpAs®) is arsenic selective adsorbent developed by Technical University, Berlin, Germany. The unit requires iron removal as pre-treatment to avoid clogging of filter bed. The proponents of the unit claims to have very high arsenic removal capacity and produces non-toxic spent granular ferric hydroxide.

2.6.9 *Read-F Arsenic Removal Unit*

Read-F is an adsorbent produced and promoted by Shin Nihon Salt Co. Ltd, Japan for arsenic removal in Bangladesh. Read-F displays high selectivity for arsenic ions under a broad range of conditions and effectively adsorbs both arsenite and arsenate without the need for pretreatment. The Read-F is Ethylene-vinyl alcohol copolymer (EVOH)-borne hydrous cerium oxide in which hydrous cerium oxide ($CeO_2 \cdot n\ H_2O$), is the adsorbent. The material contains no organic solvent or other volatile substance and is not classified as hazardous material. Laboratory test at BUET and field testing of the materials at 4 sites under the supervision of BAMWSP showed that the adsorbent is highly efficient in removing arsenic from groundwater (SNSCL, 2000).

2.6.10 *Iron Coated Sand*

BUET has constructed and tested iron coated sand based small scale unit for the removal of arsenic from groundwater. Iron coated sand has been prepared following a procedure similar to that adopted by Joshi and Choudhuri (1996). The iron content of the iron-coated sand was found to be 25 mg/g of sand. Raw water having 300 µg/L of arsenic when filtered through iron coated sand becomes essentially arsenic-free. It was found that 350 bed volumes could be treated satisfying the Bangladesh drinking water standard of 50 ppb. The saturated medium is regenerated by passing 0.2N sodium hydroxide through the column or soaking the sand in 0.2N sodium hydroxide followed by washing with distilled water. No significant change in bed volume (BV) in arsenic removal was found after 5 regeneration cycles. It was interesting to note that iron coated sand is equally effective in removing both As(III) and As(V). Iron

coated brick dust has also been developed in Bangladesh for arsenic removal from drinking water.

2.6.11 Indigenous Filters

There are several filters available in Bangladesh that use indigenous material as arsenic adsorbent. Red soil rich in oxidized iron, clay minerals, iron ore, iron scrap or fillings and processed cellulose materials are known to have capacity for arsenic adsorption. Some of the filters manufactured using these materials include:

- Sono 3-Kolshi Filter
- Granet Home-made Filter
- Chari Filter
- Adarsha Filter
- Shafi Filter
- Bijoypur Clay/Processed Cellulose filter

The Sono 3-Kolshi filter uses zero valent iron fillings and coarse sand in the top Kolshi, wood coke and fine sand in the middle Kolshi while the bottom Kolshi is the collector of the filtered water (Khan et al., 2000). Earlier Nikolaidis and Lackovic (1998) showed that 97 % arsenic can be removed by adsorption on a mixture of zero valent iron fillings and sand and recommended that arsenic species could have been removed through formation of co-precipitates, mixed precipitates and by adsorption onto the ferric hydroxide solids. The Sono 3-Kolshi unit has been found to be very effective in removing arsenic but the media habour growth of microorganism (BAMWSP, DFID and WaterAid, 2000). The one-time use unit becomes quickly clogged, if groundwater contains excessive iron.

The Garnet home-made filter contains relatively inert materials like brick chips and sand as filtering media. No chemical is added to the system. Air oxidation and adsorption on iron-rich brick chips and flocs of naturally present iron in groundwater could be the reason for arsenic removal from groundwater. The unit produced inadequate quantity of water and did not show reliable results in different areas of Bangladesh and under different operating conditions. The Chari filter also uses brick

chips and inert aggregates in different Charis as filter media. The effectiveness of this filter in arsenic removal is not known.

The Shafi and Adarsha filters use clay material as filter media in the form of candle. The Shafi filter was reported to have good arsenic removal capacity but suffered from clogging of filter media. The Adarsha filter participated in the rapid assessment program but failed to meet the technical criterion of reducing arsenic to acceptable level (BAMWSP, DFID and WaterAid, 2000). Bijoypur clay and treated cellulose were also found to adsorb arsenic from water (Khair, 2000).

2.6.12 Cartridge Filters

Filter units with cartridges filled with soptive media or ion-exchange resins are readily available in the market. These units remove arsenic like any other dissolved ions present in water. These units are not suitable for water having high impurities and iron in water. Presence of ions having higher affinity than arsenic can quickly saturate the media requiring regeneration or replacement. Two household filters were tested at BUET laboratories, These are:

- Chiyoda Arsenic Removal Unit, Japan
- Coolmart Water Purifier, Korea.

The Chiyoda Arsenic Removal Unit could treat 800 BV meeting the WHO guideline value of 10 µg/L and 1300 BV meeting the Bangladesh Standard of 50 µg/L when the feed water arsenic concentration was 300 µg/L. The Coolmart Water Purifier could treat only 20 L of water with a effluent arsenic content of 25 µg/L (Ahmed et al., 2000). The initial and operation costs of these units are high and beyond the reach of the rural people.

2.6.13 Tetrahedron ion exchange resin filter

Tetrahedron ion exchange resin filter tested under rapid assessment program in Bangladesh (BAMWSP, DFID and WaterAid, 2000) showed promising results in arsenic removal. The system needs pre-oxidation of arsenite by sodium hypochloride. The residual chlorine helps to minimize bacterial growth in the media. The saturated

resin requires regeneration by recirculating NaCl solution. The liquid wastes rich in salt and arsenic produced during regeneration require special treatment. Some other ion exchange resins were demonstrated in Bangladesh but sufficient field test results are not available on the performance of those resins.

2.6.14 MRT-1000 and Reid System Ltd.

Jago Corporation Limited promoted a household reverse osmosis water dispenser MRT-1000 manufactured by B & T Science Co. Limited, Taiwan. This system was tested at BUET and showed a arsenic (III) removal efficiency more than 80%. A wider spectrum reverse osmosis system named Reid System Limited was also promoted in Bangladesh. Experimental results showed that the system could effectively reduce arsenic content along with other impurities in water. The capital and operational costs of the reverse osmosis system would be relatively high.

2.6.15 Low-pressure Nanofiltration and Reverse Osmosis

Oh et al.(2000) applied reverse osmosis and nanofiltration membrane processes for the treatment of arsenic contaminated water applying low pressure by bicycle pump. A nanofiltration membrane process coupled with a bicycle pump could be operated under condition of low recovery and low pressure range from 0.2 to 0.7 MPa. Arsenite was found to have lower rejection than arsenate in ionized forms and hence water containing higher arsenite requires pre-oxidation for reduction of total arsenic acceptable level. In tubewell water in Bangladesh the average ratio of arsenite to total arsenic was found to be 0.25. However, the reverse osmosis process coupled with a bicycle pump system operating at 4 Mpa can be used for arsenic removal because of its high arsenite rejection. The study concluded that low-pressure nanofiltration with pre-oxidation or reverse osmosis with a bicycle pump device could be used for the treatment of arsenic contaminated groundwater in rural areas (Oh et al., 2000).

2.6.16 Comparison of Different Arsenic Removal Processes

All the technologies described have their relative merits and demerits and many technologies are being refined in order to make them more suitable for use in rural

condition. A comparison of different arsenic removal processes is shown in Table 2.4 (Ahmed, 2001).

Table 2.4 : A comparison of main arsenic removal technologies (Ahmed, 2001)

Technologies	Advantages	Disadvantages
Oxidation/ Precipitation		
▪ Air Oxidation	• Relatively simple, low-cost but slow process	• The processes remove only a part of arsenic
▪ Chemical oxidation	• Relatively simple and rapid process	
	• Oxidizes other impurities and kills microbes	
Coagulation Coprecipitation :		
• Alum Coagulation	• Relatively low capital cost,	• Produces toxic sludges
		• Low removal of As(III)
• Iron Coagulation	• Relatively simple operation	• Pre-oxidation may be required
	• Common Chemicals available	
Sorption Techniques		
• Actvated Alumina	• Relatively well known and commercially available	• Produces toxic solid waste
• Iron Coated Sand		• Replacement/regeneration required
	• Well defined technique	
• Ion Exchange Resin	• Plenty possibilities and scope of development	• High tech operation and maintenance
• Other Sorbents		• Relatively high cost
Membrane Techniques		
• Nanofiltration	• Well defined and high removal efficiency	• Very high capital and running cost
• Reverse osmosis	• No toxic solid wastes produced	• High tech operation and maintenance
• Electrodialysis	• Capable of removal of other contaminants	• Toxic wastewater produced

2.7 Summary

Arsenic removal technologies have to compete with other technologies in which cost appears to be a major determinant in the selection of a treatment option by the users. The rural people habituated in drinking tubewell water may find arsenic removal from tubewell water as a suitable option for water supply. In many arsenic affected areas, arsenic removal may be the only option in the absence of an alternative safe source of water supply.

Remarkable technological developments in arsenic removal from groundwater based on conventional arsenic removal processes have taken place during last 4-5 years. It appears that for effective removal of arsenic from groundwater coagulation co-precipitation process is a very suitable method for use in rural Bangladesh with respect to cost, availability of chemicals and operation and maintenance. Among the two most commonly and widely used coagulation co-precipitation processes, iron coagulation appears to be more suitable than alum due to less dependency on pH of water for effective removal ability. Besides in case of alum coagulation residual aluminum in the effluent water poses health risk while presence of naturally occurring iron in the tubewell water aids the coagulation process for iron coagulation.

CHAPTER 3: ARSENIC REMOVAL PROCESS DEVELOPMENT

3.1 Introduction

Various technologies have been used for removing arsenic from water. As discussed the Chapter 2, the most commonly used technologies include co-precipitation with alum or iron; adsorptive filtration (e.g., using activated alumina); ion exchange; and membrane processes such as reverse osmosis. Coagulation with ferric chloride and alum has been found to be very effective in removing arsenic from water both at bench scale and pilot scale tests (e.g., Hering et al., 1997; Hering et al., 1996; Scott et al., 1995; McNeill and Edwards, 1995; Cheng et al., 1994). In coagulation with ferric chloride, freshly precipitated amorphous ferric hydroxide, $Fe(OH)_3(am)$ is formed upon addition of the coagulant. Arsenic removal is primarily achieved by adsorption onto the surface of ferric hydroxide flocs and subsequent co-precipitation. In case of alum, removal is achieved by adsorption onto aluminum hydroxide flocs and subsequent co-precipitation. In general, ferric chloride has been found to be more effective in removing arsenic than alum on a weight basis and As(V) has been found to be more effectively removed than As(III).

In Bangladesh, arsenic removal based on coagulation-flocculation-precipitation, using alum as a coagulant has already been used in household arsenic treatment plants (sees section 2.5.1 and section 2.6). DPHE-Danida (1999) developed a simple "two bucket" system based on pre-oxidation of As(III) to As(V) using potassium permanganate and subsequent co-precipitation with alum (aluminum sulfate). Initial field testing of this system at nineteen households yielded positive results (DPHE-Danida, 1999). Arsenic removal by adsorption and co-precipitation onto coagulated flocs of ferric hydroxide could be a very effective technique for Bangladesh, particularly in view of the presence of elevated levels of iron in many regions of the country. In many affected areas, arsenic has been found to be associated with high iron concentrations (Hossain and Ali, 1997). This naturally occurring iron would increase the efficiency of any arsenic removal system based on iron coagulation. Ferric chloride based arsenic removal unit has been suggested as a promising technique by different researchers

(Ali and Chowdhury, 2000; Ahmed et al., 1999; Ali et al., 1998; Hering et al., 1997; Hering et al., 1996). Ali et al. (1998) and Hossain and Ali (1997) found very effective removal of arsenic from water by coagulation with ferric chloride; while Ahmed (1998) found good removal with alum. Recently Khoe and Emett (1999) developed an arsenic removal system based on co-precipitation with ferric sulfate. For higher arsenic concentration exceeding 0.5 mg/L, this system also involved pre-oxidation of As(III) to As(V) in a solar tray. The system is now being tested in the field. EAWAG/SANDEC (1999) also proposed a system "SORAS (Solar Oxidation and Removal of Arsenic)" for arsenic removal which is based on solar oxidation of As(III) to As(V) and co-precipitation with iron (either naturally present or added). This system is at a development stage.

In this study, effectiveness of both alum and ferric-chloride coagulation in removing arsenic from groundwater has been evaluated, in an effort to identify the more efficient coagulant for use in a household arsenic removal unit. This Chapter describes the results of batch experiments conducted to evaluate the effectiveness of alum and ferric chloride in removing both arsenite and arsenate from natural groundwater under a wide range of initial arsenic concentrations and coagulant doses. Effects of a number of water quality parameters on arsenic removal efficiency have been presented here. In addition, experiments conducted to determine the effect of mixing energy has also been described in this Chapter. Results of the experiments conducted to evaluate the effect of potassium permanganate on arsenic removal have been presented in this Chapter.

3.2 Materials and Methods

3.2.1 Effect of mixing energy on arsenic removal in the coagulation process

Arsenic removal by the coagulation-adsorption co-precipitation process involves mixing of the coagulant with the water from which arsenic is to be removed. The mixing/ agitation is required for the formation of coagulated flocs (e.g., ferric hydroxide or aluminum hydroxide flocs) and to ensure adequate contact between the flocs and the dissolved arsenic species. However, excessive agitation or mixing will break down large flocs resulting in poor settling (discrete settling) and poor arsenic

removal. Therefore, optimization of the agitating energy/process is necessary to ensure formation of larger flocs leading to better settling.

In this study, wooden stick, instead of a mechanical device, was used for mixing in order to mimic field condition in rural Bangladesh. The effect of mixing on floc formation was evaluated by varying the duration of slow mixing (i.e., by varying the number of turns of the stick used for mixing).

At first, experiments were done in 1-liter glass beakers, each containing 500-ml natural (arsenic-free) groundwater (without any pH adjustments). Ferrous sulfate and ferric chloride, available in local market, were used as coagulant. After addition of a particular dose of coagulant, stirring, at a rate of 1 turn/sec, was done manually with a glass rod. Mixing was varied by varying the number of turns from 0 to 120 (0, 5, 10, 15, 30, 45, 60, 75, 90, and 120 number of turns) at different beakers and formation of flocs were observed visually.

Then, similar experiments were conducted in 26-buckets, each containing 20-liters of natural (arsenic-free) groundwater (without any pH adjustments). As before both ferrous sulfate and ferric chloride, available in local market, were used as coagulant and a wooden stick was as a stirrer. After addition of a particular dose of coagulant, stirring was done manually with the wooden stick and the number of turns was varied as before. The water was then allowed to settle for 1 hour and 2 hours before sampling at two different sets of tests. Water samples were collected from the buckets with a pipette from a depth approximately 10-cm from the bottom. The water samples were then tested for residual iron, and percent iron removed (in the form of ferric hydroxide flocs, settled at the bottom of the bucket) was calculated in each case by subtracting the residual iron from the initial iron added as coagulant. Lower residual iron (i.e., higher removal) would indicate higher settling of iron in the form of flocs.

3.2.2 Removal system based on Alum and Ferric Chloride coagulation

Alum and ferric chloride available in the local market was used in this study. All coagulation experiments were carried out in 25-L plastic buckets with natural groundwater (without any pH adjustments) spiked with arsenite and arsenate at three

different initial concentrations. The groundwater used in the coagulation experiments was collected from the deep tubewell used for supplying water at BUET. This water was tested for detailed characterization. Removal of both arsenite and arsenate present at different initial concentrations were evaluated for different doses of alum and ferric chloride. After addition of a particular dose of a coagulant, the water in the bucket was mixed with a wooden stick. The mixing to be applied was fixed from the results of the experiments described earlier. The contents in the buckets were mixed, first vigorously for about 10 seconds (10 to 20 turns) and then slowly (approximately one turn of the wooden stick per second) for about 90 seconds (about 90 turns). Wooden stick, instead of a mechanical device, was used in order to mimic field condition in rural Bangladesh. After mixing, the flocs were allowed to settle for periods ranging from 30 minutes to 24 hours. Water samples were then collected with a pipette from a depth approximately 10 cm from the bottom of the bucket. The water samples were then tested for total arsenic. In addition a number of other parameters e.g., iron (for iron coagulation experiments), aluminum (for alum coagulation experiments) were also tested.

Similar experiments were also carried out to evaluate the effect of pre-oxidation (by different doses of potassium permanganate) on arsenite removal by alum and ferric chloride.

3.2.3 Selection and Dose of Oxidizing Agent

Potassium permanganate and bleaching powder are the two common oxidizing agents available in the local market. However, potassium permanganate is more stable than bleaching powder, and therefore was selected as the oxidizing agent in this study for the conversion of arsenite to arsenate. The dose of potassium permanganate required for oxidation of a particular concentration of arsenite can be determined from the following stoichiometry:

$$2KMnO_4 \rightarrow 2MnO_2 + K_2O + 3\,[O]$$

$$3H_3AsO_3 + 3\,[O] \rightarrow 3H_2As_2O_3^- + 3H^+$$

$$3H_3AsO_3 + 2KMnO_4 \rightarrow 3H_2As_2O_3^- + 2MnO_2 + K_2O + 3H^+$$

According to the above equation, for an arsenite concentration of 1000 ppb, required dose of potassium permanganate ($KMnO_4$) is 1.404 mg/L. However, for experiments conducted in this study, a factor of safety of 2.0 is considered. Thus actual dose of potassium permanganate used was twice that calculated from stoichiometric considerations.

3.2.4 Removal of residual Color produced by the Oxidizing Agent

Application of potassium permanganate as an oxidizing agent resulted in the development of a residual pink color in the treated water, which would be objectionable to the users.

It was observed that residual color decreases with time i.e. settling of flocs. To understand the effect of coagulant and settling time on residual color of the effluent, tests were conducted in 1-liter beakers. In this experiment residual color of the effluents were measured at different time intervals (at the end of 0 minute, 30 minutes, 60 minutes and 90 minutes) for different doses of the coagulant as well as permanganate. Alum dose of 300 mg/L and ferric chloride dose of 100 mg/L is used with different permanganate doses (0.707 mg/L, 1.404 mg/L and 2.81 mg/L) for tests of this experiment.

Color problems in typical water supply systems are solved using filters. To test color removal efficiency by sand as well as clogging of filter sand by flocs, colored effluent water from coagulation tests were passed through sand columns. Sand columns of depth 10 cm, 20 cm and 40 cm were prepared in glass burettes. Sylhet sand as well as sieved fine sand (passing #30 and retaining on #40 sieve) were used to prepare these sand columns. Tests were run with 20-liter water spiked with arsenite and high dose of iron (30 mg/L as Fe^{+++}). Permanganate concentration of 1.41 mg/L was used for 300 ppb and 500 ppb arsenite while a permanganate dose of 2.81 mg/L was used for 1000 ppb arsenite. Another run was performed with 300 mg/L of alum as coagulant with 1000 ppb of arsenite and 2.81 mg/L of permanganate. During each run flow rate and color of the effluent were measured. In most cases the run continued for one working day. Samples were collected after each liter of effluent flow. One of the

several runs continued upto 5 working days until the flow rate dropped below 5 ml/min.

3.2.5 *Effect of other water quality parameters on arsenic removal by ferric chloride coagulation*

Elevated phosphate and silicate concentrations in Bangladesh groundwater may dramatically decrease the effectiveness of arsenic removal by the co-precipitation treatment. (Meng et al., 2001). Test results of Meng et al. (2001) shown that presence of silicate alone decrease the removal of arsenic moderately. Phosphate present in the water without silicate decreases the removal of arsenic drastically. Presence of both phosphate and silicate decreases the efficiency further. They concluded that, those effects are attributed to the competition of the anions with As(V) for ferric hydroxide sorption sites. As(V), silicate and phosphate are adsorbed on ferric hydroxide through the formation of surface complexes with the surface hydroxyl groups. But the results indicated that, the affinity of silicate for ferric hydroxide was much weaker than As(V) and phosphate.

Tests were performed in 26-liter buckets with 20-liter water per batch, in this pretext. In this test $FeCl_3.6H_2O$ dose for coagulation was 20 mg/L as Fe^{3+}. $KMnO_4$ is used as oxidizer at a concentration of 1.404 mg/L. Phosphate was spiked into water in three different concentrations (2 mg/L, 4 mg/L and 6 mg/L). One batch was run as blank and no phosphate spiking had done in this batch. Reagent quality $NaH_2PO_4.2H_2O$ was used to get the phosphate. Amount of salt required for this operation was calculated from its atomic formula. For example 20 L of water required 65.684 mg of $NaH_2PO_4.2H_2O$. No tests to test the effect of silica had been performed, but presence of silica with some other impurities were measured during field testing of the unit, which has been described in chapter 4.

3.2.6 *Measurement of different parameters in the laboratory*

No speciation was made to measure As(III) and As(V) separately. In all cases total arsenic was measured. Most of the measurements were by a Graphite Furnace Atomic Absorption Spectrophotometer (Shimadzu, AA-6800). Other tests, at the initial stage

of the research work, are done either by SDDC or by HgBr method. In all cases the sample is acidified with conc. HCl (0.5 ml in 500 ml) to lower the pH well below 4.6. Iron is measured as total iron. Nessler tube method is used to measure total iron content of the samples. Color is measured with a Spectrophotometer (HACH DR/4000U Spectrophotometer). Phosphate and Manganese samples are also measured with Spectrophotometer (HACH DR/4000U Spectrophotometer). Aluminum is measured with another atomic absorption/ flame emission spectrophotometer (Shimadzu, AA-680). Fecal coliforms are measured following standard membrane filtration processes [by filtering the sample through filter paper (Millipore, 0.45μm) and then incubating in 45 degree Celsius temperature for 24 hours].

All the laboratory experiments were carried out using BUET pump house water. This water is pumped from a depth of about 392 ft. below the ground level. The water is primarily collected directly form pump house located in the south-east corner of EME (Electrical and Mechanical Engineering) building. After the preliminary experiments with mixing energy and coagulant and oxidizing agent selection, same water has been collected from the supply line at the laboratory.

3.3 Results and discussions

The laboratory experiments required a large quantity of water for coagulation experiments. It was not feasible to collect fresh natural ground water, typically containing high iron content. Therefore, BUET pump house water is used for the experiments. During the experiments water is collected from water supply fixture (bibcock) of the laboratory. Test results of different parameters of that water are presented in table 3.1:

Table 3.1 : Detailed characterization of groundwater used in laboratory experiments

Parameter	Unit	Concentration	Parameter	Unit	Concentration
pH	-	6.0	Iron	mg/L	0.07
Color	Pt-Co.	15	Manganese	mg/L	0.010
Turbidity	NTU	0.90	Potassium	mg/L	25.4
Alkalinity as $CaCO_3$	mg/L	242.0	Sodium	mg/L	131.9
Carbon di-oxide	mg/L	203.0	Arsenic	µg/L	<1.0
Dissolved Oxygen	mg/L	2.97 at 26°C 2.03 at 26.1°C	Lead	mg/L	0.0214
Conductivity	µs/cm	1054	Cadmium	mg/L	0.0018
Chloride	mg/L	165.0	Zinc	mg/L	0.0372
Hardness as $CaCO_3$	mg/L	338.0	Copper	mg/L	0.0467
Sulfate	mg/L	35.1	Nickel	mg/L	0.0074
Nitrate	mg/L	0.4	Mercury	mg/L	Nil
Phosphate	mg/L	0.14	Chromium	mg/L	0.0049
Fluoride	mg/L	0.35	Silica	mg/L	32.0

3.3.1 Fixation of mixing energy

Results of the experiments conducted to select the optimum manual mixing energy for the unit is presented in Figure 3.1 and Figure 3.2.

Visual observations during these tests indicated that stirring less than 30 turns would not result in settling of considerable flocs at the end of 2 hours. This experiment shows that increased number of gentle turns (1 turn/second) increases settling rate. But more than 90 turns does not increase the iron removal by flocculation-settling significantly. Based on these experimental results, 90 gentle turns proceeded by vigorous mixing by 10~20 turns is selected as the manual mixing energy for the household ARU. The result of this experiment is presented in Table 3.2.

Table 3.2 : Effect of mixing energy on floc formation

Fe^{+++} dose = 7.5 mg/L			No. of Turns				
			30	45	60	90	120
Ferric Chloride	Residual Iron (mg/L)	After 60 min	1.75	1.50	1.35	0.60	
			2.00	1.10	1.25	1.25	0.75
		After 120 min	1.35	1.10	0.75	0.60	
	Iron Removed %	After 60 min	76.67	80.00	82.00	92.00	
			73.33	85.33	83.33	83.33	90.00
		After 120 min	82.00	85.33	90.00	92.00	
Ferric Sulfate	Residual Iron (mg/L)	After 60 min	4.25	3.00	4.00	1.25	
		After 120 min	2.50	1.50	2.00	1.00	
	Iron Removed %	After 60 min	43.33	60.00	46.67	83.33	
		After 120 min	66.67	80.00	73.33	86.67	

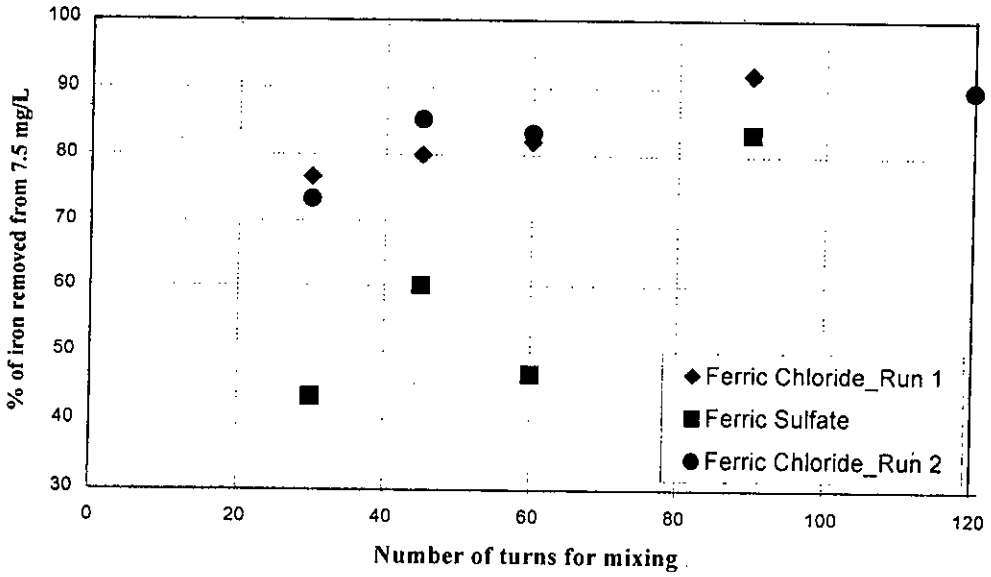

Figure 3.1 : Effect of mixing energy on iron floc formation and removal after 1 hr for different coagulants.

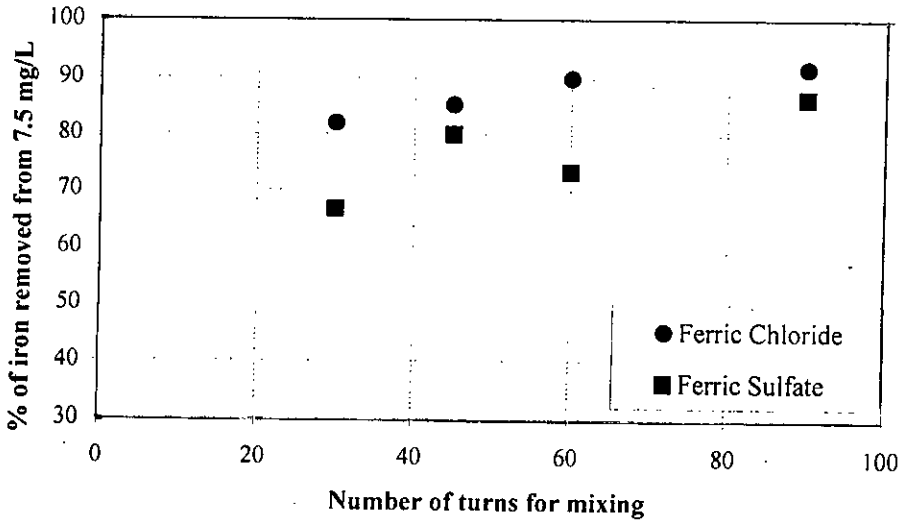

Figure 3.2 : Effect of mixing energy on iron floc formation and removal after 2 hrs for different coagulants.

3.3.2 Arsenic removal by Alum coagulation

Arsenic removal efficiency of alum is presented in figure 3.3 and figure 3.4. Tabulated results have also been presented in Table 3.3, Table 3.4 and Table 3.5.

Figure 3.3 shows removal of As(III) and As(V), present at different initial concentrations, by different doses of alum. It shows that for any particular arsenic concentration, removal efficiency increases with increasing alum dose. Removal efficiency also appears to increase with increasing settling time. As shown in Fig. 3.3(d), removal efficiency of As(III) is significantly lower than that of As(V). Even for As(V), very high doses of alum are required to bring the concentration of arsenic in the treated water below the Bangladesh standard of 50 ppb. In fact, this limit could not be achieved for a water sample with initial arsenate concentration of 1000 ppb treated with an alum dose as high as 300 mg/L. Figure 3.4 shows that removal of arsenite [As(III)], pre-oxidized with potassium permanganate, by different doses of alum was found to be similar to those achieved with As(V). In these experiments, a permanganate dose twice that required from stoichiometric consideration was used.

Raw water in actual field conditions can contain both As(V) and As(III). But alum could not bring down As(III) concentration below 50 ppb (Bangladesh standard for drinking water) from an initial concentration of 500 ppb or above within 90 minutes. Longer settling time requirement will be an operation problem if this coagulant is used for ARU.

A major concern in arsenic removal with alum coagulation is presence of high residual aluminum in the treated water. Figure 3.5 as well as Table 3.6 shows residual aluminum concentration in water treated with different doses of alum. It shows high residual aluminum concentration in the treated water ranging from around 1.0 mg/L to over 3 mg/L, against a drinking water standard of 0.20 mg/L (GoB, 1997). So the aluminum concentration of water treated by alum appears to exceed the drinking water standard by a wide margin.

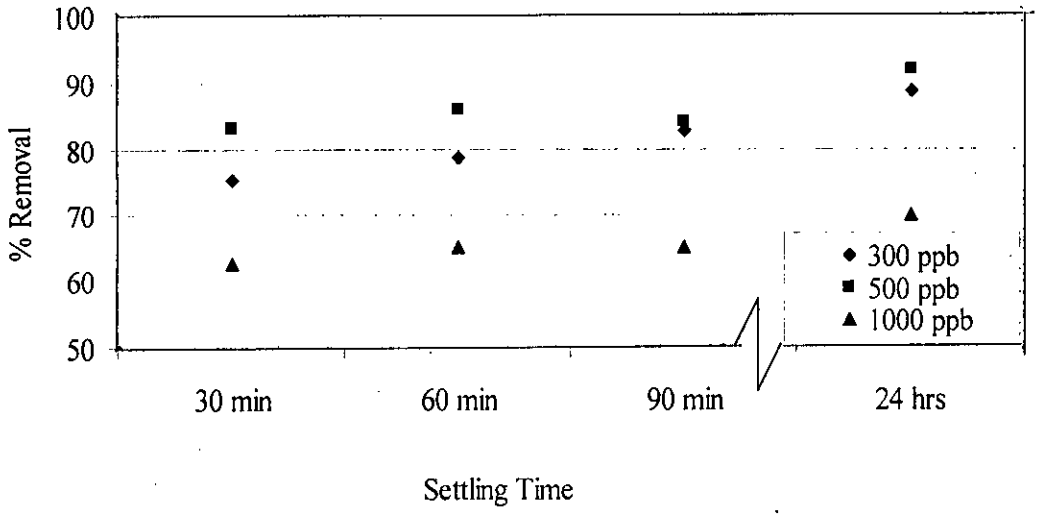

Figure 3.3 (a) : As (V) removal with 100 mg/L Alum without any oxidizing agent

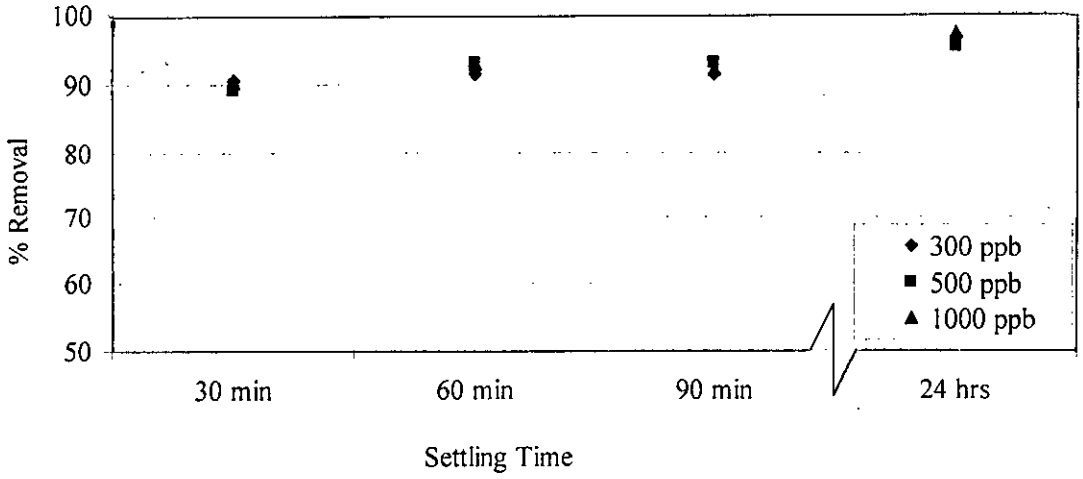

Figure 3.3 (b) : As (V) removal with 200 mg/L Alum without any oxidizing agent

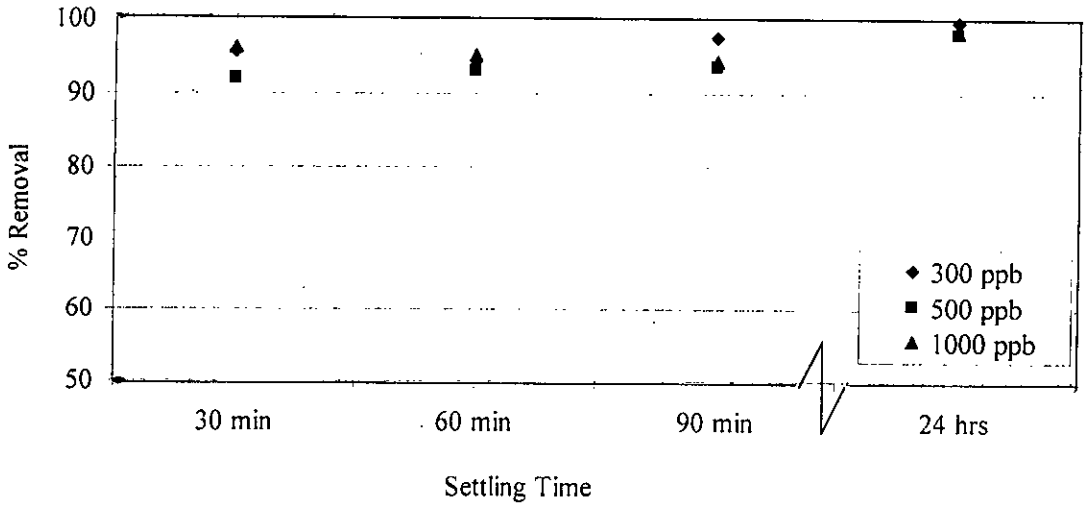

Figure 3.3 (c) : As (V) removal with 300 mg/L Alum without any oxidizing agent

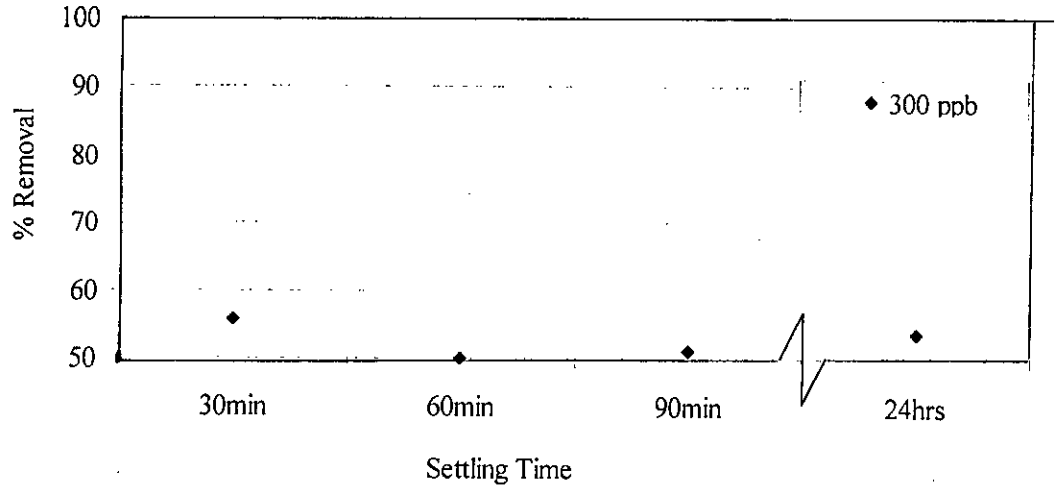

Figure 3.3 (d) : As (III) removal with 300 mg/L Alum without any oxidizing agent

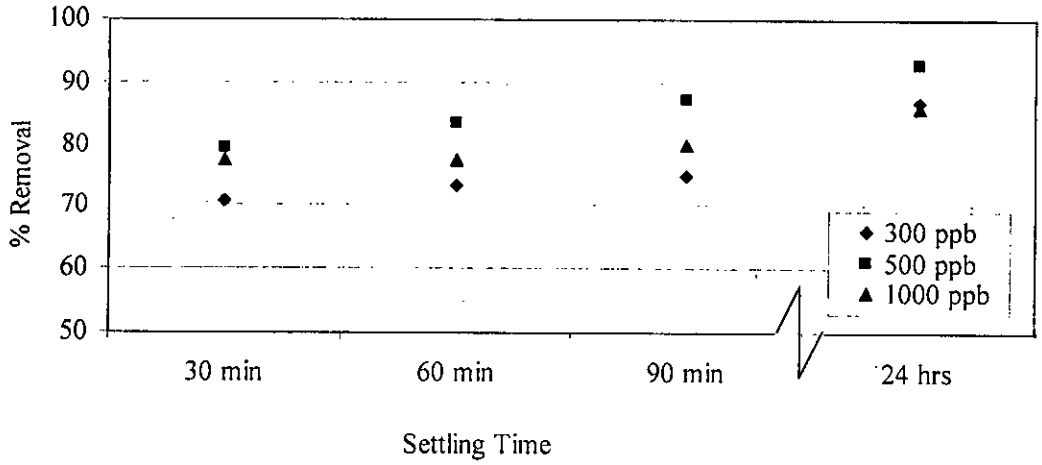

Figure 3.4 (a) : Pre-oxidized As (III) removal with 100 mg/L Alum

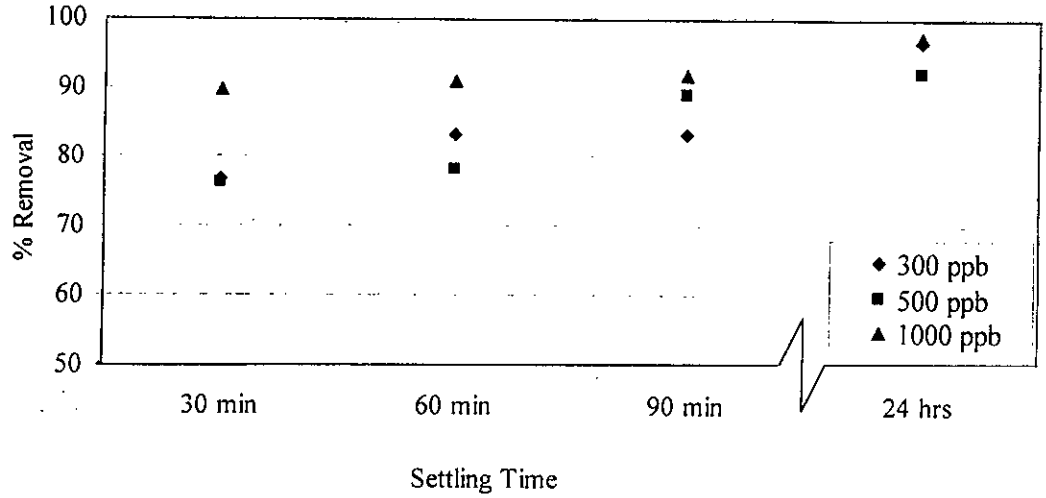

Figure 3.4 (b) : Pre-oxidized As (III) removal with 200 mg/L Alum

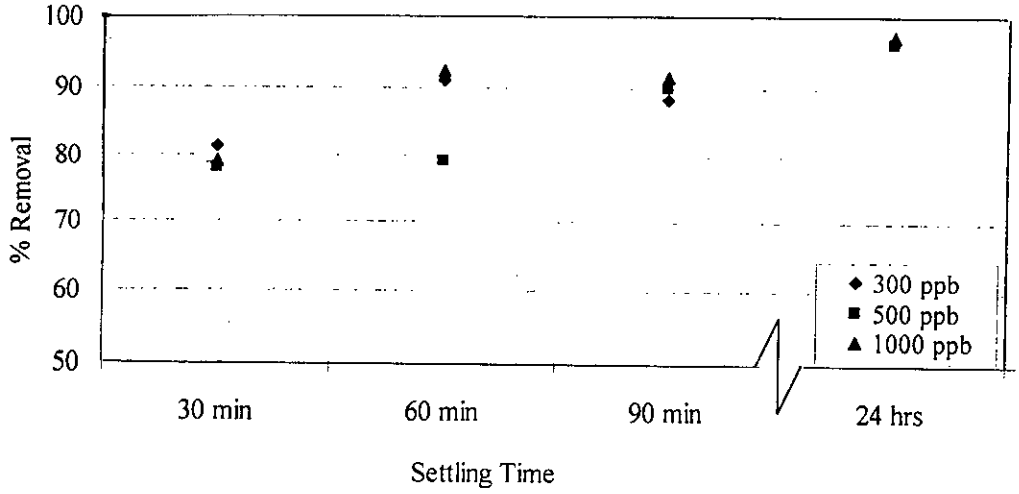

Figure 3.4 (c) : Pre-oxidized As (III) removal with 300 mg/L Alum

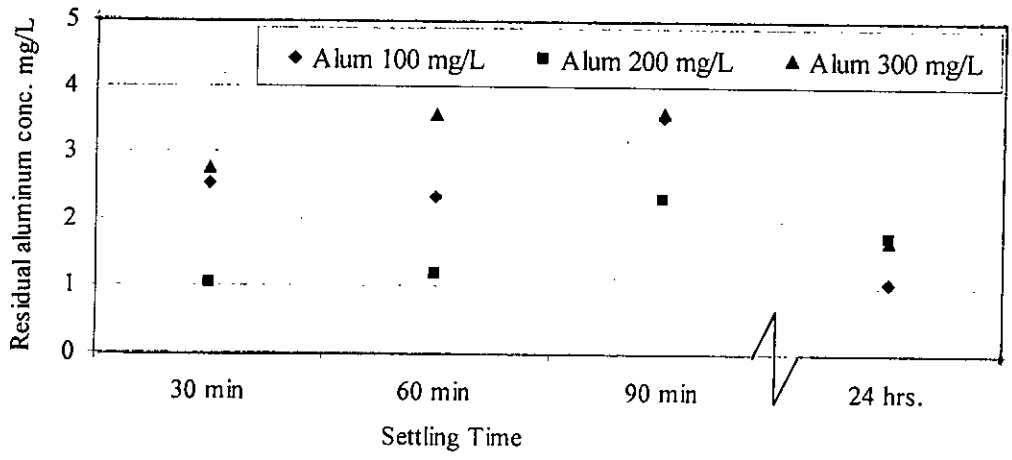

Figure 3.5 : Residual aluminum concentration in water treated with alum (initial As(V) concentration = 500 ppb)

Table 3.3 : **Removal of Arsenate [As(V)] by Alum Coagulation**

Arsenic (V) Conc. (ppb)	Settling Time	Alum dose (mg/L)					
		100		200		300	
		Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal
300	30 min	74	75.3	28	90.7	13	95.7
	60 min	64	78.7	26	91.3	17	94.3
	90 min	52	82.7	26	91.3	7	97.7
	24 hrs.	34	88.7	10	96.7	1	99.7
500	30 min	85	83.0	55	89.0	40	92.0
	60 min	70	86.0	35	93.0	35	93.0
	90 min	79	84.2	35	93.0	33	93.4
	24 hrs.	41	91.8	24	95.2	10	98.0
1000	30 min	375	62.5	98	90.2	38	96.2
	60 min	350	65.0	70	93.0	49	95.1
	90 min	350	65.0	79	92.1	57	94.3
	24 hrs.	300	70.0	28	97.2	15	98.5

Table 3.4 : **Arsenite [As(III)] removal with Alum Coagulation (without oxidizer)**

Arsenic (III) Conc. (ppb)	Settling Time	Alum dose (mg/L)	
		300	
		Residual As Conc. Ppb	% Removal
300	30 min	133	55.7
	60 min	150	50.0
	90 min	147	51.0
	24 hrs.	139	53.7

Table 3.5 : **Removal of Pre oxidized Arsenite [As(III)] by Alum Coagulation**

Arsenic (III) Conc. (ppb)	Settling Time	Alum dose (mg/L)					
		100		200		300	
		Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal
300	30 min	88	70.7	70	76.7	56	81.3
	60 min	80	73.3	50	83.3	27	91.0
	90 min	75	75.0	50	83.3	35	88.3
	24 hrs.	40	86.7	10	96.7	10	96.7
500	30 min	103	79.4	120	76.0	110	78.0
	60 min	83	83.4	110	78.0	104	79.2
	90 min	64	87.2	56	88.8	50	90.0
	24 hrs.	36	92.8	40	92.0	18	96.4
1000	30 min	225	77.5	103	89.7	210	79.0
	60 min	225	77.5	92	90.8	75	92.5
	90 min	200	80.0	81	91.9	85	91.5
	24 hrs.	140	86.0	27	97.3	25	97.5

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Table 3.6: **Residual Aluminum concentration for Alum coagulation unit**

Arsenic Conc. (ppb)	Settling Time	Alum Concentration (mg/L)					
		100		200		300	
		Residual Al Conc. ppm	Initial Al conc. ppm	Residual Al Conc. ppm	Initial Al conc. ppm	Residual Al Conc. ppm	Initial Al conc. ppm
500	30 min	2.5236	8.1	1.035	16.2	2.744	24.3
	60 min	2.3264		1.163		3.580	
	90 min	3.5519		2.309		3.595	
	24 hrs.	1.0482		1.745		1.668	

3.3.3 Arsenic removal by Ferric Chloride coagulation

Arsenic removal efficiency of ferric chloride is presented in figure 3.6 and figure 3.7. Tabulated results have also been presented in Table 3.7, Table 3.8 and Table 3.9.

Figure 3.6 shows removal of As(III) and As(V), present at three different initial concentrations, by different doses of ferric chloride. It shows very good removal of arsenate with ferric chloride. In general, removal efficiency was found to improve with increasing ferric chloride dose and longer settling times. An iron (added as ferric chloride) dose of 20 mg/L could bring down arsenate concentration below 30 ppb from an initial concentration of 1000 ppb. As shown in Figure 3.6, compared to As(V), As(III) removal was found to be significantly poor, confirming the results of previous studies and suggesting the need for pre-oxidation of As(III) to improve removal efficiency. A permanganate dose twice that required from stoichiometric consideration was used in these experiments.

Figure 3.7 shows removal of As(III), present at three different initial concentrations, by pre-oxidation of As(III) with potassium permanganate and then by different doses of ferric chloride. It shows very good removal of arsenite. In general, removal efficiency was found to improve with increasing ferric chloride dose and longer settling times. An iron (added as ferric chloride) dose of 20 mg/L could bring down arsenate concentration below 30 ppb from an initial concentration of 1000 ppb. Figure 3.6 shows that significant As(V) removal can be achieved by ferric chloride without pre-oxidation, therefore As(V) removal with pre-oxidation is not performed.

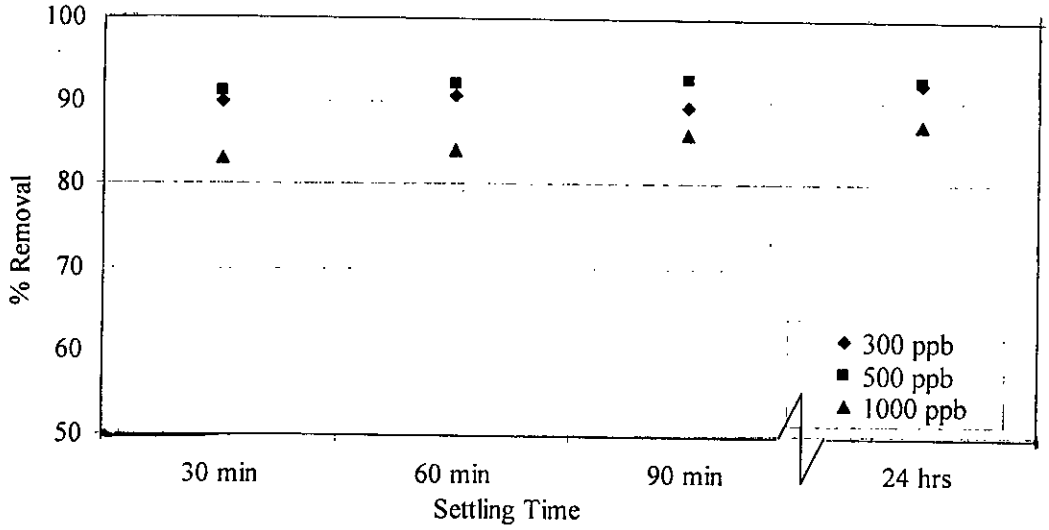

Figure 3.6 (a) : Arsenic(V) removal with 10 mg/l iron as ferric chloride.

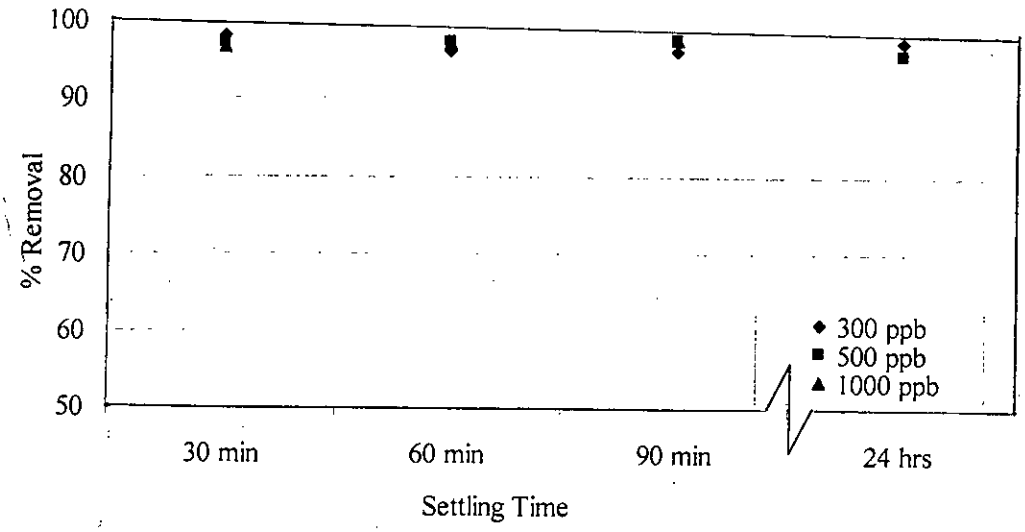

Figure 3.6 (b) : Arsenic(V) removal with 20 mg/l iron as ferric chloride.

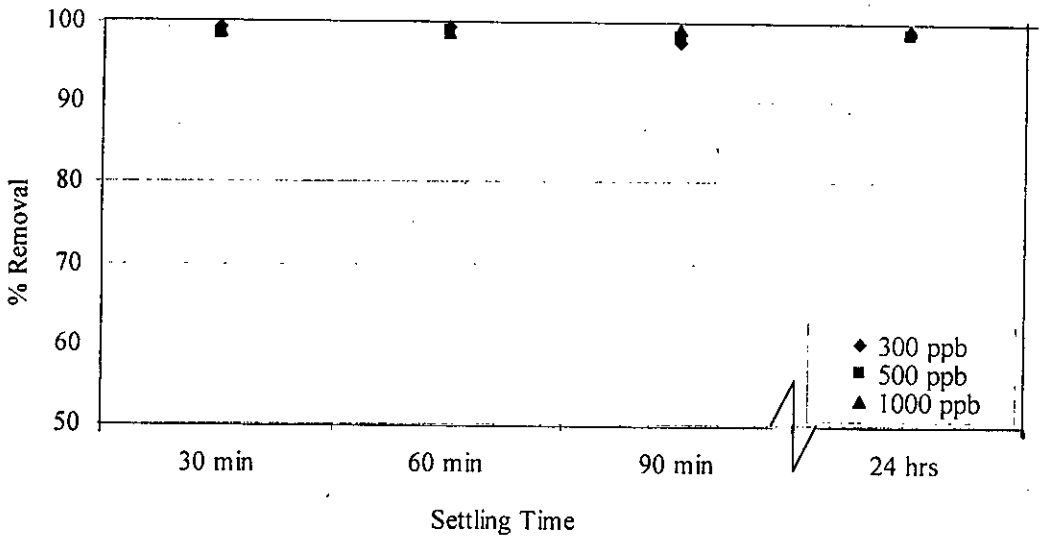

Figure 3.6 (c) : Arsenic(V) removal with 30 mg/l Iron as ferric chloride.

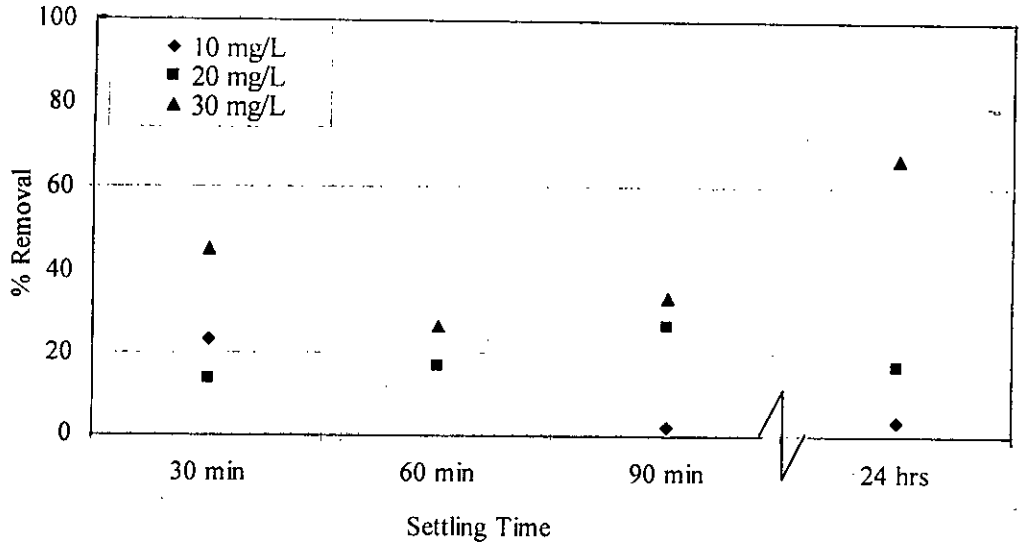

Figure 3.6 (d) : 300 ppb Arsenic(III) removal with different Iron doses (applied as ferric chloride.)

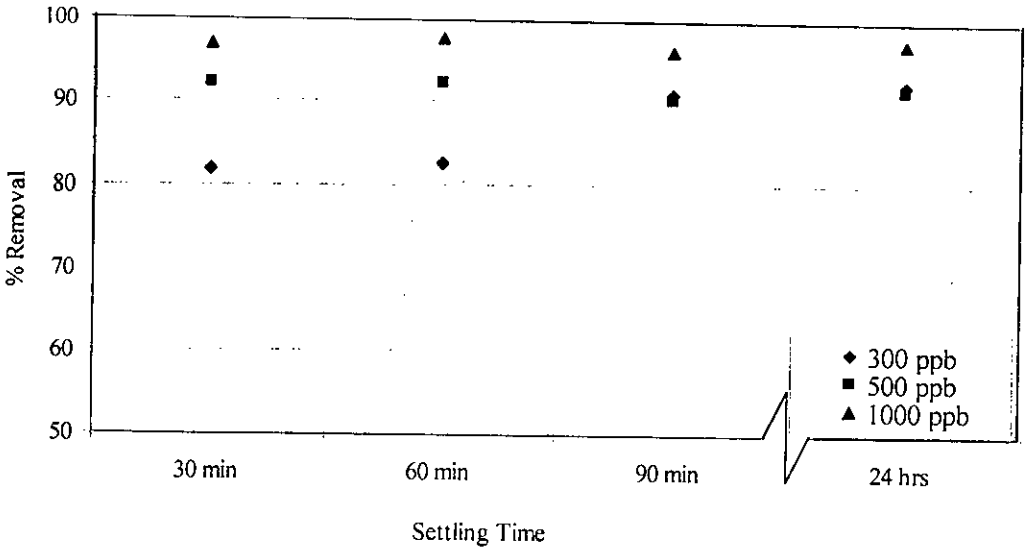

Figure 3.7 (a) : Pre-oxidized Arsenic(III) removal with 10 mg/l iron as ferric chloride.

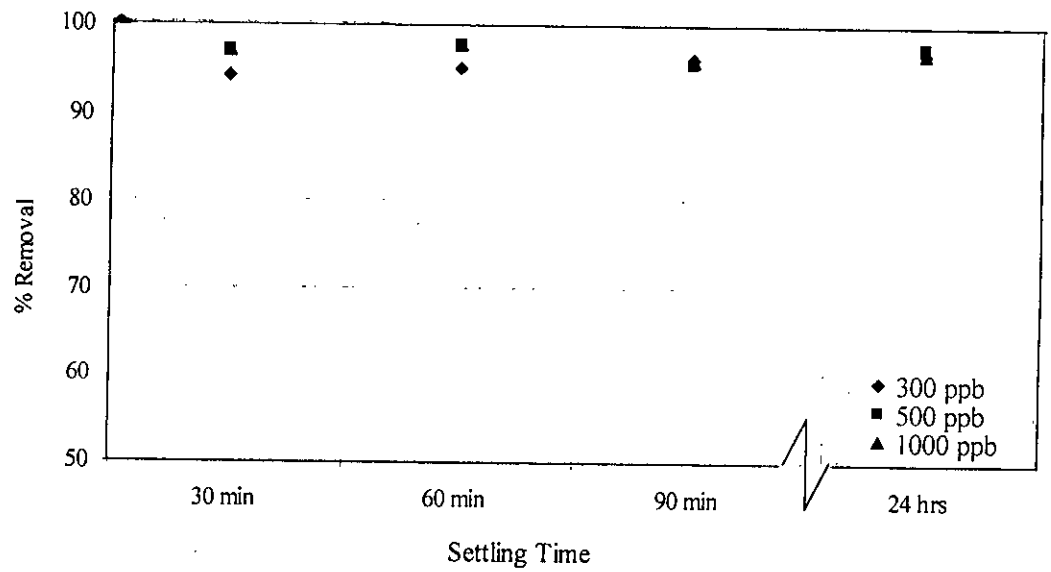

Figure 3.7 (b) : Pre-oxidized Arsenic(III) removal with 20 mg/l iron as ferric chloride.

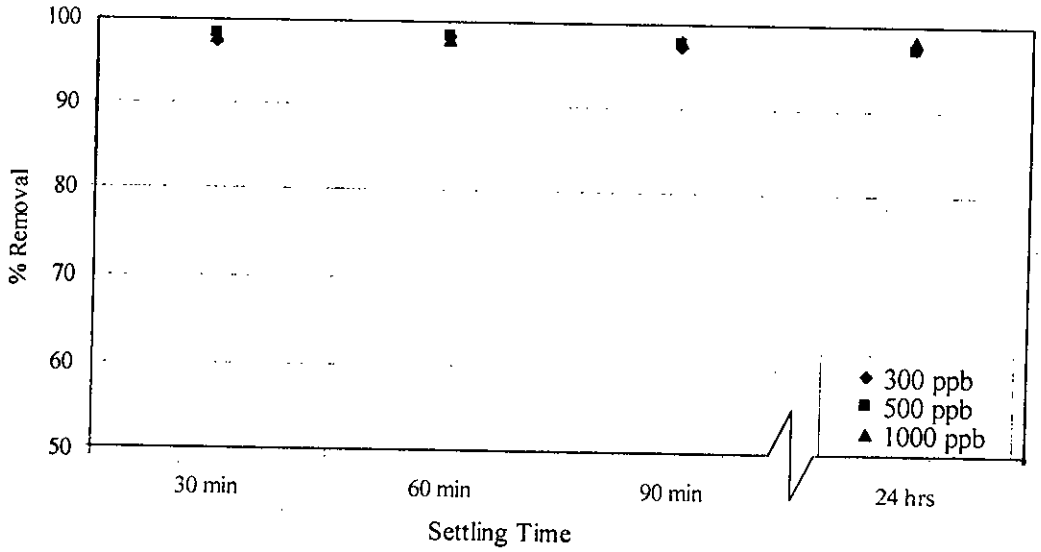

Figure 3.7 (c) : Pre-oxidized Arsenic(III) removal with 30 mg/l Iron as ferric chloride.

Table 3.7 : **Removal of Arsenate [As(V)] by Ferric Chloride Coagulation**

Arsenic Conc. (ppb)	Settling Time	Iron Concentration (added as Ferric Chloride) (mg/L)					
		10		20		30	
		Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal
300	30 min	30	90.0	6	98.0	2	99.3
	60 min	28	90.7	10	96.6	2	99.3
	90 min	32	89.3	12	96.0	8	97.3
	24 hrs.	24	92.0	8	97.3	4	98.7
500	30 min	45	91.0	15	97.0	8	97.4
	60 min	40	92.0	14	97.2	6	98.8
	90 min	38	92.4	12	97.6	10	98.0
	24 hrs.	29	94.2	22	95.6	8	98.4
1000	30 min	170	83.0	36	96.4	12	98.8
	60 min	160	84.0	30	97.0	14	98.6
	90 min	140	86.0	26	97.4	10	99.0
	24 hrs.	130	87.0	-	-	10	99.0

Table 3.8 : **Removal of Arsenite [As(III)] by Ferric Chloride Coagulation**

Arsenic Conc. (ppb)	Settling Time	Iron Concentration (added as Ferric Chloride) (mg/L)					
		10		20		30	
		Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal
300	30 min	230	23.3	260	13.3	165	45.0
	60 min	-	-	250	16.7	220	26.7
	90 min	295	1.7	220	26.7	200	33.3
	24 hrs.	290	3.3	250	16.7	100	66.7

Table 3.9 : **Removal of Pre oxidized (by 1.41 mg/L permanganate dose) Arsenite [As(III)] by Ferric Chloride Coagulation**

Arsenic Conc. (ppb)	Settling Time	Iron Concentration (added as Ferric Chloride) (mg/L)					
		10		20		30	
		Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal	Residual As Conc. ppb	% Removal
300	30 min	55	81.7	18	94.0	8	97.3
	60 min	53	82.3	15	95.0	5	98.3
	90 min	28	90.7	12	96.0	8	97.3
	24 hrs.	25	91.7	10	96.7	7	97.7
500	30 min	40	92.0	15	97.0	9	98.2
	60 min	40	92.0	12	97.6	9	98.2
	90 min	50	90.0	23	95.4	11	97.8
	24 hrs.	44	91.2	13	97.4	13	97.4
1000	30 min	120	88.0	31	96.9	23	97.7
	60 min	110	89.0	25	97.5	22	97.8
	90 min	135	86.5	42	95.8	20	98.0
	24 hrs.	75	92.5	33	96.7	15	98.5

3.3.4 Color removal

Initial test of filter column of 10 cm depth resulted lowering residual color from 68 Pt-Co unit to 7 Pt-Co unit for ferric chloride coagulant of 30 mg/L. Alum coagulation resulted in lowering color from 84 Pt-Co unit to 23 Pt-Co unit. This result failed to fulfill Bangladesh standard or 15 Pt-Co unit for color. Even though alum was not selected as the coagulant for the ARU and color removal results of the selected coagulant (ferric chloride) is within the range, to ensure the water quality against color 10 cm column was rejected for the ARU and no test with this column was done later.

Figure 3.8 shows color removal efficiency of 20 cm as well as 40 cm sand columns. Both of the sand columns (20 cm and 40 cm) showed very good color removal efficiency, bringing down color below acceptable Bangladesh standard of 15 Pt-Co unit until they were clogged.

Figure 3.9 shows reduction of flow rate with amount of water treated due to clogging by finer particles in the water. Filter columns made of fresh sand as well as columns made of used and washed column showed similar pattern of flow reduction.

Therefore providing a sand filter with minimum 20 cm deep sand filter would solve the color problem caused by permanganate; in addition, when flow rate would decrease, washing of filter media (sand) outside the column and then placing into the column again would solve the problem.

Table 3.10 : **Residual Color due to use of permanganate as an oxidizer**

	Color Readings (Pt-Co Unit)					
Coagulant	Ferric Chloride 100 mg/L			Alum 300 mg/L		
Permanganate Dose (S=1.404 mg/L)	S/2	S	2S	S/2	S	2S
Initial	26	62	75	31	61	68
After 30 minutes	4	6	20	40	38	49
After 60 minutes	6	11	20	32	30	39
After 90 minutes	8	8	23	31	29	40

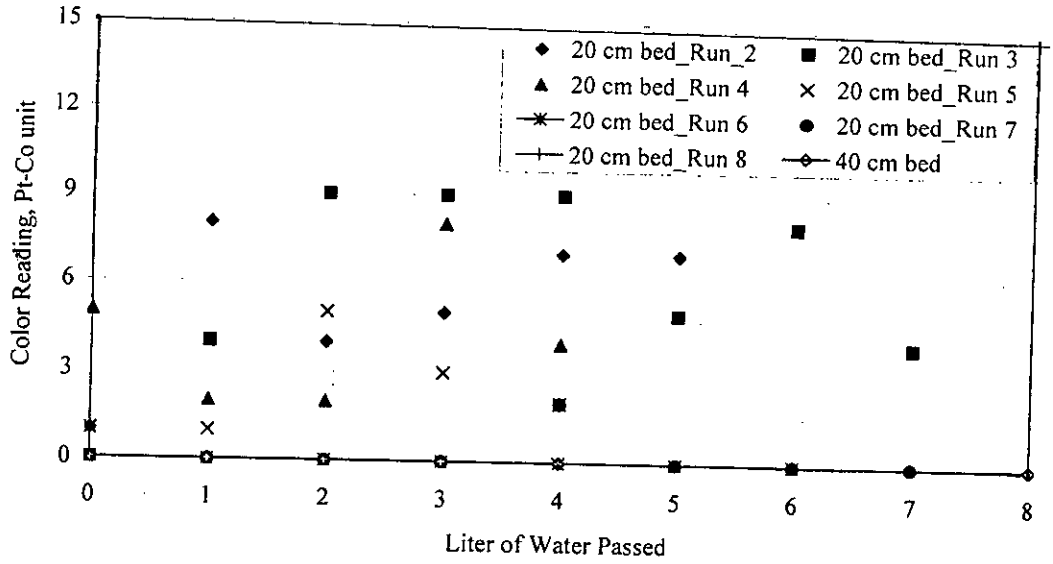

Figure 3.8 : Color removal by sand bed

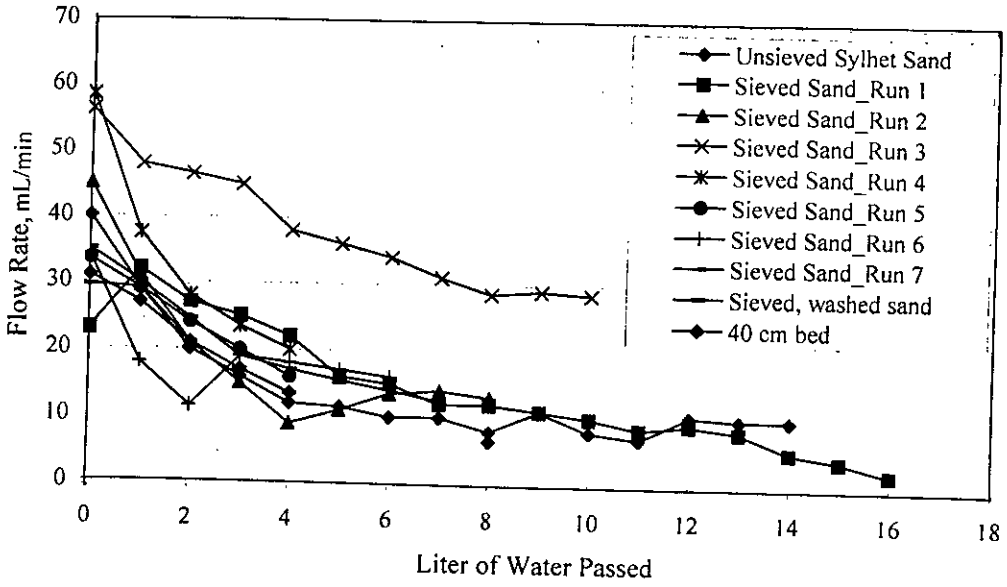

Figure 3.9 : Effect of clogging on flow rate through 20 cm sand column.

Table 3.11 : **Test of color removal efficiency of sand column**

Water Passed liters	Color Readings of Filtered effluent Pt-Co unit							
	Run - 2	Run - 3	Run - 4	Run - 5	Run - 6	Run - 7	Run - 8	Run - 9
Raw	42	10	10	12	9	10	15	11
0	1	0	5	1	0	0	0	0
1	8	4	2	1	0	0	0	0
2	4	9	2	5	0	0	0	0
3	5	9	8	3	0	0	0	0
4	7	9	4	2	0	2	0	0
5	7	5			0	0		
6		8			0	0		
7		4				0		
8								0

Table 3.12 : **Test of clogging of sand column by coagulant**

Water Passed Liters	Flow Rate mL/min									
	Run - 0	Run - 1	Run - 2	Run - 3	Run - 4	Run - 5	Run - 6	Run - 7	Run - 8	Run - 9
0	40	23.0	45.0	56.2	58.5	33.5	34.0	35.0	29.5	31.0
1	29	32.0	31.0	48.0	37.5	29.0	18.0	30.0	29.0	27.0
2	20	27.0	21.0	46.5	28.0	24.0	11.5	24.5	21.0	21.0
3	16	25.0	15.0	45.0	23.5	20.0	19.0	19.0	17.0	17.0
4	12	22.0	9.0	38.0	20.0	16.0	18.0	17.0	13.5	13.5
5	11.5	16.0	11.0	36.0			17.0	15.5		
6	10	15.0	13.5	34.0			16.0	14.0		
7	10	12.0	14.0	31.0				12.5		
8	8	12.0	13.0	28.5						6.5
9	11	11.0		29.0						
10	8	10.0		28.5						
11	7	8.5								
12	10.5	9.2								
13	10	8.2								
14	10	5.2								
15		4.0								
16		2.0								

3.3.5 *Effect of water quality parameters on performance*

Test results for effect of phosphate on arsenic removal by co-precipitation with ferric chloride is presented in figure 3.10.

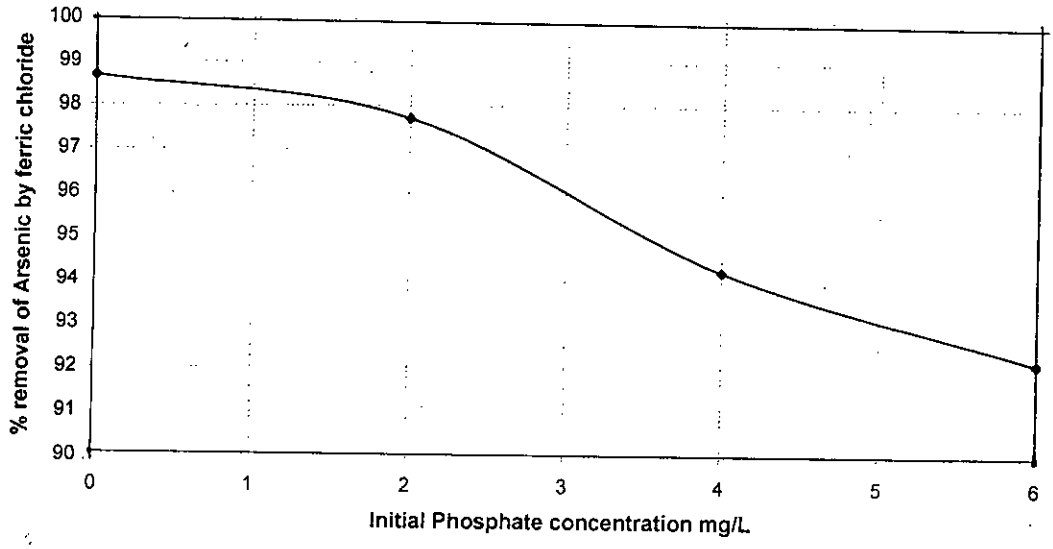

Figure 3.10 : Effect of phosphate on arsenic removal by co-precipitation with ferric chloride. .

Figure 3.10 shows that increase of phosphate concentration in the raw water decreases removal of arsenic by co-precipitation.

Table 3.13 : **Effect of Phosphate concentration in water on Arsenic removal by ferric chloride**

Initial Phosphate concentration (applied as $NaH_2PO_4.2H_2O$) mg/L	Final Phosphate concentration mg/L	Initial Arsenic(V) concentration µg/L	Final Arsenic(V) concentration µg/L	% Removal of Arsenic(V)
0	0.038	500	6.61	98.68
2	0.053	500	11.38	97.72
4	0.134	500	28.79	94.24
6	0.784	500	39.15	92.17

3.4 Summary

In this study efficiency of arsenic removal by coagulation with alum and ferric chloride has been evaluated. Coagulation-flocculation requires mixing of the coagulant with the water and the nature of mixing applied has a significant influence on formation of coagulated flocs. Experiments were conducted to determine the optimum mixing for achieving maximum floc formation. Experimental results showed that vigorous mixing for 10-15 seconds followed by 90 slow turns yielded best results. This method of mixing was adopted in all coagulation experiments conducted in this study.

Results of coagulation experiments suggest that ferric chloride is much more efficient than alum in removing arsenic from groundwater. As expected, removal of arsenate was much more efficient than arsenite. In this study potassium permanganate was used as an oxidizing agent for oxidation of arsenite to arsenate for effective removal of arsenic. Bleaching powder was not used because of its unstable nature. Experimental results suggest that a dose of potassium permanganate twice the stoichiometric requirement is sufficient for oxidation of arsenite to arsenate. However, use of potassium permanganate produced slight pink color in the treated water, which would be objectionable to the users. This color was removed using a sand filter. Laboratory test results suggest that a sand filter 20-cm deep was sufficient for removal of color. In this study, effect of phosphate on arsenic removal efficiency was evaluated in batch sorption experiments. Results show that presence of high level of phosphate can reduce the efficiency of arsenic removal by ferric chloride to some extent.

CHAPTER 4: DESIGN AND FIELD TESTING OF HOUSEHOLD ARSENIC REMOVAL UNIT

4.1 Introduction

From the results of batch experiments, described in Chapter 3, it was decided that a household arsenic removal unit (ARU) based on ferric chloride coagulation would be designed and tested in the field. It was also decided that potassium permanganate would be used as an oxidizing agent for oxidation of arsenite to arsenate for effective arsenic removal. This Chapter describes the detailed design of the household arsenic removal unit (ARU) that was developed on the basis of the results of batch experiments described earlier. This Chapter also describes the field testing of the ARU in the Adda village of Barura Thana of Comilla district. Design modifications, based on field test results, have also been described. Estimated cost of the ARU, including its operation and maintenance cost, has also been provided. This Chapter also provides a detailed assessment of the performance of the arsenic removal unit in the field, including user acceptance of the ARUs.

4.2 Design of Household ARU

4.2.1 Physical Design of the ARU

Two issues need to be considered in the design of the household arsenic removal unit. Firstly, it should efficiently remove arsenic and should also be safe with respect to other chemical and biochemical water quality parameters. Secondly, the design of the unit should be such that people, especially women in rural Bangladesh, can use and maintain it without any difficulty and that it can be replicated easily at the rural level. These issues were considered in the design of the arsenic removal units (ARU) based on ferric chloride coagulation.

It was decided that the physical design of the arsenic removal unit would be similar to that used in the DPHE-Danida two-bucket arsenic removal system. Thus, the ARU to be designed would consist of two buckets, one placed over the other. Untreated tubewell water would be placed in the upper bucket. Chemicals (coagulant and

oxidant) would be added to the groundwater in the top bucket, mixed and then allowed to flow to the lower bucket. In the lower bucket, it would flow through a sand filter and finally treated water would be collected from a tap connected to the bottom of the lower bucket. In a rural family of Bangladesh women are generally responsible for fetching and storing water for drinking and cooking. Average height of the village women in Bangladesh is below 5 feet (152 cm). Therefore, the arsenic removal unit to be designed should be of such a height that an average woman can easily pour water and chemicals in the upper bucket and mix its contents. Besides, the collection point (i.e., tap) in the lower bucket should be high enough to have space for a pitcher or jug for collection of treated water for drinking and cooking. In view of the relatively small size of average rural household, the ARU should not occupy much space and should have an aesthetically pleasing appearance.

According to Bangladesh Population Census 2001 (BBS, 2001), average size of households in rural Bangladesh is 4.8 persons per family. Therefore, 40 litres of water should be sufficient to meet drinking and cooking demand of an average family (Ahmed and Rahman, 2000) It was decided that two 35-liter plastic buckets with lids would be used in the ARUs. About 25 liters of water would be placed in the upper bucket and the design output (i.e., treated water) per run of the unit was taken to be 20 liters. Thus each unit would require two runs per day for an average sized family in rural Bangladesh. Larger buckets were selected to provide a dead volume at the bottom for sludge to settle as well as some freeboard at the top to prevent water spilling off the bucket during stirring. Each bucket used in the ARU is about 40-cm high, with a diameter of about 30-cm at the bottom and approximately 40 cm at the top.

Figure 4.1 shows a schematic representation of the designed arsenic removal unit (ARU). The upper bucket of ARU can be called as the coagulation-flocculation-settling unit. In this bucket a filling mark is provided to indicate 25 liters. A plastic tap (bibcock) placed 4 cm from the bottom provides approximately a 5-litre dead volume for accumulation of settled sludge. A flexible pipe of 1.0-cm diameter carries the effluent to the lower bucket through a small hole at its lid.

The lower bucket functions as a filtration unit, which is intended to remove any ferric hydroxide flocs (with adsorbed arsenic on it) that would come from the upper bucket and also to removal the color produced by potassium permanganate. Just below the lid of this bucket a piece of cloth is placed to aid in removing the unsettled flocs from the upper bucket. A 20-cm deep sand bed (FM=1.5) is provided in the lower bucket to filter out any flocs and color. Through the sand filter, the water flows into a piece of PVC strainer (slot 30) laid horizontally at the bottom of the lower bucket (see Figure 4.1). Both ends of the strainer are closed with appropriate PVC stoppers. A 1.0-cm diameter PVC pipe connects the strainer with a tap (bibcock), fixed 4 cm from the bottom of the bucket. Treated groundwater is collected from this tap.

Both the buckets are placed in an iron frame, made of 9 mm mild steel bars. The lower bucket sits 30 cm above the ground. This clearance is provided for the collection vessel. The top bucket is placed above the lower one. A 10-cm gap is provided between the buckets to facilitate easy operation/ functioning of the connecting pipe (Fig. 4.1).

4.2.2 Chemical Design of the ARU

Analysis of recent data on arsenic concentration in tubewell water all over Bangladesh (BGS, 2000) revealed that about 99.6% tubewells have arsenic concentration below 1000 ppb, 98.7% tubewells have arsenic concentration below 700 ppb, and 92.6% tubewells have arsenic concentration below 500 ppb. In the design of the ARU, 500 ppb is taken as the design concentration of arsenic in raw tubewell water. Results of batch experiments, presented in Chapter 3, showed that Fe^{3+} concentrations of 20 mg/L (added as ferric chloride) along with a potassium permanganate dose of 1.404 mg/l is sufficient to remove As(III) well below 50 ppb (the Bangladesh standard) from an initial concentration of 500 ppb. Thus, required dose of commercially available ferric chloride ($FeCl_3.6H_2O$) is 100-mg/L. Therefore, total chemical requirements are as follows:

Ferric Chloride (commercial grade) = *100 mg/L × 25L* = *2500 mg* = *2.5 gm*
Potassium Permanganate = *1.404 mg/L × 25L* = *35 mg*

All the chemicals were packed in airtight small polythene packets for use.

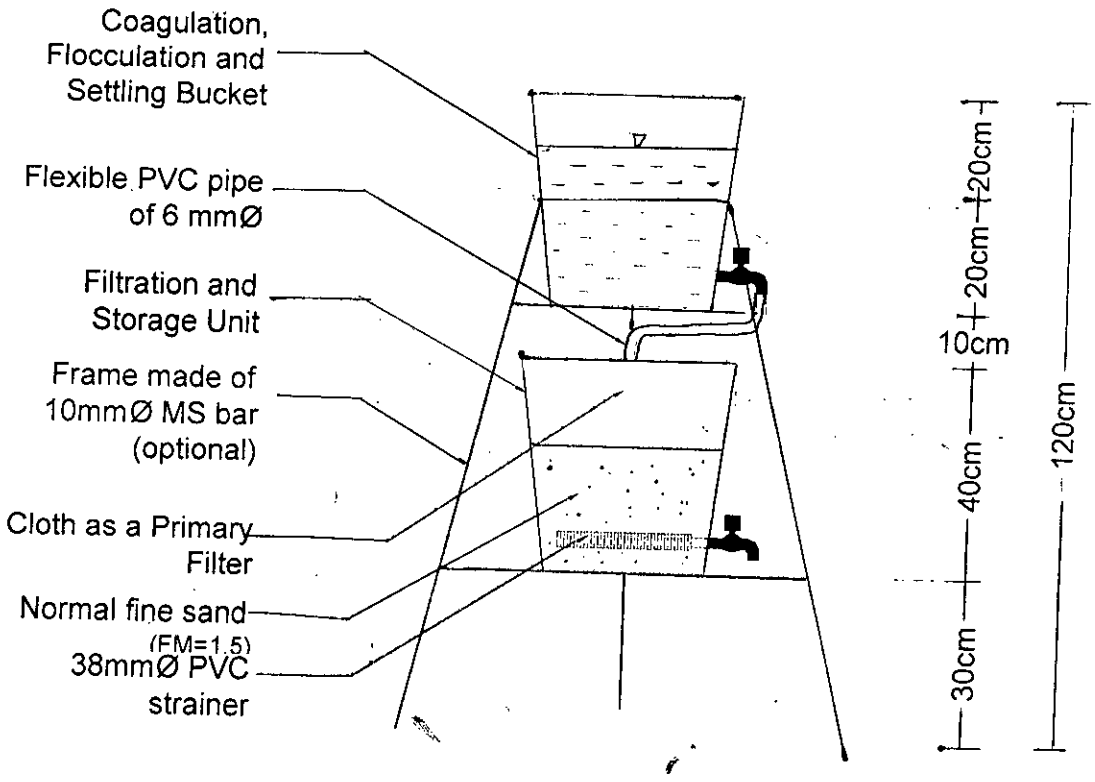

Figure 4.1 : Sectional elevation of the arsenic removal unit

4.2.3 Laboratory Testing of the ARU

Before testing in the field, the designed arsenic removal unit (ARU) based on ferric chloride coagulation was tested in the laboratory with natural (arsenic-free) groundwater spiked with 500 ppb of arsenite. Excellent arsenic removal was achieved in the laboratory testing, as all the four runs yield effluent arsenic concentration below 15 ppb averaging 7 ppb.

It should be noted that Ferric Chloride is a very hygroscopic material; therefore any leakage in the chemical packet would causes absorption of moisture by ferric chloride. This may cause part of the chemical to stick to the packet. But lab tests results suggest that it does not affect the performance of the arsenic removal unit significantly.

4.3 Field testing of Household ARU

A total of 15 arsenic removal units (ARUs) were installed in the Adda village of Barura thana of Comilla district for field testing. Adda village was selected, as most of the shallow tubewells were marked red by local NGO during their screening operations for arsenic. There were only a few public deep tubewell in that locality producing arsenic-free water. Besides, local people at Adda were very keen to have an arsenic mitigation option. For installation of the ARUs, user families were selected from different economic and social status. This was done to see if user acceptance as well as performance of the unit varies with socio-economic and educational status of the user.

The field-testing started on 21st July 2000 with installation of three such units in three different households in the village. The arsenic concentrations in the tubewells located at these households are 450 ppb, 640 ppb and 375 ppb. On 11th August 2000, two more arsenic removal units were installed at two other households of the same village. Ten more units, built by the villagers themselves, became operational on 25th September 2000.

All the ARUs were monitored for assessing the effectiveness of arsenic removal, while five ARUs were closely monitored for a wide range of parameters. Users were provided with the unit along with a one-page operation manual (see Appendix A). The operation of the ARUs basically consist of the following steps:

(i) Fill the top red bucket upto the mark with tubewell water.
(ii) Add all the medicine (chemicals) from one packet, supplied, to the water.
(iii) Stir the water with the wooden stirrer vigorously for 5~6 time to mix the chemical to water
(iv) Then gently stir the water with the wooden stirren for 40~45 times with gentle turns. (No. of turns is reduced from the laboratory findings because it was observed that users were reluctant to use 90 turns. Reduced turning did not decrease performance notably and was accepted to the users.)
(v) Cover the top bucket with lid and wait for an hour.
(vi) Open the two taps simultaneously to collect water.

(vii) Close both the taps when water collection ends.

(viii) Dispose the sludge to the drain or to cow dung pit.

(ix) Wash the cloth piece.

Users were given spot training on the operation and maintenance. They were briefed about the technique of stirring and the importance of proper mixing and waiting period before collecting treated water. They were advised to throw away the sludge collected at the bottom of the top bucket to cow dung pit. A notebook and a pen were also provided to them to record the water use. Sampling bottles were also provided to collect treated water sample (500ml) once a day for laboratory tests. The users were directed to take note of the number of buckets of water they treat each day against the date and the sampling bottle number so that the samples can be traced. After the first fifteen days of sampling the sampling frequency was changed to 2 days and after two months it was 7 days. A field supervisor (Mr. Sattar) was appointed to monitor the use of the units and recording of the information by the users. The field supervisor, who was also a user of an ARU, was responsible for collection of chemical packets (from BUET laboratory) and their distribution to all the users. The field supervisor was also responsible for carrying the water samples collected by the users to the BUET laboratory for testing. Besides, samples of both untreated and treated groundwater were collected during field visits to the site. All collected samples raw (i.e., untreated) and treated groundwater were tested for total arsenic. Samples collected from five specific households were also tested for pH, redox potential, ferrous iron and total iron, manganese, nitrate, silica, phosphate, and coliform.

Besides discussion with the users about the performance of the ARUs, a questionnaire survey also conducted to gather users' opinion about the ARUs.

4.3.1 Arsenic Removal efficiency

Treated water samples were tested for arsenic to verify the arsenic removal efficiency of the unit in field conditions. The samples were preserved in a low pH condition before testing by adding 0.5 ml of concentrated HCl to 500 ml of sample, which brought the pH to about 1.0. The dilution of the sample due to this acidification is insignificant and therefore ignored.

Very good arsenic removal efficiency was achieved in all the 15 arsenic removal units installed in the village. Analysis of arsenic concentration in the treated water samples from the 15 households have been found to be mostly below 20 ppb level, much below the Bangladesh standard of 50 ppb. The maximum average concentration in the treated water recorded is 37 ppb. 2% of the total samples exceeded Bangladesh standard value for arsenic concentration of 50 ppb. Figure 4.2 shows arsenic concentrations in the well water and average as well as maximum and minimum arsenic concentration in the treated water for the 12 out of 15 households. Detailed results of arsenic concentration in the treated water are provided in Appendix-C.

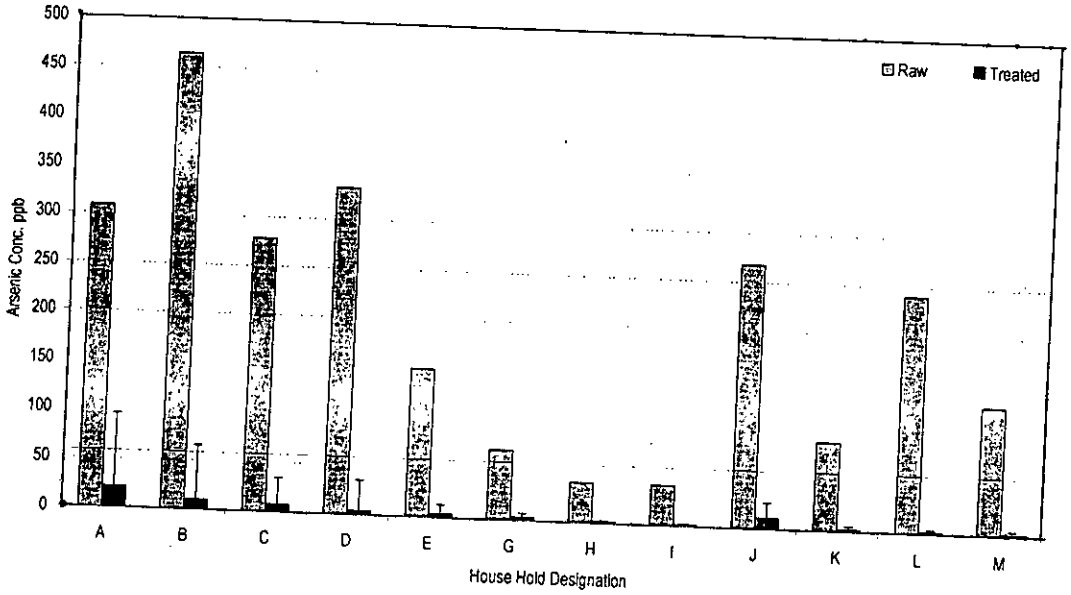

Figure 4.2 : Arsenic removal efficiency of the unit

4.3.1.1 Arsenic Removal Efficiency of Different Part of the ARU

Laboratory tests showed that careful operation of the system without the filter provides good arsenic removal efficiency. But sometimes the sludge floats in the surface instead of settling in the bottom. On the other hand sludge may flow with treated water during collection through the tap of coagulation bucket. To prevent sludge from coming with the treated water a cloth strainer and a sand filter is provided in the lower bucket. The sand filter also served to remove color developed due to the

addition of potassium permanganate (as oxidant). To optimize the design it was necessary to find the arsenic removal efficiency of the different parts of the unit.

To find efficiency of different parts, samples were collected from different stages of the treatment process within the unit for three different operating units in the field. Results of the analysis are shown in Table 4.1 and shown graphically in Fig. 4.3. Results shown in Table 4.1 show that removal of more than 70% of the total arsenic occurs in the top bucket during coagulation-flocculation-settling operation. Sand filter removed the rest. The piece of cloth, used as a primary filter, had almost no contribution in the arsenic removal process. Thus it appears that the piece of cloth can be removed from the system.

Table 4.1: **Test results for efficiency of different elements of the unit**

Unit Designation	Sampling Location	Run # 1 ppb	Run # 2 ppb	Average ppb	% Removal of initial concentration
Mr. Shamsur Rahman Installed on: 25-Sep-2000	Raw Water	132.61	132.61	132.61	0.00
	At upper tap	27.60	24.99	26.30	80.17
	Below the cloth	27.53	32.91	30.22	77.21
	Final Effluent	0.71	0.69	0.70	99.47
Mr. Abdus Sattar Installed on: 21-Jul-2000	Raw Water	133.55	133.55	133.55	0.00
	At upper tap	30.68	50.51	40.60	69.60
	Below the cloth	39.07	40.08	39.58	70.37
	Final Effluent	8.12	8.27	8.20	93.86
Dr. Delwar Hossain Installed on: 21-Jul-2000	Raw Water	129.67	129.67	129.67	0.00
	At upper tap	16.01	8.22	12.12	90.66
	Below the cloth	18.48	13.54	16.01	87.65
	Final Effluent	10.59	11.58	11.09	91.45

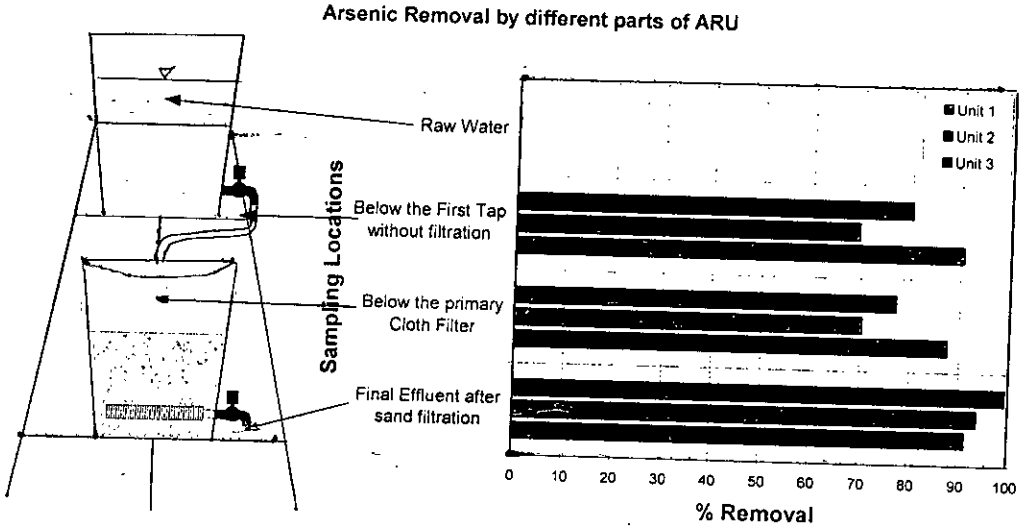

Figure 4.3 : Arsenic removal at different part of the unit

4.3.2 Other Water Quality Parameters

Besides arsenic, a number of water quality parameters (e.g., pH, Fe, Mn, Phosphate, Silica, Nitrate and redox potential) of the raw (tubewell) and treated water were measured at selected households in the field (using *Chemets* Field Kits) as well as in the laboratory. Table 4.2 shows results of field measurements of a number of parameters at selected households. These results show significant reduction of iron and phosphate concentrations in the treated water. There was also some reduction in

silica concentration. Compared to raw tubewell water, nitrate concentration of the treated water was raised and pH was slightly depressed. Redox potential data clearly show a complete shift from the reducing condition of the raw water to the oxidizing condition to the treated water. An average manganese concentration in the treated water was about 0.05 mg/L, far below the drinking water standard of 0.10 mg/L. Only one sample of treated water with Manganese concentration of 0.11 mg/L marginally exceeded the drinking water standard. It should be noted manganese concentration resulting from addition of permanganate was about 0.43 mg/L. Thus, it appears that along with arsenic, manganese was also very effectively removed from water by ferric chloride coagulation.

Table 4.2: **Field Measurement of water quality at selected households**

Parameter	Unit	Household A		Household B		Household C	
		Raw	Treated	Raw	Treated	Raw	Treated
pH	--	6.5	6.3	6.3	6.2	6.0	6.0
Redox Potential	mV	-98	102	-103	103	-112	21
Flow rate	ml/min	--	900	--	1740	--	1160
Ferrous Iron (Fe^{2+})	mg/L	2.5~5.0	0.0~0.1	2.5~5.0	0.0~0.1	6.0~7.0	0.3~0.4
Total Iron (Fe)	mg/L	2.5~5.0	0.0~0.1	3.5	0.1~0.2	8.0~10.0	0.4~0.6
Nitrate (NO_3)	mg/L	0.0~0.1	1.0~1.5	0.0~0.1	1.0~1.5	0.0~0.1	0.6~0.8
Silica (SiO_3)	mg/L	50~60	40~50	60	60~70	40~50	25~30
Phosphate (PO_4^{3-})	mg/L	8~10	1~2	7~8	0.1~0.2	7~8	0.2~0.3

4.3.3 Bacteriological quality of water

For some of the ferric-chloride-based units, presence of fecal coliform was detected in the treated water (see Table 4.3). This appears to be, primarily, due to contamination of water during transportation from the tubewell to the upper bucket in the arsenic removal unit. However, it should be mentioned that raw tubewell water samples from some households also showed presence of fecal coliform. The sand filter media appear to sustain growth of fecal coliform as was evidenced from continued presence of these organisms in some of the units. This problem was however eliminated by introducing bleaching powder (1.0 mg per packet) in the chemical packet. Continued use of chemical packets with bleaching powder for a period of about 15 days eliminated faecal coliform.

Table 4.3: **Bacteriological quality of raw and treated water from some households**

Household designation	Fecal Coliform (# per 100 ml)		
	Tubewell water	Treated Water	
		Before addition of Bleaching powder	After Addition of Bleaching powder
A	Nil	TNTC	Nil
B	Nil	nil	Nil
C	Nil	1	Nil
D	Nil	7	Nil
E	5	5	Nil
F	TNTC*	TNTC	Nil

* TNTC : Too numerous to count

4.3.4 User Acceptance

Information about user acceptance were gathered from discussion with the users and from the questionnaire survey conducted among the users. As mentioned earlier, villagers of Adda complained about lack of government or non-government initiatives for detecting arsenic in the tubewells of this village, although they suspected presence of arsenic in their tubewell water for long. When this research team confirmed the presence of high level of arsenic in many of the tubewells in the village and proposed to provide some households with arsenic removal units on a test basis, people became very enthusiastic. Initially only five units were supplied. But, many in the village made requests for more units. With an objective of transfer of technology, the villagers were trained in constructing the removal units. Following the brief training, the villagers themselves built ten more arsenic removal units locally (at the village). There is a great demand for the unit, even some of the users offered contribution for chemicals.

After more than one year of operation, the ferric chloride based unit appeared to have become very popular with the people in the village Adda. Although there are differences in the level of enthusiasm regarding these units, people in general were very eager to use these units. This was particularly true among the people who were more aware about the adverse effects of arsenic. This was evidenced by requests from many more units by the people. Many people showed their willingness to pay for the chemical packets (which were being supplied free of cost by the research project).

The easy operation and maintenance (discussed in the following section) is one aspect that appeared to have made these units popular.

Apart from the arsenic removal efficiency of these units, the aspect that impressed people most was the clarity of water produced by these units. Many households identified this aspect as the primary reason for using the unit. With relatively high iron content (upto about 10 mg/L), raw water from many tubewells in the village showed high turbidity (resulting from precipitated iron flocs). The units were very effective in removing the iron content of water (along with arsenic) and the clear water produced was very attractive aesthetically. There was another interesting aspect regarding use of these units. Some of the households informed that they did not use the treated water for drinking during winter because the water was very cold; instead they used tubewell water directly, which was much warmer.

Discussion and survey results suggest that the poorer and relatively less educated families were relatively less enthusiastic about the ARU. Some of them considered daily operation and maintenance of the ARUs as a difficult task. This was mainly due to lack of awareness about the adverse health effects of arsenic. This reveals that health awareness is necessary to make any option acceptable to any community. Some users initially complained about a chemical smell in the treated water. But it appears that they got habituated with that smell after sometime and never complained about it again.

4.3.5 Operation & Maintenance

Most of the users were comfortable with the operation of the ARUs. However, some users felt that it was difficult to fill the upper bucket with water (at the tubewell) and then put it to the iron frame of the ARU. They preferred to fill the upper bucket by carrying water from the tubewell with a smaller bucket. Some households informed that it was difficult for women in the households to stir the water in the upper bucket. They suggested that if the upper bucket is placed at a lower height, it would be easier for them to stir. People informed that they have to regularly wash the white cloth placed in the upper bucket for straining some of the iron flocs coming from the upper bucket. The users also informed that they have to periodically wash the sand in the

lower bucket to maintain a reasonable flow rate of treated water (which varied from about 1 to 2 L/min). The frequency of washing varied from twice a week to about once in every two weeks, depending on the volume of water treated, the iron content of tubewell water and operation of the unit.

During field visits, it was observed that the instructions for operation of the unit, though simple, were not strictly followed by all the users. For example the mixing (one minute of rapid mixing and one and a half minutes of slow mixing) instructions were not always followed because many felt it was too much work. The required time for settling of iron flocs (one and a half hours) was also not maintained. But it should be mentioned that the arsenic removal efficiency did not appear to have been affected much by these irregularities. All the users disposed the sludge to their normal waste drains or to the cowdung pits.

4.3.6 Cost of Water Treatment

The total cost of constructing the unit was about Tk 520/-. The cost of each packet of chemical cost around Tk. 2.00/-. Detailed breakdown of the cost is provided in Table 4.4 and Table 4.5.

Table 4.4 : **Estimated Cost of the household arsenic removal unit**

Item	Quantity	Price (Tk.)	Remarks
35 liter bucket + lid	2 pc.	350.00	Depending on the quality of the bucket material
14 liter bucket	1 pc.	55.00	
Bib Cock	2 pc.	20.00	
Jam Nut	2 pc.	10.00	
Washer	4 pc.	4.00	
Strainer	1 ft	15.00	
Stopper Cap of Strainer	2 pc.	12.00	
Pipe of 0.24 inch dia.	2 ft.	7.00	
Cloth	4 sq. ft.	9.00	
Wooden Stirrer	1 pc.	15.00	
Others (Thread tape + solvent cement + Labour)	L.S.	10.00	
Sand	1 cft.	10.00	
Sub-total		517.00	
Frame (Optional)	LS	500.00	
Total		1017.00	

Prices are subjected to temporal and spatial variation.

Table 4.5 : **Cost of Chemical Pack**

Item	Quantity	Price (Tk.)	Remarks
Ferric Chloride (commercial grade)	2.5 gm	1.25	180 packets can be prepared with 500 gm.
Potassium Permanganate	35.0 mg	0.20	
Bleaching powder	1.0 mg	0.05	
Packet and Labour	LS	0.50	
	Total Tk. =	2.00	

4.4 Summary

Performance of arsenic removal unit was evaluated in the field in order to determine their suitability as a low cost household arsenic removal unit for the rural Bangladesh. Field testing of 15 ferric chloride based units were being conducted in the village of Adda in the Barura thana of Comilla district for one year.

Field testing at Adda village showed a good arsenic removal efficiency. Arsenic concentrations in the treated water were found to be mostly below 20 ppb level, much below the Bangladesh standard; while maximum arsenic concentration in the raw was about 400 ppb. For some of the units, presence of fecal coliform was detected in the treated water. Continued use of chemical packets with bleaching powder for a period of about 15 days eliminated fecal coliform. So there appears to be a need for a disinfectant (in addition to the coagulant and the oxidant) in the chemical packet for ensuring good bacteriological quality of water. Therefore the contents of the chemical packet finally consists of 2.5 gm of commercial grade ferric chloride with 35 mg of potassium permanganate and 1 mg of bleaching powder. The cost of chemical for treatment is about Tk. 0.10 per liter.

Field-testing showed that more than 70 % of the total arsenic is removed by settling of flocs while another 20% ~ 30 % arsenic is removed in the sand bed. Therefore, the cloth, which was used as a primary filter, is not functioning as expected. So the design of the unit should reject this cloth strainer. Some of the users complained about the height of the unit during field survey. So, reducing dimensions of the frame containing the buckets can reduce height of the unit and make it more users friendly. A 10-cm reduction can be done from the top of the frame. Lowering the gap between the two buckets can do another 5-cm reduction.

CHAPTER 5 : CONCLUSIONS & RECOMMENDATIONS

5.1 General

This study was a trial to apply results of previous studies for the benefit of the rural populace of Bangladesh, affected with arsenic contaminated groundwater and badly need a solution. It was planned first to verify and adjust the results of previous studies to field conditions.

Efficiency of arsenic removal by coagulation with alum and ferric chloride has been evaluated in this study. Results of coagulation experiments suggest that ferric chloride is much more efficient than alum in removing arsenic from groundwater. As expected, removal of arsenate was much more efficient than arsenite. In this study potassium permanganate was used as an oxidant to convert arsenite to arsenate for effective removal of arsenic. Bleaching powder was not used because of its unstable nature.

The test results were used to develop a low-cost household arsenic removal unit. The unit was tested in the field to ensure its capability as well as to get input form the users to make it user friendly. Its performance was monitored and the users suggested some modifications for easy operation and maintenance. It was decided to make the unit using local materials and easy techniques so that indigenous people can replicate it for use.

5.2 Conclusions

In this study efficiency of arsenic removal by coagulation with alum and ferric chloride has been evaluated. Coagulation-flocculation requires mixing of the coagulant with the water and the nature of mixing applied has a significant influence on formation of coagulated flocs. Experiments were conducted to determine the optimum mixing for achieving maximum floc formation. Experimental results showed that vigorous mixing for 10-15 seconds followed by 90 slow turns yielded best results.

This method of mixing was adopted in all coagulation experiments conducted in this study.

Results of coagulation experiments suggest that ferric chloride is much more efficient than alum in removing arsenic from groundwater. As expected, removal of arsenate was much more efficient than arsenite. Potassium permanganate was used as an oxidizing agent for oxidation of arsenite to arsenate for effective removal of arsenic. Experimental results suggest that a dose of potassium permanganate twice the stoichiometric requirement is sufficient for oxidation of arsenite to arsenate. However, use of potassium permanganate produced slight pink color in the treated water, which would be objectionable to the users. This color was removed using a sand filter. Laboratory test results suggest that a sand filter 20-cm deep was sufficient for removal of color. In this study, effect of phosphate on arsenic removal efficiency was evaluated in batch sorption experiments. Results show that presence of high level of phosphate can reduce the efficiency of arsenic removal by ferric chloride to some extent.

Performance of arsenic removal technologies/systems was evaluated in the field in order to determine their suitability as a low cost household arsenic removal unit for the rural Bangladesh. Field testing of 15 ferric chloride based units are being conducted in the village of Adda in the Barura thana of Comilla district for one year. Field testing at Adda village showed very good arsenic removal efficiency. Arsenic concentrations in the treated water were found to be mostly below 20 ppb level, much below the Bangladesh standard; while maximum arsenic concentration in the raw was about 400 ppb.

Field-testing of the arsenic removal unit showed that more than 70 % of the total arsenic is removed by settling of flocs while another 20% ~ 30 % arsenic is removed in the sand bed. Therefore, the cloth, which was used as a primary filter, is not functioning as expected. So the design of the unit should reject this cloth strainer. Some of the users complained about the height of the unit during field survey. So, reducing dimensions of the frame containing the buckets can reduce height of the unit and make it more users friendly. A 10-cm reduction can be done from the top of the frame. Lowering the gap between the two buckets can do another 5-cm reduction.

For some of the units, presence of fecal coliform was detected in the treated water. Continued use of chemical packets with bleaching powder for a period of about 15 days eliminated fecal coliform. So there appears to be a need for a disinfectant (in addition to the coagulant and the oxidant) in the chemical packet for ensuring good bacteriological quality of water. Therefore the contents of the chemical packet finally consists of 2.5 gm of commercial grade ferric chloride with 35 mg of potassium permanganate and 1 mg of bleaching powder. The cost of chemical for treatment is about Tk. 0.10 per liter.

5.3 Recommendations

Field and laboratory testing of the arsenic removal unit showed that it is efficient in removing arsenic from water as well as is acceptable to the local community. Even local people can construct the unit themselves with locally available material. But they don't have access to the required chemicals within their locality. Therefore, to make this unit sustainable, availability of required chemicals should be ensured in the market. One or more entrepreneur can be developed from the implementing agency in this regard.

Further study can be done to evaluate the reuse of the sludge for water treatment for second or third time. This study can proceed on presumption that there may be some unused surface available in the flocs for further adsorption of arsenic.

Dose of the chemicals can be further studied to optimize it to adjust with the available naturally occurring iron.

For the total solution of the arsenic problem from Bangladesh, this arsenic removal option can be used in conjunction with other safe water options i.e., rainwater harvesting, pond sand filtration etc. Conjunctive use of different sources at different seasons at different geological conditions can be studied to determine the contribution of different options in the total water supply.

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APPENDIX A

- BTU Users Manual in Bangla

পরীক্ষামূলক আর্সেনিক দূরীকরণ ইউনিট

ব্যবহার বিধি / নির্দেশিকা ঃ

১। লাল বালতিতে কালো দাগ পর্যন্ত পানি ভর্তি করুন।

২। সরবরাহকৃত একটি প্যাকেটের সবটুকু গুঁড়া পানিতে ঢেলে দিন।

৩। গুঁড়া পানিতে মেশানোর জন্য ঘুটনী দিয়ে ৫- ৬ বার দ্রুত নাড়ুন।

৪। এরপর ঘুটনী দিয়ে ধীরে ধীরে ৪০ - ৪৫ বার নাড়ুন।

৫। ঢাকনি দিয়ে লাল বালতিটি ঢেকে দিন এবং আনুমানিক ১ঘন্টা অপেক্ষা করুন।

৬। দুটি বালতির কলই খুলে দিন এবং নিচের বালতির কল থেকে খাবার পানি সংগ্রহ করুন।

৭। পানি পড়া বন্ধ হলে কল দুটি বন্ধ করুন।

৮। লাল বালতিতে থেকে যাওয়া পানি মাটির গর্তে / গোবর-গাদায় ফেলে দিন।

৯। সবশেষে লাল বালতি এবং কাপড়টি ধুয়ে রাখুন।

সতর্কতা ঃ

কল সাবধানে খুলুন এবং বন্ধ করুন যেন কল ও বালতির সংযোগস্থলে চাপ কম পড়ে।

রক্ষনাবেক্ষণ ঃ

বালি ময়লা হওয়ার কারণে কল হতে পানি পড়া কমে গেলে ময়লা বালি বালতি থেকে তুলে পানি দিয়ে ধুয়ে আবার ঐ বালতিতে রাখুন।

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APPENDIX B

- Sample questionnaire for field survey

পরীক্ষামূলক আর্সেনিক দূরীকরণ ইউনিট

মতামত জরিপ

ব্যবহারকারীর নাম ঃ _____

পেশা ঃ _____

প্রাতিষ্ঠানিক শিক্ষা ঃ _____

পিতার নাম ঃ _____

মাতার নাম ঃ _____

গ্রাম ঃ _____

ইউনিয়ন ঃ _____

উপজেলা ঃ _____

জেলা ঃ _____

আপনার উত্তরের ঘরে টিক (✓) চিহ্ন দিন ঃ –

১. পরিশোধিত পানির স্বাদ, গন্ধ ও রঙ কেমন ?

স্বাদ		গন্ধ		রঙ	
ভালো	খারাপ	আছে	নাই	আছে	নাই

২. দিনে কোন সময় এবং কয় বালতি পানি পরিশোধন করেন ?

সকাল	দুপুর	সন্ধ্যা
বালতি	বালতি	বালতি

৩. যতটুকু পানি শোধন করেন তা কি যথেষ্ট ?

হ্যাঁ	না

৪. নিচের কল থেকে পানি পড়ার হার আগের তুলনায় কেমন ?

বেড়েছে	অপরিবর্তিত	কমেছে

৫. কি কি কাজে পরিশোধিত পানি ব্যবহার করেন ?

রান্না	পান করা	অন্যান্য

৬. ইউনিট থেকে টিউবওয়েলের দূরত্ব কত ফুট?

০-১৫	১৫-৩০	৩০-৫০	৫০-১০০	১০০ +

৭. টিউবওয়েলের পানি অন্যান্য কি কি কাজে ব্যবহার করেন ?

৮. উপরের বালতিতে গুড়া গুলানোর সময় বালতির উচ্চতা বেশী মনে হয় কি ?

অনেক বেশী	সামান্য বেশী	ঠিকই আছে

৯. গুড়া গুলানোর সময় কত বার নাড়ছেন ?

২০-৩০	৩০-৪০	৪০-৫০	৫০-৬০	৬০+

১০. গুড়া গুলানোর কত মিনিট পরে পানি নেয়া শুরু করেন ?

০-৫	৫-১৫	১৫-৩০	৩০-৪৫	৪৫+

১১. পানিতে গুড়া গুলানোর কাজ কে করেন ?

নিজে	পরিবাররের অন্য সদস্য	অন্য কোন সাহায্যকারী

১২. পানি নেয়ার পরে তলানী ময়লা পানি কোথায় ফেলেন ?

গোবর গাদায়	নর্দমায়	পুকুরে	জমিতে	অন্যান্য

১৩. আপনার পরিবারের সদস্যদের সংখ্যা এবং তাদের শিক্ষাগত অবস্থা কি ?

জন	জন	জন	জন
স্বাক্ষর	প্রাথমিক	মাধ্যমিক	মাধ্যমিক +

১৪. গুড়া ক্রয় করে ব্যবহার করতে রাজি আছেন কি ?

হ্যা	না

১৫. গুড়ার খরচ প্রতিদিন সর্বোচ্চ কেমন হলে আপনার অসুবিধা হবে না ?

২ টাকা	৪ টাকা	৬ টাকা	৮ টাকা

১৬. পরিবারের কত জন আর্সেনিক আক্রান্ত ?

১৭. আর্সেনিক আক্রান্ত হলে কি কি অসুবিধা হবে বলে আপনি মনে করেন ?

APPENDIX C

- Field testing data of arsenic removal.

Summary Table of test results showing reliability of the unit in removing arsenic form water

Unit Designation	No. of Samples Exceeding arsenic concentration of 50 ppb	No. of treated water Sample tested	% Samples exceeding 50 ppb
A	3	42	7
B	1	34	3
C	0	32	0
D	0	20	0
E	0	15	0
G	0	5	0
H	0	2	0
I	0	2	0
J	0	3	0
K	0	4	0
L	0	2	0
M	0	2	0
Total	4	163	2

Owner Md. Abdus Sattar

Raw As.

450	ppb	2-Aug-2000
234	ppb	21-Aug-2000
241	ppb	23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1	Ka - 1	3	60	21-Jul-2000	2-Aug-2000	21
2	Ka - 2	7	140	22-Jul-2000	2-Aug-2000	17
3	Ka - 3	12	240	23-Jul-2000	2-Aug-2000	16
4	Ka - 4	18	360	24-Jul-2000	2-Aug-2000	19
5	Ka - 5	22	440	25-Jul-2000	2-Aug-2000	37
6	Ka - 6	25	500	26-Jul-2000	2-Aug-2000	17
7	Ka - 7	29	580	27-Jul-2000	2-Aug-2000	38
8	Ka - 8	33	660	28-Jul-2000	2-Aug-2000	22
9	Ka - 9	38	760	29-Jul-2000	2-Aug-2000	16
10	Ka - 10	42	840	30-Jul-2000	21-Aug-2000	3
11	Ka - 11	47	940	31-Jul-2000	21-Aug-2000	3
12	Ka - 12	55	1100	1-Aug-2000	21-Aug-2000	2
13	Ka - 13	58	1160	2-Aug-2000	21-Aug-2000	3
14	Ka - 14	61	1220	3-Aug-2000	21-Aug-2000	3
15	Ka - 15	65	1300	4-Aug-2000	21-Aug-2000	2
16	Ka - 16	69	1380	5-Aug-2000	21-Aug-2000	4
17	Ka - 17	73	1460	6-Aug-2000	21-Aug-2000	2
18	Ka - 18	84	1680	7-Aug-2000	21-Aug-2000	2
19	Ka - 19	88	1760	8-Aug-2000	21-Aug-2000	1
20	Ka - 21	92	1840	10-Aug-2000	23-Aug-2000	38
21	Ka - 22	96	1920	11-Aug-2000	23-Aug-2000	32
22	Ka - 23	101	2020	12-Aug-2000	23-Aug-2000	43
23	Ka - 24	105	2100	13-Aug-2000	23-Aug-2000	74
24	Ka - 25	118	2360	16-Aug-2000	23-Aug-2000	38
25	Ka - 26	137	2740	20-Aug-2000	31-Aug-2000	40
26	Ka - 27	152	3040	24-Aug-2000	31-Aug-2000	55
27	Ka - 28	165	3300	27-Aug-2000	31-Aug-2000	52
28	Ka - 29	176	3520	30-Aug-2000	7-Sep-2000	15
29	Ka - 30	187	3740	2-Sep-2000	7-Sep-2000	42
30	Ka - 31				7-Sep-2000	9
31	Ka - 32	193	3860	4-Sep-2000	7-Sep-2000	37
32	Ka - 33	202	4040	7-Sep-2000	14-Sep-2000	41
33	Ka - 34	205	4100	9-Sep-2000	14-Sep-2000	32
34	Ka - 36	215	4300	12-Sep-2000	14-Sep-2000	40
35	Ka - 37	241	4820	20-Sep-2000	31-Oct-2000	28
36	Ka - 38	268	5360	30-Sep-2000	31-Oct-2000	33
37	Ka - 39	288	5760	6-Oct-2000		
38	Ka - 40	312	6240	14-Sep-2000		
39	Ka - 41	351	7020	25-Oct-2000		
40	Ka - 42	369	7380	30-Oct-2000		
41	Ka - 43	400	8000	9-Nov-2000	23-Nov-2000	14
42	Ka - 44				23-Nov-2000	12
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Owner Abdul Malek

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Raw As. 643 ppb 2-Aug-2000
369 ppb 21-Aug-2000
379 ppb 23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	As ppb
1	Kha - 1	2	40	21-Jul-2000	2-Aug-2000	1
2	Kha - 2	4	80	22-Jul-2000	2-Aug-2000	8
3	Kha - 3	6	120	23-Jul-2000	2-Aug-2000	10
4	Kha - 4	8	160	24-Jul-2000	2-Aug-2000	55
5	Kha - 5	10	200	25-Jul-2000	2-Aug-2000	31
6	Kha - 6	12	240	26-Jul-2000	2-Aug-2000	29
7	Kha - 7	14	280	27-Jul-2000	2-Aug-2000	8
8	Kha - 8	16	320	28-Jul-2000	2-Aug-2000	20
9	Kha - 9	18	360	29-Jul-2000	2-Aug-2000	14
10	Kha - 10	20	400	30-Jul-2000	21-Aug-2000	2
11	Kha - 11	22	440	31-Jul-2000	21-Aug-2000	6
12	Kha - 12	25	500	1-Aug-2000	21-Aug-2000	4
13	Kha - 13	27	540	2-Aug-2000	21-Aug-2000	8
14	Kha - 14	29	580	3-Aug-2000	21-Aug-2000	2
15	Kha - 15	31	620	4-Aug-2000	21-Aug-2000	2
16	Kha - 16	33	660	5-Aug-2000	21-Aug-2000	2
17	Kha - 17	35	700	6-Aug-2000	21-Aug-2000	3
18	Kha - 21	36	720	7-Aug-2000	21-Aug-2000	2
19	Kha - 22	37	740	8-Aug-2000	23-Aug-2000	7
20	Kha - 23	40	800	9-Aug-2000	23-Aug-2000	6
21	Kha - 24	42	840	13-Aug-2000	23-Aug-2000	6
22	Kha - 25	44	880	14-Aug-2000	23-Aug-2000	3
23	Kha - 26	47	940	16-Aug-2000	31-Aug-2000	4
24	Kha - 27	50	1000	17-Aug-2000	31-Aug-2000	15
25	Kha - 28	51	1020	18-Aug-2000	31-Aug-2000	35
26	Kha - 29	67	1340	26-Aug-2000	31-Aug-2000	11
27	Kha - 30	78	1560	1-Sep-2000	14-Sep-2000	2
28	Kha - 31	83	1660	4-Sep-2000	14-Sep-2000	16
29	Kha - 32	85	1700	6-Sep-2000	14-Sep-2000	9
30	Kha - 34	91	1820	12-Sep-2000	31-Oct-2000	7
31	Kha - 35				31-Oct-2000	5
32	Kha - 41				23-Nov-2000	2
33	Kha - 66				7-Nov-2000	3
34	Kha - 67				7-Nov-2000	4
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Owner Dr. Delwar Hossain sir's Country home

Raw As. 374 ppb 2-Aug-2000
227 ppb 21-Aug-2000
233 ppb 23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	As ppb
1	Ga - 1				2-Aug-2000	12
2	Ga - 2				2-Aug-2000	6
3	Ga - 3				2-Aug-2000	5
4	Ga - 4				2-Aug-2000	21
5	Ga - 5				2-Aug-2000	27
6	Ga - 6				2-Aug-2000	11
7	Ga - 7				2-Aug-2000	13
8	Ga - 8				2-Aug-2000	17
9	Ga - 9				2-Aug-2000	14
10	Ga - 10				2-Aug-2000	10
11	Ga - 11				21-Aug-2000	2
12	Ga - 12				21-Aug-2000	2
13	Ga - 13				21-Aug-2000	2
14	Ga - 14				21-Aug-2000	3
15	Ga - 15				21-Aug-2000	3
16	Ga - 16				21-Aug-2000	3
17	Ga - 17				21-Aug-2000	3
18	Ga - 18				21-Aug-2000	3
19	Ga - 19				21-Aug-2000	2
20	Ga - 20				21-Aug-2000	3
21	Ga - 21				31-Aug-2000	5
22	Ga - 22				31-Aug-2000	5
23	Ga - 23				31-Aug-2000	3
24	Ga - 24				23-Aug-2000	4
25	Ga - 25				23-Aug-2000	4
26	Ga - 26				23-Aug-2000	3
27	Ga - 27				23-Aug-2000	4
28	Ga - 28				23-Aug-2000	2
29						9
30	Ga - 29				31-Oct-2000	7
31	Ga - 30				31-Oct-2000	8
32	Ga - 41				23-Nov-2000	10
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Owner Cherag Ali & Family

घ **Raw As.** 332 ppb 23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1	Gha - 1	2	40	11-Aug-2000	21-Aug-2000	1
2	Gha - 2			11-Aug-2000	31-Aug-2000	
3	Gha - 3	6	120	13-Aug-2000		
4	Gha - 4	10	200	15-Aug-2000	31-Aug-2000	3
5	Gha - 5	14	280	17-Aug-2000	31-Aug-2000	3
6	Gha - 6	18	360	19-Aug-2000	31-Aug-2000	2
7	Gha - 7	20	400	20-Aug-2000	31-Aug-2000	2
8	Gha - 8	28	560	24-Aug-2000	31-Aug-2000	31
9	Gha - 9	32	640	26-Aug-2000	31-Aug-2000	2
10	Gha - 10			28-Aug-2000	31-Aug-2000	2
11	Gha - 11				7-Sep-2000	2
12	Gha - 12				7-Sep-2000	2
13	Gha - 13				7-Sep-2000	3
14	Gha - 14				7-Sep-2000	2
15	Gha - 15				31-Oct-2000	3
16	Gha - 16				31-Oct-2000	4
17	Gha - 17				31-Oct-2000	3
18	Gha - 18				31-Oct-2000	2
19	Gha - 19				31-Oct-2000	2
20	Gha - 20				8-Nov-2000	1
21	Gha - 22				8-Nov-2000	2
22	Gha - 24					1
23	Gha - 25					
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Owner Loni Mia

Raw As. 149 ppb 23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1	Umo - 1	2	40	11-Aug-2000	21-Aug-2000	4
2	Umo - 2	4	80	12-Aug-2000	31-Aug-2000	2
3	Umo - 3	6	120	13-Aug-2000	31-Aug-2000	2
4	Umo - 4	8	160	14-Aug-2000	31-Aug-2000	9
5	Umo - 5	10	200	15-Aug-2000	31-Aug-2000	3
6	Umo - 6	14	280	17-Aug-2000	31-Aug-2000	4
7	Umo - 7	31	620	25-Aug-2000	31-Aug-2000	3
8	Umo - 8	40	800	29-Aug-2000	7-Sep-2000	2
9	Umo - 9				7-Sep-2000	6
10	Umo - 10				31-Oct-2000	2
11	Umo - 11	46	920	1-Sep-2000	31-Oct-2000	2
12	Umo - 12	50	1000	3-Sep-2000	31-Oct-2000	3
13	Umo - 13	54	1080	5-Sep-2000	31-Oct-2000	2
14	Umo - 14	58	1160	7-Sep-2000	31-Oct-2000	3
15	Umo - 15	60	1200	9-Sep-2000	31-Oct-2000	3
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Owner Belal Hossain

Raw As. 226 ppb 23-Aug-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	As ppb
1	Cha - 1	3	60	11-Aug-2000	21-Aug-2000	2
2	Cha - 2	7	140		3-Sep-2000	2
3	Cha - 3	13	260		3-Sep-2000	2
4	Cha - 4	22	440		3-Sep-2000	2
5	Cha - 5	29	580		3-Sep-2000	2
6	Cha - 6	36	720		3-Sep-2000	2
7	Cha - 7	43	860		3-Sep-2000	2
8	Cha - 8	55	1100		3-Sep-2000	2
9	Cha - 9	62	1240		7-Sep-2000	3
10	Cha - 10				7-Sep-2000	3
11	Cha - 11	71	1420	18-Sep-2000	31-Oct-2000	3
12	Cha - 13	87	1740	2-Nov-2000	2-Nov-2000	3
13	Cha - 14	99	1980	25-Sep-2000		
14	Cha - 15	11	220	28-Sep-2000		
15	Cha - 16	123	2460	1-Oct-2000		
16	Cha - 17	145	2900	8-Oct-2000		
17	Cha - 18	187	3740	22-Oct-2000		
18	Cha - 19	207	4140	29-Oct-2000		
19	Cha - 20	230	4600	5-Nov-2000		
20	Cha - 21	244	4880	12-Nov-2000		
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Owner Mr. Sattar (worked in BRDB Rtd.)

Raw As. 70 ppb 14-Sep-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1					14-Sep-2000	4
2	Chha - 01				2-Nov-2000	3
3	Chha-1 Ru				2-Nov-2000	3
4	Chha - 08					1
5	Chha - 09					2
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	Owner		Bank	
ऊ	**Raw As.**	40	ppb	14-Sep-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1					14-Sep-2000	1
2						1
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Owner Ruhul Amin

U3 **Raw As.** 268 ppb 14-Sep-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1					14-Sep-2000	4
2	Nio - 10					14
3	Nio - 11					15
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Owner Abdul Hai

Raw As. 89 ppb 14-Sep-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	As ppb
1					14-Sep-2000	2
2	Tta - 2!				2-Nov-2000	2
3	Tta - 3!				2-Nov-2000	3
4	Tta - 21					1
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Owner Idris (At the same house of Ka)

Raw As. 240 ppb

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	**As** ppb
1	Da - 01					2
2	Da - 02					2
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Owner The Orphanage

ব **Raw As.** 129 ppb 2-Nov-2000

Sl No.	Sample No.	Cumulative water treated (Buckets)	Cumulative water treated (Liters)	Date of Collection	Date of Testing	As ppb
1					2-Nov-2000	1
2					2-Nov-2000	3
3						
4						
5						
6						
7						
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