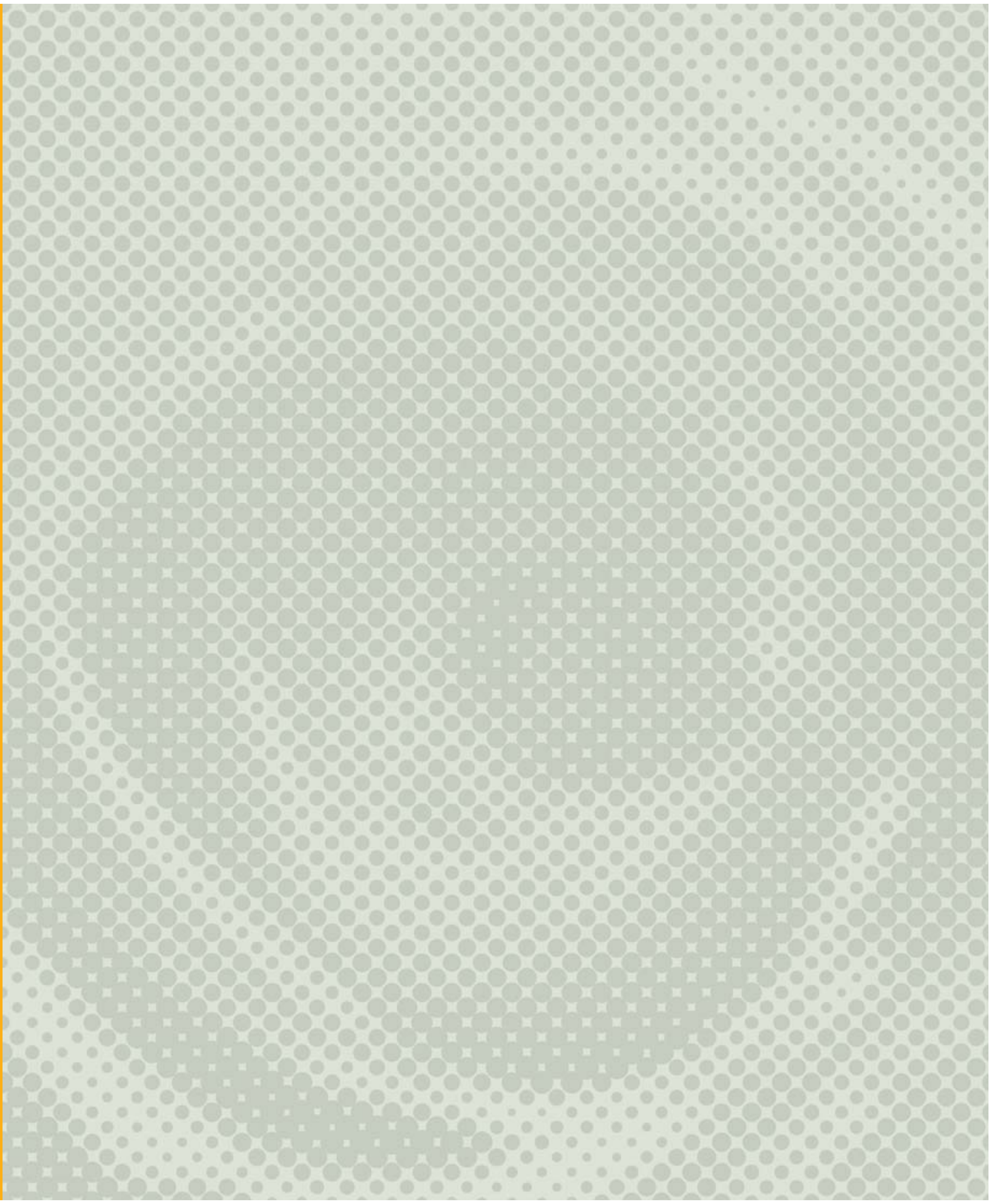
Removal of Arsenic Using Reverse Osmosis

Marie Fauskrud



NORWEGIAN UNIVERSITY OF LIFE SCIENCES
Department of Mathematical Sciences and Technology
Master Thesis 30 credits 2013

Removal of Arsenic Using Reverse Osmosis

Master Thesis at the Department of Mathematical Sciences and Technology,
Norwegian University of Life Science

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May 2013

2013

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Preface

This is my 30 credits master thesis in the Department of Mathematical Sciences and Technology at the Norwegian University of Life Science (UMB) in partial fulfilment of the requirements for the degree of Master of Technology.

I want to thank Professor Harsha Ratnaweera for guidance and supervision throughout the research. Furthermore I want to thank Karl Andreas Jensen at the Department of Plant and Environmental Sciences (IPM), for his help with analysis work, and hints during the laboratory period; and Lelum Manamperuma for instructions and help in the laboratory. I would further like to thank Tormod Kverndalen from Malthe Winje Automation ASfor instruction of the Reverse Osmosis Unit, and for troubleshooting and answering questions during the experiments. I also want to thank Hisham Jaffeer for good assistance and company in the lab.

Especially I would like to thank my friends and family who was great support, and I am grateful that Hanna Marie Volle, Birgitte Brennhovd and Stian Chan proofread and corrected the grammar.

Abstract

Arsenic is an element that naturally occurs in rocks and sediments. In some areas, arsenic is released to ground water and can contaminate drinking water sources. Since arsenic is toxic in high doses and a carcinogen, the World Health Organisation (WHO) recommends arsenic levels lower than 10 μg/L in drinking water.

This thesis describes the removal of arsenic in a reverse osmosis system. Arsenic is usually present in water in two oxidation states, as arsenite (As(III)) or arsenate (As(V)). Arsenate is usually easier to remove than arsenite, and the experiments have emphasized the removal of arsenite in double filtration process and compared to the removal of arsenate.

Previous studies at UMB have found reverse osmosis as an efficient way to remove arsenite when the water is filtrated twice, almost as efficient as arsenate removal in one filtration. One hypothesis that was tested was that arsenite was oxidized during the removal process and was removed easily as arsenate in the second filtration.

The verification of arsenate removal gave a removal efficiency of 97-99 % of As(V) after one filtration. Double filtration of arsenite gave a total arsenic removal between 91.8-94.3 %.

Oxidation of arsenite to arsenate was tested and showed a small oxidation rate of between 5 and 10 %. During the filtration processes, about 5 % of arsenite was oxidized. However, an effective membrane and RO unit rather than oxidation caused the great removal efficiency of arsenite.

Table of Contents

Abbreviations and Glossary

Arsenicosis	Chronic arsenic poisoning
Arsenic acid	H_3AsO_4, arsenic is present in pentavalent form, As(V)
Arsenite	As(III)
Arsenous acid	$HAsO_2$, arsenic is present in trivalent form, As(III)
Arsenate	As(V)
DOC	Dissolved Organic Carbon
EPA	US Environmental Protection Agency
HPLC	High-Performance Liquid Chromatography
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IE	Ion Exchange
IMT	the Department of Mathematical Sciences and Technology
IPM	the Department of Plant and Environmental Sciences
RO	Reverse Osmosis
RO1	The first filtration process in the reverse osmosis unit
RO2	The second filtration process in the reverse osmosis unit
WHO	World Health Organisation

List of Figures

List of Tables

1 Introduction

In parts all over the world the ground contains arsenic naturally, and the arsenic dissolves in contact with water. Arsenic contaminated water is being pumped out of shallow wells and is a public health problem in some countries. There medical treatment for arsenic poisoning is unknown, but early symptoms of arsenicosis may improve by drinking arsenic free water. The alternative options for water supply in areas with arsenic contamination include arsenic avoidance and treatment. The two technological options for arsenic mitigation are to switch to arsenic-free water sources, or remove arsenic from the ground water. Treatment of surface waters, rainwater harvesting and water from deep aquifers could be possible alternative sources to the arsenic contaminated shallow wells. The treated surface waters can be used for drinking, cooking and irrigation, while the arsenic contaminated water can be used for other purposes, i.e. flushing the toilet, cleaning, dishwashing, etc. Another option is to treat the well water to remove arsenic to acceptable levels (Ahmed 2001).

Arsenic is a known carcinogen; and can cause skin lesions and affect internal organs (Choong et al. 2007; Smith & Steinmaus 2009). The recommended limit set by WHO is 10 µg/L, and the removal of arsenic below this value should be strived to pursue. Even though arsenic is an important public threat, illness and death due to other waterborne diseases are also a serious health risk. Since water from arsenic contaminated wells has less bacteriological contaminants than contaminated surface water, finding a better alternative is of great importance. However, treatment of surface water and removal of arsenic are two sides of the same coin in order to provide safe drinking water (Ahmed & Talbi 2005b).

The fact that many people are affected by arsenic contaminated water and the severity of the problem make mitigation solutions interesting. This thesis examines the chemical properties of arsenic, the scope of the problem, and treatment options. The main scope of the thesis is to examine the removal rate of As(III) in a reverse osmosis unit by filtrating the contaminated water twice.

Chapter 2 describe the background for the experiment: the chemical properties, the toxicity of arsenic, the scope of the problem and mitigation solutions in order to see the severity of arsenic contamination. Several removal methods for household usage was described to give an idea of the opportunities available.

Previous studies are mentioned in the following chapter and include key findings in previous tests on arsenic removal using reverse osmosis. The removal efficiency of arsenic using reverse osmosis (RO) is well documented. As(III) is more complicated to remove than As(V) (Ahmed & Talbi 2005a; Figoli et al. 2010). Yet, previous studies at UMB have shown good results on As(III) removal using double filtration.

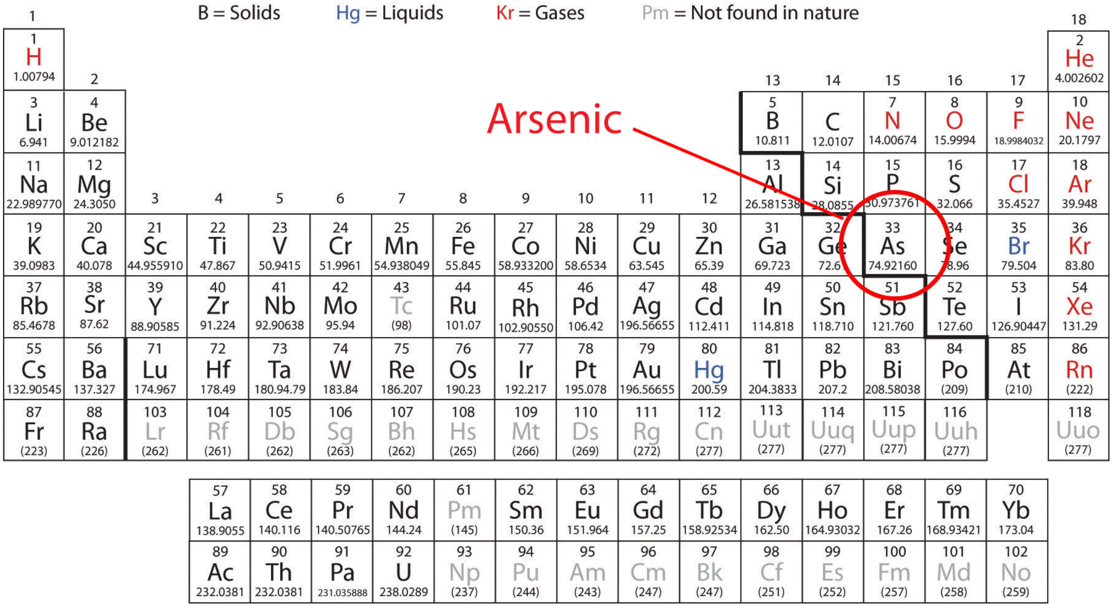
The removal of As(III) with RO was tested on pilot scale, and the effect of double filtration was examined. The experiment was designed to examine whether the high removal rates were due to oxidation of As(III) to the more easily removed As(V) or the membranes effectiveness. To have a basis of comparison, experiments using As(V) in the RO unit and tests of oxidation by stirring arsenic contaminated water samples were conducted. The method and material are described in chapter 4, then; the following chapters will give the results of the experiments and discuss the test results.

The sentences and language used in the thesis is as simple as possible, however, the reader should have some technological knowledge related to drinking water treatment.

2 Background

2.1 Chemistry of Arsenic

Arsenic is a chemical element with atomic number 33. It is a metalloid in group 15 and period 4 (Stølen 2011).



1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H 1.00794																	2 He 4.002602
3 Li 6.941	4 Be 9.012182											5 B 10.811	C 12.0107	7 N 14.00674	8 O 15.9994	9 F 18.9984032	10 Ne 20.1797
11 Na 22.989770	12 Mg 24.3050											13 Al 26.581538	14 Si 28.0855	15 P 30.973761	16 S 32.066	17 Cl 35.4527	18 Ar 39.948
19 K 39.0983	20 Ca 40.078	21 Sc 44.955910	22 Ti 47.867	23 V 50.9415	24 Cr 51.9961	25 Mn 54.938049	26 Fe 55.845	27 Co 58.933200	28 Ni 58.6534	29 Cu 63.545	30 Zn 65.39	31 Ga 69.723	32 Ge 72.6	33 As 74.92160	34 Se 78.96	35 Br 79.504	36 Kr 83.80
37 Rb 85.4678	38 Sr 87.62	39 Y 88.90585	40 Zr 91.224	41 Nb 92.90638	42 Mo 95.94	43 Tc (98)	44 Ru 101.07	45 Rh 102.90550	46 Pd 106.42	47 Ag 196.56655	48 Cd 112.411	49 In 114.818	50 Sn 118.710	51 Sb 121.760	52 Te 127.60	53 I 126.90447	54 Xe 131.29
55 Cs 132.90545	56 Ba 137.327	71 Lu 174.967	72 Hf 178.49	73 Ta 180.94.79	74 W 183.84	75 Re 186.207	76 Os 190.23	77 Ir 192.217	78 Pt 195.078	79 Au 196.56655	80 Hg 200.59	81 Tl 204.3833	82 Pb 207.2	83 Bi 208.58038	84 Po (209)	85 At (210)	86 Rn (222)
87 Fr (223)	88 Ra (226)	103 Lr (262)	104 Rf (261)	105 Db (262)	106 Sg (263)	107 Bh (262)	108 Hs (265)	109 Mt (266)	110 Ds (269)	111 Rg (272)	112 Cn (277)	113 Uut (277)	114 Uuq (277)	115 Uup (277)	116 Uuh (277)		118 Uuo (277)

57 La 138.9055	58 Ce 140.116	59 Pr 140.50765	60 Nd 144.24	61 Pm (145)	62 Sm 150.36	63 Eu 151.964	64 Gd 157.25	65 Tb 158.92534	66 Dy 162.50	67 Ho 164.93032	68 Er 167.26	69 Tm 168.93421	70 Yb 173.04
89 Ac 232.0381	90 Th 232.0381	91 Pa 231.035888	92 U 238.0289	93 Np (237)	94 Pu (244)	95 Am (243)	96 Cm (247)	97 Bk (247)	98 Cf (251)	99 Es (252)	100 Fm (257)	101 Md (258)	102 No (259)

Figure 1: Arsenic marked in the periodic table (Ball et al. 2011)

Arsenic is found in oxidation states of –3, 0, +3 and +5 in the Earth's crust, often as sulfides, arsenates or metal arsenides. In surface water, it is present as mostly arsenate, As(V). In anaerobic condition it is likely to have arsenic present as arsenite, As(III). (*Arsenic: Medical effects* 1977; Ning 2002; WHO 2011b).

Arsenic(V) acid, H_3AsO_4, and arsenous(III) acid, $HAsO_2$, and their salts can serve as model for the species predominating in natural waters (Ning 2002).

Chemistry of the two acids:

The two oxidation states, As(V) and As(III), have significant chemical differences. The equilibrium constants of dissociation are quite different:

H_3AsO_4, As(V) $pKa_1 = 2{,}19$, $pKa_2 = 6{,}94$, $pKa_3 = 11{,}5$.

H_3AsO_3, As(III) $pKa_1 = 9{,}20$ (the other constants are unknown)

At natural pH levels, arsenic acid is highly ionized and present as $H_2AsO_4^-$ and $HAsO_4^{2-}$, and arsenous acid is largely unionized. Due to the ionic charge, arsenate is more easily removed than arsenite (Ning 2002).

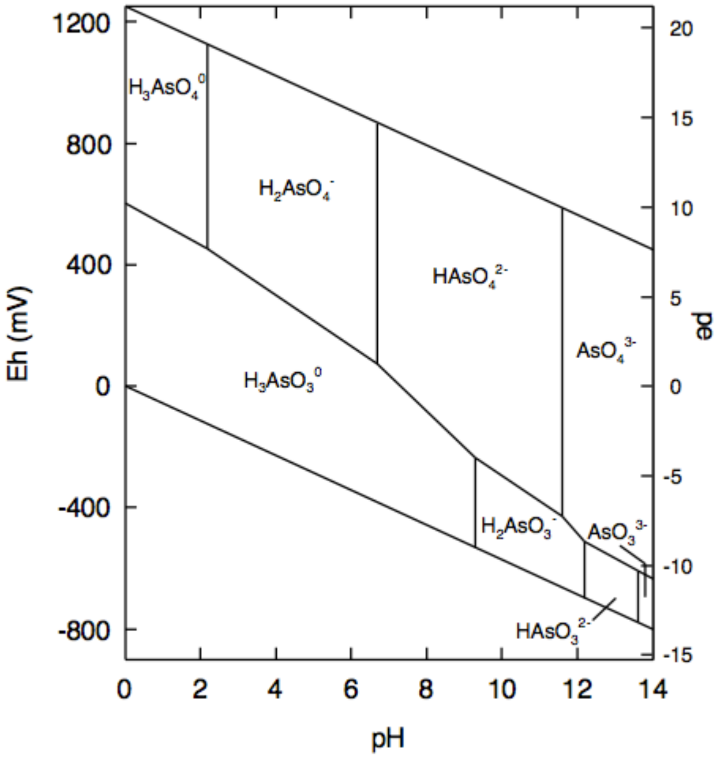


Figure 2: The Eh-pH diagram for arsenic at 1 atm and 25 °C. Displays which species of arsenite and arsenate is present at different pH (Smedley & Kinniburgh 2001)

Arsenic is a toxic compound, and the toxicity scale of arsenic is arsine > inorganic As (III) > organic As (III) > inorganic As (V) > organic As (V) > arsonium compounds and elemental arsenic (Gholami et al. 2006).

2.2 Health Effects of Arsenic

Exposure to arsenic contaminated water over a longer period of time can cause different health problems such as skin lesions, cancer (internal cancers in bladder, kidney, lung; skin cancer) neurological effects, hypertension and cardiovascular disease, pulmonary disease, peripheral vascular disease and diabetes (Smith et al. 2000). Lethal doses in humans range from 1.5 mg to 500 mg per kg bodyweight (As_2O_3). The toxicology of arsenic is divided in two classes, acute and sub-acute. Acute arsenic poisoning from consuming well water containing high concentrations (1.2-21.0 mg/L) of arsenic has been reported. The early manifestation of acute arsenic poisoning are burning and dryness of mouth and throat, vomiting, diarrhoea, dysphasia, colicky abnormal pain and haematuria (Choong et al. 2007; Smith & Steinmaus 2009; WHO 2011a; WHO 2011b).

Arsenicosis is caused by exposure of arsenic in drinking water over a longer period (5-20 years) (WHO 2001). There are four recognized stages of arsenicosis (Choong et al. 2007):

I) Preclinical:

Arsenic can be found in urine and hair, but the patient has no symptoms. Arsenic is present in every living organism. In a study conducted in 1977, the median content in human hair was found to be 0.51 ppm. If the arsenic level in human hair exceeds 2-3 ppm, it may indicate poisoning (*Arsenic: Medical effects* 1977).

II) Clinical:

Some effects can be seen on the skin. WHO estimates that this stage takes 5-10 years. Darkening of the skin (hyperpigmentation), especially on the upper chest and arms; oedema (swelling of hands and feet), and hardening of skin into nodules (keratosis) on the palms and soles of the feet are the most common signs of arsenic-caused diseases unless in very mild early cases. The diagnosis of diseases caused by chronic ingestion of inorganic arsenic is confirmed if the patient is found to have been drinking arsenic-contaminated water (Smith et al. 2000).

III) Complications:

Clinical symptoms become more pronounced, and internal organs are affected. Diabetes may be linked to arsenic exposure in this stage.

IV) Malignancy:

The affected person may develop cancer in bladder, lung, skin, or internal cancer

Skin lesions

The latency for skin lesions from the first exposure of arsenic contaminated water to manifestation of disease is typically 10 years, and the rapidity of the appearance of skin lesions appears to be dose dependent (Smith et al. 2000).

Cancer

Arsenic is a carcinogen. The latency of skin cancer is typically more than 20 years (Smith et al. 2000).

Table 1: Lifetime risk of cancer as a result of exposure to arsenic contaminated water (van Halem et al. 2008)

Lifetime cancer risk	Arsenic concentration (µg/L)	
	EPA/IRIS (1998)	NRDC (200
10-2 (1 in 100)		50
10-3 (1 in 1 000)		5
10-4 (1 in 10 000)	2	0.5
10-5 (1 in 100 000)	0.2	
10-6 (1 in 1 000 000)	0.02	

As seen in Table 1, 1 % of a population drinking water containing 50 µg/L is in risk of developing cancer. Even small concentrations can cause cancer when it is ingested over a long period.

Cardiovascular disease

Arsenic can contribute to the development of diabetes and hypertension (high blood pressure) that can lead to cardiovascular disease. Patients with arsenicosis should therefore test their urinary glucose and blood pressure should be monitored (Smith et al. 2000).

2.3 Pathways of Arsenic Exposure

Chronic arsenic-related health problems is closely linked to consumption of contaminated water. Food is another potential pathway of arsenic. Irrigation of crops is potentially vulnerable to arsenic absorption. Few results have been published, but some studies have shown a higher concentration in vegetables. Arsenic is toxic to plants and the absorption by plants may be inhibited and may therefore not be of concern. Further studies have to be carried out to explore this topic.

Burning of arsenic rich coal has caused severe chronic health problems in Guizhou province in China where chillies were dried over the coal. The exposure was from both inhalation and consumption of chillies (Ahmed & Talbi 2005b).

2.4 Natural Distribution of Arsenic

Arsenic is the twentieth element most abundant in the earth's crust (*Arsenic: Medical effects* 1977). Natural waters have a concentration of arsenic less than 1-2 µg/L, however ground waters can have elevated concentrations up to 12 mg/L due to sulfide mineral deposits and sedimentary deposits deriving from volcanic rocks (WHO 2011b). In rocks, arsenic is found in ranges from 0.5 to 2.5 mg/kg, and in sediments from 3 to 10 mg/kg (Shih 2005).

Arsenic is a worldwide problem; parts of Europe, South and East Asia, Argentina, Mongolia, Mexico, Chile, Ghana, USA, New Zealand, etc. have elevated arsenic levels in water and/or soil. Anthropogenic activities may enhance arsenic concentrations in the environment, for example from industrial effluents.

In some areas, older aquifers have been found to have lower arsenic concentration. Deep wells can therefore have none or low concentrations of arsenic. However, in other areas there is no difference. In addition, the arsenic concentration is not evenly distributed. Two wells in a distance of a few hundred meters can have very different concentrations of arsenic. Variations in concentration due to temperature have also been found. An investigation of the local conditions is necessary to recognize if the area is at possible risk (Ahmed & Talbi 2005b).

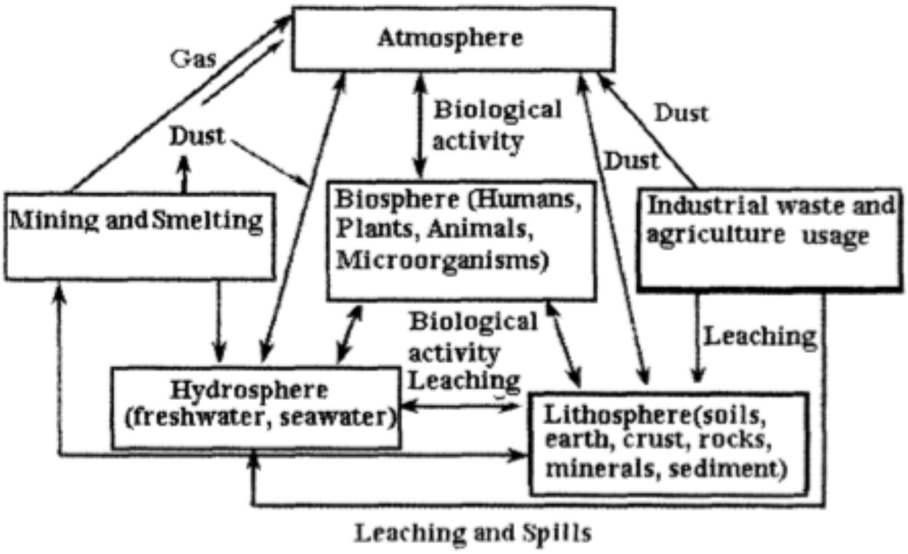


Figure 3: The arsenic cycle (Shih 2005)

Figure 3 shows the arsenic cycle, where arsenic is found and how it can change from one form to another. Arsenic is a part of the cycle; small concentrations are natural for animals and humans. The problems arise when arsenic is ingested over time.

2.5 Scale of Problem and Reason for the Arsenic Occurrence

Arsenic contaminated water is found all over the world, as seen in Figure 4. High concentrations can be found in oxidizing (under conditions of high pH) and reducing aquifers and in areas affected by geothermal, mining and industrial activity (Mandal & Suzuki 2002).

Researchers believe that about 140 million people are being poisoned by arsenic in their drinking water, mostly in developing countries. It is present in around 70 countries reports research associate in geography with Cambridge University, Peter Ravenscroft (cited by Black (2007)). Bangladesh is the worst affected country and has the highest percentage of contaminated wells. In Bangladesh alone, between 35-77 million people out of 125 million inhabitants are exposed to chronic arsenic poisoning (Figoli et al. 2010; Jackson & Jackson 2000; Leventon & Hug 2010).

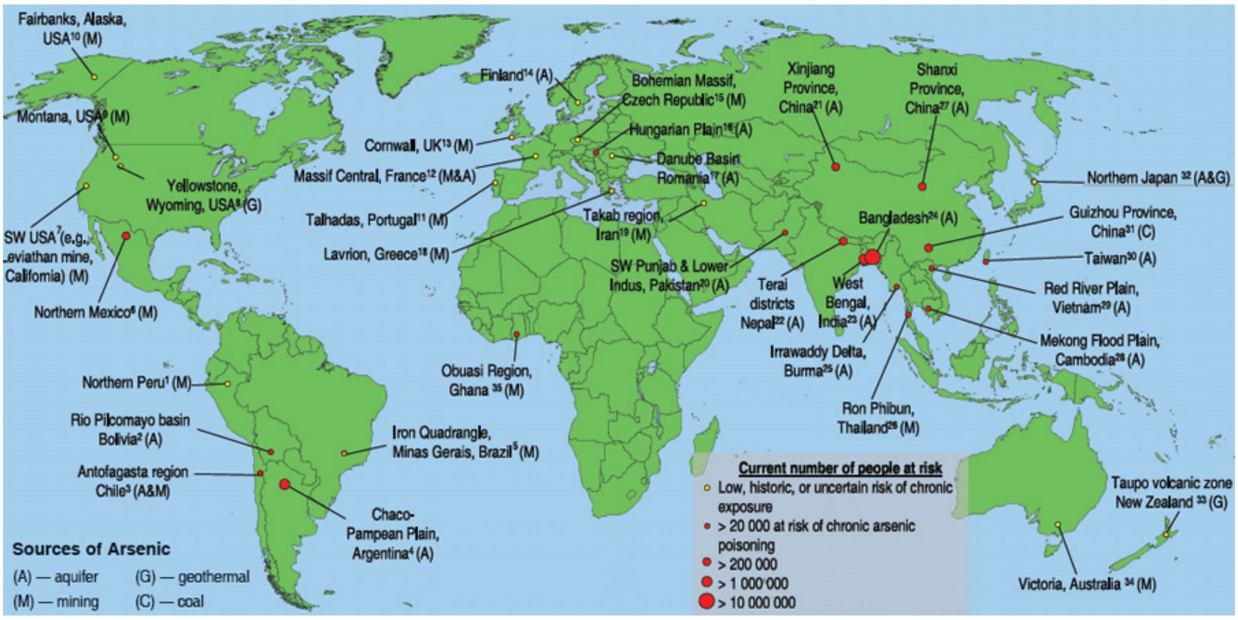


Figure 4: Areas with arsenic contaminated water and the source of the contaminant (Garelick & Jones 2008).

As seen on Figure 4, a lot of the affected areas are developing countries. Arsenic can cause problems for those having wells, treatment plants usually remove arsenic efficiently and piped water is safe if the municipal is aware of the problem. Groundwater is normally considered uncontaminated, and is therefore not usually treated. If the ground water is contaminated, the consumers will be directly affected by the arsenic contamination. To explain the scale of the problem and some of the areas and countries which have arsenic contaminated water will be explained briefly.

2.5.1 Natural Groundwater Arsenic Contamination

Most high-arsenic groundwater provinces are the result of natural occurrences of arsenic (Mandal & Suzuki 2002). Arsenic contamination can result from different geochemical mechanisms. Oxidizing conditions can mobilize arsenic from sulfides; and reducing conditions can lead to reduction of ferric oxyhydroxides and mobilize adsorbed arsenic (Jacks et al. 2010).

Asia

As seen in Figure 4, there is arsenic naturally in the ground in many of the Asian countries: Pakistan, Iran, Nepal, Myanmar, China, Japan, Taiwan, Vietnam, Cambodia, Bangladesh, India and Thailand. However, the most affected area is South – and East Asia.

Himalayan sediments contain arsenic, and rivers transport these sediments until they are deposited in low-lying regions. Desorption of arsenic from iron (and other metal-) oxides are favoured under anaerobic condition. Such conditions tend to occur in the Quaternary strata (thousands to tens of thousands of years old) underlying large alluvial and deltaic plains in South and East Asia such as the Bengal basin, Mekong valley and Red River delta (Ahmed & Talbi 2005b; Fendorf et al. 2008; Jain & Singh 2012). An estimation shows that 700 000 people in South and East Asia have been affected by arsenicosis and 60 million people are at risk from high levels of naturally occurring arsenic in groundwater (Ahmed & Talbi 2005b).

In the 1970's and 80's new wells were made in Bangladesh to give the people an alternative to surface water with high bacteria levels (Normannsen 2010). In the late 1980's and early 90's arsenic was first detected in Bangladesh and Eastern India. This drew attention to the matter. A survey in the 1990's showed that 27% of shallow tubewells in Bangladesh exceed the previous WHO arsenic limit of 50 µg/L, while 46% exceeded the WHO's guideline value of 10 µg/L. Most cities and municipalities supply water from deep aquifers free of arsenic. However, in rural areas, shallow tubewells are the principal sources of drinking water (Inauen et al. 2013).

Between 2000 and 2006, about 5 million wells were tested and painted red (arsenic contaminated water) or green (safe drinking water). Alternative sources were installed and by 2006 an expert review estimated that of the initially affected people, 29 % had switched to arsenic-safe tubewells, and another 12 % had switched to deep wells (Inauen et al. 2013).

Europe

Arsenic is also found in European ground water. Europe has a great diversity of arsenic contamination, but the occurrences have a limited human impact, with the exception of the Great Hungarian Plain (number 78 in the map in Figure 5) (Ravenscroft et al. 2009).

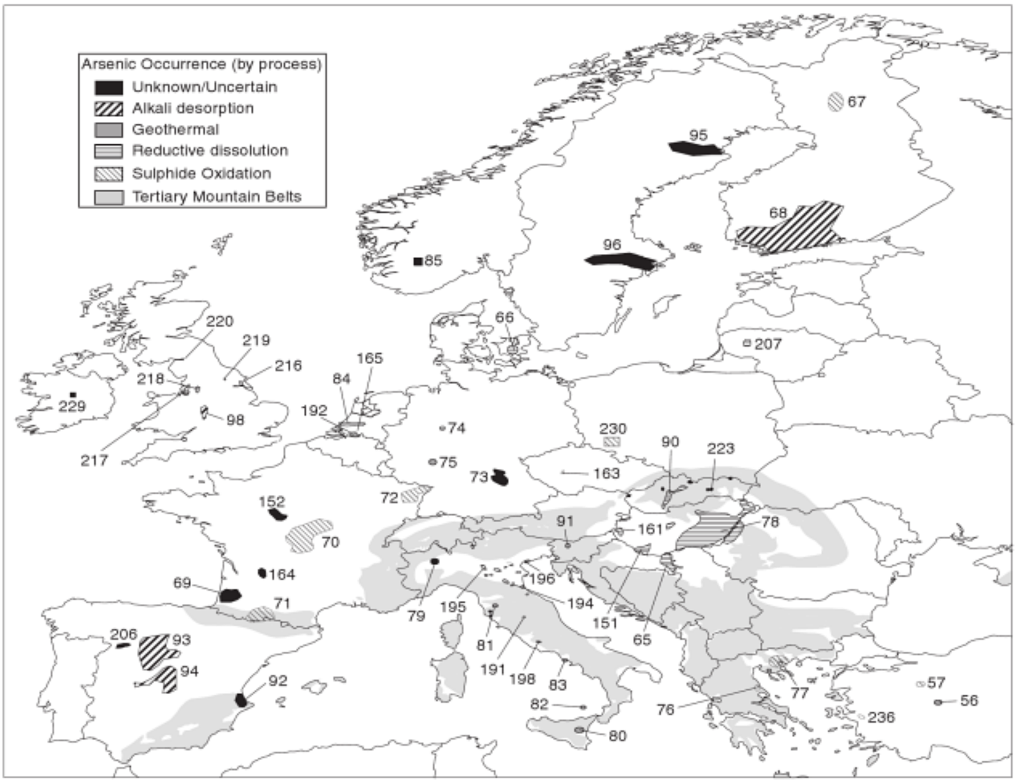


Figure 5: Arsenic contaminated areas in Europe and the underlying process (Ravenscroft et al. 2009)

Wells in northern Sweden have been found to contain arsenic, and samples were taken (in Västerbotten, number 95 in Figure 5) to identify the concentration of arsenic in the area (Jacks et al. 2010; Normannsen 2010). Finland also have areas with elevated arsenic concentrations. From Figure 5, arsenic is also found in Germany, Czech Republic, Romania, Lithuania, UK and great parts of the southern Europe.

The most affected area is the Great Hungarian Plain. Arsenic from the alluvial sediments (from the Quaternary sediments) is present in Hungarian groundwater in the range of 0.06-4.00 mg/L. A few thousands people are affected (Mandal & Suzuki 2002)

North America

USA and Mexico is the most extensively contaminated region of the world in terms of geographical extent, though not of exposed population. In North America, utilities provide arsenic-free water to the majority. The arsenic contamination is a result of all the different geochemical mobilization mechanisms (Ravenscroft et al. 2009).

The continents on the Southern hemisphere

On the Chaco-Pampean plains in Argentina, arsenic is found in groundwater caused by deposits of volcanic loess. The coastal plains of Chile have arsenic contamination caused by geothermal groundwater seeping into Andean rivers. Africa is more or less free of elevated arsenic levels. Australia has minor occurrences in coastal basins, and alluvial and geothermal arsenic is more widespread in New Zealand. These occurrences have not resulted in significant human impact (Ravenscroft et al. 2009)

2.5.2 Industrial Contamination

Mineralized areas are potentially at risk of groundwater contamination of arsenic if mining occurs. The affected area is of local extent up to a few kilometres around the mineralized zone. Water used in mining is potentially arsenic rich and can contaminate drinking water sources downstream the mining area. Run-off from waste piles can contaminate waters with both arsenic and other metals. Lead mining and smelters, gold mining and copper smelters have contaminated either water or air in respectively Toronto (Canada) and Lavrion (Greece), Obuasi (Ghana) and Montana (USA) (Ahmed & Talbi 2005b; Mandal & Suzuki 2002).

Geothermal areas may also have an increased arsenic concentration in ground water. This is also a local occurrence (Ahmed & Talbi 2005b). Even though it is a natural cause of arsenic contamination, it can be worsened by human activity. An example on this occurrence happened on a geothermal powerplant on Mt. Apo, Mindanao Island in the Philippines. The powerplant caused arsenic contamination downstream the powerplant and concentrations up to 0.1 mg/L were measured (Mandal & Suzuki 2002).

Burning of arsenic rich coal has caused severe chronic health problems in the Guizhou province in China. The same was reported from Czechoslovakia in the 1970’s (Ahmed & Talbi 2005b; Mandal & Suzuki 2002).

Industrial effluents, use of pesticides and insecticides may also contaminate air and water. 80 % of the arsenic was used for agricultural purposes in the 1970’s such as pesticides, herbicides, insecticides, wood preservatives, desiccants and feed additives. The use of arsenic in agriculture is declining, banned and phased out as among other wood preservative and pesticide in some countries (IARC 2012; Shih 2005).

In Japan, different causes have lead to arsenic contamination. A factory producing arsenic sulfide contaminated wells in Nakajo, Japan. The concentration was 0.025-4 mg/L. People in Japan have also been exposed of arsenic from manufacturing of insecticides and from run-off from a dye factory. Lead arsenate was also used in Australia as a pesticide and has caused chronic poisoning. Many other countries have contaminated water caused by industrial effluents (Mandal & Suzuki 2002; Shih 2005).

2.6 Available Technologies for Arsenic Removal in Conventional Water Treatment

Several different methods are used in large conventional treatment plants to remove arsenic. The most commonly used are different membrane techniques, oxidation, coagulation and precipitation, lime treatment and adsorption onto sorptive media (Ahmed 2001).

These different technologies are explained further in this chapter, while different inexpensive methods designed for households are described in chapter 2.7.

2.6.1 Oxidation

Oxidation will not remove arsenic. Most treatment can easily remove pentavalent form of arsenic, and oxidation can be used as a pre-treatment to oxidize arsenite (As(III)) to arsenate (As(V)) (Ahmed & Talbi 2005a; Ahmed 2001).

Aeration is an inexpensive and time-consuming method to oxidize, but chemicals like permanganate and chlorine can make the process more rapid. Oxygen, ozone, permanganate, hydrogen peroxide and hypochlorite are chemicals that can be used to oxidize arsenite faster and more effectively (Ahmed 2001).

Oxidation of As(III) by dissolved oxygen and mixing of water does not occur in a great matter (Lowry & Lowry 2002). Oxygenation of As(III) is slowest at slightly acidic conditions. This is utilized; water samples are often acidified to about pH 5 to preserve the sample. Oxidation by dissolved oxygen is a slow reaction. Eary and Schramke (1990), cited by Smedley and Kinniburgh (2001), found the half-life (the time it takes for the concentration to be reduced by 50 %) for As(III) in natural waters to 1-3 years without any oxide particles. Manganese oxides can reduce the half-life to 10-20 minutes. This is therefore used as an advantage in removal of

As(III) from drinking water. Oxidation of arsenite can also be bacterially catalyzed (Lowry & Lowry 2002; Smedley & Kinniburgh 2001).

2.6.2 Coagulation

Coagulation and flocculation are among the most used methods for arsenic removal. Ferric- and aluminium salts can be used. The process removes arsenic through three steps: formation of insoluble compounds (precipitation), soluble arsenic species are incorporated into a growing metal hydroxide phase (co-precipitation) and the electrostatic bonds formed between soluble arsenic and insoluble metal hydroxide (adsorption) (Ahmed & Talbi 2005a).

The salts are added to the water and rapidly mixed and ferric- and aluminium hydroxide flocks are formed. After a few minutes of gently stirring, larger and more easily settable flocks are created. Negative particles will attach to the flocks by electrostatic bonds. Arsenic is adsorbed onto the flocks in their pentavalent form. As(III) occurs in non-ionized form and will not be significantly removed. To ensure effective arsenic removal it is recommended to oxidize arsenic species to As(V). The flocks can be removed by sedimentation or filtration.

Coagulation with metal salts and lime followed by filtration is well documented and effective for arsenic removal and reducing turbidity, colour and odour and significantly improves the water quality. Ferric salts are found to be more effective than aluminium on a weight basis and over a wider pH range (Ahmed 2001).

2.6.3 Adsorption

Arsenic can be adsorbed if the arsenic contaminated water is filtrated through a sorptive filtration media. Arsenic and other impurities are adsorbed on the surface of the filter media grains. As with other treatment methods, arsenate is more easily removed than arsenite. Hence, the efficiency of arsenic removal is dependent on pre-treatment to oxidize arsenite to arsenate.

Activated alumina, activated carbon, iron- and manganese coated sand, silica oxide and many other natural and synthetic medias have been reported to remove arsenic from water. The different medias have different properties and efficiencies. Sorptive materials can be cost-inefficient, however some indigenous materials may be suitable for adsorption. After some time, the filter will be saturated and will not remove arsenic any longer. Some filter materials can be regenerated, but rarely to the same efficiency (Ahmed & Talbi 2005a; Ahmed 2001).

Ion exchange

Ion exchange is normally used for removal of specific cations or anions in water. It utilizes adsorption and is similar to a sorptive medium, however the medium is a synthetic material with a more defined capacity. The resin is a charged material and will exchange one ion to adsorb the unwanted ion in the water passing through. The ion exchange resin act as a chemical sponge and is effective for removal of contaminants from water (Ahmed 2001; Dow n.d.-b).

A type of resin specially intended for removal of As(V) is a strong base anion loaded with chloride:

$$2\,R_zCl^- + HAsO_4{}^{2-} \leftrightarrow (R_z)_2HAsO_4 + 2Cl^-,$$

R_z is the anionic resin.

As the impure water flows through, the resin will adsorb the unwanted ion and the product water will contain extra chlorine. As with adsorption, the resin will after extensive use be saturated or exhausted. The resin can then be regenerated using NaCl:

$$(R_z)_2HAsO_4 + 2NaCl \leftrightarrow 2\,R_zCl^- + HAsO_4{}^{2-} + 2Na^+$$

The resin will restrain all ions similar to arsenic, and the efficiency is dependent on other ions present in the water, sulfate and nitrate will be exchanged before arsenic. Compared to adsorption, ion exchange is less dependent on pH of water.

In order to remove as much arsenic as possible, As(III) has to be oxidized to As(V) to improve the efficiency. However, resins can be very sensitive to oxidants (Ahmed 2001).

Ion exchange can also be used in analysis. The efficiency is significantly better for arsenate than for arsenite, and this will be exploited in the laboratory work to distinguish between the two forms of arsenic as described later in section 4.1.

2.6.4 Reverse Osmosis and Other Membrane Technologies

Figoli et al. (2010) identifies membrane technology as a promising arsenic removal technology. Membranes are sold with different pore sizes, and are named after the pore sizes. Microfiltration has pore sizes between 100-1000 nm, ultra filtration between 10-100 nm, and nano filtration between 1-10 nm. If the membrane has pore sizes that are less than 1 nm, the membrane is a reverse osmosis membrane.

For removal of arsenic, the membrane has to have small enough pore sizes. Nano filtration, reverse osmosis and electro dialysis are therefore capable of arsenic removal. The membranes allow water to pass through and retain the impurities (Ahmed 2001).

RO is based on osmosis. If a membrane separates two solutions with different amounts of dissolved chemicals, pure water will pass through the membrane from the dilute to the more concentrated solution because of pressure differences called the osmotic pressure. In reverse osmosis, applied pressure to the most concentrated solution forces pure water to move across the membrane to the dilute (Dvorak & Skipton 2008).

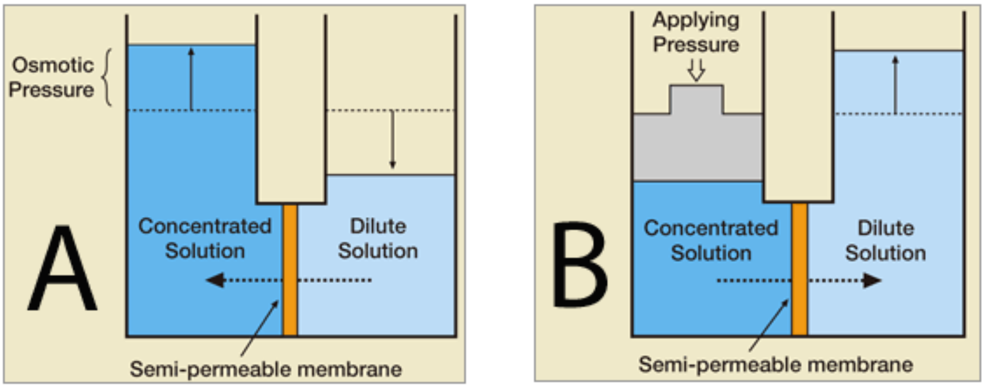


Figure 6: Picture A show the principle of osmosis, B show the principle of reverse osmosis (Nitto Denko n.d.)

Membranes usually have high energy costs because the membrane uses a lot of electricity, especially RO where pressure is applied to force the purification method. For some smaller RO units, the use of solar power or other renewable energy sources will cut the energy costs (Seibert et al. 2004).

Arsenic removal by membrane filtration is not as sensitive to pH as coagulation and sorptive medias. Besides, almost everything in a RO unit is automatically controlled; the unit can be managed without any advanced skills. There is no need for technologists or qualified people, and an RO unit can be used in a conventional treatment plant or in a small community.

However, water rejection may be an issue in water scarce regions. The US Environmental Protection Agency (EPA) has reported that nano filtration and reverse osmosis are capable of reducing the arsenic concentration with respectively 90 % and 95 % under ideal pressure (Ahmed & Talbi 2005a).

The different removal technologies have their advantages and disadvantages. The different conventional treatment processes are compared by Ahmed and Talbi (2005a) in Table 2.

Table 2: Comparison of the main arsenic removal technologies (Ahmed & Talbi 2005a)

Technology	Advantages	Disadvantages
Oxidation (air or chemical) and sedimentation	- Relatively simple and low cost - Slow oxidation process (air) or rapid process (chemical) - Oxidizes other impurities and kills microbes	- Remove only some of the arsenic - Used as pre-treatment
Coagulation and filtration (alum, iron)	- Relatively low capital cost - Relatively simple in operation - Common chemicals available	- Not ideal for anion rich water treatment - Produces toxic sludge - Low removal of As(III) - Pre-oxidation is required - Efficiencies may be inadequate to meet strict standards
Sorption techniques	- Relatively well known and commercially available - Well-defined technique - Many possibilities	- Not ideal for anion rich water treatment - Produces arsenic-rich liquid and solid wastes - Regeneration is required - High-tech operation and maintenance - Relatively high cost
Membrane techniques	- Well-defined and high removal rates - No toxic waste produced - Capable of removing other contaminants	- High-tech operational and maintenance - High capital- and running costs - Arsenic-rich reject water is produced

2.7 Inexpensive Arsenic Removal

Several technologies have been developed for a household to remove arsenic from water. They are developed and based on one or more of the four conventional treatment processes described in 2.6. Nine different household treatment options are described briefly to inform about available options in arsenic contaminated areas.

2.7.1 Technologies Based on Coagulation

The **bucket treatment unit** (**Error! Reference source not found.**) contains of two buckets, one placed above the other. The water is mixed with chemicals in the upper red buckets and flocculated by gently stirring for about 90 seconds. After settling, the water from the red bucket flows to the lower green bucket. Water is collected through a sand filter in the green bucket. The bucket treatment is found very effective in removing arsenic, as well as iron, manganese, phosphate and silica (Ahmed & Talbi 2005a).

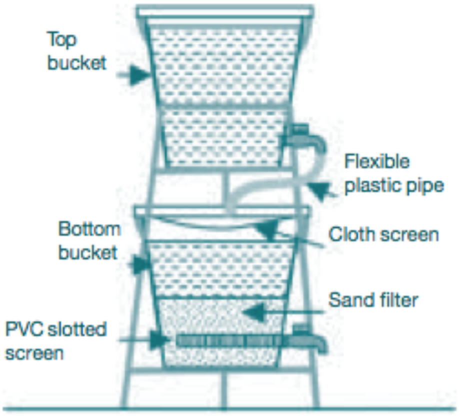


Figure 7: Double Bucket household arsenic treatment unit (Ahmed & Talbi 2005a)

Stevens Institute technology (Figure 8) also contains of two buckets. Chemicals, iron coagulant and hypochlorite, are mixed in the first bucket. The second bucket has an inner bucket with slits to keep the filter sand bed in place. A plastic pipe below the sand filter delivers the cleaned water (Ahmed & Talbi 2005a).

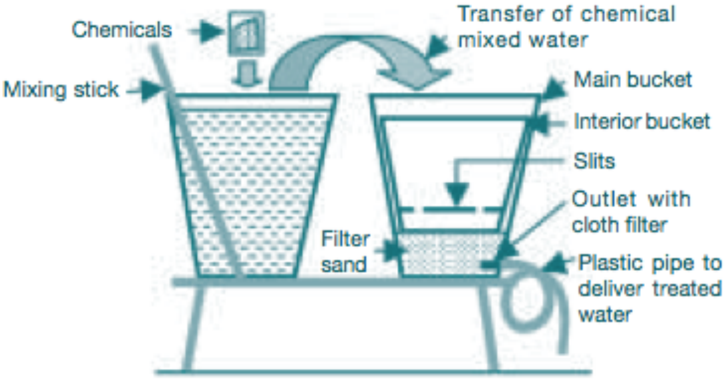


Figure 8: Bucket treatment by the Stevens Institute technology (Ahmed & Talbi 2005a)

The **fill and draw treatment** unit (Figure 9) is a treatment unit for a community. A tank is filled with water and oxidant and coagulant. After mixing, the tank is left overnight for sedimentation. The water is tapped a few centimetres from the bottom of the tank and into a sand bed (Ahmed & Talbi 2005a).

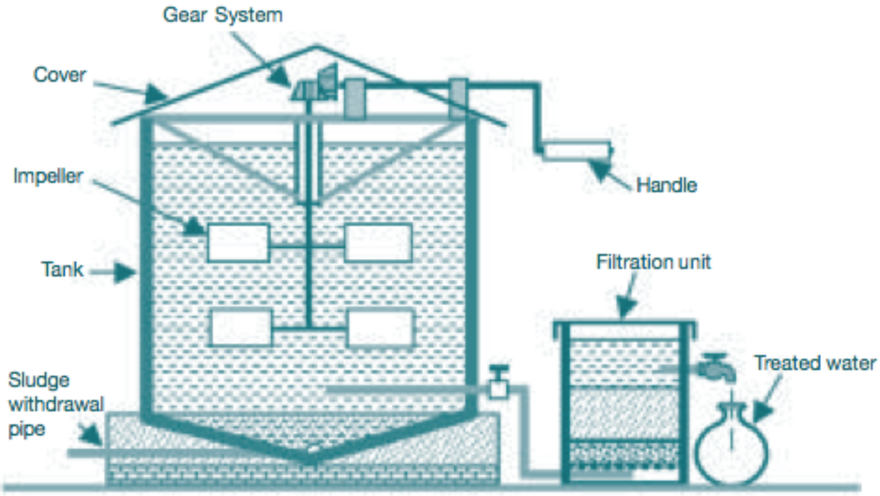


Figure 9: Fill and draw treatment (Ahmed & Talbi 2005a)

The **tubewell-attached arsenic treatment unit** (Figure 10) uses the principle of removal by alum coagulation, sedimentation and filtration. The treatment unit is compact; mixing, flocculation, sedimentation and up-flow filtration is built as one unit. This treatment has a removal rate of about 90 % (Ahmed & Talbi 2005a).

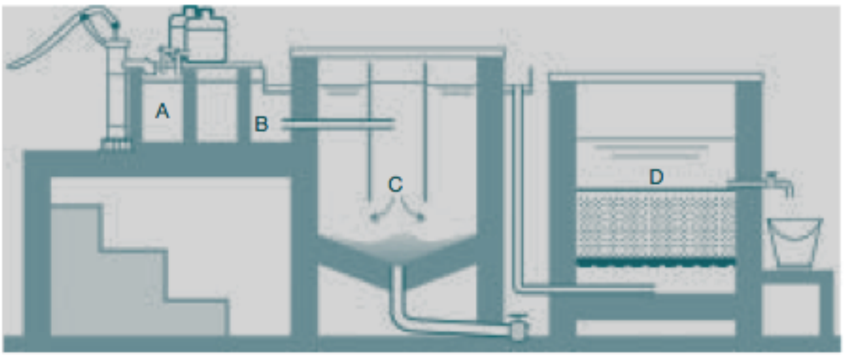


Figure 10: Treatment unit attached to a tubewell (Ahmed & Talbi 2005a)
A - mixing, B – flocculation, C – sedimentation and D – up flow filtration.

Another method is to dip a cloth wrapped alum ($K_2SO_4*Al_2(SO_4)_3*24H_2O$) in the water for a few seconds, then let the water stand overnight. This will precipitate the arsenic, and the water can be filtrated through a cloth and about 70-80 % of the arsenic is removed (Jackson & Jackson 2000).

2.7.2 Technologies Based on Adsorption

The most commonly used sorptive medias used in small treatment plants are activated alumina, metallic iron, granulated ferric oxide and hydroxide, iron-coated sand/brick dust, cerium oxide and ion exchange media (Ahmed & Talbi 2005a). **Activated alumina** is most efficient for arsenic removal at the pH range from 5.5 to 6.0. At this pH range, the surface is positively charged and the negatively charged impurities can be adsorbed. When the activated alumina is saturated, the media can be regenerated. Each regeneration will lead to capacity loss in the media and it has to be replaced after 3-4 regenerations (Ahmed & Talbi 2005a).

The **Alcan enhanced activated alumina** arrangement (Figure 11) is attached to a tubewell. No chemicals are added, and it is simple and robust. Other ions may compete with arsenic for the active sites, and the arsenic removal capacity may be reduced. 3600 liters of arsenic-reduced water can be produced per day for 100 families (Ahmed & Talbi 2005a).

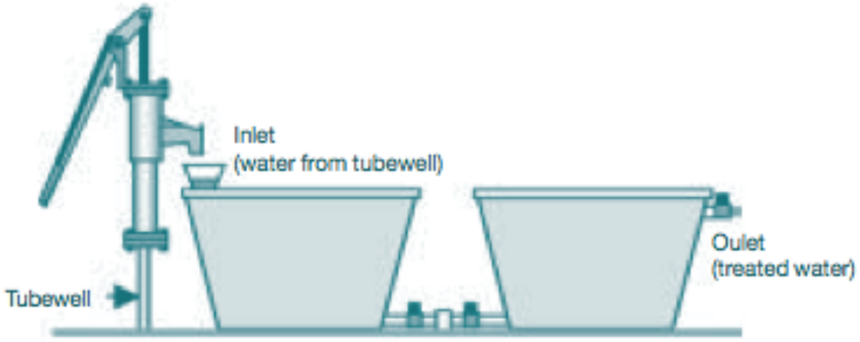


Figure 11: Alcan enhanced activated alumina unit (Ahmed & Talbi 2005a)

There are various designs which uses sorptive media for arsenic removal. Another activated alumina design is the three kalshi filtration system (Figure 12). Kalshi is a burned clay pitcher, widely used in Bangladesh. The first kalshi is filled with iron fillings and sand. Brick chips are placed around the holes to prevent the sand from leaking out. The second kalshi contains of sand, charcoal and brick ships, and the bottom kalshi collects the filtered water. Up to 97 % of arsenic can be removed. Disadvantages are clogging (Ahmed & Talbi 2005a).

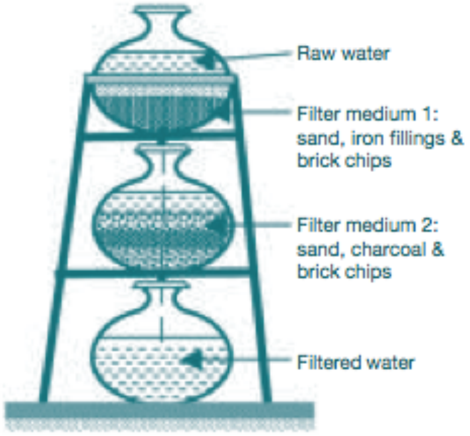
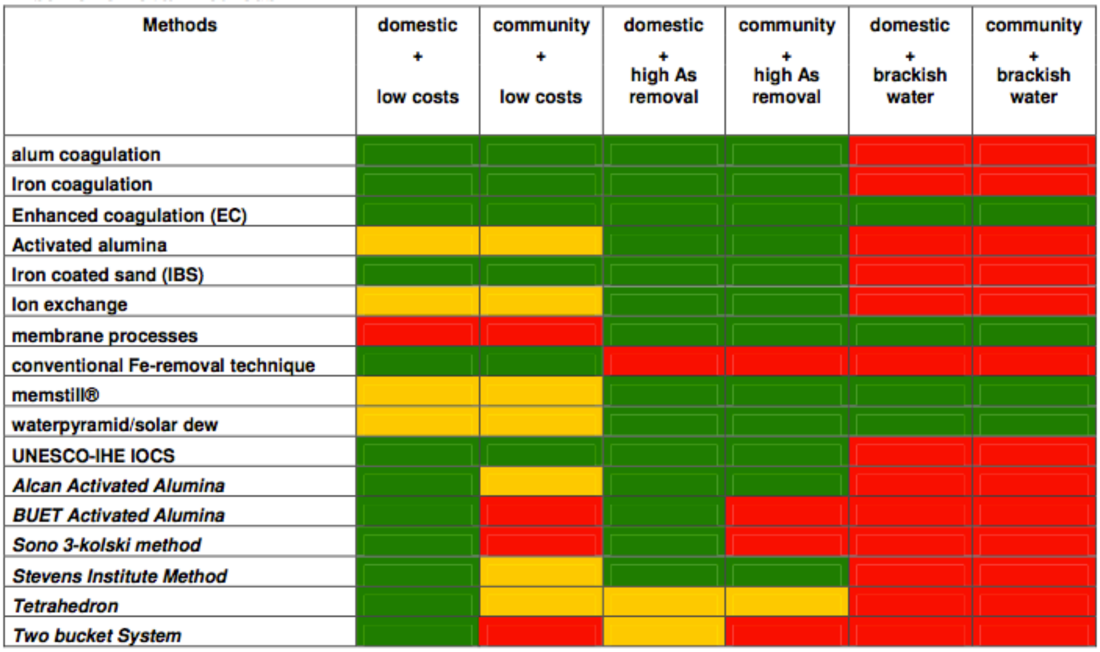


Figure 12: Three Kalshi Filter (Ahmed & Talbi 2005a)

Naturally iron-rich soil can adsorb arsenic and may oxidize As(III) to As(V). Several plants have been installed in Bangladesh. The filter bed can lower the arsenic concentration to half or up to one fifth of the original concentration if the filter is well operated (Ahmed & Talbi 2005a). A similar method is to filter the water through a tube filled with sand and iron fillings attached to the well outlet. The water is dosed with barium sulfate and filtered, and insoluble arsenopyrite (FeAsS) is formed and caught in the filter bed (Jackson & Jackson 2000).

Feenstra et al. (2007) described a number of different removal methods, both conventional and household point-of-use. The report for the International Groundwater Resources Assessment Centre (IGRAC) summarized some of the most used technologies (Figure 13).



Methods	**domestic + low costs**	**community + low costs**	**domestic + high As removal**	**community + high As removal**	**domestic + brackish water**	**community + brackish water**
alum coagulation	🟩	🟩	🟩	🟩	🟥	🟥
Iron coagulation	🟩	🟩	🟩	🟩	🟥	🟥
Enhanced coagulation (EC)	🟩	🟩	🟩	🟩	🟩	🟩
Activated alumina	🟨	🟨	🟩	🟩	🟥	🟥
Iron coated sand (IBS)	🟩	🟩	🟩	🟩	🟥	🟥
Ion exchange	🟨	🟨	🟩	🟩	🟥	🟥
membrane processes	🟥	🟥	🟩	🟩	🟩	🟩
conventional Fe-removal technique	🟩	🟩	🟥	🟥	🟥	🟥
memstill®	🟨	🟨	🟩	🟩	🟩	🟩
waterpyramid/solar dew	🟨	🟨	🟩	🟩	🟩	🟩
UNESCO-IHE IOCS	🟩	🟩	🟩	🟩	🟥	🟥
Alcan Activated Alumina	🟩	🟨	🟩	🟩	🟥	🟥
BUET Activated Alumina	🟩	🟥	🟩	🟥	🟥	🟥
Sono 3-kolski method	🟩	🟥	🟩	🟥	🟥	🟥
Stevens Institute Method	🟩	🟨	🟩	🟩	🟥	🟥
Tetrahedron	🟩	🟨	🟨	🟨	🟥	🟥
Two bucket System	🟩	🟥	🟨	🟥	🟥	🟥

Figure 13: Arsenic removal methods (Feenstra et al. 2007)

The green colour indicate a very suitable method, yellow means an average suitability and red indicates that the method is unattractive or not applicable (Feenstra et al. 2007). Memstill, Waterpyramid, UNESCO-IHE IOCS, BUET Activated Alumina and tetrahedron mentioned in Figure 13 are not described in this paper.

Figure 13 shows that membrane processes are very suitable in removal of arsenic and brackish water. Membrane processes can be quite expensive to purchase, though a small RO unit like the one used in the laboratory is intended for a small community (A-Aqua n.d.).

2.8 Analytical Methods for Arsenic in Water

The analytical methods for arsenic depend on the medium. Different methods for detecting arsenic in water is available, the majority focus on the detection of total arsenic. Field test kits are available for detecting the arsenic concentrations in areas without available laboratory based analytical methods. However, the samples in this work was analysed at a laboratory and different laboratory based analytical methods are mentioned in Table 3 (IARC 2004).

Table 3: Different analytical methods for arsenic in water (IARC 2004)

Methodology	**Detection**	**Detection limit**
Colorimetric/spectrophotometric methods	Total arsenic	~ 40 µg/L
Inductively coupled plasma – atomic emission spectrometry (ICP – AES)	Total arsenic	~ 30 µg/L
Inductively coupled plasma – mass spectrometry (ICP – MS)	Total arsenic	0.1 µg/L
High resolution (HR)-ICP-MS	Total arsenic	0.01 µg/L
Graphite furnace – atomic absorption spectrometry (GF – AAS)	Total arsenic	0.025 µg/L
Hydride generation – atomic absorption spectrometry (HG-AAS)	Total arsenic and arsenic speciation	0.6 – 6 µg/L
Hydride generation quarts furnace – atomic absorption spectrometry (HG-QF-AAS)	Total arsenic and arsenic speciation	0.003 – 0.015 µg/L
HPLC or solid phase cartridge separation combined with hydride generation-atomic fluorescence spectrometry (HPLC-HG-AFS)	Arsenic Speciation	0.05 – 0.8 µg/L
HPLC-ICP-MS	Total arsenic	0.01 µg/L

In this research, the water samples were tested using inductively coupled plasma mass spectrometry (ICP-MS) at the laboratory of IPM. The laboratory has the equipment to detect both species of arsenic in one sample. In order to do so, a high-performance liquid chromatography (HPLC) is connected to the ICP-MS. This is a chromatographic method and the ICP-MS is the detector. Even though the laboratory is able to detect the two species, it is an expensive method with a price tag of about several thousands NOK for each sample (Jensen 2012).

Since the economy of the research could not afford detection of the two different species of arsenic in the experiments, ion exchange was used to distinguish the two oxidation states. The ion exchange resin is designed to adsorb As(V), and instead of the direct analysis of As(V) the concentration of As(V) is found by subtracting the concentration of total arsenic of the sample after IE from the concentration before IE.

2.9 Legislation and Recommendations

"All people, whatever their stage of development and their social and economic conditions, have the right to have access to an adequate supply of safe drinking water" (WHO 2011a)

The phrase is one of the primary goals of the WHO. To achieve such goals, WHO proposes regulations and recommendations regarding health matters (WHO 2011a). WHO changed the guideline for arsenic level in drinking water in 1993 from 50 µg/L to 10 µg/L. Some countries have reduced the limit of arsenic in drinking water, the European Union standard arsenic level is set to 10 µg/L; and the same permissible level is used by the U.S. Environmental Protection Agency. Australia lowered the limit to 7 µg/L, France to 15 µg/L. Other countries have not, as for example Mexico, Bangladesh and Vietnam, who still have a limit of 50 µg/L arsenic (Choong et al. 2007; EPA 2001).

The WHO has a general rule that no substance may have a higher lifetime risk of more than 1 in 100 000. In regard to purely health effects, the lifetime risks found by EPA and the US Natural Resources Defence Council (NRDC) displayed in Table 1, shows that the WHO guideline of 10 µg/L is not satisfactory (van Halem et al. 2008).

3 Previous Studies

Studies of the efficiency of RO in removal of arsenic have been conducted for decades. Different membranes have been used in experiments; some of these membranes are mentioned with an abbreviation and the membrane material in a parenthesis without further explanation.

Gholami et al. (2006) added sodium arsenate in water samples in the laboratory and tested the removal of arsenic using reverse osmosis. The system performance, the effect of arsenic concentration, pressure, pH, and temperature was tested. The optimal condition found in the results were pressure 1 310 kPa – 1 448 kPa, concentration 0.2–0.5 mg/L, temperature 25–30° C and pH = 6–8 (Gholami et al. 2006).

The article written by Shih (2005) assess several studies of arsenic removal using RO. The first tests conducted by EPA during the 1980's used a cellulose-acetate RO membrane. The experiments concluded that the As(V) removal was over 90 %, but the As(III) removal efficiency was less than 70 %.

A TFC-ULP RO membrane (polyamide, from Koch Membrane System) was tested with concentration of 60 µg/L arsenic, and the concentration was reduced to 0.9 µg/L; i.e. removal efficiency of arsenic around 99 %.

A study on the correlation of pH and removal efficiency of arsenic used two different RO membranes, ES-10 (polyamide) and NTR- 729HF (polyvinyl alcohol) (both manufactured by the Nitto Electric Industrial Co., Japan). The results gave a removal efficiency of As(III) lower than As(V) in the pH range 3-10. The ES-10 membrane had removal efficiency of As(V) over 95 % on the same pH range. As(III) was removed by 75 % for pH 3, 5 and 7 and increased to around 90 % at the pH 10. The removal efficiency for As(V) using NTR-729HF membrane was around 80 % at pH 3, and around 95 % at pH 5, 7 and 10. The same membrane had removal efficiency for As(III) of around 20 % at pH 3, 5 and 7.

A study performed by American Water Works Association Research Foundation in 1998 showed that the removal efficiency of As(V) can reach 96 % and for As(III) around 5% using a flat sheet of a single element of DK2540F (from DESAL) RO membrane. Several other RO tests were performed. The removal efficiency of As(V) from groundwater with low dissolved organic carbon (DOC) was more than 90 %. With high DOC, the removal efficiency was more than 80 % using TFCL-HR membrane (polyamide). Tests from four different membranes gave removal efficiencies of As(V) by 96 % and As(III) between 60 % and 85 % depending on the membrane (Shih 2005).

Thin-film composite type membranes have been found to have better removal efficiencies than cellulose-acetate type membranes. The first mentioned has a higher permeated flow rate and hence needs lower driven-pressure than the latter (Shih 2005).

Two previous studies at UMB are done on arsenic removal using RO. Both used the same system provided by A-Aqua as the one used in this project. Ortiz (2012) found a total removal rate of 89-91 % of As(III) when the water was double filtrated at a flow rate of 100 L/h. At flow rate of 350 L/h, the total removal rate was 97 %. During one filtration, As(III) was removed by 65-88 %.

Experiments on arsenic removal conducted by Ahmad (2012) gave 95-99 % removal rate of As(V) and the removal efficiency of As(III) was between 54 % and 80 % when running the RO unit at pH 6 and 8. When double filtration, the removal efficiency was 76 % for As(III) at pH 6 and 99 % at pH 10. He also found water flow rate of 200 L/h more efficient than 50 L/h, and higher removal efficiency at pH 8 than pH 6, and As(III) removal efficiency is more dependent on pH that As(V).

Ortiz (2012) found the removal rate better at high filtration flow. The RO unit provided by A-Aqua allowed improved membrane performance by changing either the filtrate flow or the concentrate flow (reject water). The pump in front of the membrane pumped 1 m^3/h regardless of other conditions. A magnetic valve opened if there was too much pressure across the membrane. When the water flow was minimal, less water was filtrated and increased the pressure across the membrane since the pump continued to pump the same amount of water.

4 Method and Material

Some similar experiments have previously been conducted at UMB using the same RO-unit, but with different membranes. The experiments started in February 2013 and were completed by the end of March. Experiments were repeated in mid April to control and confirm previous tests.

4.1 Description of Ion Exchange

The experiments are designed to check the removal efficiency of As(III) in the RO unit, and if some As(III) gets oxidized to As(V). The analysis in a mass spectrometer can give us the concentration of total arsenic, and in order to distinguish between the two, As(III) and As(V), a sample of the solution passed through an ion exchange (IE) resin designed to adsorb all the As(V). When a sample was taken and analysed before and after the IE, the difference was the concentration of As(V).

The ion exchange resin used was Dowex 21K XLT Resin from Dow, a high efficiency, uniform particle size, strong base anion exchange resin. The matrix is of styrene-DVB gel and charged with chloride (Dow n.d.-a). Ion exchange is a reversible interchange of ions between a resin and a liquid. When arsenic contaminated water run through the resin, As(V) will be adsorbed to the resin and release chloride. The water after an ion exchange have higher chloride levels and the arsenic present is As(III).

The resin was put in a glass cylinder (depicted in Figure 14) and between 250 ml and 500 ml of the test water was run through the resin for each test to differentiate between As(III) and As(V) since the analysis conducted at IPM only measure total arsenic. 250-500 ml test water was run through the resin. The first two thirds of the water ran through the resin to rinse and replace the water in the pores. The last one third of the water was collected after the IE and sampled.

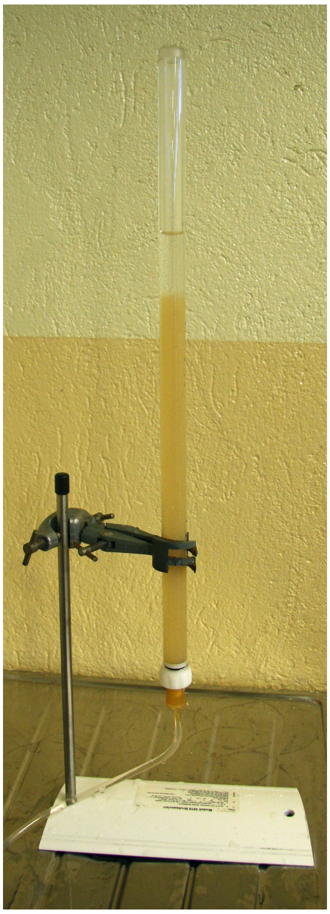


Figure 14: The glass cylinder filled with ion exchange resin (Photo: Marie Fauskrud)

4.2 Description of Reverse Osmosis Plant

4.2.1 History

The treatment unit was provided by A-Aqua, formerly known as Scan-Water, a part of the Malthe Winje Group. It is an international company that provides cost efficient drinking water systems and sanitation solutions. Since 1985 A-Aqua has supplied UN-agencies, Red Cross and NGO's. Different water treatment solutions from A-Aqua have been installed for villages in rural areas of Gabon, India and Uganda. (A-Aqua n.d.).

4.2.2 The RO-unit

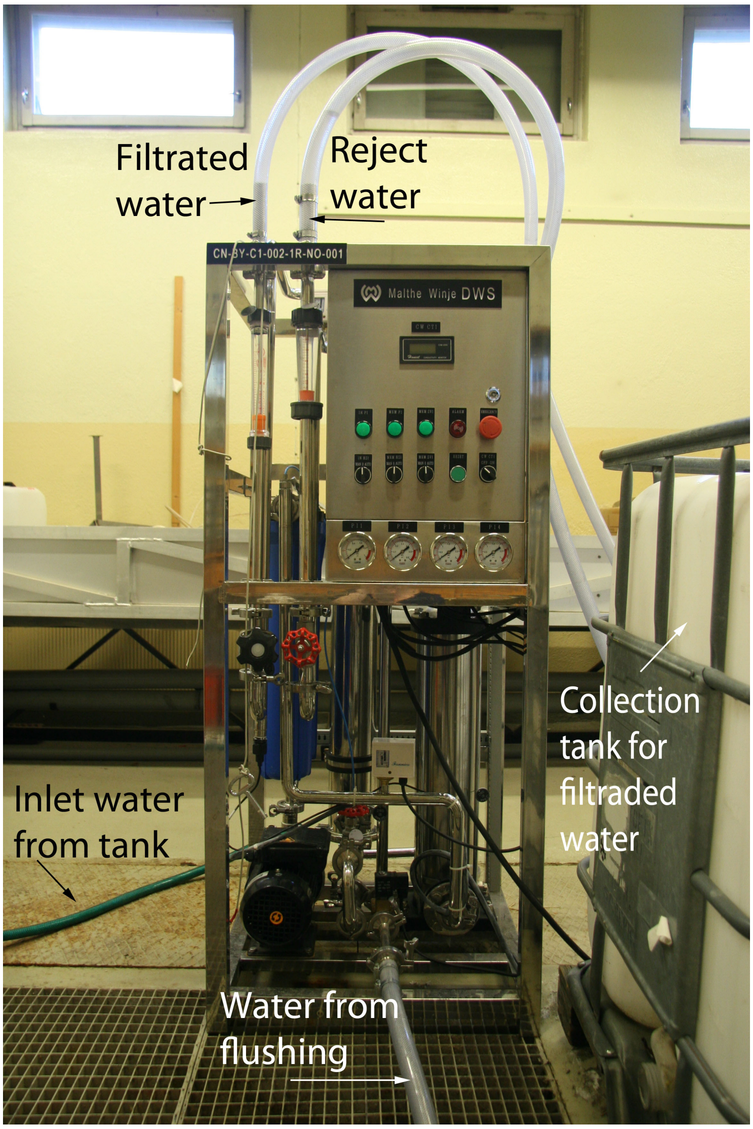


Figure 15: The RO unit installed at the laboratory (Photo: Marie Fauskrud)

The RO-unit is pictured in Figure 15 and it’s piping and instrumental design is shown in Figure 16.

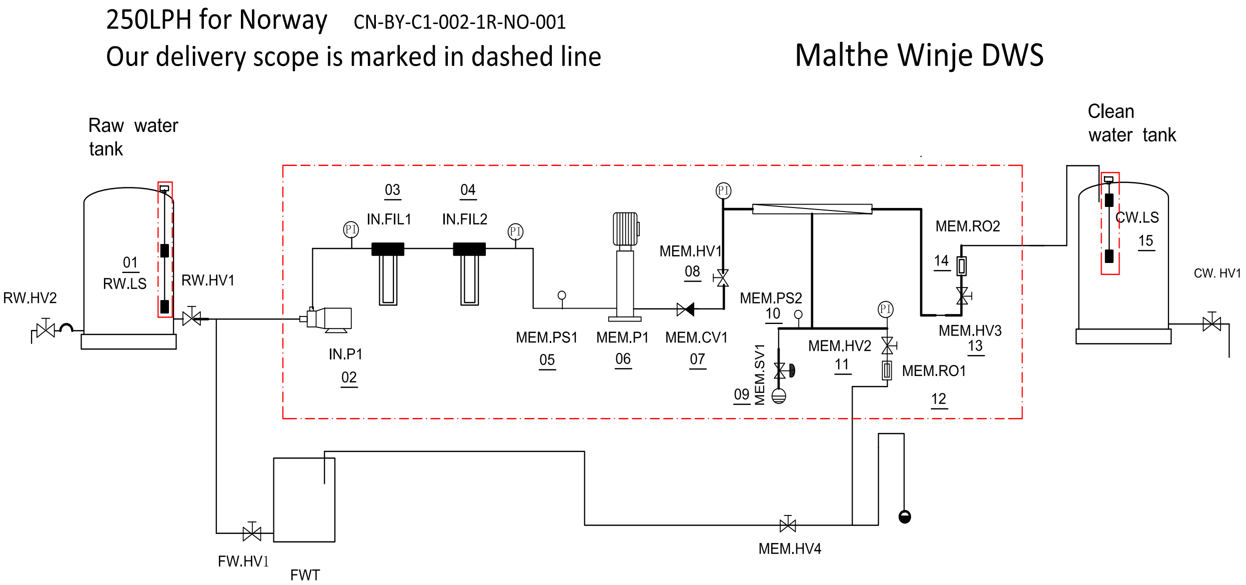


Figure 16: Instrumental design of the RO unit

In the laboratory, the raw water tank was a 1m^3 tank filled with arsenic spiked tap water. The cleaned water was collected in another tank, and used as the raw water tank in the second filtration process. The membrane used for the experiments was reverse osmosis element from FilmTec similar to the membrane in Figure 17.

Figure 17: RO membrane (Photo: from www.isopurewater.com)

The raw water was pumped (IN.P1) and went through two coarse filters (IN.FIL1 and IN.FIL2, the blue cylinders in Figure 15) as a pre treatment. Then another pump (MEM.P1) delivered high-pressured water to the membrane (MEM.RO2). The filtrate, the water cleaned by the membrane, was delivered to the clean water tank, which is depicted in Figure 15. Some water would go directly through the cylinder without being filtered, and this water would be slightly more concentrated with impurities held back by the membrane. This rejected water went to a waste tank.

Once in a while, the unit was flushed. The flushing water tank (FWT), in this situation the flushing water was taken from the raw water tank, delivered water that went the opposite direction as in the arsenic removal process.

4.3 Experimental Design

The reverse osmosis system was installed by A-Aqua at the Department of Mathematical Sciences and Technology, and the water samples were tested at the laboratory at IPM using inductively coupled plasma mass spectrometry (ICP-MS).

The arsenic solutions used were premixed at IPM to a concentration of 4 g/L. The solutions of arsenic (III) and arsenic (V) were mixed with tap water in the laboratory for each experiment in different concentrations and at different pH levels.

It is worth mentioned that the water used for the experiments using the RO-unit was already drinkable water spiked with arsenic. Natural water sources containing arsenic probably contains other contaminants that have an effect on the membrane performance.

4.3.1 Verification of the RO-system

To produce the desired flow rate, the system may use variable amounts of inlet water. It was necessary to quantify these volumes to plan the inlet water preparations. The RO system has a built-in flow meter, which is probably not that accurate. The water flow rates were verified using 2-liter cylinders and their filling times. The system was tested with tap water with a flow rate at 100 L/h and 200 L/h and the inlet water consumption and the reject water flow rates were measured.

4.3.2 Verification of As(III) and As(V) Separation

To know whether or not to rely on the resin, a verification test of the IE resin was carried out. Different solutions of tap water and arsenic salts were mixed and run through the ion exchange resin (Table 4).

Table 4: The concentration in the different tests of the ion exchange resin

Experiment	Concentration of As(III) (µg/L)	Concentration of As(V) (µg/L)
1	50	-
2	100	-
3	-	50
4	-	100
5	50	50
6	75	25
7	25	75

About 500 ml of the solution was run through the ion exchange resin as explained in chapter 4.1. The test of the sample was sent to the lab at IPM. Both the initial solution before the separation process and a sample after the ion exchange were tested.

4.3.3 Oxidation

To check if As(III) gets oxidized easily to As(V) or not, a solution of tap water and As(III) was mixed and the solution was magnetically mixed for a time. Two samples were taken after 2 hours, and two more after 24 hours. One of the two samples was run through the ion exchange resin and sent to testing. This was done with a concentration of 100 µg/L with pH levels at 6, 7.5 and 10, and with 200 µg/L with pH levels at 6 and 8.

4.3.4 Reverse Osmosis Test

The RO-unit was used to test the removal of As(V), As(III) and a combination of As(III) and As(V). The reference test of As(V) was conducted with the test parameters shown in Table 5 to have a comparison for the double filtration experiments using As(III). The test parameters for the removal of As(III) (Table 6) and for the mixed solutions (Table 7) are similar to the reference test of As(V). The reverse osmosis experiments were conducted according to the procedure described in Appendix 1.

Reference test of As(V) (Experiment 11-14 and 24)

Table 5: Test parameters for As(V) removal

	Concentration (µg/L)	Flow rate (L/h)	pH
As(V)	50	200	6
		200	8
	200	200	6
		200	8

Test of removal of As(III) (Experiment 15-18 and 27-30)

Table 6: Test parameters for As(III) removal

Concentration of As(III) (µg/L)	Concentration of As(V) (µg/L)	Flow rate (L/h)	pH	Number of filtrations
50	-	200	6	2
			8	2
200	-	200	6	2
			8	2

The experiments of double filtrating As(III) solutions were done twice, and the mean values and standard deviations were calculated for the removal rates.

The standard deviation (σ) is found by looking at the difference between the removal rate and the mean removal rate ($\bar{x}$), using the equation:

$$\sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - \bar{x})^2}\,, where\ the\ mean\ \bar{x} = \frac{1}{N}\sum_{i=1}^{N} x_i$$

Test of a combination of As(III) and As(V) (Experiment 19-22)

Table 7: Test parameters for removal of a combination of As(III) and As(V)

Concentration of As(III) (µg/L)	Concentration of As(V) (µg/L)	Flow rate (L/h)	pH	Number of filtrations
25	25	200	6	2
			8	2
100	100	200	6	2
			8	2

Test of salts

Reverse osmosis is used for desalination of seawater, and remove a lot of ions. To drink purified water from RO may be harmful. Distilled water and water free of trace minerals and electrolytes (like sodium, potassium and chloride) is very aggressive and easily dissolves minerals. This can also happen inside the body, spilling calcium, magnesium and trace minerals to the urine. Desalination processes often add a part of seawater to the purified water before it goes out to the consumer to add the minerals

Because of this, the tests from experiment 11-16 and 18 were also tested magnesium, sodium, potassium and calcium.

4.3.5 Chemicals and Equipment Data

The arsenic solutions used were mixed at IPM to solutions with concentration of 4 g/L. They were mixed using sodium arsenate, $Na_2HAsO_4 \cdot 7\ H_2O$ (CAS number 10048-95-0 from Sigma-Aldrich), and sodium arsenite, c($NaAsO_2$) (VWR 1.06277.1000 from Merck Millipore) (Merck Millipore 2013; Sigma-Aldrich 2013).

All the water contaminated by arsenic was collected in a tank and treated with potassium sulfide, $(l*K_2S)(m*K_2S_x)(n*K_2S_2O_3)$ (VWR 1.05134.1000 from Merck Millipore) to precipitate arsenic.

The pH adjustments were done with hydrochloric acid (HCl, 32 %, number 1.00319.1011, Merck Millipore) and sodium hydroxide (NaOH pellets for analysing, number 1.06498.1000, Merck Millipore)

The pH was measured using pH meter and the oxidation was tested using three magnetically mixers. The RO-system (CN-BY-C1-002-1R-NO-001 by A-Aqua) worked under different pressure, calculated in a built in water flow rate (L/h).

The RO-membrane used in the unit was a reverse osmosis element, a FilmTec membrane, from FilmTec Corporation (model TW30-4040).

The tests were sent to IPM where the analysis was done using an inductively coupled plasma mass spectrometry (ICP-MS).

5 Results

Verification of the built in flow meter and the ion exchange resin was conducted in order to know the validation of the results. Oxidation experiments were done to know the possibility of oxidation inside the RO unit. These results will be displayed before the results from the reverse osmosis experiments. All of the tests using the RO unit used flow rate 200 L/h. The results and the figures in this chapter will be explained further in chapter 0.

5.1 Verification of the RO-system

In order to know how much water needed for the different tests, the built in flow meters in the RO-unit had to be verified. The results are shown in Table 8:

Table 8: Verification of flow meters

Read Q cleaned	Read Q reject water	Measured Q cleaned water (average)	Measured Q reject water (average)
60	1400	54.6	1 366.7
100	1270	90.0	1 212.7
200	680	191.4	618.2

5.2 Verification of the Indirect Analytical Method for As(V)

The ion exchange resin is designed to adsorb As(V) and was used as an indirect analytical method for As(V) since the IPC-MS analyses the concentration of total arsenic (Table 9).

Table 9: The results from verification of the ion exchange resin

Target concentration of total arsenic (µg/L)	$C_{total\ arsenic}$ (µg/L)	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)*	Percentage error
50 µg/L As(III)	39	42	-3	-8.04
100 µg/L As(III)	116	111	5	3.97
50 µg/L As(V)	54	0.20	54.3	0.36
100 µg/L As(V)	107	0.22	107.2	0.21
50 µg/L As(III) +50 µg/L As(V)	104	53	51	-
75 µg/L As(III) + 25 µg/L As(V)	119	78	41	-
25 µg/L As(III) + 75 µg/L As(V)	114	30	84	-

* The resin was assumed to adsorb all As(V). Therefore, the concentration of As(V) was calculated using the concentration of total arsenic and subtracting the concentration of As(III).

A simple expected margin of error was calculated by dividing the actual concentration where it was expected to be 0 by the concentration of total arsenic before the ion exchange.

IE was used as an indirect method to find the concentration of As(V) in the experiments. The resin was regenerated before experiment 19. A new resin was used as of experiment 21 and regenerated before experiment 27.

Figure 18: Colour change in resin (Photo: Marie Fauskrud)

The resin to the left in Figure 18 was used for experiment 21-30 and regenerated once; the resin to the right was new.

5.3 Oxidation

The RO experiments were designed to examine the removal of As(III) inside the unit and if whether or not As(III) was oxidized to As(V). The oxidation experiments were conducted to examine the probability of oxidation of As(III) and to identify a potential estimated oxidation ratio to use in later experiments. The results of the oxidation experiments are found in Table 10.

Table 10: Results from the oxidation test and the repeated test with 200 μg/L

Condition: 100 μg/L		$C_{As(III)}$ (μg/L)	$C_{As(V)}$ (μg/L)	Oxidation ratio (%)
pH 6.02	Initial concentration	96		
pH 6.2	After 2 hours	85	8	8.70
pH 7.2	After 24 hours	89	11	10.58
pH 7.5	Initial concentration	94		
pH 8.02	After 2 hours	88	7	7.37
pH 7.65	After 24 hours	88	10	10.31
pH 9.96	Initial concentration	96		
pH 9.8	After 2 hours	83	11	11.83
pH 7.62	After 24 hours	82	19	19.11
Condition: 200 μg/L				
pH 5.97	Initial concentration	200	10	4.76
pH 5.99	After 2 hours	190	20	9.52
pH 7.47	After 24 hours	200	10	4.76
pH 7.97	Initial sample	190	20	9.52
pH 8.01	After 2 hours	180	30	14.29
pH 7.51	After 24 hours	190	20	9.52

The results are also graphically presented in Figure 19 for the oxidation of 100 μg/L As(III) and in Figure 20 for all the oxidation experiments.

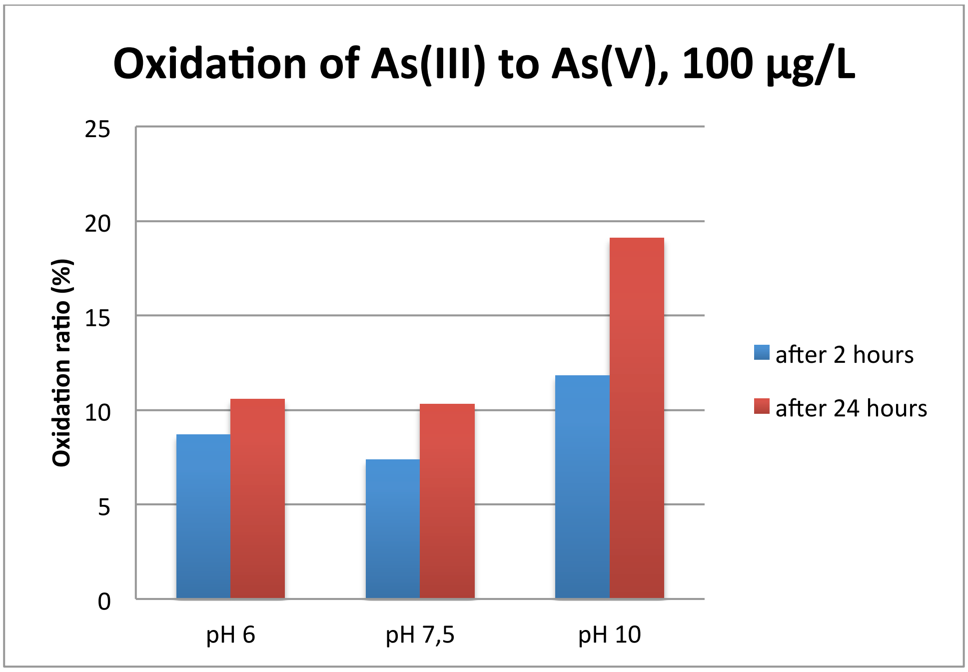


Figure 19: Chart of oxidation ratio for three different pH levels

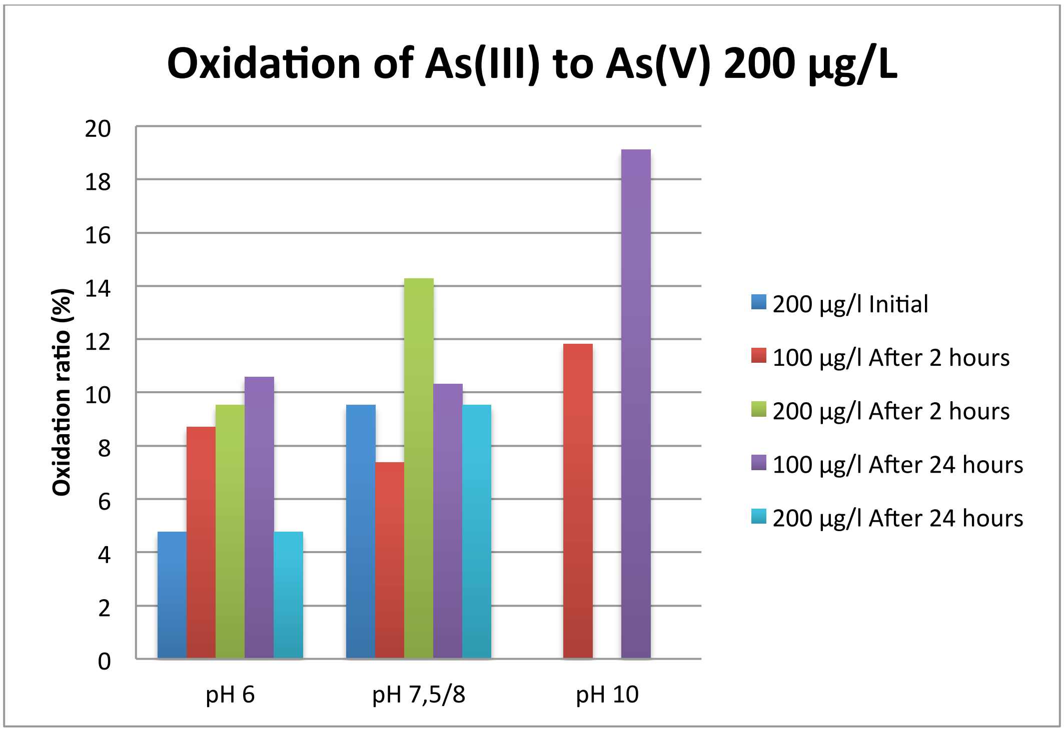


Figure 20: Oxidation of As(III) to As(V) for two different concentrations and at three different pH levels.

5.4 Proportion of As(V) in the As(III) Solution

Some of the experiments were repeated in mid April, and the initial samples of the new tests were run through the IE resin. This was not done in the first tests. These initial samples were tested for As(V) (Table 11).

Table 11: Percentage of As(V) in the As(III) solution

	$C_{\text{total As}}$ (µg/L)	$C_{\text{As(III)}}$ (µg/L)	$C_{\text{As(V)}}$ (µg/L)	Ratio As(V)/total As (%)
pH 6	57	54	3	5.26
pH 6	220	220	0	0
pH 6	210	200	10	4.76
pH 8	54	50	4	7.41
pH 8	210	190	20	9.52
pH 8	210	200	10	4.76

5.5 Reference Test Using As(V)

To have something to compare the double filtrated experiments of As(III), a reference test was conducted with removal of As(V) (Table 12).

Table 12: Results of reference test

Experiment		Concentration (µg/L)	Removal rate (%)
11	Initial sample	49	
	After 15 minutes	0.6	98.78
	After 30 minutes	0.31	99.37
	Sample from the filtrate	1.3	97.35
12	Initial sample	51	
	After 15 minutes	0.54	98.94
	After 30 minutes	0.54	98.94
	After 40 minutes	0.5	99.02
	Sample from the filtrate	1.2	97.65
13	Initial sample *	66	
	After 15 minutes	1.4	97.88
	After 30 minutes	1.2	98.18
	Sample from the filtrate	2.1	96.82

Experiment		Concentration (µg/L)	Removal rate (%)
14	Initial sample	150	
	After 15 minutes	2.2	98.53
	After 30 minutes	2.1	98.60
	Sample from the filtrate	2.9	98.07
24 (13 repeated)	Initial sample	210	
	After 15 minutes	0.69	99.67
	After 30 minutes	0.54	99.74
	After 45 minutes	0.44	99.79
	Sample from the filtrate	0.51	99.76

*The initial sample of 200 µg/L at pH 6 (experiment 13, the third test in this section) may have been taken before the solution were fully mixed. The experiment was repeated (experiment 24) and the new results replaced the results of experiment 13.

The removal efficiencies are graphically presented in Figure 21:

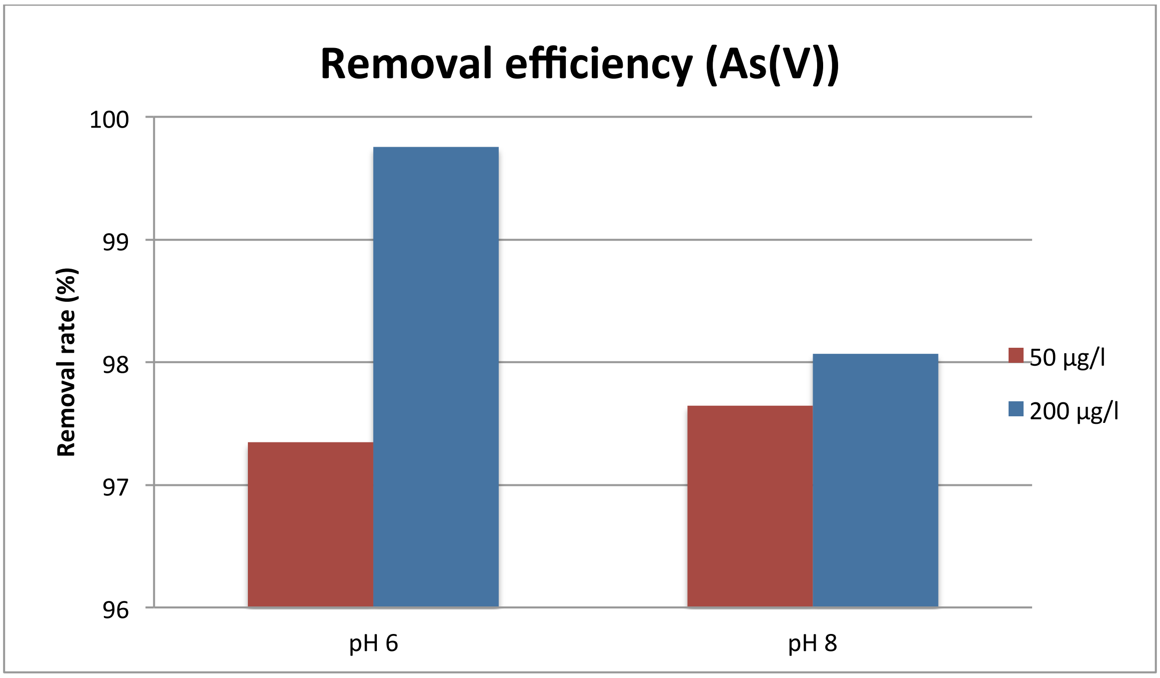


Figure 21: Removal efficiency in the reference test using As(V)

5.6 Test of As(III) Removal Using Double Filtration

Before displaying the results from the double filtration experiments, explanations of the presentations are necessary.

The concentration of As(V) was indirectly measured, assumed to be the difference of the concentration of total arsenic before and after ion exchange. This is why the concentration sometimes came out negative. Ion exchange was used to differentiate between As(III) and As(V), and there was a marginal error of about ± 8 %. In some of the analysis, a higher concentration of total arsenic was detected in the samples taken after running it through the IE resin. The results from these tests were adjusted in order to avoid negative concentrations.

The tables display three removal rates, the removal of As(III), the removal of As(V) and the removal of total arsenic. The removal of As(III) was calculated by comparing the concentration in the filtrate to the initial concentration of As(III). This does not show the concentration of As(V) in the filtrate. The removal rate of As(V) was calculated using the concentration of As(V) after the second filtration compared to the concentration of As(V) in filtrate 1. The removal rate of As(V) was only calculated where the filtrate contained As(V). The concentration of total arsenic (As(III) and As(V) at the outlet after the second filtration) was compared to the initial concentration.

The removal efficiency for the second filtration was found by taking the concentration of As(III) in the filtrate and compare it to the mean concentration of the samples taken after 10 and 20 minutes in the second filtration process.

5.6.1 Experiment 15-18

Target conditions: Experiment 15: 50 µg/L As(III), pH 6
Experiment 16: 50 µg/L As(III), pH 8
Experiment 17: 200 µg/L As(III), pH 6
Experiment 18: 200 µg/L As(III), pH 8

The results are found in Appendix 3. When the results were processed, the negative numbers were adjusted and the removal rates were calculated (Table 13) and graphically displayed

(Figure 22). The mean removal rate of total arsenic for each experiments are presented in Table 14.

Table 13: Adjusted test results of experiment 15-18 and removal rate

Experiment	Adjusted:	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)	Removal rate of total As (%)	Ratio As(V) /tot As (%)
15	Mean concentration	51.5					
	Mean 15 min	12.5	0	75.73		75.73	0.0
	Mean 30 min	11.5	0.5	77.67		76.70	4.2
	Filtrate 1	12	0	76.70		76.70	0.0
	Mean second filtration	3.3	0.15	72.50		71.25	4.3
	Total removal			93.59		93.30	
16	Mean concentration	55.5					
	Mean 15 min	14.5	2.5	73.87		69.37	14.7
	Mean 30 min	15	1	72.97		71.17	6.3
	Filtrate 1	13	3	76.58		71.17	18.8
	Mean second filtration	4.4	0.2	66.15	93.33	64.62	4.3
	Total removal			92.07		91.71	
17	Mean concentration	215					
	Mean 15 min	62	0	71.16		71.16	0.0
	Mean 30 min	62	0.5	71.16		70.93	0.8
	Filtrate 1	60	1	72.09		71.63	1.6
	Mean second filtration	19	0.5	68.33	50.00	67.50	2.6
	Total removal			91.16		90.93	
18	Mean concentration	200					
	Mean 15 min	60	0.5	70.00		69.75	0.8
	Mean 30 min	60.5	0.5	69.75		69.50	0.8
	Filtrate 1	58	3	71.00		69.50	4.9
	Mean second filtration	18	0.5	68.97	83.33	68.10	2.7
	Total removal			91.00		90.75	

Table 14: Removal rate of total arsenic for each filtration step in experiment 15 to 18

Experiment:	15	16	17	18
First filtration	76.70	70.53	71.47	69.54
Second filtration	71.25	71.25	68.03	69.67
Total removal efficiency	93.30	91.71	90.93	90.75

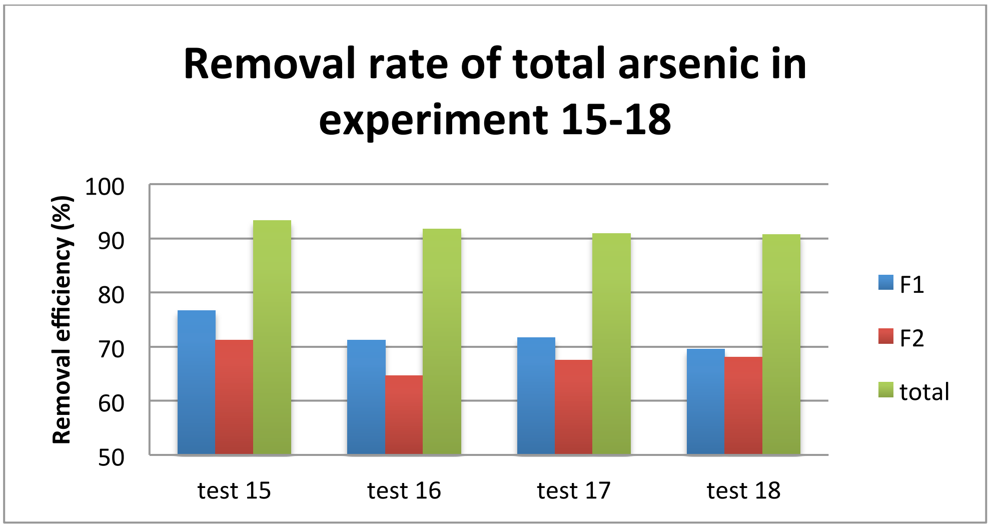


Figure 22: Removal efficiency of total arsenic for first and second filtration and the total removal rate in experiment 15 to 18

5.6.2 Experiment 27-30 (experiment 15-18 repeated)

The RO-unit changed after experiment 20 and 22. Decreased water consumption lowered the amount of rejected water. Therefore, it was only needed to use one tank as water inlet. The results are shown in Table 34 in Appendix 4 and the adjusted values were used to find the removal rates (Table 15, Figure 23).

Table 15: Adjusted test results (experiment 27-30) and the removal rate

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)	Removal rate of total As (%)	Ratio As(V) /tot As (%)
27	Concentration	54	3				5.3
	Filtrate 1	10	0	81.48	100.00	82.46	0.0
	Mean second filtration	2.65	0	73.50	-	73.50	0.0
	Total removal:			95.09	100.00	95.35	
28	Concentration	50	4				7.4
	Filtrate 1	11	0	78.00	100.00	79.63	0.0
	Mean second filtration	3.1	0.05	71.82	-	71.36	1.6
	Total removal:			93.80	98.75	94.17	
29	Concentration	220	0				0.0
	Filtrate 1	53	0	75.91	-	75.91	0.0
	Mean second filtration	16.5	0	68.87	-	68.87	0.0
	Total removal:			92.50	-	92.50	

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)	Removal rate of total As (%)	Ratio As(V) /tot As (%)
30	Concentration	200	10				4.8
	Filtrate 1	49	1	75.50	90.00	76.19	2.0
	Mean second filtration	15	0	69.39	100.00	70.00	0.0
	Total removal:			92.50	100.00	92.86	

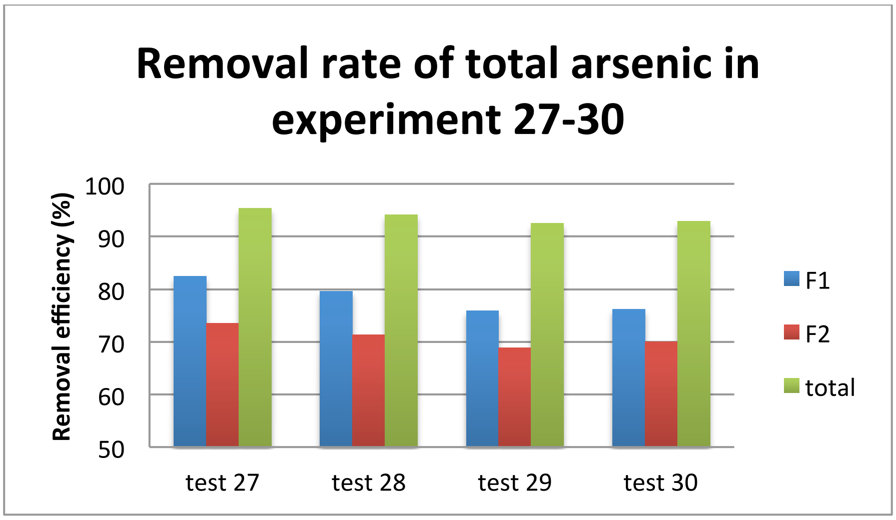


Figure 23: Removal efficiency of total arsenic in experiment 27 to 30

The original values were used in finding the removal rates for each filtration step in experiment 27-30 (Table 16).

Table 16: Removal rate for each filtration step in experiment 27 to 30.

	27	28	29	30
First filtration	82.46	79.63	75.34	76.31
Second filtration	73.50	71.36	69.81	70.00
Total removal efficiency	95.35	94.17	92.73	92.86

The results from experiment 15-18 and 27-30 were then compared graphically (Figure 24):

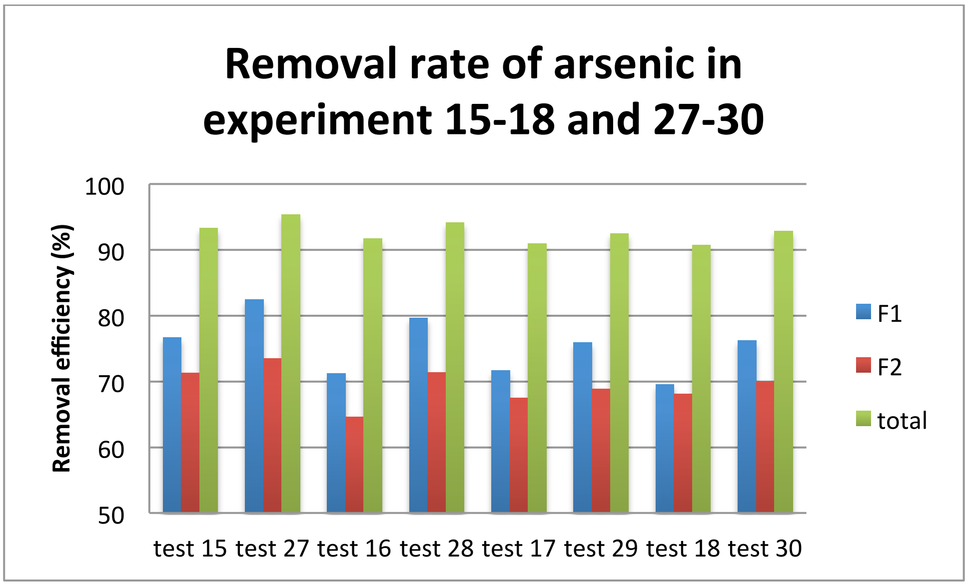


Figure 24: Removal rate of total arsenic in each step in the double filtration tests

5.6.3 Statistic - Mean Values and Standard Deviation of As(III) Removal

Experiment 27 had the same initial conditions as experiment 15; experiment 28 had the same conditions as experiment 16 and so on. The mean removal rate of the experiments was calculated using the concentrations of total arsenic (Table 17). This explains nothing of oxidation of As(III). The average of the calculations were found by using the values from the analysis and the calculations of As(V) concentration before the negative values were set to 0. However, standard deviation was found using the adjusted values (Table 17). This is tolerable since the removal rate was calculated for the removal of total arsenic and was not directly affected by the adjustments.

Table 17: The mean removal rate and their standard deviation

	50, pH 6	50, pH 8	200, pH 6	200, pH 8
RO1	79.58 ± 3.17	75.08 ± 4.59	73.41 ± 2.06	72.92 ± 3.40
RO2	72.38 ± 1.22	71.31 ± 4.74	68.92 ± 1.34	69.84 ± 1.37
Total	94.33 ± 1.02	92.94 ± 1.23	91.83 ± 0.79	91.80 ± 1.05

None of the experiments had unexplainable results or values far from any other result values. However, when the calculation gave negative As(V) concentrations, they were set to 0. Other than that, no big adjustments were done.

5.6.4 As(V) Ratio in the Double Filtration Tests

To know whether As(III) was oxidized during the filtration process or not, the ratio of As(V) to total As was calculated. The values are in the last column in Table 33 and Table 35 in Appendix 3 and 4. The original values were used, which is why some of the ratios came out negative. However, the negative values were not emphasized.

5.6.5 Experiment 23

Experiment 23 was conducted at a higher pH than the other to see how the removal efficiency varied with pH (Table 18). Target condition was 200 µg/L As(III) at pH 10.

Table 18: Test results of experiment 23

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)
23	Tank 1	220	
	After 15 min	22	-1
	After 30 min	21	1
	Tank 2	220	
	After 15 min	19	3
	After 30 min	21	0
	Filtrate 1	21	1
	After 10 min	1.2	23.8*
	After 20 min	1.3	0.3
	After 30 min	1.3	0.1

*The concentration of As(V) after 10 minutes of the second filtration does not make sense and is probably a sampling (or analytical) error.

The numbers were adjusted (Table 19) and compared to experiment 15-18 to examine if there were a correlation between removal efficiency and pH (Figure 25).

Table 19: Adjusted results in experiment 23 and the removal rate of total As.

	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate (%)
Mean concentration	220		
Mean 15 min	20.5	1.5	90.68
Mean 30 min	21	0.5	90.45
Filtrate 1	21	1	90.45
Mean second filtration	1.3	0.2	93.81
Total removal			99.32

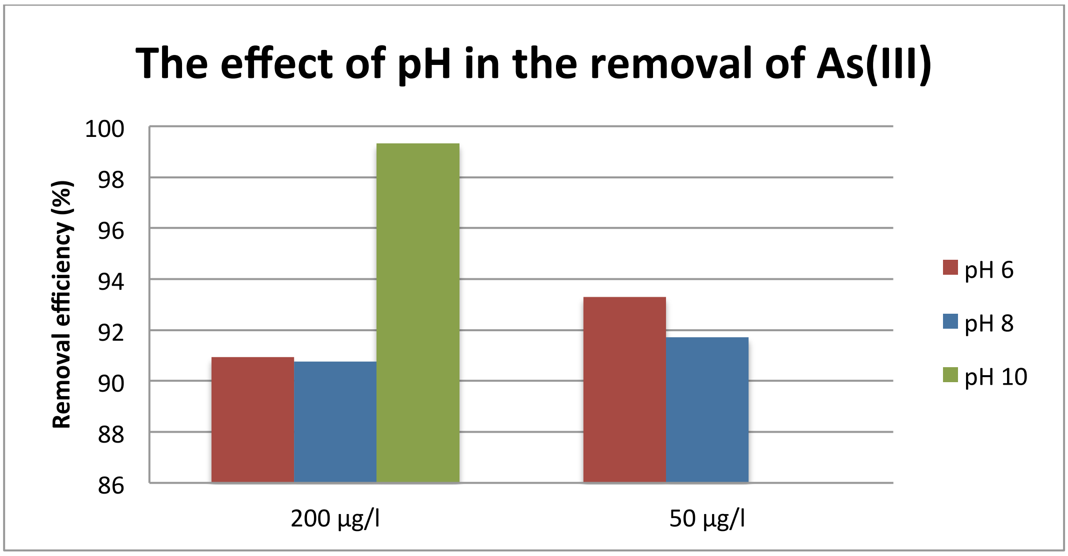


Figure 25: The effect of pH in the removal process of As(III) Experiment 15-18 and 23.

5.7 Analysed Reject Water

To know whether or not oxidation occurred inside the RO unit, samples of the reject water was taken and tested in experiment 27-30 (Table 20, Figure 26).

Table 20: Results of the experiment taken of the reject water in experiment 27-30

Experiment		$C_{total\ arsenic}$ (µg/L)	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Ratio As(V)/ tot As (%)	Increased $C_{total\ arsenic}$ (%)	Increased $C_{As(V)}$ (%) *
27	Initial sample (tank 1)	57.0	54.0	3.0	5.26		
	Reject (15 min)	85.0	76.0	9.0	10.59	49.12	200
	Reject (30 min)	84.0	77.0	7.0	8.33	47.37	133.33
	Sample of filtrate 1	10.0	10.0	0.0	0.00		
	Reject (10 min, F2)	20.0	16.0	4.0	20.00	100	-
	Reject (20 min, F2)	18.0	15.0	3.0	16.67	80	-
28	Initial sample (tank 1)	54.0	50.0	4.0	7.41		
	Reject (15 min)	88.0	79.0	9.0	10.23	62.96	125
	Reject (30 min)	88.0	80.0	8.0	9.09	62.96	100
	Sample of filtrate 1	11.0	11.0	0.0	0.00		
	Reject (10 min, F2)	24.0	18.0	6.0	25.00	118.18	-
	Reject (20 min, F2)	23.0	18.0	5.0	21.74	109.09	-

Experiment		$C_{total\ arsenic}$ (µg/L)	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Ratio As(V)/ tot As (%)	Increased $C_{total\ arsenic}$ (%)	Increased $C_{As(V)}$ (%) *
29	Initial sample (tank 1)	220.0	220.0	0.0	0.00		
	Reject (15 min)	350.0	330.0	20.0	5.71	59.09	-
	Reject (30 min)	350.0	330.0	20.0	5.71	59.09	-
	Sample of filtrate 1	53.0	53.0	0.0	0.00		
	Reject (10 min, F2)	87.0	83.0	4.0	4.60	64.15	-
	Reject (20 min, F2)	87.0	83.0	4.0	4.60	64.15	-
30	Initial sample (tank 1)	210.0	200.0	10.0	4.76		
	Reject (15 min)	330.0	310.0	20.0	6.06	57.14	100
	Reject (30 min)	340.0	320.0	20.0	5.88	61.90	100
	Sample of filtrate 1	50.0	49.0	1.0	2.00		
	Reject (10 min, F2)	84.0	77.0	7.0	8.33	68	600
	Reject (20 min, F2)	83.0	78.0	5.0	6.02	66	400

* Increased concentration (%) = (C reject-C inlet)/C inlet*100

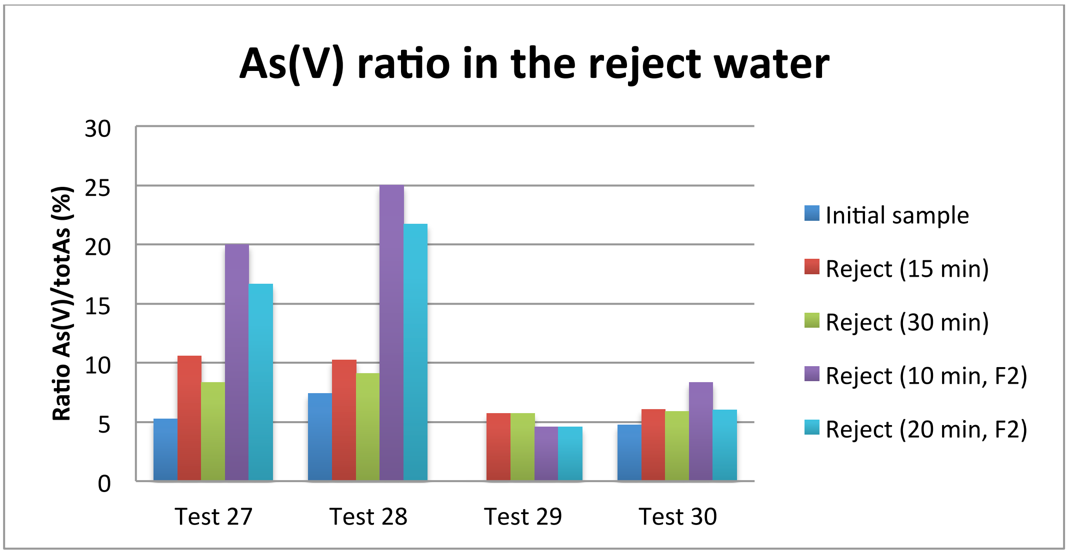


Figure 26: The As(V)-tot As – ratio in the reject water compared to the initial sample

The oxidation was calculated by looking at the theoretical concentration in the inlet and compared to the measured values. Sankey diagrams (Figure 27-Figure 30) were drawn for each test (experiment 27-30) using a demo version from www.e-sankey.com. The diagrams graphically show the flow and concentration at every step of the filtration process. The orange part of the arrows are the amount of As(III). The white part of the arrows are the amount of As(V).

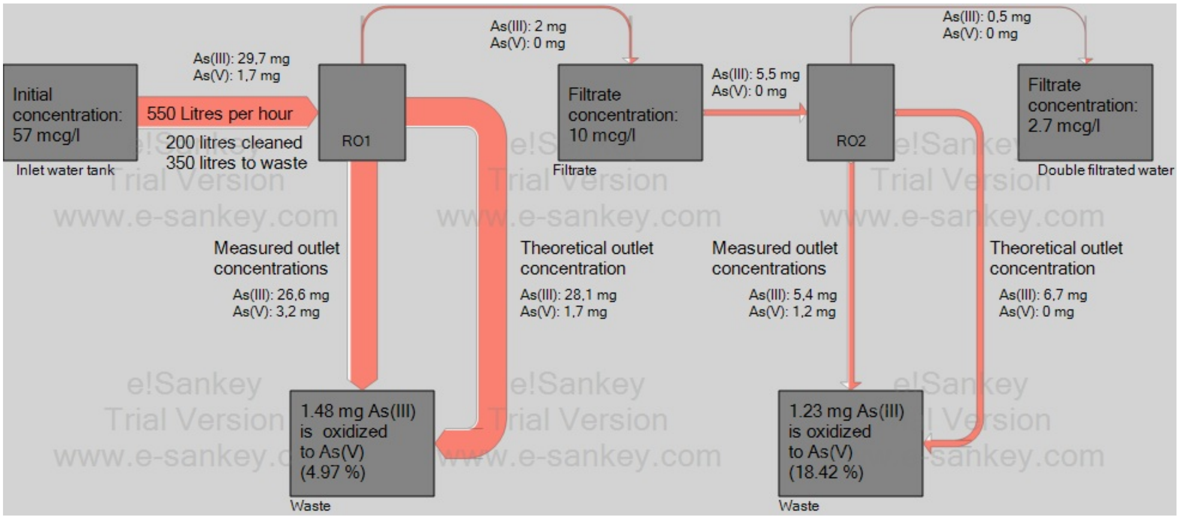


Figure 27: Sankey diagram of experiment 27

To explain the diagram (Figure 27), 200 L/h clean water was produced and 350 L/h was rejected. The initial concentration was 57 µg/L arsenic, 54 µg/L As(III) and 3 µg/L As(V). After the first filtration, the filtrate had a concentration of 10 µg/L As(III), while the reject water had a concentration of 85 µg/L arsenic, 76 µg/L As(III) and 9 µg/L As(V). The reject water contained the same concentration as the inlet water in addition to the arsenic which was held back by the membrane.

The 550 litre going into the machine in one hour contained 31.35 mg As (29.7 mg As(III) and 1.65 mg As(V)). Of the 550 litre, 200 litre went across the membrane and contained 10.8 mg As(III) and 0.6 mg As(V). The cleaned water contained 2.0 mg As(III). This means that 8.8 mg As(III) and 0.6 mg As(V) was held back by the membrane and added to the reject water.

The reject water was 350 liters of the 550 liters. Theoretically, the reject water contained 19.95 mg As (= 57 µg/L), 18.9 mg As(III) and 1.05 mg As(V). When the arsenic that was held back by the membrane was added (without any oxidation), the reject water contained 27.7 mg As(III) and 1.65 As(V).

The analysis showed that the 350 liters of reject water contained 26.6 mg As(III) and 3.15 mg As(III). The theoretical reject water had a concentration of 83.9 µg/L, 1.36 % lower than the measured concentration of 85 µg/L. If the theoretical numbers were adjusted (by multiplying the numbers with 1.0136), the theoretical reject water without oxidation contains 28.08 mg As(III) and 1.67 mg As(V). These numbers show that 1.48 mg As(III) was oxidized to As(V). 1.48 mg was 4.97 % of 29.75, and show that 4.97 % was oxidized during the first filtration process. The same calculations were done for experiment 29-30 (Figure 28-Figure 30).

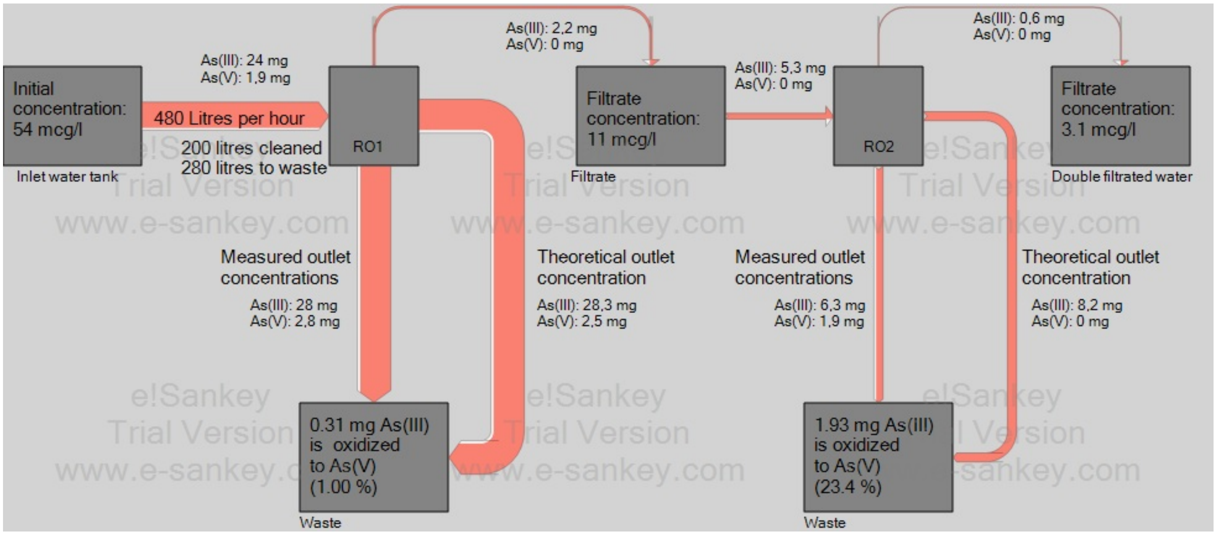


Figure 28: Sankey diagram of experiment 28

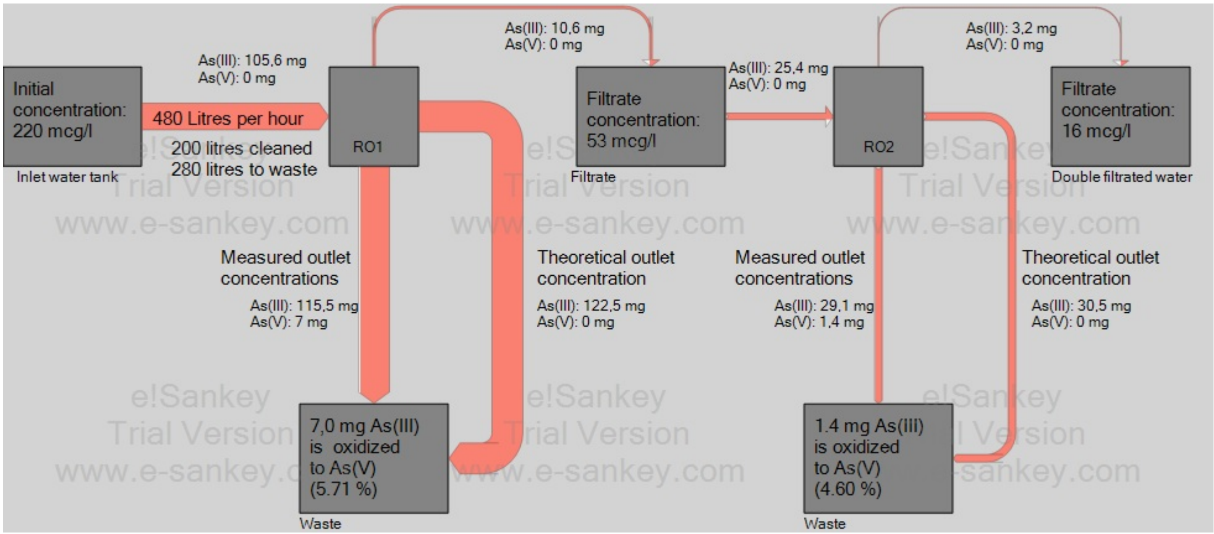


Figure 29: Sankey diagram for experiment 29

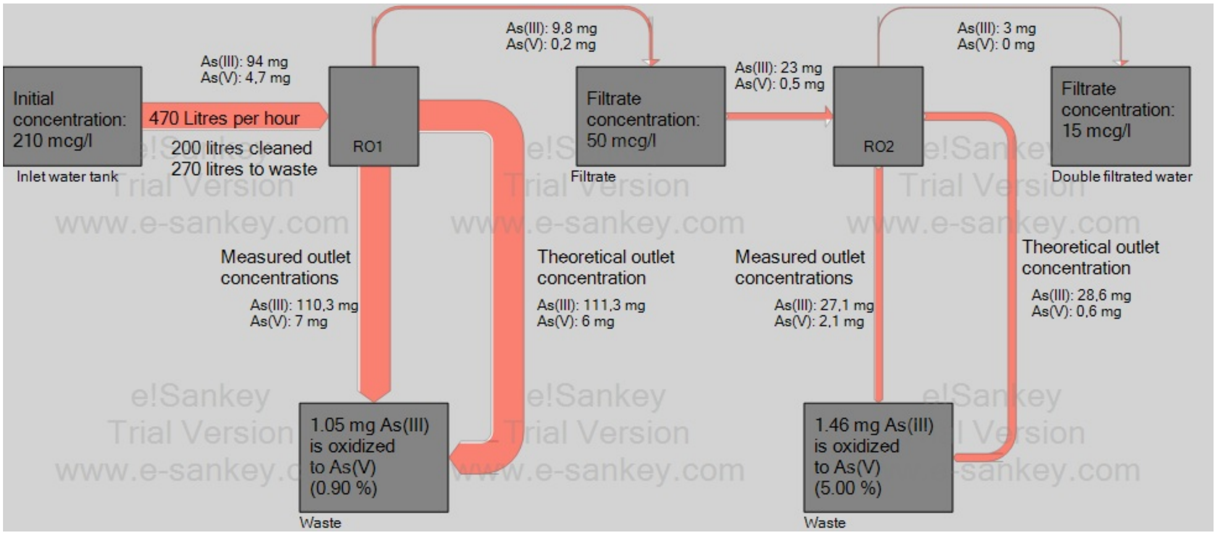


Figure 30: Sankey diagram for experiment 30

5.8 Test of a Mix of As(III) and As(V)

Experiment 19 to 22 tested a mixture of As(III) and As(V) (Table 21). The total removal of As(III) and As(V) calculated separately. The final concentration of As(III) was compared to the initial concentration of As(III).

Target conditions: Experiment 19: 25 µg/L As(III) + 25 µg/L As(V), pH 6

Experiment 20: 25 µg/L As(III) + 25 µg/L As(V), pH 8

Experiment 21: 100 µg/L As(III) + 100 µg/L As(V), pH 6

Experiment 22: 100 µg/L As(III) + 100 µg/L As(V), pH 8

Table 21: Results of the test with mixed arsenic species, experiment 19-22

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)
19	Tank 1	24	25
	After 15 min	4.3	0.1
	After 30 min	4.2	0
	Tank 2	27	23
	After 15 min	4.4	-0.1
	After 30 min	4.4	0
	Filtrate 1	4.5	-0.1
	After 10 min	1	0
	After 20 min	0.98	0.02
20	Tank 1	26	28
	After 15 min	5.4	0.3
	After 30 min	5.5	0.2
	Tank 2	24	35
	After 15 min	6	5
	After 30 min	5.4	0.1
	Filtrate 1	5.9	-0.2
	After 10 min	1.9	-0.5
	After 20 min	1.3	0.1
21	Tank 1	1.2	1.7
	After 15 min	17	0
	After 30 min	16	0
	Tank 2	120	110
	After 15 min	18	0
	After 30 min	16	2
	Filtrate 1	18	0
	After 10 min	4	0.2
	After 20 min	3.8	0.1

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)
22	Tank 1	100	120
	After 15 min	19	1
	After 30 min	20	0
	Filtrate 1	20	0
	After 10 min	4.8	0.1
	After 20 min	4.4	0.1

When the results were adjusted (Table 22), the average values were used to graphically present the results for the removal rate after the first filtration, second filtration and total removal rate (Figure 31 and Figure 32).

Table 22: Adjusted test results in the mixed arsenic species experiments

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)
19	Mean concentration	25.5	24		
	Mean 15 min	4.35	0.05	82.94	99.79
	Mean 30 min	4.3	0	83.14	100
	Filtrate 1	4.5	0	82.35	100
	Mean second filtration	0.99	0.01	78	-
	Total removal			96.12	99.96
20	Mean concentration	25	31.5		
	Mean 15 min	5.7	2.65	77.20	91.59
	Mean 30 min	5.45	0.15	78.20	99.52
	Filtrate 1	5.9	0	76.40	100
	Mean second filtration	1.6	0.05	72.88	0.00
	Total removal			93.60	99.84
21	Mean concentration	120	110		
	Mean 15 min	18	0	85	100
	Mean 30 min	16	2	86.67	98.18
	Filtrate 1	18	0	85	100
	Mean second filtration	3.9	0.15	78.33	-
	Total removal			96.75	99.86
22	Mean concentration	100	120		
	Mean 15 min	19	1	81	99.17
	Mean 30 min	20	0	80	100
	Filtrate 1	20	0	80	100
	Mean second filtration	4.6	0.1	77	-
	Total removal			95.40	99.92

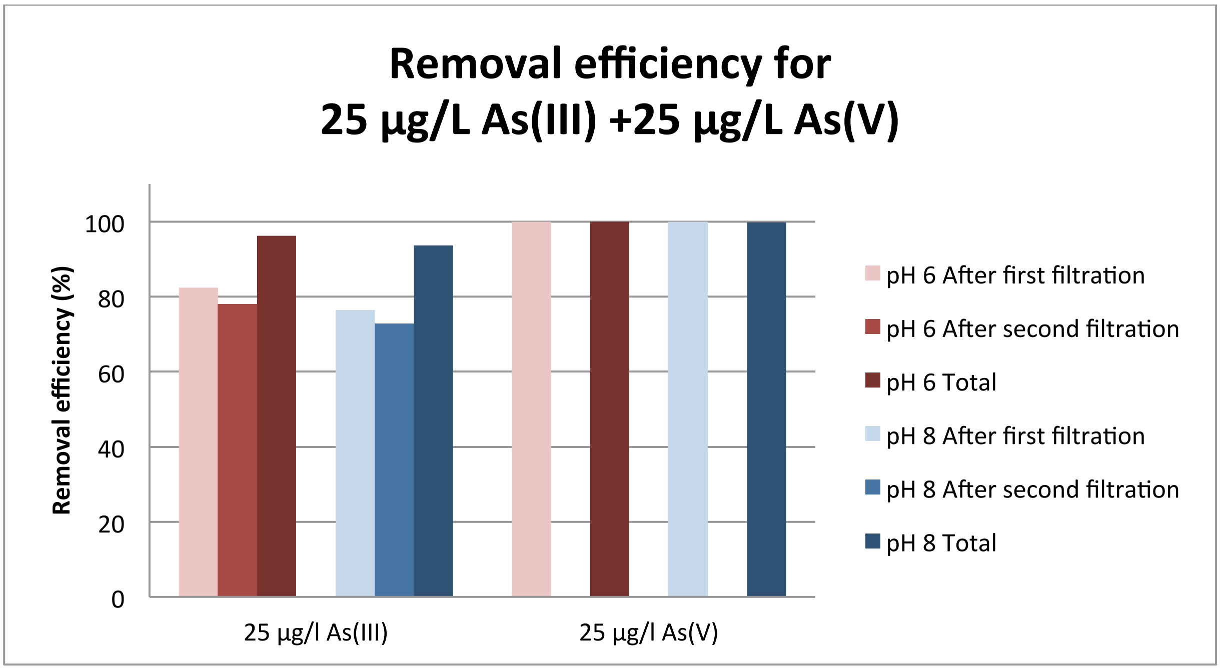


Figure 31: Removal efficiency for experiment 19 and 20

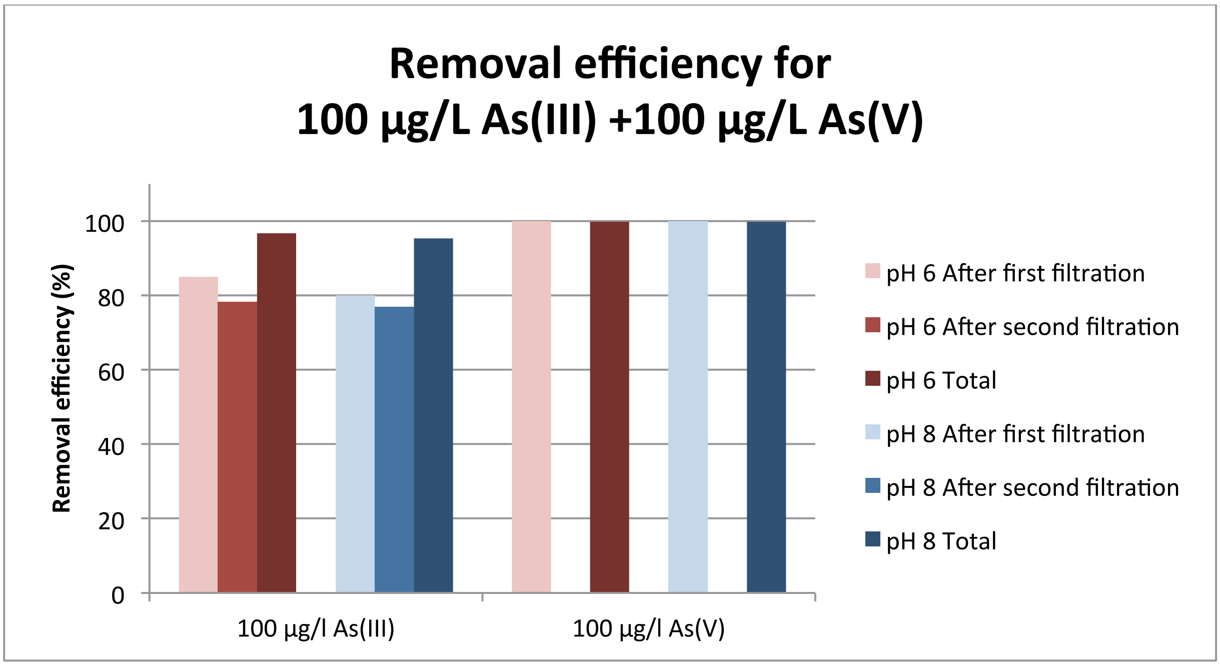


Figure 32: Removal efficiency for experiment 21 and 22

Looked at experiment 19 to 22 all together, Figure 33 displays the total removal efficiency for As(III) and As(V) at two different concentrations:

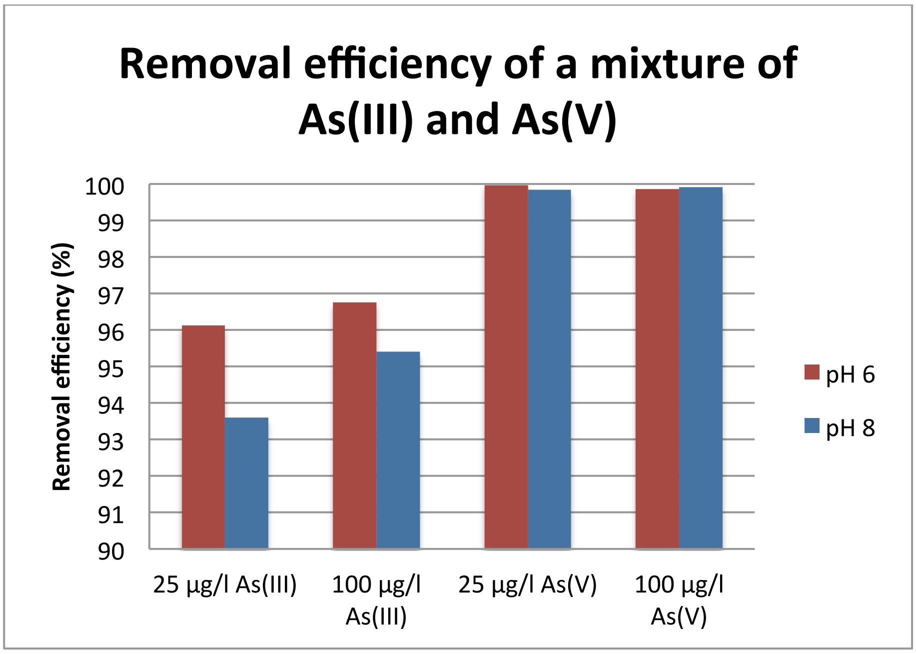


Figure 33: Removal efficiency for experiment 19-22

5.9 Removal of Salts

Experiment 11-16 and 18 were tested for some salts as well as arsenic. They were tested for sodium, magnesium, potassium and calcium (Table 23-Table 26):

Table 23: Removal rate of sodium

Sodium (Na)	Experiment						
	11	12	13	14	15	16	18
Before (C Na, mg/L)	37	29	29	30	34.5	33	30
After first filtration	4.03	1.1	1.8	1.04	1.36	1.36	1.10
Removal rate after RO1 (%)	89.10*	96.21	93.79	96.54	96.06	95.88	96.33
After second filtration (mg/L)					0.07	0.15	0.21
Removal rate after RO2 (%)					99.8	99.56	99.3

* The first test (after 15 min) showed higher values than expected. If this value is left out of the calculations, the removal rate is 95 %.

Table 24: Removal rate of magnesium

Magnesium (Mg)	Experiment						
	11	12	13	14	15	16	18
Before (mg/L)	2.9	2.9	2.9	3.1	2.95	3	3
After first filtration (mg/L)	0.11	0.03	0.06	0.03	0.06	0.02	0.02
Removal rate after RO1 (%)	96.20	98.89	97.98	98.91	98.03	99.39	99.47
After second filtration (mg/L)					0.01	0.00	0.02
Removal rate after RO2 (%)					99.80	99.91	99.47

Table 25: Removal rate of potassium

Potassium (K)	Experiment						
	11	12	13	14	15	16	18
Before (mg/L)	2.7	2.7	2.6	2.7	2.65	2.6	2.6
After first filtration (mg/L)	0.44	0.08	0.29	0.09	0.20	0.08	0.07
Removal rate after RO1 (%)	83.70	96.86	88.72	96.63	92.60	96.99	97.29
After second filtration (mg/L)					0.02	0.01	0.02
Removal rate after RO2 (%)					99.30	99.63	99.40

Table 26: Removal rate of calcium

Calcium (Ca)	Experiment						
	11	12	13	14	15	16	18
Before (mg/L)	21	21	22	24	21.5	22	21.5
After first filtration (mg/L)	0.93	0.22	0.45	0.22	0.45	0.12	0.11
Removal rate after RO1 (%)	95.59	98.94	97.94	99.08	97.92	99.46	99.50
After second filtration (mg/L)					0.09	0.02	0.06
Removal rate after RO2 (%)					99.60	99.90	99.71

6 Discussion

Arsenic is a public health problem in some countries, and Bangladesh is the most known and affected country. There are variable treatment options, both for conventional treatment plants and for households.

Different methods are compared in Figure 13 and in Table 2. The comparison reports membrane processes suitable and stable in removal of arsenic. Membrane processes treat less than half of the amount consumed and the capital cost can be quite high. Due to high energy consumption during the operation, the operational costs are high. On the other hand, there is no need for skilled people operating the unit, and the maintenance is simple. A unit can produce a large amount of potable water in a day, cover the water consumption for a small community and replace multiple household treatment units.

The RO unit provided by A-Aqua is currently used for desalination in communities where the inhabitants buy the cleaned water to finance the unit. The efficiency in removal of arsenic can make the RO unit applicable for areas with arsenic contamination, and the scope of the research was to examine the removal efficiencies of arsenic, especially the removal efficiency of As(III) by double filtration.

6.1 Sources of Errors and Uncertainty

There are several sources of error in this research. The uncertainty of the results originates from uncertainty in the RO unit, IE, sampling and analysis. There are greater uncertainties in the sampling than the analysis. According to the Senior Engineer Karl Andreas Jensen at IPM, who did the analysis, the ICP-MS had an analytical error of about 3 %.

The water inlet consisted of two tanks of 1 m^3 each filled with arsenic spiked water. The marks that marked 1000 litres on both of the tanks were not accurate. The target concentration of 25 µg/L, 50 µg/L, 100 µg/L and 200 µg/L was not easy to get accurate since the tank was filled with an estimated amount of water approximately 1000 litres ± 10 %.

The arsenic solutions of 4 g/L were measured to mix with exactly 1000 litres. To measure the exact volume of arsenic solution, a measuring cylinder was used. The uncertainty is set to

0.25 ml for the measuring cylinder. The concentrations in the inlet water are expected to be within the range shown in Table 27.

Table 27: The uncertainty of inlet water concentrations

Measured arsenic solution á 4 g/L (mL)	C in 1000 L (microgram/L)	C in 900 L	C in 1100 L
6.25	25	27.78	22.73
± 0.25		28.89	21.82
12.5	50	55.56	45.45
± 0.25		56.67	44.55
25	100	111.11	90.91
± 0.25		112.22	90
50	200	222.22	181.82
± 0.25		223.33	180.91

The actual concentration could be expected to be in the range of 44.6 – 56.7 µg/L when the intended concentration was 50 µg/L. The actual concentration was rarely the intended concentration. The intended 50 µg/L was between 49-59 µg/L and 200 µg/L was from 150-220 µg/L. The sample of 150 µg/L, in test 14, was taken from a tank before the experiment started; it might not have been stirred thoroughly. The concentration of 59 µg/L is over the uncertainty margin. The other initial concentrations are within the range of uncertainty as seen in Table 27. The concentrations of the premixed arsenic solutions were assumed to be 4 g/L. Although this solution was not analysed, prepared test water solutions were analysed thus any impacts are minimized. The premixed solutions were stored in a refrigerator, and some might have oxidized while stored.

Uncertainties could also come from sampling. The pipette used to take a test sample from the beaker into the test bottle, or the beaker itself, despite all the care taken, may have been contaminated. Mistakes in execution or sampling (human error) could also occur.

A colour change in the IE resin was observed (Figure 18). After a regeneration of the resin, the colour stayed the same. Therefore, the discolouration did not come from accumulation of arsenate in the resin. The discolouration may have originated from the colour of the tap water. There were no differences in the results before and after regeneration and change of resin, hence the resin was changed or regenerated before the resin was saturated.

The experiments were designed for different concentrations at different pH levels. The pH was measured using a pH meter. The meter had to be calibrated to show the correct pH at all times. Lab demonstrators did this frequently without notice. At what time the pH meter was calibrated was therefore unknown, although the pH meter was assumed to be correct at all times.

The pH was adjusted using hydrochloric acid and caustic soda by adding little by little, the pH meter settled before adding more chemicals. The tap water at Ås has a low buffer capacity, and the pH can easily fluctuate. The pH therefore changed during experiments. The pH in the experiments using the RO unit had pH values up to ± 0.5. Most of the experiments had pH changes less than 0.2. Since the pH is logarithmic, this was quite a lot.

In the first experiments, the sum of the filtrated water and reject water was about 800 litres per hour, not 1 m^3/h. However, the pressure changed after experiment 20 and 22 and the sum of it was as low as about 500 litres per hour at the last experiments.

6.2 Verification of the RO-system

The verification of the built-in flow meters on the RO-unit showed that in one hour when the flow meter displayed 200 L/h, the real amount was actually 191 litres of water. At the same time, 620 litres went through the RO-unit untreated (reject water).

In order to get enough water for a second filtration, two tanks of 1 m^3 were filled up and spiked with arsenic. Since two tanks of 1 m^3 were used as water inlet, the verification of the RO-unit's flow meters made it clear that if the experiment would be executed with low flow rates (below 100 L/h) the two water tanks were insufficient in order to produce enough filtrate to do a second filtration. Hence, the experiments used only one flow rate. The rest of the experiments were done with a flow rate of 200 litres per hour according to the built-in flow meter, in other words 191 litres per hour.

6.3 Verification of the Indirect Analytical Method for As(V)

The As(V) was indirectly estimated as the difference between the total As and As(III), which were found as the total As analysis of the test water and water treated by ion exchange which selectively extracted As(III). The results from the verification of the indirect analytical method for As(V) are found in Table 9. In the two first lines the concentration of As(V) should be 0, and in the next two lines the concentration of As(III) should be 0 because the test water assumingly contained only one of the two oxidation states.

In the first experiment, the concentration of As(III) was calculated to -3, that could be an analytical error. The fact that the concentration was negative confirms this. The second experiment gave a 3.97 % of As(V) content. This may be within the uncertainty of the resin, however, the initial samples of experiment 27-30 were run through the ion exchange and showed that the As(III)-solution contained between 0 and 9.5 % As(V). Karl Andreas Jensen at IPM mixed the arsenic solutions; and it was assumed that the arsenic solutions were pure without further testing.

The experiments were only done once for each concentration. There was not enough information to conduct a statistical analysis. Therefore, a simple expected margin of error was calculated by dividing the actual concentration where it was expected to be 0 by the concentration of total arsenic before the ion exchange.

The verification gave a margin of error of the ion exchange resin as high as 8 %, but the resin was nevertheless found to be reliable to distinguish between As(III) and As(V).

6.4 Oxidation

The literature shows that oxidation of As(III) by dissolved oxygen and mixing of water does not occur in a great matter (Lowry & Lowry 2002). Smedley and Kinniburgh (2001) reported that the oxidation rate of As(III) is dependent of pH and is lowest at acidic pH levels. The results from the first experiment at the lab presented that approximately 10 per cent of As(III) was oxidized at pH 6 and 8, and up to 20 per cent at pH 10 during 24 hours (Figure 25).

When the experiment was repeated at concentration 200 µg/L, the initial sample was run through the IE. This was not done the first time. Before the magnetic mixers had started on the second oxidation experiment, the initial sample showed 5 % As(V) content at pH 6 and 9.5 % at pH 8. The As(V) content increased to respectively 9.5 % and 14.3 % after two hours, before the ratio was down to the initial oxidation ratios after 24 hours.

The behaviour of the repeated experiment showed a small amount that got oxidized, however after 24 hours, the As(V) content was down to the initial concentration. A change from As(III) to As(V) was expected at all times, and it is unknown if this happened because of change in pH, sampling errors or any other reason. The initial concentration of As(V) was not tested in the first oxidation experiment. Nonetheless, the results of the first experiment showed an increased concentration of As(V) after 2 hours and then after 24 hours. There might have been errors in the repeated experiments, since the As(V) increased during the first 2 hours and then decreased.

The pH was measured at the same time as the samples were taken. The values are listed in Table 31 in Appendix 2. Regardless of the intended pH, the pH was between 7 and 7.5 after 24 hours in both experiments. The fluctuation of pH might originate from the low buffer capacity of the tap water and its' carbonate system. Change of temperatures and mixing will convert some carbonates and bicarbonates to CO_2, which increase the pH. The pH levels were close to the initial pH levels after two hours, and the exact reason for the change in pH during 24 hours is unknown.

If oxidation occurred during the filtration process, the proportion is unknown and was not shown in the results. However, the results of the analysis of the reject water showed small signs of oxidation in the reject water. This will be discussed later in chapter 6.7.2. The oxidation experiment showed that up to 10 percent of As(III) was oxidized to As(V). It is likely to assume that a small amount, ± 5-10 %, was oxidized based on the oxidation experiment.

6.5 Ratio of As(V) in the As(III) Solution

The concentration of the premixed solutions were not controlled and checked. It was assumed that the solutions were accurately 4 g/L and did not contain other species of arsenic. When experiment 27-30 were conducted, the initial samples were run through the IE resin. Of six samples, only one showed no content of As(V) while the other samples had a As(V) ratio of 4.8-9.5 %.

When the verification of the IE resin was conducted, the first As(III) solution contained no As(V), while the second solution contained 4 % As(V). Besides, one of the last experiment did not contain As(V).

The ratio was higher at pH 8 than pH 6. This might be because a larger part of As(III) was present in its ionized form and got more easily oxidized (Smedley & Kinniburgh 2001). The As(V) solution was tested in the verification test of the resin which indicated < 1 % As(III) in the solution. This is within the uncertainty range of the analysis.

6.6 Reference Test Using As(V)

In order to be able to compare the removal rate of As(III), four experiments were conducted of As(V). As(V) is easier to remove from water, mostly because it is ionized in normal pH conditions. The four experiments were carried out by filtrating the solutions once.

The third experiment, experiment 13, had an inlet concentration of 66 μg/L instead of the wanted 200 μg/L. The rest of the test results matched an initial concentration of about 200 μg/L, however, the experiment was repeated in experiment 24 and experiment 13 was not used further. The reason for an unexpected low inlet concentration might be that the sample was taken before the added arsenic solution was fully mixed in the tank. However, the pressure changed after test 20 and 22, and test 13 was repeated after the change of the RO unit.

At the first four experiments, the concentration of the filtrate is higher than expected compared to the other experiments. The sample from the filtrate may show greater concentrations of total arsenic because some water came out of the cleaned water tube when the machine was flushed. The tube gave water that had gone through the membrane; therefore, the probability of untreated water coming out of the tube is small. However, the filtrate eventually got a more expected concentration. The first experiment had a 286 % higher filtrate concentration than the average concentration of the samples taking after 15 and 30 minutes. The second filtrate was 226 % higher, and the fourth filtrate was 135 % higher. The repetition of experiment 13 resulted in a filtrate concentration of 91 % of the average concentration of the samples taking during the filtration process. The membrane was brand new, and even though the RO unit was run with tap water for twenty minutes before the experiments started, the membrane might have needed longer time to work optimally and might have lead to a higher share of arsenic passing through the membrane.

The removal rate was calculated by comparing the filtrate to the initial concentration. The filtrate had higher concentrations in experiment 11, 12 and 14 compared to the samples taken from the cleaned water tube after 15 and 30 minutes. This resulted in lower removal rates than the calculated removal rates in the samples taken after 15 and 30 minutes. The reference test gave a 97.3 – 99.8 % removal of As(V), and all the experiments had a concentration lower than the recommended limit of 10 µg/L after the filtration. The experiments were conducted once for each condition, and there was not enough data to give a statistical analysis of the results, and therefore no basis to see a relationship between removal rate and pH or concentration. Anyway, Figure 21 shows that a higher concentration gave a higher removal rate.

6.7 Double Filtration of As(III)

The first experiments of double filtration were as expected, with the exception of two of the samples. The initial level of arsenic in tank 1 in experiment 16 was 59 µg/L, a value outside of the uncertainty range (Table 27). The concentration of As(V) after 15 minutes of filtrating from the same tank was also higher (4 µg/L) compared to the other results.

Some of the concentrations of As(V) came out negative because of the uncertainty in the research. However, the magnitude of the error was not greater than the uncertainty limit.

The second time the experiments were conducted, the pressure had changed. These tests were the first where the initial samples were run through IE as well as discussed in 0. Besides from that, none of the samples were outside the limit range except for two negative concentrations of As(V) within the uncertainty limit.

6.7.1 Removal Rate of Total Arsenic

The RO-unit changed after experiment 20 and 22 and produced less reject water and became more efficient, as Ortiz explained, because of the decreased pressure. The removal rates for arsenic at the different conditions were calculated, using a mean of experiments 15-18 and 27-30 (Table 28).

Table 28: The removal rate for total arsenic in the four different condition, and for As(III) and As(V)

Mean removal rate (%)								
	50, pH 6		50, pH 8		200, pH 6		200, pH 8	
	As(III)	As(V)	As(III)	As(V)	As(III)	As(V)	As(III)	As(V)
RO1	79.58		75.08		73.41		72.92	
RO2	72.38		71.31		68.92		69.84	
Total	94.33	97.35	92.94	97.65	91.83	99.76	91.80	98.07

The removal rate for the first filtration step was between 72.9 % and 79.6 % when the arsenic was a As(III) solution, and between 97.4 and 99.8 % for As(V) (Table 28). One filtration step for As(V) was sufficient compared to the limit of 10 µg/L recommended by WHO.

One filtration step was not sufficient for As(III) since the concentration was above 10 µg/L in the filtrate. However, the double filtration process removed enough arsenic when the concentration was about 50 µg/L. The arsenic concentration after two filtration processes was between 15 and 18 µg/L when the initial concentration was approximately 200 µg/L, and the concentration was higher than the recommended limit set by WHO.

6.7.2 Oxidation Inside the Unit

If oxidation occurs during the filtration process, the proportion is unknown and not shown in the results. From the reference test, it can be assumed that the removal of the potential oxidized As(V) was 97-99 %. The ratio of As(V) to total As was calculated in order to see if oxidation happened (Table 29).

Table 29: The ratio of the concentration of As(V) to total arsenic in the different filtrations in experiment 15-18 and 27-30

Experiment		Ratio As(V)/total As
15	RO1	0-4.2 %
	RO2	4.3 %
16	RO1	6.3-18.8 %
	RO2	4.3 %
17	RO1	0-1.8 %
	RO2	2.6 %
18	RO1	0.8-4.9 %
	RO2	2.7 %
27	RO1	0 %
	RO2	0 %
28	RO1	0 %
	RO2	1.6 %
29	RO1	0-1.8 %
	RO2	0 %
30	RO1	0-2 %
	RO2	0 %

Experiment 16 stand out with a much higher ratio of As(V). The other experiments have a ratio between 0 and 4.9 % As(V) after the RO processes. There was a small As(V) content in experiment 27-30, the highest ratio was 2 %. This is within the uncertainty limit. The uncertainty margin for the analysis is 3 %, and the results in table 29 cannot confirm that oxidation occurred. However, if 10 % of the As(III) was oxidized and 97-99 % of the oxidized As(V) was removed during the RO process, there would be 0.001-0.003 % As(V) in the filtrate after RO1. This amount is not even detectable.

A new analysis was conducted to check if oxidation occurred. The reject water was analyzed in experiment 27-30 to see if As(III) was oxidized when the water passed the unit. Where the ratio of As(V) to total As in the inlet water was in the range of 4.76-7.41 %, the ratio was 5.88-10.59 % in the reject water after RO1. During the RO2, the ratio of As(V) to total As was in the range 6.02-25.0 %. The As(V) concentration increased with 100-200 % while the total As concentration increased with 47.4-63.0 % during RO1.

Since the As(V) concentration increased more than As(III), oxidation was one valid explanation. Another reason could have been that all As(V) was rejected while some As(III) got through the membrane. However, a greater amount of As(V) was in the reject water than the theoretical amount without oxidation as seen in the Sankey diagrams (Figure 27-Figure 30). The oxidation ratios are found by using the amount of As(III) that is oxidized (the difference between the theoretical and the measured amount of As(III) in the reject water was assumed to have oxidized) divided by the total amount of arsenic in the reject water. These numbers are also shown in the Sankey diagrams.

In these numbers, experiment 29 is left out because the inlet water had no content of As(V), and had lower ratios in RO1 and RO2. During RO1, the ratio was 5.71 and during RO2 the ratio was 4.6.

From the analysis of the reject water, the oxidation rates in the different steps are summed up in Table 30.

Table 30: Oxidation in the reject water

Experiment	27	28	29	30
RO1	4.97	1.00	5.71	0.90
RO2	18.42	23.40	4.60	5.00

These calculations do not take into account the uncertainty of the experiments and analysis. The oxidation rates in RO1 are not clear enough to conclude that oxidation happened during the process. Experiment 27 and 28 had a target concentration of about 50 µg/L in the inlet, and after the second filtration process, the concentration was about 2.6-3.1 µg/L. Small changes to these numbers will have a great impact on the oxidation calculations. Nonetheless, there was a minor amount of As(III) that was oxidized, but not in the same magnitude as in the oxidation experiment. The oxidation rate was closer to 5 % than 10 %.

6.7.3 Statistics

Two experiments were conducted for the four different conditions using As(III), and there were basis for comparison. However, the two experiments of each condition had different pressures across the membrane. The results were compared anyways.

The mean removal rate was calculated using the concentrations of total As before adjustments. The standard deviation was found using the adjusted values where the negative concentrations of As(V) was put to 0. This is tolerable since the removal rate was calculated for the removal of total arsenic and was not directly affected by the adjustments. The removal rate would have been less if the “real” values were used because the As(V) concentration got a negative value where the sample after IE had a higher arsenic content than the sample before. Of the two, the lowest concentration was used finding the standard deviation.

15 of the 56 removal rates were outside the mean values ± σ (Table 36 in Appendix 5). Because the samples were small, there were a greater uncertainty to the results and higher standard deviations than for a greater sample.

6.7.4 Correlation Between pH and Removal Rate or Between Concentration and Removal Rate?

The effect of pH is compared in Figure 25 and the effect of concentration in Figure 34. It is clear that pH 10 had higher removal efficiency than lower pH levels. This was also discovered in previous studies using membranes from Nitto Electric Industrial Co. At pH 10, most of As(III) is in an ionized form, and was more easily removed.

However, there are no clear correlation between pH 6 and 8 and the removal rate. There might be a slightly higher efficiency at pH 6 compared to pH 8, however the sampling size is too small to confirm this.

The results have shown a higher removal rate at the concentration of 50 µg/L than 200 µg/L, but not more than around 1 percentage point (Figure 34). This was not enough to confirm a correlation, especially not when the sample size was as small as this.

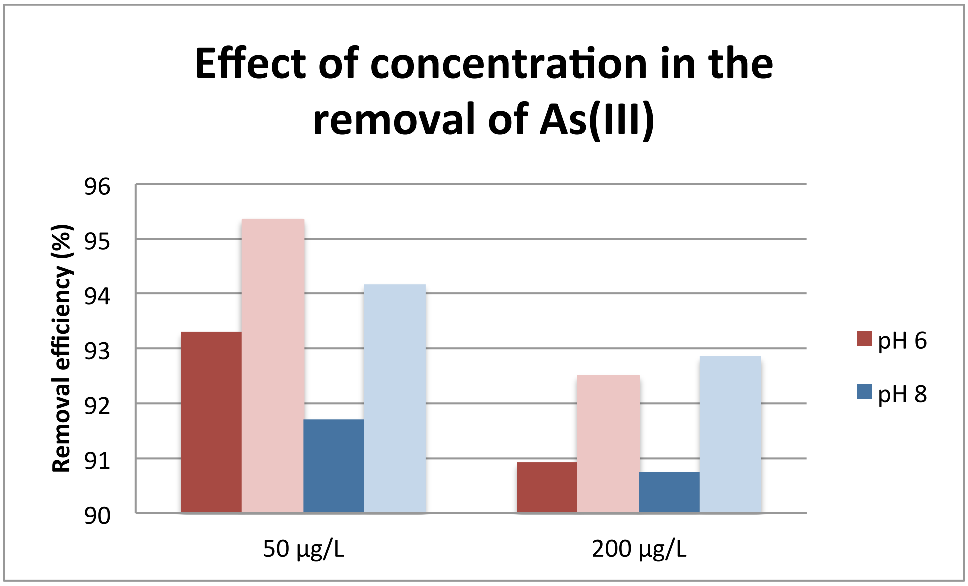


Figure 34: Effect of concentration in removal of As(III), put together from Figure 24. The darker hues are from experiment 15-18, the lighter hues are from experiment 27-30

6.8 Test of a Mix of As(III) and As(V)

In experiment 19 to 22, the RO unit was tested with water with a mixed concentration of As(III) and As(V). From Figure 31 and Figure 32, the removal efficiency for As(V) after the second filtration is not displayed because As(V) was totally removed during the first filtration and the concentration of As(V) in the filtrate was 0.

In the sample taken after 15 minutes from tank 2 in experiment 20, the concentration was higher than expected compared to the other experiments taken from the first tank. The reason for this may have been sampling error, because the error was higher than the uncertainty in the analysis. The inlet concentration in tank 2 in experiment 20 was also relatively high. While the As(III) was 24 μg/L as wanted, the As(V) concentration was 35 μg/L. There might be several reasons for the elevated concentration. First of all, As(III) could have got converted to As(V) while mixing. The second explanation could be error in the way the sample was run through the resin or fault in the sampling procedure (i.e. contamination of the pipette, sampling bottle or the beaker where the water was collected after IE).

The initial concentration in tank 1 in experiment 21 was a hundred times lower than the intended concentration. The other results from tank 1 were not different from expected values. This could mean that the sample was taken before the tank was fully mixed. The results from tank 1 was therefore not used in further calculations even though the filtrate from tank 1 was mixed with the filtrate from tank 2 and used as inlet water in the second filtration.

The results of experiment 19 to 22 was calculated by dividing As(III) and As(V). Because of the two elevated concentrations explained in experiment 20, the As(V) removal was 91.6 % after 15 minutes in experiment 20, tank 2. Besides that, the As(V) removal was between 98.2 and 100 % for the first filtration. Because the filtrate did not contain any As(V) in any of the four experiments, the removal efficiency regarding As(V) was not calculated in the second filtration process. This was also the reason for missing columns in Figure 31 and Figure 32. However, the overall removal rate was 99.84-99.96 % for As(V). The removal efficiency for As(III) was in the first filtration between 76.4 % and 86.7 %. The second filtration process had a 72.9-78.3 % removal efficiency and gave a total removal of 93.6-96.75 %.

The removal rates of As(III) and As(V) were greater when the inlet water contained both As(III) and As(V). The reference test gave 97.3 – 99.8 % removal of As(V), the mixed solution gave the same and even a little bit better results. The removal rate of As(III) after two filtrations were 91.8-94.33 %, the removal of As(III) in the mixed solutions gave two percentage points higher removal rates.

Oxidation might have happened during the mixed solution experiments as seen by the concentration of As(V) after the second filtration. Since the removal efficiency of As(V) is close to 100 %, the concentration of As(V) in the second filtrate came from either oxidation or from the uncertainty margin. The calculated values for As(V) after RO2 was a few tenths, some results came out negative and some were positive. This could mean that the uncertainty might be a more valid explanation than oxidations. However, the previous experiments have shown oxidation rate of about 5-10 %.

6.9 Removal of Salts

The samples from experiment 11-16 and 18 were tested for sodium, magnesium, potassium and calcium in addition to arsenic. Table 23 to Table 26 shows the results of the analysis, and it is worth noticing the low concentrations of the four elements after double filtration.

In order to use this water as drinking water, it is highly recommended to add salts or increase the values of the trace minerals and electrolytes in other ways.

7 Conclusion

The verification of arsenate (V) removal gave a removal efficiency of 97-99 % of As(V) after one filtration and it is evident that removal of As(V) is close to 100 % efficient.

Arsenite (III) removal is normally 30-60 % during RO processes, according to the literature. However, the double filtration concept evaluated in this thesis resulted in arsenic removals up to 94.3 %. The total arsenic removal was found to be between 91.8-94.3 %. The removal of arsenite in the first filtration was in the range 72.9-79.6 % and in the second filtration between 68.9 % and 72.4 %. For test water consisting both with arsenite and arsenate, the RO system gave a total arsenite removal between 93.6-96.7 %. The arsenate removal was 99.8-99.9 % after two filtrations.

Double filtration is an efficient method to treat water that contains arsenite. The removal of arsenite was sufficient when the initial concentration was 50 µg/L. At initial concentration of 200 µg/L, the concentration after two filtrations was higher than the recommended limit of 10 µg/L.

One of the theories for the removal efficiency of As(III) was oxidation. The oxidation experiments showed that 5-10 % of arsenite could be expected to oxidize to arsenate. Aeration was found to be a slow and inefficient way to oxidize As(III). Tests of the reject water were tested and showed a small oxidation rate of up to 5 % in the first filtration and between 5-23 % in the second. However, the concentrations in the tests taken during the second filtration were so small that the results would have been drastically different if the analysis were inaccurate. The conclusion is that assumingly only about 5 % of arsenite was oxidized. An effective membrane and RO unit, rather than oxidation, caused the great removal efficiency of arsenite.

The removal rates were found to be higher at lower concentration, but there was no evidence to support a clear correlation between concentration and removal rates. There is no correlation between pH and removal efficiencies in the pH rate of 6 to 8, however, the removal rate is noticeably higher at pH 10 when As(III) is ionized.

Recommendations: In order to meet the recommended limit set by WHO, water sources with high concentrations of As(III) should be chemically oxidized before filtrating or filtrated at pH > 10 based on this research. However, this requires trained operators and monitoring. The RO unit was very efficient in removal of As(V) and when double filtrating water with concentrations of As(III) around 50 μg/L. Due to high water rejection, the water should be recycled in water scarce areas.

Further experiments should look into the oxidation process and examine it more closely. The reject water was only tested in the last experiments in this research, and there was not enough information to determine whether the As(V) concentration in the reject water was a single case or due to oxidation.

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9 Appendix

Appendix 1: Procedure for experiment 15-23 and 27-30

Appendix 2: Experimental conditions

Appendix 3: Test results for experiment 15-18

Appendix 4: Test results for experiment 27-30

Appendix 5: The uncertainty in experiment 15-18 and 27-30

Appendix 1

Procedure for experiment 15-23 and 27-30:

Flush the membrane for 15-20 minutes with tap water in the beginning of each day

Fill one or two tanks of 1m^3 with tap water and mix the As solution in the tanks in wanted concentration.

Take a water sample from the tank(s). Measure pH at the beginning of each test series and several times per day

This is the inlet water for the first filtration (F1)

Start the filtration process and gather the filtrate for the second filtration

Take the first sample after 15 minutes

Take the second sample (replicate) after 30 minutes

Stop the system to change inlet. The filtrate is now the inlet water for the second filtration (and the outlet can go directly to the waste tank)

Take a test of the filtrate (the new inlet), measure and adjust the pH

Start the second filtration (F2)

Take the first sample after 10 minutes

Take second sample after 20-30 minutes (at least 5 minutes after the first sample is taken)

Stop the system and flush if necessary between test series

Appendix 2

Table 31: Experimental conditions

Experiment	pH	Conductivity	P11	P12	P13	P14	Water flow Cleaned water	Water flow Reject water
11	6.16-6.23	17-18	0.35-0.37	0.33-0.35	1.4-1.67	1.41-1.72	200	550-750
12	8.07-8.35	8-10	0.37	0.34-0.35	1.6	1.61-1.68	200	650-700
13	6.17-6.49	17	0.36	0.34-0.35	1.5-1.51	1.5-1.57	200	650
14	7.81-7.98	7-9	0.32-0.375	0.32-0.355	1.58-1.7	1.6-1.73	200	550-625
15	5.68-5.88 (4.89-5.4)	17-19 (8-9)	0.35-0.375	0.34-0.355	1.55-1.6	1.6-1.7	200	600-640
16	7.93-7.96 (7.81-8.12)	8-10 (3-4)	0.36	0.34-0.35	1.5-1.6	1.6-1.67	200	570-620
17	5.94-5.98 (5.98-6.05)	16-17 (7)	0.36	0.34-0.35	1.55-1.6	1.6-1.65	200	590-600
18	8.03-8.06 (8.31-8.55)	7-8 (3-4)	0.36-0.37	0.34-0.35	1.55-1.6	1.6-1.62	200	600-650
19	6.02-6.09 (6.06-6.12)	13-14 (7)	0.37-0.38	0.35-0.36	1.6-1.69	1.7-1.71	200	500-550
20	7.97-8.02 (8.04-8.12)	6-7 (1-2)	0.38	0.36	1.05-1.3	1.1-1.35	200	400-450
21	6.07-6.09 (6.03-6.09)	11-12 (6)	0.37-0.38	0.35-0.36	1.5-1.58	1.55-1.61	200	550-600
22	7.93-7.98 (7.83-7.94)	6-7 (3)	0.37-0.38	0.35-0.36	0.7-0.9	0.7-0.9	200	300-400
23	9.93-10.26 (10.17-10.36)	6-8 (5)	0.37-0.38	0.35-0.36	0.6-0.8	0.65-0.82	200	450-500
24	5.91-5.99	14-16	0.38	0.36	0.8	0.85	200	400
27	6.00-6.09	11-13 (7)	0.38	0.36	0.65-0.8	0.7-0.82	200	300-380
28	7.87-8.10 (7.56-7.9)	7-9 (3-5)	0.38	0.36-0.365	0.6-0.78	0.7-0.8	200	260-290
29	6.05-6.10 (6.03-6.16)	13-15 (6-12)	0.37-0.38	0.35-0.36	0.55-0.65	0.6-0.7	200	280
30	7.86-7.94 (7.68-7.99)	7-9 (3-4)	0.37-0.38	0.35-0.36	0.55-0.7	0.6-0.7	200	260-280

Appendix 3

Table 32: Test results of experiment 15-18

Experiment		$C_{As(III)}$ (μg/L)	$C_{As(V)}$ (μg/L)
15	Tank 1	52	
	After 15 min	13	-1
	After 30 min	12	0
	Tank 2	51	
	After 15 min	12	0
	After 30 min	11	1
	Filtrate 1	12	0
	After 10 min	3.3	0.2
	After 20 min	3.3	0.1
16	Tank 1	59	
	After 15 min	14	4
	After 30 min	15	1
	Tank 2	52	
	After 15 min	15	1
	After 30 min	15	1
	Filtrate 1	13	3
	After 10 min	4.4	0.2
	After 20 min	4.4	0.2
17	Tank 1	210	
	After 15 min	61	-1
	After 30 min	62	-1
	Tank 2	220	
	After 15 min	63	-1
	After 30 min	62	1
	Filtrate 1	60	1
	After 10 min	19	1
	After 20 min	19	0
18	Tank 1	190	
	After 15 min	60	0
	After 30 min	61	0
	Tank 2	210	
	After 15 min	60	1
	After 30 min	60	1
	Filtrate 1	58	3
	After 10 min	18	0
	After 20 min	18	1

Table 33: Adjusted test results for experiment 15-18 and calculations

Experiment	Adjusted:	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)	Removal rate of total As (%)	Ratio As(V)/tot As (%)
15	Tank 1	52					
	After 15 min	13	0	75.00		75.00	0
	After 30 min	12	0	76.92		76.92	0
	Tank 2	51					
	After 15 min	12	0	76.47		76.47	0.0
	After 30 min	11	1	78.43		76.47	8.3
	Filtrate 1	12	0	76.70		88.35	0.0
	After 10 min	3.3	0.2	72.50		70.83	5.7
	After 20 min	3.3	0.1	72.50		71.67	2.9
	Total removal			93.59		93.30	
	Mean concentration	51.5					
	Mean 15	12.5	0	75.73		75.73	0.0
	Mean 30 min	11.5	0.5	77.67		76.70	4.2
	Filtrate 1	12	0	76.70		76.70	0.0
	Mean second filtration	3.3	0.15	72.50		71.25	4.3
	Total removal			93.59		93.30	
16	Tank 1	59					
	After 15 min	14	4	76.27		69.49	22.2
	After 30 min	15	1	74.58		72.88	6.3
	Tank 2	52					
	After 15 min	15	1	71.15		69.23	6.3
	After 30 min	15	1	71.15		69.23	6.3
	Filtrate 1	13	3	76.58		88.29	18.8
	After 10 min	4.4	0.2	66.15	93.33	64.62	4.3
	After 20 min	4.4	0.2	66.15	93.33	64.62	4.3
	Total removal			92.07		91.71	
	Mean concentration	55.5					
	Mean 15	14.5	2.5	73.87		69.37	14.7
	Mean 30 min	15	1	72.97		71.17	6.3
	Filtrate 1	13	3	76.58		71.17	18.8
	Mean second filtration	4.4	0.2	66.15	93.33	64.62	4.3
	Total removal			92.07		91.71	
17	Tank 1	210					
	After 15 min	61	0	70.95		70.95	0.0
	After 30 min	62	0	70.48		70.48	0.0
	Tank 2	220					

	After 15 min	63	0	71.36		71.36	0.0
	After 30 min	62	1	71.82		71.36	1.6
	Filtrate 1	60	1	72.09		86.05	1.6
	After 10 min	19	1	68.33	0.00	66.67	5.0
	After 20 min	19	0	68.33	100.00	68.33	0.0
	Total removal			91.16		90.93	
	Mean concentration	215					
	Mean 15	62	0	71.16		71.16	0.0
	Mean 30 min	62	0.5	71.16		70.93	0.8
	Filtrate 1	60	1	72.09		71.63	1.6
	Mean second filtration	19	0.5	68.33	50.00	67.50	2.6
	Total removal			91.16		90.93	
18	Tank 1	190					
	After 15 min	60	0	68.42		68.42	0.0
	After 30 min	61	0	67.89		67.89	0.0
	Tank 2	210					
	After 15 min	60	1	71.43		70.95	1.6
	After 30 min	60	1	71.43		70.95	1.6
	Filtrate 1	58	3	71.00		85.50	4.9
	After 10 min	18	0	68.97	100.00	68.97	0.0
	After 20 min	18	1	68.97	66.67	67.24	5.3
	Total removal			91.00		90.75	
	Mean concentration	200					
	Mean 15	60	0.5	70.00		69.75	0.8
	Mean 30 min	60.5	0.5	69.75		69.50	0.8
	Filtrate 1	58	3	71.00		69.50	4.9
	Mean second filtration	18	0.5	68.97	83.33	68.10	2.7
	Total removal			91.00		90.75	

Appendix 4

Table 34: Test results of experiment 27-30

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)
27	Tank 1	54	3
	After 15 min	10	0
	After 30 min	10	0
	Filtrate 1	10	0
	After 10 min	2.7	0
	After 20 min	2.6	0
28	Tank 1	50	4
	After 15 min	11	0
	After 30 min	11	0
	Filtrate 1	11	0
	After 10 min	3.1	0
	After 20 min	3.1	0.1
29	Tank 1	220	0
	After 15 min	57	-1
	After 30 min	54	1
	Filtrate 1	53	0
	After 10 min	17	-1
	After 20 min	16	0
30	Tank 1	200	10
	After 15 min	49	0
	After 30 min	50	0
	Filtrate 1	49	1
	After 10 min	15	0
	After 20 min	15	0

Table 35: Adjusted test results (experiment 27-30) and calculations

Experiment	Adjusted	$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of As(III) (%)	Removal rate of As(V) (%)	Removal rate of total As (%)	Ratio As(V)/ tot As (%)
27	Tank 1	54	3				5.3
	After 15 min	10	0	81.48	100.00	82.46	0.0
	After 30 min	10	0	81.48	100.00	82.46	0.0
	Filtrate 1	10	0	81.48	100.00	82.46	0.0
	After 10 min	2.7	0	73.00	-	73.00	0.0
	After 20 min	2.6	0	74.00	-	74.00	0.0
	Total removal:			95.09	100.00	95.35	
	Mean concentration	54	3				5.3
	Mean 15 min	10	0	81.48	100.00	82.46	0.0
	Mean 30 min	10	0	81.48	100.00	82.46	0.0
	Filtrate 1	10	0	81.48	100.00	82.46	0.0
	Mean second filtration	2.65	0	73.50	-	73.50	0.0
	Total removal:			95.09	100.00	95.35	
28	Tank 1	50	4				7.4
	After 15 min	11	0	78.00	100.00	79.63	0.0
	After 30 min	11	0	78.00	100.00	79.63	0.0
	Filtrate 1	11	0	78.00	100.00	79.63	0.0
	After 10 min	3.1	0	71.82	-	71.82	0.0
	After 20 min	3.1	0.1	71.82	-	70.91	3.1
	Total removal:			93.80	98.75	94.17	
	Mean concentration	50	4				7.4
	Mean 15 min	11	0	78.00	100.00	79.63	0.0
	Mean 30 min	11	0	78.00	100.00	79.63	0.0
	Filtrate 1	11	0	78.00	100.00	79.63	0.0
	Mean second filtration	3.1	0.05	71.82	-	71.36	1.6
	Total removal:			93.80	98.75	94.17	
29	Tank 1	220	0				0.0
	After 15 min	57	0	74.09	-	74.09	0.0
	After 30 min	54	1	75.45	-	75.00	1.8
	Filtrate 1	53	0	75.91	-	75.91	0.0
	After 10 min	17	0	67.92	-	67.92	0.0
	After 20 min	16	0	69.81	-	69.81	0.0
	Total removal:			92.50	-	92.50	
	Mean concentration	220	0				0.0
	Mean 15 min	57	0	74.09	-	74.09	0.0
	Mean 30 min	54	1	75.45	-	75.00	1.8

	Filtrate 1	53	0	75.91	-	75.91	0.0
	Mean second filtration	16.5	0	68.87	-	68.87	0.0
	Total removal:			92.50	-	92.50	
30	Tank 1	200	10				4.8
	After 15 min	49	0	75.50	100.00	76.67	0.0
	After 30 min	50	0	75.00	100.00	76.19	0.0
	Filtrate 1	49	1	75.50	90.00	76.19	2.0
	After 10 min	15	0	69.39	100.00	70.00	0.0
	After 20 min	15	0	69.39	100.00	70.00	0.0
	Total removal:			92.50	100.00	92.86	
	Mean concentration	200	10				4.8
	Mean 15 min	49	0	75.50	100.00	76.67	0.0
	Mean 30 min	50	0	75.00	100.00	76.19	0.0
	Filtrate 1	49	1	75.50	90.00	76.19	2.0
	Mean second filtration	15	0	69.39	100.00	70.00	0.0
	Total removal			92.50	100.00	92.86	

Appendix 5

Table 36: The uncertainty in the experiment 15-18, and 27-30 using the adjusted values

Experiment		$C_{As(III)}$ (µg/L)	$C_{As(V)}$ (µg/L)	Removal rate of total As (%)	Difference between the removal rate and the average removal rate	Standard deviation
15	Tank 1	52				
	After 15 min	13	0	75.00	4.58	3.17
	After 30 min	12	0	76.92	2.65	3.17
	Tank 2	51				
	After 15 min	12	0	76.47	3.11	3.17
	After 30 min	11	1	76.47	3.11	3.17
	Filtrate 1	12	0	76.70	2.88	3.17
	After 10 min	3.3	0.2	70.83	1.54	1.22
	After 20 min	3.3	0.1	71.67	0.71	1.22
	Total removal:	0		93.30	1.02	1.02
16	Tank 1	59				
	After 15 min	14	4	69.49	5.59	4.59
	After 30 min	15	1	72.88	2.20	4.59
	Tank 2	52				
	After 15 min	15	1	69.23	5.85	4.59
	After 30 min	15	1	69.23	5.85	4.59
	Filtrate 1	13	3	76.58	-1.50	4.59
	After 10 min	4.4	0.2	64.62	6.69	4.74
	After 20 min	4.4	0.2	64.62	6.69	4.74
	Total removal:			91.71	1.23	1.23
17	Tank 1	210				
	After 15 min	61	0	70.95	2.45	2.06
	After 30 min	62	0	70.48	2.93	2.06
	Tank 2	220				
	After 15 min	63	0	71.36	2.04	2.06
	After 30 min	62	1	71.36	2.04	2.06
	Filtrate 1	60	1	72.09	1.31	2.06
	After 10 min	19	1	66.67	2.26	1.34
	After 20 min	19	0	68.33	0.59	1.34
	Total removal:			90.93	0.90	0.79
18	Tank 1	190				
	After 15 min	60	0	68.42	4.50	3.40
	After 30 min	61	0	67.89	5.03	3.40
	Tank 2	210				
	After 15 min	60	1	70.95	1.97	3.40
	After 30 min	60	1	70.95	1.97	3.40

	Filtrate 1	58	3	71.00	1.92	3.40
	After 10 min	18	0	68.97	0.87	1.37
	After 20 min	18	1	67.24	2.59	1.37
	Total removal:			90.75	1.05	1.05
27	Tank 1	54	3			
	After 15 min	10	0	82.46	-2.88	3.17
	After 30 min	10	0	82.46	-2.88	3.17
	Filtrate 1	10	0	82.46	-2.88	3.17
	After 10 min	2.7	0	73.00	-0.63	1.22
	After 20 min	2.6	0	74.00	-1.63	1.22
	Total removal:			95.35	-1.02	1.02
28	Tank 1	50	4			
	After 15 min	11	0	79.63	-4.55	4.59
	After 30 min	11	0	79.63	-4.55	4.59
	Filtrate 1	11	0	79.63	-4.55	4.59
	After 10 min	3.1	0	71.82	-0.51	4.74
	After 20 min	3.1	0.1	70.91	0.40	4.74
	Total removal:			94.17	-1.23	1.23
29	Tank 1	220	0			
	After 15 min	57	0	74.09	-0.69	2.06
	After 30 min	54	1	75.00	-1.59	2.06
	Filtrate 1	53	0	75.91	-2.50	2.06
	After 10 min	17	0	67.92	1.00	1.34
	After 20 min	16	0	69.81	-0.89	1.34
	Total removal:			92.50	-0.67	0.79
30	Tank 1	200	10			
	After 15 min	49	0	76.67	-3.74	3.40
	After 30 min	50	0	76.19	-3.27	3.40
	Filtrate 1	49	1	76.19	-3.27	3.40
	After 10 min	15	0	70.00	-0.16	1.37
	After 20 min	15	0	70.00	-0.16	1.37
	Total removal:			92.86	-1.05	1.05