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ARSENIC REMOVAL BY SAND FILTRATION FOR
POTABLE WATER IN NEWFOUNDLAND

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August 2012



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FINAL REPORT

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IN NEWFOUNDLAND**

Submitted By,

Mr. Danial Bin Rohail and Dr. Cynthia Coles

27 August 2012

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Research and Methods

Introduction

The purpose of this research was to study sand filtration as a treatment technology for drinking water sources in Newfoundland and Labrador, so as to reduce the arsenic concentration to the level of $7\mu\text{g/L}$ without using chemicals. Further, the effect of various ions present in groundwater have on the arsenic removal efficiency of sand filters, was also investigated. Owing to the carcinogenic effects of arsenic, Health and Medical Research Council (2003), NHMRC, of Australia sets the maximum acceptable concentration (MAC) for Arsenic in drinking water as $7\mu\text{g/L}$. While, as per Health Canada guidelines, the MAC for arsenic is $10\mu\text{g/L}$. Due to the carcinogenic effects of arsenic, we have followed more stringent guidelines in this study.

Water Sampling

The iron to arsenic ratio (wt/wt) is the most important parameter in successfully removing arsenic from groundwater to the level below an acceptable concentration (Berg et al. 2006). In this study water sampling locations were narrowed down based on the composition of groundwater provided by the Department of Environment and Conservation, Newfoundland and Labrador (courtesy of Mr. Keith Guzzwell). Two different water samples were collected from the town of the Wabana on Bell Island. One with the high arsenic, As and iron, Fe concentrations and second was the Wabana normal supply water. Approximately 120 litres of water sample with high As and Fe was collected, whereas, 60 litres of normal supply water was collected. Similarly, 120 litres of water sample was collected from the Town of Freshwater in Carbonear. The composition of the water samples (arsenic, iron and other element concentrations) was determined using Inductively Coupled Plasma Mass Spectrometry (ICPMS) from the Department of Earth Sciences at the Memorial University of Newfoundland. (*Please see Appendix-A for composition of the water samples*).

Sand

Since the aim of this research project was to use the locally available materials to treat the groundwater, hence, locally available sand manufacturers were contacted. M/s Capital Ready Mix supplied the washed sand required for this project. (*Please see Appendix-B for a Sieve Analysis of Sand*). The sand had a finesse modulus, FM, of 2.9. The fineness modulus or FM is defined as the characteristic coefficient obtained by adding percentages of the sample retained on

the specified series of sieves divided by 100. The sieve sizes are: 0.16 – 0.315 – 0.63 – 1.25 – 2.5 – 5 – 10 – 20 – 40 and 80. “The fineness modulus allows to appreciate the quantity of fine elements contained in the sand” (Kurtz, 2004). The Canadian Standard Association, CSA standards requires the FM between 2.3 and 3.1.

Test Equipment

Batch Column tests were conducted for treating the arsenic contaminated water. The test equipment was manufactured by Technical Services in the Faculty of Engineering and Applied Sciences. Two columns of dimensions 6.7cm x 14cm (small) and 12.5cm x 24cm (large) (Dia. X Length) were fabricated to be used in this research. (*Please refer to Figure 1 for details of the Apparatus*).



Figure 1: Experimental Setup

Experiment Design

i) Sand Preparation:

The sand for all the experiments was first washed with 60 °C hot distilled water to dissolve all the impurities. The water was then drained to collect the washed sand. Further, the sand was dried in the oven at 105°C for 24 hours to remove all the moisture.

ii) Column Preparation:

The height of the sand column was kept at 6.7cm equivalent to the diameter of the smaller diameter column. The columns were partially filled with the washed and dried sand to the height of 6.7 cm. In order to uniformly distribute the water along the whole cross section and to control the flow of solution into the column, either a Ceramic disk or the cloth was used. The ceramic disk had an approximate porosity of 50% by volume, pore size of 6µm, and hydraulic conductivity of 5.11×10^{-5} . The cloth was a mixture of 55% cotton and 45% polyester and had a thread count of 180. Similarly, at the bottom of the column, either the ceramic disk or cloth was used. The purpose of using either the ceramic disc or the cloth at the bottom of the column was to allow the flow of the solution out of the column and to retain the sand. One layer of cloth was used at the top to make sure that all the precipitates, if any, passed through it and should not be retained on the cloth; while, three layers of cloth were used at the bottom to stop the passage of sand particles through the pores of cloth. 1000mL of distilled water was passed through the column prior to commencing the batch tests to remove all the very fine particles which results in turbidity. Once the clear water was collected at the bottom of the column, only then the tests were initiated.

Experimentation

Various tests that were conducted during the course of this study are tabulated below in the Table-1. The run-out test is defined as the minimum volume of the water sample that can be treated through the sand filled column without exceeding the limit of 7µg/L of arsenic in the effluent water defined for this study.

Table 1: Type of Column Test

S/No.	Type of Test	Purpose
1	Test conducted using ceramic and cotton cloth at the top and bottom of the columns	To determine the effect of ceramic disk and the cloth on the Arsenic removal and to find out whether ceramic plate or the cloth is better suited to be used at the top and bottom of the columns
2	Run Out 1 test (RO-1)	To determine the amount of high iron and arsenic Wabana water that can be passed through the column without exceeding the limit of 7 µg/L.
3	Run Out 2 test (RO-2)	To study the boundary layer effects i.e. either the larger column (12.5cm x 24cm) is better or the smaller column of size 6.7cm x 14cm
4	Run Out 3 test (RO-3)	To study the effect of aeration on the arsenic removal efficiency from the high iron and arsenic Wabana water of the sand
5	Run Out 3(b) test (RO-3(b))	To study the effect of uniformity coefficient of the sand on the arsenic removal efficiency
6	Run Out 4 (RO-4) and Run Out 4(b) (RO-4(b)) test	Effect of dilution and aeration on the arsenic removal efficiency from the Wabana water of the sand. RO-4 includes one to one ratio of high iron and arsenic Wabana water and normal Wabana water. While, RO-4(b) includes one to four ratio of high arsenic and iron Wabana water to normal Wabana water
7	Run Out 5 test (RO-5)	To determine if removing the top layer of the sand, after run-out, would revive the arsenic removal efficiency of the sand
8	Freshwater	Run out test was conducted for the Freshwater water sample to determine the amount of water that could be passed without exceeding the limit of 7µg/L
9	Breakthrough Test	Breakthrough test for the Wabana water, Mix-1, Mix-2 and Mix-3.
10	Bulk Density and the Pore Volume	To determine the Bulk density and Pore Volume of the sand

Bulk Density and Pore Volume Measurement

a) Bulk Density

Bulk density is defined as the “weight per volume in the graduated cylinder including both the particulate volume and the pore volume” (Gad, 2008). The bulk density of the sand was measured using the standard ASTM test method for bulk density measurement i.e. ASTM-D4531-86. The beaker of a known volume i.e. 40(cm³), weighing 26.4422(g), was filled with wet sand and left in an oven for 48 hours at 105°C. The sample was taken out of the oven after 48 hours and weight was calculated to be 85.5642(g). The sand after drying in oven is known as the dry sand. The weight of the dry sand was calculated to be 59.122(g). Bulk Density can be calculated using the following equation:

$$\rho_b \left(\frac{g}{cm^3} \right) = m_{sand}(g) \div V_T(cm^3) \quad \text{Eq-1}$$

Where, ρ_b is the bulk density and m_{sand} is the mass of the dry sand. V_T is the total volume and it is defined as the sum of the volume of the solids or sand (V_S) and volume of the Voids (V_V). Using equation-1, the Bulk Density for the sand was calculated to be 1.47 g/cm³.

b) Pore Volume Measurement

Pore Volume is defined as “Pore Space”. And pore space or porosity is defined as follows: “The portion of soil bulk volume occupied by soil pores”(Kirkham, 2005). In order to calculate the pore volume three containers of known volume (12 cm³) and weight were used. They were filled with dry sand and their weights were recorded. The density of the sand was taken as 2.65g/cm³. The density and resultantly the volume of the sand occupying the container were calculated using the following relation:

$$V_S(cm^3) = m_{sand}(g) \div \rho \left(\frac{g}{cm^3} \right) \quad \text{Eq-2}$$

Where, m_{sand} is the mass and ρ is the density of the sand. The mass of the sand is calculated as

$$m_{sand}(g) = m_{sand \text{ with container}}(g) - m_{container}(g) \quad \text{Eq-3}$$

Pore volume is the difference in the volume calculated using above equation (equation-2) and the volume of cylinder. The volume can also be expressed as the percentage (Table-2)

Table 2: Calculation of Pore Volume

	6.559		27.588		21.029		7.935		0.3387 (33.87%)	
mass of container (g)	6.661	mass of sand and Container (g)	28.108	Mass of sand (g)	21.447	Volume of sand (cm ³)	8.093	Porosity (Fraction or %age)	0.3255 (32.55%)	Average 0.34183 (34.183%)
	6.586		29.898		20.312		7.664		0.3612 (36.12 %)	

Results and Discussions

a) Preliminary Tests

The results have been summarized below for the experiments that had been conducted during this study. All the tests were conducted using the small columns except for the RO-2 test. This test was conducted to compare the efficiency of large and small diameter columns. The details are provided later in the report. Gravity filtration was used for all the tests except for the breakthrough tests for which the vacuum pump was used owing to the large amount of water that was required to pass through the sand filled columns. Gravity filtration could have taken months to treat the same volume of water that was filtered in weeks using the vacuum pump.

Two samples were collected from the town of Wabana on Bell Island i.e. one with high arsenic (As) and iron (Fe) content (base water sample) and one from the normal water supply distribution network. The water samples were tested and the arsenic and iron contents for the base sample i.e. sample with high iron and arsenic content were found out to be 62.91µg/L and 11825.84µg/L, respectively (Figure2). While, the arsenic and iron concentration for Wabana normal supply water was 4.50 and 86.33, respectively (Figure2).

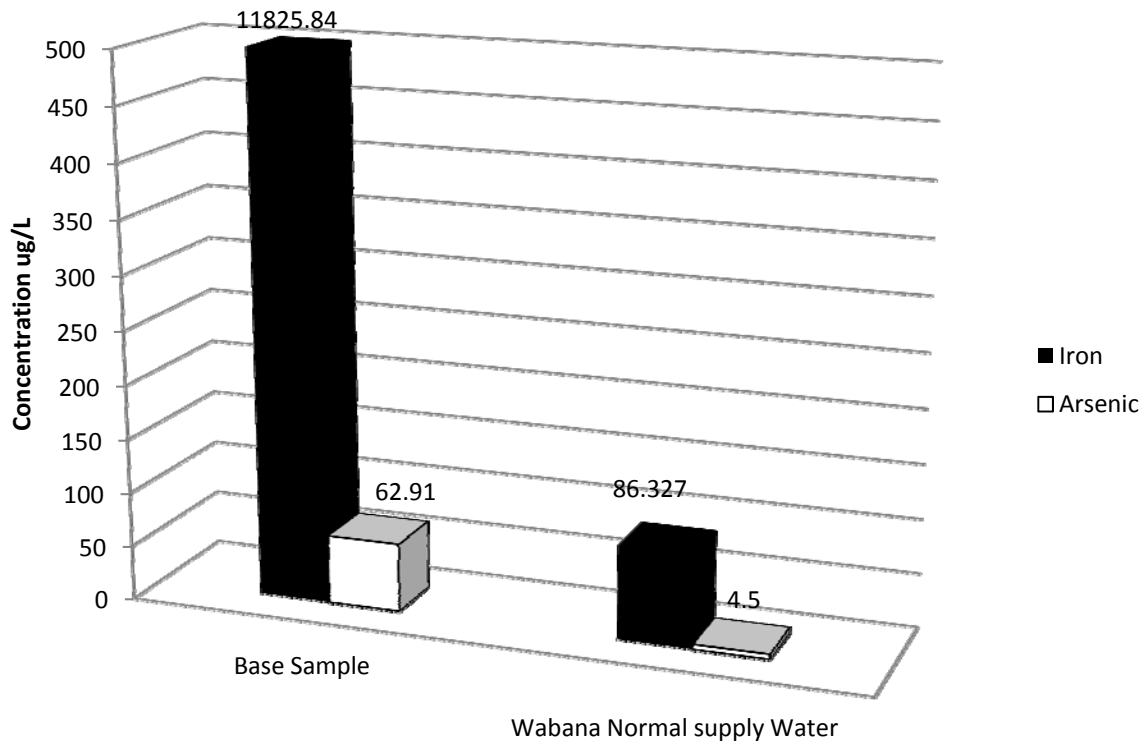


Figure 2: Results for Wabana's Base water (High Fe and As content) and Normal Supply Water (Potable Water) Sample

Two tests were carried out with the base water sample. In one test, the small column was filled with sand and ceramic discs were used as a retainer at the top and bottom of the column. In the second test, only ceramic discs were used and column was not filled with the sand (Blank). The purpose of this test was to determine if the ceramic discs were interfering with the results or not. 200 mL of base water sample was passed through both the columns and the samples were analyzed. The ceramic discs were found to be interfering with the arsenic adsorption as the arsenic concentration for the base sample reduced from 62.91 $\mu\text{g/L}$ to 28.49 $\mu\text{g/L}$ (Figure3) for the column in which no sand was used. Although, the result for the column filled with the sand and the ceramic discs was satisfactory as the iron and arsenic concentration reduced to 0 and 0.75 $\mu\text{g/L}$ respectively (Figure3). However, this orientation could not be used for later experiments. Ceramic disc was found out to be interfering with the removal of arsenic and this was not the objective of this study. Hence, it was decided to use the cotton cloth (180 Thread Count) in place of a ceramic plate as it had larger pore size and cotton bags filled with sand are commonly found in the water treatment applications. This orientation would give more realistic results as compared to the one that had the ceramic discs.

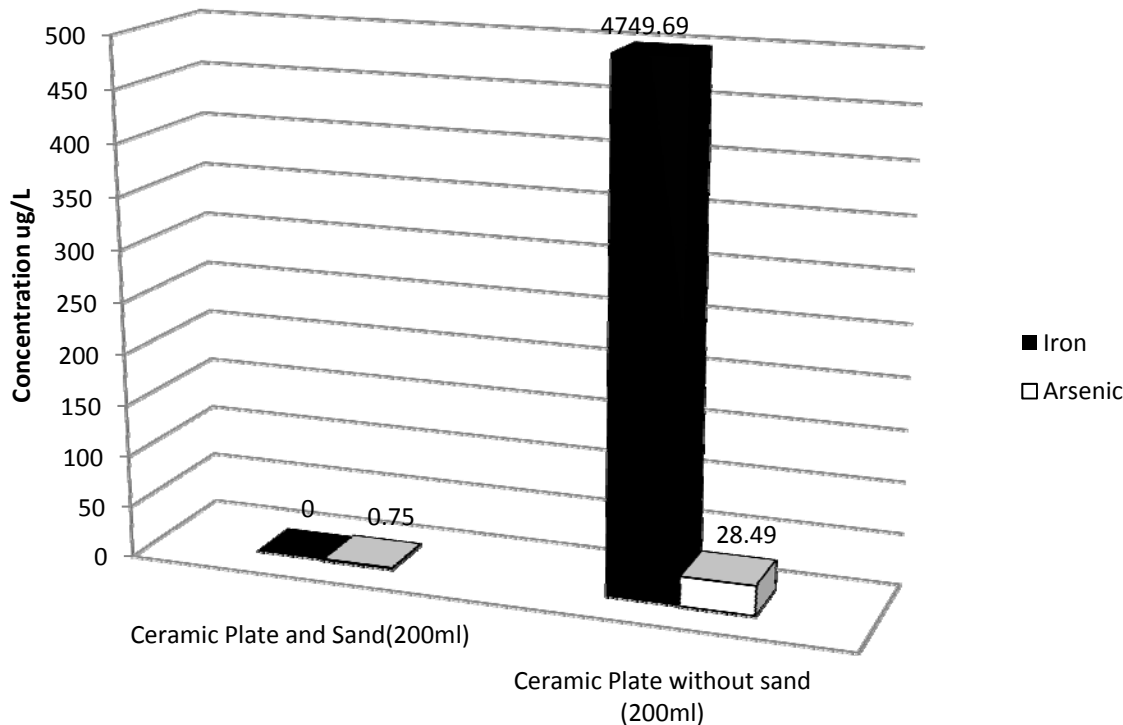


Figure 3: Effect of using Ceramic Plates on top and bottom of the Column as a sand retainer; one filled with sand and the other is without sand.

After the issue of interference of ceramic plates with the experimental results was resolved, fresh tests were conducted with the cotton cloth as the flow distributor and sand retainer in the column. 200mL of high arsenic and iron Wabana water was passed through the small column and the sample was submitted for analysis to determine how the sand was aiding in arsenic removal. The arsenic concentration in the effluent after passing 200mL through the partially filled column (height of sand column was 6.7cm), was lowered to 0.607 μ g/L (Cotton with sand (200mL), Figure 4). The next step was to determine the volume of water that could be treated with the small column that contains 246.91 cm³ of sand. It was determined that 1400 mL ((RO-7), Figure 4) could easily be treated to keep the arsenic level close to the limit of 7 μ g/L. When a total of 2000 mL of water was passed through the small column, the arsenic concentration spiked to 19.73 μ g/L ((RO-10), Figure 4). The deposition of high iron contents on the top layer of the sand might have restricted the flow of water and water might have passed through the space between column wall and sand boundary. Further, the lack of aeration in the column might have restricted the oxidation of Fe²⁺ to Fe³⁺ ions that adsorbed the arsenic (Jovanovic et al., 2011).

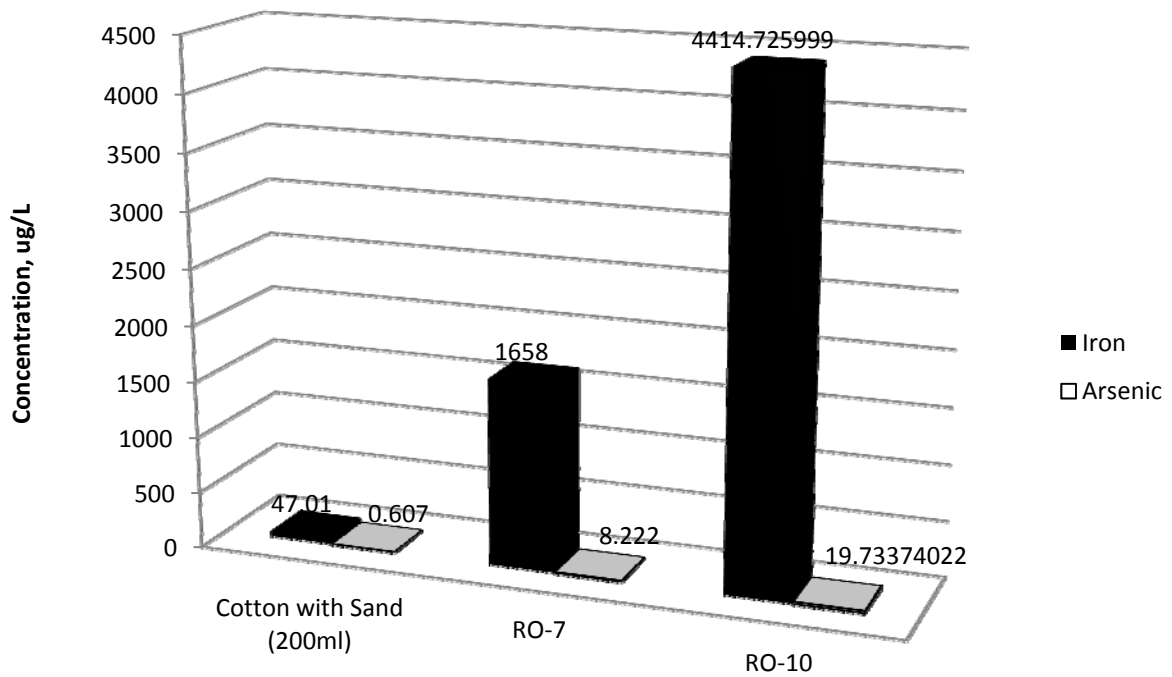


Figure 4: Run out Test 1 using cotton as a retainer (200 mL, RO-7 and RO-10 represents the Fe and As concentration determined in the treated water after passing 200mL 1400mL and 2000mL of water containing 11825.84 $\mu\text{g/L}$ of Fe and 62.91 $\mu\text{g/L}$ of As)

Next, it was decided to study the effect of column diameter on the arsenic removal efficiency of the column that was partially filled with sand. Hence, two tests were simultaneously conducted with small and large diameter columns. Both the columns were filled with sand to the same height i.e. $\sim 6.7\text{cm}$. The volume of sand in the large column was 834.33cm^3 and that occupied by the sand in small column was 246.91cm^3 . In order to compare and contrast the results, the liquid to sand ratio was kept same for both the columns i.e. 5.7:1. It was determined by the preliminary experiments that the small column could treat 1400 mL of water to keep the arsenic concentration in close approximation to the limit of $7\mu\text{g/L}$ (RO-7, Figure 4) which gave the liquid to sand ratio of 5.7:1. A total volume of 1400 mL was passed through the small column; whereas, 4730 mL was passed through the large column. The samples, RO2-S7 (Figure 5) corresponding to the volume of 1400 mL for small diameter column and RO2-L4730 (Figure 5) was corresponding to the volume of 4730 mL for large column. The small column performed better than the large column as the arsenic concentration was lowered to $4.475\mu\text{g/L}$ as compared to $5.827\mu\text{g/L}$ for large column. Further, the filtration rate through large column was very slow as compared to the small column. The higher concentration for large column could have meant that the solution concentrated around the middle of the column and the full diameter was not utilized

and hence accumulation around the centre slowed down the filtration. Hence, it was decided to use a small column with cotton as a retainer for the remainder of this study. It is pertinent to mention here that the difference in iron and arsenic concentrations in water collected at the downstream of filter after treating 1400 mL of water for RO-1 test (As: 8.22 $\mu\text{g/L}$, Fe: 1658 $\mu\text{g/L}$, Figure 4) and RO-2 test (As: 4.475 $\mu\text{g/L}$ and Fe: 1112 $\mu\text{g/L}$, Figure 5) was due to the change in the composition of the Wabana water sample with time as a result of oxidation of soluble Fe^{2+} ions into insoluble Fe^{3+} ions. These Fe^{3+} ions adsorbed the arsenic and settled at the bottom of the sample container due to which the concentration of the arsenic in the sample changed. The initial arsenic concentration dropped from 62.91 $\mu\text{g/L}$ for RO-1 test (Figure 4) to 42.906 $\mu\text{g/L}$ for RO-2 test.

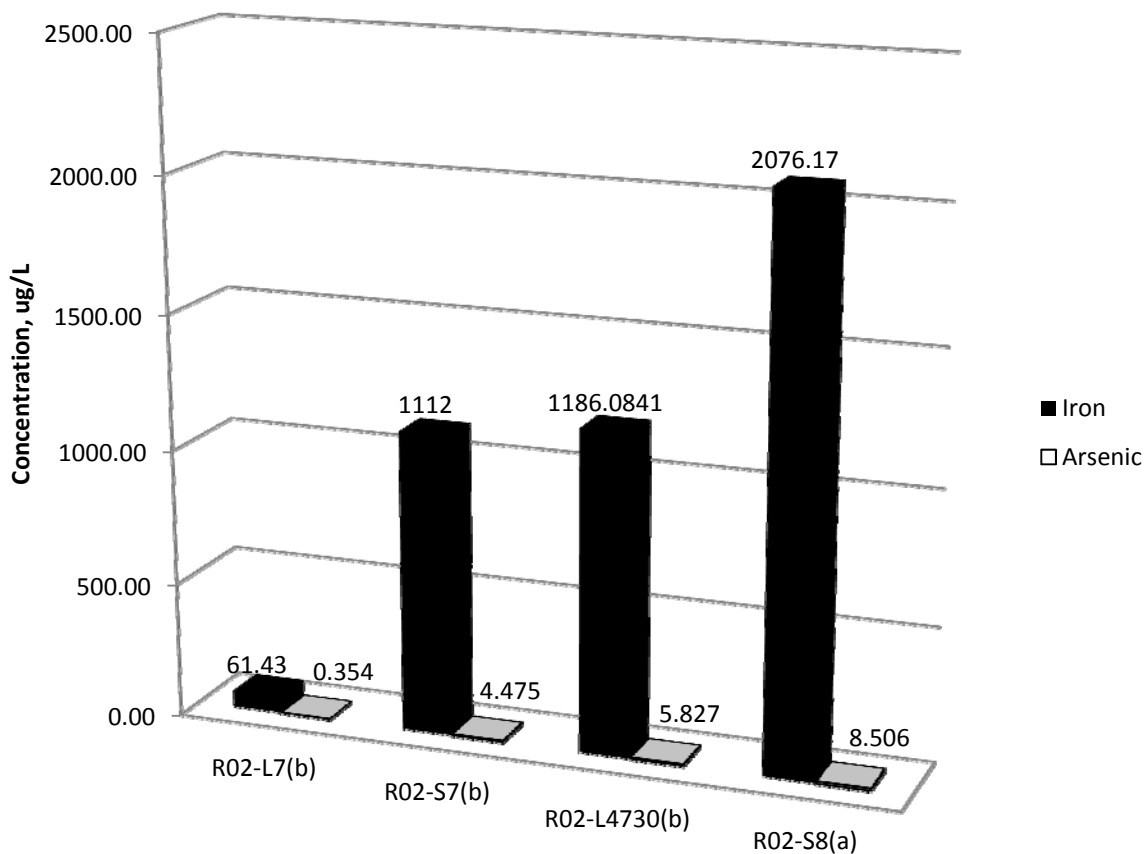


Figure 5: Boundary Layer Effect: Comparison of arsenic removal efficiency between small and large column (RO2-L7 and RO2-S7 represents the Fe and As concentration determined in the treated water after passing 1400mL through large and smaller diameter columns, respectively, RO2-S(8) for 1600mL through the small column and RO2-L4730 for 4730mL through the large diameter column for water containing 11825.84 $\mu\text{g/L}$ of Fe and 62.91 $\mu\text{g/L}$ of As)

Preliminary experiments were also carried out with the removal of a top layer of sand to determine if the removal of sand layer revived the arsenic holding ability or not. The small diameter column that was used for conducting the RO-2 test (results presented in Figure 5) was used for the RO-5 test (to study the effect of removal of top layer of sand on arsenic removal efficiency). In this test, the top layer of sand, approximately 1cm, was removed to expose the fresh surface of sand for the filtration. 200 mL of water was passed through it making the cumulative volume to 1800mL (RO5-1, Figure 6) as 1600 mL had already been passed through the column during RO-2 test. The arsenic concentration was higher than the level of 7 μ g/L; hence, it was decided to remove approximately 1 more cm of sand layer. The 200 mL of water was passed through the column again to make the cumulative volume to 2000mL (RO5-2, Figure 6). The results suggested that (Figure 6), removing the top layer of sand did actually enhance the arsenic removal efficiency of the sand, when compared with the arsenic concentration recorded in the effluent water for the RO-1 test (Figure 4). The spike in the arsenic that was observed in the case of run out test 1 in the effluent water between 1400mL and 2000mL was not observed in this case. In fact, arsenic concentration stays at an approximately same level i.e. 9.626 and 9.616 respectively as is evident from Figure 6. Since, removing the top layer did not actually reduce the arsenic concentration in the effluent below the limit of 7 μ g/L considering the change in water composition with time as well, further such experiments were not conducted.

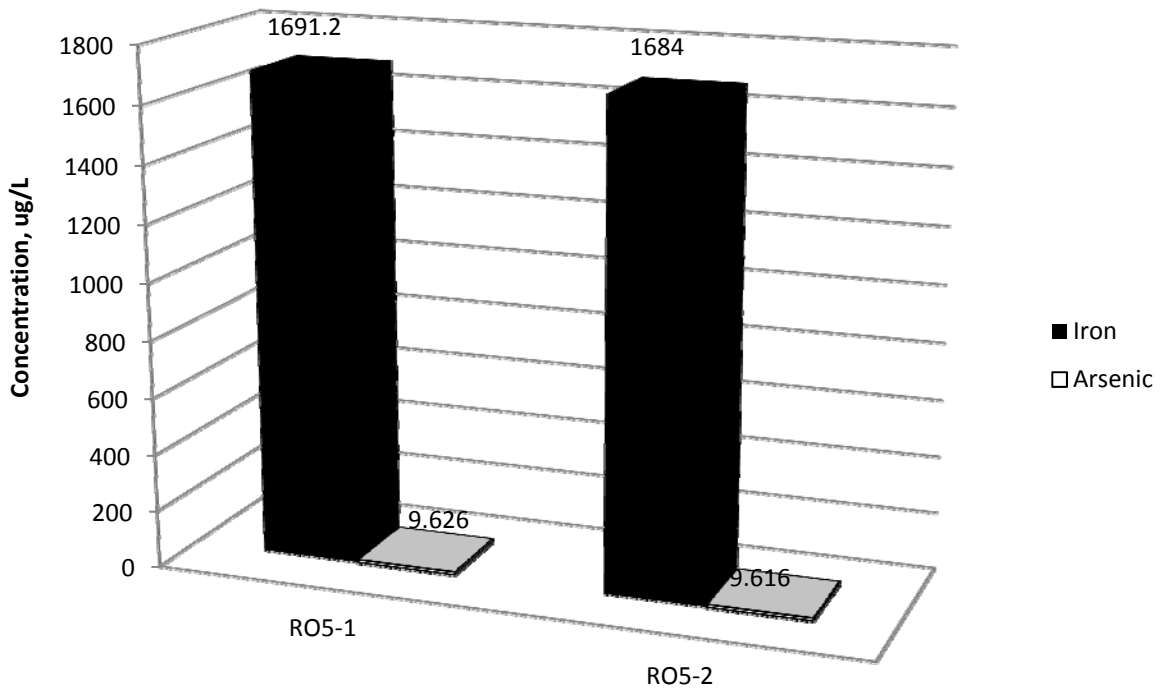


Figure 6: Run Out Test 5 (RO5-1 and RO5-2 represents the Fe and As concentration determined in the treated water after passing 1800mL and 2000mL of high As and Fe water, respectively, after removing the top layer of sand).

It can be seen from the Figure 4 that arsenic concentration in the treated water suddenly increased as the volume of treated water increased from the 1400mL. The possible cause could be the lack of aeration which inhibited the oxidation of Fe^{2+} ions to Fe^{3+} ions and hence, arsenic removal efficiency started to decrease. In order to study the effect of aeration on arsenic removal efficiency, an experiment was designed to aerate the column for two hours with the vacuum pump after passing each batch of 500 mL of high iron and arsenic Wabana water. The vacuum pump manufactured by the Gast manufacturing was a non-lubricated diaphragm type pump capable of generating 25.5 in-Hg of vacuum at a pressure of 60psi with the maximum flow output of 1.90cfm. The results were very promising and the sample collected after passing 2400mL of water i.e. RO3-2400, had arsenic and iron contents of 1.69 and 14.4 μ g/L, respectively (Figure 7). The mechanism involved the oxidation of Fe^{2+} ions to Fe^{3+} ions in the presence of oxidizing agent (atmospheric oxygen or chemicals can also be added) followed by the oxidation of some of As(III) (arsenite) to better adsorbable As(V) (arsenate). Further, arsenite and arsenates were adsorbed on hydrous ferric oxide (HFO) particles followed by the formation of $FeAsO_4(s)$ that is precipitated along with the HFO. One study found out that the adsorption affinity of As(V) to HFO is higher as compared to the As(III) by a factor of 100 (Roberts et al., 2004). Arsenic in Newfoundland groundwater resources exists primarily in the form of arsenate, hence, it is comparatively easy to mitigate the arsenic concern in the Newfoundland due to the better adsorption characteristics of ionic As (V) species ($H_2AsO_4^{4-}$ and $HadO_4^{-2}$) (Rageh, 2008).

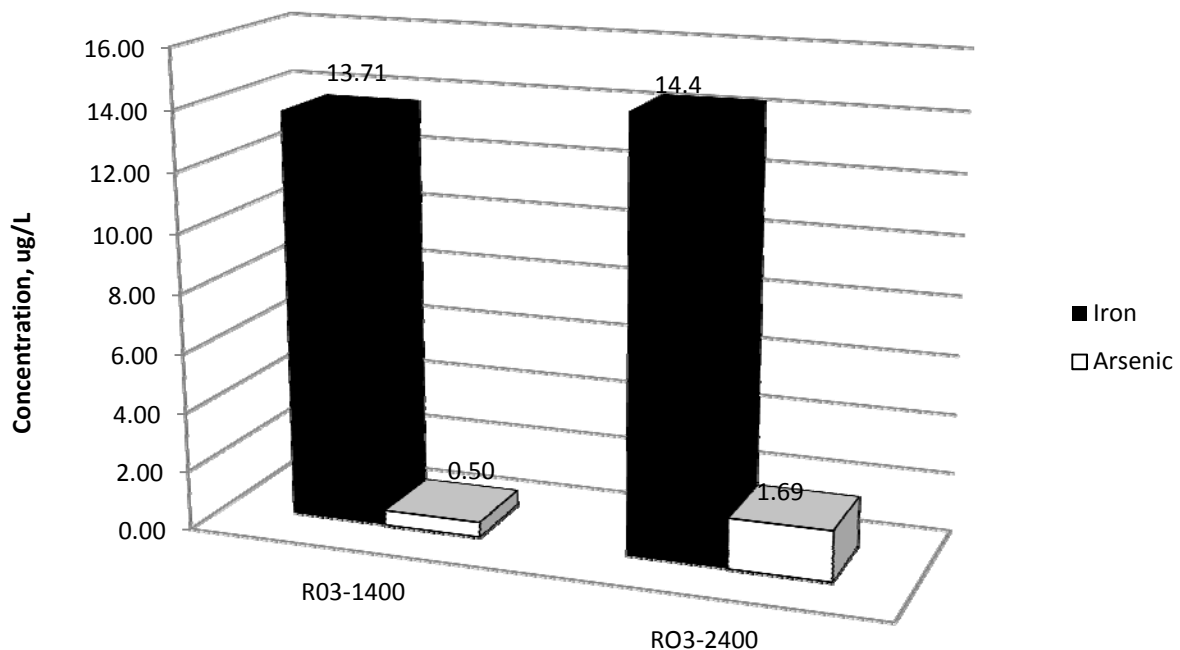


Figure 7: Run Out Test 3 (RO3-1400, RO3-2400 represents the Fe and As concentration determined in the treated water after treating 1400mL and 2400mL of base water containing 11825.84 μ g/L of Fe and 62.91 μ g/L of As using aeration.

Berg et al., (2006) suggested that an Fe/As ratio of 50 was required to achieve the residual arsenic concentration of 50 $\mu\text{g/L}$ and in order to comply with WHO guidelines (10 $\mu\text{g/L}$) an Fe/As ratio of 250 was required. In our water sample the Fe/As ratio was 187.98 which was less than the 250. This suggests that there were other mechanisms, besides HFO adsorption, in effect, that aided in achieving the arsenic concentration to the level of 7 $\mu\text{g/L}$. It is believed that the removal of arsenic was not only dependent on the iron concentration but also on the presence of manganese. Manganese dioxide (MnO_2) oxidizes the As (III) to better adsorbable As(V) in the water bodies (Mohan & Pittman, 2007). The study conducted by Driehaus et al., (1995) revealed that manganese had the ability to form arsenate-manganese ion complexes which are favorable for arsenic removal as it provides adsorption site for removal of arsenic in addition to the HFO. Hence, the presence of high manganese content (1561.51 $\mu\text{g/L}$) along with the high iron content could be responsible for achieving the better removal rates of arsenic in our case. The sand had a high uniformity coefficient (UC) of >7.5 and an iron content of 11825.84 $\mu\text{g/L}$. Uniformity Coefficient is defined as the “ratio of the size of grain which has 60 percent of the sample finer than itself to the size which had 10 percent finer than itself” (Dake, 2009). Due to the high UC of sand and very high iron content, the fine particles in the sand and the conversion of soluble Fe^{2+} ions to insoluble Fe^{3+} ions iron particles occupied the pore space during the vacuum pump operation and later on slowed down the water filtration rate to a great extent. Hence, it was not possible to conduct future tests and hence further experiments were discontinued.

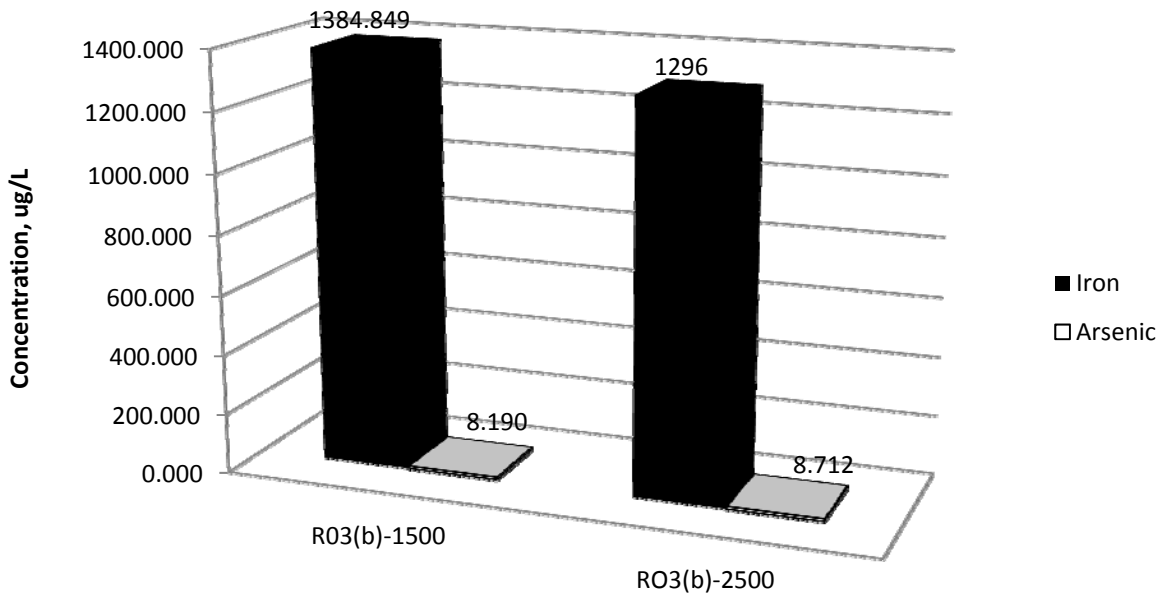


Figure 8: Run Out Test 3(b) (RO3(b)-1500, RO3(b)-2500 represents the Fe and As concentration determined in the treated water after treating 1500mL and 2500mL of base water containing 11825.84 $\mu\text{g/L}$ of Fe and 62.91 $\mu\text{g/L}$ of As using sand of Uniformity Coefficient 4 in combination with aeration)

An experiment was designed to study the effect of lower UC on the arsenic removal efficiency of the sand. The sand of UC of 4 was used and the results are presented in Figure 8. It can be seen from Figure 8 that arsenic concentration after treating 1500 mL and 2500 mL of water was 8.190 $\mu\text{g/L}$ and 8.712 $\mu\text{g/L}$, respectively. This concentration was higher than that recorded for the RO-3 test for the same volume (Figure 7). Further, the arsenic concentration after passing 2500 mL was found to be 8.712 $\mu\text{g/L}$, which was low compared to the arsenic concentration recorded for the RO-1 test for 2000 mL i.e. 19.73 $\mu\text{g/L}$ (Figure 4). It was therefore concluded that aeration aided in reducing the arsenic concentration in the effluent. Further, the lower the UC, the efficiency of arsenic removal decreased as the porosity was increased and the capacity of sand to retain HFO particles decreased. As can be seen from the Figure 8 that iron content was 1384.8 $\mu\text{g/L}$ and 1296 $\mu\text{g/L}$ for the treated volumes of 1500mL and 2500mL, respectively for RO-3(b) tests as compared to the iron content of 13.71 $\mu\text{g/L}$ and 14.4 $\mu\text{g/L}$ for RO(3) tests for the volumes of 1400mL and 2400mL, respectively (Figure 7). The increased iron concentration in the water corresponded directly to the increased arsenic concentration as the HFO particles have the high adsorption capacity for the arsenate species (Roberts et al., 2004).

After conducting preliminary experiments it was decided to use combination of dilution and aeration to conduct the later experiments to reduce the arsenic concentration down to the limit of 7 $\mu\text{g/L}$. This helped in increasing the life of the sand bed by increasing the volume of the water that could be treated and the sand replacement frequency could be greatly reduced.

b) Detailed Experiments

Run Out Test-4

After conducting preliminary analysis, it was concluded that the combination of dilution and aeration could be helpful in utilizing the sand bed for a longer duration of time. The run-out test was designed entirely for this purpose. The Run out 4 tests involved the dilution of high arsenic and iron Wabana water (base water sample) with Wabana normal supply water system in a ratio of one to one (1:1). The objective was to study the effect of water dilution on the arsenic removal efficiency of the sand and to determine if this approach could be useful in practical applications. The groundwater wells that have a high arsenic concentration can flow commingle with the other wells tied into the supply network without abandoning them and this may result in better conservation of water resources. The iron and arsenic concentration of the freshly collected Wabana water sample was 10045 $\mu\text{g/L}$ and 65.579 $\mu\text{g/L}$, respectively at the beginning of the test. The water sample was divided into small containers to retard the oxidation of Fe^{2+} ions to Fe^{3+} ions by filling them to zero air space and using the water from only one container at a time and keeping the rest sealed.

The results were very promising and suggested that 4000mL of diluted water sample could be treated (Figure 9) without replacing the sand instead of 1400mL for the base water sample (Figure 4). After treating 4000mL the arsenic concentration in the treated water approached the mark of 7 μ g/L.

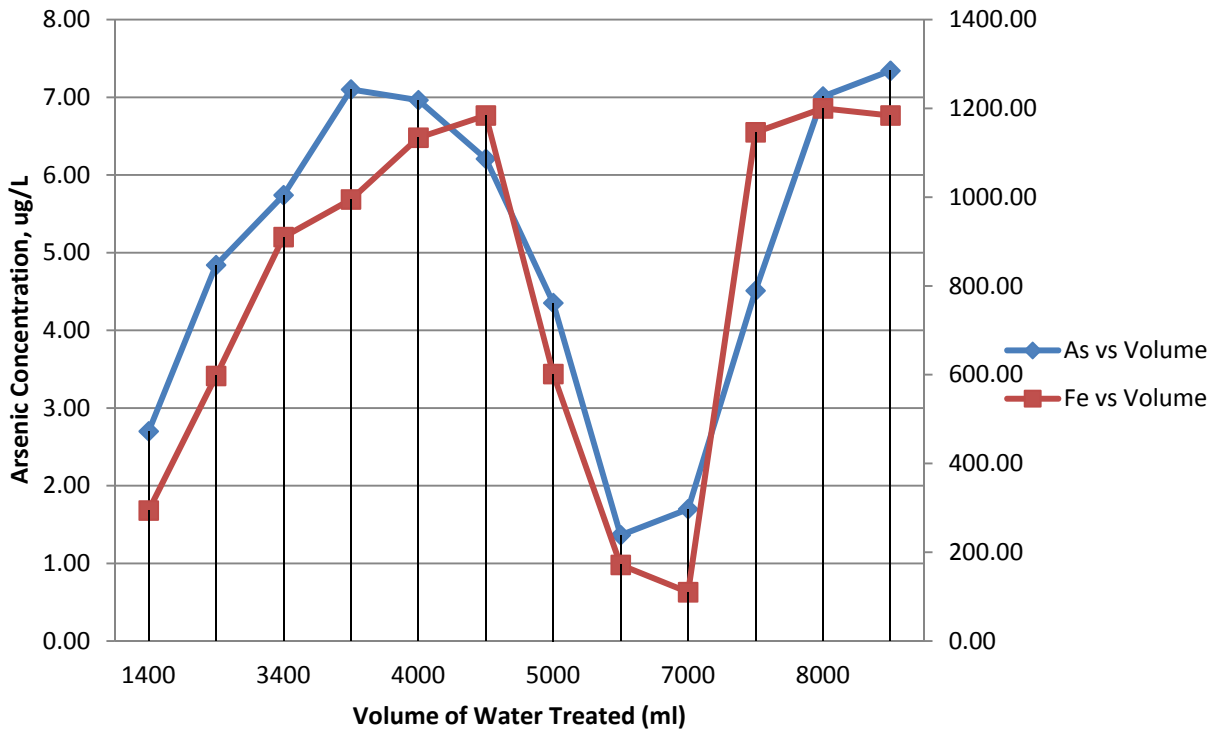


Figure 9: Change in Iron and Arsenic concentration with volume for RO-4 test in combination with the dilution of 1:1 and aeration.

Next, the column was aerated for one hour using the vacuum pump to help oxidize the Fe^{2+} ions to Fe^{3+} ions. 200 mL of water was passed through the column. After aeration the arsenic concentration in treated water dropped down from 6.96 μ g/L for 4000mL to 6.21 μ g/L for 4200mL (Figure 9). Hence, it was decided to aerate the column for one hour after treating every batch of 500mL of water. After treating 4500mL, the vacuum pump was taken into the service again and 500mL of more water was filtered through the column and so on. The downward trend in the arsenic concentration was observed and it was dropped from 6.96 μ g/L for 4000 mL to 1.36 μ g/L for 6000mL (Figure 9). Moreover, the downward trend in the iron concentration of the effluent water was also observed from 1133 μ g/L for 4000mL to 110 μ g/L for the volume of 6000mL (Figure 9). This could be due to the oxidation of Fe^{2+} to Fe^{3+} ions (insoluble form of iron) due to which iron was accumulating in the sand rather than being carried out with the water. The results suggested that arsenic removal efficiency was enhanced because of the oxidation of Fe^{2+} ions to Fe^{3+} ions. The Fe^{3+} ions had better adsorption characteristics for the arsenite and arsenate species. After treating further water the increasing trend in the As and Fe concentrations was observed. The arsenic concentration was increased to the 7.34 μ g/L after

treating 9000mL of this mix (Figure 9). Hence, in total 121.58 micrograms of arsenic was removed from treating 4000mL of water, while, the input iron concentration was 5065.66µg/L. The amount of arsenic adsorbed (µg) was calculated using the following formula and tabulated below in the Table 3:

$$\text{Arsenic adsorbed}(\mu\text{g}) =$$

$$\Sigma \left[\left\{ \text{Influent As} \left(\frac{\mu\text{g}}{\text{L}} \right) \times \text{Vol} (\text{L}) \right\} - \left\{ \text{Effluent As} (\mu\text{g}) \right\} \right] \quad \text{Eq 4}$$

The effluent arsenic concentration (µg) was calculated using following relation:

$$\text{Effluent As} (\mu\text{g}) =$$

$$\Sigma_{i=1}^n \left[\text{Eff. As Conc}_i \left(\frac{\mu\text{g}}{\text{L}} \right) \times \{ \text{Vol. Treated}_i (\text{L}) - \text{Vol. Treated}_{i-1} (\text{L}) \} \right] \quad \text{Eq 5}$$

Whereas, the influent arsenic concentration was calculated as:

$$\text{Influent Arsenic} \left(\frac{\mu\text{g}}{\text{L}} \right) = 0.5 \times 65.579 \left(\frac{\mu\text{g}}{\text{L}} \right) + 0.5 \times 4.5 \left(\frac{\mu\text{g}}{\text{L}} \right) \quad \text{Eq 6}$$

The constant i.e. 0.5 was the ratio in which samples were mixed. 65.579µg/L and 4.5µg/L were the arsenic concentrations of the high arsenic and iron Wabana water and Wabana normal supply water, respectively. The input arsenic concentration was calculated to be 35.0395µg/L.

The values for the effluent arsenic concentration (µg) at various analysis intervals are tabulated below in Table 3.

Table 3: Calculation of effluent Arsenic concentration (ug) and the Amount of Arsenic removed (ug) for RO-4 test

Volume (mL)	Effluent As (µg/L)	Volume (L) (Col1/1000)	Volume (L) between Successive Intervals	Input As (µg/L)	Influent As (ug) (Col3 x Col5)	Effluent As (ug) (Col2 x Col3)	As Removed (ug) (Col5 - Col6)
1400	2.70	1.4	1.4	35.04	49.06	3.78	45.28
2400	4.84	2.4	1		35.04	4.84	30.20
3400	5.74	3.4	1		35.04	5.74	29.30
3700	7.10	3.7	0.3		10.51	2.13	8.38
4000	6.96	4	0.3		10.51	2.0889	8.42
Cumulative Arsenic Removed (ug)							121.58

Similarly, the input iron concentration was calculated using following equation:

$$\text{Influent Iron } \left(\frac{\mu\text{g}}{\text{L}}\right) = 0.5 \times 10045 \left(\frac{\mu\text{g}}{\text{L}}\right) + 0.5 \times 86.327 \left(\frac{\mu\text{g}}{\text{L}}\right) \quad \text{Eq 7}$$

The constant i.e. 0.5 is the ratio in which samples were mixed. 10045 $\mu\text{g/L}$ and 86.327 $\mu\text{g/L}$ was the iron concentration of high iron and arsenic Wabana water and Wabana normal supply water, respectively. The Input iron was calculated to be 5065.66 $\mu\text{g/L}$.

Minitab was used to determine the correlation between arsenic and different elements' concentrations. It was found out that arsenic levels were significantly correlated with the iron and chloride content (Table-4). The relation between iron and arsenic is positive, while, that of arsenic and chlorine is negative.

Table 4: Concentration of elements significantly correlated to Arsenic in the effluent of RO-4 test

Volume Treated (mL)	As Content ($\mu\text{g/L}$)	Fe Content ($\mu\text{g/L}$)	Cl Content ($\mu\text{g/L}$)
1400	2.70	294.04	41653
2400	4.84	597.20	44073
3400	5.74	910.04	38313
3700	7.10	994.86	55154
4000	6.96	1133.93	36137
4200	6.21	1184.00	50802
5000	4.35	601.50	41741
6000	1.36	171.44	60653
7000	1.70	110.00	106519
7500	4.51	1184.00	35589
8000	7.01	1200	37104
9000	7.34	1146	33418
Correlation with As	1	0.906	-0.597

According to Vincent (1995), for data of 12 different samples, a significant correlation should be of value 0.576 or more. The Fe based adsorbents are widely used in water treatment applications to adsorb the arsenic and the high positive correlation between Fe and As in Table 3 was in accordance to the literature cited (Leupin and Hug 2005, Gu et al. 2005 and Berg, et al. 2006). Further, the effect of chlorine (Cl) on arsenic adsorption is still unknown. But it is suggested that chlorine has the same effect as that of other anions like phosphate, silicate and bicarbonates i.e. it competes with arsenic for the iron oxides adsorption sites (Meng, et al. 2002). The effect of chloride is more significant for As (III) (Meng, et al. 2002).

Run Out Test 4(b)

The Run out 4(b) test was similar to the Run-out-4 test with only difference being the ratio of the mixture. It involved the dilution of high arsenic and iron Wabana water (base water sample) with Wabana normal supply water system in a ratio of one to four (1:4). Two tests, RO4 and RO4(b), were designed to study the combined effect of aeration and dilution on the arsenic removal efficiency of the sand medium. Later, the results for these two tests are compared with the RO-1 test (Figure 4).

The results for the RO-4(b) test were very promising and suggested that approximately 11000mL of diluted water sample could be treated (Figure 10) without replacing the sand instead of 1400mL for the base water sample (Figure 4) and 4000mL for RO4 test (Figure 9). After treating 10,500 mL the arsenic concentration in the treated water was found to be 6.232 $\mu\text{g/L}$. After this, the column was aerated for one hour using the vacuum pump to help oxidize the Fe^{2+} ions to Fe^{3+} ions. 500 mL of water was passed through the column. After aeration, the arsenic concentration in treated water dropped down from 6.232 $\mu\text{g/L}$ for 10,500mL to 4.751 $\mu\text{g/L}$ for 11,000mL (RO4-4200) (Figure 10). Hence, it was decided to aerate the column for one hour after treating every batch of 500mL of water. After treating 11,500mL, the vacuum pump was taken into the service again and 500mL more water was filtered through the column and so on. The downward trend in the arsenic concentration was observed and it was dropped from 6.232 $\mu\text{g/L}$ for 10,500 mL to 2.513 $\mu\text{g/L}$ for 14,000mL (Figure 10).

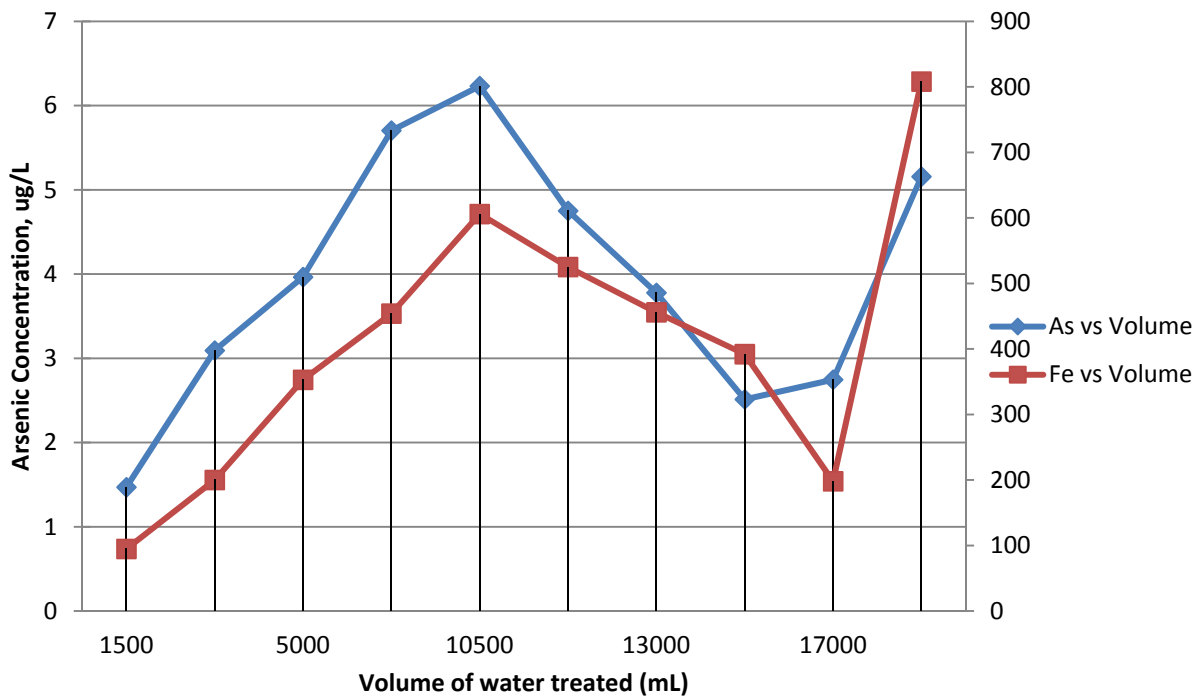


Figure 10: Change in Arsenic concentration with volume for RO-4(b) test in combination with the dilution of 1:3 and aeration

After treating 17000mL, the upward trend in As concentration was observed. Arsenic concentration was 2.747µg/L corresponding to the treated volume of 17000mL (Figure 11). Moreover, the downward trend in the iron concentration of the effluent water was also observed which dropped from 605.99µg/L for 10,500mL to 392.14µg/L for the treated volume of 14000mL (Figure 10). This suggested that oxidation of Fe²⁺ ions to Fe³⁺ ions (insoluble form of iron) was taking place and iron was accumulating in the sand rather than being carried out with the water. The results suggested that the arsenic removal efficiency was enhanced because of the oxidation of Fe²⁺ ions to Fe³⁺ ions that had better adsorption characteristics for the arsenite and arsenate species (Roberts et al., 2004). For 18000mL, the iron content was raised to 808.2µg/L (Figure 10), while the arsenic concentration increased to 5.157µg/L (Figure 10). Hence, in total 166.50 micrograms of arsenic was removed from treating 10,500mL of water, while, the input iron concentration was only 2576µg/L. The amount of arsenic adsorbed (µg) was calculated using equation 4 and the effluent arsenic concentration (µg) was determined using equation 5 presented earlier in the report. The values for the effluent arsenic concentration (µg) at various analysis intervals are tabulated below in Table 5.

The input arsenic concentration was calculated as:

$$\text{Influent Arsenic } \left(\frac{\mu\text{g}}{\text{L}}\right) = 0.25 \times 65.579 \left(\frac{\mu\text{g}}{\text{L}}\right) + 0.75 \times 4.5 \left(\frac{\mu\text{g}}{\text{L}}\right) \quad \text{Eq 8}$$

The constant i.e. 0.25, 0.75 was the ratio in which samples were mixed. 65.579µg/L and 4.5µg/L were the arsenic concentrations of high arsenic and iron Wabana water and Wabana normal supply water, respectively. The input arsenic concentration was calculated to be 19.770µg/L.

Table 5: Calculation of effluent Arsenic concentration (ug) and the Amount of Arsenic removed (ug) for RO-4(b) test

Volume (mL)	Effluent As (µg/L)	Volume (L) (Col1/1000)	Volume (L) between Successive Intervals	Input As (µg/L)	Influent As (ug) (Col3 x Col5)	Effluent As (ug) (Col2 x Col3)	As Removed (ug) (Col5 - Col6)
1500	1.47	1.5	1.5	19.77	29.65	2.205	27.45
4000	3.093	4	2.5		49.42	7.7325	41.69
5000	3.965	5	1		19.77	3.965	15.80
7500	3.817	7.5	2.5		49.42	9.5425	39.88
9500	5.704	9.5	2		39.54	11.408	28.13
10500	6.232	10.5	1		19.76975	6.232	13.53775
Cumulative Arsenic Removed (ug)							166.50

Similarly, the input iron concentration was calculated using following equation:

$$\text{Influent Iron } \left(\frac{\mu\text{g}}{\text{L}}\right) = 0.25 \times 10045 \left(\frac{\mu\text{g}}{\text{L}}\right) + 0.75 \times 86.327 \left(\frac{\mu\text{g}}{\text{L}}\right) \text{ Eq 9}$$

The constant i.e. 0.25 and 0.75 is the ratio in which samples were mixed. 10045 $\mu\text{g/L}$ and 86.327 $\mu\text{g/L}$ was the Fe concentration of high iron and arsenic Wabana water and Wabana normal supply water, respectively. The Input iron was calculated to be 2576 $\mu\text{g/L}$.

Minitab was used to determine the correlation between As and different elements' concentrations. It was found that the arsenic levels were significantly correlated with the iron and lithium content (Table-6). The relation between both arsenic and iron and arsenic and lithium was positive.

Table 6: Concentration of elements significantly correlated to Arsenic in effluent of RO-4(b) test.

Volume Treated (mL)	As Content ($\mu\text{g/L}$)	Fe Content ($\mu\text{g/L}$)	Li Content ($\mu\text{g/L}$)
1500	1.470	94.90	2.642
4000	3.093	200.000	13.936
5000	3.965	353.031	12.572
9500	5.704	454	14.416
10500	6.232	605.99	15.758
11000	4.751	525	15.106
13000	3.778	456	11.154
14000	2.513	392	1.906
17000	2.747	198.201	18.100
18000	5.157	808.230	15.426
Correlation with As	1	0.802	0.641

According to Vincent (1995), for data of 10 different samples, a significant correlation should be of value 0.632 or more. The affinity of oxides of iron to adsorb arsenic was mentioned in the literature cited and a lot of work had been carried out in this field (Leupin and Hug 2005, Gu, et al. 2005 and Berg, et al. 2006). However, the interaction between lithium (Li) and As is still unknown and has not yet been mentioned in the literature. This is one of the topic that still needs to be explored. Although, the correlation observed between Li and As was not very strong but it could be due to the effect of Li in reducing the adverse effects of anions in inhibiting the arsenic co-precipitation with oxides of iron either by affecting the surface charge or enhancing the floc aggregation of iron oxides.

It can be concluded from Figure 11 that 76.56 micrograms of As was removed or 1400 mL of high arsenic and iron content Wabana water was treated for the RO-1 test (Figure 4). However, after incorporating dilution 121.58 micrograms of arsenic was removed or 4000mL of one to one mixture of Wabana’s high iron and arsenic content water with Wabana’s normal supply water was treated for RO4 test. Similarly, 166.50 micrograms of As was removed or 10,500mL of one to four mixture of Wabana’s high iron and arsenic content water with Wabana’s normal supply water could be treated for RO4(b) test before the arsenic concentration approached the allowable limit of $7\mu\text{g/L}$. It is evident from Figure 11 that as the dilution was increased, more quantity of water could be filtered and sand replacement frequency could be decreased as the concentration of inlet iron was decreased. The higher the concentration of the iron, the more quickly it occupied the pore space in the sand bed and therefore, it retarded the iron holding capacity of the sand and in turn influenced the arsenic removal efficiency. Hence, it was concluded that if there are one or two wells in the water supply system with high arsenic content, the water from these wells can be used for potable purposes if diluted with the groundwater wells containing low arsenic concentration, provided that they have high concentration of iron and manganese. Further, if dilution is combined with aeration, the efficiency of filtration system can further be augmented.

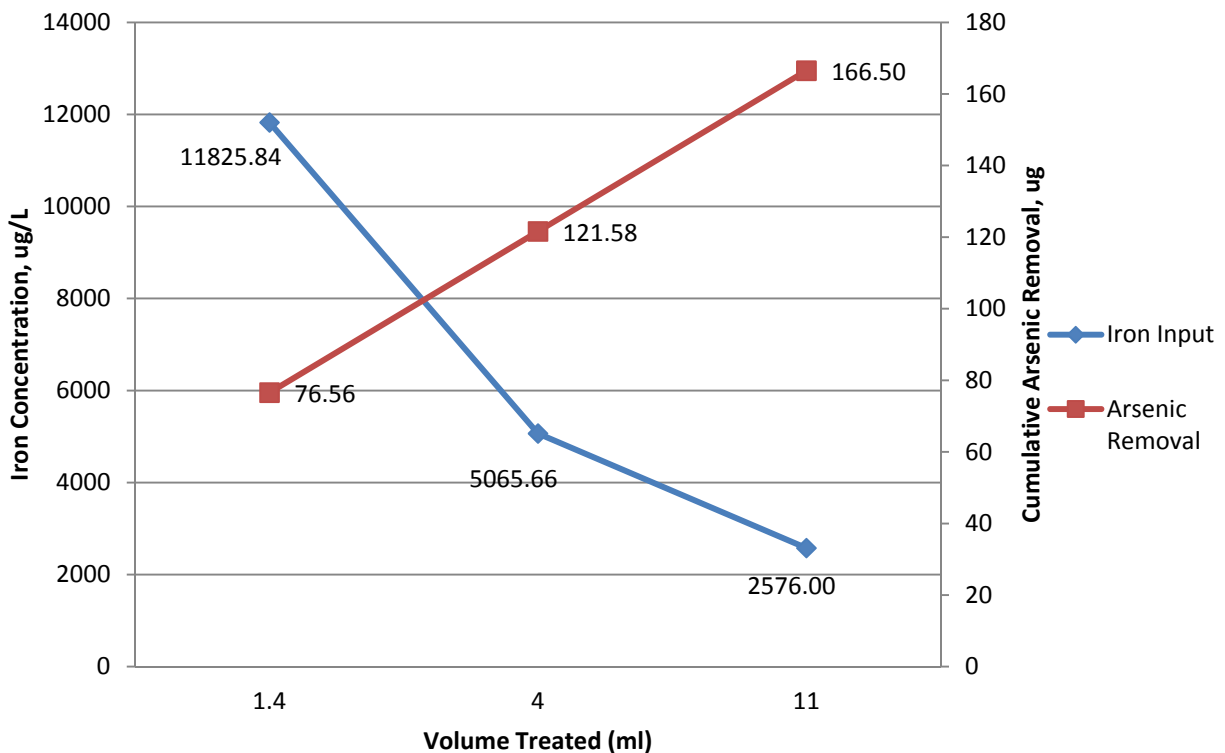


Figure 11: Comparison of Cumulative As removal vs Iron Input for RO-1, RO4 and RO4(b) test without considering the effect of aeration

Breakthrough Curves

A Breakthrough curve is an 'S' shaped curve that is generated by plotting adsorbate concentration against pore volume or time (U.S. Army Corps of Engineers 2001). The break through analysis was conducted for the high iron and arsenic containing Wabana water. The initial concentration of iron and arsenic at the start of breakthrough analysis was found out to be 8486.688 $\mu\text{g/L}$ and 37.787 $\mu\text{g/L}$, respectively using ICPMS analysis. The drop in the iron and arsenic concentration from 62.91 $\mu\text{g/L}$ and 11825.84 $\mu\text{g/L}$, respectively for the freshly collected water sample was associated with the instability of this water sample. In the ground, the anoxic condition prevailed that avoided the oxidation of soluble Fe^{2+} ions to insoluble Fe^{3+} ions. Hence, as this water was exposed to oxic conditions, the oxidation of soluble Fe^{2+} ions to insoluble Fe^{3+} ions started taking place. Since Fe^{3+} had good adsorption characteristics for arsenic species, they started adsorbing arsenic from the water and settled at the bottom of the container. Therefore, the drop in the concentration of iron and arsenic concentration was observed. In order to avoid the further change in the concentration, the water was transferred from the 20 litre capacity gallon to small bottles of two litre capacity. The bottles were filled to the top for zero air space and were tightly capped. The iron to arsenic ratio for this water sample was 224.59. The column and sand was prepared in the same way as described earlier in the report. The test was continued for 120 hours until approximately 11500 mL of water had passed. The average filtration rate was approximately 79.1 mL/h. The filtration rate at the start of experiment was 125 mL/hr which slowly dropped down as the test progresses because the pore space in the sand bed was starting to fill up with the adsorbing species. The pump was used to conduct the breakthrough analysis to increase the filtration rate. The pump was supplied by the M/s Cole-Parmer. It was a Masterflex variable speed drive pump with the revolutions per minute (RPM) in the range of 6-600. The flow rate could be adjusted in the range of 0.36mL/min to 3400mL/min depending upon the tubing size and the RPM. The parameters such as flow rate were not changed throughout the duration of the test.

The concentration of elements present in the effluent and having significant correlation with As are tabulated in Table 7. According to Vincent (1995), for data of 8 different samples, a significant correlation should be of value 0.707 or more. The results showed that the arsenic concentration dropped from 37.787 $\mu\text{g/L}$ to 4.4 $\mu\text{g/L}$ in the first 4 hours and then the uptake rate decreased gradually. After 150 hours, the Arsenic concentration in the effluent went up to 32.44 $\mu\text{g/L}$. The breakthrough occurred after approximately 36 hours where a value of 17.83 $\mu\text{g/L}$ was obtained (Figure 12). The breakthrough point is defined as the point on the breakthrough curve where the concentration of the adsorbate in the effluent reaches to the maximum allowable concentration (U.S. Army Corps of Engineers, 2001). The breakthrough point for this study was defined as the 50 percent of the influent adsorbent concentration as is the case with geo-environmental engineering (Yong, 2001). The concentration was taken as the 50% of the influent concentration because the flow at this point in the adsorbent bed was considered as a steady state (Shackelford, 1993). The filter could be taken out of operation for servicing at the breakthrough

point. The breakthrough curves was drawn using $\frac{C_i}{C_o}$ vs Time (h) for As, Fe and Mn. (Figure 12). The C_i was defined as the the concentration at time 'i'; while, C_o was defined as the initial concentration.

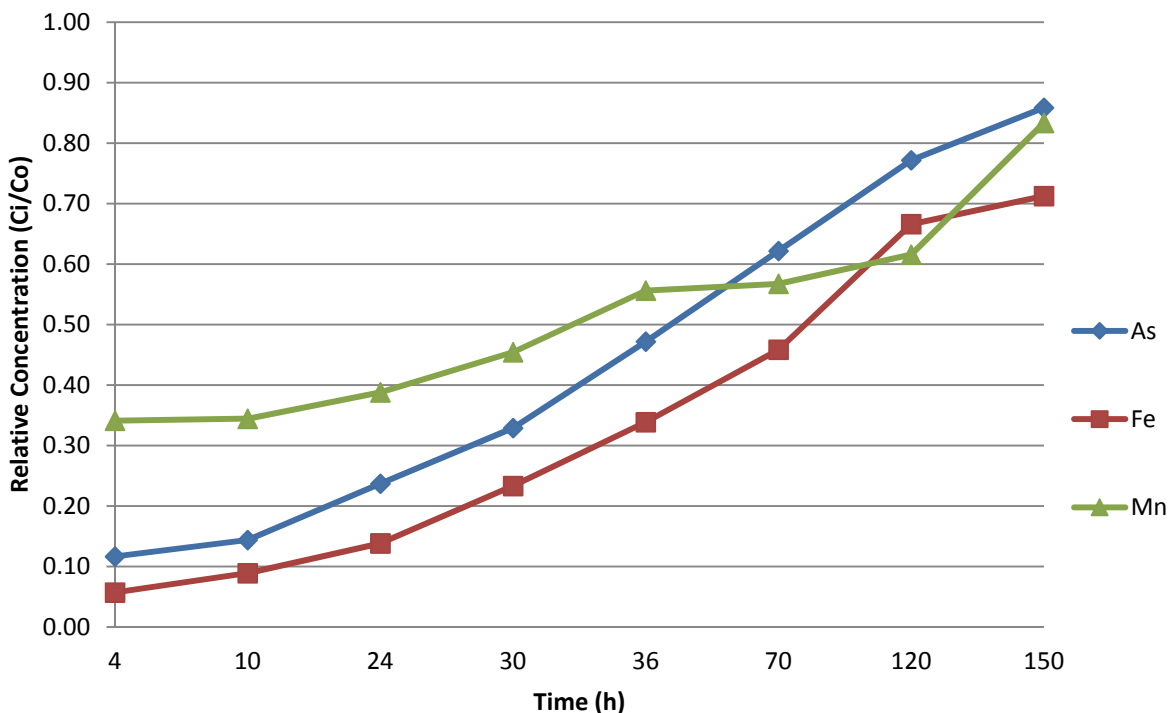


Figure 12: Breakthrough curves for High Iron and Arsenic Wabana water in combination with Fe and Mn content

Minitab was used to find the significantly correlated elements' concentration with the arsenic concentration. The elements such as Be, P, V, Cr, Fe and Mn were found to be statistically correlated with As. The correlation values are shown at the bottom of the Table 7. Other than Fe and Mn, the rest of the elements were negatively correlated with As. Iron hydroxide based adsorbents could be used to adsorb SO_4^{-2} , SeO_3^{-2} , PO_4^{-3} and CrO_4^{-2} ions (Meng and Letterman, 1996; Goldberg, 1985, Zachara et al., 1987 and Chowdhury and Yanful 2010). Therefore, in the presence of sulphates, phosphates and chromate ions, the arsenate species competed with them for the HFO adsorption sites and hence they were found negatively correlated with the arsenic. Similarly, it was reported that vanadate anions have the ability to adsorb on the iron based adsorbents (Naeem et al., 2007). They also competed with the arsenic anions for the HFO adsorption sites. It could be seen from the Table 7 that as the concentration of the arsenic was increased in the effluent, the concentration of V, P and Cr decreased due to which these anions were found negatively correlated with arsenic.

Table 7: Concentration of elements significantly correlated to Arsenic in the effluent of Wabana water

Time (h)	Initial As ($\mu\text{g/L}$)	As ($\mu\text{g/L}$)	Be ($\mu\text{g/L}$)	P ($\mu\text{g/L}$)	V ($\mu\text{g/L}$)	Cr ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)	Mn ($\mu\text{g/L}$)
4	37.79	4.4	37.30	358.10	8.03	5.710	483.05	1154.00
10		5.43	37.64	361.30	8.11	5.760	754.68	1165.00
24		8.95	37.65	361.40	8.11	5.770	1172.5	1312.60
30		12.43	3.41	248.50	2.46	4.045	1977.6	1537.32
36		17.83	4.48	326.00	3.24	3.297	2873.6	1882.10
70		23.49	9.91	158.80	2.67	3.255	3889.4	1919.75
120		29.16	3.59	60.323	1.99	1.029	5652.1	2082.96
150		32.44	3.410	34.60	2.17	0.928	6046.4	2820.3
Correlation with Arsenic		1.00	-0.803	-0.942	-0.846	-0.972	0.996	0.954

Run-Out Test Conducted for water Collected from Town of Freshwater

The water collected from the Town of Freshwater was filtered through the column partially filled with sand to determine the amount of water that could be treated before the As concentration exceeded the level of $7\mu\text{g/L}$. The column and sand was prepared in the same manner as described earlier in the report.

It can be seen from Figure 13 that after treating 1000mL of water collected from the town of Freshwater, the Fe and As concentration dropped from $507.52\mu\text{g/L}$ and $29.707\mu\text{g/L}$ to $117.46\mu\text{g/L}$ and $4.88\mu\text{g/L}$, respectively. As additional water was passed through the column, the As concentration increased to $8.217\mu\text{g/L}$ which was higher than the limit of $7\mu\text{g/L}$ set for this project. It was concluded that the low removal rates were subjected to the low Fe concentration which is the main source of providing the adsorption sites for the As in form of HFO. Therefore, it was decided to conduct future tests by mixing the water sample collected from the town of Wabana and Freshwater in three different ratios to get three different Fe/As ratios, as the Fe/As ratio was the most important factor in removing the As from the water (Berg, et al., 2006). Hence, the two water samples were mixed in a ratio of 1:10, 2:10 and 3:10 and were named as Mix-1, Mix-2 and Mix-3, respectively.

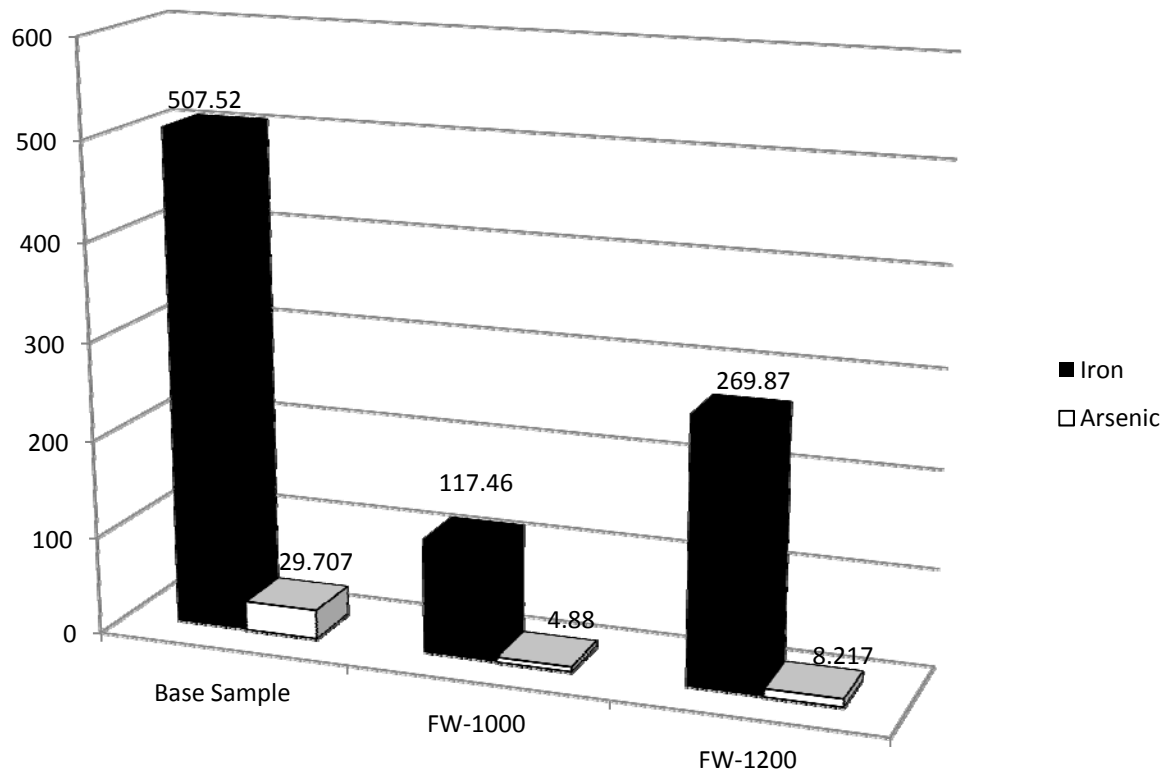


Figure 13: Run-Out test for the water collected from the Town of Freshwater (Base Sample represents the concentration of Fe and As in the groundwater sample, whereas, FW-1000 and FW-1200 represents the Fe and As concentration after treating 1000mL and 1200mL of water through the partially filled sand column)

The composition of these samples is attached in the Appendix-C. The Fe/As ratio (wt/wt) for Mix-1, Mix-2 and Mix-3 was 17.282, 32.227 and 64.957, respectively. The purpose of carrying out these tests was to determine the minimum Fe/As ratio that could be used to control the arsenic concentration in the effluent water after passing the water sample through the small column.

Mix-1

The sample mix-1 that had the Fe/As concentration of 17.282 and the initial arsenic concentration of $33.58\mu\text{g/L}$ was passed through the small column continuously at the filtration rate of 125mL/h . The pump supplied by the M/s Cole-palmer was used to carry out the test. Since, the sample had the Fe concentration of only $580.26\mu\text{g/L}$, hence no significant change in flow rate was observed. Total of $38,500\text{mL}$ water was passed through the column in approximately 325h. Hence, the average flow rate was 118.5mL/h . The concentration of elements present in the effluent having significant correlation with As are tabulated below in Table 8.

Table 8: Concentration of elements significantly correlated to Arsenic in the effluent of 1:10 mixture of the water collected from the towns of Wabana and Freshwater

Time (h)	Initial As ($\mu\text{g/L}$)	As ($\mu\text{g/L}$)	Li ($\mu\text{g/L}$)	S ($\mu\text{g/L}$)	Cl ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)	Ni ($\mu\text{g/L}$)	Cu ($\mu\text{g/L}$)	Zn ($\mu\text{g/L}$)
8	33.58	2	0.95	14318	115175.3	11	47.94	103.82	1069.12
36		4.50	1.93	6247.27	92021.03	18.98	18.91	51.92	595.80
68		6.35	2.94	5437.46	91051.09	21.16	14.14	59.93	665.71
84		5.86	2.48	13509.2	92373.52	13.42	13.59	42.88	291.16
200		11.54	2.84	4370.74	88251.65	16.67	9.07	44.87	328.41
290		16.94	4.67	63426.3	140799.8	3.06	2.82	60.39	397.94
315		20.37	3.69	68199.6	132160.2	66.68	1.51	14.58	117.87
325		23.93	4.31	71532.2	141403.9	203.18	2.26	16.39	98.15
Correlation with Arsenic		1.00	0.879	0.901	0.768	0.777	-0.780	-0.757	-0.805

According to Vincent (1995), for data of 8 different samples, a significant correlation should be of value 0.707 or more. The initial As concentration dropped from $33.58\mu\text{g/L}$ after 8 hours to $2\mu\text{g/L}$ and then the As uptake decreased gradually. After 325 hours, the arsenic concentration reached to the maximum of $23.93\mu\text{g/L}$. The As concentration limit of $7\mu\text{g/L}$ set for this project was exceeded after 84 hours and the breakthrough point was reached after approximately 290 hours with the As concentration of $16.94\mu\text{g/L}$ (Figure 14).

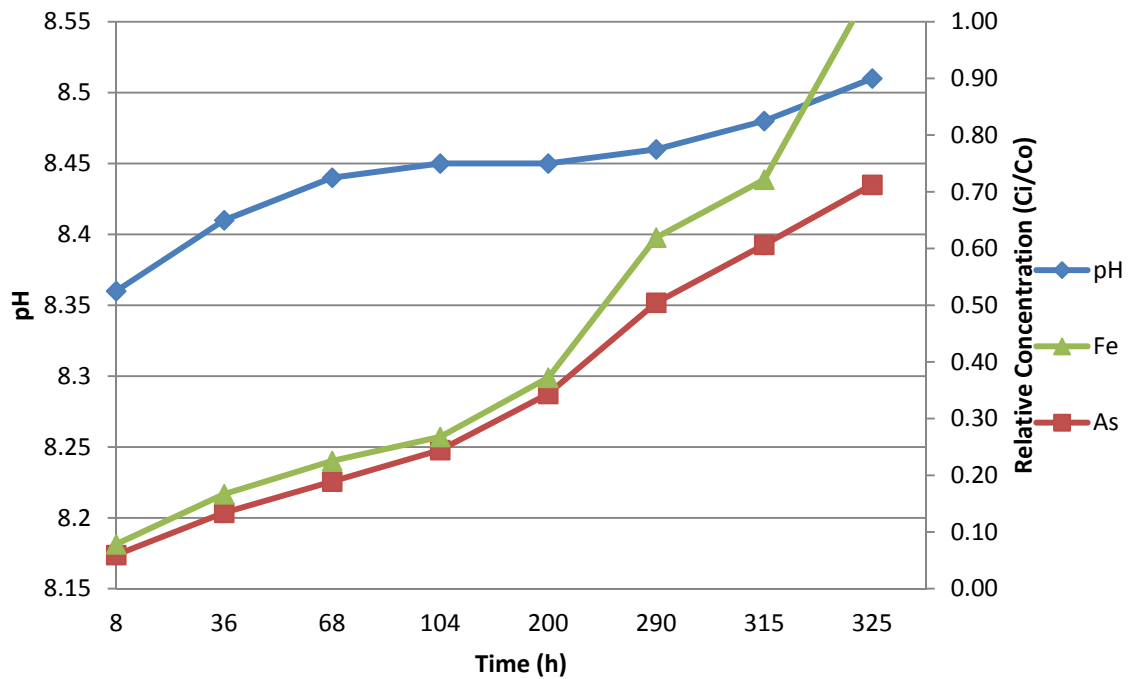
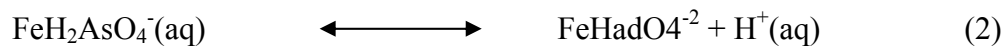


Figure 14: Breakthrough curve for Mix-1 test showing iron, arsenic and pH trends

Table 9: Change in pH at different times in relation with the Fe and arsenic concentration in the effluent during the Mix-1 test.

Time (h)	pH	As ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)
0	7.97	33.58	580.26
8	8.36	2	11
36	8.41	4.50	18.98
68	8.44	6.35	21.16
84	8.45	5.86	13.42
200	8.45	11.54	16.67
290	8.46	16.94	3.06
315	8.48	20.37	66.68
325	8.51	23.93	203.18

It is reported in the literature that with the increase in pH, the adsorption of arsenic decreases which suggested that it is an anion exchange material (Streat et al., 2008b & Driehaus et al., 1998). The pH values are listed in the Table 9 and pH was found to increase with the decrease in arsenic adsorption. The isoelectric point (IEP) for the hydrous ferric oxide is in the pH range of 7-8. The IEP is defined as “the pH value of the dispersion medium of a colloidal suspension at which the colloidal particles do not move in an electric field” (McGraw-Hill Science and Technology Dictionary). Therefore, below this pH, the surface of adsorbent remained positively charged and it attracted anions. Stumm & Morgan (1981) suggested the mechanism of ligand exchange for protonated arsenic species for the pH below the IEP with the help of following equations.



The release of OH^- increased the pH of the solution and hence, decreased the positive surface charge; thus, lowering the forces to attract anionic species. The proton was dissociated from the acid surface to carry out the neutral adsorption. Qian, et al. (2009) reported that the higher pH leads to the higher adsorption of copper ions in the aqueous solution using iron based adsorbents. Due to the increase in the pH, more metal binding sites were created that resulted in the high metal ion adsorption. Hence, it may be suggested that the change in pH increased the adsorption of Cu, Ni and Zn ions on to the HFO adsorption sites and the adsorption of arsenic along with the other anionic species (S and Cl) was decreased. Further, Boujelben et al., (2009), studied the adsorption of nickel on to the iron oxide coated sand from aqueous solution in the pH range of 2-9. The optimum adsorption took place at pH:7 but the adsorption was also reported to take place between the pH range of 7-9. Moreover, Srivastava and Srivastava (1990) studied the adsorption of Zn(II) on to the Fe(III) hydroxide sites at three different pH levels of 5.5, 6.85 and 8.2. They concluded that adsorption of the Zn(II) increased with the increase in pH. Hence, It could be concluded that HFO adsorption sites were not only capable of adsorbing anionic species but cationic species as well provided that the suitable conditions were met. The decrease in

concentration of the Cu, Ni and Zn with the increase in pH suggested that under these conditions Cu, Ni and Zn ions were more favorable to adsorb on HFO sites as compared to the arsenic. This is reflected in Table 8, where it could be seen that the arsenic concentration was increasing and Cu, Ni and Zn concentrations were decreasing with the rise in pH.

Mix-2 and Mix-3

The sample mix-2 and mix-3 that had the Fe/As concentrations of 32.227 and 64.957 and the initial arsenic concentrations of 34.336 $\mu\text{g/L}$ and 34.322, respectively, were passed through the column continuously at the filtration rate of 125mL/h. The pump supplied by the M/s Colepalmer was used to carry out the test. The mix-2 and mix-3 had the Fe concentrations of 1106.53 $\mu\text{g/L}$ and 2229.43 $\mu\text{g/L}$, respectively. The RPM of the pump was kept constant throughout the duration of the tests. A total of 39000mL of water was passed through the column for both mix-2 and mix-3 tests. The tests were continuously run for 315h for both mix-2 and mix-3. Hence, the average flow rate was 123.8mL/h.

Table 10: Concentration of elements significantly correlated to Arsenic in the effluent of 2:10 mixture of the water collected from the town of Wabana and Freshwater (Mix-2)

Time (h)	Initial As ($\mu\text{g/L}$)	As ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)	Be ($\mu\text{g/L}$)	V ($\mu\text{g/L}$)	Ni ($\mu\text{g/L}$)	Zn ($\mu\text{g/L}$)	Se ($\mu\text{g/L}$)
8	34.34	1.910	23.00	10.020	14.17	153.800	2037	31.350
40		3.399	17.90	3.32	3.200	56.110	901.948	7.140
56		5.709	18.50	3.09	3.020	5.678	283.038	6.770
80		10.965	717.07	1.790	1	2.633	107.261	4.460
120		7.005	122.00	3.17	3.050	23.05	145.094	4.560
160		9.556	156.71	3.180	3.02	12.534	135.257	4.570
280		6.559	137.35	3.60	2.140	18.02	161.906	7.830
296		8.382	180.62	3.28	1.950	20.44	238.983	7.130
315		8.735	87.08	3.46	2.050	37.13	119.374	7.520
Correlation with Arsenic		1.00	0.693	-0.722	-0.729	-0.766	-0.828	-0.699

According to Vincent (1995), for data of 9 different samples, a significant correlation should be of value 0.666 or more. It could be seen from the Table 10 that arsenic was significantly correlated with Fe, Be, V, Ni and Zn. Arsenic was only positively correlated with the Fe and with the rest of the elements, it was negatively correlated. The mechanism suggested to be occurring related to the Ni and Zn adsorption on the Fe adsorption sites discussed under the section Mix-1 could very well be responsible in this case as well. Similarly, it was reported that the vanadate anions have the ability to adsorb on the iron based adsorbents (Naeem et al., 2007). Further, Jeong, et al. (2007) reported that the adsorption of As(V) on Fe adsorption sites was

affected by the presence of Se(IV) and V(V) which was exactly in accordance with the results presented in the Table 10.

The pH values recorded at different instances during the test are listed below in Table-11. The arsenic concentration remained below the level of 10µg/L even after treating 39000mL. The arsenic concentration past 80h period increased to 10.965µg/L, but, after that the drop in arsenic concentration was observed and after 315h the arsenic concentration was recorded to be 8.735µg/L. The Fe concentration dropped from 717µg/L at 80h to 87.08µg/L at 315h. It was suggested that the aeration of the small column as a result of operation of pump was aiding in oxidizing the soluble Fe²⁺ ions to insoluble Fe³⁺ ions. The formation of Fe³⁺ ions might be responsible for the improved adsorption of arsenic due to the availability of increased adsorption sites and hence lower arsenic concentration was recorded in the effluent. The breakthrough curve in relation with the pH values are depicted in Figure 15. The breakthrough did not occur for arsenic even after treating 39000mL of water for Mix-2 as it only reached 8.735µg/L.

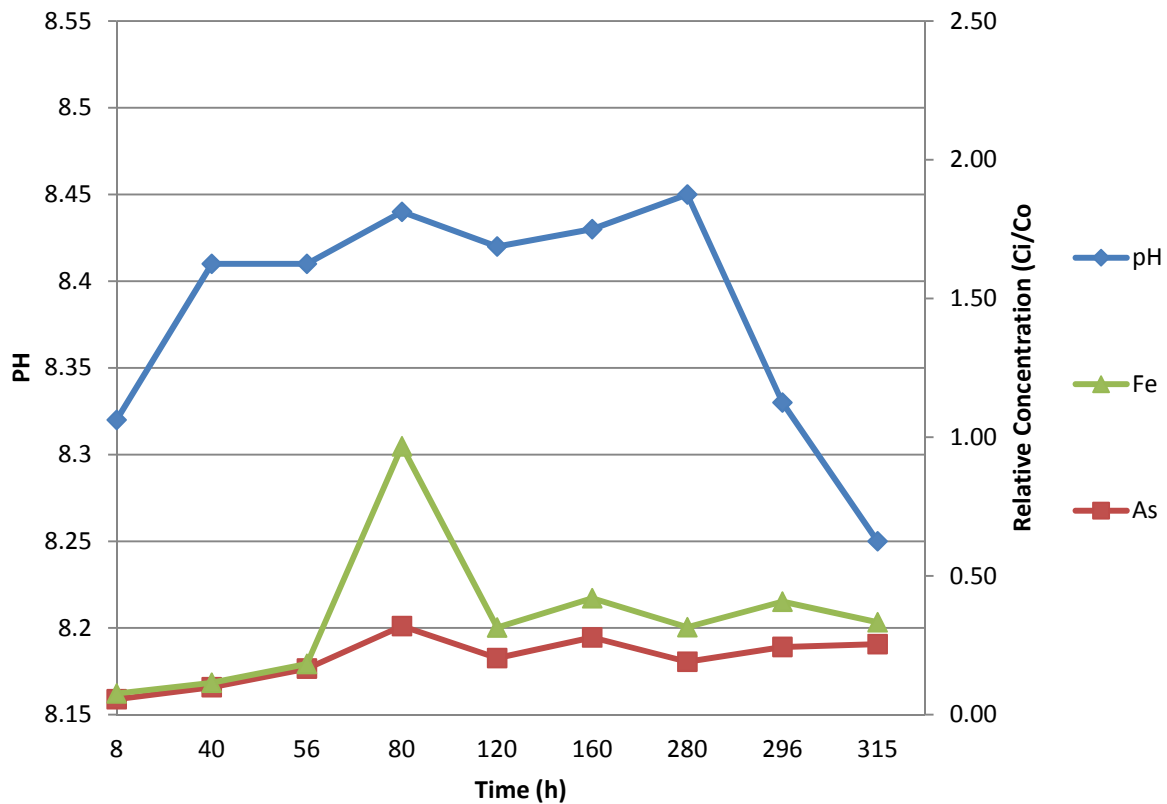


Figure 15: Breakthrough curve for Mix-2 test showing iron, arsenic and pH trends.

Table 11: Change in pH at different times in relation with the Fe and arsenic concentration in the effluent during the Mix-2 test.

Time (h)	pH	As (µg/L)	Fe (µg/L)
0	7.91	34.34	1106.53
8	8.32	1.910	23.00
40	8.41	3.399	17.90
56	8.41	5.709	18.50
80	8.44	10.965	717.07
120	8.42	7.005	122.00
160	8.43	9.556	156.71
280	8.45	6.559	137.35
296	8.33	8.382	180.62
315	8.25	8.735	87.08

The drop in the pH noted in Table-11 between 280h and 315h could be due to the utilization of OH⁻ ions released as a result of a ligand exchange reaction for the protonated arsenic species presented under the section Mix-1 by the Fe³⁺ ions released as a result of oxidation of Fe²⁺ ions (Figure 15). The increasing trend observed in the arsenic concentration during this time interval could be due to the competition provided by the V, Ni, Zn, Se for the HFO adsorption sites. The change in pH can change the selectivity or the relative affinity preferences among various species.

Table 12: Concentration of elements significantly correlated to Arsenic in the effluent of 3:10 mixture of the water collected from the town of Wabana and Freshwater (Mix-3)

Time (h)	Initial As (µg/L)	As (µg/L)	Al (µg/L)	Fe (µg/L)
8	34.32	1.540	8.563	100.00
40		2.677	5.076	17.180
56		5.781	16.338	86.659
80		10.944	23.02	588.169
120		11.457	34.274	665.795
160		14.338	35.631	1086.34
280		7.677	15.274	164.30
296		13.270	30.207	690.039
315		12.315	21.529	518.16
Correlation with Arsenic		1.000	0.907	0.915

According to the Vincent (1995), for data of 9 different samples, a significant correlation should be of value 0.666 or more. It could be seen from Table 12 that arsenic was significantly

correlated to Fe and aluminum (Al). A similar kind of response was noted in this case as that of the sample mix-2. The As concentration at 160h increased to 14.338 $\mu\text{g/L}$ which dropped to 12.315 $\mu\text{g/L}$ after 315h. Similarly, the Fe concentration dropped from 1086.34 $\mu\text{g/L}$ to 518.16 $\mu\text{g/L}$ during the same period. The oxidation of soluble Fe^{2+} ions to the insoluble Fe^{3+} ions as a result of operation of the pump could be the main reason for this drop in arsenic concentration. Further, in the literature reviewed, the oxides of aluminum have been found to have a positive effect in mitigating the arsenic concern (Jeong, et al. 2007 and Giles, et al. 2011). But, the authors reported that the oxides of iron were more suited in adsorbing As(V) over oxides of aluminum. Therefore, the positive correlation found between Al and arsenic in this study was in accordance with the literature cited. The breakthrough curve in relation with the pH values is depicted in Figure 16. Arsenic breakthrough did not occur as the arsenic level even after treating 39000mL of water for Mix-3 reached to 12.315 $\mu\text{g/L}$.

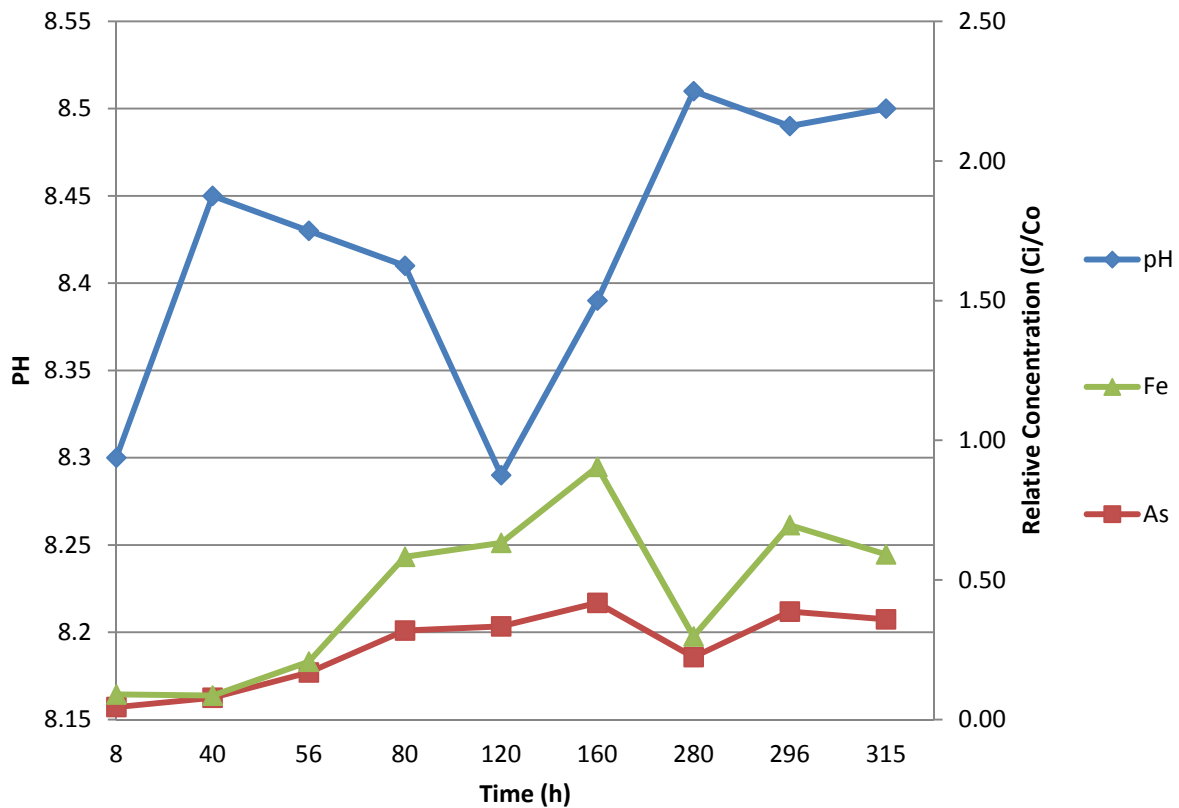


Figure 16: Breakthrough Curve for Mix-3 test showing iron, arsenic and pH trends.

The pH values taken during the course of experiment for Mix-3 are listed in the Table-13.

Table 13: Change in pH at different times in relation with the Fe and arsenic concentration in the effluent during the Mix-3 test.

Time (h)	pH	As ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)
0	7.80	34.32	2229.43
8	8.30	1.540	100.00
40	8.45	2.677	17.180
56	8.43	5.781	86.659
80	8.41	10.944	588.169
120	8.29	11.457	665.795
160	8.39	14.338	1086.34
280	8.51	7.677	164.30
296	8.49	13.270	690.039
315	8.50	12.315	518.16

It is concluded from the above discussion that the sample Mix-2 performed best as compared to the Mix-1 and Mix-3. After running the experiments for approximately 325h for Mix-1 and 315h for both Mix-2 and Mix-3 and passing 38500mL for Mix-1 and 39000mL each for Mix-2 and Mix-3, it was concluded that Fe/As ratio (wt/wt) of 32.227 in the case of Mix-2 gave the most desired results. The Mix-1 test was low in Fe concentration and further, the competition arsenic ions faced from other competing ions such as Cu, Ni, and Zn for HFO adsorption sites hampered the efficiency of arsenic removal. Moreover, the Fe concentration in case of Mix-3 test was on the higher end that resulted in quickly filling up the pore spaces in the sand bed and consequently; decreased the arsenic adsorption. Even though, the drop in pH noted during the period of 80h and 160h did not improve the As removal as the Fe concentration in the effluent was found to be increased. The higher the Fe content recorded in the effluent, the higher the arsenic concentration was reported. On the other hand, the best combination of Fe and As concentration was found in the case of Mix-2 and the results presented in Table 8 reflected that even after running the experiments for 315h the arsenic concentration, although it stayed above the limit of $7\mu\text{g/L}$ set for this study, still it stayed below the limit of $10\mu\text{g/L}$ defined by Health Canada.

It can be seen from Figure 13 that with a Fe input of $1106.53\mu\text{g/L}$, a total of $960.73\mu\text{g}$ of arsenic was removed. Similarly, $900.48\mu\text{g}$ of arsenic was removed with the Fe input of $2229.43\mu\text{g/L}$ for Mix-3. Moreover, in the case of Mix-1, a total of $783.62\mu\text{g}$ of arsenic was removed with the Fe input of $580.26\mu\text{g/L}$ (Figure 17). The comparison was made for the treated volume of 35000mL for each of the Mix-1, Mix-2 and Mix-3 tests. The amount of arsenic removed was calculated in the same way as described earlier in the report for RO-4 and RO-4(b) tests. It could be concluded that Mix-2 performed better than either of the Mix-1 or Mix-3 water samples. Further, the effluent arsenic concentration in Mix-2 ($8.74\mu\text{g/L}$) for the treated volume of 39000mL was lower than either the Mix-1 ($23.93\mu\text{g/L}$) or Mix-3 ($12.32\mu\text{g/L}$) for the treated volumes of 38500mL and

39000mL, respectively. Therefore, it was proposed that the Mix-2 water sample had the ideal composition due to which increased arsenic adsorption was noted as compared to either of the Mix-1 or Mix-3. The study of the combination of the elements of Mix-2 water sample responsible for this remarked behaviour is beyond the scope of this research and hence; it was not explored any further.

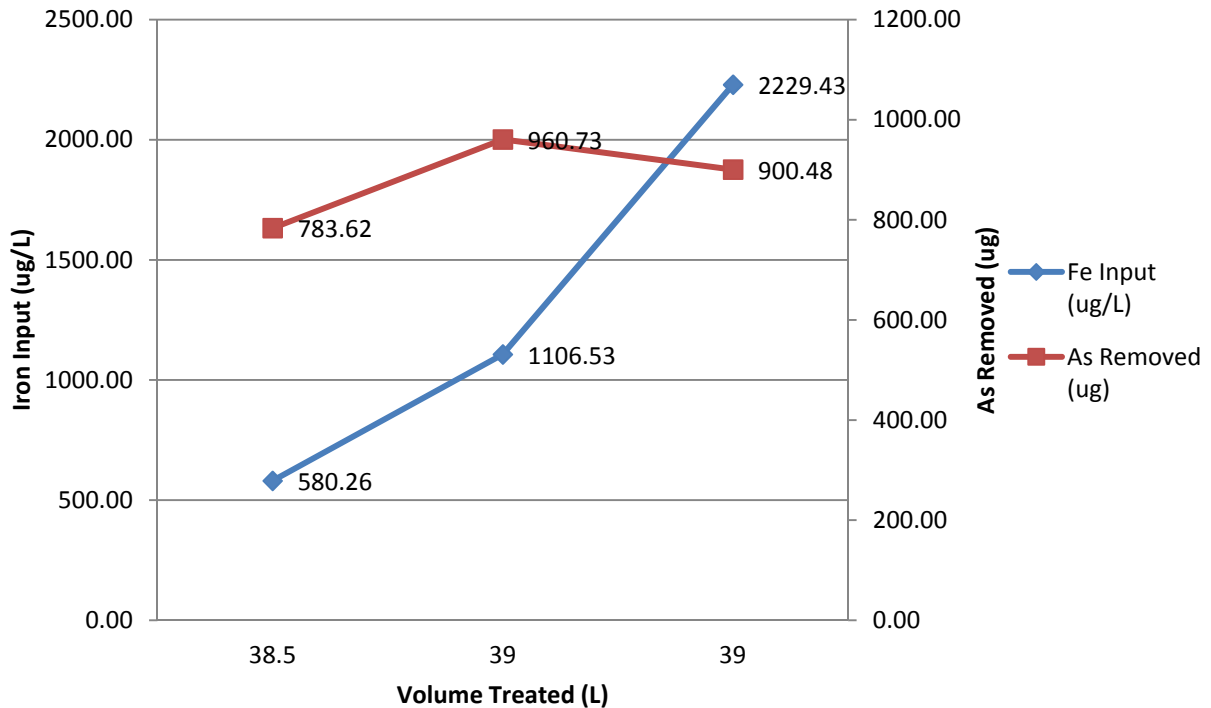


Figure 17: Comparison of Cumulative arsenic removal vs Iron Input for Mix-1, Mix-2 and Mix-3 water sample

Conclusions

It is concluded that HFO can play a very vital role in mitigating the arsenic concern in the groundwater of the Newfoundland and Labrador. The higher the iron content, the greater will be the arsenic removal but the sand filter replacement frequency may decrease. As a result of the deposition of iron oxides in the pore space, the filtration rate decreases along with the filtration efficiency. Further, the incorporation of aeration in the filtration system can significantly enhance the arsenic adsorption. Moreover, the dilution of the high iron and arsenic containing water sample with the low iron and arsenic containing water can significantly improve the arsenic adsorption. The mixture should neither be too lean nor too rich in the iron concentration

as both have negative impact on arsenic adsorption. It is therefore suggested to use sand filtration in combination with aeration and dilution to deal with the growing arsenic concern in the province of the Newfoundland and Labrador.

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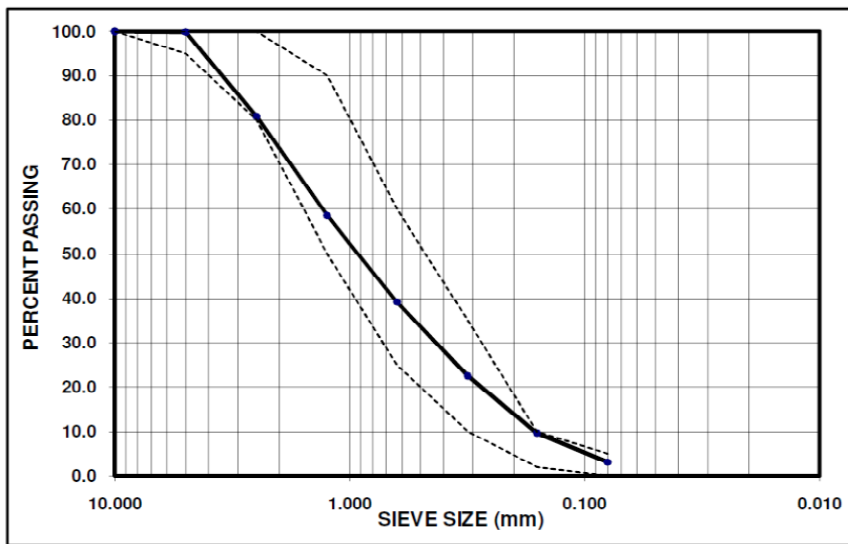
APPENDIX-A

S/N	Sample	Units	Wabana	Freshwater	Wabana Normal
1	Li 7	µg/L	7.31	4.13	27.16
2	Be	µg/L	<5.26	0.28	<1.55
3	B	µg/L	21.02	18.19	37.12
4	Mg	µg/L	3151.95	7894.09	5352.69
5	Al	µg/L	34.69	5.64	<2.3
6	Si	µg/L	5986.58	<12165.6	4932.59
7	P	µg/L	78.94	<909.5	18.20
8	S	µg/L	<7235	11979.47	10655.62
9	Cl	µg/L	37387.19	100101.95	36489.57
10	Ca 43	µg/L	25516.55	36224.42	38389.71
11	Ti	µg/L	7.31	2.11	13.20
12	V	µg/L	0.46	<16.94	<0.64
13	Cr 52	µg/L	2.02	6.05	1.39
14	Fe 54	µg/L	11436.92	<507.52	86.33
15	Mn	µg/L	2819.01	26.05	179.60
16	Co	µg/L	6.04	0.33	<0.21
17	Ni	µg/L	11.64	1.88	<1.68
18	Cu	µg/L	9.55	9.25	<0.57
19	Zn	µg/L	95.74	25.63	4.45
20	As	µg/L	62.70	29.71	4.50
21	Se	µg/L	12.82	<105.65	<6.23
22	Br	µg/L	68.19	131.69	89.15
23	Rb	µg/L	2.25	9.55	2.46
24	Sr	µg/L	157.55	203.08	425.45
25	Mo	µg/L	0.86	2.98	0.23
26	Ag	µg/L	<0.05	<0.75	<0.05
27	Cd	µg/L	<0.36	<3.31	0.15
28	Sn	µg/L	<0.23	<1.14	<0.20
29	Sb	µg/L	<0.09	4.90	<0.05
30	I	µg/L	24.57	8.39	14.49
31	Cs	µg/L	0.05	0.12	0.07
32	Ba	µg/L	162.18	45.07	91.57
33	La	µg/L	0.23	1.32	0.02
34	Ce	µg/L	1.09	0.01	0.01
35	Hg	µg/L	<0.13	<2.01	<0.07
36	Tl	µg/L	0.03	0.03	0.00
37	Pb	µg/L	3.35	22.03	0.03
38	Bi	µg/L	<0.03	<0.17	<0.01
39	U	µg/L	0.08	0.48	0.02



GRADATION ANALYSIS REPORT

Project No:	TF1030119	Lab ID:	3516
Project:	Aggregate Gradation Analysis Foxtrap, NL	Sample Type:	Washed Concrete Sand
Client:	Capital Ready Mix	Date Sampled:	10-Jun-11
Sampled By:	M. Lynch of AMEC	Date Tested:	10-Jun-11
Location:	Loading Face of Working Stockpile	Sample Source:	Black Mountain



SIEVE SIZE (mm)	PERCENT PASSING	GRADING LIMITS	
		Upper	Lower
10.000	100.0	100	100
5.000	99.8	95	100
2.500	80.8	80	100
1.250	58.5	50	90
0.630	39.2	25	60
0.315	22.5	10	35
0.160	9.7	2	10
0.080	3.1	0	5

Comments: *The sample gradation meets the project specifications.*

The FM has been determined to be 2.9; CSA standard requires an FM between 2.3 and 3.1.

Reporting of these test results constitutes a testing service only.
Engineering interpretation or evaluation of the test results is provided only on written request.

AMEC Earth & Environmental

Per: *[Signature]*

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FM-MAT-57-03

APPENDIX-C

S/N	Sample	Units	Mix-1	Mix-2	Mix-3
1	Li 7	µg/L	3.53	3.79	4.43
2	Be	µg/L	<3.41	<3.62	<3.24
3	B	µg/L	13.45	12.76	15.87
4	Mg	µg/L	8754.15	8229.46	7324.02
5	Al	µg/L	5.77	7.48	14.72
6	Si	µg/L	3704.35	4218.99	3794.10
7	P	µg/L	<64.9	<68.9	<61.7
8	S	µg/L	<66807.7	<70919.3	<63484.1
9	Cl	µg/L	129649.26	121401.90	107490.37
10	Ca 43	µg/L	47306.64	46486.27	43786.58
11	Ti	µg/L	<16.64	<17.67	<15.81
12	V	µg/L	<2.02	<2.15	<1.92
13	Cr 52	µg/L	0.71	<2.96	<2.65
14	Fe 54	µg/L	580.26	1106.53	2229.43
15	Mn	µg/L	195.92	375.47	607.32
16	Co	µg/L	3.62	1.65	0.28
17	Ni	µg/L	2.10	4.18	3.90
18	Cu	µg/L	8.26	7.44	9.81
19	Zn	µg/L	54.92	119.69	173.81
20	As	µg/L	33.58	34.34	34.32
21	Se	µg/L	<7.40	<7.86	<7.03
22	Br	µg/L	163.07	164.08	137.46
23	Rb	µg/L	0.93	1.24	1.46
24	Sr	µg/L	273.53	270.59	254.22
25	Mo	µg/L	3.10	2.93	2.50
26	Ag	µg/L	<0.04	0.04	<0.04
27	Cd	µg/L	<0.46	<0.49	<0.44
28	Sn	µg/L	<0.37	<0.39	0.39
29	Sb	µg/L	2.53	2.27	2.03
30	I	µg/L	10.41	<11.37	12.00
31	Cs	µg/L	0.11	0.12	0.13
32	Ba	µg/L	14.60	27.03	44.30
33	La	µg/L	0.04	0.07	0.10
34	Ce	µg/L	0.10	0.15	0.22
35	Hg	µg/L	<0.25	<0.27	<0.24
36	Tl	µg/L	0.03	0.02	0.04
37	Pb	µg/L	2.00	4.72	1.79
38	Bi	µg/L	<0.03	<0.03	<0.03
39	U	µg/L	0.65	0.73	0.50



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