

Arsenic Removal from Groundwater using Ferric

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Abstract

Arsenic is a carcinogenic metalloid that is currently regulated in drinking water. The levels of arsenic in finished water in an existing water treatment plant are exceeding the current regulation of 10 µg/L. One of the available technologies for arsenic removal from groundwater is adsorption onto coagulated flocs and in this field, ferric chloride is the most commonly used coagulant for arsenic removal. This research work was conducted to explore a suitable conventional treatment technology for arsenic removal from given groundwater in order to reduce the filtrate arsenic concentration to less than 10 µg/L.

Bench scale jar test experiments and pilot-scale investigations were carried out to evaluate and improve the coagulation/flocculation process for arsenic removal using ferric chloride. Model water that represented the water from the existing water treatment plant was used to investigate the effects of different conditions of pH, coagulant doses, arsenic speciation, initial arsenic concentration, temperature, and flocculation conditions on the arsenic removal efficiency by coagulation/flocculation process. Based on these bench scale experiments, a direct filtration technique to separate the formed flocs was considered as the most suitable floc separation system to be applied after coagulation/flocculation process. A direct filtration pilot plant was operated to evaluate the efficiency of arsenic removal.

The results of series of jar test experiments showed that As(V) could be completely removed with iron doses higher than 2 mg/L for filtered samples and at pH value about 7.0. The lower efficiencies obtained for unfiltered samples indicate that settling mechanisms are not effective enough to ensure complete removal of As(V), even when using very high doses of coagulant. In agreement with the results of previous studies, it was found that As removal efficiency increased with the coagulant dose. Additionally it was also observed that under the given conditions As(III) removal efficiency was much lower (up to 60%) compared to As(V) removal efficiency (90 - 100%).

Direct filtration with iron doses of 2 mg/L at pH value about 7.0, could reduce As(V) levels from 50 µg/L to 4 µg/L or less without any risk of iron or turbidity increasing in the filtered water within a reasonable filterrun length.

Direct filtration using ferric chloride as coagulant, could be an appropriate technology to reduce arsenic levels below 10 µg/L for the given groundwater.

Key words:

arsenic removal – groundwater - ferric chloride – coagulation – flocculation – direct filtration.

1. Introduction

Arsenic (As) is the 20th most abundant element present in the earth's crust. It is a common pollutant in groundwater and industrial wastes. It may be under mineral form and mainly as impurities from other materials. Moreover, it may be present in water, air and living beings (sea-living species)

Due to its oxidation state (As III or As V), it will represent more or less risk for the human health. Most of the arsenic present in the nature is in the As(V) form, which has relatively low toxicity. However in contact with the air it generates oxidised forms which are more toxic. The health effects have a large diversity, from skin pigmentation, skin damages to skin and respiratory cancer.

Arsenic may be found in water, which has flowed through arsenic-rich rocks. Severe health effects have been observed in populations drinking arsenic-rich water over long periods in countries world-wide (WHO, 2001)

One of the available technologies for arsenic removal from groundwater is adsorption onto coagulated flocs (Hering, 1996-1997; Edwards, 1994; Chen et al, 1999). Alum and ferric salts are examples of coagulants in use. However, there are some uncertainties regarding to the best coagulation process to be used when arsenic is present. Research works on available technologies for arsenic removal reported that ferric chloride seems to be the most appropriate coagulant to be used for arsenic removal. (Gupta & Cheng, 1978; Cheng et al, 1994; Hering et al, 1997) When a technology for arsenic removal must to be chosen, some limiting factors must to be considered. In the present research work ferric chloride is used as coagulant (although there are many others) and since the results of this research will be applied to a real case, the downstream process carried out in the plant must be also considered.

1.2 Objectives

The goal of this research work is to propose a suitable conventional treatment technology for arsenic removal from given groundwater in order to reduce the filtrate arsenic concentration to less than 10 µg/L. This study, based on bench scale and pilot scale experiments, outlines some preliminary design parameters for full-scale treatment plant. The proposed technology should be able to achieve a removal efficiency that can fulfil the current regulations, and to anticipate the new restriction planned to the Maximum Contamination Level (MCL) of arsenic.

The specific objectives of this research work are:

- **To optimise conditions for coagulation/flocculation process using Ferric Chloride for the given groundwater.**
- **To examine, at pilot scale, the performance of direct filtration to minimise residual As concentration.**

2. Literature Review

Arsenic Removal Technologies

Various treatment methods have been adopted to remove arsenic from drinking water. These methods include co-precipitation, sorption techniques and membranes filtration techniques. Most of them can be implemented at domestic, small, medium and large sized facilities. The technologies under review perform most effectively when treating arsenic in the form of As(V). Moreover As(III) must be converted through pre-oxidation to As(V) before these methods can be effectively used. The following table shows the efficiency of different methods. It is clearly appreciated that the pentavalent form of arsenic is easier to remove than the trivalent one.

Table 1. Efficiency of As removal using different technologies.

As form	Coagulation		Ion Exchange	Adsorption (Alumina, Carbon)	Reverse Osmosis	Electrodialysis
	Al ₂ (SO ₄) ₃	FeCl ₃				
As (III)	0-20%	40-70%	20-40%	40-70%	70-80%	70-80%
As (V)	70-80%	80-100%	80-100%	80-100%	80-100%	80-100%

(Source: Chwirka et al, 1999)

Arsenic removal by coagulation

Speciation / Oxidation

Inorganic As speciation is an important factor in the efficiency of the process of removal of arsenic from water. As(V) is much more effectively removed than As(III) because the former exists in natural waters as mono or bivalent anion and the latter exists, predominantly, in an anionic form. For this reason, previous oxidation of As(III) to As(V) may be an important stage of the treatment process in order to achieve more efficient removal (Sancha, 2000).

The coagulation technology is based on the principle that As(V) adsorbs onto coagulated flocs which are then removed from solution by filtration. Coagulation is an effective treatment process for removal of As(V). The type of coagulant and dosage used affects the efficiency of the process. Aluminium and iron salts can be used as coagulants to achieve efficient As(V) removal (Hering et al, 1997).

Although the potential to obtain high efficiency arsenic removal by coagulation is unquestioned, an understanding of the mechanisms and optimisation strategies is currently lacking. Addition of iron or aluminium coagulants to water can facilitate the conversion of soluble As(V) and As(III) into insoluble reaction products. These products might form through Precipitation, Co-precipitation or Adsorption mechanisms.

Regarding the coagulant dosages it is illustrative to present the dosages required to obtain a given effluent arsenic concentration. Edwards (1994) reported that for an initial As(V) concentration of 20 µg/l, a coagulant dose of 7 mg/l as FeCl₃ (14 mg/l as alum) would meet a 10 µg/l standard for most samples. However, a wide range of dosages-removal values could be referred since several factors such as turbidity, pH, initial concentrations and other ionic compounds present in the water to be treated affect the arsenic removal efficiency.

Flocs separation systems

Coagulation/flocculation can be only used successfully when the flocs which are formed can be separated out reliably and when most of the coagulant is also removed. In general, coagulation/filtration method of arsenic removal could mainly be of the following three types: (Fields et al, 2000; Chwirka et al, 2000; Madiec et al, 2000; Jekel & Seith, 2000; Sancha, 2000)

coagulation, sedimentation, rapid sand filtration, direct filtration (coagulation – rapid sand filtration), coagulation followed by microfiltration.

Factors affecting arsenic removal by coagulation

The efficiency of arsenic removal by coagulation filtration process depends on water quality and process conditions applied. The water quality includes pH, temperature, initial arsenic concentration and speciation and the presence of other competing ions. The process conditions include type and dose of coagulant, flocculation conditions and method of floc separation applied.

Other Ions

Adsorption of As(V) and As(III) oxyanions by ferric chloride may be adversely affected by anions such as phosphate, silicate, sulphate, carbonate and natural organic matter. The effects of competing ions in arsenic removal can be summarised as follows (WHO, 2001)

- effects of co-occurring solutes will be more pronounced when adsorption density is nearing saturation.
- As(III) makes a weaker bond than As(V) with metal oxides, and is thus more likely to be displaced by competing anions.
- Surface complexation chemistry is complex, particularly when multiple anions are present.

3. Materials and Methods

The research work was carried out in two phases. Firstly bench scale tests were performed to represent different coagulation conditions by means of a jar-test. Subsequently based on the results obtained from the jar-test experiments, a pilot plant representing a direct filtration process was also evaluated.

3.2 Bench studies

Experimental set up

A series of 20 jar tests was performed for arsenic removal evaluation from November 2001 to February 2002. The main objective of the series was to find the optimum conditions that could achieve the maximum arsenic removal efficiency. The experiments represented the present conditions at second filtration step in the treatment line, so, the model water to be analysed should have the same characteristics as the samples obtained at the outlet of filters 1 and 2 (RSF 1+2). To fulfil this aim model water was prepared from demi-water by adjusting some parameters as shown in the following table:

Table 2. Adjusted parameters to represent case study water.

Parameter	Unit	RSF (1+2)	Model Water	By adding
Temperature	°C	27	20/30	Heater
pH	mg/L	6.7 - 7.1	7.50	HCl
Calcium	mg/L	40	40	CaCl ₂ .2H ₂ O
Magnesium	mg/L	13	13	MgCl ₂ .6H ₂ O
Bicarbonate	mg/L	225	225	NaHCO ₃
Arsenic	µg/L	50	50	As ⁵⁺

Jar Test apparatus

The jar-test experiments were conducted using a modified Jar Test apparatus containing six jars of 1.8 litres. It consists of six paddle stirrers that keep a constant uniform stirring in all beakers. The device is provided with a double wall to let warm water pass through to allow experiments at different temperatures. The glass jars contain baffles to avoid vortices and a vacuum system that allows to take samples of the supernatant of each beakers at the same time. The different dosages of coagulant were added to all jar tests in parallel.

Standards and reagents

All chemicals were reagents-grade and were used without purification. All solutions were prepared using de-mineralised water. All glasswares were acid-washed. As(V) standard was prepared from As₂O₅ and As(III) standard was prepared from As₂O₃, both dried at 105°C and kept in dessicator. Primary stock solutions for As(V) and As(III) (1000 mg/L) were prepared, from which a secondary stock (10 mg/L) was prepared to make the model water. For

coagulation experiments, ferric chloride stock solution (0.1 M) was prepared from $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and stored in a dark bottle, the pH of this solution was maintained below 1.70. Background electrolyte solutions were prepared from the salts $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, NaHCO_3 and stock solutions (0.1 M) were used for addition to de-mineralised water to make model water. HCl was used to adjust the pH of the model water. A vacuum device with 0.45 μm pore size membrane filter was used to obtain filtered samples from the Jar Test experiments.

Experimental equipment

Jar test set up, Electronic balance of sensitivity 0.00001 g, Vacuum flask assembly, pH meter (WTW pH 340), Turbidimeter (Lange LTP4)

Process conditions

Mixing conditions: For all jar tests performed in this research work rapid mixing conditions (300 rpm, $G = 750$ 1/sec, $t = 1$ min) was applied to ensure the uniform mixing of the coagulant into the beakers.

Flocculation conditions: Three flocculation conditions were used in this research work. For jar tests a G value lower than 10 1/sec was applied for those jar tests where effects of different equilibrium pH was evaluated. Another G value (50 1/sec) was used for evaluation of effects of arsenic speciation on removal efficiency. At last, a tapered flocculation conditions was applied as follow: $G = 70$ 1/sec, $t = 5$ min + $G = 50$ 1/sec, $t = 5$ min + $G = 20$ 1/sec, $t = 10$ min.

pH conditions: A total of ten pH conditions were tested in this study. The equilibrium pH value (pH measured after settling) was ranged between 6 and 8.

Arsenic speciation: Different jar tests were performed using As(V) spiked model water and As(III) spiked model water. The pH for these jar tests was adjusted at 7.0 and the initial concentration for both arsenic species was around 50 $\mu\text{g/L}$.

Temperature conditions: All jar test were performed at 20°C, except one of them performed at 32°C in order to compare As(V) removal efficiency at different temperature conditions.

Initial As(V) concentration conditions: An experiment to observe the influence of initial arsenic concentration on arsenic removal efficiency was carried out. The coagulant dose used in that case was 6.9 mg/L Fe^{3+} (20 mg/L FeCl_3) and pH was maintained at 7.0.

Settling conditions for As(III): In order to observe an improvement in the As(III) removal efficiency an experiment with a longer settling time was performed. The settling time was increased from 30 min to 2 hours.

Fe^{2+} present in model water: Preliminary experiment was carried out in order to observe arsenic removal efficiency in waters containing ferrous iron at high concentration. The reagent used to make this model water was ferrous sulphate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) and the initial concentrations were fixed at 2, 5, 10 and 20 mg/L Fe^{2+} .

Experimental procedure

The general experimental procedure involved adding ferric chloride coagulant to arsenic containing model water, applying a certain G value by means of stirring, settling and taking samples (filtered and unfiltered ones). The experiments differed in the G value applied, the initial and final pH, arsenic oxidation state, the initial arsenic concentration and the

temperature conditions. The doses of ferric chloride were in the range from 2 mg/L to 40 mg/L (0.7 mg/L to 13.8 mg/L Fe^{3+}) for a given jar test condition.

The coagulants were added to each jar, mixed at 300 rpm ($G = 750$ 1/sec) for 1 min, flocculated (for standard conditions) at 50 rpm ($G = 50$ 1/sec) for 20 min, and 30 min quiescent settling. After the settling period, approximately 300 mL of solution was collected from 3 cm below the liquid surface; 100 mL of this solution was then filtered through a 0.45 μm membrane filter using a vacuum filter flask assembly.

The pH was adjusted in the model water prepared specifically for each jar test. It was measured in the container of model water, after addition to the beakers (initial pH) and after settling (final or equilibrium pH). The different jar tests were characterised and named by the equilibrium pH.

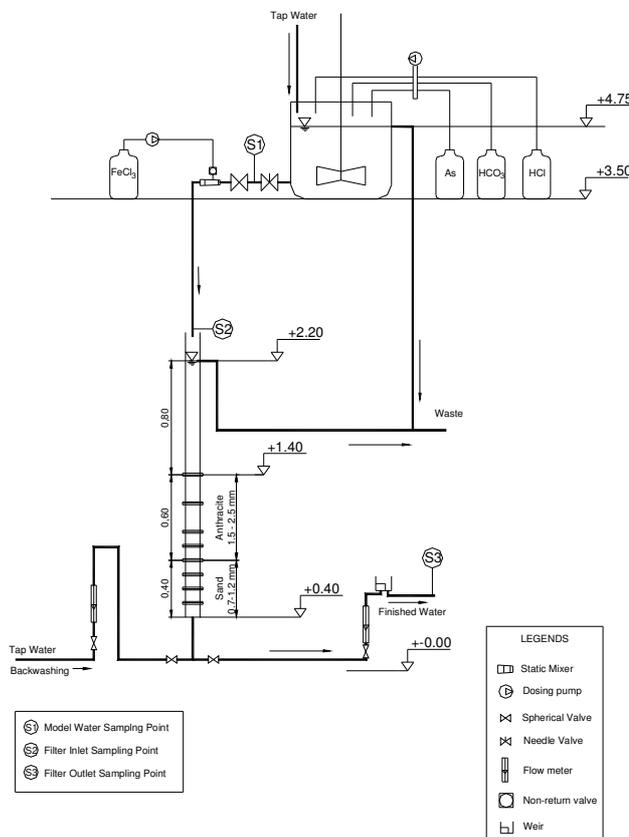
Parameters to be analysed

For both filtered and unfiltered samples from each beaker and model water, total iron, total arsenic and turbidity were determined.

3.3 Filtration column studies

Experimental set up

From the jar tests experiments it was clear that high arsenic removal efficiencies can be achieved using ferric chloride as coagulant for the given groundwater under lab conditions, when the flocs are separated by means of filtration through 0.45 μm pore size membrane. Thus, a direct filtration process to achieve the flocs separation was decided to be carried out to collect operational data on this filtration process. The main goal of the filtration column studies was to analyse the feasibility of applying *coagulation-direct filtration process* under similar and extreme conditions regarding to the case study treatment plant.



The model water used for filtration experiments was from tap water spiked with different chemicals to represent filtrate characteristics of RSF (1+2) in the existing GWTP. The coagulant was dosed in line to the stream before it passed to the filter (see figure).

Column set up

The pilot plant consist of one filter column, 100 mm in diameter, with associated flow measuring and regulating devices and chemical dosing and mixing equipment (in-line static mixer). The filter column was provided of sampling points and manometer points along its length for measurement. The filter was operated with a constant supernatant depth of 0.80 m. A constant filtration rate (6 m/h) was

maintained with help of flow regulating hand-operated needle valve and rotameter installed on filtrate line. The filter was operated for a run length of approximately 8 hours before backwashing.

The model raw water was prepared in a 100 L plastic tank with continuous mixing at 200 rpm. The addition of HCl, NaHCO₃ and As were performed by means of peristaltic pump WT 503S and for the addition of ferric chloride a peristaltic pump WT 101 U/R was used. Fresh stock dilutions of 4 L of chemicals and fresh stock dilution of 1L of ferric chloride were prepared for each run.

Process conditions

The pilot plant was operated under the following conditions:

pH inlet: 6.8 to 7.1
Water Temperature: 20°C
Backwash frequency: 8-10 hours
Break-through by: head losses
Filter bed depth: 1 m (0.6 m anthracite + 0.4 m sand)
Coagulant dose: 2 mg/L Fe³⁺
Filter control: constant raw water level, constant filtration rate.
Backwash: Media expansion: 15%, V_B: approx. 60 m/h, t: 6 min.

Experimental procedure

The general experimental procedure consisted of running the filter with certain conditions of pH and coagulant dose, taking samples from the sampling points (see scheme pilot plant) every hour, analysing turbidity, arsenic and iron concentration. At the beginning of the run (first hour, ripening period) the samples were taken every 20 min. After the daily run was finished, a backwashing procedure with tap water was applied. It consisted of 6 minutes of stabilised velocity of approx. 60 m/h which guaranteed an expansion of the filter from 15 to 20 %.

Analytical methods

Arsenic samples were analysed by means of an Atomic Absorption Spectrometer Graphite Furnace (AAS-GF), Perkin-Elmer 1100 B, Range of measurement: 0 to 60 µg/L (of total As), that is As(V)+As(III), Detection Limit (for total As): 4 µg/L, Sensitivity: +- 3 µg/L. Iron, calcium and manganese were measured using an AAS-Flame Perkin Elmer 3110, with detection limit 0.03 mg/L for Fe (Standards Methods, 1995).

4. Results and Discussion

4.1 Bench studies

Effect of pH

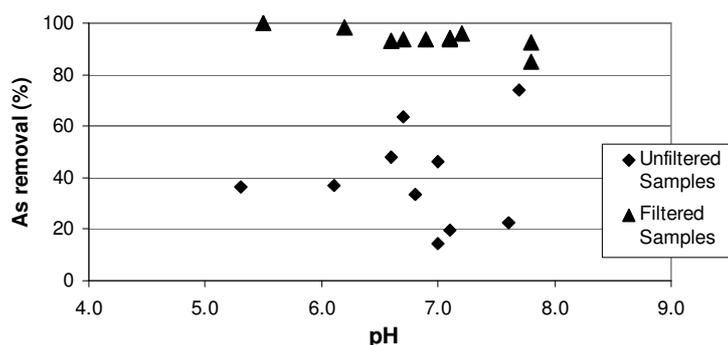


Figure 1. As(V) removal efficiency. (Initial As(V) concentration: 40 – 50 µg/L / pH equilibrium: 5.5/7.8 - Temperature: 20°C / Dose of Fe³⁺: 3.4 mg/L (10 mg/L FeCl₃) – G < 10 l/sec)

Effect of initial arsenic concentration

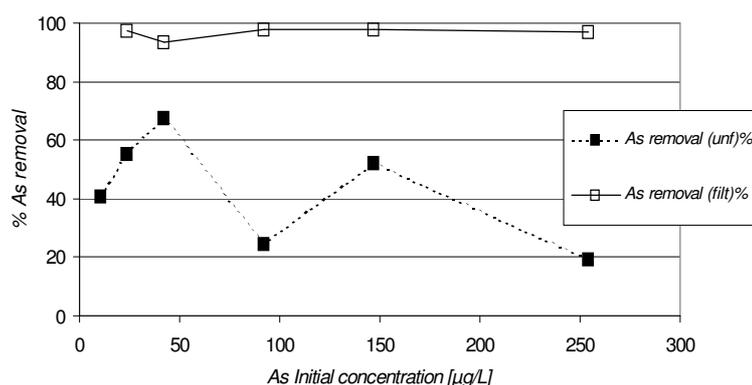


Figure 2. As(V) removal efficiency for different initial concentrations. (Initial As(V) concentration: 10 – 250 µg/L / pH equilibrium: 7.0 - Temperature: 20°C / Dose of Fe³⁺: 6.9 mg/L (20 mg/L FeCl₃) – G < 10 1/sec)

Over the initial arsenic concentration range examined, removal efficiencies were independent of initial As(V)

concentration. This result is consistent with previous works (Cheng et al, 1994; Hering et al, 1997). This lack of dependence is consistent with the expected adsorption behaviour of arsenic when the surface of freshly precipitated amorphous metal hydroxide formed upon coagulant addition is not saturated.

Effect of coagulant dose and arsenic speciation

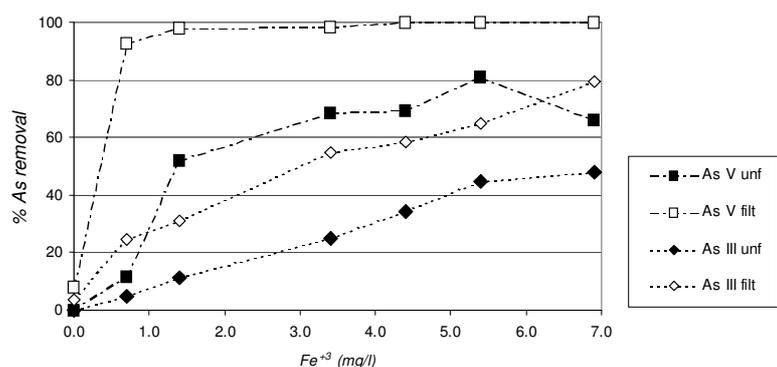


Figure 3. As(V) removal efficiency for different dose of Fe³⁺. (Initial As(V/III) concentration: 50 µg/L / pH equilibrium: 7.0 - Temperature: 20°C - G = 50 1/sec)

It is shown in Figure 3 that As(V) removal by ferric chloride was improved by increasing coagulant dose. At a coagulant dose of 0.7 mg/L Fe³⁺, As(V) removal was 93% for filtered samples. Complete removal of As(V) was achieved for doses above 4.0 mg/L Fe³⁺, that is consistent with previous research work (Hering et al, 1996). A removal better than 97% could be reached for filtered samples with a coagulant dose higher than 2.0 mg/L Fe³⁺. This value is relevant for the present study since an efficiency of 90% is required to be reached. For lower values of coagulant doses (< 1.0 mg/L Fe³⁺), the removal efficiency increases rapidly (from 0 to 80%) and does not guarantee a constant and reliable data to be applied. With respect to unfiltered samples, it is clear that also an improvement in removal efficiency can be achieved by increasing coagulant dose. A removal efficiency higher than 60 % could be achieved for As(V) with a settling time of 30 minutes and for coagulant doses higher than 2.0 mg/L Fe³⁺.

4.2 Filtration column studies

A total of 6 runs were performed in the filtration column studies described in section 3.3. From those runs, two of the most representative ones were analysed and the average values are informed in the charts as follows.

Overall behaviour of direct filtration set up

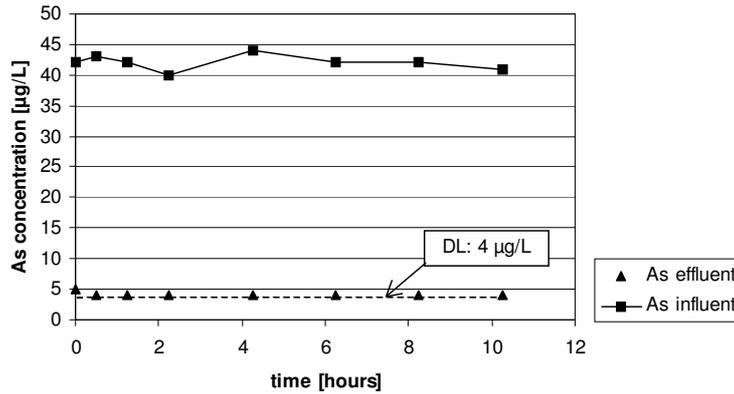


Figure 4. As(V) removal efficiency during direct filtration experiments. (Inlet As(V) concentration: 42 µg/L / pH model water: 6.6/7.1 - Temperature: 20°C / Dose of Fe³⁺: 2.4 mg/L - Filtration rate: 6 m/h)

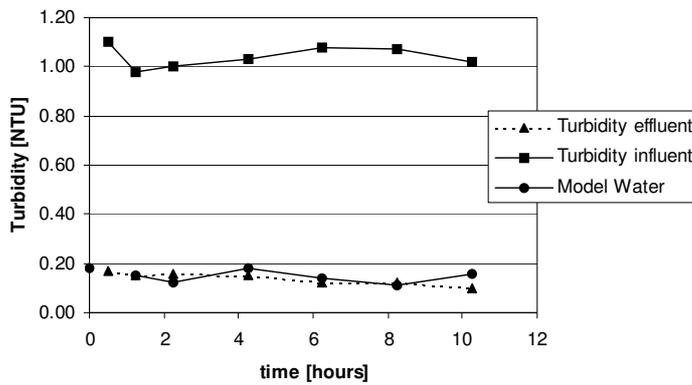


Figure 5. Turbidity during direct filtration experiments. (Inlet As(V) concentration: 42 µg/L / pH model water: 6.6/7.1 - Temperature: 20°C / Dose of Fe³⁺: 2.4 mg/L - Filtration rate: 6 m/h)

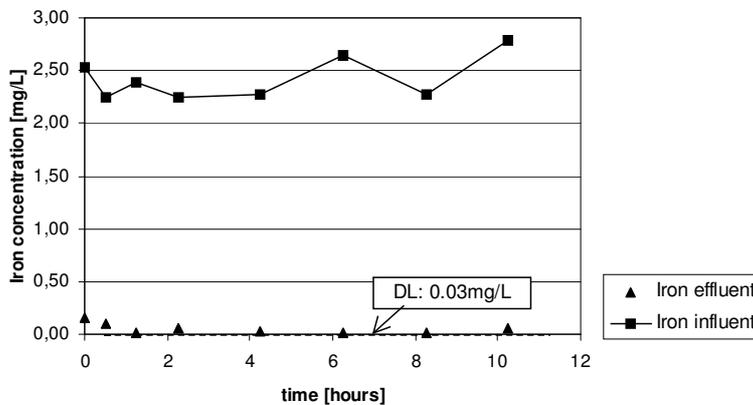


Figure 6. Iron concentration during direct filtration experiments. (Inlet As(V) concentration: 42 µg/L / pH model water: 6.6/7.1 - Temperature: 20°C / Dose of Fe³⁺: 2.4 mg/L - Filtration rate: 6 m/h)

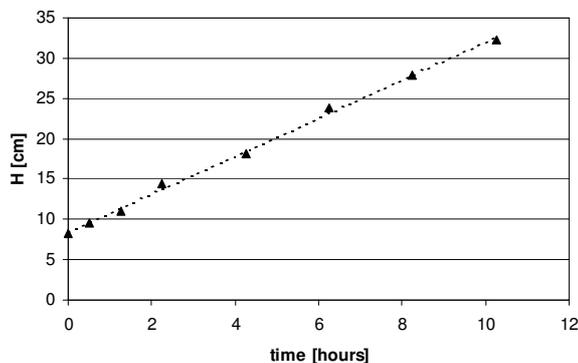


Figure 7. Head loss development. (Inlet As(V) concentration: 42 µg/L / pH model water: 6.6/7.1 - Temperature: 20°C / Dose of Fe³⁺: 2.4 mg/L - Filtration rate: 6 m/h)

5. Conclusions and recommendations

An increase in pH value from 5.5 to 8.0 had no significant effect on As(V) removal efficiency. However, optimal pH value was close to 7.0. During coagulation / flocculation experiments, As(V) removal was independent of initial As(V) concentration. This is helpful when the WTP deals with wells with different raw water quality. Under similar conditions of pH, temperature, coagulant dose and flocculation, As(V) is easier to be removed than As(III) for the given model water. At the lowest coagulant dose applied for filtered samples (1.4 mg/L Fe³⁺), As(V) removal efficiency was higher than 95 % at pH value 7.0. At the highest coagulant dose applied for unfiltered samples (13.8 mg/L Fe³⁺), As(V) removal efficiency was higher than 90 % at pH value 6.8, with a settling time of 30 minutes. However iron concentration in supernatant was above 1 mg/L. Marginal improvement was observed in As(V) removal when water temperature was increased from 20°C to 32°C, for unfiltered samples. There was no detectable improvement in As(V) removal efficiencies for different flocculation conditions applied (tapered flocculation and constant G value). It was not possible to achieve complete As(V) removal by coagulation-sedimentation process without exceeding iron concentration of 0.1 mg/L in supernatant. Complete removal of As(V) was achieved for iron doses higher than 2 mg/L in samples filtered through 0.45 µm membrane. That suggests direct filtration or microfiltration process as feasible technique to achieve residual As(V) lower than 5 µg/L. Pilot tests results indicate that under these conditions (pH inflow 7.0, filtration rate 6 m/h, initial As(V) 50 µg/L, iron doses 2 mg/L, length of filter run 10 hours) arsenic removal efficiency was higher than (< 4 µg/L) 90%. With regards to iron concentration in the filtrate it was consistently lower than 0.10 mg/L. Direct filtration could be an appropriate technique to achieve efficient arsenic removal, specifically when the formed flocs are difficult to be removed by settling.

Optimisation of filter bed (depth, composition), filter rate and backwashing conditions must to be investigated. Direct filtration experiment should be conducted at higher initial As (for example 200 µg/L) in order to assess the capability limit of the direct filtration process for arsenic removal.

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