



AQUATIC ARSENIC TOXICITY AND TREATMENT

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Chapter 8

Presence and removal of arsenic from rural water supplies in Canada

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Abstract

The urgent need to find removal techniques for arsenic (As) in drinking water supplies has focused on physical and chemical removal techniques. This includes conventional processes, such as coagulation, iron-manganese removal and sorption processes including ion exchange, activated alumina, and iron-coated sand. Different types of membranes, micro-, ultra-, nano- and reverse osmosis, are also used. The potential for using biological treatment for arsenic removal has not been addressed. There are, however, several characteristics of arsenic in water that makes it suitable for microbial removal processes. Arsenic exists in several different states with the major health concern centred around As^{3+} and to a lesser extent As^{5+} , which are present as inorganic ions. Organic arsenic compounds are of a lesser health concern and are also typically only a small percentage of the arsenic pool in groundwater. As^{3+} is much more difficult to remove from water than As^{5+} and one of the key pretreatments in arsenic removal is to oxidize the As^{3+} to As^{5+} . Conventional processes use oxidizing compounds, such as chlorine, ozone, potassium permanganate etc. However, microorganisms can both reduce As^{5+} to As^{3+} and they can oxidize As^{3+} to As^{5+} . In addition, they can metabolize organic arsenic complexes and potentially release As^{3+} or As^{5+} into the water. The treatment processes described here require no chemicals and rely on several different mechanisms with biological and chemical processes interacting to remove arsenic and several other compounds from solution. By taking advantage of naturally induced removal of arsenic, inexpensive and effective technologies suitable for the treatment of arsenic in rural areas around the world may become a reality. For simplicity we have termed the designed treatment units biological reactors. The goal with biological treatment of arsenic is to move all the dissolved arsenic into a particulate state either as part of the microbial biomass or as organic/inorganic flocs that can be easily separated out. The key in a biological reactor is therefore to establish conditions where these reactions are achieved. Biological reactors have been used in rural Canada on an experimental basis for 7 years with consistent arsenic removal rates exceeding 90% irrespective of season. These reactors remove 99% of the iron, and can also remove dissolved organic material, convert ammonium to nitrate, and when properly designed can remove manganese. The arsenic-enriched biomass and organic-inorganic floc can be removed without replacing the support material in contrast to several sorption technologies.

While the presence of arsenic in regional water supplies in Western Canada were considered to be of quite limited concern, new guidelines for allowable arsenic levels have prompted a re-evaluation of its importance in rural ground water sources across the Province of Saskatchewan. It is anticipated that future guidelines will be less than 10 micrograms/L and 16% of Saskatchewan well waters will then need to be treated to be in regulatory compliance. For Western Canada, arsenic has therefore gone from not being a problem to an emerging contaminant potentially affecting the health of many rural people. Efforts to proactively avoid negative health effects of arsenic-tainted water will therefore be required.

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Monitoring of arsenic levels combined with R&D to improve and implement arsenic removal processes are essential for the protection of the health of rural water users.

Introduction

Background

The National Research Council's report "Arsenic in drinking water" (1999) outlines several issues associated with the chronic ingestion of arsenic. It can cause bladder, lung and skin cancer as well as having non-cancer effects, such as cutaneous lesions. Arsenic can pass through the placenta and can affect fetal growth and prenatal viability. Arsenic has not been found to be required for any essential biochemical process. Humans absorb arsenic from the gastrointestinal tract and transport it in the blood bound to sulfhydryl groups in proteins, amino acids and peptides. Arsenic has a half-life of about four days in the body and is mainly excreted through the urine. Humans and some animals can methylate arsenic into less toxic and more easily excreted compounds. The ability to methylate arsenic is variable depending on many factors including nutritional status, age, etc.

On the basis of its work the National Research Council (1999) recommended that a level of 50 micrograms/L ($\mu\text{g/L}$) does not achieve the U.S. Environmental Protection Agency's goal for public health protection requiring prompt downward revision. Consumption of water with 50 $\mu\text{g/L}$ arsenic carries an estimated increased risk of dying from cancer of 20 per 1000. The extent to which the arsenic level needs to be lowered is hotly debated around the world. Canada decreased its arsenic guideline to 25 $\mu\text{g/L}$. WHO decreased its 1984 guideline of 50 $\mu\text{g/L}$ to 10 $\mu\text{g/L}$ in 1993 (WHO 1993). Suggestions that arsenic levels need to be revised to levels below 2 $\mu\text{g/L}$ has been made by the U.S. Environmental Protection Agency (Macler and Tikkanen, 1994), but for now the U.S. arsenic limit has been set to 10 $\mu\text{g/L}$. The guideline remains at 50 $\mu\text{g/L}$ in Bangladesh where 92% of districts surveyed exceeded this level with a potentially exposed population of 24 million (ACIC, 1998). The guideline in China is also 50 $\mu\text{g/L}$.

There are many water supplies around the world that have arsenic levels in excess of 50 $\mu\text{g/L}$. The most notable region is Bangladesh where the presence of around 50,000 wells during British colonial rule were expanded to between 3-5 million today accounting for 97% of rural drinking water supplies (UNICEF 1999). Around 30% of these wells have arsenic levels in excess of 50 $\mu\text{g/L}$ (ACIC 1998). This has been considered the largest mass-poisoning in the world. Arsenic has no taste and no smell and is most prevalent in groundwater supplies. These waters may therefore leave no hint that something is amiss. Chemical analysis is required to pinpoint the presence of arsenic.

Most countries around the world likely have arsenic problems in some parts of the country. The reason for such widespread contamination by arsenic is the common occurrence of arsenic in minerals, ores, and sediments which when in contact with water can dissolve arsenic leading to contamination. Pesticides and some agricultural practices can also result in arsenic contamination of water supplies. As a result of this, arsenic contamination has been reported from Bangladesh, Chile, United

States, Canada, Mexico, Thailand, Japan, United Kingdom, India, Hungary, China, Argentina, Taiwan, Philippines, New Zealand, Inner Mongolia and Australia.

What is required is a proper assessment of the chemistry of the water, and its content of arsenic. After 5-15 years of arsenic exposure, the human health effects become irreversible. Proper analysis and actions are required to ensure that people are not consuming arsenic-tainted water.

Arsenic chemistry and microbiology

Arsenic exists in several different states with the major health concern centred around As^{3+} and to a lesser extent As^{5+} , which are present as inorganic ions. Organic arsenic compounds are of lesser health concern and are also typically only a small percentage of the arsenic pool. As^{3+} is much more difficult to remove from water than As^{5+} and one of the key goals in arsenic removal is to oxidize the As^{3+} to As^{5+} . As^{3+} exists mainly under conditions of low oxygen and as soon as oxygen is introduced to the water some physical precipitation will take place especially in the presence of iron. Also under aerobic conditions some bacteria will oxidize As^{3+} to As^{5+} (Wilkie and Hering 1998, Langner et al. 2001). Under anaerobic conditions other types of bacteria will reduce the As^{5+} to As^{3+} ; this reaction can even solubilize precipitated arsenic. An opposite reaction can also occur under anaerobic conditions where sulphate-reducing bacteria can generate sulphides resulting in precipitation of arsenic-sulphides (Castro et al. 1999).

Arsenic is not very toxic to microorganisms with other metals, such as copper and lead, exhibiting greater toxicity (Aoyama and Nagumo 1997). Indeed constructed microbial mats have been shown to tolerate up to 350 mg As/L (Bender and Phillips 1995). In addition to being able to oxidize arsenic, microbes produce metal-binding flocculents (polyanionic polysaccharides) capable of arsenic-sequestration (Bender and Phillips, 1995).

Microorganisms can metabolize organic arsenic complexes and potentially release As^{3+} or As^{5+} into the water (Hanaoka and Kaise 1999). Frankenberger et al. (1995) described the bio-oxidation and methylation of arsenic resulting in sorption into biomass (biosorption), or onto soil particles, and volatilization (methylation).

The goal in biological removal of arsenic is to move all the dissolved arsenic into a particulate state either as part of the microbial biomass or as organic/inorganic flocs that can be easily separated out.

Ground water treatment using microorganisms

Biological treatment of water is based on establishing a microbial population on a suitable substrate, such as sand, and then ensuring that ideal conditions for the microbes are constantly maintained. Several kinds of bacteria are important components of effective iron removal, including different species of the families *Chlamydo-bacteriaceae*, *Crenotrichaceae*, *Sidercapsaceae*, and *Gallionellaceae* (Degremont 1991). Organisms in these groups are generally present in ground water and their establishment on water treatment filters should be rapid.

Biological manganese removal is a more complex process than iron removal (Mouchet 1992). However, most of the organisms responsible for manganese removal are also associated with iron removal, except the most prominent iron removal genus, *Gallionella*, which exclusively uses iron. Some organisms, such as *Pseudomonas manganoxidans*, exclusively oxidize manganese. Iron and manganese removal can take place by enzymatic intracellular oxidation, by adsorption of the dissolved iron and manganese on the cell membrane followed by enzymatic oxidation, or by catalysis close to the bacterial cells. The biological iron removal process can be optimized at low oxygen and low redox potentials. In contrast, the manganese removal process generally requires ample oxygen and high redox potentials.

When iron and manganese are oxidized (Fe^{2+} to Fe^{3+} , Mn^{2+} to Mn^{3+}) energy is released. The above bacteria can use this energy source for their metabolism and some, such as *Gallionella* and *Thiobacillus*, use this energy source exclusively, while others additionally use DOC as an energy source. The oxidation of iron and manganese does not release a lot of energy and to satisfy their energy demands, the bacteria need to process a large quantity of metals. Therefore, large volumes of water can be processed in relationship to the amount of bacterial biomass allowing for rapid filter flow rates.

Precipitates are produced during biological metal removal and they form around the bacterial cells, or around their biopolymer secretions, which can look like sheaths or stalks. These precipitates are more compact and less likely to clog filters compared with the amorphous precipitates formed during conventional treatment (Mouchet 1992). It is likely that arsenic may be co-metabolized or co-precipitated in the water treatment reactors used here. Malcolm and Pirnie Inc. (1993) and Ghurye 2001) examined physical and chemical treatment technologies for arsenic removal and we would like to add some less expensive technologies to these lists of technologies. We therefore examine the potential for using biological and chemical processes interacting to remove arsenic and several other compounds from solution without the use of chemical additions.

Materials and methods

Samples around Saskatchewan were collected from rural water wells typically serving individual rural households (1,108 wells were examined). Samples for metals were preserved with nitric acid upon collection. Most samples were analyzed at the Saskatchewan Research Council Analytical Laboratories using ion coupled plasma-atomic emission spectrometry (ICP-AES). Samples that were not analyzed by ICP-AES, were analyzed by WateResearch Corp. using direct air-acetylene flame atomic absorption spectrometry. Low levels of arsenic were either determined by hydride atomic adsorption spectrometry or by ultrasonic nebulizer/ICP.

Dissolved organic carbon (DOC) analysis was carried out on filtered unpreserved samples at the Saskatchewan Research Council Analytical Laboratories using UV persulphate oxidation with IR detection. Some of the DOC analysis was carried out by the Saskatchewan Research Council, Water Quality Section or by WateResearch Corp. using UV persulfate oxidation with a phenolphthalein colour reagent.

Sulphate analysis was carried out on filtered and unpreserved samples at the Saskatchewan Research Council Analytical Laboratories either using a turbidimetric/centrifugal analyzer or more recently by ICP-AES. Some samples were analyzed at WaterResearch Corp. using a turbidimetric/spectrophotometric analysis using barium chloride.

Results

Regional arsenic levels across Saskatchewan, Canada

Seventy-five percent of wells in rural Saskatchewan have arsenic levels below 5 µg/L (Table 1). Nine percent are between 5 and 9 µg/L with 16% being above the WHO arsenic standard of 10 µg/L. The Canadian Guideline (Health Canada, 1996) for arsenic (which has been adopted by Saskatchewan) is 25 micrograms/L and 6% of the wells were above this limit. The previous arsenic limit was 50 µg/L and only 1% of the water supplies were above this limit. In Saskatchewan, therefore, arsenic has gone from being "no problem" to an emerging problem possibly affecting a quarter of the rural wells depending on where the "safe" guideline will end up. It may be noted that WHO (1993) states in terms of its provisional 10 µg/L guideline the following: Based on health criteria, the guideline value for arsenic in drinking-water would be less than 0.01 mg/L.

When biological treatment is used to remove arsenic several other compounds will be affected during the treatment notably iron, manganese, dissolved organic carbon, ammonium, oxygen and sulphide. In Figure 1 the levels of some of these compounds are plotted as a function of the arsenic level. The DOC concentration varied around 7 mg/L for all arsenic levels except for the 25 to 50 microgram level. For that arsenic category the average DOC level was 9 mg/L. Only 1% of the wells analyzed for arsenic were above 50 micrograms/L and it is difficult to ascertain from this data whether there is a positive correlation with the level of arsenic and DOC.

The iron concentrations were lowest at the lowest arsenic concentrations with iron levels at least quadrupling as soon as arsenic levels of 5 µg/L or higher were reached. If iron levels are above 2 mg/L this may therefore be a potential warning sign for the presence of arsenic in the water.

In contrast to iron the average manganese concentration was highest in the lowest level of arsenic with average levels close to 0.7 mg/L. The manganese levels fluctuate around 0.5 mg/L for wells with higher than 5 µg/L arsenic.

The sulphate levels were around 500 mg/L for arsenic levels up to 10 µg/L, but as higher levels of arsenic were reached the sulphate levels steadily climbed to almost 800 mg/L.

Some general conclusions can therefore be made in regard to the presence of arsenic in Saskatchewan well water. It does not appear to be closely associated with the level of DOC, but the levels of iron and sulphate appear to increase in concert with increased arsenic levels. In contrast, there is an inverse relationship between arsenic and manganese levels with the highest manganese levels occurring at the low-

Table 1. Arsenic levels in 1,108 wells tested across rural Saskatchewan

Arsenic concentration range	Percentage of wells
Less than 2 µg/L	63
2 to 5 µg/L	12
5 to 10 µg/L	9
10 to 25 µg/L	10
25 to 50 µg/L	5
Greater than 50 µg/L	1

est arsenic concentrations. Other factors that may possibly help in characterizing Saskatchewan's arsenic problems are presently being investigated. Compounds associated with the arsenic can potentially affect what type of treatment is most suitable and defining such issues is a prerequisite to the implementation of generalized solutions.

Biological treatment of ground water

Experimental biological water treatment units were designed during 1990-95 (Peterson et al. 1996, Corkal et al. 2001). The constructed treatment reactors were designed to deal with water quality problems encountered by rural water users when trying to make ground or surface water suitable for human consumption on-site. A major problem in surface water is the high level of dissolved organic carbon with removal of this DOC remaining the major priority. For ground water several problems were of concern and targeted for biological treatment were DOC, iron, manganese, arsenic and ammonium ions. The treatment systems were not designed to deal with some other problem compounds, such as sulphate ions. The first biological surface and ground water treatment systems were installed on individual farms, where the water supplies were not in use because of extremely poor quality mainly due to high levels of iron, manganese and dissolved organic carbon (nobody at that time had tested for arsenic). The water was considered to be aesthetically unacceptable and the farmers were trucking in water from elsewhere and filling on-site tanks. Even failure of the biological treatment systems would therefore not negatively affect the farm user. However, instead of failing, these water treatment units have been producing high quality water since 1993 (surface water) and 1995 (ground water). A photo of a biological groundwater system is shown in Figure 2.

The ground water biological water treatment system consists of several units with the first being a backwashable down-flow gravity sand filter where water to be treated is introduced directly from the well to the top of the filter. Depending on the pH and redox-potential of the incoming water, air may be passively introduced to the reactor to generate suitable redox conditions for iron-removing microorganisms, but typically low oxygen levels are beneficial for biological iron removal (Mouchet 1992). If hydrogen sulphide is present, this needs to be removed before the water enters the biological reactor, and the stronger aeration required will push the biological iron removal to become increasingly straight chemical oxidation (Mouchet 1992). Even

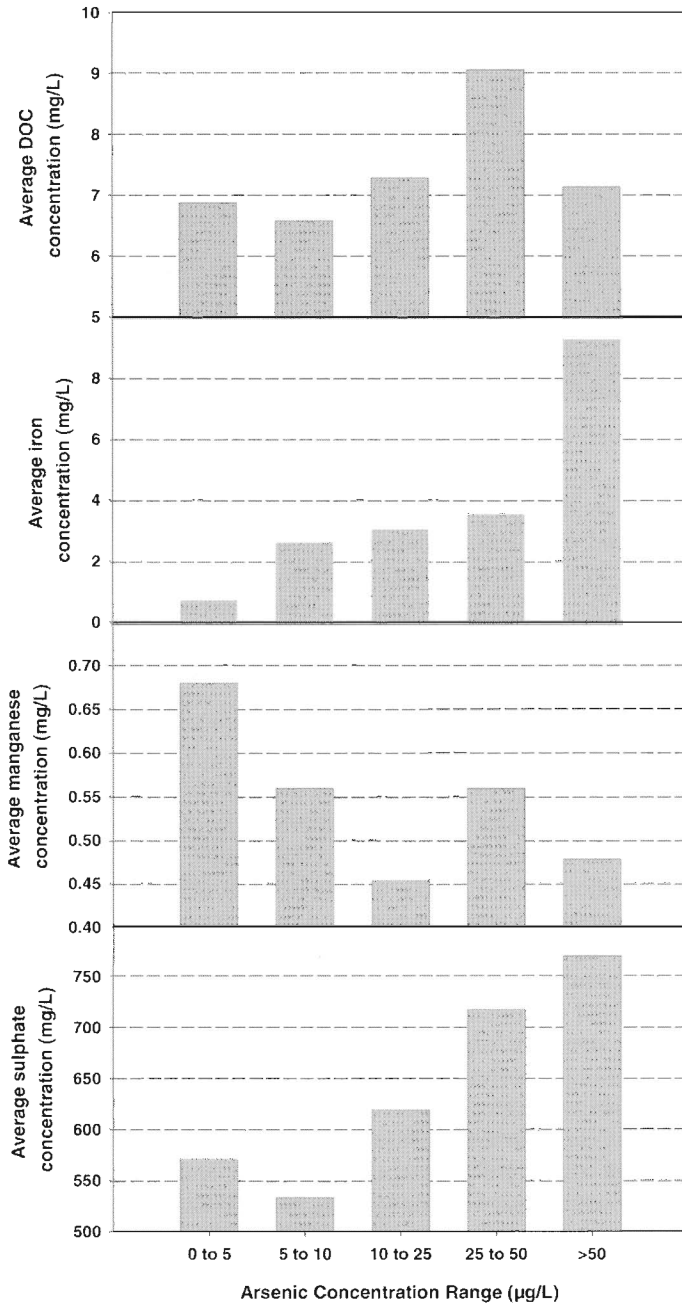


Fig. 1. DOC, iron, manganese and sulphate levels in correlation to arsenic concentrations.



Fig. 2. An experimental groundwater treatment system manufactured by Mainstream BMS Ltd.

during this process condition no chemicals (other than air) is introduced to the reactor and biological and chemical oxidation combine to remove iron and arsenic.

The contact time in this first treatment unit is typically several hours during normal consumption at the farm household. The water that has been treated by the biological sand filter is then gravity fed to the top of a Biological Active Carbon (BAC) filter similar in size to the sand filter. The BAC filter is designed to remove DOC, manganese, ammonium, and any remaining arsenic. High levels of oxygen and strongly positive redox conditions are required for the microorganisms to carry out the required functions in the BAC reactor. Water from the BAC filter is then supplied to a similarly sized water storage tank, which feeds the house-distribution system to points of use. This water is now considered acceptable for general household use and the goal is to decrease undesirable compounds, such as DOC and ammonium, so that the water can be safely disinfected. In addition, a further goal is to decrease fouling compounds, such as iron, and manganese to levels where the water is not staining clothing etc. Further, this water is now suitable for treatment with membranes as the quantity of membrane fouling components have been decreased.

Membranes are used to polish this water and make it safe for human consumption and meet the most stringent regulations anywhere in the world. The biologically treated water is feeding a series of cartridge filters followed by a reverse osmosis membrane unit typically located under the kitchen sink. A separate tap is used for this water. This way safe drinking water can be produced in a sustainable fashion with cartridge filters and membranes lasting months and years. Instead of hauling water, several rural water users are now using these water treatment systems for all household purposes including human consumption.

Data from several years are presented for one ground water system in Figure 3. The data presented show the water entering the first unit (raw water) and exiting the second unit (tap water). Membrane treated water is not shown. Iron was removed almost completely (>98%) in the biological reactors. Most arsenic was also removed with levels decreasing from around 30 $\mu\text{g/L}$ to less than 2 $\mu\text{g/L}$. DOC removal decreased with time reaching low levels of removal after a couple of years. The manganese removal rates were initially poor, but increased with time although the last

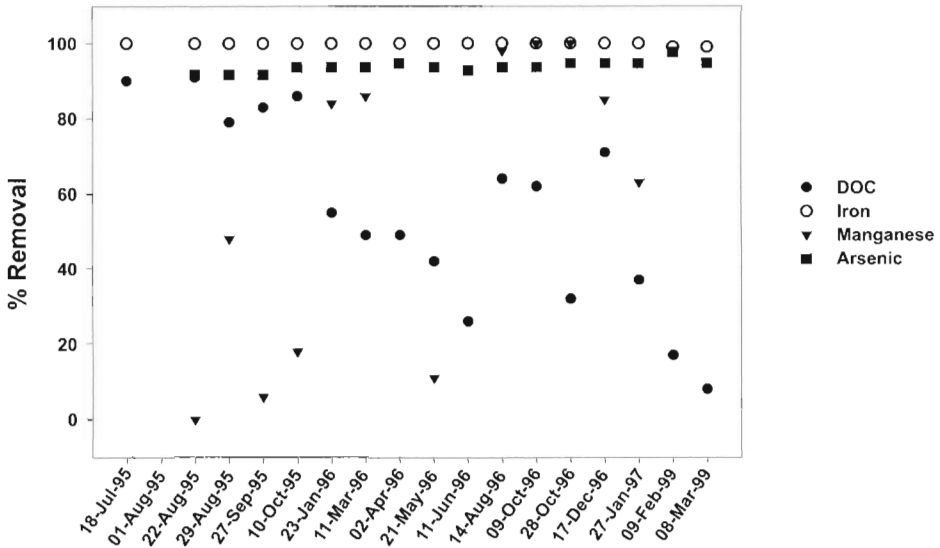


Fig. 3. Arsenic, DOC, iron and manganese removals from a biological treatment system.

few determinations again showed decreased removal rates. Work presently carried out is addressing how to establish the correct conditions for sustainable manganese removal and what alterations need to be made to have sustainable DOC removal. Other problem compounds in groundwater supplies, such as sulphate ions are not removed with the present system. Since most groundwater supplies in Saskatchewan have a high salt content requiring salt removal, it is essential to remove compounds that can foul membranes that are used to achieve salt reduction.

Discussion

While Canada maintained a 50 $\mu\text{g/L}$ guideline level for arsenic rural Saskatchewan appeared to have little concern with the arsenic levels in its drinking water. However, geological data on arsenic from the prairie region should have pointed to the likelihood of discrete areas with arsenic problems as Dudas (1987) measured arsenic levels as high as 450 mg/kg in soil from this geographic area. These arsenic levels in the soil are as high as in Bangladesh. Dudas (1987) also showed that arsenic partitioned into silt and clay fractions apparently related to the distribution of iron oxides. Hydrous ferric oxides are capable of binding both As^{3+} and As^{5+} (Cummings et al. 1999). Geological maps can be used to pinpoint areas of potential drinking water concern.

The levels of iron and sulphate appeared to increase in concert with increased arsenic levels in a survey across Saskatchewan. In contrast, there was an inverse relationship between arsenic and manganese levels with the highest manganese levels occurring at some of the lowest arsenic concentrations. At high manganese levels

both the sulphate and iron contents were lower than average. There are some interactions between arsenic and other elements that may partly explain the above. For example, sulphate can compete with As^{5+} for sites on aluminum flocs, which can decrease the amount of As^{5+} that will sorb or co-precipitate to the alum floc (McNeill and Edwards 1995). Even in the aquifer such interactions may be important and high sulphate levels may therefore force a larger proportion of the arsenic to be in dissolved form, which may partly explain the association of high sulphate and arsenic.

If the arsenic in the water is associated with high levels of iron (as it is in Saskatchewan) then formation of $\text{Fe}(\text{OH})_3$ precipitates during the oxidation of the iron is expected to remove considerable amounts of soluble As^{5+} (McNeill and Edwards 1995, 1997a,b). In contrast, the presence of high Mn concentrations at low arsenic levels is not expected to remove significant concentrations of dissolved arsenic even when this Mn is producing $\text{MnO}(\text{OH})_2$ precipitates (Edwards 1994). Indeed, McNeill and Edwards (1995) showed that significant arsenic removal was associated with significant iron removal with low arsenic removal rates if oxidation reactions were not associated with iron removal. It is therefore possible that it is easier to remove arsenic from high arsenic/high iron waters compared with low arsenic/low iron waters. The precipitation of iron and the concomitant removal of arsenic can explain part of the arsenic removals in the present study.

If a softening process resulting in the production of $\text{Mg}(\text{OH})_2$ formation is implemented then very high levels of arsenic removal can be achieved as arsenic is effectively sorbed to such solids (McNeill and Edwards 1995). In contrast, little arsenic is removed during calcite precipitation. Amy et al. (2000) also evaluated treatment options for arsenic.

In a separate study Peterson et al. (1997) used mini-columns with various contents (empty column controls, clean sand and Biological Active Sand) and passed water collected from wells through these columns using in-situ (low oxygen levels) and aerated oxygen conditions. Biological removal of both arsenic and manganese was demonstrated while clean sand actually increased the manganese concentrations although some arsenic removal was achieved (31% removal, while Biological Active Sand on average removed 74%). Under low-oxygen conditions the Biological Active Sand did not remove iron, while the clean sand removed 60%. When aerated, all columns removed iron including the glass-wool stoppered empty columns. Arsenic and manganese removals were lower for both clean sand and Biological Active Sand under aerated conditions. The interactions between physical, chemical and biological conditions are therefore not always clear-cut and defining the optimum conditions of water treatment systems will require careful selection of process conditions to take advantage of both biological and chemical removal processes.

The production of various inorganic arsenic precipitates will interact with microbial arsenic removal processes, and in the present study a high iron/high arsenic groundwater was studied. The removal of both iron and arsenic using a combination of sand/GAC biological reactors was shown to be highly effective during the 4-year study period with more than 90% removal rates. There were no additions of chemicals (other than straight aeration) throughout the treatment processes and interactions between physico-chemical and biological factors accounted for the high removals. The media in the treatment units (sand and Biological Granular Active Carbon) support the attachment of microorganisms; it will not get exhausted and was not replaced

during the study period (7 year old media). The expansion of treatment processes that take advantage of biological and naturally occurring physico-chemical removal processes eliminates the need for expensive chemicals and they can also be simple to operate. The greatest challenge in the production of safe drinking water for rural people around the world is the development of technologies that are robust, simple, effective and inexpensive. The optimization of biological treatment combined with enhancing naturally occurring physico-chemical removal processes offer a remarkable set of promises that need to be carefully evaluated and implemented.

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