Arsenic Removal by Adsorption and Multilayer Sand Filtration

(CIE 5050-09 Additional Graduation Work)

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Abstract

The presence of Arsenic in the groundwater, and eventually in the drinking water is a serious problem in many of the Asian countries. Due to it high toxicity to its removal is very essential. There are many removal technologies for the removal of arsenic such as adsorption, coagulation-filtration, membrane filtration, ion exchange and precipitation. Adsorption is one such technology which is mainly used as it's a very simple technique. the extent of adsorption is affected by pH, nature of the adsorbent, surface area of the adsorbents. Iron is widely used for the removal of Arsenic from groundwater.

The iron which is present in dissolved form in groundwater needs to first get oxidised to hydrous ferric oxides as flocs which provide adsorption sites for arsenic. In this study the effect of pH and Iron dosage on the arsenic removal by adsorption with the available dissolved Iron in the groundwater. The effect of pH on arsenic removal by floc formation of iron with a series of Jar tests were done for Arsenite and Arsenate. Also, the removal of arsenic in a multilayer sand bed (3 layers with Anthracite, Sand and Garnet) was evaluated at three different pH.

It was found that pH plays a very important role on Arsenic removal. Arsenite removal was increased with pH whereas Arsenate removal decreased with increase in pH. At pH 5 and at high Fe concentrations, the removal decreases but the effective iron or the iron that flocculated to greater than 0.45µm was quite similar. This was also seen in the particle counter analysis. Iron concentration of 5mg/l was enough to remove Arsenate upto 90% whereas Arsenite needed higher doses of upto 20mg/l to reach 90%. Clear increase in the flocculation was observed in particle counts with the increase of iron concentration.

Before the dosing of Iron Arsenic oxidation was seen in the filter bed maybe due to the presence of Arsenic Oxidising Bacteria (AOB). High oxidation efficiency was seen Sand and Garnet layer. The oxidation of Arsenite to Arsenate before dosing iron in the filter was less at pH 8. High resistance was observed in the filter bed within 3 days after backwashing although the removal was quite stable in the three days. After dosing of Iron, high removal of arsenic was seen in Anthracite layer due to the high adsorption of Iron in the Anthracite layer.

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Chapter 1

1 Introduction

1.1 Global Problem

In the modern world, with the decline of natural resources the humans are exposed to various types of threats. There are numerous hazardous chemicals around us which can affect the human health. Arsenic (As) which is a toxic metalloid and is 20th most abundant element in the Earth's crust has adverse health effects on humans in areas where it is found in high concentrations in soil, water and air (Sarkar & Paul, 2016). Humans can be exposed to As through contaminated drinking water, using contaminated water in food preparations, smoking tobacco or anthropogenic sources (use of As in medicine, industrial pollution, mining etc.). (Centeno et al., 2007). Arsenic has four oxidation states (+V, +III, 0 and -III) depending on the redox potential and pH. Among which +V (Arsenate) and +III (Arsenite) are most common in aqueous environment. As(V) is found in oxidative environment and As (III) is found in reductive environment. As(III) exists as uncharged species (H_3AsO_3) or anionic species ($H_2AsO_3^-$) and As(V) exists as $H_2AsO_4^-$ or $HAsO_4^{2-}$ anions in the natural water (Babaeivelni et al., 2014). Arsenic is released into the environment due to natural and anthropogenic activities. Natural processes include weathering of arsenic containing rocks, mineral dissolution, volcanic activities and Geothermal waters (Tabbal, 2003). Anthropogenic activities include mining, smelting of arsenic bearing minerals, discharge of industrial As waste and application of arsenic herbicides and pesticides (Sarkar & Paul, 2016; Wan et al., 2011).

Considering the toxicity of arsenic, the World Health Organization has set the maximum limit of acceptable level in drinking water at 10µg/l (World health organisation, 2008). It is estimated that 140 million people worldwide are drinking As contaminated water (World health organisation, 2018, February 15). Arsenic is largely found in the alluviums of the Indian states of West Bengal, Assam, Bihar, Jharkhand, Uttar Pradesh and Bangladesh. Many other parts of Asian countries like Nepal, China, Mongolia, Myanmar, Thailand, Taiwan, Vietnam, Cambodia, Sri Lanka, Mexico, Bolivia and Argentina are fairly affected by arsenic toxicity. Bangladesh and North Eastern parts of India are the worst hit with concentrations of 50 µg/l. Almost half of the Bangladesh citizens are at risk of consuming arsenic contaminated tube wells. It is estimated that in 2012 about 39 million people in Bangladesh were exposed high concentrations of Arsenic (World health organisation, 2018, February 15) . The problem is not confined to only these countries. High concentrations of As are found in communities in Iran, Australia, New Zealand, parts of European Union, Iceland Brazil, Canada, USA and many other countries. A map of arsenic affected countries is shown in Figure 1 and the approximate number of people affected by groundwater containing Arsenic is given in Table 1.

Figure 1 Arsenic affected countries of the World (Centeno et al., 2007)

Table 1 Groundwater arsenic concentration and approximate number of affected people (Sarkar & Paul, 2016)

Country/Region	**Groundwater As Concentration in µg/l**	**Approximate Size of population at risk**
India	<10-3200	70,400,000
Bangladesh	<1-2500	32,000,000
China (mainland china)	50-2000	>2,300,000
Vietnam	1-3050	>100,000
Thailand	1->5000	15,000
Taiwan	10-1820	200,000
Inner Mongolia	<1-2400	600,000
Argentina	<1-9900	2,000,000
Chile	100-1000	400,000
Mexico	8-620	400,000
Hungary, Romania	<2-176	400,000
Greece	1-1840	150,000
Spain	<1-100	>50,000
U. K	<1-80	Minimal
U.S.A., Canada	<1->10,000	Minimal
Ghana	<1-175	<100,000

1.2 Toxicity of Arsenic

The International Agency for Research on Cancer (IARC) has classified arsenic and its compounds as toxic, mutagenic and skin and lung carcinogenic to humans. Many different parts of the body are affected by chronic exposure of inorganic arsenic. Arsenic enters human body through various ways. One way of human exposure to arsenic is by ingestion of As contaminated food and water. Arsenic can also enter due to occupational circumstances due to the exposure of Arsine gas, which is a colorless, odorless, tasteless, non-irritating gas causing rapid destruction of red blood cells (Saha et al., 1999). Arsenic effects humans differently depending on the diet, health and causes methylation in the body upon oxidizing Arsenite and Arsenate. Arsenic can also be absorbed by inhalation and dermal absorption (Karim, 2000). It is undetectable in the primary stage whereas long term exposure to arsenic leads to chronic poisoning and occurs between the age of 8 to 14 years (Jha et al., 2017). The toxicity of arsenic is of three types – acute, subacute and chronic. Acute poisoning involves vomiting, abdominal pain and diarrhea followed by numbness, muscle crumpling and death in extreme cases (World health organisation, 2018, February 15). Chronic arsenic poisoning (arsenicosis) is mostly observed in people who live endemic areas with high arsenic concentrations in drinking water or in burning coal and occupational exposure (Ng et al., 2003). Skin, liver, nervous system, respiratory system, renal system are vulnerable to chronic arsenic poisoning (Sarkar & Paul, 2016). Arsenicosis includes several kinds of skin problems, skin cancer, lung and kidney cancer and diabetes. A severe disease of the blood vessels has been identified in Taiwan known as "black foot disease". Malnutrition contributes to the development of this disease (World Health Organisation, 2001). Other effects of Arsenic include infertility, infant mortality, negative impacts on cognitive development and intelligence (World health organisation, 2018, February 15).

Arsenic exposure to humans can also be due to diet. About 90% of the dietary arsenic in US comes from seafood (Borak & Hosgood, 2007). It is usually present in organic form in the marine life. Inorganic arsenic is absorbed when rice, grains and other vegetable are cooked in arsenic contaminated water. It is found as Arsenobetaine (AB), Arenosugars (As Sugars) and Arsenolipids (As Lipids) which are metabolized when taken up by humans. The toxicity of these compounds on humans and experimental animals has not been characterized yet (Taylor et al., 2017). The level of arsenic in the marine organisms lie within the range of 5-100 µg/g dry mass. Terrestrial food contains less than 0.05µg/g. Rice can typically contain about 0.1-0.4µg/g (Francesconi, 2010). Lower rate exposure of the order 0.01mg/kg per day or higher can also lead to hyperpigmentation after intervals of 5 to 15 years.

Although there are uncertainties of the role of Arsenic in the human health risks, the research done is enough to clearly say that the presence of Arsenic in extremely low concentrations is also very unsafe to humans.

1.3 Arsenic Removal Technologies

Removal of Arsenic depends on the chemistry and composition of the As contaminated water. Arsenic exists in water in two inorganic forms – Pentavalent arsenate, As (V) and Trivalent Arsenite, As (III). Under environmental relevant pH range 4-10, As (III) is dominant and exists as neutrally charged compared to As(V) that are negatively charged. Hence it has been found that for effective removal of arsenic As (III) has to be first oxidized to As(V) (Nicomel et al., 2015). Most available technologies for Arsenic removal are by Oxidation, Coagulation-flocculation, Membrane Filtration, Adsorption and ion exchange. Scientists have also developed biological processes for arsenic removal by ingenious bacteria to catalyze the chemical processes. Phytoremediation, microbial remediation, bio-filtration process have given promising results under the lab scale and need to be tested under real-scale conditions (Duarte et al., 2009; Sarkar & Paul, 2016).

1.3.1 Pre – Oxidation

In the drinking water pH arsenic exists as As (III) species and must be oxidized to As(V) to be efficiently removed. In order to increase the oxidation process ozone, chlorine, chlorine dioxide, Monochloramine, Hydrogen peroxide are used as oxidants (Bissen & Frimmel, 2003). Oxidation through atmospheric oxygen takes several weeks to oxidize (Ahmed, 2001). Photocatalytic oxidation (combination of UV and H_2O_2) gave 70% oxidation efficiency at a UV dose of 2,000mJ/cm^2) (Sorlini et al., 2010). The oxidation rate is inhibited due to the presence of Fe (II), Mn(II), Sulphur and Total organic carbon are present in the water (Shankar & Shanker, 2014). The use of chlorine can lead to the formation of chlorinated byproducts which are hazardous to the human. Thus, suitable oxidants must be chosen for efficient oxidation of As (III) to As(V).

1.3.2 Coagulation – Flocculation

Coagulation and flocculation (or simply flocculation) are the most common techniques used for arsenic removal. Coagulation involves addition of a coagulant (aluminum sulphate or ferric sulphate) which destabilizes the negatively charged colloids. Flocculation acts as a bridge between the flocs and helps in binding into large clumps which can then be filtered in later processes (Choong et al., 2007). The pH of the water influences the removal. Optimum pH for effective removal varies with the type of coagulant added. Iron works efficiently under a pH range of 7.2-7.5 (Ahmed, 2001; Nicomel et al., 2015). Fe based coagulants are more efficient in arsenic removal than alum with a removal efficiency of 90% and 70% of As(V) and As(III) respectively (Shankar & Shanker, 2014). For an efficient removal, As (III) must be pre-oxidized to As(V) to get adsorbed onto the coagulant flocs (EPA, 2000). Coagulants such as zirconium (IV) chloride, titanium chloride (III and V), Ferric and Titanium Sulphate have showed moderate removal for As(V). Management of the sludge produced during this process is costly which makes this process less feasible (Sarkar & Paul, 2016).

1.3.3 Adsorption and Ion Exchange

Adsorption is a traditional method to separate the solutes from solvent or gases by the accumulation at the surface of adsorbent. Adsorption is driven by Vander Waal forces and electrostatic forces between the adsorbate and adsorbent. Adsorption is widely used in the treatment of drinking water as it has high efficiency, low cost, sludge free and easy to handle. This process can be described into two processes – Coagulation Adsorption and Ion Exchange adsorption. Coagulation Adsorption involves the formation of colloidal particles with the addition of coagulants and thus getting adsorbed onto the coagulate (Gallegos-Garcia et al., 2012). Activated alumina is packed in beds to remove contaminants. Activated alumina is considered to be adsorption process although it removes Arsenic by the exchange of As ions to the surface hydroxides on the Alumina (EPA, 2000). Ion exchange is a special type of adsorption where adsorbed ion is the displaced by the dissolved ion and can remove Arsenic up to 95%. Strong base anion exchange resins are used which are effective over a large range of pH. These resins have high affinity towards As(V) (Donia et al., 2011) . The efficiency of Arsenic removal by Ion exchange is affected by the competition of other background ions which are present in the water. Due to this competition of other ions over Arsenic, arsenic is not removed from the water which often leads to chromatography peaking. To avoid this the bed must be regularly monitored and regenerated (EPA, 2000). The presence of high Fe(III) in the water leads to formation of complex forms of Arsenic with iron and are difficult to remove witthese resins (EPA, 2000).

1.3.4 Membrane Filtration

Membrane filtration uses membranes which are synthetic materials with many pores which act as a barrier to avoid the passing of some constituents through them. Pressure difference between the feed water and the permeate water is used as the driving force for these barriers (Oakes, 2005). Based on the pore size and operating pressures of the membranes they are classified into four types namely – microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) (Oakes, 2005). The removal efficiency in UF can be increased by adding a cationic surfactant can bind to the negatively charged arsenic (Nicomel et al., 2015). Membrane technologies are efficient in removing Arsenate than Arsenite, thus pretreatment for oxidizing As (III) to As(V) is needed (Mudhoo et al., 2011) . Arsenic is removed by filtration, electric repulsion and adsorption in these processes. The repulsion of the membranes for arsenic depends on the size and chemical characteristics of arsenic. Membrane technologies are expensive if treated at high pressures. Sufficient research is needed for removing As(III) at low pressures (Mudhoo et al., 2011).

1.4 Study Objective

The aim of this research was to study the process of adsorption of As (III) and As (V) with the available Fe (II) in the water. Fe (II) present in the water is first needs to be oxidized to Fe (III) to be able to remove Arsenic by adsorption. Iron flocs are formed which act as the sites for adsorption of Arsenic and are pH dependent.

Thus, the following objectives are taken in this research

1. To see the effect of pH on arsenic removal by floc formation of iron by performing a series of jar tests using As (III) and As(V).
2. To see the performance of Arsenic removal by multi-layer sand filtration using a pilot setup.

The following steps were undertaken to complete this study: -

- Preparation of the Stock solutions of Arsenic and Iron for Jar tests and Multilayer sand filtration were done separately.
- For Jar test pH range from 5-9 with Fe concentration of 0, 0.1, 0.5, 1,2 ,5 ,10 and 20 mg/l were chosen. Arsenic concentration of 5, 10, 50, 100, 500 and 1000 µg/l were taken for both As (III) and As(V) separately.
- Particle counter analysis of the Jar test solutions at the end of 60min was done to see the floc formation.
- Arsenic removal in the Multi – layer Sand filtration Pilot consisting of 6 columns with 3 layers (Anthracite, Sand and Garnet) at 3 different pH.

Chapter 2

2 Review of Literature

In 2005, Olivier and Stephan have done batch studies to reduce As (III) from 500 µg/l to 50µg/l in synthetic groundwater using zero valent iron. The studies were done for developing an efficient filtration column for households in Bangladesh. The studies were done with synthetic groundwater containing 500µg/l As (III), 2-3 mg/l P, 20mg/l Si, 8.2 mM HCO_3^-, 2.5 mM Ca^{2+}, 1.6 mM Mg^{2+}, at pH 7. The experiments were done in columns with 1.5g iron and 3-4g quartz sand and found that by passing the water four times into the filter arsenic concentration was reduced. It was found that due to subsequent filtration led to oxidation of As (III). Due to the decrease in the dissolved oxygen Fe (II) was oxidized in the column and formed hydrous ferrous oxide. The oxidation of Fe (II) lead to the increase in the oxidation of As (III). Phosphate acted as a corrosion inhibitor forming scales on the surface of iron fillings, which lowered the Fe (II) release and As (V) adsorption in the column. As (III) removal by Fe (II) was almost 90% even along with phosphate removal. But with addition of Fe (III), As (III) oxidation was not observed and as a result only 20% was removed. By adding 15mg/l Fe (II) it was enough to remove 90% of Arsenic (III) without any addition of oxidant but with Fe (III) almost 80mg/l was needed.

Kanel et al., (2006) studied As (V) removal by using Nano scale zero valent iron (NZVI). Batch experiments were done to investigate the influence of pH, adsorption kinetics, sorption mechanism and anion effects. NZVI are capable to remove variety of pollutants with halogenated hydrocarbons. A wide range of pH was investigated from pH 3-11 to see the adsorption of Arsenic. It was found that 100% total arsenic sorption was observed to decrease to 84% at pH 9 and 37.9% at pH 11 which shows that NZVI are effective in acidic and neutral pH. Due to the presence of high dissolved carbon, sulphate and phosphate high amounts of NZVI was needed to remove for complete removal of As(V) from the groundwater. 0.4g/l of NZVI was required to remove 1mg/l Arsenic (V) from the groundwaters of Bangladesh and West Bengal to reach 100% removal.

Kanel et al., (2005) also did batch studies on removal of As (III) from the groundwater of Bangladesh and Nepal. No As (III) removal was seen without the addition of NZVI. At 4.5g/l NZVI was needed for 100% removal. But a low concentration of NZVI was enough if combined with $FeCl_3$. A greater amount of NZVI was needed for As (III) as compared to As (V) due to the presence of anions and trace elements of silica.

Robert, L. C., et al (2004) did studies to compare the removal efficiencies of arsenic with Fe (II) with Fe (III) in groundwater. The study was done on the synthetic water which represented the composition of the groundwater in Bangladesh. The studies show that a concentration of 3.8mg/l Fe (II) was enough to remove 90 % As (V). The studies show that with the addition of Fe (II) the As (III) was decreased. The decrease was not linear but showed a threshold at 0.4 – 0.5. The overall oxidation of As (III) is limited with single addition of Fe (II) as even Fe (II) consumes reactive oxidants. Multiple addition of Fe (II) led to more oxidation of As (III) and removal. For practical treatment of groundwater in Bangladesh, the Fe (II) which is present in the groundwater is mostly less than 30mg/l which is not enough for passive arsenic removal. The addition of Fe (II) or Fe (III) is needed to oxidize and remove arsenic.

Chapter 3

3 Materials and Methods

The details of the experimental study in terms of the chemical and their properties, setup used for the study and experimental procedure are given in this section. The research was conducted in the Sanitary lab of TU Delft between 6th July to 3rd September 2018.

3.1 Chemicals

The chemicals for the experiment were obtained from Sigma Aldric were used as such. All the reagents and standards were prepared using demi water. Salts of Sodium (Meta) Arsenite ($NaAsO_2$) and Sodium Arsenate dibasic heptahydrate (Na2HAsO4.7H2O) were used for the preparation of As (III) and As(V) stock solutions respectively. To avoid the oxidation of the arsenic, hydrochloric acid (2M) was added to the stock solution lower the pH and in the prevailing acidic condition arsenic oxidation will be lower. Salts of Iron Sulphate Heptahydrate (FeSO4.7H2O) were used for Fe (II) stock solution. The Stock solutions prepared for the experiments are shown in Table 2. The pH during the running of the experiment was adjusted by the addition of 0.1M Hydrochloric acid and Sodium Hydroxide (NaOH) solutions as per the requirement.

Table 2 Stock Solutions for Jar test

Parameter	Chemical added
Arsenic (III)	173 mg/ 100 ml
Arsenic (V)	416 mg/ 100 ml
Iron (II)	500 g / 100ml

3.2 Setup

3.2.1 Jar Test

A typical Jar testing apparatus was used for the experiment with 6 different baffled jars of capacity of 2litre each filled with 1liter of tap water each with initial pH value of 7.5-8. The arsenic concentration was added to each jar with increased amount of the stock solution as shown in Table 3. The experiment was carried out in an ambient temperature. 40 different runs were performed with varying Fe (II) concentration and pH for As (III). 30 different runs were performed with varying Fe (II) concentration and pH for As (V) as 100% removal was seen at 5mg/l of Fe. Fe (II) solution was added after 1-2 min after adding Arsenic till the colorless solutions were well mixed. The water composition evaluated in the Jar test are shown in Table 4. The pH in each jar was adjusted at the beginning of each run and maintained constant throughout the run by adding HCl or NaOH as required. The mixing speed of the stirrer was kept at 55rpm and each run was performed for 60min. Figure 2 shows the Jar test setup used in the lab.

Table 3 Stock Solution added for reaching the desired concentration of Arsenic in the Jars

Jar	µl of As Stock added	µg/l Arsenic dosage
1	5	5
2	10	10
3	50	50
4	100	100
5	500	500
6	1000	1000

Table 4 Parameters used for Jar test analysis

Parameters	Range of values
Initial Arsenic concentration	5, 10, 50, 100, 500 and 1000 µg/l
Arsenic Oxidation state	As (III) and As (V)
pH	5, 6, 7, 8, 9
Iron (Fe 2+)	0, 0.1, 0.5, 1, 2, 5, 10, 20

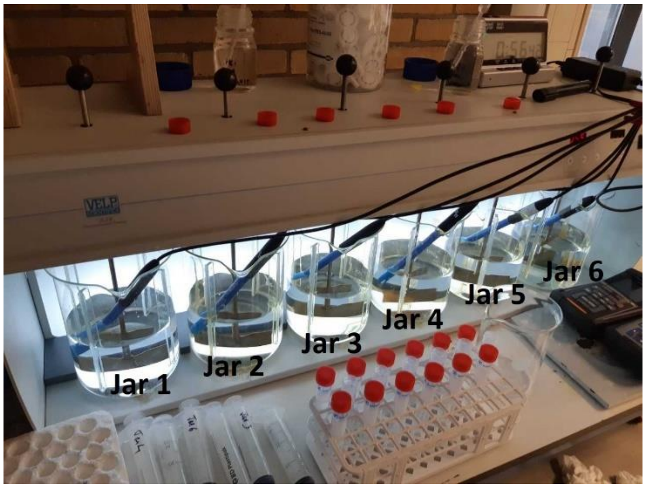


Figure 2 Jar Test Apparatus with individual pH meters in each jar

3.2.2 Multilayer Sand Filtration

Column tests were performed for multilayer sand filtration. Six columns of 2m height and 9.2cm inner diameter were set up with 3 different layers of materials with three different pH. 2 columns for each pH. Sand of different grain size were used in the layers. The bottom 1m of the column was filled with three layers - 30cm of Anthracite, 40 cm of sand and 30 cm of Garnet. Properties of the filter material used are given in Table 5. The arsenic solution and Iron solution were dosed through a main pipe which distributed 20µg/l and 2mg/l respectively in each column at a rate of 27 – 30 l/h. 20l of stock solutions of Arsenic solution and 10l of iron solution were prepared. Column 1 and 2 were maintained at a pH 8, column 3 and 4 were kept at pH7.5 and column 5 and 6 at pH 7. Backwashing was done with clean tap water at a velocity of 105m/h. Sampling points were made at every 10 cm through the bed for collecting the samples. Figure 3 shows the multilayer sand filtration setup.

Table 5 Properties of the materials used in the Multi-layer sand Filtration columns

	Grain size (mm)	Porosity (%)
Anthracite	0.8-2	47-52
Sand	0.4-0.8	40-43
Garnet	0.2-0.4	45-58

Figure 3 Multilayer Sand Filtration columns

3.3 Sampling

3.3.1 Jar Test

The sampling process is quite straight forward. Mainly samples were collected for three tests from during the performance of Jar test. The three type of test and processes of sampling are given below.

- ***Sampling for ICP-MS (Inductive Coupled Plasma Mass Spectrometry)*** – the mass spectrometry can detect the metals and non-metals at a very low concentration. For this research only, the concentration of Arsenic and Iron are of interest. Samples were collected at the start (before adding Fe solution) and at the end of the run. 10ml samples from all the 6 jars were collected separately in test tubes for analyzing through ICP-MS (Inductive Coupled Plasma Mass Spectrometry). 10ml Samples taken at the end of the run were filtered through a 0.45µm polyethersulphone syringe (PESS) filter to remove any solid particles which might affect the ICP-MS machine. The samples so collected were acidified with 100µl of 69% Nitric acid to inhibit the precipitation of the components.

- ***Sampling for Fe concentration*** – 5ml unfiltered samples were collected after addition of Fe to the jars to calculate the initial amount of Fe present in the solution during the run. The Fe concentration was measured using Merck Millipore 114761 Iron test which can measure a concentration of 0.0025 – 5mg/l of Fe. Figure 4 shows the samples prepared for the Fe test.

Figure 4 Prepared samples for Fe test from Jar Test

- ***Sampling for PSD (Particle Size Distribution)*** – Particle size distribution was carried out with the particle counter OLS-4031 from PAMAS. The samples were taken directly from the jar when the mixing was still performed to avoid any error in the PSD measurement due to sedimentation of particle without mixing. Each PSD measurement consists of 10 runs with 5ml per run, hence 50 ml pf unfiltered samples were taken at the end of 60 min from each jar. The particle counter measured the number particles of specific particle diameters. The lower analysis limit of the particle counter was 1µm and the higher limit is 100µm. The data was analyzed considering the average of the only the last 7 runs for each sample to avoid background contamination.

3.3.2 Multilayer Sand Filtration

- ***Sampling for ICP-MS (Inductive Coupled Plasma Mass Spectrometry)*** – the samples from the multilayer sand filtration were collected only for the analysis of Arsenic and Iron in the filter columns by ICP-MS. Samples were also collected at every 10 cm through the bed from the sampling points to see the working of the bed and the filter material. A total of 66 samples were collected from the 6 columns, 11 points per column (9 points through the bed, influent and effluent). For the speciation of As (III) and As(V), 10ml samples were taken with and without resin. The samples collected without resin will give the total arsenic in the water. These samples were filtered through a 0.45µm polyethersulphone syringe (PESS) filter to remove any solid particles which might affect the ICP-MS machine. For As (III) concentration - a strong basic anion exchange resin Amberlite® IRA-400 chloride form was used to adsorb As(V). When the sample is passed through the resin the As(V) gets adsorbed to the resin and the remaining arsenic concentration would give only unoxidized As (III) concentration. Figure 5 shows the passing of collected sample through the resin. The samples were then acidified by adding 100µl of 69% Nitric Acid. The so collected samples were then analyzed under ICP-MS.
- ***Samples for Fe concentration*** - 5ml unfiltered samples were taken from the sampling points to see the effect of removal at each layer. These unfiltered Fe samples were measured using Merck Millipore 114761 Iron test which can measure a concentration of 0.0025 – 5mg/l of Fe. The ICP-MS gave the Fe concentration of the filtered samples with 0.45µm filter.

Figure 5 Passing of samples from Multilayer sand filtration through resin for Arsenic Speciation

Chapter 4

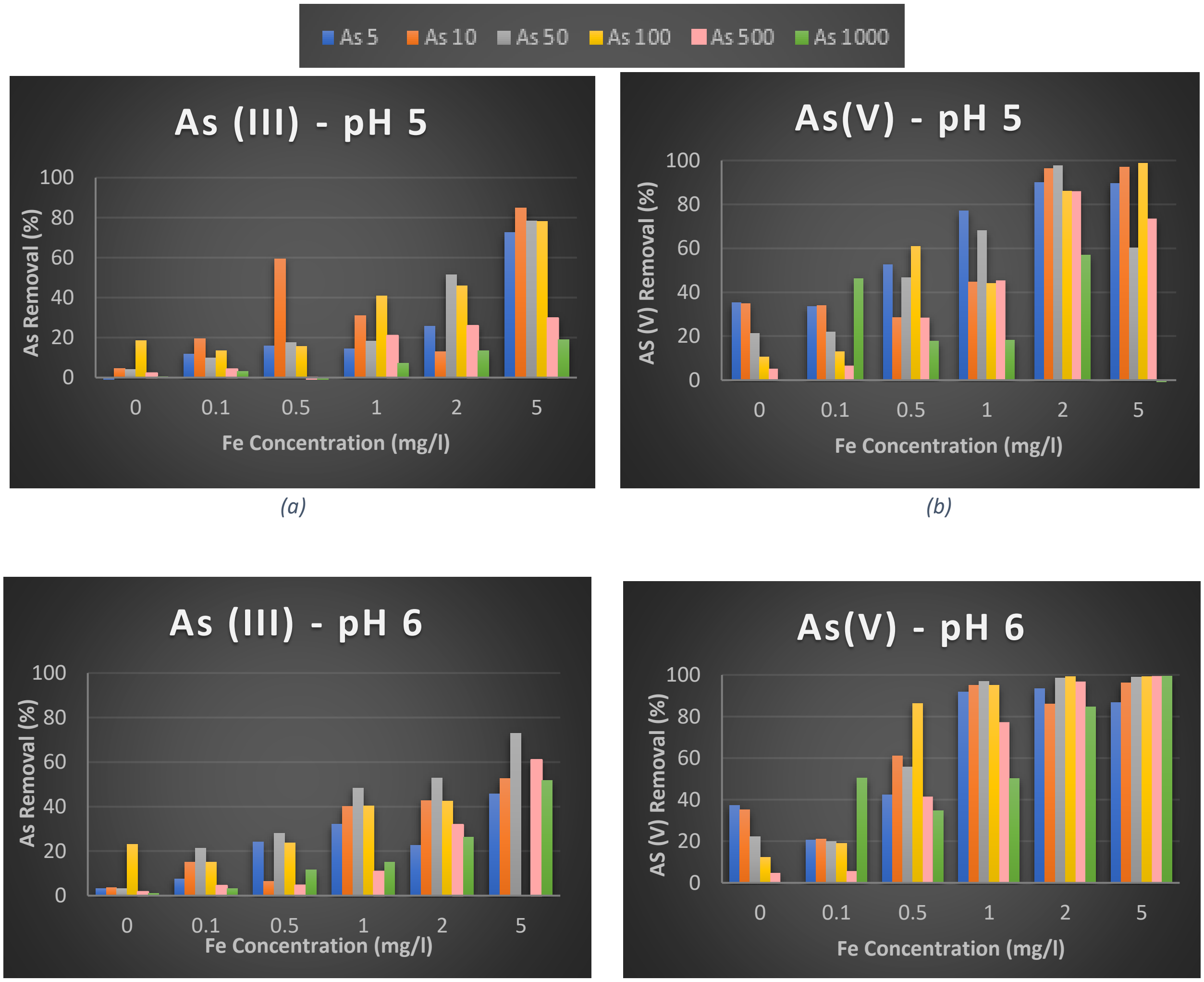
4 Results and Discussion

In this section all test results from the tests are presented separately for the Jar tests and Multilayer Sand Filtration.

4.1 Jar Test

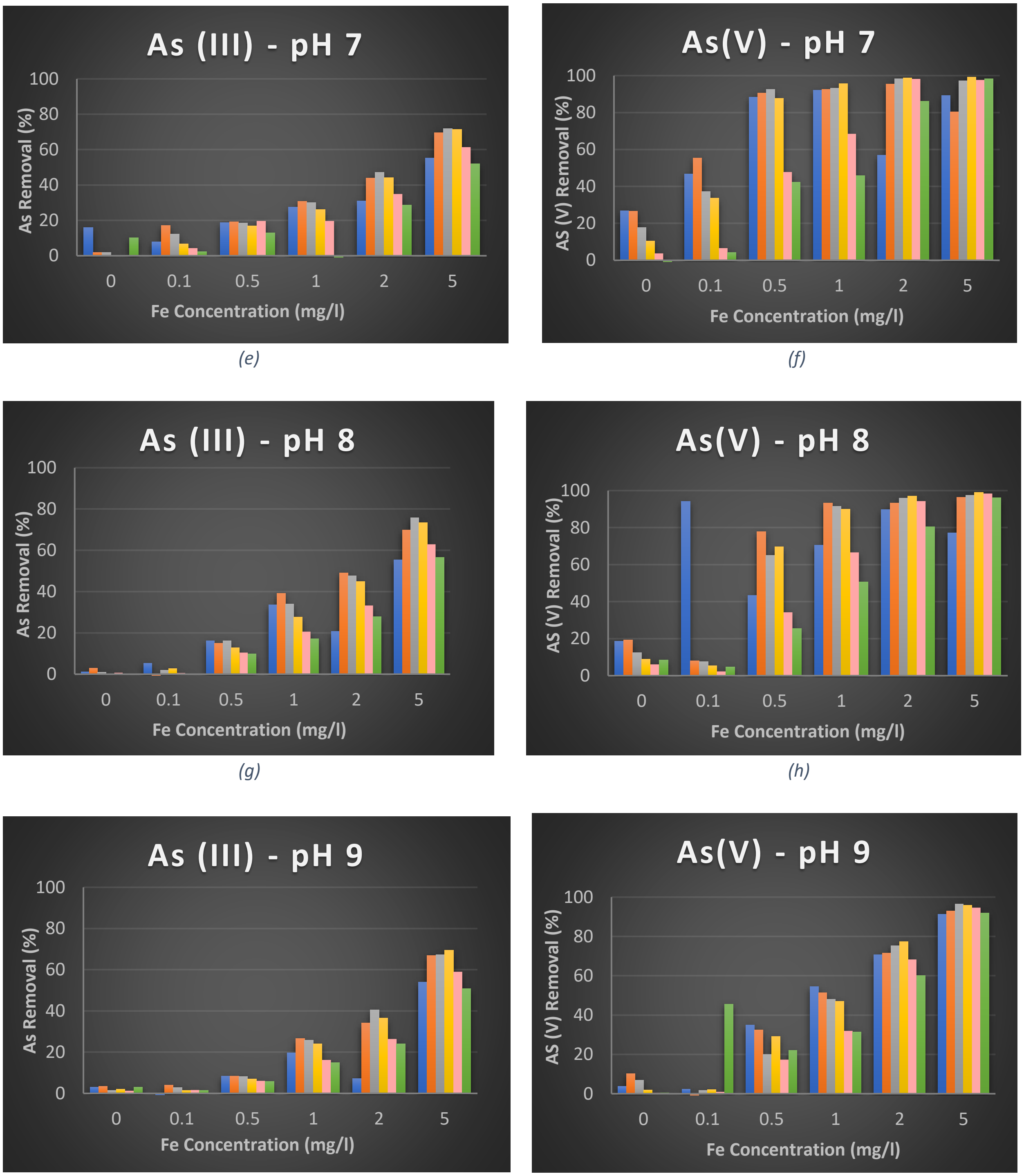
4.1.1 Effect of pH on As Removal

The removal rate of Arsenic was affected by pH. Figure 6 shows the percentage of As (III) and As (V) removal at different pH and varying Fe concentration. The removal of Arsenate was higher than that of Arsenite over the pH range of 5-9. 99% Arsenic (V) was completely removed at Fe dose of 5mg/l, hence the Fe dose with 10 and 20mg/l were not done.



(a) (b)

(c) (d)



(e) *(f)*

(g) *(h)*

(i) *(j)*

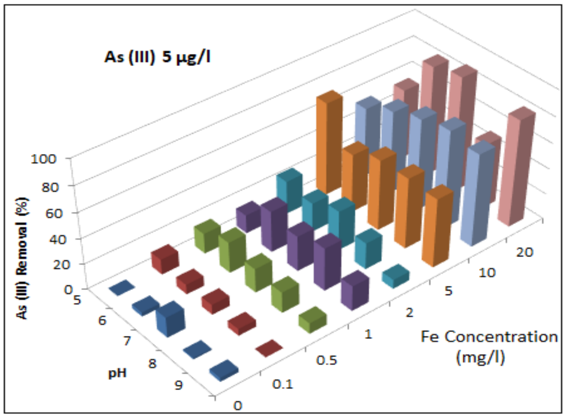
Figure 6 (a) to (j) Comparison of As (III) and As (V) removal to see the effect of pH at various Fe concentrations

When the pH was decreased from 9 to 6, As(V) removal increased by approximately 10% at all Fe concentrations. This might due to the presence of other anions such as silicates or phosphates as these decreases the adsorption of arsenate above a pH of 8.5 (Mohan & Pittman Jr, 2007). In contrast to As(V) removal, As (III) removal increased when pH was increased from 5 to 7 and maintained stable at pH 7-9.

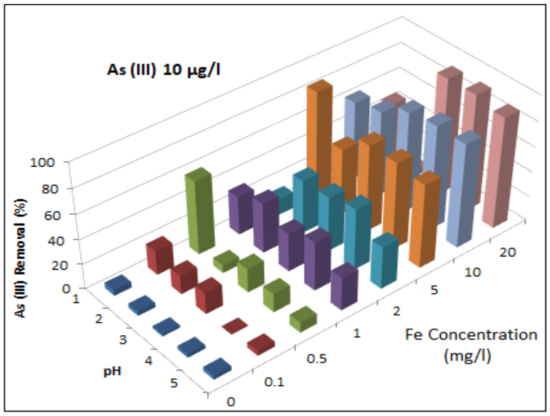
A maximum of As (III) removal was observed at pH 7. This result is consistent with the previous works. (Fernandez & Petrusevski; Hering et al., 1997; Meng et al., 2000)

4.1.2 Effect of Initial Arsenic Concentration and Oxidation state on As Removal

The percentage of removal of As (III) and As(V) are shown in the Figure 7 and Figure 8 respectively. As expected, the removal of As (V) is higher than that of As (III).



(a)



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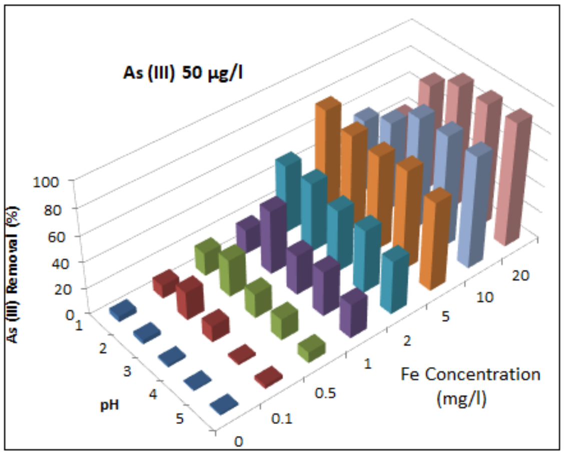
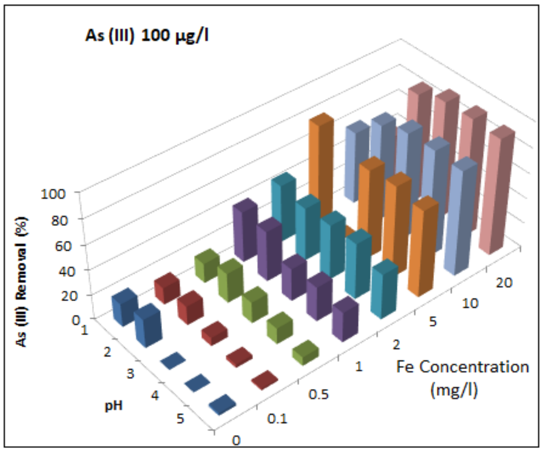
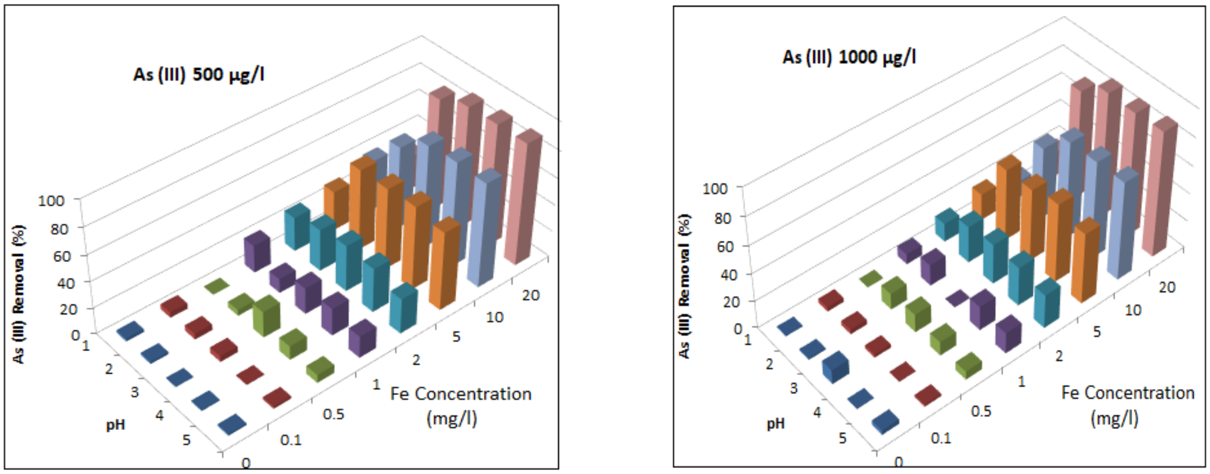


(c)



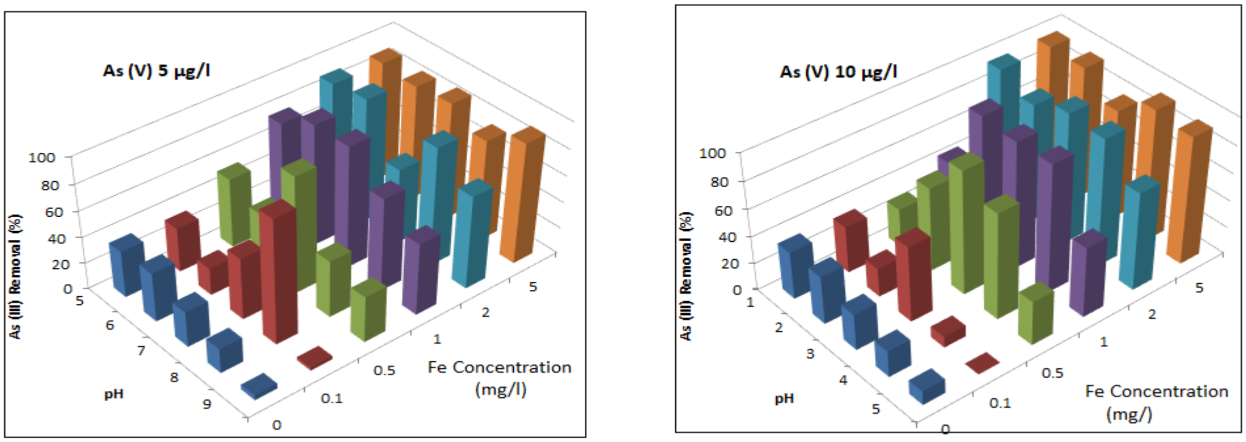
(d)



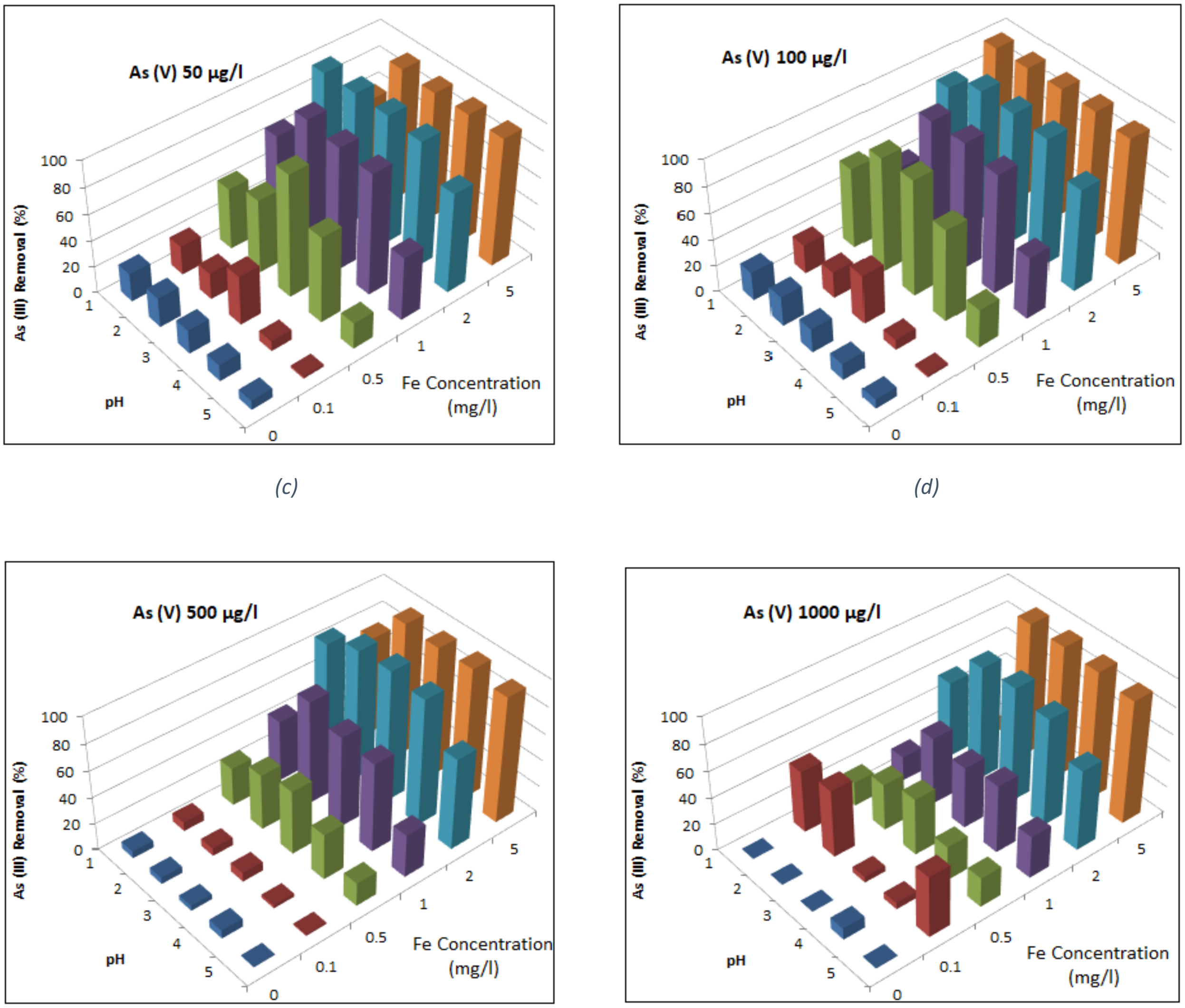
(e) *(f)*

Figure 7 (a) to (f) Effect of initial concentration and As (III) removal at various Fe concentrations

As (III) removal reduced with increasing As (III) concentration. The overall As (III) removal rates were about 70 - 80%. The presence of more than 50µg/l does not have any pronounced effect on the Arsenic removal at low Fe concentrations. This is in consistent with the study (Liu et al., 2009) "removal of As (III) by several types of Ferrated Salts in Aqueous form". This could be due to the less adsorption sites for As (III) as the Fe dosage was less. When initial As concentrations were higher, more iron is needed to decrease the dissolved Arsenic concentrations.



(a) *(b)*



(c) (d)

(e) (f)

Figure 8 (a) to (j) Effects of initial concentration and As (V) at various Fe concentrations

As (V) removal is usually faster than As (III) removal. It can be seen in Figure 8 As (V) is efficiently removed than As (III) during coagulation with Fe. 90 - 99% removal of As (V) are seen at Fe concentration of 5mg/l in contrast with As (III) where about 70% removal is observed. Thus, a dosage of 5mg/l is enough to remove As (V) above 50 µg/l. The removal efficiencies of As (V) during the coagulation are independent of the initial Arsenic concentration range examined. In all the experiments using Arsenic removal by coagulation shows higher removal rates for As (V) (Choi et al., 2010; Hering et al., 1997).

4.1.3 Effective Iron

The initial concentration of iron present in the solution was measured by the Fe Test and the final concentration was measured using ICP-MS. The amount of iron removed in percentage after filtration through 0.45 µm filter is shown in Appendix 6.1.2.1 and 6.1.2.2 for As (III) and As (V) respectively. The effective Fe or the actual amount of Fe in mg/l that flocculated to greater than 0.45 µm is calculated. Table 6 and Table 7 shows the Effective Iron for As (III) and As (V) respectively.

Table 6 Effective Fe or Fe > 0.45 µm for As (III)

Effective Fe (Fe > 0.45µm) for As (III) (mg/l)						
Fe (mg/l)	**As Start (µg/l)**	**pH**				
		5	**6**	**7**	**8**	**9**
0.1	5	0.00	0.01	0.05	0.05	0.07
	10	0.00	0.01	0.05	0.06	0.05
	50	0.00	0.05	0.06	0.06	0.06
	100	0.00	0.03	0.06	0.04	0.06
	500	0.00	0.01	0.07	0.06	0.03
	1000	0.00	0.02	0.07	0.07	0.06
0.5	5	0.06	0.36	0.48	0.48	0.45
	10	0.24	0.09	0.48	0.48	0.45
	50	0.05	0.34	0.48	0.47	0.44
	100	0.08	0.26	0.48	0.47	0.45
	500	0.05	0.07	0.48	0.47	0.46
	1000	0.07	0.34	0.48	0.48	0.46
1	5	0.1	0.8	1.0	1.0	1.0
	10	0.1	0.1	1.0	1.0	1.0
	50	0.1	0.9	1.0	0.9	1.0
	100	0.3	0.9	1.0	0.9	1.0

	500	0.5	0.2	1.0	0.9	0.9
	1000	0.3	0.4	1.0	1.0	0.9
2	5	1.3	1.9	2.0	2.0	2.0
	10	1.5	1.8	2.0	2.0	2.0
	50	1.6	1.8	2.0	2.0	2.0
	100	1.4	1.6	2.0	2.0	2.0
	500	1.8	1.5	2.0	2.0	2.0
	1000	0.9	1.7	2.0	1.9	1.9
5	5	3.6	5.0	5.0	4.9	5.0
	10	2.6	4.7	5.0	4.9	5.0
	50	2.7	4.9	5.0	5.0	5.0
	100	3.4	4.6	5.0	5.0	5.0
	500	2.2	4.5	5.0	5.0	5.0
	1000	1.9	4.6	5.0	5.0	4.9
10	5	1.5	7.3	10.0	10.0	10.0
	10	2.1	6.1	10.0	10.0	10.0
	50	2.1	5.4	10.0	10.0	10.0
	100	3.4	8.7	10.0	10.0	10.0
	500	3.5	6.1	10.0	10.0	10.0
	1000	1.2	5.5	10.0	10.0	10.0
20	5	6.2	17.2	20.0	20.0	20.0

	10	4.5	12.6	20.0	20.0	20.0
	50	5.7	12.6	20.0	20.0	20.0
	100	4.4	16.2	20.0	20.0	20.0
	500	4.2	15.3	20.0	20.0	20.0
	1000	2.0	13.2	20.0	20.0	20.0

Table 7 Effective Fe or Fe > 0.45 µm for As (V)

Effective Fe (Fe > 0.45µm) for As (V) (mg/l)

Fe (mg/l)	As Start (µg/l)	pH				
		5	**6**	**7**	**8**	**9**
0.1	5	0.00	0.01	0.07	0.10	-0.01
	10	0.00	0.01	0.06	-0.01	-0.01
	50	0.01	0.01	0.07	-0.03	-0.01
	100	0.01	0.03	0.07	-0.01	-0.01
	500	0.01	0.02	0.06	-0.01	-0.03
	1000	0.00	0.03	0.07	-0.01	-0.01
0.5	5	23	32	96	95	95
	10	0.12	0.16	0.48	0.47	0.48
	50	-0.02	0.23	0.48	0.47	0.47
	100	0.10	0.18	0.49	0.48	0.48
	500	0.22	0.36	0.48	0.48	0.48

	1000	0.26	0.35	0.45	0.44	0.47
1	5	0.39	0.95	0.99	0.99	0.99
	10	0.22	0.94	0.99	0.99	0.99
	50	0.37	0.90	0.98	0.99	0.98
	100	0.28	0.86	0.99	0.99	0.98
	500	0.52	0.90	0.97	0.97	0.97
	1000	0.33	0.88	0.93	0.95	0.98
2	5	1.22	1.79	1.99	1.99	1.99
	10	1.50	1.66	1.99	1.99	1.99
	50	1.56	1.41	1.99	1.99	1.99
	100	0.91	1.68	1.99	1.99	1.99
	500	1.53	1.41	1.99	1.99	1.98
	1000	1.35	1.61	1.97	1.97	1.97
5	5	1.53	4.50	4.99	4.99	4.99
	10	2.58	3.79	4.99	4.99	4.99
	50	0.55	4.44	4.98	4.99	4.99
	100	3.09	4.28	4.99	4.99	4.99
	500	2.00	3.55	4.98	4.99	4.99
	1000	2.08	4.61	4.98	4.98	4.99

From the Table 6 and Table 7 it can be observed that at higher Fe dose and at pH 5 and 6, the removal of Iron is reduced, although the actual amount of Fe greater than 0.45µm is quite stable for both As (III) and As (V). At pH 7-9 the removal of iron was about 95 – 99% indicating complete conversion of

Ferrous ions to Ferric. The process of floc formation is dependent on pH. At pH 7- 9 the floc is stable irrelevant of the Fe concentration. It agrees with the process of iron removal by oxidation-floc formation is dominant at pH 8.5 as explained by Sharma, S.K. in his thesis. (Sharma, 2001). This can also be seen clearly in the particle size distribution which is shown in the next section.

4.1.4 Particle Size Distribution

The complete data of the particle size analysis is not shown here. The results for the standard conditions only (pH 7, 1 mg/l Fe and 100µg/l As) are shown here by varying one of the parameters. Although the range of particle size analysis was 1 - 100µm, the results shown here are until the size of 20µm as the number of particles bigger than 20µm were negligible.

4.1.4.1 Effect of pH

Figure 9 and Figure 10 shows the effect of pH on the flocculation of iron for As (III) and As(V) respectively. This proves that there is poor flocculation at pH 5 and slight improvement at pH 6. Similar results were observed with other Arsenic concentrations and Fe concentrations (data not shown here). At pH 7- 9 all the Fe was flocculated, which agrees that the floc formation is dominant at higher pH.

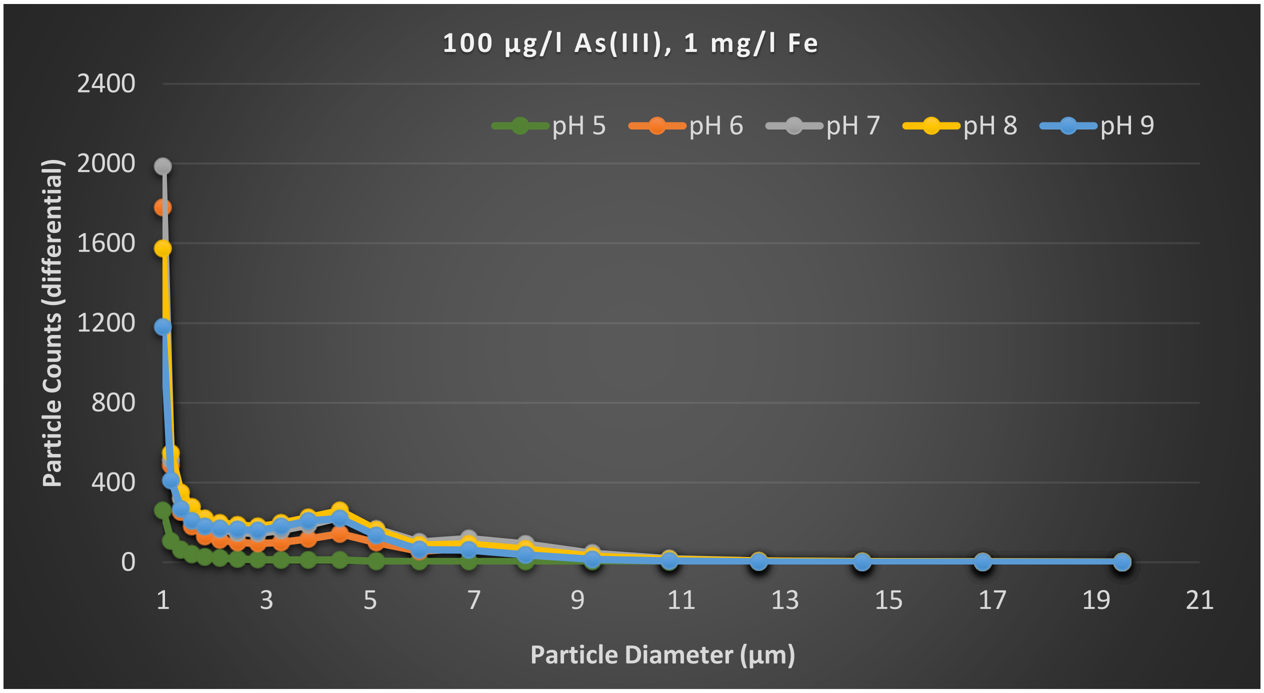


Figure 9 Particle size distribution to see the effect of pH on flocculation for 100 µg/l As (III) and 1 mg/l Fe

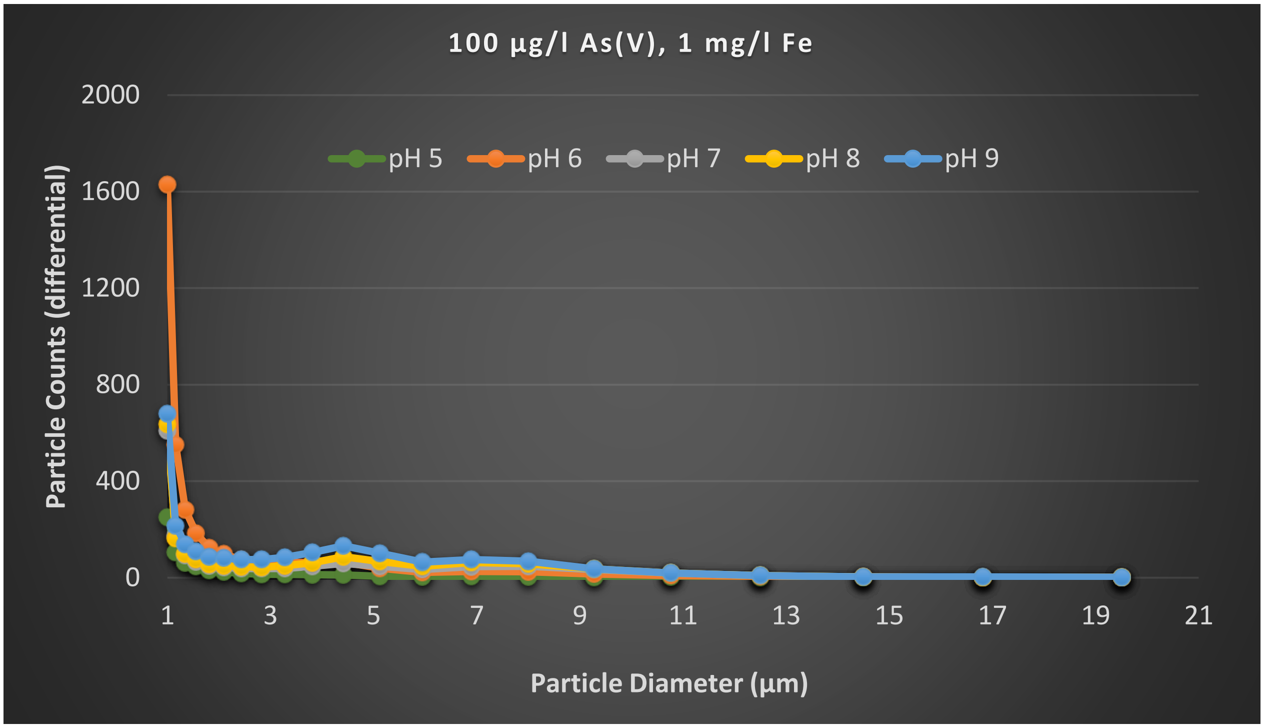


Figure 10 Particle size distribution to see the effect of pH on flocculation for 100 µg/l As (V) and 1 mg/l Fe

4.1.4.2 Effect of As Concentration

The effect of arsenic concentration on Fe flocculation was not clear enough. Slight differences were seen in few cases, but the trend was not followed, hence the data is not shown here. This could be due the experimental errors of residual iron in the Jar or issues with the pH measuring. These differences could also be due to the competition within the Arsenic itself rather than with the Fe flocculation.

4.1.4.3 Effect of Fe Dosage

Figure 11 and Figure 12 shows the effect of Fe dosage on flocculation for As (III) and As(V) respectively. The graphs show clear increase in the particle counts with the increase in the Fe concentrations. The number of particles at a diameter increases with the same factor as the Fe concentration. For example, the counts at 1µm with 20mg/l Fe is double the counts with 10mg/l Fe. Also, there seems to be an increase in the peak at around 4 µm which is observed at all the Fe concentrations. The same trend is seen in other concentrations of Arsenic (data not shown here).

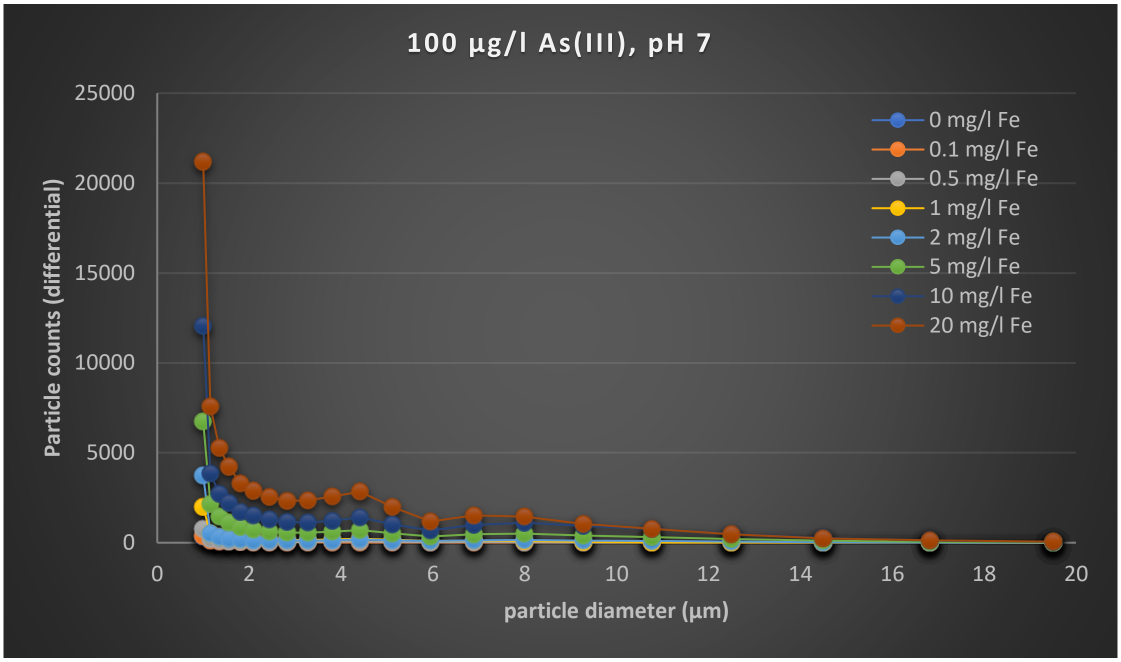


Figure 11 Particle size distribution to see the effect of Fe dosage on flocculation for 100 µg/l As (III) and pH 7

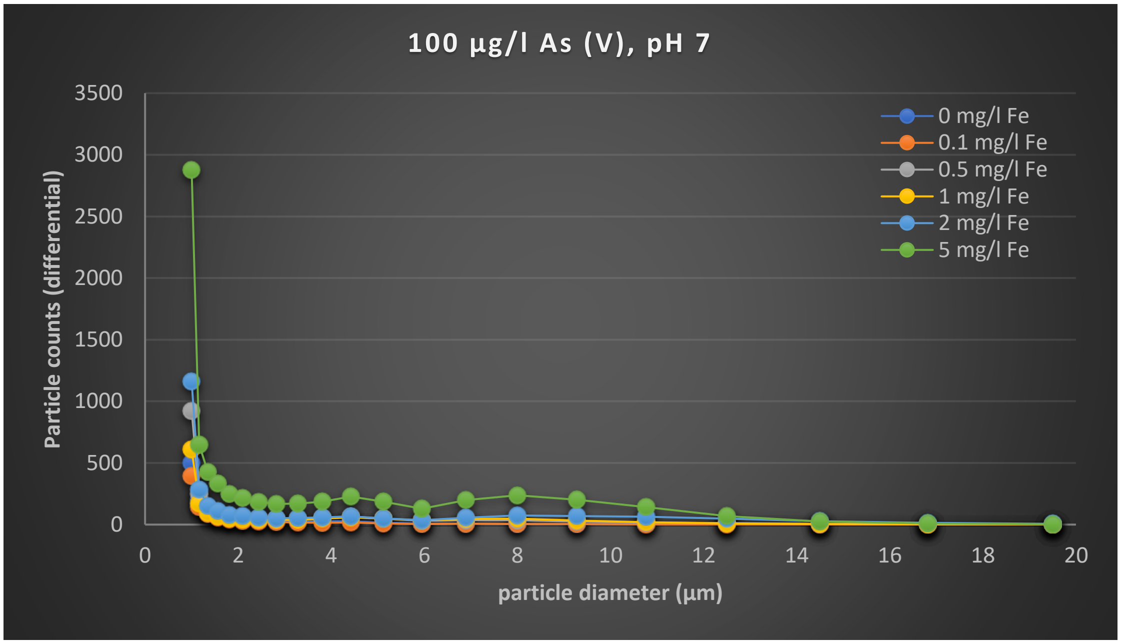


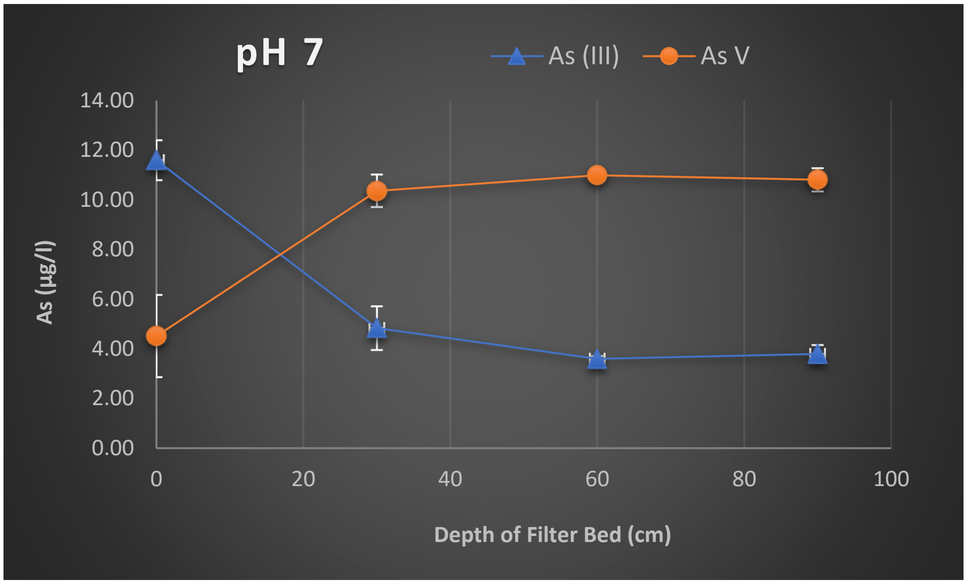
Figure 12 Particle size distribution to see the effect of Fe dosage on flocculation for 100 µg/l As (V) and pH 7

4.2 Multilayer Sand Filtration

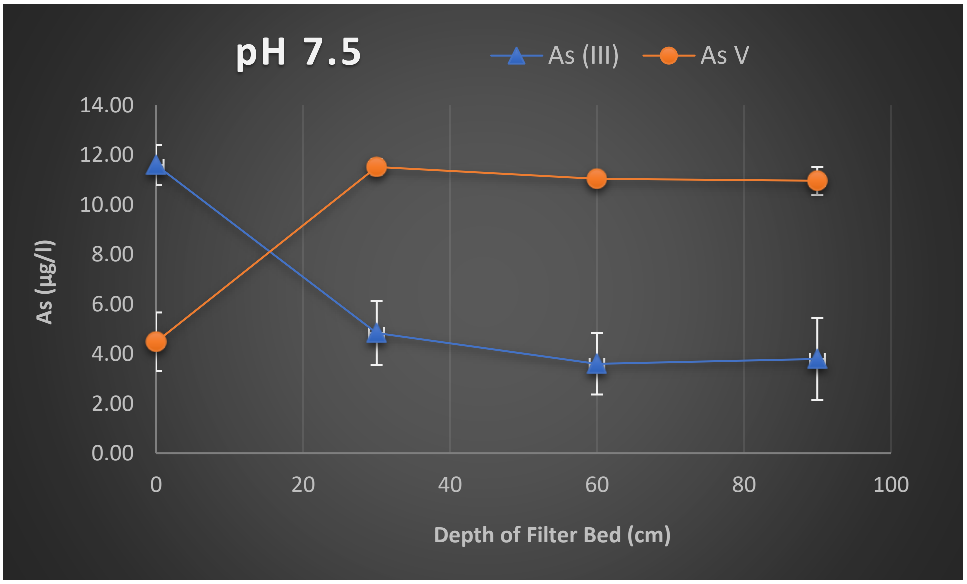
The removal of Arsenic in the multilayer sand filtration columns were observed over a period. Arsenic removal before and after the addition of iron were also compared. The averaged value for the 2 columns with same pH were taken for the analysis of the data.

4.2.1 Biological Oxidation

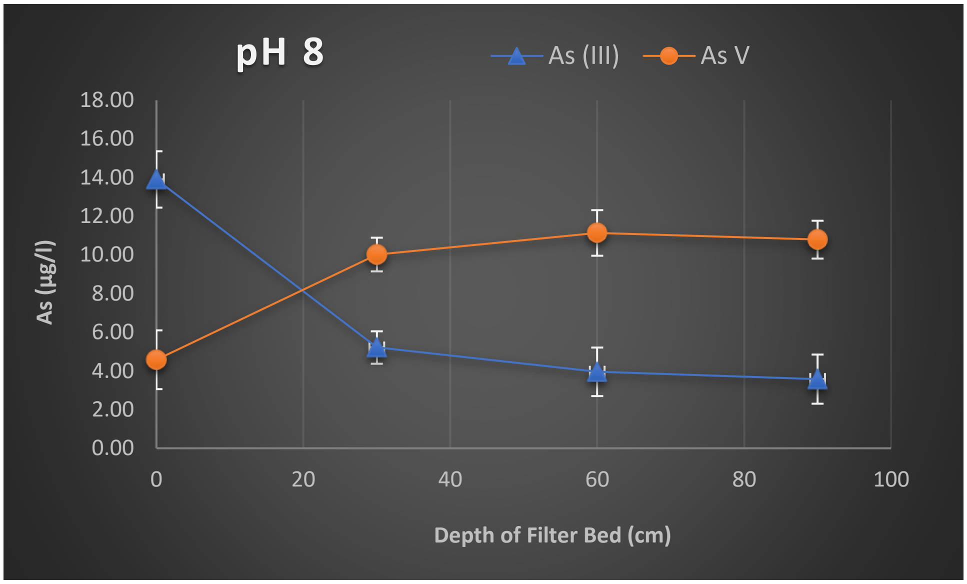
Arsenite oxidation was observed in the columns before the addition of Iron. The arsenic oxidation in the water dosed through the column for all different pH taken are shown in the Figure 13. As (III) was oxidized into the equivalent concentration of As (V) in the filter bed. These results indicate the presence of Arsenic Oxidizing Bacteria (AOB) that has been developed in the filter bed. Arsenic oxidation is less at pH 8 than that of pH 7, this contradicts the study done by (Ike et al., 2008) that oxidation by AOB is similar at pH 7 and pH 8.



(a)



(b)



(c)

Figure 13 (a) (b) and (c) - Oxidation of Arsenite to Arsenate before the addition of iron at pH 7, pH 7.5 and pH 8

The oxidation efficiency for the columns is shown in Table 8. The oxidation efficiency was calculated according to the equation (Crognale et al., 2019):-

$$Oxidation\ Efficiency\ (\%) = \left(1 - \frac{As(III)}{As\ total}\right) * 100$$

Table 8 Oxidation Efficiency by Arsenic Oxidizing Bacteria at pH 7, pH 7.5 and pH 8

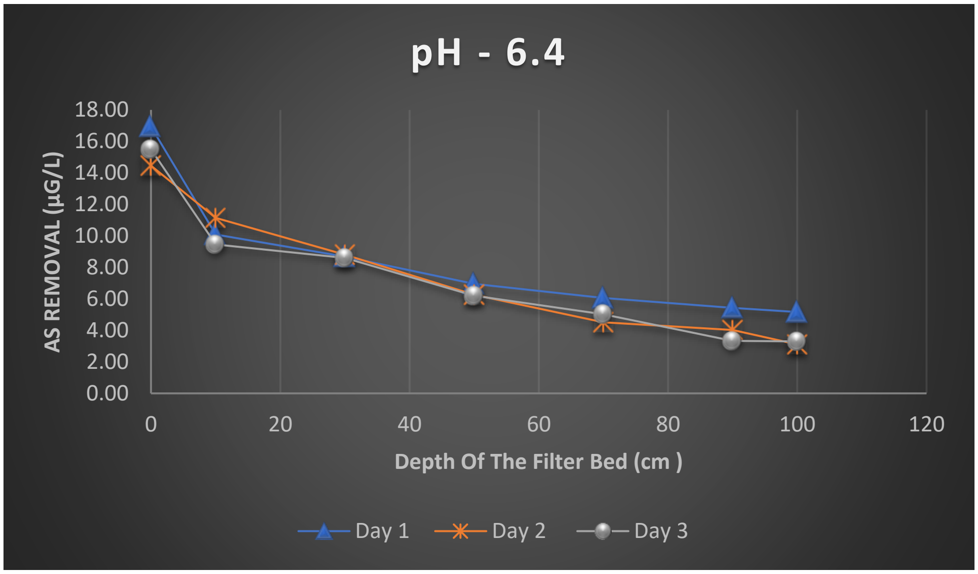
Depth of the Bed (cm)	pH 7	pH 7.5	pH 8
Influent - 0	24.38	24.38	24.77
Anthracite - 30	68.20	68.85	55.24
Sand – 60	75.34	73.63	64.10
Garnet - 90	74.00	75.40	65.35

Microbial oxidation efficiency in a bio sand filters is typically in the range of 60-80% (Crognale et al., 2019; Smith et al., 2017). The oxidation efficiency is highest in the Sand and Garnet layer as compared to Anthracite layer. Possible reason for this could be due to large colonization surface area for acclimation of biofilm on Sand and Garnet (Crognale et al., 2019). As (III) is oxidized by the process of detoxification mechanism in the presence of heterotrophic oxidizing bacteria using Oxygen as electron acceptor (Crognale et al., 2019; Ike et al., 2008; Nguyen et al., 2017). The stochiometric equation with oxygen is given by the equation (Ike et al., 2008) :-

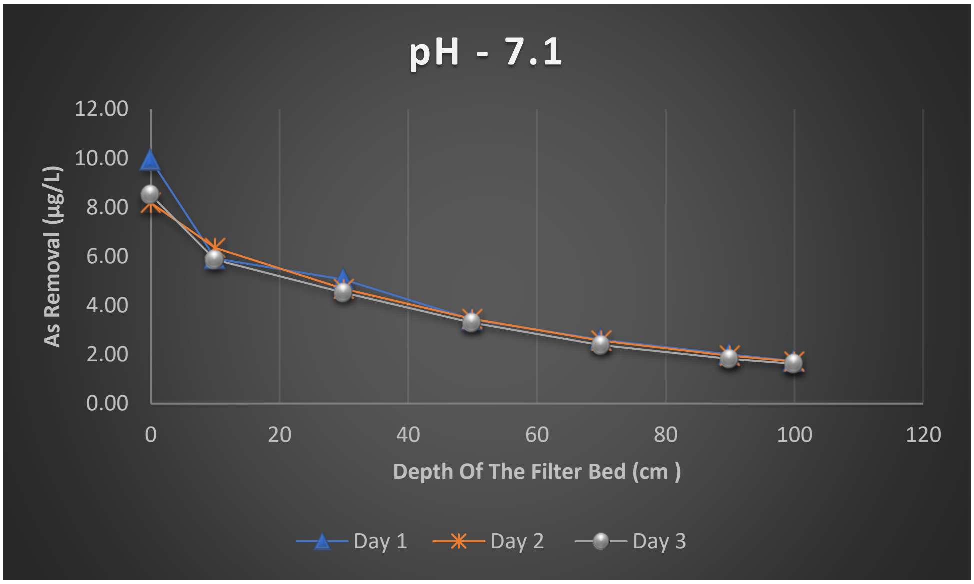
$$H_2AsO_3^- + O_2 + H^+ \rightarrow HAsO_4^{2-} + H_2O$$

4.2.2 Arsenic Removal

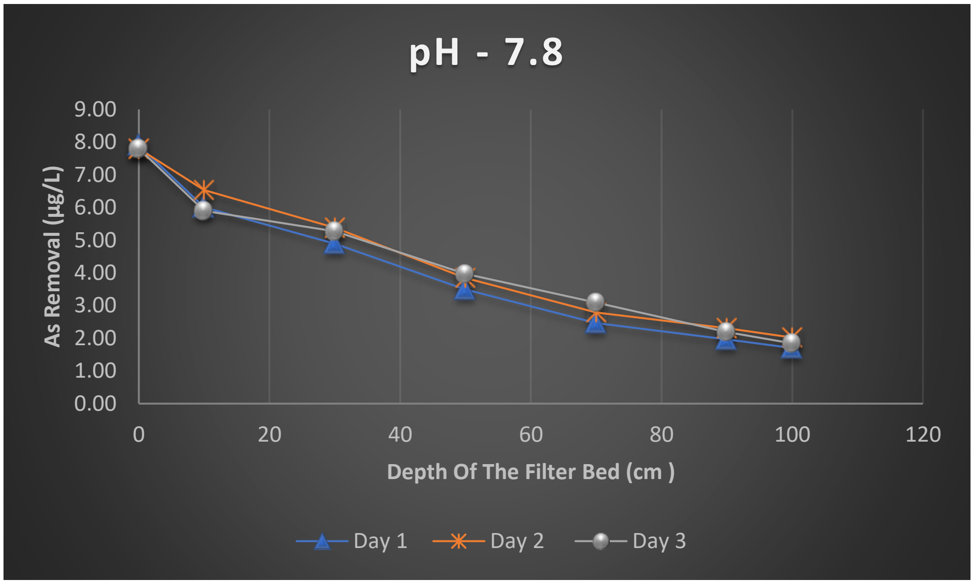
The removal percentage in each column for Arsenic and Iron are shown in Table 9. After dosing iron solution, sampling was done once a week for two weeks. Due to the increase in the filter bed resistance which led to the overflowing of the columns, they were backwashed after every 3 days. Due to the increase of pore clogging the resistance in the filtration bed increases. Also, by virtual inspection a layer (about 2-3cm) of iron was deposited on the top layer of the bed. In the third week after the dosing of Iron sampling was done for three consecutive days after backwash to see the effect of pore clogging in the three different layers. Due to the addition of Iron to the columns the pH in the columns dropped down. Hence, pH was then adjusted in the acid solutions dosed (for Column 3 and 4 - 400ml 96% Sulphuric Acid in 10l, for Column 5 and 6 - 100ml 96% Sulphuric Acid in 10l). The new pH in the columns were then 6.4, 7.1, 8. Total Arsenic removal for all the three consecutive days though the bed is shown the Figure 14



(a)



(b)



(c)

Figure 14 (a) (b) and (c) - Removal of Arsenic in the columns on Day 1, 2 and 3 after backwash at various depths in the filter bed

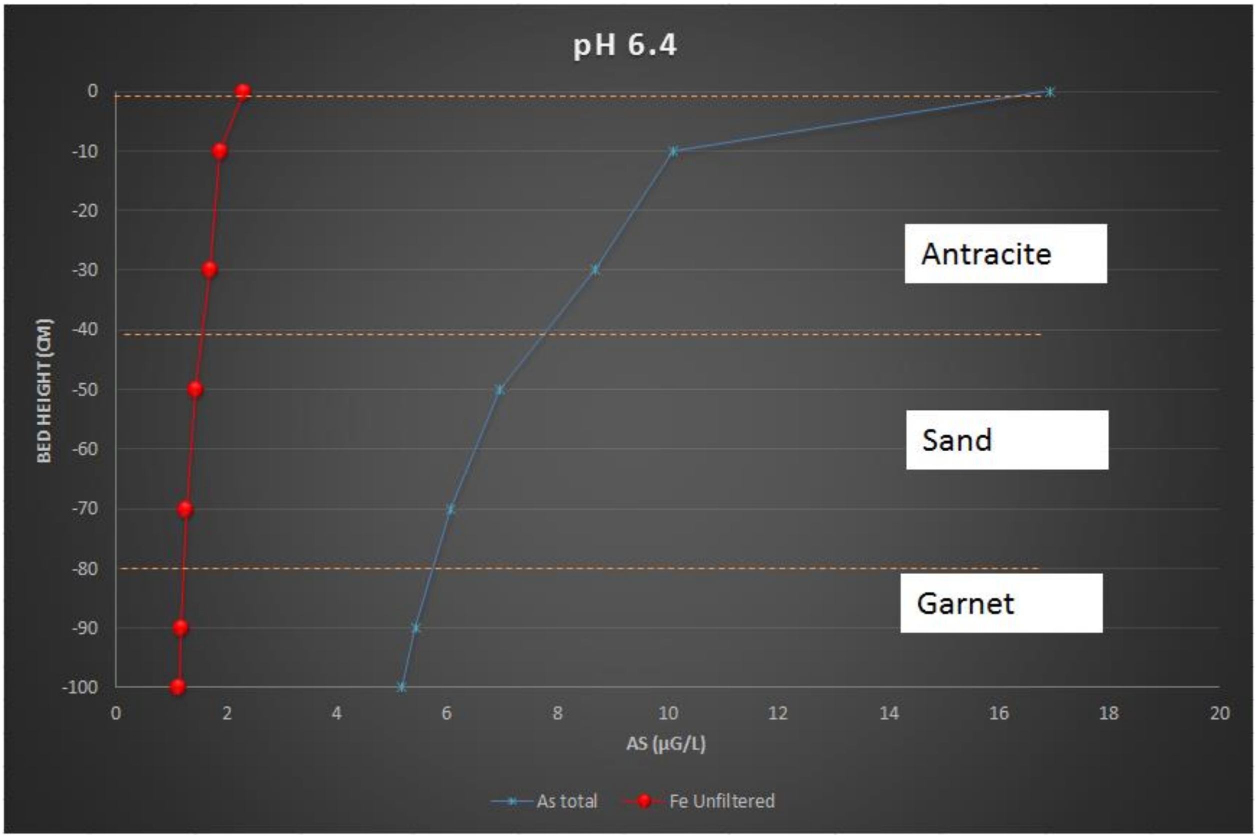
The removal for all the three days is quite stable though out the bed. The highest removal is seen in the top layer of Anthracite. The complete removal data on the arsenic removal for the three days is shown in the Appendix in Figure A. 1, Figure A. 2 and Figure A. 3 for day 1, 2 and 3 respectively.

Table 9 Arsenic and Iron removal percentages in each column on day 1,2 and 3 at pH 7.8, 7 and 6.4

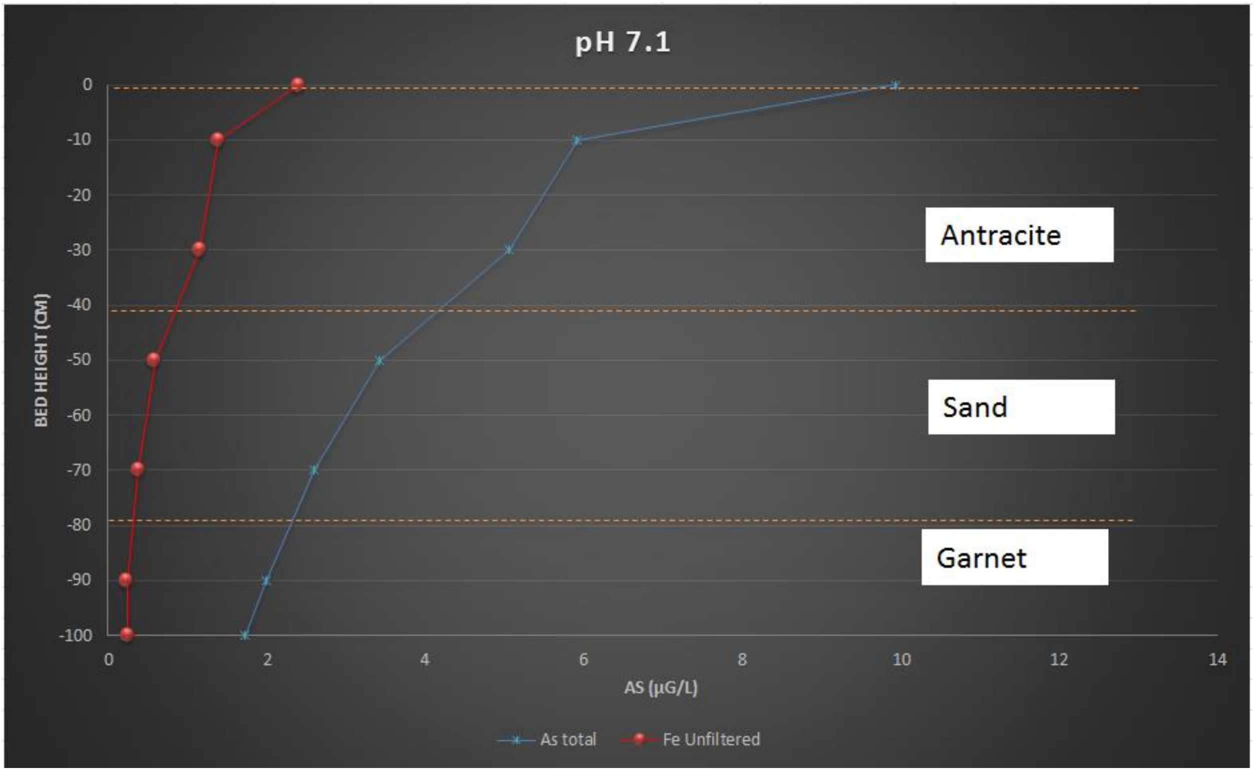
Removal Percentage (%)							
pH	**Column**	**Day 1**		**Day 2**		**Day 3**	
		As	**Fe**	**As**	**Fe**	**As**	**Fe**
7.8	1	79.38	95.85	72.53	92.95	75.22	69.90
	2	77.56	96.1	75.40	94.76	77.59	78.10
7.1	3	83.29	88.16	80.53	91.67	84.68	91.43
	4	81.83	91.95	77.52	94.37	76.72	93.55
6.4	5	66.78	5.29	81.68	49.51	76.54	66.35
	6	73.32	87.5	73.66	90.46	80.88	74.60

4.2.3 Removal Efficiency at different layers

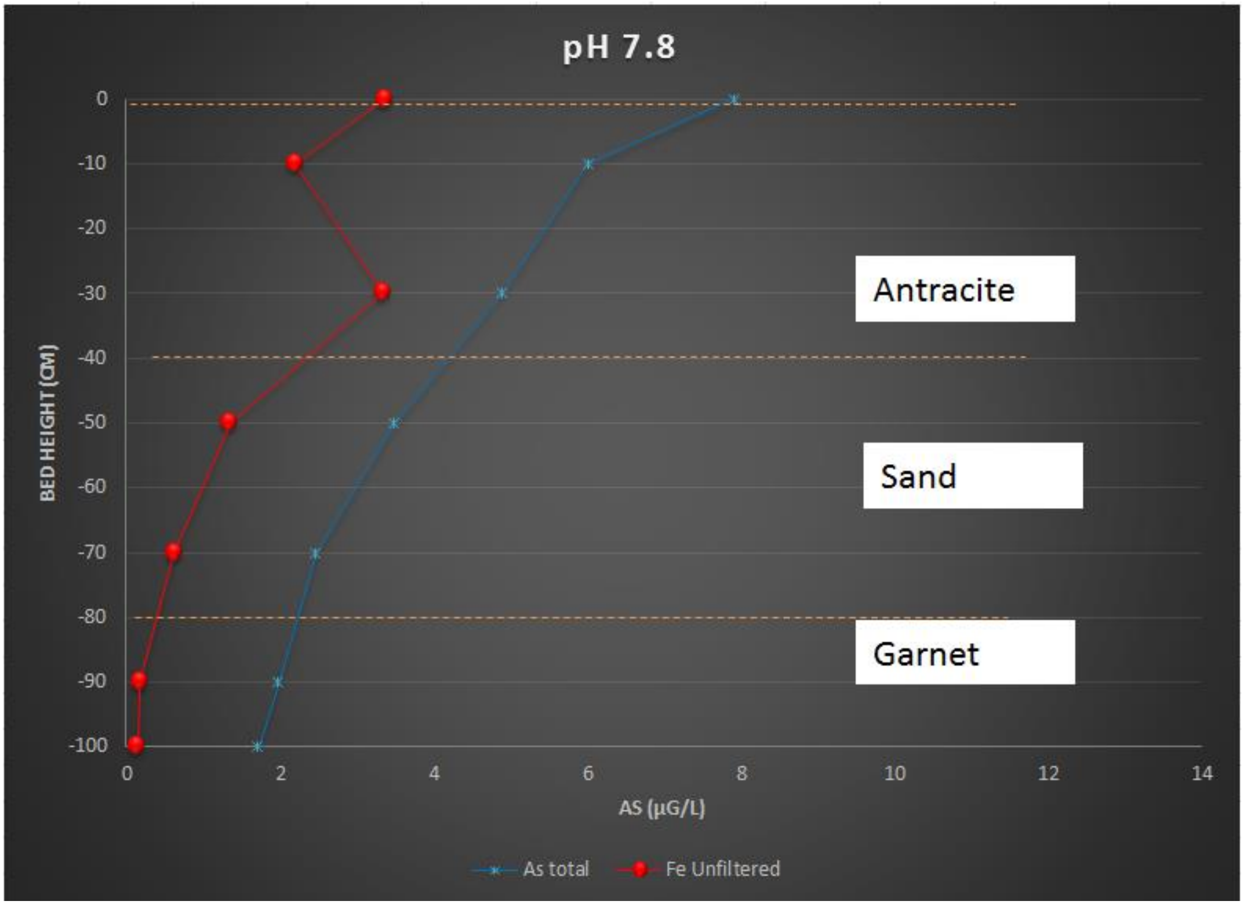
The removal of total Arsenic with respect to iron for day 1 in the layer bed are shown in Figure 15. Day 2 and day 3 are shown in Appendix Figure A. 4 and Figure A. 5 respectively. From the figures it is observed that Arsenic removal was high at Anthracite layer as compared to the Sand and Garnet. This is due to oxidation of iron in the anthracite layer and forming adsorption sites for arsenic. Some difference in the Iron removal are seen in Anthracite layer (Figure 15 (c)), this might be due to fact that the water has not flowed through the bed rather it was attached to the wall of the column. This variation is seen at day 2 and day 3 as well, but this does not affect the removal of Arsenic overall.


pH 6.4
BED HEIGHT (CM)
0
-10
-20
-30
-40
-50
-60
-70
-80
-90
-100
0
2
4
6
8
10
12
14
16
18
20
AS (µG/L)
Antracite
Sand
Garnet
As total
Fe Unfiltered

(a)


pH 7.1
BED HEIGHT (CM)
0
-10
-20
-30
-40
-50
-60
-70
-80
-90
-100
0
2
4
6
8
10
12
14
AS (µG/L)
Antracite
Sand
Garnet
As total
Fe Unfiltered

(b)



(c)

Figure 15 (a), (b) and (c) Arsenic removal in the bed with respect to Iron at Day 1 after backwash for three pH

Chapter 5

5 Conclusion and Recommendations

5.1 Conclusions

Based on the presented research the following main conclusions can be drawn: -

5.1.1 Jar Test

- pH has a positive effect on the arsenic removal. As (V) removal is decreased with the decrease in pH whereas As (III) removal is increased with increase in pH.
- The initial concentration of Arsenic (V) does not have any effect on removal. For As (III), higher the initial concentration more adsorption sites are needed thus a higher amount of Fe is needed.
- As (V) is 90-99% removed with about 5mg/l Fe whereas As (III) needs higher doses of Fe.
- Although the Fe removal was low at pH Fe that flocculated to greater than 0.45µm was quite stable at all pH. This has been observed with particle counter as well at pH 5 the flocculation was poor with slight improvement with the increase in pH.
- Effect of arsenic concentration on floc formation was not clear enough as there was no trend that followed. The slight difference was seen due to experimental error of residual iron in the Jar. There could also be competition within the Arsenic itself.
- Effect of Fe on flocculation showed a clear increase in the particle counts with the increase in Fe. The number of particles at a diameter increased in the same factor as the Fe concentration.

5.1.2 Multilayer Sand Filtration

- The results observed before dosing of iron in the filter show that Arsenic Oxidizing Bacteria could have been the reason for oxidation of Arsenite to Arsenate, although the presence was not tested.
- High oxidation efficiency was seen at Sand and Garnet layer due to the large surface area.
- The oxidation was less at pH 8 than of pH 7 which contradicts the results of previous study.
- The oxidation efficiency was higher in sand and garnet layer than that of anthracite layer due to the large surface area of sand and garnet.
- After the dosage of iron, the resistance in the filter bed increased and needed backwash after every three days.
- Although the resistance was increased, the total removal observed after backwash for three consecutive days was quite stable.
- The highest removal of arsenic was seen in Anthracite layer although the oxidation of As (III) to As (V) was more dominant in Sand and Garnet layer. This is due to the high adsorption of Iron in the Anthracite layer.

5.2 Recommendations

The results obtained are quite satisfactory as Fe (II) can remove arsenic to a large extent. Further the research could be improved by

5.2.1 Jar tests

- By performing duplicates or triplicates for these tests.
- The presence of other compounds such as sulphates, silicates, phosphates could be seen.
- Samples could be taken at different times to see the dissolving rates of the As (III) and As (V).
- The Fe dosage below 10mg/l could be used to see the effect in a more pronounced way. As most of the Fe in the groundwater present in Bangladesh is around 5mg/l.

5.2.2 Multilayer sand filtration

- Microbial analysis could be done to see the biological activity in the filtration process.
- The filter could be tested for higher concentrations of Arsenic and varied concentration of iron as the concentration used here is less compared.
- The influence of other parameters such as silica and phosphate on the removal of iron and arsenic could be determined.
- The process conditions could be changed to see the removal in the filter.

5.3 Acknowledgements

I would like to thank Dr. Ir. Doris van Halem and Dr. David Ridder for their continuous inputs and guidance throughout the work. I would also like to thank my co master student Mr. Mrinal Roy for his help during the sampling in the laboratory. I would also like to thank lab technicians Mr. Mohammed Jafar and Mr. Armand Middeldorp who were supporting me during the research in the lab.

Chapter 6

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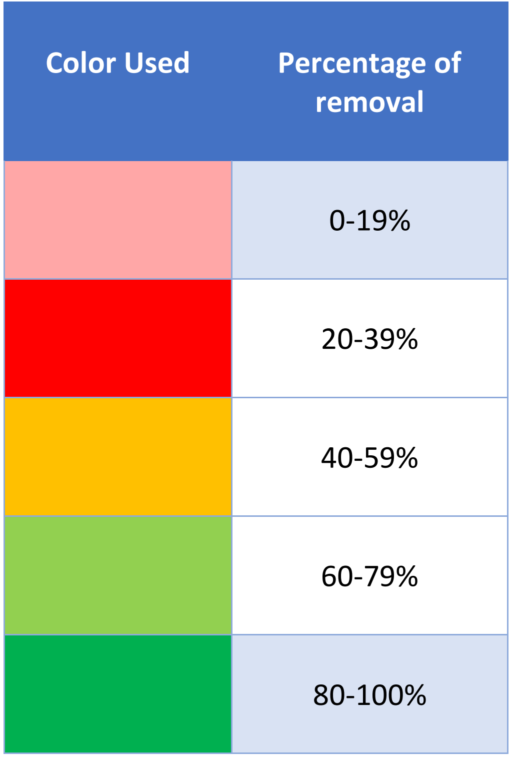
Appendix

6.1 Jar Test

6.1.1 Arsenic Removal

For the easy understanding of the data obtained color table has been used. The color code used for analyzing the data is shown in Table A. 1.

Table A. 1 Color table to explain the removal percentage



Color Used	Percentage of removal
🩷	0-19%
🟥	20-39%
🟨	40-59%
🟩	60-79%
🟩	80-100%

6.1.1.1 Arsenite

Table A. 2 Removal percentage of As (III) with various Fe concentrations at pH 5-9

Arsenic (III) removal (%)							
Fe (mg/l)	**As Start (µg/l)**	**pH**					**Removal needed for < 10 µg/l (%)**
		5	**6**	**7**	**8**	**9**	
0	5	-3	3	16	1	3	0
	10	5	4	2	3	3	0
	50	4	3	2	1	1	80
	100	19	23	0	0	2	90
	500	2	2	0	1	1	98
	1000	1	1	10	0	3	99
0.1	5	12	8	8	5	-2	0
	10	20	15	17	-9	4	0
	50	10	21	12	2	3	80
	100	14	15	7	3	1	90
	500	4	4	4	1	2	98
	1000	3	3	2	0	1	99
0.5	5	16	24	19	16	8	0
	10	59	6	19	15	8	0
	50	17	28	18	16	8	80
	100	16	24	17	13	7	90

	500	-2	5	20	10	6	98
	1000	-2	12	13	10	6	99
1	5	14	32	28	34	20	0
	10	31	40	31	39	27	0
	50	18	48	30	34	26	80
	100	41	40	26	28	24	90
	500	21	11	20	21	16	98
	1000	7	15	-3	17	15	99
2	5	26	23	31	21	7	0
	10	13	43	44	49	34	0
	50	52	53	47	48	41	80
	100	46	43	44	45	37	90
	500	26	32	35	33	26	98
	1000	13	26	29	28	24	99
5	5	73	46	55	55	54	0
	10	85	53	70	70	67	0
	50	78	73	72	76	67	80
	100	78		71	73	70	90
	500	30	61	61	63	59	98
	1000	19	52	52	57	51	99

10	5	54	64	72	77	72	0
	10	63	68	80	84	83	0
	50	55	67	85	86	85	80
	100	58	77	85	85	84	90
	500	38	63	78	80	79	98
	1000	12	52	70	71	71	99
20	5	55	85	90	51	83	0
	10	47	5	93	93	89	0
	50	43	81	93	94	94	80
	100	39	87	94	95	93	90
	500	26	84	91	92	92	98
	1000	19	78	90	89	90	99

6.1.1.2 Arsenate

Table A. 3 Removal percentage of As (V) with various Fe concentrations at pH 5-9

Arsenic (V) removal (%)							
Fe (mg/l)	**As Start (µg/l)**	**pH**					**Removal needed for < 10 µg/l (%)**
		5	**6**	**7**	**8**	**9**	
0	5	35	37	27	19	4	0
	10	35	35	27	19	10	0
	50	21	22	18	12	7	80
	100	11	12	10	9	2	90
	500	5	5	4	6	0	98
	1000	0	0	-2	9	0	99
0.1	5	34	21	47	94	2	0
	10	34	21	55	8	-21	0
	50	22	20	37	8	2	80
	100	13	19	34	5	2	90
	500	7	6	7	2	1	98
	1000	46	50	4	5	46	99
0.5	5	53	42	88	43	35	0
	10	28	61	91	78	32	0
	50	47	56	93	65	20	80
	100	61	86	88	70	29	90
	500	29	41	48	34	17	98

	1000	18	35	42	26	22	99
1	5	77	92	92	71	54	0
	10	45	95	93	93	51	0
	50	68	97	93	92	48	80
	100	44	95	96	90	47	90
	500	45	77	68	67	32	98
	1000	18	50	46	51	32	99
2	5	90	94	57	90	71	0
	10	96	86	95	93	72	0
	50	98	98	98	96	75	80
	100	86	99	99	97	77	90
	500	86	97	98	94	68	98
	1000	57	85	86	80	60	99
5	5	90	87	89	77	91	0
	10	97	96	80	96	93	0
	50	60	99	97	97	97	80
	100	99	99	99	99	96	90
	500	74	100	98	98	95	98
	1000	-284	100	98	96	92	99

6.1.2 Fe Removal

6.1.2.1 In As (III) Test

Table A. 4 Removal percentage of Iron in As (III) Jar tests at pH 5-9

Iron Removal with As (III) (%)						
Fe (mg/l)	**As Start (µg/l)**	**pH**				
		5	**6**	**7**	**8**	**9**
0.1	5	0	6	48	47	67
	10	0	7	51	64	50
	50	0	50	61	63	58
	100	0	34	65	44	58
	500	0	11	68	61	34
	1000	0	18	71	70	57
0.5	5	12	73	96	96	90
	10	48	19	97	96	90
	50	10	68	96	94	88
	100	17	53	96	95	89
	500	9	13	97	94	91
	1000	14	69	97	96	92
1	5	10	78	97	96	96
	10	11	10	98	96	97

	50	10	93	98	94	96
	100	33	94	97	95	95
	500	53	17	96	94	92
	1000	33	45	98	96	93
2	5	66	94	99	98	98
	10	73	91	99	98	99
	50	79	92	99	98	98
	100	69	82	98	98	99
	500	92	73	99	98	98
	1000	46	87	99	96	97
5	5	72	99	100	99	99
	10	52	94	100	98	99
	50	54	98	99	100	99
	100	68	91	100	100	99
	500	43	89	100	100	99
	1000	39	93	99	100	98
10	5	15	73	100	100	100
	10	21	61	100	100	100
	50	21	54	100	100	100
	100	34	87	100	100	100
	500	35	61	100	100	100
	1000	12	55	100	100	100

20	5	31	86	100	100	100
	10	23	63	100	100	100
	50	28	63	100	100	100
	100	22	81	100	100	100
	500	21	76	100	100	100
	1000	10	66	100	100	100

6.1.2.2 In As (V) Test

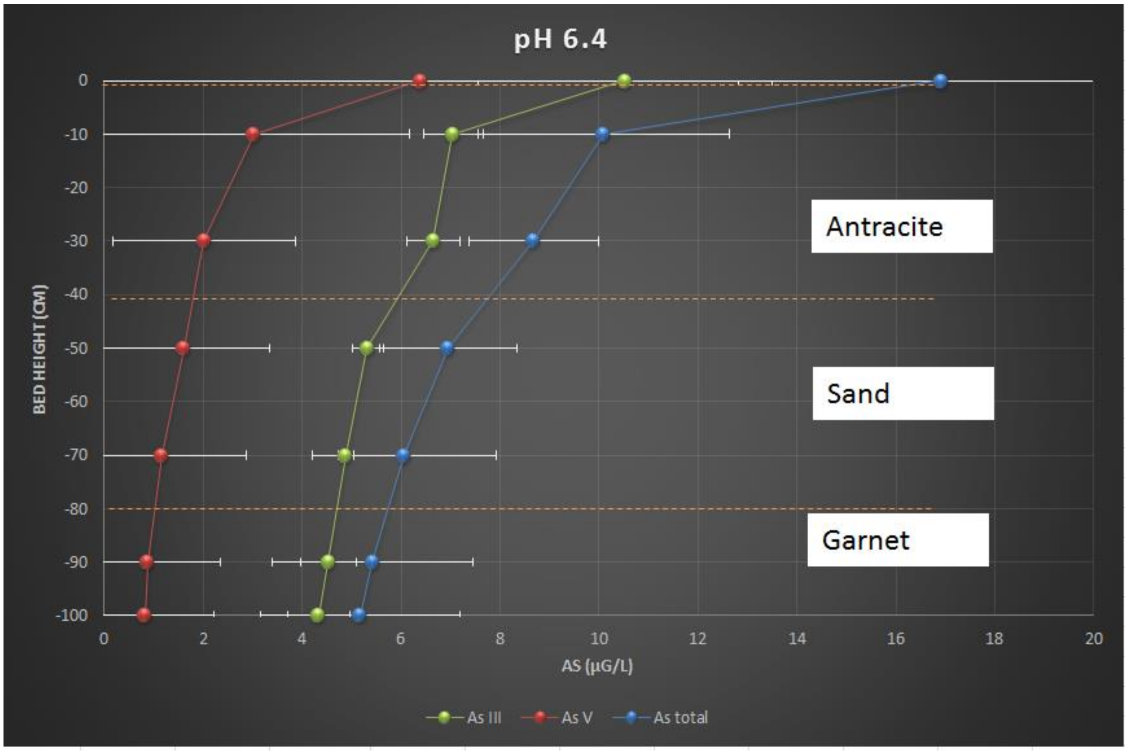
Table A. 5 Removal percentage of Iron in As (V) Jar tests at pH 5-9

Iron Removal with As (V) (%)						
Fe (mg/l)	**As Start (µg/l)**	**pH**				
		5	**6**	**7**	**8**	**9**
0.1	5	4	5	65	100	-11
	10	-1	9	61	-7	-10
	50	7	15	68	-25	-12
	100	7	31	73	-11	-12
	500	11	25	56	-12	-27
	1000	4	33	75	-11	-9
0.5	5	23	32	96	95	95
	10	-4	47	96	95	95
	50	20	36	97	96	96

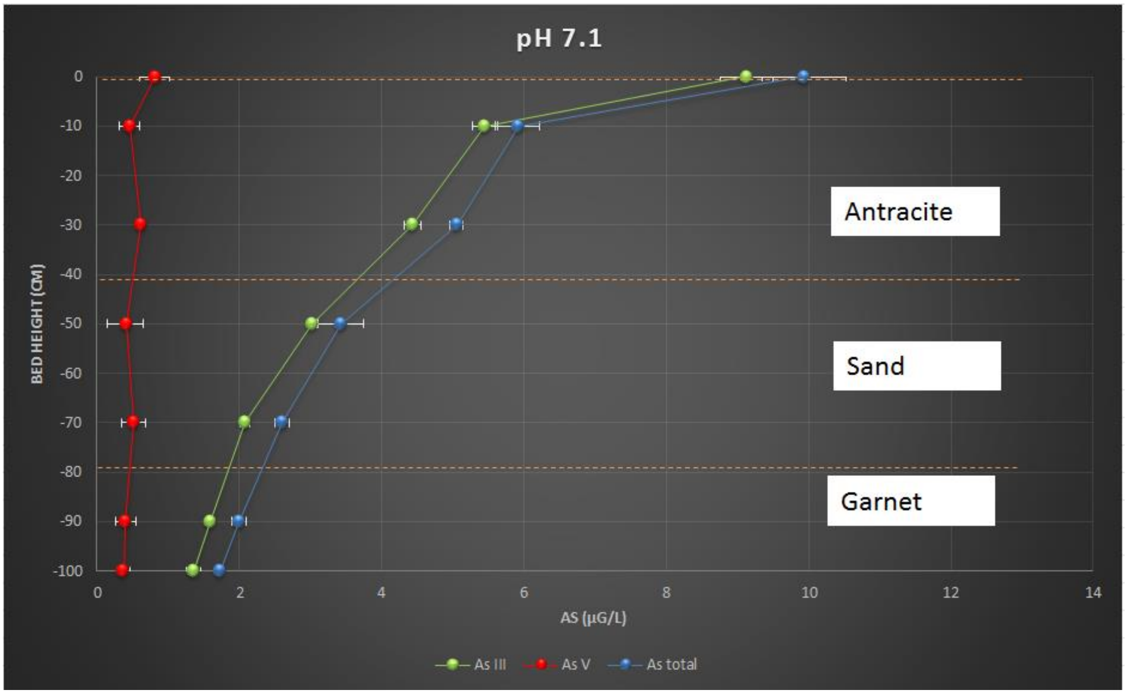
	100	43	72	95	96	96
	500	53	70	89	88	94
	1000	32	84	94	87	95
1	5	39	95	99	99	99
	10	22	94	99	99	99
	50	37	90	98	99	98
	100	28	86	99	99	98
	500	52	90	97	97	97
	1000	33	88	93	95	98
2	5	61	89	100	100	99
	10	75	83	100	100	99
	50	78	71	99	100	99
	100	46	84	100	100	99
	500	76	71	99	99	99
	1000	68	81	99	99	99
5	5	31	90	100	100	100
	10	52	76	100	100	100
	50	11	89	100	100	100
	100	62	86	100	100	100
	500	40	71	100	100	100
	1000	42	92	100	100	100

6.2 Multilayer Sand Filtration

6.2.1 Oxidation and arsenic removal Day 1



(a)



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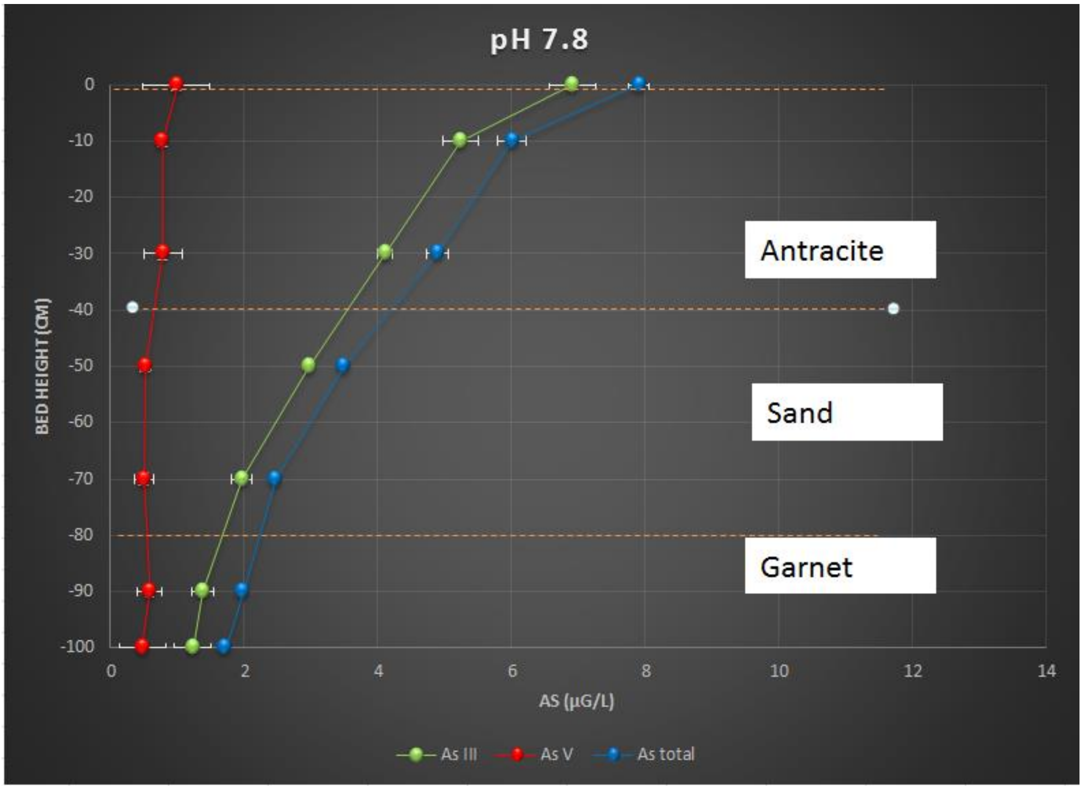
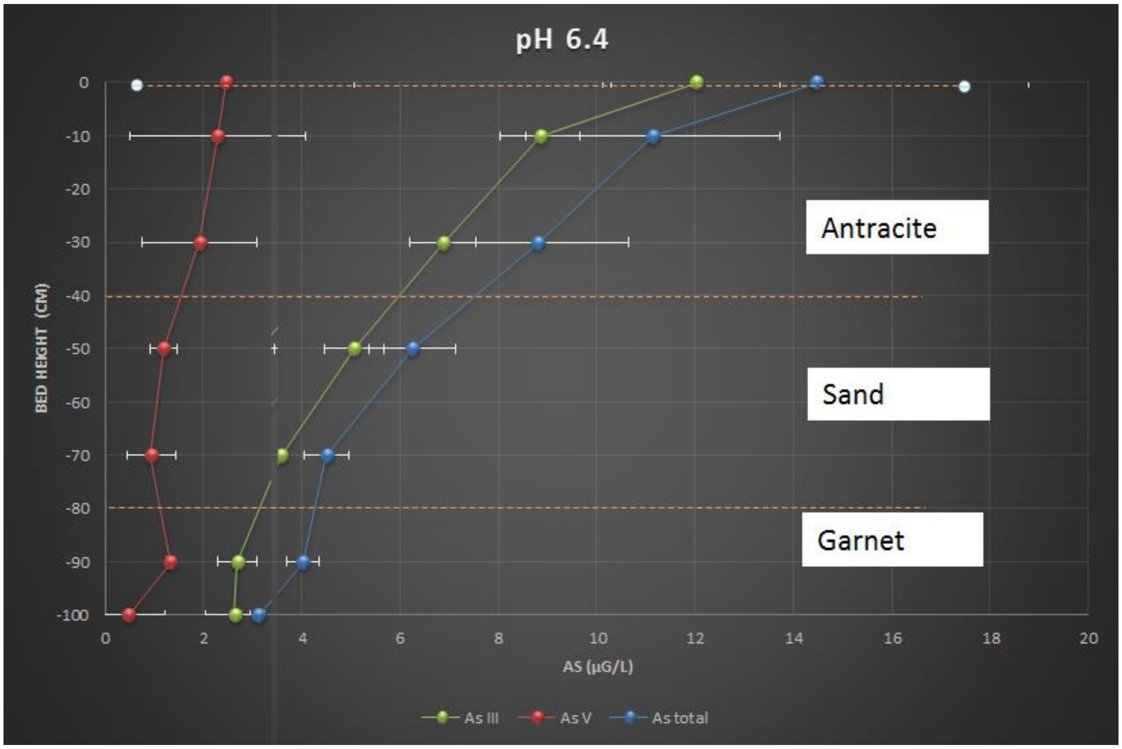


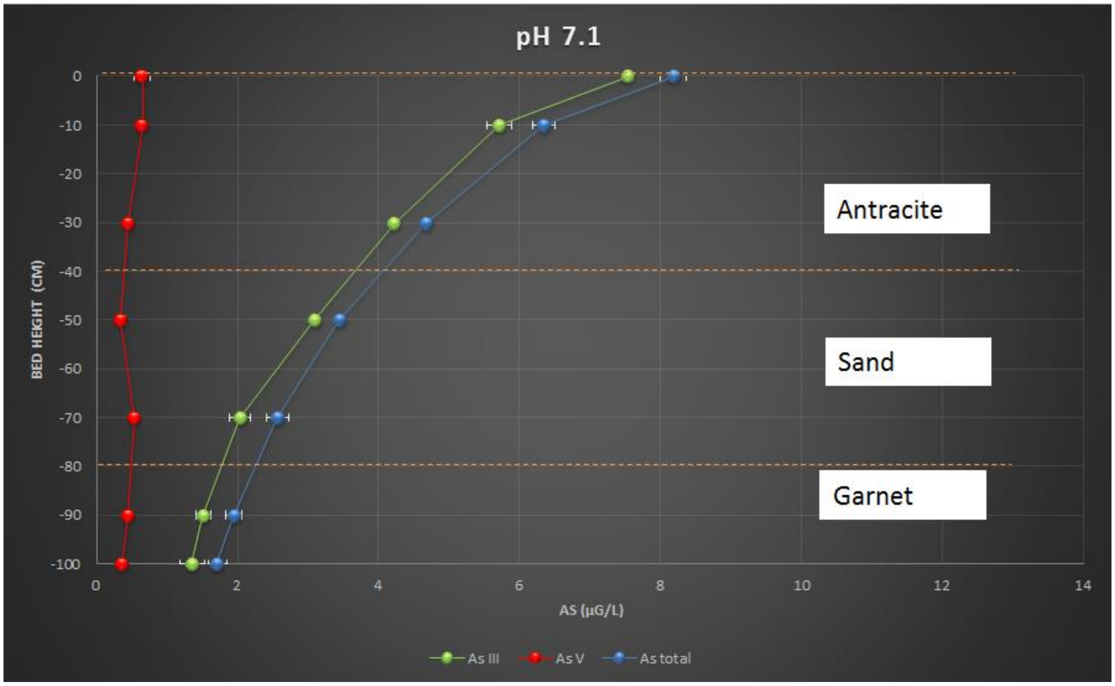
(c)

Figure A. 1 (a), (b) and (c) Total arsenic removal in the bed with speciation at Day 1 after backwash at three different pH

6.2.2 Oxidation and arsenic removal Day 2



(a)



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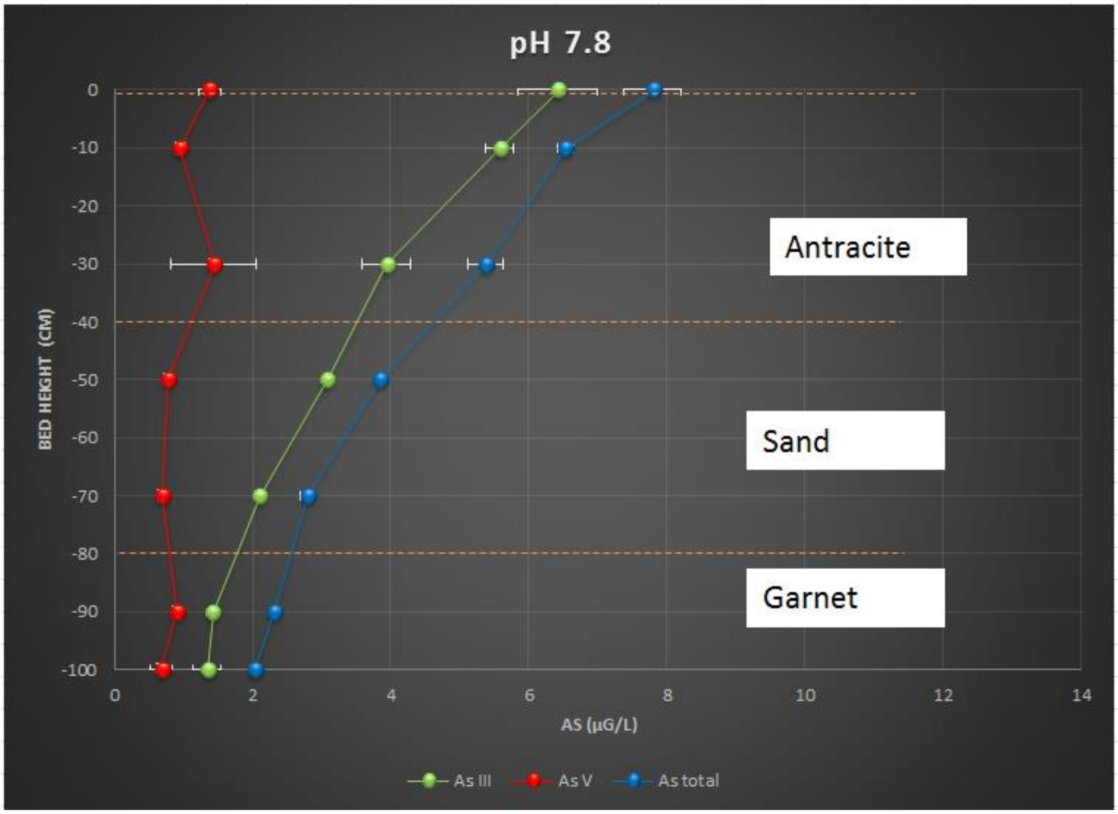
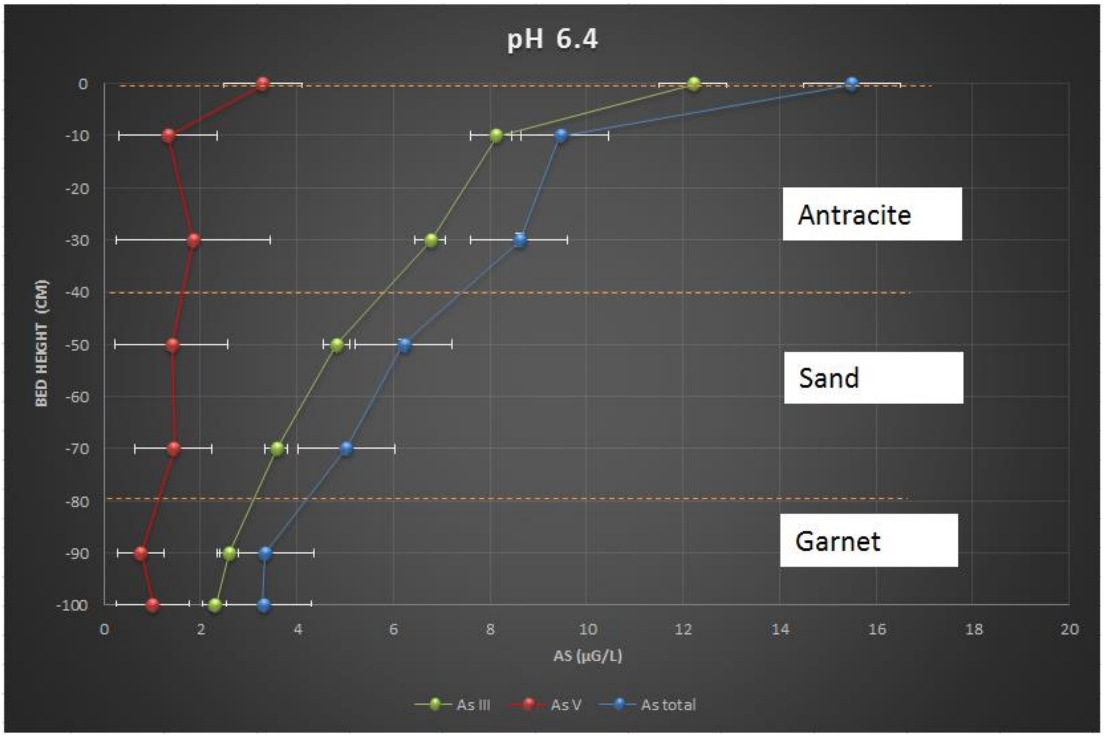


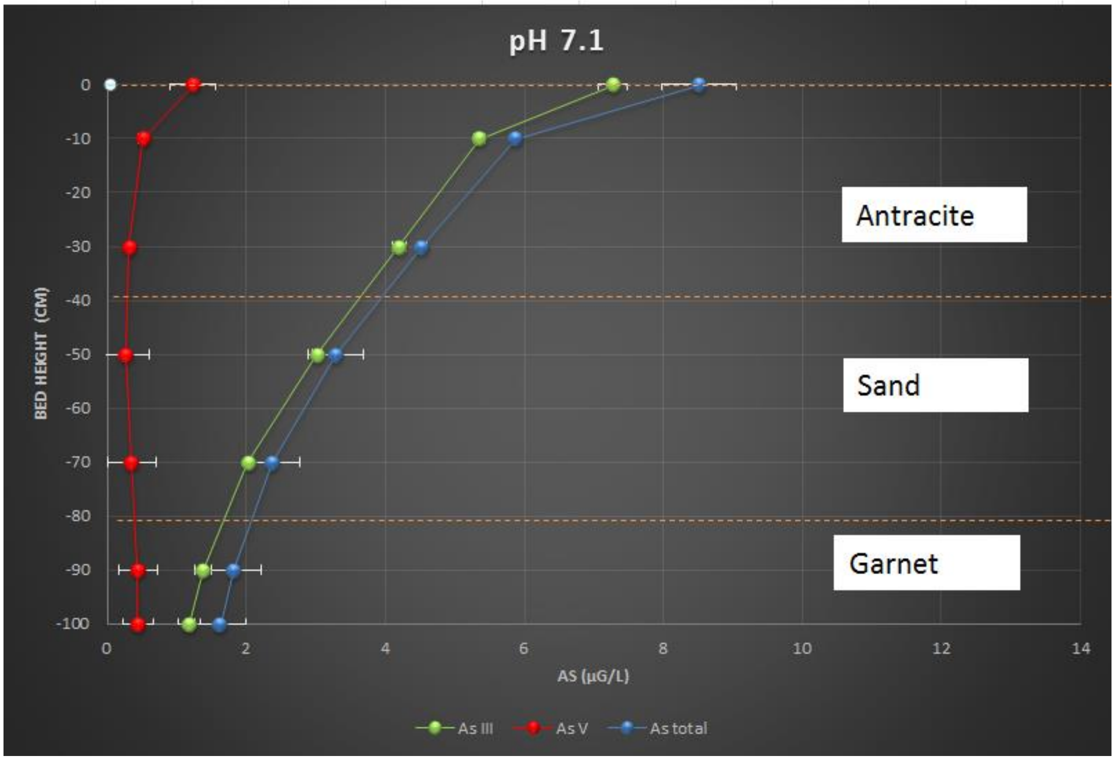
(C)

Figure A. 2 (a), (b) and (c) Total arsenic removal in the bed with speciation at Day 2 after backwash at three different pH

6.2.3 Oxidation and arsenic removal Day 3



(a)



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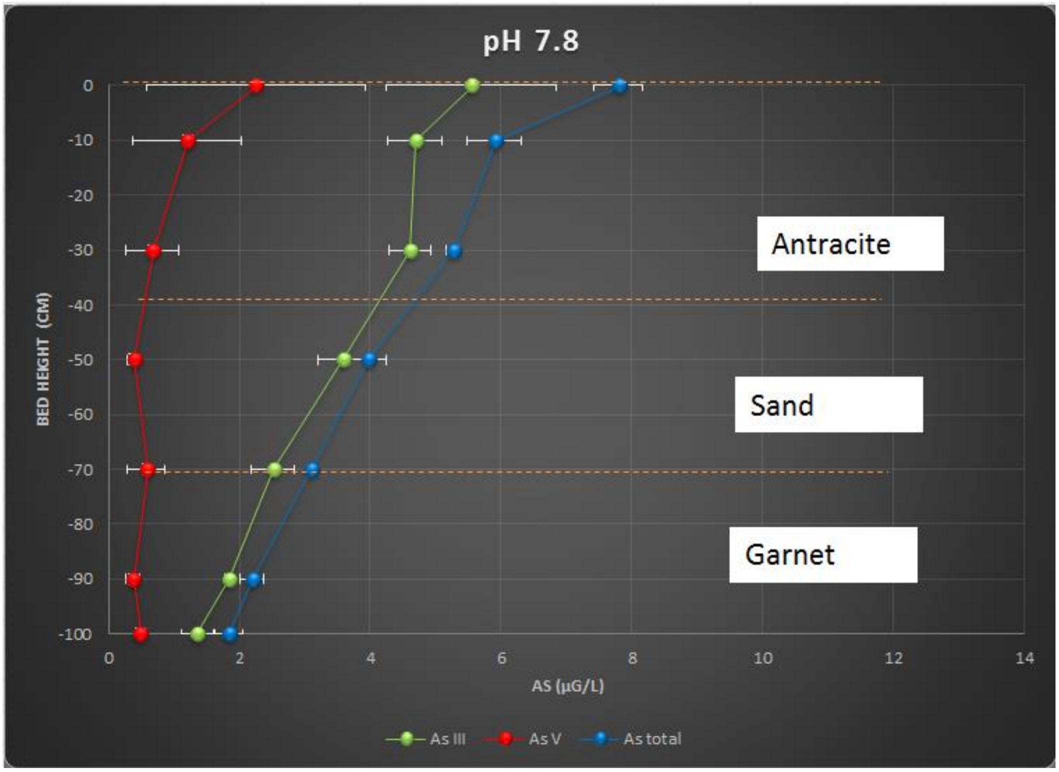
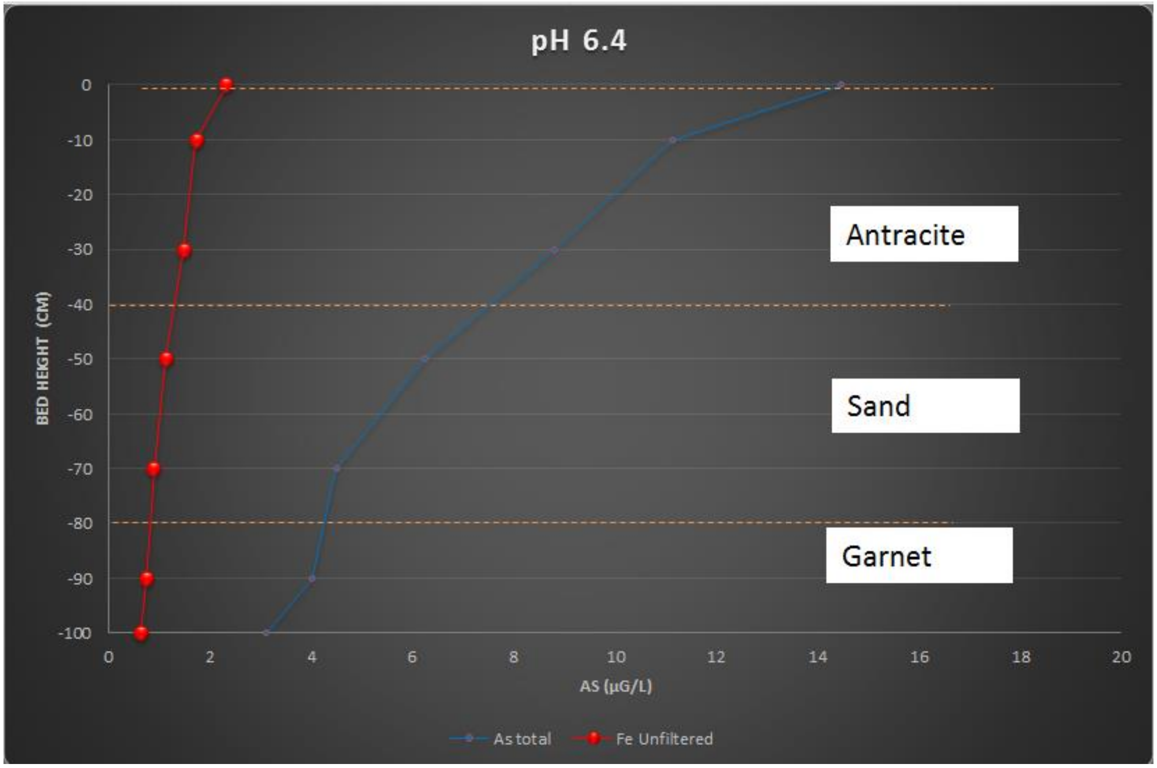


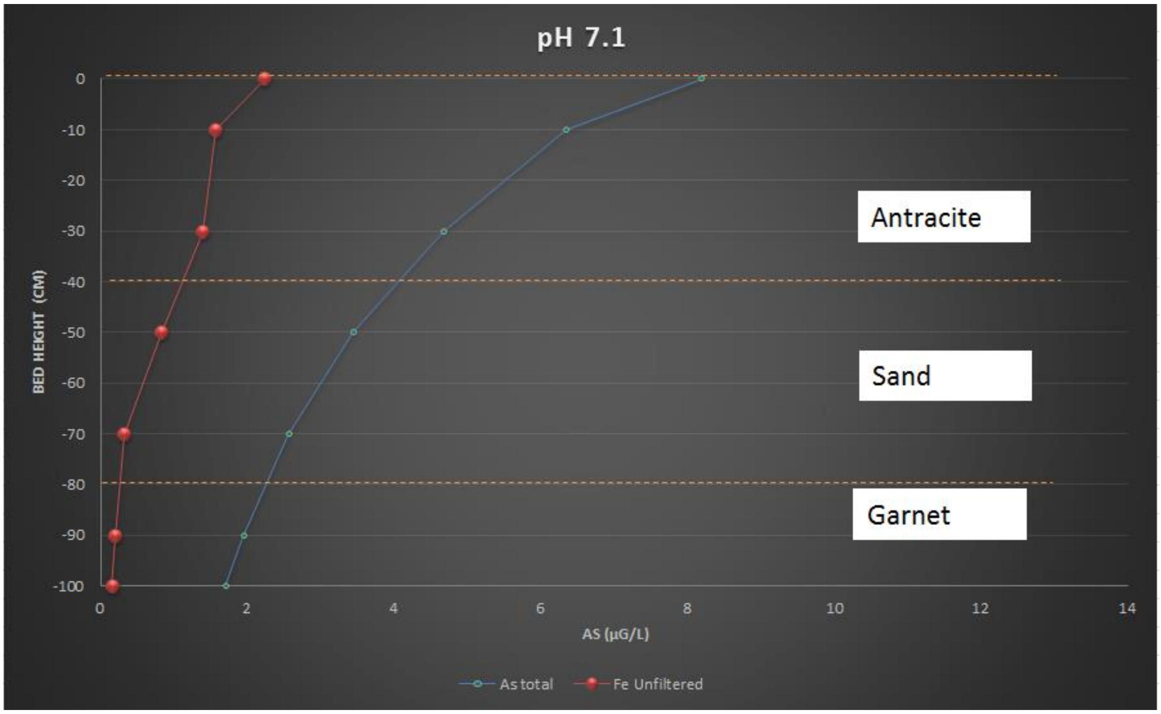
(C)

Figure A. 3 (a), (b) and (c) Total arsenic removal in the bed with speciation at Day 3 after backwash at three different pH

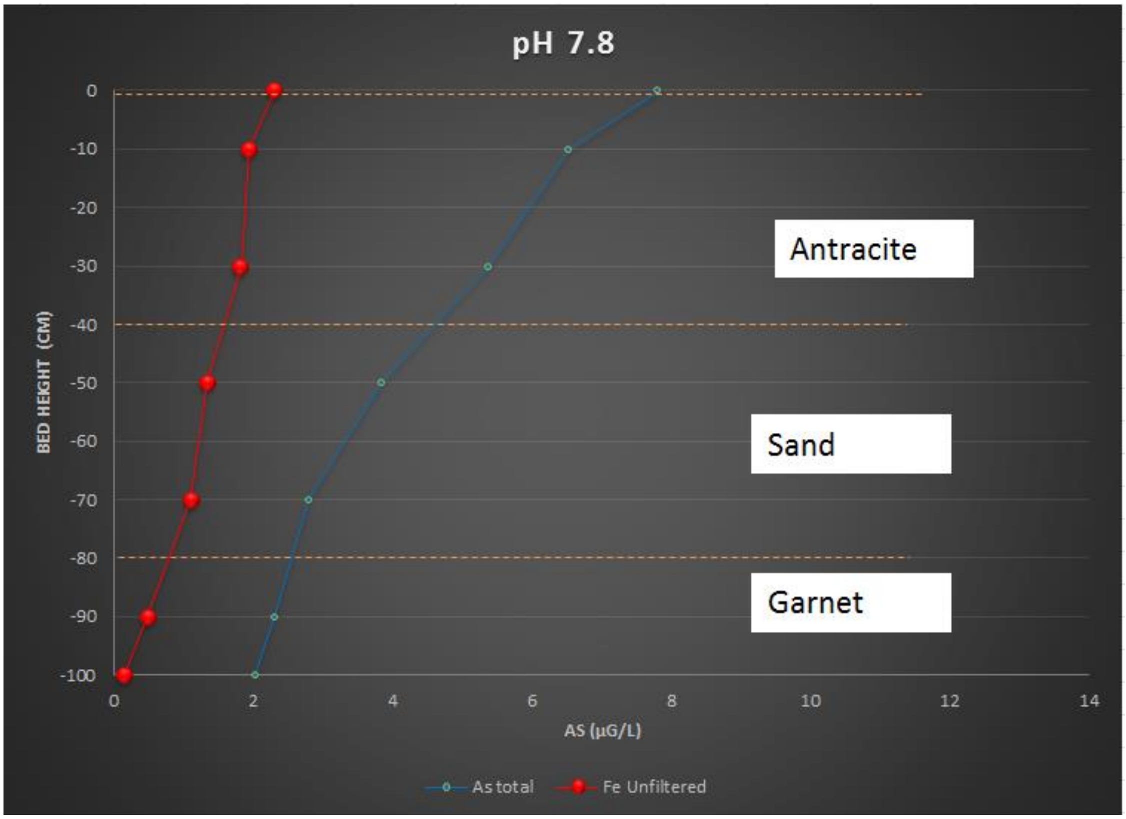
6.2.4 Arsenic removal with respect to iron Day 2



(a)



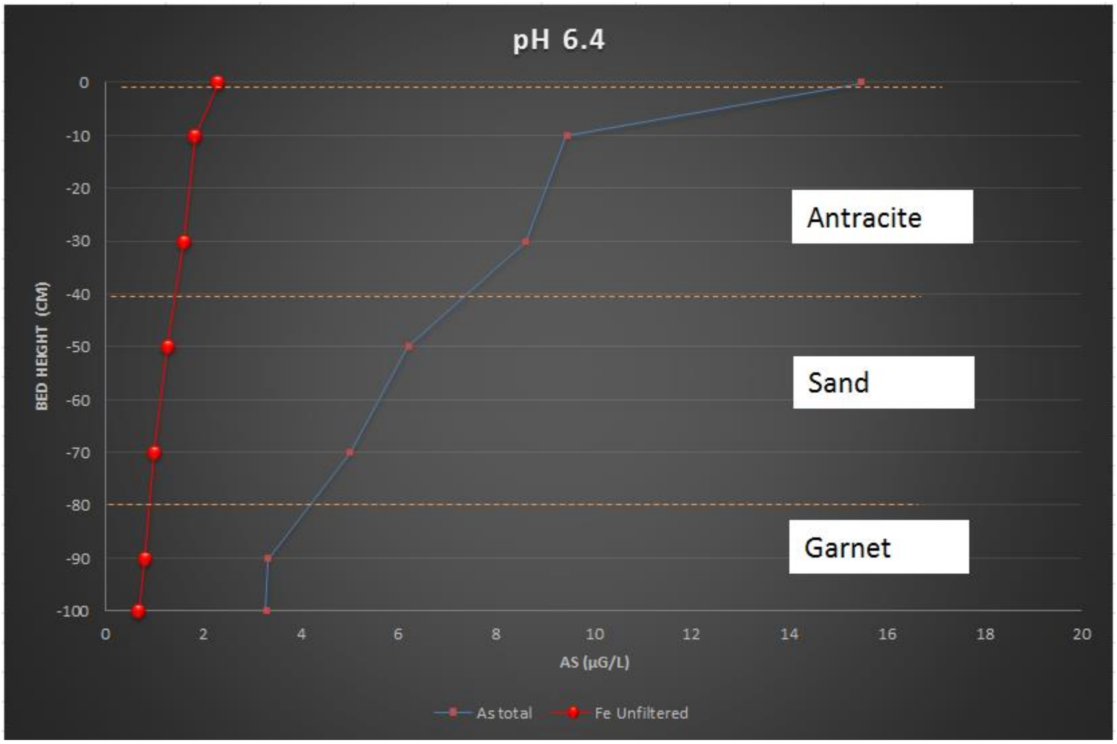
(b)



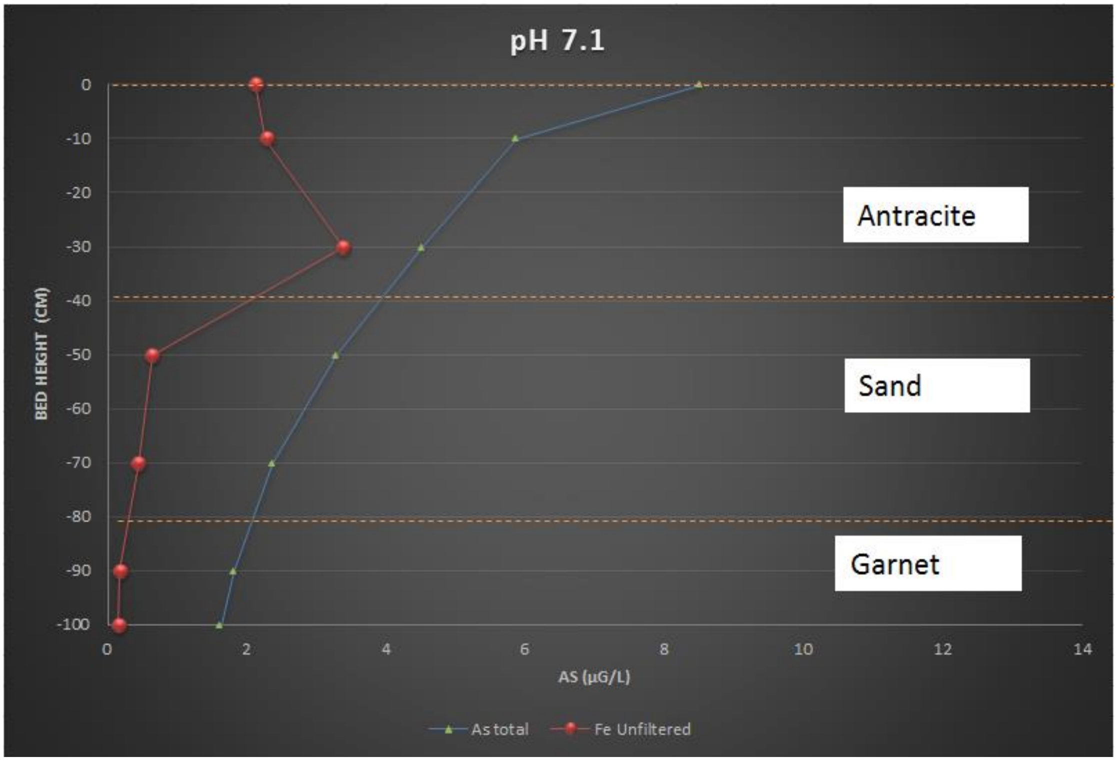
(c)

Figure A. 4 (a), (b) and (c) Total Arsenic removal with respect to Iron at day 2 for the three pH

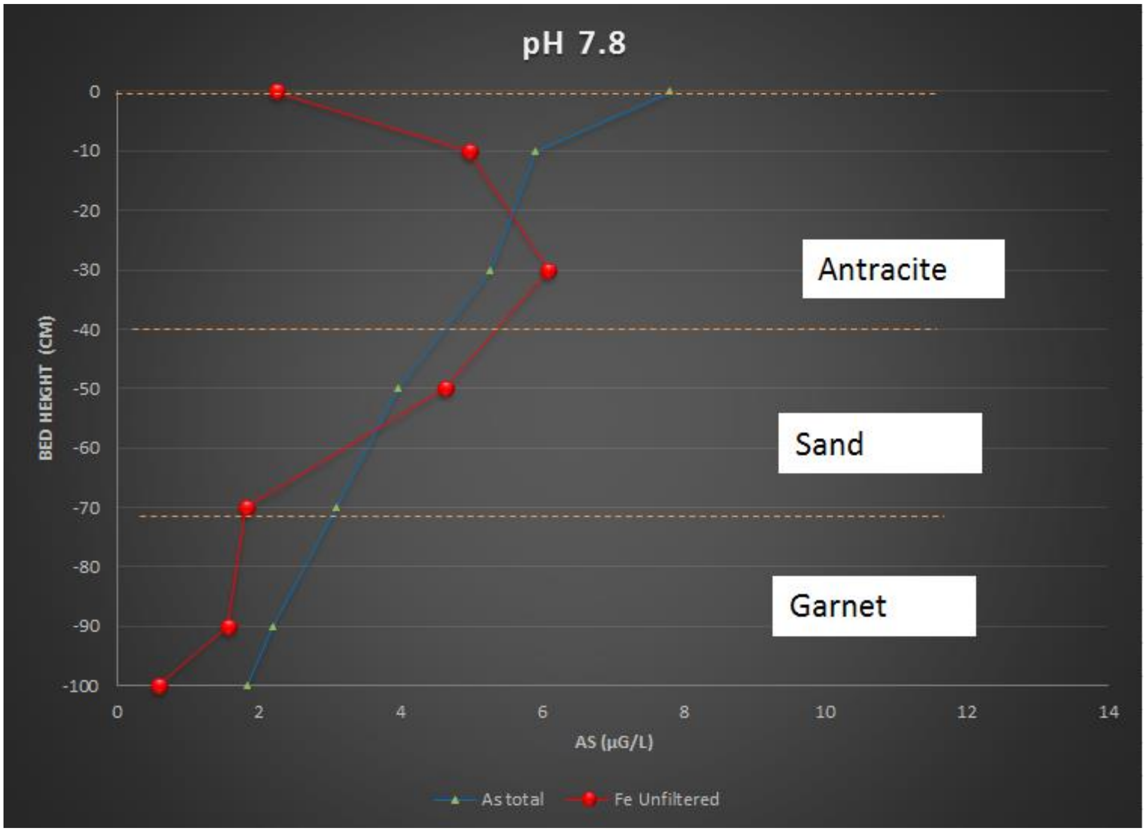
6.2.5 Arsenic removal with respect to Iron Day 3



(a)



(b)



(c)

Figure A. 5 (a), (b) and (c) Total Arsenic removal with respect to Iron at day 3 for the three pH