ARSENIC SPECIATION AND EVALUATION OF **AN** ADSORPTION **MEDIA** IN **RUPANDEHI AND** NAWALPARASI DISTRICTS OF **NEPAL**

by

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Submitted to the Department of Civil and Environmental Engineering in Partial Fulfillment of the Requirements for the Degree of

MASTER OF **ENGINEERING IN** CIVIL **AND ENVIRONMENTAL ENGINEERING** at the **MASSACHUSETTS INSTITUTE** OF **TECHNOLOGY**

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ABSTRACT

A study of arsenic speciation in the drinking water of Nepal and an evaluation of an arsenic adsorption media was conducted as part of the 2001-2002 Massachusetts Institute of Technology Nepal Water Project. Over 40 tubewell water samples were collected in the Nawalparasi and Rupandehi Districts of Nepal where arsenic contamination is known to occur. Arsenic **(III)** and arsenic (V) in the raw water were separated using Bio-Rad **AG1-X8** strong ion exchange resin. Arsenic concentration was analyzed in Nepal using Industrial Test Systems Quick Arsenic Test Kit. Samples were also preserved and brought back to MIT for analysis using Graphite Furnace Atomic Absorption Spectrometry. Results showed that arsenic **(III)** is the dominant inorganic arsenic species. On average, **79%** of all inorganic arsenic is in the **(+III)** oxidation state, while the rest is in (+V) oxidation state. There **is** a strong correlation between arsenic speciation and the oxidation reductive potential (ORP) as measured **by** a Digi-Sense Digital pH/mV/ORP Meter.

An activated alumina manganese oxide (A/M) adsorption media was tested at **9** locations with total arsenic in the raw water ranging from 147 μ g/L to 863 μ g/L. In all 9 runs, the treated water contained arsenic below detection limit $(<5 \mu g/L)$. Based on these promising results, further evaluation of the A/M media on its technical, social, and economic aspects is recommended.

Thesis Supervisor: Susan Murcott, Title: Lecturer, Department of Civil and Environmental Engineering

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CHAPTER 1 INTRODUCTION

1.1 Nepal Water Project Overview

The Massachusetts Institute of Technology (MIT) Nepal Water Project 2001-2002 is a program intended to increase the awareness of water quality problems in the country of Nepal, and to provide assistance in solving these problems. Since **1999,** 20 Master of Engineering students at the MIT Civil and Environmental Engineering Department traveled to Nepal to study various water quality and household/community level water treatment issues. This year's project is a collective effort of eight students: Heather Lukas, Chian Siong Low, Hannah Sullivan, Yong Xuan Gao, Luca Morganti, Barika Poole, Jeff Hwang, and Tommy Ngai.

Starting in September 2001, these eight students conducted background research into the water issues in Nepal. They traveled with Susan Murcott, their project advisor, to Nepal for three weeks in Jan 2002 to carry out their field studies. While in Nepal, these students worked closely with local agencies including Environment and Public Health Organization **(ENPHO)** in Kathmandu and in Parasi, International Buddist Society (IBS) in Lumbini, and the Finnish Department of International Development Co-operation **(DIDC),** formerly called the Finnish International Development Agency **(FINNIDA),** in Butwal.

The MIT Nepal Water Project is completed as part of the degree requirements for the Master of Engineering program in the Civil and Environmental Engineering Department of MIT. Each student writes an independent thesis and the team produces a group report. This thesis is one of those independent theses.

1.2 Project Motivation

Nepal is a developing country in south central Asia landlocked between China to the north and India to the south. The land area is 140 **000 km2** and the year 2000 population is **23** millions, of which 20 millions is rural'. Nepal is one of the world's poorest and least developed countries. The average annual income per capita is \$210 US². About 42% of the people live below the national poverty line³. Due to the poor economic conditions and ineffective institutional programs, proper water and sanitation services are inadequate, resulting in serious health concerns. The severity of the water problems is even more prominent in the rural remote villages. The infant mortality rate is very high at 74/1000 live births, compared with **5/1000** in the **U.S.** The under five mortality is even higher at 105/1000 birth⁴. 54% of the children suffers moderate to severe stunting on account of water borne diseases⁵. Diarrheal diseases kill 44000 children annually. The average life expectancy is only **58,** compared with **77** in the **U.S. 6.** Their serious health concern is the main motivation for this project

1.3 Project Goal

The MIT Nepal Water Project 2001-2002 is a continuation and expansion of the two previous Nepal Water Projects. As in past years, the aim of the project is to improve the health situation in Nepal through improvement in water supply quality. For this year, there are three main goals. The first goal is to continue to monitor microbial and arsenic contamination in Nepal. The second goal is to study the appropriateness of various pointof-use household water treatment systems based on technical performance, social acceptability, and affordability. Technologies investigated include filtration, and arsenic removal. The last goal is to evaluate past pilot studies and/or implement new projects, including the Lumbini pilot study, a tubewell maintanence program, and a chlorine generation micro-enterprise project.

Arsenic is a toxin that causes serious health effects such as cancer. The evaluation of an arsenic removal technology, as well as arsenic speciation in tubewell water are the focus

of this thesis. Figure 1 and Figure 2 show two very common types of tubewells, a lift tubewell and a suction tubewell, for which arsenic contaminated groundwater has been sampled and tested.

Figure 1 **A** Typical Lift Tubewell

Figure 2 **A** Typical Suction Tubewell

CHAPTER 2 BACKGROUND

2.1 Arsenic in the Environment

Arsenic is a naturally occurring element widely distributed in the earth's crust. It is the **51 st** most abundant element on earth, with an average level of **1.8** mg/kg in the earth's crust. Normal background concentration are **0.2-15** mg/kg in the lithosphere, less than **¹⁵** mg/kg in soils, 0.02 -2.8 ng/m³ in the atmosphere, and less than 1 μ g/L in aquatic **⁷** environment **.**

Arsenic can be released into the environment **by** either natural or anthropogenic processes. Dominant natural processes are volcanic activities and weathering of arsenic bearing rocks, including realgar (AsS), orpiment (As₂S₃), arsenopyrite (FeAsS), and lollingite (FeAs₂)^{8, 9}. Anthropogenic sources of arsenic are numerous. They include the application of arsenical pesticides on land, incineration of arsenic containing substances, industrial wastewater discharge, mine tailing/landfill leaching, and manufacturing of arsenic compounds $^{10, 11, 12}$.

Since arsenic is an element, it is indestructible. It can only change from one form to another, and be transported from one medium to another¹³. Arsenic in air will settle to the ground or be washed out **by** rain. Arsenic in water may precipitate out of the solution, or adsorbed onto rocks and soils. Arsenic-containing rocks and soils may release arsenic into the water by dissolution or desorption¹⁴. Of the above media, aqueous arsenic is of most concern because high levels arsenic have found in drinking water in various places around the world, and are known to cause serious adverse health effects. Therefore aqueous arsenic will be the main focus in this report.

2.2 Arsenic Chemistry

Arsenic is a group V element with an atomic mass of 74.9 **g/mo1¹⁵ .** Contrary to popular belief, arsenic is not a metal, but a metalloid. Arsenic forms a number of inorganic and

organic compounds. Naturally occurring inorganic arsenic is stable in oxidation states of -III as in arsine gas (AsH3), **0** as in crystalline arsenic, **+111** as in arsenite, and +V as in arsenate¹⁶. Organic arsenic species include monomethylarsonic acid (MMAA), and dimethylarsonic acid (DMAA)¹⁷. They may be produced by biological activity, mostly in surface waters, but are rarely quantitatively important. Organic forms may, however, occur where waters are significantly impacted **by** industrial pollution'⁸ . In the relatively pristine natural ground water environment, arsenic **(III)** and arsenic (V) are typically the dominant forms of arsenic.

2.2.1 Acid-base Chemistry

Depending on pH, different forms of arsenic (III) are H_3AsO_3 , H_2AsO_3 ^{*}, $HAsO_3^2$ ^{*}, and AsO_3^3 whereas different forms of arsenic (V) are H_3AsO_4 , H_2AsO_4^7 , H_3AsO_4^2 , and AsO4 **3-.** Refer to Figure **3** and Figure 4 for the protonation forms of arsenic **(III)** and arsenic (V) at various **pH.** These diagrams are generated **by** the following equilibrium relationships¹⁹.

For arsenic **(III),**

Figure **3** Protonation Forms of Arsenic (III) at Various **pH**

Figure 4 Protonation Forms of Arsenic (V) at Various **pH**

The amount of protonation of both arsenic **(III)** and arsenic (V) is an important factor governing the mobility of these chemical species. For example, the **pH** of groundwater is often between **6** to **8.** Within this range, arsenic **(III)** is uncharged while arsenic (V) is negatively charged. As a result, arsenic **(III)** is more mobile than arsenic (V). The movement of arsenic (V) is retarded **by** electrostatic attraction to positively charged particles, such as iron hydroxides²⁰. This information is also useful in designing effective arsenic removal technologies and in determining the arsenic speciation **by** ion exchange separation technique.

2.2.2 Reduction-Oxidation (redox) Chemistry

Given the acid-base chemistry, combined with Gibb's free energy change of the oxidation/reduction reactions between arsenic (III) and arsenic (V), an **Eh-pH** diagram (Pourbaix diagram) can be constructed^{21,22}. Refer to Figure 5 for an Eh-pH diagram of arsenic.

The **Eh-pH** diagram shows the exact arsenic speciation and oxidation states at a particular pH and redox potential²³. The diagram also shows the expected change in arsenic state when environmental conditions differ. For example, anoxic groundwater usually has a low redox potential. When the water is pumped to the ground surface and exposed to the atmosphere, the presence of dissolved oxygen increases the redox potential. As a result, arsenic **(III)** will naturally oxidize to arsenic (V).

This information is very useful because different oxidation states of arsenic also have different toxicities. Studies show arsenic **(III)** can be a degree of magnitude more toxic than arsenic $(V)^{24,25}$. Refer to Chapter 3 Health Effects, for more information on toxicology. Refer to Chapter **7** Results and Discussions, for more information on the kinetics of arsenic **(III)** oxidation.

Figure **5 Eh-pH** Diagram of Aqueous Arsenic Species in the System As-02-H20 at 25°C and 1 bar Total Pressure²⁶

2.3 Arsenic Situation in South Asia

In the early 1970s, most of the rural population of Nepal and the neighboring countries of Bangladesh and India received their drinking water from surface sources, including lakes, rivers, and ponds. Because this water became increasingly polluted, water-borne diseases were prevalent. Nearly a quarter of a million children died each year from these diseases²⁷. Throughout the late 1970s and the 1980s, there was a worldwide push to improve the general health condition of the people in developing nations. Infant mortality was one of the target health indicators. It was well understood that access to clean drinking water source could reduce infant mortality²⁸. As a result, many international aid agencies such as the World Bank, and the United Nations International Children's and Educational Fund **(UNICEF)** became involved in funding the installation of tubewells for domestic water supply. Since groundwater contains relatively less microbial contamination than surface water, groundwater was believed to be a better source of drinking water supply²⁹. Tubewell construction was simple and cheap. In addition, tubewells could be installed close to people's home, saving time for them to travel to surface water sources. Furthermore, tubewells provided rural villagers with a year round source of water to irrigate their fields, greatly contributing to the green revolution of food self-sufficiency. Millions of wells were built during the period of the late 1970s to 1980s^{30, 31}. In Bangladesh, the tubewell construction programs by various foreign aid agencies were successful in improving coverage of "safe" drinking water to over **97%32.** Partly due to the results of various successful tubewell installation programs, infant mortality in Bangladesh dropped significantly from **151/1000** live births in 1960 to $83/1000$ live births in 1996^{33} .

At the time, however, the standard water quality testing procedures did not include tests for arsenic. Starting in **1983,** cases of arsenic-induced skin diseases began to appear in West Bengal, India³⁴. However, the issue remained unnoticed. In 1993, the Bangladesh Department of Public Health Engineering (DPHE) identified their first arsenic-affected patient. Well testing was conducted in the western districts of Bangladesh in September of the same year. **Of** the **33** tubewell samples tested, **13** had arsenic level more than WHO guideline of 10 μ g/L. A committee consisting of representatives from DPHE,

National Institute of Preventive and Social Medicine **(NIPSOM),** Atomic Energy Commission, Geological Survey of Bangladesh, **UNICEF,** and other concerned organization was formed in 1994 to review the situation of arsenic in drinking water in Bangladesh. Further well survey was recommended. The Government of Bangladesh was contacted as well, but the government remained silent. The arsenic situation again remained unnoticed. In June **1996,** a non-profit **NGO** called the Dhaka Community Hospital (DCH) received several patients showing signs of arsenical skin lesions³⁵. DCH promptly tested tubewells in the area, and found high arsenic content in the water. **DCH** informed the local officials and made newspaper reports. Arsenic contamination concern began to surface in Bangladesh. Finally in **1997,** the Bangladesh Government officially confirmed that its country's drinking water supply is seriously contaminated with arsenic. Arsenic in Bangladesh made headlines around the world³⁶.

Bangladesh currently faces the world's most serious arsenic crisis. It is estimated between **20-60** million people of the country's total population of *125* million are at risk of exposure to arsenic in their drinking water over the Bangladesh standard of **50 pg/L37,38 .** At least **100 000** cases of debilitating skin lesions are believed to have already occurred³⁹. Another 4-7 million people are at risk in neighboring West Bengal.⁴⁰ Figure **6** shows the relative locations of Nepal, Bangladesh, and West Bengal (India).

Figure **6** Relative Location of Nepal, Bangladesh and West Bengal

2.4 Arsenic Situation in Nepal

The Nepal Terai is the flat plain region in the southern part of the country, a part of the Gangetic watershed. Due to its proximity to Bangladesh and West Bengal, various groups became concerned that arsenic can be a problem in Nepali drinking water. In **1999,** the Department of Water Supply and Sewerage **(DWSS)** received financial support of World Health Organization (WHO) and **UNICEF** to initiate various arsenic monitoring programmes⁴¹. These programmes were conducted on the groundwater of southern Nepal **by** a number of national and international organizations. In the **1999** study **by DWSS** in

the districts of Jhapa, Morang, and Sunsari in eastern Nepal, it was found that **9%** of the **268** tube well water samples contained arsenic above the WHO guideline of **10** pg/L. The highest concentration was found to be $75 \mu g/L^{42}$. In January 2000, a study by Halsey of the MIT Nepal Water Project **1999-2000** showed that **18%** of the **172** tested tube wells of the Terai region were contaminated with arsenic level above the WHO guideline⁴³. In the same year, in study **by** the Nepal Red Cross Society **(NRCS)** in the three districts of eastern Terai namely Jhapa, Saptari and Sarlahi, it was found that **5.1%** of the tube wells has arsenic contamination above the WHO guideline. The highest level detected was **56 pg/L 44.** The **NRCS** also conducted a study in 2001 in eight other districts of the Terai region including Rautahat, Bara, Parsa, Nawalparasi, Rupandehi, Kapilvastu, Banke, and Bardiya. It was found that 22% of the total investigated tube wells have arsenic level exceeding the WHO guideline, and the maximum level of contamination was found to be **165** tg/L **45.** Also in 2001, the Rural Water Supply and Sanitation Support Program (RWSSSP) in collaboration with the Finnish International Development Co-operation **(DIDC),** formerly called the Finnish International Development Agency **(FINNIDA)** showed that **9.8%** of **1508** samples in the districts of Rupandehi, Nawalparasi, and Palpa have over 10 μ g/L of arsenic ⁴⁶. The highest was measured in the village development community **(VDC)** of Devdaha of Rupandhi district where two wells have over 2000 μ g/L of arsenic⁴⁷. From these studies, it is clear that arsenic is a problem in the groundwater of the Nepal Terai region. It can potentially escalate into a serious health issue if the problem is not addressed properly.

2.5 Origins of Arsenic

Due to the low level of industrial activities in Nepal and Bangladesh, it is generally agreed that natural processes, as opposed to anthropogenic activities, are the dominant source of arsenic in these regions. Several hypotheses have been put forward as to the source and release mechanisms of arsenic in groundwater. The two most popular hypotheses are pyrite oxidation and iron oxyhydroxide reduction⁴⁸.

2.5.1 Pyrite Oxidation Hypothesis

The pyrite oxidation hypothesis was proposed **by** Dr. Dipankar Chakraborti and his group⁴⁹. Studies have shown that there is a high level of arsenopyrites in the alluvial regions of Bangladesh⁵⁰. After tubewells were installed, the water table was lowered due to increased pumping. As a result, atmospheric oxygen invaded the aquifer **by** diffusing into the pore spaces of the soil and into the groundwater. The oxygen interacts with the arsenopyrite, turns the mineral into water-soluble form, thus releaseing arsenic into the groundwater $51, 52, 53$.

However, this hypothesis was not consistent with the observation that shallow, oxic wells generally contained much lower level of arsenic than deeper, anoxic wells⁵⁴. In addition, if pyrite was oxidized, the aquifer should be rich in sulfate, but it was not⁵⁵. Recent works **by** Harvey et al found sulfate levels were lower at locations with higher arsenic concentration **6.** In the conference on arsenic mitigation in Dhaka on January 14th -16th 2002, there was a widespread feeling that the oxidation theory was not reasonable⁵⁷.

2.5.2 Iron Oxyhydroxide Reduction Hypothesis

According to the iron oxyhydroxide reduction hypothesis, the source of arsenic was from the Ganges source region upstream of Bangladesh, where arsenic-rich sulfide minerals were weathered during the late Pleistocene-Recent times. As these weathered minerals traveled down the Ganges, arsenic was adsorbed to iron oxyhydroxide (FeOOH). The arsenic-rich iron oxyhydroxide were deposited at the Gangetic delta, and formed an alluvial aquifer. Due to a wide variety of processes, such as agriculture or groundwater pumping, organic carbon was introduced into the aquifer. The mineralization of organic carbon caused anoxic condition in the aquifer, and lowered the redox potential. The reducing environment caused the breakdown of iron oxyhydroxide, and released the previously adsorbed arsenic $58, 59$.

One of the arguments supporting this hypothesis was that studies showed arsenic correlated well with iron and dissolved bicarbonate⁶⁰. Arsenic concentration also

increased with depth 61 . These observations suggested that arsenic was released when arsenic-rich iron oxyhydroxides were reduced in the anoxic condition, a process that mobilized iron and its absorbed load, and increased bicarbonate concentration⁶².

Recent works **by** Harvey et a163 further supports this hypothesis **by** showing strong correlations between arsenic and several key chemical species, including calcium, ammonium, organic carbon, sulfate, and iron in groundwater at various depth. For example, arsenic and sulfate shows a strong negative correlation, meaning that arsenic concentrations are high when sulfate concentrations are low. One explanation for low sulfate concentration at a particular depth is that sulfate reduction is taking place. According to the ecological redox sequence, sulfate reduction should occur after iron reduction, because sulfate reduction is less energetically favorable then iron reduction⁶⁴. Therefore, at depths with low sulfate, iron reduction should have already occurred. The reduction of iron released arsenic into the groundwater. As a result, arsenic concentrations are high when sulfate concentrations are low.

2.6 Arsenic Treatment Technologies

Since the discovery of arsenic in Bangladesh and West Bengal, many researchers worldwide have focused their efforts on developing appropriate arsenic removal technologies to treat the arsenic contaminated groundwater. These technologies can be broadly categorized according to their dominant removal process. They include oxidation, coagulation/precipitation, filtration, adsorption, ion exchange, membrane processes, and biological processes⁶⁵. Although many of these technologies are theoretically sound and work well in a controlled laboratory environment, many of them have not been field tested for their performance and appropriateness. In the MIT Nepal Water Project 2000-2001, Hurd evaluated the performance and appropriateness of three promising technologies: Three-Gagri System, Jerry Can System, and Arsenic Treatment Unit (ATU)⁶⁶. The Three-Gagri System was found to be both effective and appropriate, but clogging and possible microbial growth were potentially the main problems. The Jerry Can System was found to be ineffective and inappropriate with its current design.

The **ATU** was quite effective, but too expensive for mass implementation in South Asian contexts.

2.7 Alternative Water Source

Besides removing arsenic from groundwater, alternative water sources such as rainwater harvesting, dugwell, and deep wells may provide arsenic-free drinking water to the Nepali people. Rainwater harvesting is under studied **by DIDC** and the preliminary results are encouraging^{67}. The town of Parasi in the Nawalparasi district currently taps into a deep aquifer to provide drinking water of low arsenic concentration to the townspeople. However, construction of deep wells requires more sophisticated equipment and is thus more expensive, at about US\$500 per well⁶⁸.

CHAPTER 3 HEALTH EFFECTS

Arsenic has been long known as a poison. Even at low concentration, it can produce devastating human health effects⁶⁹. The toxic character of arsenic species mainly depends upon their chemical form. The most toxic form is arsine gas, followed **by** inorganic trivalent compounds, organic trivalent compounds, inorganic pentavalent compounds, organic pentavalent compounds and elemental arsenic^{70, 71}. Both the WHO and EPA have classified inorganic arsenic as a toxin and carcinogen⁷².

3.1 Route of Entry

Given that arsenic can be found in different environmental media, possible routes of entry include inhalation of arsenic contaminated air, ingestion of arsenic containing food and water, and skin contact⁷³. Air borne arsenic concentration is usually between 0.02 and 4 ng/m3 . This concentration is too low to induce any noticeable health effects **by** inhalation⁷⁴. As for skin contact, arsenic does not readily absorb into skin upon contact. Therefore, inhalation and skin contact are negligible source of entry for arsenic. The ingestion of arsenic containing food and/or water is the most important route of entry. **Of** the many food categories, fish and shellfish contain the highest level of arsenic 75. **Up** to 40 tg of arsenic per gram of dry weight fish can be found. Fortunately, over **90%** of the arsenic is in organic form, which is only very mildly toxic⁷⁶. In contrast, for arsenic contaminated drinking water, most of the arsenic is in the more toxic inorganic form. Arsenic levels in groundwater typically average around 1 to 2 μ g/L. However, in areas with volcanic rock and sulphide mineral deposits, arsenic levels in excess of **3000** ug/L have been measured⁷⁷. Therefore, arsenic in drinking water is of the most concern.

3.2 Acute Toxicity of Arsenic (III) and (V)

Ingestion of large doses of arsenic usually results in symptoms within **30** to **60** minutes, but may be delayed when taken with food. Acute arsenic poisoning usually starts with a metallic or garlic-like taste, burning lips and dysphagia. Then, violent vomiting and

hematemesis may occur⁷⁸. These gastrointestinal symptoms are a result of intestinal injury caused **by** dilatation of splanchnic vessels leading to mucosal vesiculation. After the initial gastrointestinal problems, multi-organ failures may occur, followed **by** death. Survivors of acute arsenic poisoning commonly incur damage to their peripheral nervous **79** system

Arsenic **(III)** and (V) behaves differently in acute poisoning. Arsenic **(III)** binds and inactivates sulfhydryl-containing enzymes necessary for proper body functions. On the other hand, arsenic (V) elicits toxicity **by** mimicking phosphate and interfering with ATP **b** production in the mitochondria⁸⁰.

Acute poisoning has a mortality rate of *50-75%* and death usually occurs within 48 hours. **A** lethal does will vary with the arsenic *form,* but **0.2-0.3 g** of arsenic trioxide is usually fatal for adult humans⁸¹. Reported arsenic (V) LD_{50} values in rats are 110 mg/kg, while the LD50 values in rats for arsenic **(III)** varies from **15** mg/kg to **110** mg/kg. Therefore, arsenic (III) is a magnitude more acutely toxic than arsenic $(V)^{82}$. However, in the context of drinking water supply, acute poisoning is less common than chronic exposure.

3.3 Chronic Toxicity of Arsenic (I1) and (V)

Chronic exposure to low level of arsenic has long since been linked to adverse health effects in human. There are contradictory beliefs on the relative chronic toxicity of arsenic (III) and (V). On one hand, arsenic **(III)** should be more toxic than (V), as an extension of acute toxicity data. On the other hand, some believe that chronic toxicity at low arsenic levels, as found in most groundwater, is influenced only **by** total arsenic concentration, not speciation **83.** No matter which hypothesis is correct, long-term exposure to arsenic has proven to cause dermal, vascular, and cancer effects⁸⁴

3.3.1 Dermal

Initially, chronic exposure to arsenic causes skin changes such as hyperpigmentation and keratosis. Hyperpigmentation is an alteration in color resulting in spots on the skin and

keratosis is a hardening of skin bulges, usually found in palms and soles. Following hyperkeratosis and hyperpigmentation, cancer may occur. After **10** years of exposure, cancer of the skin may develop⁸⁵. Figure 2 shows a keratosis victim. Recent studies from West Bengal, India and Bangladesh in populations showed that that the ageadjusted prevalence of keratosis rose from zero in the lowest exposure level $(< 50 \mu g/L)$ to 8.3 per 100 women drinking water containing $> 800 \mu g/L^{86}$. For men, the age-adjusted prevalence rates rose from 0.2 per **100** in the lowest exposure category to **10.7** per **100** in the high exposure group **7.** For hyperpigmentation prevalence, similar results were reported⁸⁸. Figure 7 shows a victim of keratosis.

Figure **7 A** Keratosis Victim

3.3.2 Vascular Effects

Exposure to arsenic has been linked to various vascular diseases affecting both the large (cardio-vascular) and small blood vessels (peripheral vascular). Blackfoot disease (BFD) in parts of Taiwan is an example of peripheral vascular disease. BFD is characterized **by** coldness and numbness in the feet, followed **by** ulceration, black discoloration and

subsequently dry gangrene of the affected parts. In addition many of the BDF-patients have shown significantly higher death rate from cardio-vasuclar problems⁸⁹.

3.3.3 Cancer

In additional to skin cancer, arsenic exposure in drinking water causes lung, bladder and kidney cancer may appear after 20 years or more years⁹⁰. Studies have consistently shown high mortality risks from lung, bladder and kidney cancers among populations exposed to arsenic via drinking water. Moreover, the risk of cancer for these sites increases with increasing exposure⁹¹.

CHAPTER 4 ANALYTIC METHOD

Numerous relevant water quality parameters were measured in this arsenic study. These parameters include:

- Total inorganic arsenic
- Arsenic speciation
- **" pH**
- Oxidation-reduction potential (ORP)
- Total alkalinity
- Total hardness as CaCO₃
- Total soluble iron
- **"** Manganese

A description of each of the above water quality parameters and their test methods are explained in this section.

4.1 Total Inorganic Arsenic

Description

Total inorganic arsenic in Rupandehi and Nawalparasi districts of Nepal was comprised mainly of arsenic (III) and arsenic (V). Total inorganic arsenic was first measured with an Industrial Test Systems Inc. Arsenic CheckTM Field Test Kit, and then confirmed with a Graphite Furnace Atomic Adsorption Spectrometry **(GFAAS).** Other forms of arsenic were irrelevant at these sites, thus were not measured. For example, other inorganic forms of arsenic such as arsine or arsenic element were not found in natural waters. Organic arsenic was formed only by bacteria methylation of arsenite^{92, 93}. The level of organic arsenic was believed to be low in anoxic groundwater. Furthermore, the toxicity of organic arsenic was very low compared to inorganic forms; therefore, organic arsenic was ignored.

4.1.1 Industrial Test Systems Inc. Arsenic CheckTM Field Test Kit 4811964

This arsenic field test kit shown in Figure **8** provides a safe, simple, and reliable method to test for aqueous inorganic arsenic. It requires no electricity and no refrigeration. The detection range is from **0** to **800** mg/L. The upper detection limit can be extended to 4000 mg/L with a simple 1 to *5* dilution. **All** components are supplied in the kit, including a detailed description of the test method, a color chart, three chemical reagents with material safety data sheets, three measuring spoons for the reagents, test strips, two reaction bottles, four bottle caps, a thermometer, and a zip-loc test strips disposal bag.

Figure 8 Industrial Test Systems Arsenic Check[™] Field Test Kit 481196

The test results are determined **by** colorimetry. The color chart is standardized at *25'C,* starting at pure white for **0 ppb** arsenic, a tint of yellow for *5* **ppb** arsenic, to slightly more yellow at **10** mg/L, and increasing yellow intensity at 20, *50,* **100,** 200, and **500** mg/L. At **500** mg/L arsenic, the color is dark brown. It is recommended that the water sample temperature be between **20'C** and **30'C** for accurate reading off the color chart. This test tolerates up to 2 mg/L hydrogen sulfide and *0.5* mg/L antimony without test result

interference. No interference from iron or sulfate was found. There are **100** tests per kit, selling at \$120 per kit.

Procedure

The chemistry of the reaction is based on the conversion of inorganic arsenic compounds in water to arsine gas $(AsH₃)$ by the reaction of zinc dust and tartaric acid⁹⁵.

- **1.** First, a clean reaction bottle is filled with raw water to the **100** mL mark. Then, three level pink spoons of reagent **1,** tartaric acid, are added to the bottle. The bottle is capped and shaken vigorously for **15** seconds. The tartaric acid is dissolved. The contents are allowed to sit for **15** seconds.
- 2. Three level red spoons of reagent 2, which contains a mixture of ferrous salts as reaction catalyst, are introduced. Again, the bottle is capped and shaken vigorously for **15** seconds. The metal salts are dissolved. The content is allowed to sit for 2 minutes.
- **3.** Then three level white spoons of reagent **3,** zinc dust, is added to the bottle and shaken vigorously for **15** seconds. The bottle cap is replaced with another cap that allows a test strip containing mercuric bromide to be inserted into the bottle.
- 4. The bottle is capped for the next **30** minutes. Both hydrogen gas and arsine gas will bubble from the solution. The arsine gas then reacts with mercuric bromide on the test strip to form mixed mercury halogens (such as AsH_2HgBr) that appear with a color change from white to yellow or brown.
- **5.** At the end of **30** minutes, the test strip is taken out of the bottle and compared to the color chart to determine the arsenic concentration. Because hydrogen may cause an explosion, and arsine gas is toxic, it is **highly** recommended the test be conducted in a well-ventilated* area away from fire and other sources of ignition⁹⁶.

This test kit was simple and easy to use while in remote locations in Nepal. No complications or problems occurred in its use under these challenging field conditions.

^{*} Study **by** Hussam et al showed the maximum arsine concentration in the immediate vicinity of some arsenic test kits can be more than **35** times the threshold limiting value (TLV) of **50** ppbv of arsenic from a single experiment with **100 ppb** of total arsenic in solution.

4.1.2 Graphite Furnace Atomic Adsorption Spectrometry (GFAAS)⁹⁷

GFAAS is one of the **USEPA** methods for measuring arsenic in drinking water. Atomic adsorption is based on the principle that atoms will absorb light at some characteristic wavelength. These wavelengths are related to the atomic structure of an element and the energy required for the promotion of its electrons from one quantum level to another. Therefore each element has its own characteristic wavelength. **A** wavelength of **193.7** nm is recommended for arsenic. The amount of light absorbed **by** an element at a certain wavelength can be correlated to the concentration of the element within the linear calibration range. The reliable calibration range is as low as *1-5* mg/L, and as high as 200 mg/L. Dilution of samples with higher arsenic concentrations may be required. Chemical interference may occur, but it can be ameliorated **by** the use of matrix modifiers. This method is not suitable for field use because of its large size and the sensitivity of the delicate components to transportation. The **GFAAS** requires electricity, but no refrigeration. **A GFAAS** instrument is available for use in the R.M. Parsons Laboratory at MIT.

Procedure

- **1.** Water samples from Nepal were first preserved **by** adding a drop of strong hydrochloric acid **(HCl)** per **10** mL of sample at the time of collection. The acidification prevents the precipitation of aqueous iron that would otherwise adsorb arsenic.
- 2. In the MIT laboratory, 1 mL of the water sample is carefully pipetted into a specially made plastic vial for **GFAAS** analysis. The vial is placed on the sampling dish. **A** set of standard arsenic solutions and matrix modifiers are also placed on the sampling dish. The dish has a capacity of **75** samples.
- **3. A** pre-programmed arsenic testing procedure is loaded on the **GFAAS** computer control. Information on the test samples are entered into the computer.
- 4. Click to start the test procedure. The instrument then automatically takes a small amount of the sample and volatilizes the arsenic atoms **by** intense heating. Once the atoms are excited, a monochrome lamp at **193.7** nm sends an optical beam through the headspace above the sample. The instrument measures the absorption and reports it as a peak focused around the **193.7** nm wavelength. The area under the peak is numerically integrated. The integrated results are displayed on the computer screen and on a printer printout.
- **5. By** comparing the area under the peak of the sample water with standard arsenic solutions, the concentration of the sample can be determined. Because of the variances associated with the instrument, a standard calibration curve was developed for every six to eight samples analyzed to maintain accurate measurements.

4.2 Arsenic Speciation

Description

Arsenic speciation refers to the proportion of arsenic **(III)** to arsenic (V) in water. Speciation is dependent on the groundwater chemistry. Speciation is an important measure in many respects. For example, arsenic (III) is much more toxic than arsenic **(V) 98.** Arsenic **(III)** is charged thus more mobile than neutral arsenic (V) under normal groundwater conditions. Furthermore, many current arsenic removal technologies can effectively remove arsenic (V), but not arsenic **(III).** The arsenic speciation test was performed using Bio-Rad Laboratory's **AGL-X8** ion exchange resin, 100-200 mesh, acetate form.

4.2.1 Bio-Rad Laboratory's AG1-X8 Ion-exchange Resin

The fundamental requirement for the arsenic speciation test is the separation of arsenic (III) from (V) in the water^{99, 100}. While there are many separation methods, the Bio-Rad Laboratory's **AG1-X8** ion exchange resin, 100-200 mesh, acetate form is based on ionexchange. Ion exchange processes are chemical reactions between ions in solution and ions in an insoluble solid phase. Depending on the ion-exchange media, certain aqueous ionic species are removed **by** the ion-exchange solid. Since electron-neutrality must be maintained, the solid releases replacement ions to the solution 101 . For the case of arsenic speciation at a **pH** range of 2 to **9,** arsenic **(III)** is an uncharged aqueous species while arsenic (V) is a negatively charged aqueous species. The **AG1-X8** resin is an ion exchange resin that only attracts and binds arsenic (V) in the solution. Virtually all arsenic (V) can be removed **by** the resin, leaving behind only arsenic **(1II)** in the solution. Then the arsenic **(III)** solution can be tested for arsenic content. **By** comparing this result with water sample containing both arsenic **(III)** and (V), the amount of arsenic (V) can be easily determined **by** a simple subtraction.

Procedure

- **1.** The **pH** of the water sample is checked with a **pH** meter. **If** the **pH** falls within 2- **9,** then no **pH** adjustment is required. Otherwise, **pH** adjustment using strong acid such as HCl or strong base such as NaOH is necessary.
- 2. For every 10mL of water sample collected, 0.2 **g** of resin is needed. In case that a balance is not available, the resin has a density of **0.75** g/mL. The desired resin weight can be determined **by** measuring the required volume in a graduated cylinder. When in doubt, use more resin than prescribed.
- **3.** For samples collected in mailer tubes that have a volume of about 14 mL, 0.3-0.4 **g** of resin was added to the sample. The tube is capped and shaken vigorously for *5* minutes. The shaking action allows efficient contact between the resin and arsenate species. Virtually all arsenate is adsorbed onto the ion-exchange resin within *5* minutes. Because arsenic (V) is very strongly adsorbed onto the resin, the vigorous shaking is not going to release the arsenic (V) back into the solution.
- 4. The content is then left undisturbed for *5* minutes. The resin settles to the bottom of the tube. The supernatant, containing only arsenic **(III),** is decanted into a clean mailer tube. This water sample is preserved **by** acidification with **HCl** as described before.
- *5.* Another mailer tube is used to collect a water sample containing total inorganic arsenic. This sample is preserved **by** acidification with **HCl** as well.
- **6.** The two sample sets (arsenic **(III)** only and total arsenic) are then brought back to MIT to be analyzed **by** the **GFAAS.**

4.3 PH

Description

pH is a measure of the acidity of a solution. It is a very important water quality parameter because in natural water, many chemical reactions associated with the formation, alteration, and dissolution of minerals are pH dependent¹⁰². This is especially true for carbonate minerals, as the equilibrium of carbon dioxide (CO_2) , bicarbonate (HCO₃²), and carbonate (CO₃²) are dictated by pH^{103} . Many biological activities, such as photosynthesis and respiration, are also influenced **by pH** through their respective abilities to decrease and increase the concentration of dissolved carbon dioxide¹⁰⁴. As a result, an observed change in **pH** may provide clues on the types and direction of reactions occurring in a solution. Another use of **pH** is that the protonation of many species are dependent on **pH,** such as arsenic **(III)** and (V). In addition to pH's significance in chemical reactions, **pH** is also a key aesthetic objective in drinking water. For example, the **US** National Secondary Drinking Water Regulation (NSDWR) sets a **pH** guideline of *6.5* to **8.5** 105. In this arsenic study, **pH** in the field site was tested with two methods: *Digi-Sense Digital pH/mV/ORP meter* **by** Cole Parmer, and *pH Test Strips* **by** Industrial Test Systems.

4.3.1 Cole Parmer Digi-Sense Digital pH/mV/ORP Meter EW-5938-50

The Cole Parmer **pH** meter measures **pH** using a combination electrode. The electrode incorporates both glass and reference electrodes in one body¹⁰⁶. The pH-sensitive part of the electrode is the thin glass bulb at the bottom of the electrode. When the electrode is immersed in a solution, the electric potential difference across the two electrodes is measured and converted to a pH reading¹⁰⁷.
Procedure

- **1.** The **pH** meter must be calibrated before use. Calibration solutions are prepared with **pH** buffer tablets. Three buffer solutions must be prepared, at **pH =** *4,* **pH** = **7,** and **pH = 10.**
- 2. The temperature of the buffer solutions are measured and entered into the **pH** meter.
- **3.** The electrode is dipped into the **pH = 7** buffer solution. The **pH** reading on the meter is set to **7.**
- 4. Then the electrode is dipped into the **pH =** 4 buffer solution. Adjust the slope screw on the meter to a **pH** reading of 4. Now the **pH** meter is ready for use, with accuracy optimized for solutions with **pH** between 4 to **7. If** the expected solution **pH** is above **7,** then the **pH = 10** buffer solution is used instead of the **pH =** *4* buffer solution. The slope screw is adjusted to a **pH** reading of **10.**
- *5.* Since **pH** is temperature dependent, it is important to enter the solution temperature into the meter in order to obtain the best readings.
- **6.** The **pH** electrode should be immersed in a strong KCl solution (use tap water if KCl is not available) when not in use.

4.3.2 Industrial Test Systems pH Test Strips 4800005

The test strips are very easy for field study use. The test strips come in a bottle of **50** strips. Printed on the bottle is a color chart corresponding to **pH** values from **6.0** to **9.0** at a *0.5* interval. Due to the proprietary nature of the product, the author was unsuccessful in obtaining details on the underlying chemistry of how the strips work.

Procedure

- **1. A** test strip is taken out from the bottle, and immersed in the sample solution for **10** seconds without any motion.
- 2. Remove and immediately match the color on the strip with the color chart. Color on the strip will start to fade after **15** seconds.

4.3 Oxidation-Reduction Potential (ORP)

Description

Oxidation and reduction (redox) reactions involve the transfer of electrons. Redox reactions are important because they mediate the behavior of many chemical constituents in drinking water¹⁰⁸. Arsenic is an excellent example. The speciation of arsenic is dependent on the redox potential of the solution. While redox potential is an exact chemical quantity, ORP is an approximate measurement of the redox potential of a solution. Digi-Sense Digital pH/mV/ORP meter **by** Cole Parmer was used in the field study to measure ORP.

4.3.1 Cole Parmer Digi-Sense Digital pH/mV/ORP Meter EW-5938-50

The measurement of ORP is based on the potential difference measured between a measuring electrode and a reference electrode. For the Cole Parmer meter, the reference electrode for ORP is identical to that for **pH** measurements. When the electrode is immersed in a solution containing a reversible chemical reaction system, a migration of electrons is established between the electrode and the system. The electron flow is measured and converted to an ORP reading¹⁰⁹.

Procedure

- **1.** ORP calibration is not perfromed on the meter because the procedue is not explained in the user manual.
- 2. Place the electrode into a sample solution. Select ORP mode on the meter. Record ORP value when the reading stablize.

4.4 Total Alkalinity

Description

Alkalinity of water is its acid-neutralizing capacity. Total alkalinity can be defined as the sum of all the titratable bases^{110, 111}. In groundwater, where the carbonate system is the dominant buffering system, alkalinity is primarily a function of **pH,** bicarbonate, and

carbonate*. Therefore, it can be taken as an indication of the concentration of these **12,113** constituents **12 .** In this arsenic study, the total alkalinity was measured with *Total Alkalinity Test Strips* **by** Industrial Test Systems.

4.4.1 Industrial Test Systems Total Alkalinity Test Strips

The test strips are very easy for field study use. The test strips come in a bottle of **50** strips. Printed on the bottle is a color chart corresponding to total alkalinity values of **0, 80,** 120, **180,** 240, and **360** mg/L. Due to the proprietary nature of the product, the author was unsuccessful in obtaining details on the underlying chemistry of how the strips work.

Procedure

- **1. A** test strip is taken out from the bottle, and immersed in the sample solution for **10** seconds without any motion.
- 2. Remove and immediately match the color on the strip with the color chart. Color on the strip will start to fade after **15** seconds.

4.5 Total Hardness as CaCO 3

Description

Hardness is a measure of the total amount of calcium and magnesium^{**} that has naturally leached into the water, as mg/L of calcium carbonate. **If** the subsurface contains abundant calcium and magnesium rich formations, such as calcite $(CaCO₃)$ and dolomites $(CaCO₃·MgCO₃)$, then the groundwater from that region may have high hardness. Hardness of more than 300 mg/L as CaCO₃ is consider excessive for public use because of high soap consumption and formation of scale in water fixtures¹¹⁴. Most people prefer

^{*} Total Alkalinity \approx -[H⁺] + [OH⁻] + [HCO₃⁻] + 2[CO₃²⁻] given that the carbonate system is the only significant buffer system

^{}** Although ions of iron, manganese, strontium, and aluminum also produce hardness, they are not present in significant quantities in natural water.

Source: Viessman, W., Hammer, M. "Water Supply and Pollution Control." 6th Edition. Addison-Wesley Longman, Inc.: Menlo Park, **CA. USA. 1998.**

water of less than 150 mg/L hardness¹¹⁵. Hardness was measured in this study with *Total Hardness (as calcium carbonate) Test Strips made by WaterWorksTM*

4.5.1 WaterWorksTM Total Hardness (as calcium carbonate) Test Strips

The test strips are very easy for field study use. The test strips come in a pack of **30** individually wrapped strips. Found in the pack is a color chart corresponding to hardness values of **0,** 40, **80,** 120, **180, 250,** and *425* mg/L as CaCO3. Due to the proprietary nature of the product, the author was again unsuccessful in obtaining details on the underlying chemistry of how the strips work.

Procedure

- **1. A** test strip is immersed in the sample solution for three seconds.
- 2. Remove and immediately match the color on the strip with the color chart. Color on the strip is stable for one minute.

4.6 Total Soluble Iron

Description

The two most common oxidation states for groundwater iron are $Fe(+II)$ and $Fe(+III)^{116}$. Soluble iron refers to mostly Fe(II) because it is **highly** soluble. The solubility of Fe(III) is very low. Iron is a very important water quality parameter in this arsenic study because iron hydroxide, $Fe(OH)_3$, is a strong adsorbent for arsenic¹¹⁷. Therefore, the presence of high iron can aid arsenic removal. Iron was measured in the field sites in Nepal using Chemetics Inc.'s CHEMets^R Iron Kit K-6010.

4.6.1 CHEMetsTM Iron Kit K-6010

The iron test kit is simple to use. The test kit contains **30** ampoules, a sample cup, a bottle of **A-6000** Activator Solution, and two sets of colour comparator. The first set of color comparator is for iron range of **0** to **1.0** mg/L at **0.1** mg/L interval. The second set is for iron range of **1** to **10** mg/L at **1** mg/L interval. The iron concentration quantification is based on colorimetry. Each molecule of soluble iron in water is chelated **by** three molecules of phenanthroline in the reagent ampoules to produce an orange-red complex¹¹⁸. The colored solution obeys Beer's law; the higher the iron concentration, the intenser the orange-red colour. Quantitative manganese concentration is determined **by** comparison to the standardized colour comparator.

Procedure

- **1.** Fill the sample cup to the 25mL mark with sample water
- 2. Place the reagent ampoule in the sample cup and snap the tip **by** pressing the glass ampoule against the side of the cup. Because the ampoule is vacummized, sample water would flow into the ampoule. The ampoule contains phenanthroline.
- **3.** Invert the ampoule several times to thoroughly mix its contents. Wait one minute for colour development
- 4. Compare the colour of the ampoule with the standard colour comparators to determine the iron concentration.

4.7 Manganese

Description

Manganese is an important water quality parameter because it can catalyze the oxidation of arsenic **(III)** to arsenic (V) **119,120.** Manganese was measured in the field sites in Nepal using Chemetics Inc.'s CHEMets^R Manganese Kit K-6502.

4.7.1 CHEMetsTM Manganese Kit K-6502

The manganese test kit is simple to use. The test kit contains **30** ampoules, a sample cup, a bottle of **A-6500** Activator Solution, and a color comparator. The kit has a detection range of **0** to 2 ppm of manganese. The kit is suitable for field study use. The manganese concentration quantification is based on colorimetry. Manganese in water reacts with sodium meta periodate in reagent ampoules, and the manganese activator solution to produce a purple color. The higher the manganese concentration, the intenser the purple colour. Quantitative manganese concentration is determined **by** comparison to the standardized colour comparator.

Procedure

- **1.** Fill the sample cup to the 5mL mark with sample water.
- 2. Five drops of the **A-6500** Manganese Activator Solution is added to the sample cup. The Activator Solution contains citric acid, sodium sulfate, sodium phosphate, and deionized water. Use the tip of a glass ampoule to mix the content of the sample cup.
- **3.** Place the ampoule in the sample cup and snap the tip **by** pressing the glass ampoule against the side of the cup. Because the ampoule is under negative pressure, sample water will flow into the ampoule.
- 4. Invert the ampoule several times to thoroughly mix its contents. Wait one minute for colour development
- **5.** Compare the colour of the ampoule with the standard colour comparator to determine the manganese concentration.

CHAPTER 5 ARSENIC REMOVAL TECHNOLOGY

The arsenic removal technology evaluated in this thesis is based on two proprietary products designed, manufactured, and distributed **by** a Massachusetts firm, Aquatic Treatment Systems, Inc¹²¹. The two products are *Poly-Benzyl Pyridinium Tri-Iodide* (BP/I₃) and Activated Alumina Manganese Oxide (A/M). The description of the BP/I₃ and A/M is explained first, followed **by** the apparatus setup. Refer to Appendix **A. 1, A.2** for the company profile, and the material safety data sheets **(MSDS)** for both media.

5.1 Poly-Benzvl Pyridinium Tri-lodide (BP/ 3)

Description

BP/13 media is designed to provide an on-demand* oxidant to convert arsenic **(III)** to arsenic (V). It is made from a water insoluble, porous polymer (poly-benzyl pyridinium). BP/13 media is a quaternary amine (pyridinium) divinylbenzene copolymer in the triiodide form, with 35-65% water¹²². Figure 9 shows the structure of the BP/I₃ media.

BP/13 is a dark brown, free flowing polymer bead with mean diameter of *568* micron **(28** 123 . It is easily handled in open air and has no odor. Fluid containing reducing species such as sulfide, sulfite, iron **(II)** and arsenic **(III)** are readily oxidized on-demand when contacted by $BP/I3^{124}$. The dark brown color of the media turns into a light tan color as its oxidizing capacity is exhausted. Therefore, the remaining useful life of the $BP/I₃$ can be easily determined.

^{*} On-demand means that the BP/I₃ oxidizes only when in contact with a reducing agent. BP/I3 remains inert when reducing agents are not present.

Figure **9** The Chemical Structure of Poly-Benzyl Pyridinium Tri-Iodide

 $BP/I₃$ is very stable, and does not deteriorate upon drying. Iodide on the media is released during oxidation. However, the iodide is subsequently re-captured on the benzyl pyridinium polymer. No detectable iodide should be found in the treated water. As a result, unlike ion-exchange, no by-products are found in the treated water 125 . BP/I₃ is produced in a manufacturing facility exclusively for Aquatic Treatment Systems, Inc. under supervision and quality control **by** the company. It is suitable for use as a food grade material.

Oxidation Mechanism

For arsenic **(III),** a contact time of as little of one second can completely oxidize it into arsenic $(V)^{126}$. The quick oxidation is due to iodide. In the presence of oxygen, the oxidation of arsenic **(III)** to (V) is catalyzed **by** iodide **127.** To achieve the one-second reaction rate, arsenic must be in contact with iodide. One of the best contact methods is in a flowing bed. Ideally, water containing arsenic should flow through a BP/13 bed. **If** the bed is not flowing, as in standing water, arsenic in the water can only reach the iodide by diffusion mass transfer. Diffusion can be a slow process¹²⁸

Oxidation Capacity

The capacity of **BP/13** in terms of equivalents of oxidizing power is **0.250** mol/L. This means every liter of BP/13 can accept up to *0.25* moles of electrons for oxidation

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purpose¹²⁹. The volume of water treated depends on the nature of the water and amount of dissolved reducing species. The following calculation shows the theoretical amount of arsenic **(III)** that can be oxidized with **1** L of BP/13-

Consider a 1 L solution containing pure water and **1000** pg of arsenic **(III).** The molecular weight of arsenic is 74.9 g/mol^{130} . Therefore 1000 μ g arsenic = 0.00001335 mol arsenic. In this solution, arsenic **(III)** is the only reducing agent. To convert arsenic (III) to (V), two moles of electrons are given up per mole of arsenic. As a result, 0.0000267 mol of electrons is released. Given that 1 L of BP/I₃ can accept up to 0.25 mol of electrons, therefore 1 L of BP/I₃ can treat up to about 9360 L of a 1000 μ g/L arsenic **(III)** solution. For a household that uses about **50** L of drinking water per day*, 9460 L represents about **187** days (or **6** months) of water use**.

In actual practice, the volume of water treated can be much lowered. This is because the presence of other reducing agents in the solution will deplete BP/I_3 as well. For example, groundwater may contain **10** mg/L of iron **(II).** The molecular weight of iron is **55.8** $g/mol¹³¹$. Therefore 10 mg of iron (II) = 0.000179 mole of iron. Each mole of iron (II) releases one mole of electron to oxidize to iron **(III).** As a result, **0.000179** mole of electrons is released. Given that $1 L$ of BP/I_3 can accept up to 0.25 mol of electrons, therefore 1 L of BP/I₃ can treat only 1400 L of a 10 mg/L iron (II) solution. For a household that uses about **50** L of drinking water per day, 1400 L represents about **28** days (or $\lt 1$ month) of water use^{***}. If there are other reducing agents in the water, such as sulfides and arsenic (III), then the volume treated by BP/I₃ can be even less. Removal of iron, sulfide, and other non-arsenic reducing agents **by** alternative methods is therefore recommended in order to prolong the life of $BP/I₃¹³²$.

^{*} Based on a household of **5** people, each use **10** L of drinking water for drinking, cooking and other purposes.

^{**} Based on 1000 µg/L of arsenic (III) in raw water and no other reducing agent.

^{***} Based on 10 mg/L of iron (II) in raw water and no other reducing agent.

Disinfection

In addition to its oxidizing capacity, BP/13 has also demonstrated ability to disinfect. Preliminary studies **by** the company showed positive disinfection results with *Pseudomonas Aeruonosa, Legionella,* total coliform, and *E-Coli133 .* However, the disinfecting properties of BP/I_3 will not be studied in this thesis.

Disinfection Mechanism

In a packed bed of BP/13, there are intra-granular pores on the media surface and intergranular pores between the particles. When raw water containing pathogens passes through a packed bed of BP/13, some of the pathogens are trapped in the inter-granular pores, while some are trapped in the intra-granular pores. Trapping in either type of pores is the primary mechanism to remove pathogens¹³⁴. If the water is not flowing, as in standing water, or if the BP/I_3 is used in a jar test, then pathogens are not trapped. In addition to the above mechanism, a second pathogen removal process is **by** disinfection/oxidation¹³⁵. The triiodide (I_3) is a disinfectant because of its strong oxidative capability. Pathogens can be inactivated **by** oxidation.

Cost

BP/I₃ is sold by Aquatic Treatment Systems, Inc. at a price of US\$1000 per cubic foot¹³⁶. It is currently available only in the mainland **U.S.** Tim Badger of Aquatic Treatment Systems, Inc. is currently in negotiating with a firm in Hyderabad, India to manufacture *BP/I3.* The selling price in the Indian-subcontinent will be at least *25%* less than the U.S.¹³⁷ However, Paul Smith of Aquatic Treatment Systes, Inc. does not think the Indian deal will be successful, casting doubt that BP/I₃ will be available in Asia in the near future¹³⁸.

5.2 Activated Alumina Manganese Oxide (AM)

Description

The A/M media is designed to adsorb arsenic (V). It also has a limited ability to oxidize arsenic (III) to (V). The chemical structure of the media is not available, but it is based on some proprietary modification of activated alumina.

The A/M media appears as very dark brown granules. It is easily handled in open air and has no odor¹³⁹. It requires no chemical additions to achieve complete arsenic removal. Temperature, **pH,** and contaminants affect little or no change in the adsorption performance of the A/M media¹⁴⁰. Because arsenic binds so strongly to the A/M media, arsenic will not leach off. The A/M media is not designed for regeneration. This media is approved by the USEPA under NSF 61 protocol as a safe non-hazardous solid waste¹⁴¹. Direct disposal can be a safe alternative to the generation of arsenic waste upon regeneration.

Adsorption Mechanism

Water containing arsenic **(III)** is first oxidized to arsenic (V). Then, arsenic (V) is permanently adsorbed on the A/M media **by** complexation reactions with the A/M media 142 . The new complex is stable. In order to improve the efficiency of arsenic removal, water containing arsenic should flow through a pack bed of A/M media. The water flowrate should not exceed **0.8** bed volumes per minute for arsenic concentration above **2500** pg/L, and should not exceed **1.0** bed volumes per minute for lower arsenic concentration to achieve optimal arsenic removal 143 .

Adsorption Capacity

The company claims each volume of A/M media can treat in excess of **80 000** volumes of water contaminated with 100 μ g/L arsenic. The presence of sulfate and/or fluoride does not significantly affect the arsenic removal results. The treated water should consistently remove arsenic to below detectable levels of $2 \mu g/L$. Figure 10 shows an experiment

done **by** the company to determine the oxidation and adsorption capacity of the A/M media.

Figure **10** Oxidizing and Adsorption Capacity of **A/M** Media

Cost

A/M is sold by Aquatic Treatment Systems, Inc. at a price of US\$100 per cubic foot¹⁴⁴. It is currently available only in the mainland **U.S.** Tim Badger of Aquatic Treatment Systems, Inc. is currently in negotiating with a firm in Hyderabad, India to manufacture BP/13 . The selling price in the Indian-subcontinent will be at least **25%** less than the U.S.¹⁴⁵ However, Paul Smith of Aquatic Treatment Systes, Inc. does not think the Indian deal will be successful, casting doubt that A/M will be available in Asia in the near $future¹⁴⁶$.

5.3 Apparatus Setup

5.3.1 A/M Only System

The A/M only system consisted of a clear plastic column, a spigot, a piece of felt, A/M media, and a metal retort stand with clamps. The column was 2 3/4 inch **(7** cm) in diameter and 94 cm tall. **A** spigot was connected to the bottom end of the column to control the water flow. **A** round piece of felt **0.5** cm thick and **7** cm in diameter sat inside the column just above the spigot. **800** mL of prepared AIM media was contained in the column above the felt. The purpose of the felt was to block any A/M from leaving the column. The A/M occupied about 34 cm height of column. The top **60** cm height of the column served as a water reservoir. The column was secured to the metal retort stand with 2 plastic clamps.

On account of its current production method, the A/M must be prepared before use. The raw A/M media contained impurities such as dirt that may foul the system and/or discolor the water. The A/M preparation was simple. **800** mL of A/M was first measured in a 1 L plastic beaker. Arsenic-free water was then added to the beaker. The content was stirred and the cloudy supernatant was discarded. The rinsing procedure was repeated for **¹⁰** times, using about **10** L of water in total. Now the A/M was ready for use.

The cost of **800** mL of A/M media was about **US\$2.83** based on **US\$100** per cubic foot*.

5.3.2 BP/13 & A/M System

The BP/13 **&** A/M system setup was similar to the A/M only system except that an extra 400 mL of BP/13 was used. The BP/13 **&** A/M system consisted of a clear plastic column, a spigot, a piece of felt, A/M media, BP/13 media, and a metal retort stand with clamps. The column was 2 3/4 inch **(7** cm) in diameter and **100** cm in height. **A** spigot was connected to the bottom end of the column to control the water flow. **A** round piece of felt **0.5** cm thick and **6** cm in diameter sat inside the column just above the spigot. **800**

 $*$ 1 cubic feet = about 28.3 L

mL of prepared A/M was contained in the column above the felt. 400 mL of BP/I₃ was placed directly on top of the A/M. The purpose of the felt was to block any A/M from escaping the system. The A/M and the BP/13 occupied about **33** cm and 20 cm height of column respectively. The top 47 cm height of the column served as a water reservoir. The column was secured to the metal retort stand with 2 plastic clamps. See Figure **¹¹** for a schematic diagram showing the setup of the A/M only and the BP/13 **&** A/M systems. Figure 12 shows a picture of the actual BP/13 **&** A/M systems.

Figure **11** Schematic Diagram Showing the Setup of the A/M Only and the BP/13 **&** A/M Treatment Systems.

Figure 12 The Actual Column Used in this Arsenic Study

The preparation of A/M was the same as was previously discussed. The BP/I₃ must also be prepared before use in order to remove impurities. The preparation procedure for BP/I₃ was identical to A/M.

The cost of 400 mL of A/M media was about **US\$14.13** based on **US\$100** per cubic foot^{*}.

 $*$ 1 cubic feet = about 28.3 L

CHAPTER 6 EXPERIMENT SETUP AND PROCEDURES

There are three main part of the field study. They are

- **"** Arsenic Removal Technology Evaluation
- Arsenic Speciation of Tubewell Water
- **"** Arsenic Oxidation Kinetics

6.1 Arsenic Removal Technology Evaluation

Two treatment systems were setup Parasi, one with A/M media only, the other with BP/13 **&** A/M media. The technical performance of both systems were measured and compared.

6.1.1 Apparatus Setup

A/M only system

Equipment needed to evaluate the A/M only arsenic removal system included a pH/ORP meter, a thermometer, some mailer tubes, some sampling bottles of at least **100** mL volume, and the A/M only treatment system column as described in Chapter **5.**

BP/13 **&** A/M system

Equipments needed to evaluate the BP/13 **&** A/M arsenic removal system included a pH/ORP meter, a thermometer, some mailer tubes, some sampling bottles of at least **100** mL volume, an the BP/13 **&** A/M treatment system column as described in Chapter **5.**

6.1.2 Field Experiment

A/M only system

The A/M only system was tested four times in the town of Parasi in the Nawalparasi District. Refer to Figure **13** for a map showing the different districts of Nepal.

Nawalparasi and Rupandehi are in central south. Parasi is a town in the Nawalparasi District. This is the town where the author stayed from Jan **6,** 2002 to Jan 14, 2002. The first test was conducted on Jan **10,** 2002 with raw water from the tubewell in the backyard of the author's residence. The second test was conducted on Jan **11,** 2002 with raw water from a tubewell in front of a restaurant in the town center. The third test was conducted on Jan **13,** 2002 with raw water from a tubewell in front of a shop across the street from the author's residence. The fourth test was conducted on Jan 14, 2002 with raw water from the backyard tubewell again. See Appendix B for a map showing these well locations. In all the above tests, water quality parameters for the raw water were measured including **pH,** temperature, oxidation-reduction-potential (ORP), total soluble iron, total arsenic, arsenic **(III),** manganese, hardness, and alkalinity. Then the column was flushed with three litres of raw water before a treated water sample was taken. The flushing was intended to purge any water remaining from prior tests (i.e. memory effect). This ensures that the treated water sample comes from freshly input raw water. The treated water was also analyzed for the above water quality parameters. In addition to these water quality tests, two to three flowrate measurements were done on each run using a **100** mL plastic graduated cylinder.

BP/13 **&** AIM system

The A/M only system and the BP/13 **&** A/M system were tested simultaneously on the four occasions in the Parasi. In addition to these four Parasi tests, the BP/13 **&** A/M system was tested at five more locations, while the author was staying in the city of Butwal in Rupandehi District from Jan *15,* 2002 to Jan 22, 2002. The fifth test was conducted on Jan **16,** 2002 with raw water from a well in the village of Madangram in the Devdaha village development committee **(VDC)** in the Rupandehi district. The sixth test was conducted on Jan **16,** 2002 in the village of Madangram as well. The seventh test was conducted on Jan **17,** 2002 in the village of Bangali in the Devdaha VDCof Rupandehi District. The eighth test was conducted on Jan 20, 2002 in the village of Kirtipur-1 in the Sunwal **VDC** of Nawalparasi District. The ninth test was conducted on Jan 21, 2002 in the village of Khakaribari in the Sunwal **VDC.** These wells were chosen because of their

known high arsenic concentrations. The test procedure for the A/M **&** BP/13 system was identical to the A/M only system. Table **1** summarizes all the test dates and locations.

Run	Date	Village/	VDC	District	Test on	Test on
#		Town			A/M only	$BP/I_3 \& A/M$
$\mathbf{1}$	Jan 10, 02	Parasi	Parasi	Nawalparasi	Yes	Yes
2	Jan 11,02	Parasi	Parasi	Nawalparasi	Yes	Yes
3	Jan 13, 02	Parasi	Parasi	Nawalparasi	Yes	Yes
$\overline{4}$	Jan 14, 02	Parasi	Parasi	Nawalparasi	Yes	Yes
5	Jan 16, 02	Madangram	Devdaha	Rupandehi	No	Yes
6	Jan 16, 02	Madangram	Devdaha	Rupandehi	N _o	Yes
$\overline{7}$	Jan 17, 02	Bangali	Devdaha	Rupandehi	N ₀	Yes
8	Jan 20, 02	Kirtipur-1	Sunwal	Nawalparasi	N _o	Yes
9	Jan 21, 02	Khakaribari	Sunwal	Nawalparasi	N _o	Yes

Table 1 Summary of Arsenic Removal Technology Test Dates and Locations

6.2 Arsenic Speciation of Tubewell Water

Raw water speciation refers to the determination of the ratio of arsenic **(III)** to total arsenic. This test was conducted at **37** tubewells throughout Rupandehi and Nawalparasi Districts between Jan **16,** 2002 and Jan 21, 2002.

6.2.1 Apparatus Setup

Equipment needed to carry out raw water speciation tests included a pH/ORP meter, a thermometer, some mailer tubes, some sampling bottles of at least **100** mL volume, strong-ion exchange resin.

6.2.2 Field Experiment

At each of the wells, the **pH** and ORP of the well water was measured. **A** water sample containing total arsenic and a water sample containing arsenic **(III)** were collected in mailer tubes. The thermometer was used to adjust the **pH** and ORP readings as they were temperature dependent. Refer to Chapter 4 **-** Analytical Methods for detailed description on how to obtain a water sample of arsenic **(III)** only. After on-site tests and sample collections, detailed records on each of the visited wells were gathered at the office of **DIDC** (formerly **FINNIDA).** Refer to Appendix **G.** 1 and **G.2** for the records. Their database contained the well location (i.e. village/town, **VDC,** district), well number, contact person, well type, number of household sharing the well, actual number of users, well depth, well age, agencies who constructed the well, previous arsenic records, number of cases of arsenic health effects, and microbiological safety. Table 2 shows the test dates and locations of these **37** wells.

6.3 Arsenic Oxidation Kinetics

The objective of the kinetics experiment was to determine the rate at which arsenic (III) oxidizes to arsenic (V) under natural aeration in an undisturbed environment.

6.3.1 Apparatus Setup

The apparatus required included a **1** L plastic beaker, a pH/ORP meter, a thermometer, and a clock.

6.3.2 Field Experiment

On Jan 12, 2002, the 1 L plastic beaker was filled with raw water from the backyard well in Parasi. The beaker was left undisturbed and uncovered for the next three days. The water temperature, **pH,** and ORP were measured at **18** different intervals over an approximately three days time period after the initial water collection. Arsenic speciation tests were performed at all of these time intervals. Table **3** shows the time and date for these **18** data points.

Data	Date	Time	Hours since initial
point #			water collection
1	Jan 12,02	12 noon	0
$\overline{2}$	Jan 12, 02	12:15 pm	0.25
3	Jan 12, 02	12:30 pm	0.5
$\overline{4}$	Jan 12, 02	12:45 pm	0.75
5	Jan 12, 02	$1:00$ pm	1.0
6	Jan 12, 02	$1:30$ pm	1.5
7	Jan 12, 02	$2:00$ pm	2.0
8	Jan 12, 02	$2:30$ pm	2.5
9	Jan 12, 02	3:30 pm	3.5

Table **3** Summary of Oxidation Kinetics Tests Dates and Time

CHAPTER 7 RESULTS AND DISCUSSIONS

In this section, the results for the arsenic removal technology evaluation will be discussed first, followed **by** the arsenic speciation of tubewell results and then the arsenic oxidation kinetics results.

7.1 Arsenic Removal Technology Evalution

The A/M only system was tested on four occasions in Nepal. The BP/I₃ & A/M system was tested at five additional locations. For each of the nine runs, the following items were measured:

- Arsenic removal
- * **pH**
- **"** Oxidation-Reduction Potential (ORP)
- **"** Total alkalinity
- Total hardness as CaCO₃
- Total soluble iron
- Manganese
- **"** Flowrate

Detail results for each of these individual tests are in Appendix F.

7.1.1 Arsenic Removal Results

Table 4 shows a summary of the arsenic removal results. For the nine runs, the raw water total arsenic ranged from 147 μ g/L to 863 μ g/L. The well with 863 μ g/L total arsenic was the highest arsenic concentration in DIDC's (formerly FINNIDA's) **1508** well data set. The percentage of arsenic **(III)** ranged from **73%** to **100%. All** treated water from the A/M only system contained non-detect level of total arsenic (i.e. $\lt 5 \mu g/L$) from the field test kit. Therefore the A/M was **97-99%** effective, and consistently reduce total

arsenic to less than the WHO standard of 10 μ g/L. For the BP/I₃ & A/M system, all treated water contains non-detect level of total arsenic as well.

Run	Raw water	As(III)	A/M only	$\%$	$BP/I_3 \& A/M$	$\%$
#	total	as $%$ of	treatment	arsenic	treatment total	arsenic
	arsenic	total	total arsenic	removal	arsenic $(\mu g/L)$	removal
	$(\mu g/L)$	arsenic	$(\mu g/L)$			
$\mathbf{1}$	242	91	< 5	> 98	≤ 5	> 98
$\overline{2}$	152	89	< 5	> 97	< 5	> 97
3	337	91	$<$ 5	> 99	< 5	> 99
$\overline{4}$	323	73	< 5	> 98	< 5	> 98
5	863	94	Not tested	N/A	< 5	> 99
6	328	98	Not tested	N/A	≤ 5	> 98
$\overline{7}$	149	77	Not tested	N/A	< 5	> 97
8	328	81	Not tested	N/A	< 5	> 98
9	147	100	Not tested	N/A	≤ 5	> 97

Table 4 Summary of Arsenic Removal Results

Discussion of results

By comparing the arsenic removal results between the A/M only system and the BP/13 **&** A/M systems, it appeared that the A/M **by** itself was sufficient for arsenic removal. BP/13 was not required. Both systems reduced total arsenic to less than the WHO standard for all runs. However, this was a short term study. The A/M system was tested for only 4 runs with an approximately **15-25** L of water treated; the BP/13 **&** A/M system was tested for only **9** runs with an approximately 40-50 L of water treated. When the A/M was subjected to a long-term use, its oxidizing capacity might soon be exhausted, leading to incomplete arsenic **(III)** to arsenic (V) oxidation. As a result, the arsenic removal effectiveness may be lowered. In that case, the use of BP/I_3 , a strong oxidizing agent, may prove to be useful in improving the removal effectiveness. **A** long-term study was recommended.

Another observation is that the raw water arsenic speciation does not seem to affect the removal effectiveness of both treatment systems. Two hypotheses could be deduced from this. First, both A/M and BP/13 could **100%** oxidize arsenic **(III)** into arsenic (V). The A/M then adsorbed all arsenic (V). The second hypothesis was that the A/M could **100%** adsorb all forms of arsenic regardless of speciation. The exact explanation was unknown. It might be a combination of these two hypotheses. In order to better understand the mechanism, it was suggested that an A/M only system be tested with an arsenite solution of sufficiently high concentration such that the treated water had appreciable amount of arsenic. The treated water was then speciation tested. **If** all the arsenic is in the **(+III)** oxidation state, then it implied that A/M cannot **100%** oxidize arsenic **(III)** into arsenic (V). **If** all the arsenic was in the **+5** state, then **it** implied that A/M could **100%** oxidize arsenic **(III)** into arsenic (V), but not able to adsorb all arsenic (V). **A** similar test could also be performed on the BP/13 **&** A/M system to determine the oxidative power of BP/13, and the adsorption effectiveness of A/M.

It was also observed that the top two centimeters of the BP/I_3 changed color from deep brown to light yellow after the nine runs. This was an indication that the oxidizing capacity of the top two centimeters has been exhausted. The capacity was used up so quickly not only because of the arsenic, but also because of hydrogen sulfide 14 . Hydrogen sulfide is a reducing agent. It was oxidized **by** BP/13 into sulfate. Hydrogen sulfide was found in over 40% of the tubewells in the Terai¹⁴⁸. Arsenic oxidation alone could not justify the quick exhaustion of oxidizing capacity¹⁴⁹. It was recommended that oxidizing capacities of both the BP/13 and A/M media be further studied.

Discussion of errors

Although the **GFAAS** should accurately detect arsenic to the **1 pg/L** level, it was never observed in the instrument used in this arsenic study. The detection limit determined in the author's analysis of arsenic sample was **10** ig/L. **All** arsenic reading from **0** pg/L to about **10 pg/L** were questionable. For example, when the instrument was calibrated, the standard 0 μ g/L arsenic solution can give an arsenic reading of as high as 10 μ g/L. For a

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reading between 10 μ g/L to 20 μ g/L, the accuracy is better; however, confirmation with a secondary test method (e.g. test kit) was advisable. The most accurate readings were for arsenic concentration ranging from about 20 μ g/L to about 75 μ g/L. The calibration was approximately linear for this arsenic range. For arsenic readings above $75 \mu g/L$, the accuracy drops. The reason was that the calibration curve is non-linear at this region. Therefore, all water samples above **75** pig/L should be diluted to between 20 and **75 pig/L** in order to get more accurate readings. For this arsenic study, all water samples above **60** ptg/L were diluted and retested. **All** water samples below 20 **ig/L** were regarded unreliable unless confirmed **by** the test kit, which had a detection limit of **5 pg/L. All** water samples between 20 and 60 μ g/L were considered at their face value. Refer to Appendix **E.** 1 for calibration curves and **E.2** for interpolated results.

Fluctuation in lamp intensity resulting in fluctuation in the arsenic atomic signal from the **GFAAS** with time was another concern. To illustrate the level of fluctuation, the eight average-calibration curves obtained during the test on March 12, 2002 are shown in Figure 14. **All** of these curves had a different slope and/or intercept. For example, consider a water sample with peak area of **0.100.** The top calibration curve translated this peak area to an arsenic concentration of about **55 tg/L,** while the bottom calibration curve translated the same peak area to an arsenic concentration of about 70 μ g/L. The difference between 55 μ g/L and 70 μ g/L was approximately 20-30%. To reduce errors associated with signal fluctuation in this arsenic study, a new calibration curve was plotted for every **6** to **10** sample tested. These **6** to **10** samples were calibrated with the average of the calibration curves before and after (i.e. average-calibration curve).

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Figure 14 Fluctuation in **8** Calibration Curves obtained on March 12, 2002.

7.1.2 pH Results

Table **5** shows a summary of the **pH** results. For the nine runs, the raw water **pH** ranged from 6.49 to **7.60.** The treated water for A/M only system ranged from **6.60** to *7.05;* BP/13 **&** A/M system ranged from **5.89** to **7.03. A pH** drop was observed in all runs, except run **#1.**

Run	Raw water	A/M only	Change in pH	BP/I3 & A/M	Change in pH
#	pH	treated water	after treatment	treated water	after treatment
	7.00	7.05	$+0.05$	7.03	$+0.03$
\cdot 2	6.35	Not measured	N/A	Not measured	N/A
3	6.90	6.60	-0.30	6.16	-0.74

Table *5* Summary of **pH** Results

Discussion of results

It was found that both the A/M and the BP/I_3 media increased the acidity of water. For example, for the A/M only system, **pH** drop was observed in runs **# 3** and 4 except for a very slight **pH** increase in run **#1.** This indicated that the A/M media gave acidity. **By** comparing the **pH** drop between the A/M only system and the BP/13 **&** A/M system for runs **# 1, 3,** and 4, it was concluded that the BP/13 was acidic as well. These observations agreed with the explanation given by Paul Smith¹⁵⁰, one of the inventors of this media. Paul Smith explained that both A/M and BP/I₃ are acidic in nature. The acidic nature was related to the medias' proprietary chemical structure. Both media could be prepared in the manufacturing process to be pre-treated with a **pH** buffer to eliminate the **pH** drop. The media used in this arsenic study was not **pH** pre-treated.

One implication of the acidic nature of both A/M and BP/I₃ was that the A/M and BP/I₃ may need to be **pH** pre-treated. In the case of runs **# 3** to **8,** the treated water pHs were lower than the **US** National Secondary Drinking Water Regulation (NSDWR) of **6.5** to **8.5. Of** the over **50** wells visited for this arsenic study, none had **pH** greater than **8.** Therefore, the media pre-treatment to eliminate **pH** drop was preferred in many locations of Rupendehi and Nawalparasi water conditions.

Discussion of errors

The **pH** meter was calibrated daily in the morning. It was linearly calibrated with prepared **pH =** 4 and **pH = 7** buffer solutions. These buffer solutions were prepared with Cole Parmer **pH** buffer tablets. Sometimes, the **pH** meter was recalibrated in the afternoon. It was observed that the **pH** reading could be off **by** as much as 0.2 **pH** units.

In general, the fluctuation was less than **0.1 pH** unit. Therefore, **pH** fluctuation was not a significant issue.

Another potential problem was the slow **pH** meter response time. It could be due to a variety of reasons such as drying of the **pH** probe tip, or aged probe, or low battery. The issue of the **pH** probe drying up was prevented **by** inserting the probe into a bottle of strong KCl solution whenever the probe is not in use. The problem of an aged probe created some problems in the field*. The **pH** meter started to give erroneous results starting on Jan 20, 2002. As a result, no further **pH** reading from the Cole Palmer **pH** meter was possible. Subsequently, test strips were used to semi-quantitatively determine the **pH** level of water. The test strips color chart could be difficult to compare **pH** values. The **pH** intervals were every *0.5* **pH** units. Therefore, the **pH** measurements starting on Jan 20, 2002 were not very accurate.

7.1.3 Oxidation-Reduction Potential (ORP) Results

Table **6** shows a summary of the ORP results. For the nine runs, the raw water ORP ranged from **-78** mV to **-37** mV. The treated water ORP for A/M only system ranged from -62 to -33 mV; BP/I₃ & A/M system ranged from -47 mV to -3 mV. It was observed the treated water was, in general, more oxidizing than the raw water.

^{*} Note: Although a new probe has been purchased prior to this field study, it did not manage to arrive at the author's field site.

Run#	Raw	A/M only	Change in ORP	BP/I_3 & A/M	Change in	Apparent
	water	treated water	after treatment	treated water	ORP after	BP/I_3
	(mV)	(mV)	(mV)	(mV)	treatment	contribution
					(mV)	(mV)
$\mathbf{1}$	-45	-62	-17	-47	-2	$+15$
$\overline{2}$	-37	Not measured	N/A	Not measured	N/A	N/A
3	-61	-45	$+16$	-13	$+48$	$+32$
4	-78	-33	$+45$	-6	$+72$	$+27$
5	-60	Not tested	N/A	-3	$+57$	N/A
6	-45	Not tested	N/A	-12	$+33$	N/A
$\overline{7}$	-40	Not tested	N/A	-10	$+30$	N/A
8	Meter	Not tested	N/A	Meter broken	N/A	N/A
	broken					
9	Meter	Not tested	N/A	Meter broken	N/A	N/A
	broken					

Table **6** Summary of ORP Results

Discussion of results

Because A/M and the BP/I₃ media were both oxidizing agents, it was logical that the treated water is more oxidizing than the raw water, except for the anomaly in run **#1.** The anomaly in run **#1** could be due to the fact that the A/M media was not fully functional initially. It took some time (or some volume of water) for the A/M to begin its oxidizing role. This delay in the A/M to fully function was analogous to the process whereby a machine that needed to "warm up". **A** comparison between the A/M only system and the BP/13 **&** A/M system showed that the BP/13 further contributed to water oxidation. For runs **# 1, 3,** and 4, the ORP of the BP/13 **&** A/M system was higher than the A/M only system. The apparent contribution of BP/13 was **+15** mV for run **#1, +32** mV for run #2, and $+ 27$ mV for run #4. The apparent BP/I₃ contribution was an estimate only. No actual measurement of ORP was performed at the interface of the $BP/I₃$ and A/M media of the BP/13 **&** A/M system. The apparent BP/13 contribution assumed that the A/M media in both columns behaved identically.

It was noted that for different runs, the change in ORP was also different. One reason was that the water chemistry was different at different wells, thus requiring different levels of oxidants to achieve the same change in ORP. Another reason was that the oxidizing rate of the A/M and BP/I_3 were dependent on water chemistry, flowrate, temperature, and other factors such as the "warm-up" effect discussed above. Therefore the amount of oxidation varied for each run. Further investigation into the oxidizing rate could be done, but was not necessary. It was because the A/M and $BP/I₃$ had already achieved their main objective of arsenic removal. How much the ORP changed does not matter. The ORP of drinking water is not regulated, nor is a health concern¹⁵².

Discussion of errors

Unlike **pH,** the ORP reading on the meter was not calibrated for this arsenic study. Because the meter was not calibrated to a known reference state, all readings are relative. Therefore, an ORP reading had no meaning **by** itself, except when compared to another reading. One implication was that the same pH/ORP probe should be used throughout the entire field experiment. **If** not, the ORP readings using one probe might not be compatible with readings using another probe.

One problem with the pH/ORP probe was the slow response time. The causes and corresponding remediation techniques are similar to the slow **pH** response, thus not discussed here. As mentioned earlier, the pH/ORP meter started to give erroneous results starting on Jan 20, 2002. As a result, no further ORP readings were possible.

7.1.4 Total Alkalinity Results

For the A/M only system, the total alkalinity between the raw and the treated water was hardly changed. For the BP/13 **&** A/M, the total alkalinity change ranged from slightly lowered to significantly lowered. Refer to Table **7** for a summary for the alkalinity results.

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Run	Raw Water	A/M only	Change in Alk	$BP/I_3 \& A/M$	Change in Alk after
#	(mg/L)	treated water	after treatment	treated water	treatment
		(mg/L)		(mg/L)	
$\mathbf{1}$	180-240	120-180	Slightly lowered	$0 - 80$	Moderately lowered
2	120-180	Not measured	N/A	Not measured	N/A
3	180-240	180-240	Not at all	$0 - 80$	Moderately lowered
$\boldsymbol{4}$	80	80	Not al all	$0 - 80$	Slightly lowered
5	240-360	Not tested	N/A	120-180	Moderately lowered
6	240-360	Not tested	N/A	80-120	Significantly lowered
7	Not measured	Not tested	N/A	Not measured	N/A
8	240	Not tested	N/A	120	Moderately lowered
9	240-360	Not tested	N/A	120	Significantly lowered

Table **7** Summary of Total Alkalinity Results

Discussion of results

Since alkalinity was a measure of water's acid-neutralizing capacity, the drop in alkalinity indicated acid was introduced to the water. This observation was in agreement with fact that A/M and BP/I₃ gives acidity. In an attempt to neutralize the additional acidity, alkalinity was consumed, thus lowered. For the A/M only system, the drop in alkalinity was minimal. For the BP/13 system, the drop in alkalinity was moderate. It was because the BP/I_3 gives acidity in addition to the A/M. On the other hand, if both media were pre-treated with a **pH** buffer, then alkalinity might not drop.

Other factors such as the presence of other buffering systems, and addition/removal of strong cations/anions, might affect alkalinity as well¹⁵³. However, these factors were not measured in this study.

The drop in alkalinity was not a concern from the perspective of providing a safe, drinkable water. Alkalinity was not regulated and has no health consequences.

Discussion of errors

The total alkalinity test results were based on color matching between the test strips and a color chart. Frequently, none of the colors on the color chart matched with the color on the test strip. In those cases, the average perceptions among two or three of the author's teammates were taken as the alkalinity reading. Often there was wide variability among peoples' perception; therefore, the accuracy of the total alkalinity results was low. It was better to regard total alkalinity results as more of qualitative than quantitative measurements. Nevertheless, total alkalinity readings were far from purely random. For those runs with moderate to significant drops in alkalinity, it could be clearly seen that there were color change before and after treatment.

7.1.5 Hardness Results

For most of the runs, total hardness was unchanged **by** the A/M or BP/13 treatment. Total hardness was slightly to moderately lowered in other cases. Refer to Table **8** for a summary of the hardness results.

Run	Raw Water	A/M only treated	Change in	$BP/I_3 \& A/M$	Change in
#	Hardness (mg/L)	water (mg/L) as	Hardness after	treated water	Hardness after
	as $CaCO3$)	$CaCO3$)	treatment	$(mg/L \text{ as } CaCO3)$	treatment
$\mathbf{1}$	250	180	Slightly lowered	250	No change
$\overline{2}$	250-425	Not measured	N/A	Not measured	N/A
3	250	120-180	Moderately	180-250	Slightly lowered
			lowered		
4	180-250	180-250	No change	180-250	No change
$\mathbf{5}$	80-120	Not tested	N/A	80-120	No change
6	120-180	Not tested	N/A	120-180	No change
7	Not measured	Not tested	N/A	Not measured	N/A
8	180-250	Not tested	N/A	180-250	No change
9	Not measured	Not tested	N/A	Not measured	N/A

Table **8** Summary of Total Hardness Results

Discussion of results

It was concluded that A/M and BP/13 had no or minimal effect on total hardness. The observed lowering of hardness in a few runs might be attributed to errors in measuring hardness. This was explained in more detail in the following paragraph.

Discussion of errors

Because total hardness was determined in the field in Nepal using test strips similar in nature to the alkalinity test strips, many of the problems associated with the alkalinity test strips applied to the total hardness test. For example, the color on the test strip did not match any of the colors on the chart. Therefore, hardness measurements were again more qualitative than quantitative measurements. **All** of the above hardness results were questionable unless they are reconfirmed with more accurate tests.

7.1.6 Total Soluble Iron Results

Due to an insufficient number of available iron test ampoules, total soluble iron was measured only for runs **#1** and **3.** For both cases, the raw water contained **2.5-3.5** mg/L of soluble iron, while the treated water contained non-detect level **(<0.1** mg/L) of soluble iron. Refer to Table **9** for a summary of total soluble iron results.

Run	Raw water	A/M only treated	$BP/I_3 \& A/M$ treated
#	(mg/L)	water (mg/L)	water (mg/L)
	2.5	<0.1	< 0.1
	3.5	<0.1	< 0.1

Table **9** Summary of Total Soluble Iron Results

Discussion of results

The two most common oxidation states for groundwater iron were Fe(+II) and Fe($+III$)¹⁵⁴. Soluble iron refered mostly to Fe(II) because it is highly soluble. The solubility of Fe(III) was very low. The reduction of soluble iron could be explained **by** two mechanisms. In the first mechanism, Fe(II) was oxidized into Fe(III). The oxidation could be accomplished **by** exposure to the oxygen rich atmosphere, or **by** contact with BP/I_3 and/or A/M. Fe(III) then formed a precipitate, Fe(OH)₃, and was trapped within the BP/13 and A/M media bed **by** physical filtration. The second iron removal mechanism was its adsorption to the A/M media. Paul Smith¹⁵⁵ confirmed that the A/M media has an ability to adsorb iron. One drawback of the A/M's ability to adsorb iron was that adsorption sites occupied **by** iron could not be used to adsorb arsenic. Therefore, high iron levels in the raw water could seriously reduce A/M's capacity to remove arsenic.

On the other hand, iron adsorption **by** the A/M media had its benefits. Currently in the **U.S.** and Nepal, iron concentration in drinking water was recommended to be less than or equal to **0.3** mg/L. The water in runs **#1** and **3** were about **10** times the recommended iron level. Although high iron was not a health concern, high iron gave an orange-red color to the water. Water containers and cooking utensils could be stained. High iron also gave taste and odor problems. As a result, people might incorrectly perceive high iron water to be unsafe for drinking. In addition, numerous studies have shown that iron is an excellent absorbent for arsenic^{156, 157, 158, 159, 160}. Iron adsorbed on the A/M media could in turn adsorb arsenic. Therefore, the loss in A/M's capacity to remove arsenic **¹⁶¹** could be compensated **by** the gain in iron's capacity to adsorb arsenic **.** In contrast, if iron was not adsorbed **by** the A/M media, but rather leached into the treated water; then arsenic might be found together with the iron. Arsenic was thus not removed.

Discussion of errors

The determination of iron concentration was **by** color comparison. It was sometimes difficult to match the colour of the iron test ampoules to the standard iron ampoules. Different shades of orange could be hard to distinguish, especially for iron concentrations above 1 mg/L. The color could also be affected **by** sunlight intensity and ampoule orientation. Therefore, the raw water iron measurements were not very accurate.

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However, the iron concentration of the treated water should be more reliable. It was because the colour for an iron-free sample has a distinctive colorless color.

7.1.7 Manganese Results

In testing for the presence of manganese in four runs, manganese was found to be below detection limit **(0.1** mg/L) in either the raw water or the treated samples. Refer to Table **10** for a summery of the manganese results.

Run	Raw water	A/M only treated	BP/I3 & A/M treated
#	(mg/L)	water (mg/L)	water (mg/L)
	< 0.1	< 0.1	< 0.1
3	< 0.1	< 0.1	< 0.1
4	< 0.1	< 0.1	< 0.1
Q	< 0.1	Not tested	< 0.1

Table **10** Summary of Manganese Results

Discussion of results

Because no manganese was found in the raw water, the effect of manganese on the treatment systems is unknown.

Discussion of errors

The manganese results should be accurate. It was because the color for a manganese-free sample has a distinctive colorless color compared to manganese positive sample.

7.1.8 Flow Rate Results

Table **11** shows the summary for flow rate results. The columns with multiple entries indicate multiple flow rate measurements were taken. **All** flow rate measurements were taken when the water level in the column was at maximum (i.e. water reservoir

completely filled). For the A/M only system, the flow rate increased with each subsequent run. For the BP/13 **&** A/M system, the flow rate gradually decreased with each subsequent run.

Run	A/M only	A/M only	$BP/I_3 \& A/M$	$BP/I_3 \& A/M$
#	flowrate (mL/min)	flowrate (L/hr)	flowrate (mL/min)	flowrate (L/hr)
1	228	13.7	324	19.4
2	240	14.4	444	26.6
3	384	23.0	378, 414	22.7, 24.8
$\overline{4}$	408, 408	24.5, 24.5	396, 372	23.8, 22.3
5	Not tested	Not tested	360, 372	21.6, 22.3
6	Not tested	Not tested	348, 371	20.9, 22.3
τ	Not tested	Not tested	348, 354, 384	20.9, 21.2, 23.0
8	Not tested	Not tested	354, 336	21.2, 20.2
9	Not tested	Not tested	324, 342, 324	19.4, 20.5, 19.4
avg	334	20.0	364	21.8

Table **11** Summary of Flow Rate Results

Discussion of results

In general, a flow rate of about 20 Uhr was more than sufficient for an average household need of about **20-30** Uday*. However, the flowrate was **highly** dependent on the make of the media material, media porosity, trapped air bubbles, the column diameter and length, the column orientation, and other factors. In another column configuration, the flow rate would be completely different. Therefore the flow rate results were not readily transferable to another column configuration unless detailed hydrodynamics experiments were performed to understand the characteristics of the media. The use of Darcy's law¹⁶² or Ergun's equation¹⁶³ for flow through a porous bed might prove helpful.

^{*} Based on a family of **5,** with drinking/cooking water requirement of 4 L/person/day

The exact reason for the increasing flow rate for the A/M media was unknown. One explanation was that the media bed was loosely compacted for the first two runs. As a result, air bubbles were present in the media bed, which restricted water flow. **By** run **# 3** and 4, the bed became more compact, and resulted in no air bubble and better water flow.

For the BP/13 **&** A/M system, compaction might explain the jump in flow rate between runs **#** 1 and 2. The subsequent gradual decrease in flow rate was due to the accumulation of dirt, sand, soil, and other materials in the column. These foreign materials were from the raw water and were trapped as a layer above the $BP/I₃$ media. **By** the end of run **#9,** a layer of sand and dirt and soil could be clearly seen. The accumulation of these materials in the column represented a barrier to water flow. Therefore clogging could potentially be a serious problem, especially in long-term operation. This problem could be ameliorated **by** periodic clearing of the accumulated sludge, pre-filtering of sediments, using raw water that contains low level of solid particulates, or take special precautions to better prepare and develop the well during construction¹⁶⁴.

Discussion of error

The flow rate measurements were carried out **by** collecting treated water using a **100** mL plastic graduated cylinder over time intervals of **10** seconds, **15** seconds, or 20 seconds. Error might be introduced due to the accuracy tolerance of the cylinder marking. Error could also be due to imperfect timing. That is, the timing might be off **by** a second. Nevertheless, these random errors associated with the above two reasons were relatively insignificant. This was illustrated **by** runs **#7** and **9** of which the three flowrate measurements are very close to each other.

7.1.9 Summary of Important Results on the Arsenic Removal Technology

• Both the A/M only system and the BP/I₃ & A/M system were capable of removing total arsenic to below the WHO standard of 10 µg/L.

- The use of BP/I₃ to enhance arsenic removal was apparently not required for the water samples tested in the field site, at least in the short duration of the tests described in this arsenic study.
- **"** The change in **pH,** ORP, total alkalinity, total hardness, total soluble iron, manganese after treatment was not a concern in the water samples tested in the field sites.
- **"** Any effect of these parameters on arsenic removal was not observed.
- Iron might compete with arsenic in adsorption onto A/M media, raising the issue of long-term capacity of A/M. However, iron binded on A/M can adsorb arsenic, replacing the loss in A/M's arsenic adsorbing capacity.
- **"** In addition, iron removal was advantageous because the high iron content in the raw water was unpleasant, as iron gave color, taste, and perhaps odor.
- **"** Clogging of the system with turbid raw water could reduce flowrate. Cleaning might be required unless raw water has low turbidity.

7.2 Arsenic Speciation in Tubewell Water

Thirty-seven arsenic contaminated wells throughout the Nawalparasi and Rupandehi Districts were tested in order to obtain arsenic speciation results. For each of these wells, the total arsenic, arsenic **(III), pH,** and ORP of the raw water were measured. Correlations between arsenic **(III)** as **%** of total arsenic and the following parameters were investigated and discussed:

- **"** Total arsenic
- **" pH**
- Oxidation-Reduction Potential (ORP)
- Well depth
- Well age
- Number of users
- Number of households sharing the well

Refer to Appendix H.1 and H.2 for more details on the above correlations. Appendix H.3 contains correlations between total arsenic and the above parameters.

7.2.1 Total Arsenic and Arsenic Speciation

Results for total arsenic and speciation are summarized in Table 12, Figure *15,* Figure **16,** and Figure **17** The highest concentration was **863** pg/L in Bir Bahadur Gurung's well. The range of arsenic **(III)** as **%** of total arsenic was from 40% to **100%** with a mean of **79%.**

Table 12 Results for Total Arsenic for the **37** Wells in Rupandehi and Nawalparasi **Districts**

 $\sim 10^{-1}$

Figure 15 Results for Total Arsenic for the 37 Wells in Various Villages.

Each bar represents one well.

Figure **16** Arsenic **(III)** as **%** of Total Arsenic for the **37** Wells in Various Villages. Each bar represents one well.

The three highest total arsenic concentrations were **863, 572,** and **328** pg/L. They were all found in the village of Madangram. Because the village of Madangram was a known high arsenic area, **10** out of **37** wells were chosen in this village to better understand the extent of arsenic contamination. Kirtipur-1 was another village with high arsenic in its groundwater. The fourth, fifth, and sixth highest total arsenic concentration **(328,** 242, **233** pg/L) were found in this village. Nine of **37** wells were chosen in this village. These results showed that the occurrence of high total arsenic was not random, but was concentrated in specific villages. When installing a new well, extra care should be given to the well location and its proximity to high arsenic concentration areas. This could reduce the chance of the new well containing high arsenic.

On the other hand, the speciation results seemed to be random across the villages. No village had a consistent level of arsenic (III) as a **%** of total arsenic. The range within

each village could be fairly wide. Therefore, it was concluded that arsenic speciation was not village/area specific.

Figure **17** Correlation Between Total Arsenic and **%** Arsenic (III)

The comparison between total arsenic and arsenic speciation showed no correlation. This implied that arsenic speciation could not be predicted **by** total arsenic. It was impossible to derive any information on speciation using arsenic test methods that measured total arsenic only. Additional techniques such as separation of arsenic **(III)** from arsenic (V) were necessary to determine speciation.

The high percentage of arsenic **(III)** as a percentage of total arsenic in the tubewell water was also a health concern. It was because arsenic (III) was the more toxic form of arsenic compared to arsenic (V). Therefore, the arsenic positive wells should never be used for drinking and cooking purposes.

Discussion of errors

The above arsenic results were determined **by** the **GFAAS** in the Ralph M Parson's laboratory at MIT. As discussed before, the **GFAAS** accuracy was best in the **20-75** pg/L range. Many of the above results were also tested with the field test kits. In general, the two results showed fairly high degree of agreement of **78%.** Table **13** shows the comparison of the **GFAAS** results with the field test kit results.

Test Kit (µg/L)	GFAAS (µg/L)	Agreement
500	572	yes
800	863	yes
100-200	$\overline{121}$	yes
200	328	no
50-100	158	no
100-200	149	yes
100	140	yes
50-100	93	yes
100	154	yes
100-200	102	yes
50-100	103	yes
100-200	96	no
50	95	no
50-100	56	yes
100-200	150	yes
200	91	no
300-500	328	yes
200-300	233	yes
300	242	yes
$20 - 50$	45	yes

Table **13** Comparion of test kit and **GFAAS** arsenic results

7.2.2 Correlation with pH

Figure 18 shows the correlation between pH and arsenic speciation. The pH ranges from 5.76 to 7.27.

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Figure 18 Correlation Between pH and Arsenic Speciation

Arsenic speciation appeared independent of the water **pH.** The square of correlation coefficient was only **0.05,** indicating no correlation. According to the **pH-Eh** diagram, **pH** could only affect the amount of protonation of arsenic **(III)** and arsenic (V), but not the speciation of arsenic. Therefore, **pH** should have no correlation with speciation, as indicated **by** the above results.

Discussion of errors

Some of the errors associated with **pH** measurements were already discussed in the previous section concerning arsenic removal technology. An additional source of error in this experiment concerning arsenic speciation in tubewell water was that the collected water was not representative of the actual water condition in the well. For example, some of the wells visited **by** the author had not been used for weeks or months. In order to collect a water sample, these wells were primed. The priming procedure required pouring a bucket of water into the well. This additional source of external water might interfere with the existing water in the well. When water was pumped from the well, the collected water could be a mixture of well water and priming water, thus not representative of the actual water condition in the well.

7.2.3 Correlation with ORP

Figure **19** shows the correlation between ORP and arsenic speciation. The percent arsenic **(III)** to total arsenic appeared to decrease with increasing ORP.

Figure **19** Correlation Between ORP and Arsenic Speciation

The observed trend of percent arsenic **(III)** decreasing with increasing ORP matched with theoretical prediction. The higher the ORP, the more oxidizing the water was. More arsenic would appear at its oxidized form, i.e. arsenic (V), than at its reduced form, i.e. arsenic **(III).** In other words, the percent arsenic **(III)** decreased with increasing ORP as more arsenic was in arsenic (V) form at high $ORP¹⁶⁵$.

Discussion of error

Some of the errors associated with ORP measurements are already discussed in the previous section concerning arsenic removal technology. An additional source of error in this arsenic speciation in tubewell water experiment is that the collected water is not representative of the actual water condition in the well. For example, some of the wells visited **by** the author had not been used for weeks or months. In order to collect a water sample, these wells must be primed. The priming procedure required pouring a bucket of

water into the well. This additional source of external water might interfere with the existing water in the well. When water was pumped from the well, the collected water could be a mixture of well water and priming water, thus not representative of the actual water condition in the well.

Another serious error with ORP measurement was its applicability to arsenic speciation. ORP measurement was based on the dominant redox reaction couple in the water¹⁶⁶. The presence of multiple couples could limit the interpretation of **ORP167.** For example, if a water contains both Fe(II)/Fe(III) and As(III)/As(V) couples, the speciation of each of these couples should reach equilibrium according to the given water redox potential. However, redox reactions could be very slow¹⁶⁸. The As(III)/As(V) couple might have reached equilibrium, while $Fe(II)/Fe(III)$ has not. This implied that the $Fe(II)/Fe(III)$ couple did not reflect the true redox potential of the water. **If,** unfortunately, Fe(II)/Fe(III) was the dominant redox reaction couple in the water, then the ORP measurement would not reflect the true redox potential of the water. On the other hand, if the iron couple has reached equilibrium but the arsenic couple has not, then the ORP measurement based on the iron couple would not reflect the true arsenic speciation condition in the water. In actual groundwater, many different redox couples existed, further complicating the ORP interpretation. In addition, arsenic was usually not the dominant couple due to its low concentration compared to other couples, such as iron. Therefore, the ORP measurement in this arsenic speciation in tubewell water experiment was not very accurate in predicting arsenic speciation, as suggested in works **by** Holm and Curtiss¹⁶⁹.

7.2.4 Correlation with Well Depth

Figure 20 shows the correlation between well depth and arsenic speciation. The well depth ranged from **28 ft** to 200 **ft.**

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Figure 20 Correlation Between Well Depth and Arsenic Speciation

Arsenic speciation appeared to be independent of the well depth with the square of correlation coefficient of only 0.04. However, a comparison between well depth and ORP showed a moderate correlation of $R^2 = 0.24$, as shown in Figure 21. This correlation showed the deeper the well, the more reducing the water was. This could be explained **by** the ecological redox sequence, which stated the redox should decrease with an increase in depth given the presence of organic matter¹⁷⁰. This ecological redox sequence is governed by the relative change in the standard free energy change (ΔG°) of the common environmental half-reactions. Refer to Table 14 for these common halfreactions.

Figure 21 Correlation Between Well Depth and ORP

At shallow depth, oxygen is present due to easy diffusion from atmospheric oxygen. The half-reaction involving oxygen also has one of the lowest ΔG° , indicating the reaction is highly favorable compared to those half-reactions with less negative ΔG° . As a result, the oxidation of organic matter **by** oxygen (aerobic respiration) is preferred over other oxidants at shallow depth. At a deeper level where oxygen is depleted, the oxidation of organic matter is dominated **by** half-reactions involving nitrate (denitrification) and manganese oxide (Manganese reduction). At even deeper level, the less favorable iron reduction, sulfate reduction, and methanogenesis may occur. The redox potential of the water is defined by the dominant redox reaction occurring¹⁷². For aerobic respiration, the pe is around +12 to *+15.* For denitrification and manganese reduction, the pe are around **+10** to +12 and **+8** to **+11** respectively. For iron reduction, sulfate reduction, and methanogenesis, the pe are approximately -2 to $+1$, -5 to -2 and -7 to -4 respectively¹⁷³. Since pe is directly proportional to **Eh** (redox potential)* and ORP is a rough approximate measure of **Eh,** the decrease in pe with increasing depth should correspond to a decrease in ORP. This explained the moderate correlation between well depth and ORP^{174} .

Recall from the previous discussion of ORP vs. arsenic speciation, it was observed that the percentage arsenic **(1II)** as total arsenic increased as ORP was lowered. Combining this observation with the observation that ORP decreased with depth should yield the conclusion that the **%** arsenic **(III)** should increase with depth. However, this was not observed. This surprising result might be due to the limitation of ORP measurement to predict arsenic speciation under the interference of multiple redox couples.

Discussion of errors

The errors associated with well depth should be minimal. Well depth was easy to measure.

 $*$ **Eh** = 2.3 RT $*$ pe /F where R = universal gas constant, T = temperature in Kelvin, F = Faraday's constant **- 96500,** and **Eh** is the redox potential.

Source: Strumm W., Morgan, J. "Aquatic Chemistry." 2nd edition

7.2.5 Correlation with Number of Users

Figure 22 shows the correlation between the number of users and arsenic speciation. The number of users ranged from **6** to **160.**

Figure 22 Correlation Between Number of Users and Arsenic Speciation

Discussion of results

Arsenic speciation appeared independent of the number of users. The square of correlation coefficient was only **0.06,** indicating no correlation. **If** it was assumed that the number of users was approximately direct proportional to the amount of water use, then water use did not correlate with arsenic speciation.

Discussion of errors

The number of users for a particular well was dependent on factors such as the population in a village, the proximity to other wells, the well condition, the well water quality, and the well ownership^{175,176}. All wells visited by the author were public wells, accessible to the entire village population. The number of users for a well might change as a result of population growth, construction of a new well in the village, or changing water-collecting habits of the villagers due to the closing of certain arsenic contaminated wells. As a result, the actual number of users for a well might change from day to day. In addition, it is common that villagers choose different water sources for different purposes¹⁷⁷. This further complicated the measurement of number of users. Therefore, the measurement was not very accurate.

7.2.6 Correlation with Number of Households Sharing the Well

Figure **23** shows the correlation between the number of households and arsenic speciation. The number of users per well ranges from 1 to **30.**

Figure **23** Correlation Between Number of Households and Arsenic Speciation

Arsenic speciation appeared independent of the number of households. This was consistent with the observation of no correlations between number of users and speciation. It was because there was strong correlation between the number of households and the number of users, as shown in Figure 24.

Figure 24 Correlation Between the Number of Households and Users

Discussion of errors

Similar to the number of users, the number of households for a particular well was subject to changes in village population, water-collection patterns of the villagers, addition or closure of wells, and other factors. Therefore, the data on number of households was not very accurate as it fluctuates daily.

7.2.7 Summary of Important Results on Arsenic Speciation in Tubewell

- Total arsenic appeared to be village/region dependent, while arsenic speciation was not. This has implications for site selection in the constructing new wells to obtain arsenic-free water.
- * On average, **79%** of the total arsenic was in the more toxic arsenic (III) form. The removal of arsenic was therefore **highly** necessary. In addition, pre-oxidation of

arsenic **(III)** to (V) was essential to improve arsenic removal efficiency for many adsorption based treatment technologies.

- * There was a strong correlation between ORP and arsenic speciation. The lower the ORP, the more arsenic was in the arsenic **(III)** form.
- * There was no correlation across sites between total arsenic, **pH,** well depth, well age, number of users, and number of households on arsenic speciation. It was concluded that ORP was a better predictor for arsenic speciation.

7.3 Arsenic Oxidation Kinetics

Arsenic oxidation kinetics was studied to investigate weather natural aeration was sufficiently quick to oxidize arsenic **(III)** to (V). **A** beaker containing arsenic contaminated water was exposed to the atmosphere for about three days. The total arsenic, arsenic **(III), pH,** and ORP of the raw water were measured at **18** different times within the three days. Changes in the above four parameters overtime were recorded. Refer to Appendix **I** for more details on the test results.

7.3.1 Total arsenic and speciation results

Figure **25** shows the change in total arsenic and arsenic (III) over time. Figure **26** shows the change in percent of arsenic **(III)** with time. Total arsenic decreased very slightly with time. The decrease in arsenic (III) and percent arsenic **(III)** over time was quicker.

Figure **25** Change in Total Arsenic and Arsenic (III) with Time

Figure **26** Change in **%** Arsenic (III) with Time

The decrease in total arsenic could be due to adsorption to iron. The total soluble iron concentration in the water was about **2.5** mg/L at the start of the kinetics experiment. Over the next three days, as a result of exposure to the atmosphere, the soluble iron was slowly oxidized into insoluble ferric hydroxide. Since ferric hydroxide was an excellent adsorbent for arsenic^{178, 179}, arsenic was removed from the solution.

The decrease in arsenic **(III)** could be explained **by** two hypotheses. First, arsenic (III) was oxidized to arsenic (V). It could be due to oxygen in the air, or **by** other oxidation means such as photo-oxidation. Second, arsenic **(III)** was adsorbed to ferric hydroxide. **Of** the two hypotheses, the former is more likely. It is because arsenic **(III)** was not adsorbed **by** iron hydroxide as readily as arsenic (V). If the latter hypothesis was true,

then arsenic **(III)** should decrease at a rate slower than that of total arsenic, which contains both arsenic **(III)** and (V). However, the contrary was observed. The rate of arsenic **(III)** decrease was higher than that of total arsenic. This disproved the latter hypothesis as the main mechanism. Therefore, arsenic oxidation was the main reason for the drop in arsenic (III) with time. It follows that arsenic **(III)** as a percentage of total arsenic decreased with time. It should be noted that the trend lines in **Fig 7.11** and **7.12** are intended to highlight the direction of the trend. Theoretically, these trends might not be straight lines. They could be first order decay, or any other mathematical functions. The actual equation describing these trends could only be determined **by** a carefully monitored experiment, and sophisticated analysis to understand the exact mechanisms. These were beyond the scope of this arsenic speciation kinetics study.

It was concluded that the arsenic oxidation kinetics was slow¹⁸⁰. Over the three day period, the percent arsenic **(III)** decreased from about **85%** to **65%. By** extrapolation, complete arsenic oxidation **by** natural aeration could take weeks. Because arsenic (V) was more readily adsorbed than arsenic (III)¹⁸¹, many arsenic remediation technologies, such as iron coated sand, activated alumina, iron filings, and arsenic treatment plants (ATP) required a pre-oxidation step to improve arsenic removal efficiency.

The results from this experiment show that natural aeration might not be effective at oxidation. The pre-oxidation must be accomplished **by** other means, such as the addition of an oxidant. On the other hand, the slow oxidation kinetics allows the author to take his time when performing speciation tests on this particular water. Since the speciation conditions changed so slowly, no significant error was introduced if the speciation test was delay for a few hours. One implication was that the speciation test might not needed to be performed at the moment the water is collected at a tubewell. Samples could be collected in the morning, and speciation tests carried out in the afternoon with no significant error. However, another water could contain a catalyst for aeration oxidation. Therefore, the speciation test protocols should not be changed unless more experimental results confirmed the slow kinetics.

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Discussion of errors

The fluctuation in total arsenic and arsenic **(III)** could be attributed to the accuracy of the **GFAAS** analysis. In addition, the time between when the samples were collected and acidified and then analyzed in the laboratory was about two months. Water might have vaporized within the headspace of the mailer tubes such that the arsenic concentration in the solution appearsedhigher (arsenic is not volatile). Another potential error \was that the speciation procedure required vigorous shaking of the mailing tube for five minutes. During the five minutes, arsenic **(III)** might have oxidized into arsenic (V) due to the presence of oxygen in the mailer tube. Therefore, the measured percent arsenic (III) could be 10-20% lower than the actual percent arsenic $(III)^{182}$.

7.3.1 pH and ORP results

Figure **27** and Figure **28** show the change of **pH** and ORP with time. It was observed that **pH** increased with time, while ORP decreased with time.

Figure **27** Change in **pH** with Time

The increase in **pH** with time could be explained **by** the decarbonation of groundwater. When water was underground, it might contain high total carbonate, C_T^* , as a result of dissolution of underlying geologic formations such as calcite $(CaCO₃)$, and dolomite $(CaCO_3 \cdot MgCO_3)^{183}$. As the groundwater was taken out from the ground and was exposed to the atmosphere, new equilibrium with the atmospheric carbon dioxide $(CO₂)$ would be established. Usually, a lower level of total carbonate was expected for the new equilibrium¹⁸⁴. Decarbonation must occur. Of the different species in the carbonate system, only $[CO_2]_{(aq)}$ could escape to the atmosphere as $[CO_2]_{(g)}$. To compensate for the loss of $[CO_2]_{(aq)}$, the other carbonate species must be protonated to $[H_2CO_3]$ and $[CO_2]_{(aq)}$ in order to retain the carbonate system equilibrium. The protonation reactions consumed $[H^+]$ in the solution. With less $[H^+]$, the water became less acidic. It followed that pH should increase.

 * **CT** = $[H_2CO_3^*]$ + $[HCO_3^-]$ + $[CO_3^2]$ where $[H_2CO_3^*]$ = $[CO2]_{(aq)}$ + $[H_2CO_3]_{(aq)}$

Figure **28** Change in ORP with Time

The decrease in ORP with time was puzzling. This observation was completely contrary to the expected increase in ORP with time. The water should be more oxidizing with time, as suggested **by** the arsenic oxidation results; therefore, ORP should increase with time. The anomaly might be explained **by** the interference of the ORP **by pH.** Redox potential was sensitive to **pH** if hydrogen ion or hydroxide ion was involved in the redox half-cells. The ORP tended to decrease when pH increases¹⁸⁵. As a result, the ORP measurements were **highly** distorted and did not truly reflect the water condition. **A** comparison between **pH** and ORP, as in Figure **29,** shows a strong negative correlation between **pH** and ORP.

Figure **29** Correlation Between **pH** and ORP

Discussion of errors

The errors associated with **pH** and ORP measurements were thoroughly examined earlier, thus not repeated here.

7.3.2 Summary of Important Results on Arsenic Oxidation Kinetics

- The oxidation of arsenic (III) to (V) was slow under natural aeration for this particular water. At least one week was needed for complete oxidation.
- **"** Natural aeration can be ineffective as an arsenic pre-oxidation step for many arsenic removal technologies. Other oxidation techniques are needed.

CHAPTER 8 CONCLUSIONS AND RECOMMENDATIONS

8.1 Arsenic Removal Technology Evaluation

The arsenic removal technology was very successful in removing arsenic. The A/M only and the $BP/I_3 \& A/M$ systems were tested at four and nine wells respectively, with raw water total arsenic between 147 to $863 \mu g/L$. In all of the above tests, the treated water contains non-detect level \ll $\frac{1}{g}(L)$ of total arsenic, which is below the WHO guideline of **10** pg/L total arsenic. **By** comparing the arsenic removal results between the A/M only system and the BP/13 **&** A/M systems, it appeared that the A/M **by** itself was sufficient for arsenic removal. BP/I_3 is not required.

Iron was also oxidized and subsequently removed **by** the treatment systems, from **2.5-3.5** mg/L in the raw water to below detection limit of **0.1** mg/L in the treated water. On the one hand, iron is undesirable because iron adsorption reduced the A/M's capacity to bind arsenic. On the other hand, the iron bound on the A/M surface could adsorb arsenic, replacing the loss in A/M's arsenic adsorbing capacity. In addition, iron removal was advantageous because the **high** iron content in the raw water was unpleasant, as iron gave color, taste, and perhaps odor.

The flow rate through the system was about 20 L/hour, which was more than sufficient for an average household need of about **20-30** L/day. However, clogging of the treatment system due to the accumulation of dirt, sand, and other matter found in the raw water could potentially be a serious problem. Pre-filtering of the raw water might be needed.

Several key recommendations can be made based on this study. They are:

• Further investigations in the A/M media regarding its technical, social and economic aspects is needed.

- **"** For the technical aspect, a practical filter column design using local materials should be developed. Examples include the use of buckets, or plastic water bottles. The plastic column setup used in this arsenic study was not locally available and was therefore not appropriate for household use.
- The long-term oxidation and adsorption capacity of A/M should be assessed.
- The treated water should be tested for the full set of water quality parameters in the drinking water standards.
- **"** For the social aspect, issues regarding the ease of setup, operation and maintenance, availability of the media, and the perceived quality of the treated water should be addressed.
- For the economic aspect, a users' willingness-to-pay study is suggested to understand how much the villagers can afford.
- Long-term cost of the A/M media based on its arsenic adsorption capacity, and the capital cost of the treatment system should be estimated in order to evaluate its economic viability, especially relative to other possible systems appropriate for Nepal.

8.2 Arsenic Speciation in Tubewell Water

Speciation tests at **37** wells showed, on average, **79%** of the total arsenic was in the more toxic arsenic **(III)** form. The removal of arsenic is therefore **highly** necessary. In addition, pre-oxidation of arsenic **(III)** to (V) was essential to improve arsenic removal efficiency for many adsorption-based treatment technologies. Simple aeration was not effective to oxidize arsenic.

There was a strong correlation between ORP and arsenic speciation. The lower the ORP, the more arsenic was in the arsenic **(III)** form. On the other hand, there was no correlation between total arsenic, **pH,** well depth, well age, number of users, and number of households on arsenic speciation based on the well water tested. It was concluded that ORP was a suitable predictor for arsenic speciation.

Several key recommendations can be made based on the speciation aspect of this study. They are:

- * The currently available scientific data on the toxicity of arsenic **(III)** vs. (V) is still primitive. Further studies are needed.
- The speciation test method used in this study should be checked with an alternative test method in order to confirm the validity of the results.
- **"** More wells should be speciation tested to explore undiscovered correlations and trends.

Access to adequate safe water supply is a basic human right. Every effort should be taken to protect this right.

APPENDIX A AQUATIC TREATMENT SYSTEMS, INC.

Company Profile $A.1$

Company Profile

50 Cole Parkway, Suite 26, Scituate, MA 02066 Phone: (781) 545-8588 or 888-772-AQUA (2782) Fax: (781) 545-8488 www.aquatictreatment.com

C Aquatic Treatment Systems, Inc. 2001

OVERVIEW

Aquatic Treatment Systems, Inc. "ATS" was founded in 1992 as a boutique consulting company to provide cost effective solutions for purifying contaminated water. The Company typically made use of existing water treatment technologies in the design of treatment solutions. Through its initial work as a consulting company, the Company determined that existing technologies did not have the capabilities to remove a number of priority pollutants to safe drinking water levels. Given growing health concerns, a very large unmet world market need and imminently tougher regulatory standards, at the end of 1999 ATS made the decision to focus its total efforts on developing enabling technologies that would completely remove priority pollutants from water supplies.

The Company's initial target contaminant was arsenic, one of the highest priority contaminants in the United States and in many other parts of the world. Arsenic is a naturally occurring element in the earth's crust. Arsenic in groundwater is largely the result of minerals dissolving from weathered rocks and soils. The health effects from arsenic consumption have been well documented. The more scrious effects from chronic toxicity include various forms of skin cancer, organ cancers and neurological disorders. Arsenic can enter the body by ingesting food or water, dermal contact and inhalation. When arsenic is ingested it is quickly distributed to body tissues and organs including the spleen, kidneys, liver, lungs, bladder, reproductive organs and skin.

Because the market lacked a technology that adequately and efficiently removed arsenic, ATS invented, developed, filed for patents, field-tested and has sold a proprietary water purification product that removes total arsenic from contaminated water to nondetectable levels (less than 1 part per billion (ppb)). Based upon the latest scientific studies undertaken for the United States Environmental Protection Agency (USEPA), negligible exposure to arsenic in water occurs at a concentration of <2ppb of arsenic.

The Company has also invented, developed and filed for patents on technology that has direct application in another high health risk area - water borne microbial contamination. Water borne disease occurs in both the Western world, where the most current water treatment technologies are used, as well as in much of the third world. Contamination can come from the water source or from the piping systems carrying water to or within the home. The Company's technology has the capability to disinfect and filter enteric pathogens that cause waterborne and water based diseases. A study by the Harvard School of Public Health pointed out that children and the elderly are most susceptible to infectious water borne disease. Many pathogens known to cause gastrointestinal illnesses are resistant to chlorine, the primary disinfectant used. Chlorine can also impart harmful residual chemicals into the water. The Company's technology disinfects on contact with no chemical residuals.

1

In developing new applications of its filtration technology, ATS is following a disciplined strategy that focuses on the successful customization of its filtration media to target specific high priority pollutants. The Company believes that each new application will be patentable and accordingly intends to file patent applications, as it has for its arsenic removal and disinfection technologies.

In developing each application, the Company will use its formidable capabilities in research and development, product and system design, pilot testing and initial market introduction in order to define the operating parameters and total capabilities of the technology.

By following this product development cycle, the Company will be able to offer field-tested filtration media or devices to existing manufacturers and distributors of water treatment systems for broad scale distribution and sales. These customers include suppliers for household and community treatment systems, as well as suppliers of pointof-use systems, both under the counter and carafe type. The Company will also provide filtration media or systems to water utilities and to community water systems. It will also market filtration media through appropriate strategic alliances with resin and/or chemical manufacturers. In all cases, technical support and consulting will be provided by the Company as required.

As the Company has already put in place quality contract manufacturing capabilities that allow it to maintain adequate capacity to supply all demand, it will focus near term on the commercialization of its proven arsenic technology and on the validation and field testing of its disinfection technology, prior to proceeding with any new product development. The Company has conducted preliminary feasibility testing on a number of other priority pollutants, which include: MTBE, TCE, Perchlorate, and 2-Methylisoborneol. Market analysis projects MTBE to have a potential market demand similar to arsenic, and the Company has scheduled MTBF as its next priority for development and market introduction.

The Company's core technology is based upon proprietary new chemistries and methodology to modify and convert commonly used water treatment materials into high performance filtration media with a strong affinity for target contaminants. The Company also leverages the experience of its principals in designing cost effective water treatment systems that enable its treatment media to be applied across the complete range of contaminated water sources.

Arsenic Products Description

The Company's first products utilize its arsenic removal media, A/I Complex 2000 and A/M Complex 2001, to completely remove arsenic from contaminated water These systems range in size from under the counter point-of-use systems, to whole house point-of-entry systems, to small community systems. The media is equally effective and the systems are scalable for larger community systems. All arsenic removal systems can also be expanded to remove other contaminants if needed, which may include: fluoride, iron, hydrogen sulfide, manganese, sulfide and pH adjust.

The Company also plans to sell its arsenic removal media in bulk to water treatment systems manufacturers and distributors, to cartridge manufactures including carafe point-of-use filters, to water utilities servicing community water systems and for industrial applications such as the food and beverage industry and the bottled water industry.

The ATS family of arsenic removal media can also be used to remove a number of other specific contaminants, including mercury, selenium, antimony, cobalt, eesium, radium 226 and radium 227. Certain distributors are interested to pursue one or more of these applications.

Disinfection Products Description

The Company has installed filter devices that utilize its BPI3 Disinfection Media for non-potable water applications. These filter systems are used to disinfect water features such as fountains and water walls in hospitals, restaurants and office buildings. Test results have confirmed that E. Coli, Pseudomonas, and Legionella bacteria were effectively eliminated.

The Company plans to initiate microbiological reduction testing as required by the United States Environmental Protection Agency Office of Drinking Water and Office of Pesticide Programs to prove that our technology will remove, kill or inactivate all types of disease-causing micro-organisms in water.

3
Community System For Total Arsenic Removal

Benefits of the Community System for Total Arsenic Removal

- Reduces total arsenic to below 2 parts per billion
- Provides a good tasting beverage
- Provides bright clear water with no staining
- Automated flow through system
- Modular system with flexibility for additional treatment options
- · Fail safe redundant system design
- Minimal space requirements (small foot print)
- No use or storage of hazardous chemicals
- No hazardous waste
- Lifetime performance warranty available
- + Affordable annual cost for safe water: S0.30 to \$0.50 per 1,000 gallons

As/1200 Point-of-Entry Total Arsenic Removal System

- Minimal space requirements (small foot print)
- No maintenance requirements for homeowner
- . No use or storage of hazardous chemicals in the home
- Periodic service to dispose of arsenic waste and replenish media
- · Lifetime performance warranty available
- Annual cost for safe water: 0.3¢ to 0.8¢ per gallon

5

As/100 Point-of-Use Total Arsenic Removal System

ATS ARSENIC ADSORPTION MEDIA FEATURES & PERFORMANCE

Aquatic Treatment Systems, Inc. has developed a comprehensive and practical approach to provide both community and non-community water systems with a safe, cost-effective and versatile arsenic removal technology. ATS has invented, successfully field-tested and introduced to market patent pending adsorption/complexing media for the rapid and complete removal of arsenic from aqueous streams.

Each volume of ATS adsorption/complexing media will treat in excess of 80,000 volumes of water contaminated with 100 parts per billion of arsenic over a wide pH range and in the presence of sulfate and/or fluoride. ATS systems are designed to consistently remove arsenic to below detectable levels (<1.0 ppb) at high flow rates of 1 bed volume per minute (1 BV/M).

Extensive field-testing and third party evaluation of the ATS arsenic removal media have demonstrated a number of superior product features and competitive advantages:

Non-Detectable Arsenic Results - Test results have proven that the ATS adsorption media will achieve complete removal of total arsenic in concentrations over 1,000 parts per billion and in a wide range of water conditions. Our first system in Wisconsin, a whole house point-of-entry system, has processed well over 100,000 gallons of safe, good tasting water during its first 14 month of operation, with all treated water testing "non-detectable" for Total Arsenic.

- Adsorption Capacity Test results show high arsenic adsorption levels over a wide range of **pH** levels **(6.0** to **9.0).** When other typically competing parameters are present, (i.e. fluoride, sulphate, phosphate) no significant changes in performance are shown The **ATS** media has been tested under a wide range **of** water conditions. Under controlled tests no significant difference in adsorption rate or capacity is found. This results in a significani pcrformance advantge when water conditions are known to change, or source water pH is high.
- **Through Put/Flow** Rate **,** The rate of adsorption is significantly higher than any other media **A** rate of up to two (2) **bed** volumes per minute (BV/M) is possible with **1.0** BVM standard. Other medias typically operate at flow rates **of 0 1** to **0.2** BV/M. In other words, **ATS** medias are able to effectively remove **total** arsonic contaminants at flows at least **5** times faster and under some conditions **10** times faster.
- Oxidatio. *Capacity* **--** The **ATS** adsorption media will oxidize arsenic **111** and convert it to arsenic V. The oxidation capacity of the media is sufficient for many applikations where there is a limited daily volume, such as Point-of-Use (POU) devices, or when a low to moderate level of arsemic III is present. ATS also has a companion oxidant media. This on demand oxidant is effective in any water including industrial streams where high concentrations of arsemic III are present. **NO** CHLORINEMDECHLORIN1ZAT1ON **STEPS** ARE **REQUIRED.**
- Safe and Reliable ATS arsenic removal media requires no chemical additions to achieve complete arsenic removal. The **ATS** media is not designed for regeneration which eliminates any residual hazardous liquid waste. Binding characteristics of the **ATS** media are without equal; the bound arsenic will not leach and re-contaminate the environment. When the media reaches capacity it is simply disposed oFin **a** non-hazardous landfill **ATS** arsenic removal media has **hcen** tested for safety under NSF 61 protocol.
- **Effective in the Presence of Interferences** Unlike exchange resins that give up significant capacity when other ions are present, **ATS** media complexes target ions. **As** a result, the *ATS* adsorption media binds arsenic without giving **up** capacity to common ions such as fluoride and sulphate.
- **Durable and Tolerant To Changing Water Conditions No chemical or operational** modifications are necessary when water conditions change. Temperature, pH, and contaminants affect little or no change in adsorption performance of the ATS media.
- **Variely of Media Dimensions and Shapes The standard media is graded in a** flake/grain form ranging from 20 to 40 mesh sizes. Spherical shapes are also avaiiable. When there are **high** volume **flows** from municipal or industrial applications, large bed sizes are required. To reduce pressure drop multidimension spherical shapes are available.
- Robust Media Unlike other media such as granulated ferric hydroxide, ATS media do not crumble and disintegrate with installation and use. ATS rigid, high composition strength media allow for simpler, more rapid installation and will not progressively compact, which causes loss of flow and pressure drop.
- Non-hazardous Solid Waste The arsenic binds so tightly to the ATS media that the arsenic will not leach off. A safe non-hazardous solid waste that meets USEPA, Toxicity Characteristic Leaching Procedure [TCLP] requirements is a safe alternative to the regeneration chemicals and hazardous waste streams common to resin regeneration.
- **Ease Of Operation** ATS systems operate automatically on demand. The installation and management of replacing media is simple and does not require special training or equipment. Whether the ATS media is used in residential, community or industrial applications it is easy and safe to use.
- Small Space Requirements ATS media systems provide adsorption capacities many times greater than other known technologies. As a result, the total space or "foot print" requirement of an ATS media system is usually less than one half that of other equipment installations. This is a significant factor in homes and with small community systems where existing space is often limited.
- Certified For Use ATS media has been certified by the state of Wisconsin for use in homes and community water systems.

COMPETITION

ATS has compared all competitive arsenic adsorption media for general effectiveness in removing total arsenic and for relative cost. The per gallon cost comparison assumed media use for a point-of-entry household system, including disposal costs for spent media and liquid waste from regeneration.

ATS arsenic adsorption media is substantially the best value and most effective technology.

Timothy J. Badger, Founder and CEO

Badger received his doctoral degree from Boston University in **1979.** Is graduate degrees were in environmental sciences and natural resource management, specializing in aquatic environment studies. He has held senior positions at environmental firms with responsibilitics in sales, marketing, environmental risk management, financial services, operations, field engineering and project management. **Dr.** Badger has years of applications experience, which has **led** to his co-development of key inventions and proprietary know-how that are the basis for the Company's water treatment technologies.

Dr. Badger is a member of the National Sanitary Foundation **(NSF)** Task Group on Arsenic. He'is a member of the Wisconsin State Arsenic Subcommittee under the Department of Natural Resources (NR) and the Department of Commerce. Dr. Badger is a member of the State of New Ilampshire Advisory Board for Arsenic. Ile is a *member* of the American Water Works Association *(AWWA),* the National Ground Water Association (NGWA) and the Water Quality Association (WQA),

Phillip *C.* **Henderson, President**

henderson received his BS in engineering from the **US** Naval Academy, and his M413A from Harvard Business School in **1969.** He has held top management responsibilities in public and private companies. He provides the Company with broad management experience in technology based, high growth businesses. He has industry experience with leading edge technologies in water treatment, separations technologies and health care, with expertise **in** start-ups and turnarounds. **fie also** has extensive experience in system and process design. Henderson is responsible for building the management team and overseeing all business and financial endeavors of the Company. **He** is **a** member of the Water Quality Association **(WQA).**

Paul Smith, Senior Synthetic Organic Chemist

Smith has over **30** years of Research **&** Development experience in the chemical industry, with extensive expertise in the chemical activation/derivation of solid supports for high purity separations. In addition to graduate work **in Chemistry aid Organmic** Synthesis', Smith studied patent law. He is the inventor of 20 patents and has co-authored numerous technical and product papers and one **book** related to novel chemistries based on natural and synthetic products Smith **is** co-inventor **of** the Company's technology with Dr. Badger. He is a member of the Water Quality Association (WQA).

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A.2 MSDS

For BP/I_3

For A/M

APPENDIX B PARASI SURVIVAL GUIDE

Three members of the MIT Nepal Water Project Team 2001-2002 were based on Parasi for part of their study. Parasi is a small town of approximately 2000 people in the Nawalparasi district. It is **6** hours car ride from Kathmandu, or 1hour car drive from Butwal or Lumbini.

The three major streets in Parasi are lined on both sides with general stores and shops. Simple essential items used **by** the Nepali can be brought there. For example, toothpaste, bar soap, mosquito coils, clothing, bedding, rugs, bottled water, and water buckets can be easily found. On the other hand, medical supplies or Western goods are hardly found. Figure **30** shows a schematic map of Parasi town center and well locations. Figure **31** shows the street scene of the main street looking east.

Figure **30** Schematic Map of Parasi Town Centre and Well Locations

There are two Nepali restaurants (shacks) in Parasi, serving mostly spicy Indian or Nepali dishes. Besides eating out, cooking at home is another option. Kerosene stove, cooking utensils, pots and pans, and fuel can be easily bought. Fresh vegetables, rice, eggs, chicken, oranges, cooking oil, and seasonings can be found throughout the town as well.

The **ENPHO** house is a typical Nepali house. It is minimally furnished, with only beds, chairs and tables in the house. The rooms are lit **by** dim electric light bulbs, with intermittent power supply. There is a squat style latrine in the backyard separated from the house. The latrine has no light, and must be pour flushed (manually flushed with a bucket of water). Water is available from a tubewell in the backyard about **10** meters away from the latrine, as shown in Figure **32.** The water is not safe for dinking, as it contains about **300** pg/L arsenic and is microbiologically contaminated. Drinking water is collected from the town water supply pipe in the front yard next to the main street. Town water is generally available daily from 7am to 8am and 4pm to 4:30pm. It comes from a deep boring well northeast of town. Besides drinking and cooking, this water supply is usually used for washing or taking a shower. Most Nepalis drink directly from this water supply; however, drinking without boiling is not advised.

The weather in Parasi during January is very comfortable. It is usually suuny and dry with highs around 20-25 °C. Nevertheless, it can be chilly at night, as the mercury dips to about **10 'C.** Mosquitos are always a problem, even in the middle of winter. Therefore, malaria pills should be taken. Mosquito coils and pads sold in Parasi are effective in keeping mosquitos away.

Phone calls to Kathmandu can be made at numerous places in the town, but international calls are fairly expensive **(US\$2-3** per minute) and not easy to make.

It is recommended to bring the following items: toilet paper, flash light with extra batteries, matches or lighter, detergens or dishwashing liquid, chlorine disinfectant solution called Piyush or equivalent disinfection agents, warm clothes, a pair of boots and sandals. Expect to spend less than NRs **500** per day* **(-US\$6.7)** in Parasi for normal daily needs. Bringing electronic equipment is not advised. Electricity is unreliable and blackouts occur frequently.

Figure **31** Street Scene of Parasi

Figure **32** Backyard Well

^{*} In Jan 2002, **US\$1 =** NRs **75** (Nepali Rupees)

APPENDIX C TRIP SCHEDULE AND EXPENSE

C.1 Schedule

C.2 Project Related Expense

NRs **75= US\$ 1**

Note: Room and food in Butwal was provided free of charge **by DIDC** (formerly **FINNIDA).**

APPENDIX D CONTACTS IN NEPAL

Roshan R. Shrestha Executive Chairman **ENPHO** Thapagaon, New Baneshwor, Kathmandu, Nepal. P.O.Box 4102 e-mail: rshrestha@mos.com.np phone (home): **272590** phone (office): *491052,* **491376**

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APPENDIX E GFAAS RESULTS

E.1 Calibration Curves

Calibration curve group **1**

Calibration curve group 2

Calibration curve group **3**

Calibration curve group 4

Calibration curve group **5**

Calibration curve group **6**

Calibration curve group **7**

Calibration curve group **8**

Calibration curve group **9**

Calibration curve group **10**

Calibration curve group **11**

Calibration curve group 12

Calibration curve group **13**

Calibration curve group 14

Calibration curve group 15

Calibration curve group **16**

Calibration curve group **17**

Calibration curve group **19**

Calibration curve group 20

Calibration curve group 21

Calibration curve group 22

Calibration curve group **23**

Calibration curve group 24

Calibration curve group **25**

Calibration curve group **26**

Calibration curve group **27**

Calibration curve group **28**

Calibration curve group **29**

Calibration curve group **30**

Calibration curve group **31**

E.2 Interpolated Results

Analyzed on **7** March 2002

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Analyzed on 12 March 2002

 \bar{z}

Analyzed on 14 March 2002

Group 20 slope int **0.0013 0.006**

Group 21 **slope** int
0.0013 0.003 **0.0013 0.003**

Group **23** slope int 0.0012 **0.013**

Analyzed on 21 March 2002

Group 27 **block c** slope int **c** 0.001 **c** 0.008 **0.001 0.008**

 $\mathcal{L}_{\mathcal{A}}$

Group 30 slope int **0.0011** 0.014

APPENDIX F ARSENIC REMOVAL TECHNOLOGY DATA

Run #8

 $\mathcal{A}^{\mathcal{A}}$

APPENDIX G DIDC WELL DATA MASTER LIST

G.1 Basic Well Information

RWSSSP **- Ill**

Updated on **29.8.2001**

District: Rupandehi **VDC:** Devdaha

Print date: 22-Jan-02

New Wells Jan-Feb, 2001

New Wells Jan-Feb, 2001

Old Wells Jan-Feb, 2001

Old Wells Jan-Feb, 2001

11-Nov-01

23-Nov-01

RWSSSP **- III**

Updated on **9.9.2001**

Old Wells

District: Nawalparasi

VDC: Sunwal

Print date: 21-Jan-02

Jul-01

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11-Nov-01

21-Jan-02

21-Jan-02

23-Jan-02

Legend:

- **Pop.** ⁼ Population
- STW = Suction Tubewell
- LTW = Lift Tubewel
- DW = Dug Well
- R-l= RWSSSP phase **1**
- R-11 = RWSSSP phase 2
- R-1ll = RWSSSP phase **3**
- **VDC** = Village Development Committee
- DWS = Department of Water Supply and Sewerage, Royal Government of Nepal

NRC = Nepal Red Cross

G.2 Arsenic Information

RWSSSP **- Ill**

Updated on **29.8.2001** District: Rupandehi **VDC:** Devdaha Print date: 22-Jan-02

New Wells Jan-Feb, 2001

New Wells Jan-Feb, 2001

Old Wells Jan-Feb, 2001

Old Wells Jan-Feb, 2001

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Old Wells Jan-Feb, 2001

11-Nov-01

23-Nov-01

23-Jan-02

RWSSSP **- Ill**

Updated on **9.9.2001**

Old Wells

District: Nawalparasi

VDC: Sunwal

Print date: 21-Jan-02

11-Nov-01

21-Jan-02

21-Jan-02

23-Jan-02

Legend:

Pop. ⁼ Population

ND = Non-detect (detection level unknown)

APPENDIX H TUBEWELL SPECIATION DATA

H.1 Basic Well Information

RWSSSP **- Ill**

Updated on **29.8.2001**

District: Rupandehi **+** Nawalparasi

VDC: Devdaha **+** others

H.2 Site Visit Information

RWSSSP **- III**

Updated on **29.8.2001**

District: Rupandehi **+** Nawalparasi

VDC: Devdaha **+** others

H.3 Total Arsenic Correlations

Total arsenic is correlated with the followings:

- **" DIDC** (formerly **FINNIDA)** total arsenic records
- **"** PH
- **"** ORP
- Well depth
- Well age
- **"** Population
- Number of households

Except for **DIDC** total arsenic records, no correlations were observed.

Fig **H.3.1** Correlation between **DIDC** (formerly **FINNIDA)** total arsenic records with the author's **GFAAS** results

Fig **H.3.2** Correlation between total arsenic and **pH**

Fig **H.3.3** Correlation between total arsenic and ORP

Fig H.3.4 Correlation between total arsenic and well depth

Fig **H.3.5** Correlation between total arsenic and well age

Fig **H.3.6** Correlation between total arsenic and population (number of users)

Fig **H.3.7** Correlation between total arsenic and number of households
APPENDIX I OXIDATION KINETICS DATA

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