

ON TWO SIMPLE ARSENIC REMOVAL METHODS FOR GROUNDWATER OF BANGLADESH

A. H. Khan

Department of Chemistry, University of Dhaka, Dhaka-1000, Bangladesh.

S. B. Rasul and A. K. M. Munir

Sono Diagnostic Center Environment Initiative (SDCEI), Courtpara,
Kushtia, Bangladesh.

M. Alauddin

Chemistry Department, Wagner College, Staten Island, NY 10301, USA.

M. Habibuddowlah

Natural Resources and Environmental Protection Cabinet, Commonwealth of Kentucky,
Kentucky, USA.

A. Hussam*

Chemistry Department, George Mason University, Fairfax, VA 22030, USA

ABSTRACT

Two of the simplest arsenic removal methods are evaluated by measuring concentrations of As(III), As(total), Fe, Mn, Pb, Ca, Ba, Cu, Al, Zn, Cd, Na, K, Cl, ionic conductivity, E_h , pH, temperature and flow rate at various stages of the filtration process. The first method is simply leaving the groundwater for a few hours in the container and collecting the water by decanting. This method can be used to remove 50-70% arsenic from drinking water containing soluble iron. The efficiency of this method is discussed in relation to water chemistry parameters and chemical equilibrium models. The second method consists of a simple three-pitcher (locally known as '3-kalshi') filtration assembly made entirely from readily available local materials. In a 3- kalshi assembly, the first kalshi has iron chips and coarse sand, the second kalshi has wood charcoal and fine sand, and the third kalshi is the collector for filtered water. About 240 L of arsenic contaminated groundwater and groundwater spiked with high concentrations of both As(III) and As(V) were filtered.. It has been shown that more toxic As(III) can be removed from 800 ppb to below the detection limit of 2 ppb. The As(total) can be removed to a level below 10 ppb for most samples even at the highest input concentration of 1000 ppb As(total). The dissolved iron concentration decreased from an average 6000 ppb to 200 ppb. Calculations based on compound formation and arsenic adsorption on hydrous ferric oxide show that, with a constant input of dissolved iron the arsenic removal capacity increases linearly with each kalshi of filtration. The decrease in conductivity by 35% of the original value indicates substantial removal of dissolved ions. The final water quality was comparable to that of the guideline values suggested by World Health Organization and Bangladesh.

Key Words: Arsenic removal, groundwater, Bangladesh

INTRODUCTION

Recently, a high concentration of arsenic was found in the groundwater of Bangladesh and neighboring India (Chatterjee, et. al., 1995; Das et. al., 1995; Samanta, et. al., 1999). The geochemical nature of arsenic contamination of the aquifers of Bengal deltaic region is of serious concern because groundwater is the primary source of drinking water. Hyperkeratosis on the palms and feet is the common symptoms of arsenic poisoning. Long term exposure to low concentrations of arsenic has been reported to cause cancer of bladder, skin and other internal organs (Tseng, et. al., 1968; International Agency for Research on Cancer, 1980; Smith, 1992). Also, arsenic in its various ionic forms is known to be very toxic to most microorganisms (Cervantes, et. al., 1994). World Health Organization (WHO, 1999) reports that there are about 2.5 million tubewells in Bangladesh and more than 95% of the Bangladesh population of 120 million drinks well water. The present crisis may have affected more than 50 million of people in Bangladesh and the neighboring India. According to WHO, the maximum contamination level (MCL) of arsenic in drinking water is 50 $\mu\text{g/L}$ (or ppb: part-per-billion) and 10 $\mu\text{g/L}$ as a provisional guideline value. Total arsenic measurement shows that more than 60% of water from shallow and deep tubewells (wells with a metal casing) has above 10 ppb guideline value (International Conference, 1998).

The primary concern now is the measurement and purification of drinking water from arsenic. Because arsenic is found only in the well water, the obvious choice would be to drink surface water (river, pond etc.). However, this is also limited by the extensive contamination of surface water by fecal matters. Although Bangladesh has one of the world's largest freshwater resources, the lack of appropriate water treatment facilities prevents majority of the population from clean drinking water. An array of methods and technologies are available for the purification of water from arsenic and other trace metal contaminants (EPA, 1999). These are coagulation- filtration, lime softening, activated alumina, ion exchange, reverse osmosis, electro dialysis, nanofiltration and in-situ treatment of well water (Rott, et. al., 1993). The USEPA (United States Environmental Protection Agency) has identified three prospective methods: ion exchange with brine recycle, iron addition-coagulation with direct filtration, and attenuation by naturally occurring iron in water. Although, USEPA technologies are generally developed for large scale treatment facilities, the basic chemistry for the later method can be utilized in a small scale.

This work is motivated by the need to develop a simple, low cost technique for the removal of arsenic from the groundwater of Bangladesh by using locally available materials. Instead of using methods mentioned above, we decided to test two of the simplest water purification methods. The first method is simply leaving the groundwater for a few hours in the container and collecting the water by decanting. The efficiency of this method is discussed in relation to water chemistry parameters and chemical equilibrium models. The second method consists of a simple three-pitcher (locally known as '3-kalshi') filtration assembly made entirely from readily available local

materials. In a 3- kalshi assembly, the first kalshi has iron chips and coarse sand, the second kalshi has wood charcoal and fine sand, and the third kalshi is the collector for filtered water. This work is a scientific evaluation of the two methods for the purification of water and its viability in Bangladesh. The methods are evaluated by measuring the water chemistry parameters such as concentrations of As(III), As(total), Fe(total), Mn, Pb, Ca, Ba, Cu, Al, Zn, Cd, Na, K, Cl, ionic conductivity, E_h , pH, temperature and flow rate at various stages of the filtration process.

EXPERIMENTAL SECTION

In the first method, groundwater was left in polyethylene bottles without a headspace for 4 hours to 6 weeks. The same water was acidified with HCl and kept in similar bottles without a headspace. The water from both samples were analyzed for As(total) and other water chemistry parameters as described later. In The second method known as the 3-pitcher (hereafter referred as '3-kalshi', the local name) water purification setup is shown in *Figure 1*. It consists of three kalshis made of fired unglazed clay used as reservoir for drinking water by 80% of the people in Bangladesh. Local artisans make a variety of shapes of these kalshis. We have used the ones with narrow mouth, round bottom, and have a volume of about 18 L. The three kalshis are stacked on top of each other. The bottom of top two kalshis has a small hole covered with 100% polyester cloth or similar synthetic material. The first kalshi contains 3 kg (approx. 1/6 kalshi volume) of iron filings (chips) and 2 kg of coarse sand. The low carbon iron chips were obtained form a local foundry (Renwick Ironworks, Kushtia). Both coarse and fine sand was obtained from the local Garai River. The river sand has about 0.05% iron oxide. The second kalshi consists of 1 kg wood charcoal and 2 kg of fine sand. The wood charcoal was collected from burned wood for cooking. All precautions were taken to avoid the fine wood ash, which may dissolve in water and produce a basic solution. The function of the wood charcoal is to adsorb organic impurities that may be present in groundwater. The third kalshi was the collector.

The concentrations of As(III), and As(total) were measured by a Model HQ-2040 (Advanced Analytics, Virginia, USA) personal computer controlled electrochemical analyzer. This multifunctional computer controlled potentiostat is capable performing a variety of electrochemical experiments and has a magnetic stirrer controlled by the computer (Hussam, 1988; Hussam et. al. 1988). For arsenic analysis the instrument uses a staircase anodic stripping voltammetry (AS V) which has been shown to be more sensitive than differential pulse anodic stripping voltammetry (Christie, et. al., 1976; Eisner, et. al. 1976). Glass electrochemical cells with Teflon top and Teflon magnetic stir bar were used throughout the experiment. The top has provisions to insert Teflon purging tube and micropipet tips for standard additions.

Analytical protocols are based on published methods (Davis et. Al., 1978; Sun, et. Al., 1997; Hamilton, et. Al., 1980; Huiliang, et. Al., 1988) and that of a modified

EPA method 7063: Arsenic in Aqueous Samples and Extracts by Anodic Stripping Voltammetry (ASV) (Pyles, et. Al., 1999). In the method, As(III) is deposited from a 6M HCl acidic solution on a preplated gold coated glassy carbon electrode at -150 mV vs. Ag/AgCl, saturated KCl reference electrode for 60 s. The deposited arsenic is stripped off the electrode by a linear potential ramp from -150 to 500 mV in the same solution while the oxidation current is recorded as a function of the potential. The resulting linear scan anodic stripping voltammogram (LSASV) was used to measure the concentration of arsenic. The method of standard addition was employed to eliminate matrix effect of solution. The electrochemical method can be used to measure As(III) in presence of As(V) at all concentrations. Total As concentration was measured after reduction of As(V) to As(III) by Na₂SO₃. The analytical performance of the method can be summarized as follows: precision 1-10% relative standard deviation (rsd) of three replicate runs, accuracy 10% rsd (maximum) for 10 ppb quality control sample, three standard deviation of the signal detection limit 1.2 ppb at 95% confidence level at 120 s deposition time, dilution and aliquot addition errors 10% max, and sample carry over 0-4 ppb max. Details of the speciation analysis procedure are described elsewhere (Rasul, et. al. 1999; Rasul, et. al. 1999). The same instrument was used to obtain the redox potential, E_h, of water samples by measuring the rest potential of a polished platinum button electrode against the Ag/AgCl(s), satd. KCl reference electrode (197 mV vs. standard hydrogen electrode, SHE) and corrected for SHE.

Total soluble iron (Fe(II) and Fe(III)) was measured by visible spectrophotometric procedure based on 1,10-phenanthroline colored complex of Fe(II). The detection limit for soluble iron was 50 ppb. Standard digital meters measured total ionic conductivity, pH, and temperature. Na⁺, K⁺, and Cl⁻ were measured with ion selective electrodes on a serum electrolyte analyzer (Model-EasyLite Plus, M.I.T Services Inc., USA). The profile of trace metal ions was also measured by Inductively Coupled Plasma Spectroscopy (ICP) and Graphite Furnace Atomic Absorption Spectroscopy with Zeeman background correction (GFAASZ). The effluent flow rate was measured by collecting water in a measuring cylinder for a fixed time.

RESULTS AND DISCUSSION

The present experiment is performed with groundwater obtained from a tubewell in Kushtia Sadar, which has been continuously monitored for As(III) and As(total). About 40% of groundwater in Kushtia were contaminated with more than 50 ppb As(total). The water quality of the tubewell is representative of the groundwater used by about 400 thousand people of Kushtia Sadar (area 316 sq. km) and may have been used by many for years.

Table 1 shows the results for the first method where water samples are analyzed by GFAASZ for total arsenic.

The data shows that water left in the container without acid has lower arsenic concentrations. Visual examination of natural water left for a few hours shows brownish precipitate due to hydrous ferric oxide (HFO) while acid preserved water was clear. The acid preserved water shows the original concentration of arsenic in water. Arsenic concentrations analyzed after decanting the water shows reduction of arsenic by 50 to 70%. Therefore, simply leaving the water to precipitate out the arsenic with HFO did not remove arsenic below the MCL.

In the second method, two series of experiments were performed to evaluate the efficacy of the filtration system. In the first series, arsenic contaminated tubewell water and the same water spiked with higher concentrations of As(III) and As(V) were filtered. Iron and sand in A- kalshi were renewed in the second series of experiments. Also, the spiking level of As(III) and As(V) was increased. Table 1 shows the compilation of physicochemical data for first and second series of experiments. The data comprising temperature, pH, ionic conductivity, ppb of As(III), ppb of As(total), ppb of total soluble iron, and flow rate at various stages of the filtration process for both series of experiments. Redox potential, and concentrations of Na^+ , K^+ , and Cl^- were measured only in the second series of experiments.

In both series of experiments about 12 L of water passed through A and B and collected in C. In series 1, the first five experiments were done with tubewell water containing arsenic at indicated levels. Experiments were also performed by spiking tubewell water with sodium salts of arsenite, As(III), and arsenate, As(V).

Efficiency of arsenic removal. In both series of experiments As(III) was nearly completely removed from a maximum value of 800 ppb to below the detection limit of the instrument (ca. 2 ppb) for all influx. It appears that most of the As(III) is oxidized into more stable As(V) and precipitated in A and B kalshis. It has been recognized that As(III) is more prevalent in groundwater than was previously believed which is a concern because As(III) is more toxic than As(V) (Korte, et. al., 1991; Knowles, et. al., 1983). In Bangladesh, the groundwater contains 43-98% of arsenic in the form of As(III). For direct consumption, this is possibly one of the most toxic groundwater known today. Therefore, the removal of As(III) by any filtration procedure is crucial. In contrast, negligible removal of As(III) from drinking water was achieved by coagulation with alum (Hering, et. al., 1996). The overall arsenic removal capability is shown in *Figure 2*. In the first series of experiments the concentration of total As in the filtered water is reduced to 13.6 ± 3 ppb for all cases. This is much below 50 ppb and nearly 10 ppb within sample spread. The concentration of total arsenic in the filtered water is nearly independent of the initial concentration (79 - 723 ppb) is indicative of a

very efficient filtration mechanism. The second series of experiment started after leaving the filter assembly for a month without use. About 120 L of water was passed in each series of experiments. In the second series, the first two experiments were performed with the original tubewell water which show removal of As(total) below 10 ppb. This is indicative of a reproducible filtration process with new iron and sand. To further test the integrity and stability of the system, groundwater spiked with As(III) and As(V) were filtered. We measured that As(total) in the filtered water has decreased from 1005 ppb to 7 ppb with an average value of 6 ± 3 ppb for all samples. In series 1 and series 2, the As(total) removed were 44.0 mg and 72.5 mg respectively, for 120 L water filtered in each series.

Role of soluble iron in arsenic removal. Dissolved iron is a natural component of most groundwater. The dissolved iron concentration range in Bangladesh groundwater is 0.1 -7.0 mg/L (100 - 7000 ppbs) obtained from ICP measurement of 15 selected samples from Kushtia and near regions.

The maximum desirable concentration of iron in water is 300 ppb and the maximum permissible concentration is 1000 ppb (see Table 2). Besides causing pots and pans to become brown, at high concentration it can be toxic to small infants (Miah, 1996). The concentration of soluble iron originally present in the well water has decreased significantly, from 6000 ppb to a range of 0 - 480 ppb with an average of 200 ppb which is below the permissible level for most cases.

Dissolved iron, primarily present as Fe(II) in groundwater plays a very significant role in removing arsenic and other trace metals. In contact with air Fe(II) is oxidized to Fe(III) and precipitates as $\text{Fe}(\text{OH})_3$, hydrous ferric oxide (HFO: $\text{Fe}_2\text{O}_3 \cdot 2-3 \text{H}_2\text{O}$), $\text{Fe}(\text{HCO}_3)_2$ etc. Also, As(III) in contact with air and in the presence of zero valent iron, Mn^{2+} in ground water, and MnO_2 in the sand is catalytically oxidized to As(V) in the heterogeneous media at indicated pH range (Scott, et. al., 1995; Oscarson, et. al., 1980). It is well known that HFO binds arsenate formed during the slow percolation process (Sullivan, et. al., 1996; Seyler, et. al., 1989). In addition to As(V), recent data also shows As(III) is strongly sorbed by iron(III) oxides such as amorphous $\text{Fe}(\text{OH})_3$, HFO and Goethite (Manning et. al., 1998; Pierce, et. al., 1980). Arsenic bound to HFO can form common naturally occurring arsenate minerals $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$ (Scorodite) and $\text{FeHASO}_4 \cdot 8\text{H}_2\text{O}$ (Symplectite) as the dominant solid phase (Azcue, et. al., 1994). Recently, it has been shown that 0.267 mols of arsenic per kg of HFO are removed by sorption on HFO below pH 7.5 (Raven, et. al., 1998). Therefore, arsenic is removed by iron species either by compound formation or by adsorption or both. We also note that phyllosilicate, an abundant mineral component of sand has an affinity for arsenic at $9.62 \mu\text{g As(III)/g phyllosilicate}$ (Frost et. al., 1977). If phyllosilicate plays a significant role in removing As(III), then it may have removed a significant portion of As(III) in the first kalshi. However, saturation of phyllosilicate by iron and other ions in solution may limit its role as a sole agent for arsenic removal.

We find that there is excess Fe present in the groundwater than is required by stoichiometry of FeAsO_4 (s), other insoluble arsenic species or adsorption. The excess iron is accumulating in the filtering media (sand) after each kalshi of water is filtered. *Figure 3* shows the excess arsenic removal capacity due to the excess iron accumulating in the kalshi after filtration of each liter of water. *Figure 3* clearly shows that compound formation and precipitation as FeAsO_4 (s) are the most effective means of arsenic removal compared to adsorption. The compound formation (solid circle) is calculated on the basis of As/Fe mole ratio of unity and the adsorption calculation is based on the literature data (Pierce, et. al., 1982; Raven, et. al., 1998). The excess capacity due to compound formation or adsorption can decrease with decreasing soluble iron in the water. But with a constant input of soluble iron, naturally present or deliberately added, the capacity for arsenic removal increases linearly with each extraction for both cases. This calculation shows that this simple process without decreasing the efficiency of the system can remove very large quantities of arsenic.

Even with such increased capacity arsenic cannot be completely removed from water. The lower limit is possibly determined by other more soluble iron-arsenic species in equilibrium with solution. Factors such as leakage, mechanical bed failure due to dry bed, and bed clogging by hydrous ferric oxide may, however, limit the efficiency of the process in the long run.

Role of metallic iron. Elemental iron (or zero valent iron) is one of the most effective agents for environmental remediation of inorganic (Wilson, 1995) and organic (chlorinated solvents, nitroaromatics etc.) (Powel, et. al., 1995) species because it is a strong reducer. The use of metallic iron to remediate metal contaminated sites has increased because it is non-toxic and inexpensive. Literature data clearly demonstrate that zero valent iron can be useful for arsenic remediation at low pH and high sulfide containing water (Nikolaidis, et. al. 1998). The reducing property of metallic iron is most effective at low pH and slows down significantly at neutral pH. However, the hydroxide species formed on the metallic iron function as active adsorption sites for anions of arsenate and arsenite at neutral and basic pH. Although the exact role of zero valent iron in the present work cannot be quantitated, it is used to provide a constant input of iron (soluble or surface precipitate) for groundwater with low in soluble iron.

Redox potential change. To compare the redox quality of water produced by the filtration process electron activities are calculated and expressed by a nondimensional scale, pe using the relation (Morel, 1983; Hostettler, 1984): $pe = (F/2.303RT)E_h$. We observe that the outflow E_h is more oxidizing than the inflow water by 100 mV. The inflow water does not, however, represent the true groundwater condition because of its collection, spiking, and residence in the first kalshi during which some oxygenation occurred. Water in contact with air will have an E_h in the range of 350 - 500 mV and a pe between 5-8 are in agreement with the measured values (Wagemann, 1978). Due to large excess of soluble iron present in the inflow water, the

average inflow $E_h = 351 \pm 39$ and $\text{pH} = 6.8 \pm 0.12$ represents iron species $\text{Fe}^{2+}/\text{Fe}(\text{OH})_2^+$ in equilibrium. The 100 mV shift in the E_h and a pH shift to 7.7 for the filtered water shifts the equilibrium towards oxidizing condition where $\text{Fe}(\text{OH})_3(\text{s})/\text{Fe}(\text{OH})_2^+$ is the predominant equilibrium.

Effect on pH of water. The pH of the filtered water increased from 6.8 ± 0.1 to 7.8 ± 0.1 . This is caused by two factors (i) the 95% decrease in soluble iron concentration as a Lewis acid, and (ii) the increase in bicarbonate concentration due to increase in dissolved CO_2 (16-26 mg/L) from the ambient atmosphere after a prolonged contact time. The pH of the filtered water is almost the same as many bottled or spring water.

Removal of other ionic species. Measurement of ionic conductivity is essential to understand the overall filtration process with regard to soluble ionic components passing through the filtering media. The ionic conductivity of solution decreased consistently from A to B to C. On average a 25% decrease in conductivity occurred from A to B. This is indicative of a process where majority of Fe(II), Fe(III) and other metal ions are sorbed or precipitated in the first kalshi. The decrease in conductivity from B to C is about 12%. The decrease in conductivity in the final filtered water indicates that filtering media itself (sand in particular) did not dissolve or contribute to any excess ions in solution. This is further supported by the measurement of highly conductive ions Na^+ , K^+ , and Cl^- .

Flow rate and daily capacity. The flow rate of effluent during filtration is shown in the last column. The average outflow rate for both series of experiments is 27 ± 3 mL/min and did not decrease throughout the experiment in either A or B kalshi. This flow rate amounts to 1.6 L/hour or 39 L/day, which is adequate for the supply of drinking and cooking water for a middle size family of 4.

CONCLUSION

The first method of arsenic purification is not adequate for obtaining water with arsenic below MCL. However, it is a method where no other means for water purification are available. This method should be considered with caution. On the other hand, we show that arsenic in groundwater can be removed below 10 ppb by a simple 3-kalshi filtration procedure using locally available material and without adding any chemicals. The overall water quality obtained from the 3-kalshi setup is compared with that of standards in **Table 2**.

Clearly, the water quality obtained from 3-kalshi setup meets the WHO and Bangladesh standard and made potable water from contaminated water of near wastewater quality. The present system can be further optimized to increase the flow rate, the

efficiency, and the aesthetics for a wider acceptance and use. In this respect local innovation and local participation are essential. We urge immediate use of the 3-kalshi method to mitigate part of the present arsenic crisis.

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TABLE 1
Total arsenic concentration with method 1 for the purification of groundwater

Sample id	Sample characteristics	As(total), ppb
2BN	Natural tubewell water, decanted	39
2BN	Natural tubewell water, shaken	50
2BA	Natural tubewell water, acid preserved	158
5BN	Spiked tubewell water, decanted	194
5BN	Spiked tubewell water, shaken	191
5BA	Spiked tubewell water, acid preserved	552
9BN	Spiked tubewell water, decanted	358
9BN	Spiked tubewell water, shaken	378
9BA	Spiked tubewell water, acid preserved	776

TABLE 2
Drinking water Inorganic Quality Parameters: Comparison of 3-Kalshi Water with Limits Set by World Health Organization (WHO) and Bangladesh Standards. ^a

Constituent	WHO	Bangladesh	3-Kalshi water
Arsenic (total)- mg/L	0.05	0.05	0.01
Iron (total) - mg/L	0.3	0.3 (1.0)	0.07 - 0.49
pH	6.5-8.5	6.5-8.5	7.74 ± 0.1
Sodium - mg/L	200		107 ^b
Potassium - mg/L			8.2
Chloride - mg/L	250	200 (600)	130 - 350
Total dissolved solids - mg/L	1000	500 (1500)	208
E _h (mV vs. SHE)			454 ± 54
Conductivity - μS/cm			416 ± 40

a. WHO guideline values. Bangladesh standard values are given as maximum desirable concentration with maximum permissible concentration in parentheses. TDS for 3-kalshi was calculated from the conductivity data excluding silica present in the filtered water. b. Provisional value.

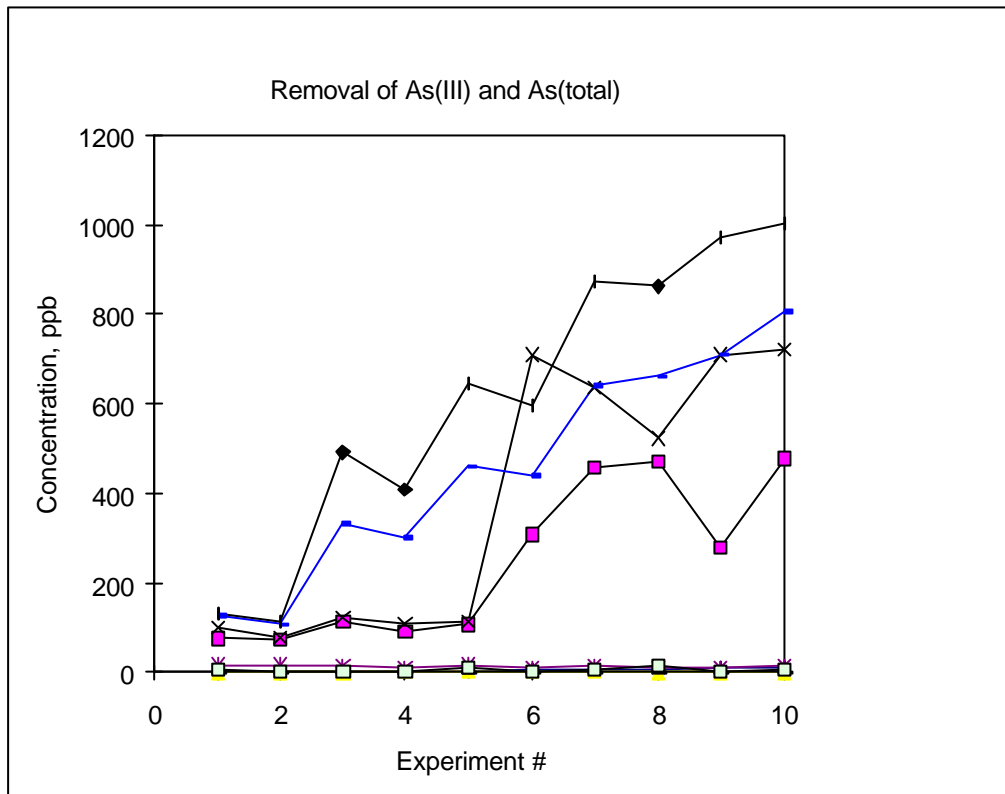


Figure 2. Efficiency of arsenic removal by 3-kalshi method. Series 1 experiment: X - As(total), ~ -As(III). Series 2: diamond: As(total), arrow - As(III) for input water. The filtered water concentrations for both series of experiments are shown near zero, parallel to abscissa.

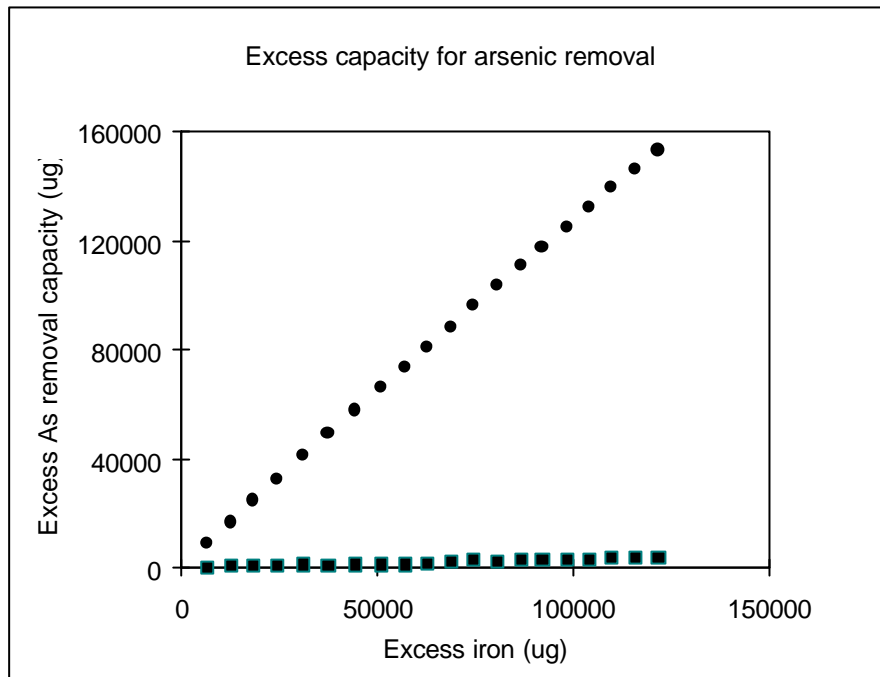


Figure 3. Plot showing excess capacity for arsenic removal by the 3-kalshi method. Capacity calculation is based on formation of (●) - $\text{Fe}(\text{AsO}_4)$, (■) - adsorption data.