

Evaluation of Treatment Systems for the Removal of Arsenic from Groundwater

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Abstract: High concentrations of iron and arsenic are common in Saskatchewan groundwater. Their cost effective removal from groundwater is a challenge for small communities. Four small treatment systems were tested for iron and arsenic removal with and without ozone pretreatment using groundwater high in iron (6.4–8.4 mg/L) and arsenic (14.5–27.2 µg/L). The systems tested were (1) coagulation, (2) biological activated carbon (BAC) filter, (3) slow sand filter, and (4) rapid sand filter. Coagulation removed 70% of arsenic and 85% of iron. With ozone pretreatment, the removal increased to 95% for arsenic and 93% for iron. The BAC system was highly effective in the removal of both arsenic and iron. On average, it removed 97% of arsenic and 99.8% of iron. With the ozone pretreatment, the average removal increased to 99% for arsenic and 99.9% for iron. The increase in iron removal with ozone was not statistically significant at $P=0.1$. The slow sand filter system was also highly effective in the removal of both arsenic and iron. On average, it removed 96% of arsenic and 99.8% of iron. The ozone pretreatment did not significantly change removal rates. The rapid sand treatment system removed an average of 50% of arsenic and 99% of iron. With the addition of ozone, the average arsenic removal increased to about 65%; however, the average iron removal was only 50%. The removal of arsenic by the rapid sand filter was extremely variable and this was possibly related to media saturation and some surge in the system as the ozone off gas valve was opened and closed.

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Introduction

Arsenic is a toxic compound present in nature. As(III) is reported to be 25–60 times more toxic than As(V) and several hundred times more toxic than methylated arsenicals (Korte and Fernando 1991). The degree of toxicity of arsenic compounds is as follows: arsine > As(III) > As(V) > methylated arsenicals (Korte and Fernando 1991). Arsenic may occur naturally (dissolution of arsenic-bearing minerals) or it may be produced by anthropogenic activities (industrial discharge: electroplating, gold mining, fossil fuels, and others). Arsenic enters into the human body through arsenic-bearing food, drink, or air. The major passage of entrance is through drinking water. Arsenic contamination of groundwater and associated health risks have been reported in many parts of the world such as Bangladesh (BGS 1999; Watanabe et al. 2001, 2003; Anwar et al. 2002; Yokota et al. 2002; Milton et al. 2003; Chowdhury et al. 2003); India (Mazumder 2003; Rahman et al.

2003); China (Zhang 2003; Lianfang and Shenling 2003), Taiwan (Guo 2003), Thailand (Choprapawon and Rodcline 1997); United States (Kim et al. 2002), Latin America (De Esparza 2003; Karschunke et al. 2000), Japan (Kondo et al. 1999), Nepal (Shrestha et al. 2003), Canada (Peterson and Sketchell 2003; Viraraghavan et al. 1999; BGS 1999).

Health implications due to ingestion of arsenic contaminated drinking water are well known as “black foot disease” in Taiwan, “Bell Ville disease” in Argentina, and “Kai Dam” in Thailand (BGS 1999). The poisoning of arsenic in human body was reported to show various health disorders such as skin and liver cancer, nervous system damage, hepatic haematological disruption, and even death for dosages ranging from 70 to 180 mg (Galvin 2003). Various physiologic effects may occur due to chronic and acute intoxication of arsenic [Agency for Toxic Substances and Disease Registry (ATSDR) 2002].

Follow-up studies of Japanese children who were exposed for a long period by consuming arsenic-contaminated milk revealed an increased incidence of severe hearing loss, mental retardation, epilepsy, and other brain damage (ASTDR 2002). Rodriguez et al. (2003) reported nervous system disturbances such as polyneuropathy, EEC abnormalities and, in extreme cases, hallucinations, disorientation, and agitation due to human exposure of arsenic. The World Health Organization (WHO) in 1993 and U.S. Environmental Protection Agency (USEPA) in 2001, therefore lowered limit of arsenic in drinking water to 10 µg/L. The interim Canadian drinking water limit for arsenic is 25 µg/L (Canadian Drinking Water Guidelines 1996).

Treatment systems such as coagulation/filtration, adsorption on activated alumina, ion exchange resin, adsorption by hydrous ferric oxides, iron oxide coated sand, iron filings, manganese green-sand filtration have been reported for arsenic removal (Joshi and

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Table 1. Characteristics of Groundwater (unit: mg/L except for arsenic, pH, and turbidity)

pH	As ($\mu\text{g/L}$)	Iron	Total alkalinity	Hardness (CaCO_3)	DOC	Na	Mg	Chloride	Sulfate	TDS	Turbidity NTU	Ca	Mn
7.2–7.5	14.5–27.2	6.4–8.4	388–393	1060–1100	4.2–4.9	243–249	114–119	335–348	739–745	2030–2110	106–136	238–251	0.93–0.99

Note: All results except for arsenic and iron are based on tests conducted at the Saskatchewan Research Council; the tests for arsenic and iron are conducted at the University of Regina.

Chaudhuri 1996; Subramanian et al. 1996; Viraraghavan et al. 1999; Karshunke et al. 2000; Khan et al. 2000; Ramaswami et al. 2001; Petrusevski et al. 2002; Thirunavukkarasu et al. 2001, 2002, 2003; Selvin et al. 2002; Mokashi and Paknikar 2002; Nikolaidos et al. 2003; Vaishya and Gupta 2003). Ion exchange, activated alumina, reverse osmosis, modified coagulation filtration, modified lime softening, electro dialysis reversal, and oxidation/filtration are considered as the best available technologies by the USEPA (2001). Iron oxide-coated sand, manganese greensand filtration, iron filings, and modified iron and granular ferric hydroxide are classified as emerging technologies by the USEPA (Thirunavukkarasu and Viraraghavan 2003). In a comparative evaluation of various adsorbents to remove arsenic, Saha et al. (2001) reported that iron oxide-coated sand, activated alumina, CalSiCo (conglomeration of calcium silicate impregnated with copper sulfate solution), and hydrous granular ferric oxide were highly efficient adsorbents. Siddiqi (2003) reported granular ferric hydroxide as the best and cost-effective technology to remove arsenic from groundwater in Bangladesh. The author claimed that this technology was already in use and accepted by the communities with satisfactory operation of the system. Souter et al. (2003) examined a new point-of-use treatment system that is based on flocculation, sedimentation, and disinfection for removal of microorganisms and arsenic. The authors reported 99.8% removal of arsenic in a laboratory experiment. All these systems require chemical pretreatment of the filter media or use chemicals in the system and are expensive in initial investment as well as operation and maintenance of the system.

There is a need to find a cheaper, reliable, and easy-to-operate technology, which is affordable by small communities. Sand filtration appears to be better than chemically assisted technologies and is a more cost-effective technology to remove arsenic from groundwater, provided the water is rich in iron content. Lipton et al. (1994) reported 99% removal of arsenic from groundwater by combined anaerobic and aerobic bioreactors. This was accomplished by adding iron in the system. Arsenic removal in conjunction with the biological oxidation of iron or manganese or both were reported (Lehimas et al. 2001; Zouboulis and Katsoyiannis 2002; Katsoyiannis et al. 2002; Katsoyiannis and Zouboulis 2004).

Biologically active filtration units developed by the Mainstream BMS Ltd., Vanscoy, Saskatchewan and Davnor Water Treatment Technologies Ltd., Calgary, Alberta, have been used in rural Saskatchewan on an experimental basis for seven years with consistent arsenic removal exceeding 90% irrespective of the seasons (Peterson and Sketchell 2003). These systems could remove more than 99% of iron and could also remove dissolved organic material. The objective of the present study was to compare the removal of arsenic and iron from groundwater using four treatment systems with and without an ozone pretreatment. The four treatment systems included those two tested by Peterson and Sketchell (2003), Davnor slow sand filter and Mainstream BAC system, as well a batch coagulation system and a rapid sand filter. These treatment systems along with an ozone water treatment system were installed in a trailer near Swift Current,

Saskatchewan, Canada. The specific objective of preozonation in this study was to examine whether oxidation of iron to iron oxide by ozone would enhance As removal through iron oxide coating of the media (Kim and Nriagu 2000; Nishimura and Umetsu 2001). This study also included speciation of arsenic to determine arsenite [As(III)] and arsenate [As(V)] concentration in the raw and certain treated waters.

Materials and Methods

The Mainstream BAC filter, Davnor slow sand filter, rapid sand filter, the Tec-Water commercial coagulation systems, and the ozone system were housed in a trailer that could be moved to various sites. No nutrients or any specific microorganism was added to any of the filter units. Table 1 shows the characteristics of the groundwater used in the study.

Mainstream Biological Activated Carbon Filter

This system contained three tanks, the first two contained filter media and the third tank was a treated water storage tank. The first tank was a slow sand filter 660 mm in diameter and 1740 mm high with a filter depth of 1060 mm. The second tank was 450 mm in diameter, 1740 mm high with 965 mm of granular activated carbon (GAC), 0.5 mm in mean size. The two filters were continuously aerated to enhance the growth of biological activity within the filter media. The last tank was a storage tank for the treated water.

The source water, either raw or ozonated, passed through the slow sand filter and, then the GAC filter to the storage tank by gravity. The system was operated at 4.5 L/min (1 Imp. gal/min). The empty bed contact times of the sand filter and the carbon filter were found to be 80.6 and 34.1 min, respectively. The particle size distribution of the sand filter ranged from 0.2 to 1 mm. The sand filter was backwashed once a month whereas the carbon filter was backwashed twice a year. Backwashing was carried out by purging air through the bottom and expanding the media followed by rinsing with water.

Davnor Slow Sand Filter

The system consisted of three tanks; the source water was sprayed into an elevated tank, which aerated the water and oxidized some of the iron and organic matter. The first tank was 560 mm in internal diameter with a height of 850 mm. A water level of 670 mm was maintained in this tank. From the elevated tank the water flowed by gravity to the second tank which contained the filter media. The second tank was 914 mm high with an internal diameter of 620 mm. In the second tank, the media consisted of 6 mm \times 3 mm subgravel on the bottom followed by 12 mm crushed quartz and then topped off with 8 mm crushed quartz followed by sand of size 0.2–1 mm. The total depth of the filter media was 550 mm and a constant water head of 230 mm was maintained in this tank. Any oxidized iron particles that did not

settle out in the elevated tank (first tank) was removed as the water slowly moved through the filter media. At the bottom of the tank was a drainpipe that transported the treated water to a storage tank. After a backwash cycle the system was able to produce about 4.5 L/min. After about 4,500 L of water, the media started to plug up and the outflow from the system was reduced. The empty bed contact time of this filter was 36.9 min and the filter was backwashed at least every two days during the trials.

Rapid Sand Filter

The rapid sand filter unit (Model 1044) used in this project was manufactured by Waterrite Inc., Winnipeg, Manitoba. The filter was 1180 mm high and 260 mm in internal diameter. The media depth was 864 mm. The average flow rate of the system was 4.5 L/min and the empty bed contact time of the filter was 10.2 min.

Commercial Coagulation System

Coagulation was accomplished using a Floc System 100, manufactured by Tec-Water Inc., Tisdale, Saskatchewan. It was a point of entry (POE) coagulation system, which can be used to treat domestic water from surface water sources. This system consisted of a pressure tank and pump, to transport water from an external storage tank into 450 L settling tank. As the water was pumped into the settling tank a coagulant chemical was injected in line. Mixing occurred as the water enters the settling tank. Polyaluminum chloride (brand name Clear Pac) was used as a coagulant with a dose of 8.56 mg Al/L. Mixing continued until the tank was full. The momentum created during the filling process continued to mix the water in a swirling fashion for an additional 10 min. Once the initial filling was complete, the water was allowed to settle for 3 h. After 3 h, all the water except the bottom 0.3 m was transferred by pump to a storage tank. Once the transfer was completed, raw water was injected into the settling tank to agitate the sludge in the bottom 0.3 m of the tank and pumped into a sewer system for disposal. The system was able to provide approximately 27,000 L of good quality water over a 24 h period.

Ozone Generator

The ozone generator used in the treatment process was an Azcozon VMUS-4, silent pulse injection corona discharge unit manufactured by Azco Industries Ltd., Surrey, B.C. The ozone production was 4 g/h from dry air at 5 L/min air flow and an ozone dosage of 6.2–8.5 mg ozone/L of water was applied during ozonation.

Sampling, Arsenic Analysis, and Speciation

Five/six samples of raw water and treated water at various stages of treatment were collected. Samples were preserved in the field with 5 drops of nitric acid (EMD, Canada). The pH values of the acidified samples were found to vary from 2.15 to 3.7 which were within the pH ranges (1.8–5.0) mentioned by Edwards et al. (1998) for arsenic speciation. The acidified samples were filtered using 0.4 μm polycarbonate filters (Osmonics Inc). Arsenic in the filtered water samples was speciated in to As(III) and As(V) [Thirunavukkarasu et al. 2001]. They found that nitric acid did not affect speciation; similar recoveries of As(III) were found for samples acidified with H_2SO_4 and HNO_3 . Resins in chloride form were obtained from Sigma-Aldrich, Inc., USA, converted into

acetate form and 19.5 g was packed into a chromatography column, 1.5 cm in diameter and 10 cm in height. To determine the breakthrough volume, a 30 $\mu\text{g/L}$ As(V) aqueous solution was prepared and injected by syringe through the resin-packed chromatography column at a flow rate of 10 mL/min. The breakthrough volume was found to be 380 mL.

The water samples were analyzed for arsenic then 10 mL of raw water was passed through a clean dry chromatography column (Sigma-Aldrich, Inc.) packed with 19.5 g of resin in the acetate form. Arsenic (V) in the water was obtained by the difference between the total soluble arsenic (before passing through the column) and the arsenic in the filtrate (arsenite).

Samples were analyzed for arsenic by graphite furnace atomic absorption spectrometer (GFAAS) [Varian type SpectrAA-600 Zeeman GFAAS equipped with GTA 100-graphite tube atomizer and PSD-00 sample dispenser]. Iron was analyzed by Varian AA10 atomic absorption spectrometer using an air-acetylene flame. However, samples having low iron concentration (below 1 mg/L) were diluted and were analyzed by GFAAS as the accuracy of the flame atomic absorption spectrometer was not high for low concentrations of iron. In order to understand the iron oxidation and subsequent coating of the iron oxide to the filter media, 1 g each of the media from Mainstream BAC (sand and activated carbon) and Davnor filter (sand) was digested in 50 mL of 10% HNO_3 solution (AWWARF 1993) and the solution was made up to 1 L after digestion and was analyzed for iron.

Results and Discussion

General

Table 2 provides the arsenic and iron profiles through the various treatment systems, respectively, based on mean values. In general, all treatments reduced the concentrations of arsenic and iron; pretreatment using ozone improved removal. The BAC and the slow sand filter (Davnor) were so effective in arsenic removal, the incremental benefit of ozone was not always evident.

The arsenic content in the raw water collected at different times showed varying results. The water samples of raw water used in the coagulation treatment system were found to contain relatively higher (35% higher) arsenic than the samples collected during the operation of other treatment systems. This may be related to the withdrawal rate from the well. With higher and more frequent pumping, water from a greater depth could be drawn from the aquifer. However, the iron concentration in the raw water was quite consistent. The variable arsenic concentration may be the result of multiple factors including pumping rates, pumping duration, sampling procedure, or laboratory tests.

Coagulation was effective in removing an average of 70% of the arsenic and 85% of the iron. When ozone was added as a pretreatment, the average removal increased to 95% of the arsenic and 93% of the iron. Ozone is a strong oxidant known to oxidize metals such as arsenic, iron, and manganese converting them into a particulate forms. Speciation of the arsenic showed that coagulation was highly effective in the removal of As(V) and removed close to 100% of the As(V) with or without ozone. The addition of ozone enhanced the As(III) removal from approximately 50 to 90%. It was likely that As(III) may have been partially oxidized to As(V) by ozonation increasing its removal rate.

The BAC system was highly effective in removing both arsenic and iron. Arsenic removal was about 97% and iron removal was about 99.8%. The addition of ozone improved removal

Table 2. Arsenic and Iron Removal by Various Treatment Systems [Mean Value \pm SD:^a As(μ g/L) and Iron (mg/L)]

Treatment system	Sample description	No. of samples	Total arsenic (soluble)	As(III) (soluble)	As(V) (soluble)	Iron (soluble)
Coagulation	Raw water	5	24.3 \pm 4.1	13.5 \pm 1.5	10.8 \pm 5.1	8.3 \pm 0.1
	Post coag.-no ozone	5	7.2 \pm 0.9	7.2 \pm 0.6	0.3 \pm 0.5	1.3 \pm 0.1
	Post coag, preozonation	5	1.1 \pm 0.4	1.2 \pm 0.4	0.0 \pm 0.1	0.6 \pm 0.0
Mainstream (BAC)	Raw water	6	17.4 \pm 1.7	11.6 \pm 1.5	5.8 \pm 2.2	7.9 \pm 0.7
	Post BAC-no ozone	6	0.7 \pm 0.5	0.6 \pm 0.6	0.2 \pm 0.2	0.0 \pm 0.0
	Post BAC, preozonation	6	0.2 \pm 0.1	0.1 \pm 0.2	0.2 \pm 0.3	0.0 \pm 0.0
Slow sand filter (Davnor)	Raw water	6	17.4 \pm 1.7	11.6 \pm 1.5	5.8 \pm 2.2	7.9 \pm 0.7
	Post Davnor-no ozone	6	0.8 \pm 0.5	0.8 \pm 0.6	0.1 \pm 0.2	0.0 \pm 0.0
	Post Davnor, preozonation	6	0.7 \pm 0.4	0.9 \pm 0.3	0.0 \pm 0.0	0.0 \pm 0.0
Rapid sand filter	Raw water	6	18.2 \pm 0.3	12.4 \pm 2.2	5.9 \pm 2.2	8.3 \pm 0.0
	Post rapid sand-no ozone	6	8.6 \pm 0.5	5.4 \pm 1.1	3.1 \pm 0.7	0.0 \pm 0.0
	Post rapid sand, preozonation	6	6.6 \pm 5.6	2.1 \pm 0.5	5.0 \pm 5.1	4.2 \pm 1.6

Note: All tests conducted on field acidified and lab filtered samples.

^aSD=standard deviation.

slightly to an average of 99% for arsenic and 99.9% for iron. Statistical analysis using the t-test indicated that the pretreatment of ozone did not significantly improve iron removals at the 90% confidence level. As(III) removal increased from about 95 to 100% with the pretreatment of ozone. The t-test indicated that additional As(III) removal with ozone was significant at a 90% confidence level. The significant increase in As(III) removal may be due to (1) its oxidation to As(V) and (2) increased contact time through a two-filter system. The iron content in the sand filter was found to be 2.5 mg/g of sand whereas iron in the activated carbon filter was found to be 1.3 mg/g of activated carbon.

Arsenic and iron removals with the slow sand filter (Davnor unit) were high. Removals of arsenic and iron were 96 and 99.8%, respectively. Pretreatment by ozone did not show a significant improvement. As with the BAC system, removal of As(V) was essentially complete. Removal of As(III) was about 93% with and without ozone pretreatment. Ozone was expected to transform As(III) to As(V), however, this may not have occurred because of possible low dosage of ozone. Further tests are needed to understand the role of ozone prior to slow sand filtration. Slow sand filters were efficient in removing arsenic. The quantity of the iron in the filter media was found to be 1.4 mg/g of sand.

The rapid sand filter removed approximately 50% of arsenic and 99% of iron. There was no iron in the filtrate leading to possible iron oxide coating of the media. No measurement of iron in the sand was made. The low arsenic removal in this case may be due to the short contact time (only 10.2 min). With the addition of ozone, the resulting data showed high variability and were not considered fit for analysis. The possible reasons for the fluctuation in treatment levels included breakthrough of the filter, fluctuating water flow rate from the opening and closing of the ozone offgas valve and possibly improper sampling technique. The samples were taken with a larger container and poured into sample bottles. Some of the arsenic and iron could have settled more in some samples than others. The data for the rapid sand filter treatment without ozone showed consistent results. The As(V) removal was only about 47%, compared with close to 100% for the BAC and the slow sand filter.

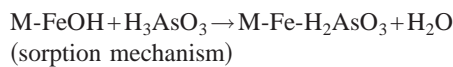
Approximately 63% of the arsenic content in raw water was

dominated by As(III). It is common that groundwaters are often dominated by As(III) because of anoxic conditions.

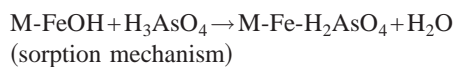
Possible Arsenic Removal Mechanism

The oxidation of iron can be carried out by either aeration, chemical oxidation or by biological means. It was reported that biological oxidation of iron by iron oxidizing bacteria oxidizes iron at a rate of 50×10^3 times faster than chemical oxidation of iron (Sharmin 2001). Biological treatment of iron and manganese or both from drinking water was reported by many authors (Bergel 1999; Borggine et al. 1994; Mouchet 1992; Hatva 1989). Lehimas et al. (2001) reported that arsenic was reduced below 50 μ g/L from an initial arsenic concentration of 400 μ g/L from a groundwater rich in iron content using a sand filtration system with an application of iron oxidizing bacteria (*Gallionella*). The filtration rate of the system was between 3 and 4 m/h. Iron was almost eliminated in the final effluent (<0.01 mg/L). Arsenic removal from the groundwater in conjunction with biological oxidation of iron was also reported by some other authors (Zouboulis and Katsoyiannis 2002; Katsoyiannis and Zouboulis 2004). These authors used polystyrene beads as the filtration and biological growth media and iron oxidizing bacteria *Gallionella ferruginea* and *Leptothrix ochracea* were used to oxidize iron present in the groundwater. The authors indicated that As(V) was easily removed compared to As(III). Maintaining redox potential greater than 300 mV was important in order to achieve greater than 90% As(III) removal efficiency. The presence of iron may have played a great role with removal of arsenic in all cases of filtration in these studies. Iron may have been biologically or chemically oxidized to iron oxide and coated to the media and the arsenic may have been adsorbed on the iron-coated media. Katsoyiannis et al. (2002) reported iron and manganese oxidation by biological or physicochemical means and the coating of these oxides in the filter media played an important role to adsorb arsenic from the groundwater. The authors also reported that oxidation of As(III) to

As(V), partially increased the arsenic removal efficiency. The arsenic chemistry shows that As(III) exists in aqueous solution in H_3AsO_3 form and As(V) in H_3AsO_4 . It is believed that following mechanism enhances the process of arsenic removal from the groundwater during biological oxidation (Katsoyiannis et al. 2002; Zouboulis and Katsoyiannis 2002; Katsoyiannis and Zouboulis 2004)



and arsenite is oxidized partially by biological means



where "M" stands for media.

The mechanism of arsenic removal in coagulation may also be enmeshing of arsenic in the floc. Fan et al. (2003) examined As(V) removal by various coagulants. The authors showed that polymeric coagulants have better arsenic removal capacity than conventional coagulants. In an initial pH of 5.5 and a coagulant dosage of 70 μM Al/L, polymeric aluminum chloride, polymeric aluminum sulfate, aluminum chloride, and aluminum sulfate removed 89.2, 88.2, 84, and 82.8%, respectively, of arsenic by coagulation and sedimentation process in lab-scale experiments. In the present study, polyaluminum chloride was used with a dose of 8.56 mg Al/L and it resulted in 70% removal of arsenic without preozonation.

Conclusions

The following conclusions can be drawn from the study:

1. The Mainstream BAC and the Davnor slow sand filter were very efficient in the removal of iron and arsenic. The effect of preozonation could not be interpreted clearly in these two systems as the values of iron and arsenic with and without preozonation are very low and similar.
2. Speciation of arsenic indicated that essentially all As(V) was removed with coagulation and both biological systems. Addition of an ozone pretreatment significantly improved the As(III) removal for coagulation. The Mainstream BAC system showed some benefit with an ozone pretreatment in removal of As(III), however, no benefit was realized with the Davonor slow sand filter system.
3. Further studies are needed to identify the optimum ratio of iron to arsenic for arsenic removal; additional studies are needed to examine whether the addition of iron oxidizing bacteria would enhance arsenic removal in biological systems.

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