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DOI
10.1016/j.jwpe.2017.12.003

Publication date
2018

Document Version
Final published version

Published in
Journal of Water Process Engineering

Journal of Water Process Engineering 21 (2018) 107–115

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Biological As(III) oxidation in rapid sand filters

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ARTICLE INFO

Keywords:
Groundwater treatment
Rapid sand filtration
Arsenic removal
Biological As(III) oxidation

ABSTRACT

The objective of this study was to investigate whether arsenic-oxidising bacteria (AsOB) will grow and survive in rapid sand filters. Additionally, the interdependence of other groundwater constituents (Fe(II), Mn(II), NH_4) with biological As(III) oxidation was investigated. For this purpose As(III) oxidation was monitored in pilot-scale filter sand columns fed with raw groundwater, as well as treated groundwater (drinking water) with spikes of either As(III), Mn(II) or NH_4.

It was concluded that biological As(III) oxidation rapidly developed in the rapid sand filter columns. With a typical lag and log phase, decreasing As(III) and increasing As(V) concentrations in the effluent of the sand columns were observed in a timeframe of weeks. The growth of biomass in the sand columns was confirmed with ATP analysis. ATP concentrations on the sand grains increased from 0.7 ng/g to 16, 8 and 2 ng/g filter sand stratified from the top of the sand filter to the bottom, respectively. Additionally, a microbial community analysis (16S rRNA) showed a high relative abundance of α- and β-Proteobacteria; the same classes where most AsOB are phylogenetically placed.

This study establishes that AsOB are able to grow and maintain their population on low As(III) concentrations, either in presence, or absence, of other common groundwater bacteria and mineral precipitates, directly leading to an increased As removal in the filter bed.

1. Introduction

Conventional groundwater treatment plants for drinking water production, consisting of aeration and rapid sand filtration, are primarily designed for removal of dissolved iron (Fe), manganese (Mn) and ammonium (NH_4). However, when present, arsenic (As) is often only partially removed by this technology, potentially affecting drinking water safety. Therefore, drinking water companies in the Netherlands are currently considering a new target of 1 μg/L. To prevent costly modifications to an otherwise simple treatment set-up, better understanding of As removal mechanisms in rapid sand filters needs to be gained.

Prior to rapid sand filtration, anaerobic groundwater is aerated with spray or cascade aeration, equilibrating the water with oxygen (O_2), and removing (part of the) carbon dioxide (CO_2), hydrogen sulphite (H_2S) and methane (CH_4). O_2 facilitates the oxidative removal of Fe, Mn and NH_4 in the subsequent filter bed. The aerated water is supplied to the supernatant water of a rapid sand filter, that typically consists of a sand bed with a height of 1.5–2.5 m, and is operated with a downward filtration velocity of 3–8 m/h. Pressure drop or effluent water quality trigger a periodical backwash procedure to remove the retained solids [1].

In the filter bed, Fe(II) can be removed via homogeneous, heterogeneous and biological oxidation, depending on operational parameters such as, supernatant water level, filtration velocity, pH and O_2 concentration [2]. The backwashed solids mainly consist of hydrous ferric oxides (HFO) and its adsorbed substances [3]. Homogeneous Mn(II) oxidation is slow [4] and therefore removal of Mn (mainly) occurs in the filter bed via a biological oxidation pathway to Mn oxides (MnO_2) [5–7]. NH_4 is also biologically oxidised in the filter bed [8], in a two-step process, executed by bacteria and archaea, via nitrite (NO_2) to nitrate (NO_3) [9].

As, when dissolved in anaerobic groundwater, is at neutral pH and slightly acidic water typically present as $H_3AsO_3^*$, the reduced, uncharged, trivalent form [10,11]. While, after filtration, the remaining As is present as $H_2AsO_4^-$ [3]. This is the oxidised, charged, pentavalent form. At neutral pH, As(V) is, in contrast to As(III), negatively charged and is therefore more efficiently removed via adsorption to HFO [12,13]. Measurements over the height of the filter bed pointed out that As(III) oxidises rapidly in the top layer of a rapid sand filter [3]. Since homogeneous As(III) oxidation occurs on a time scale of days [12,14], the observed rapid oxidation was hypothesised to be either biological, by MnO_2 present on the filter grains, or a combination of the two. However, although MnO_2 is capable of oxidising As(III) [15], it can be inhibited in presence of Fe(II) and Mn(II) [13,16,17]. The alternative explanation for the accelerated As(III) oxidation during rapid sand

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https://doi.org/10.1016/j.jwpe.2017.12.003
Received 18 September 2017; Received in revised form 4 December 2017; Accepted 8 December 2017
Available online 14 December 2017

filtration, is the presence of As(III) oxidising bacteria (AsOB). At least 50 phylogenetically diverse As(III)-oxidizing strains, distributed over 25 genera, have been isolated from various environments [18], and aerobic AsOB have been found in groundwater aquifers [19,20] and groundwater treatment filters [21].

AsOB can be subdivided into: heterotrophic bacteria [22–24] and chemolithoautotrophic bacteria [25,26,24]. Heterotrophic bacteria oxidize As(III) as a detoxification mechanism [27–29] and require organic matter for growth and As(III) oxidation. Chemolithoautotrophic bacteria, on the other hand, can use As(III) as the principal electron donor in catabolism, and use inorganic carbon as their carbon source [30]. Phylogenetic studies classified most AsOB in the Proteobacteria phylum. Within this phylum they can be subdivided as follows: most AsOB in the α-Proteobacteria class are chemolithoautotrophic, most in the β-Proteobacteria class are heterotrophic and all AsOB in the ɣ-Proteobacteria class are heterotrophic [31,32].

So far As(III) oxidation in rapid sand filters has only been studied in the relation to other biological processes, whereby As(III) oxidation was regarded to rely on co-occurring (bio-)chemical processes, including oxidation with Fe [33], Mn [34,35], NH_4 [36] or in combination with biogenic MnO_2 [37]. Other studies have focussed on using various inoculates to invoke biological As(III) oxidation [38,37]. Although AsOB are found in rapid sand filters [21], it is however, far less researched whether these bacteria are responsible for As(III) oxidation independent of co-occurring chemical and biological processes, as a result of Fe, Mn and NH_4 removal. The oxidation of As(III) by AsOB is of great interest for As(III) removal, because the resulting As(V) formation is imperative for subsequent adsorption onto HFO in filters [12,13].

Therefore, the objective of this study was firstly to investigate whether AsOB will grow on As(III) substrate in rapid sand filters, in absence of other biological (ammonium, manganese and ferrous oxidising bacteria) and chemical (HFO, MnO_2) processes. And secondly, it was the aim to assess whether a mature AsOB population can survive in rapid sand filters with low As(III) concentrations ($< 10\,\mu g/L$) amidst the other major groundwater constituents (Fe(II), Mn(II), NH_4). For this purpose As(III) oxidation was investigated in pilot-scale sand filter columns fed with raw groundwater, as well as treated groundwater (drinking water) with spikes of either As(III), Mn(II) or NH_4.

2. Materials and methods

2.1. Experimental procedure

For the purpose of accumulating different biomasses that were tested on their ability to oxidise As(III), multiple experiments were executed by preloading sand columns with different feed water qualities, i.e., preloading consisted of gently flowing a specific water quality through the columns to establish a biomass in the column. As(III) oxidation was tested with five different water quality settings of which the first two can be considered as control settings: (1) “virgin sand” (no preloading), (2) “blank” (sand preloaded with drinking water), (3) “As (III)” (sand preloaded with drinking water and added As(III)), (4) “NH_4” (sand preloaded with drinking water and added NH_4), and (5) “Mn” (sand preloaded with drinking water and added Mn(II)). Additionally an experiment with naturally As(III)-containing groundwater was done to find the influence of the natural groundwater matrix on the AsOB. Here, the filtrate of sand filters was compared for 50 days by using virgin sand (no preloading) and sand with an As(III) oxidising biomass (preloaded by drinking water spiked with As(III)). All experiments were executed as triplicates.

2.2. Experimental column set-up

The experimental set-up consisted of 12 identical columns, therefore 4 settings could run simultaneously in triplicates (Fig. 1). Each column has a diameter of 90 mm and a height of 1m. The columns were filled with 0.5 m (± 2%) quartz sand (0.4–0.8 mm) obtained from ‘Aqua Techniek’, which is typically used for rapid sand filtration. Before starting the experiment the columns were extensively backwashed with drinking water until the supernatant was visually clear. The flowrate used for all experiments was set to 105 mL/min per column, resulting in a filtration velocity of 1 m/h (± 10%). Supernatant water level in the columns was kept at 10 cm during the drinking water experiments for the purpose of sufficient chemical mixing. Chemicals, when used, were directly dosed in the supernatant water, using peristatic dosing pumps (Cole-Parmer Masterflex L/S) at a continuous flow of 1 mL/min. For the aerated groundwater experiments the supernatant water level was lowered to an initial height of 2 cm, which, as a result of filter clogging, rose to 15 cm just before backwashing. Backwashing procedure was executed with drinking water and consisted of expanding the filter bed by 20% until the supernatant water was visually clear. The anaerobic groundwater was aerated by cascading the groundwater directly into the supernatant water. No chemicals were dosed in the natural groundwater. Throughout the experiment, the sand columns were continuously fed with (spiked) drinking water or groundwater and covered to prevent direct (sun)light influencing the results.

2.3. Water quality

The column experiments were performed at water treatment plant Dorst (Brabant Water). Natural groundwater was used to perform the experiments in the presence of Fe, Mn and NH_4. Preloading was executed with drinking water produced at this production location spiked with the prior mentioned desired components. Both the quality of the groundwater and the drinking water are shown in Table 1. The groundwater is anaerobic and abstracted from 140 m depth. Drinking water is produced by aeration and rapid sand filtration, including the dosage of 2 mg/L $NaMnO_4$ and 2 mg/L $FeCl_3$ to stimulate As removal.

2.4. Chemicals, addition and concentrations

The As(III), Mn(II) and NH_4 dosing solutions were prepared from the following reagent grade chemicals: As(III)Cl_3 (Aldrich chemistry, 99.99% trace metals basis), Mn(II)Cl_2, (Aldrich chemistry, 98% beads) and NH_4Cl (Emsure, 99.8%). The chemicals were diluted in drinking water to 10.6, 212 and 106 mg/L, respectively. To prevent oxidation in the dosing vessels (25L), the vessels containing As(III) and Mn(II) were acidified to pH 3–4 by adding 8–12 mL 5 M HNO_3. The chemicals were continuously pumped into the supernatant water, with a flow rate of 1 mL/min, targeting an influent concentration of approximately 100 µg/L As(III), 1 mg/L NH_4 and 2 mg/L Mn(II).

2.5. Sampling and analytical methods

Samples from supernatant water were collected with a syringe from the lowest part of the supernatant water. Filtrate was obtained from the discharge tube after the overflow to prevent changing the filtration rate. pH, electrical conductivity (EC) and O_2 were measured with WTW electrodes (SenTix 940, TerraCon 925 and FDO925). As, Fe and Mn were analysed with Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Thermo X2-series), NH_4, NO_2, NO_3 and PO_4 were analysed by a discrete analyser spectophotometry (Aquakem 250, Thermo Scientific). As speciation was done according the Clifford [47] method. Here, 150 mL sample is passed through an anionic resin (80 mL Amberlite® IRA-400 chlorite form resin in a 100-mL syringe) that retains only the charged As(V) species. The filtrate from the resin is considered to be As(III) only. As(V) is then calculated by subtracting As(III) from the measured total As concentration. The first 50 mL was always discarded, the remaining 100 mL was collected and analysed using ICP-MS. The Clifford method is a robust method, however it was found that unavoidably the resin retained 14% of As(III), min = 7%, max = 23%; n = 24.

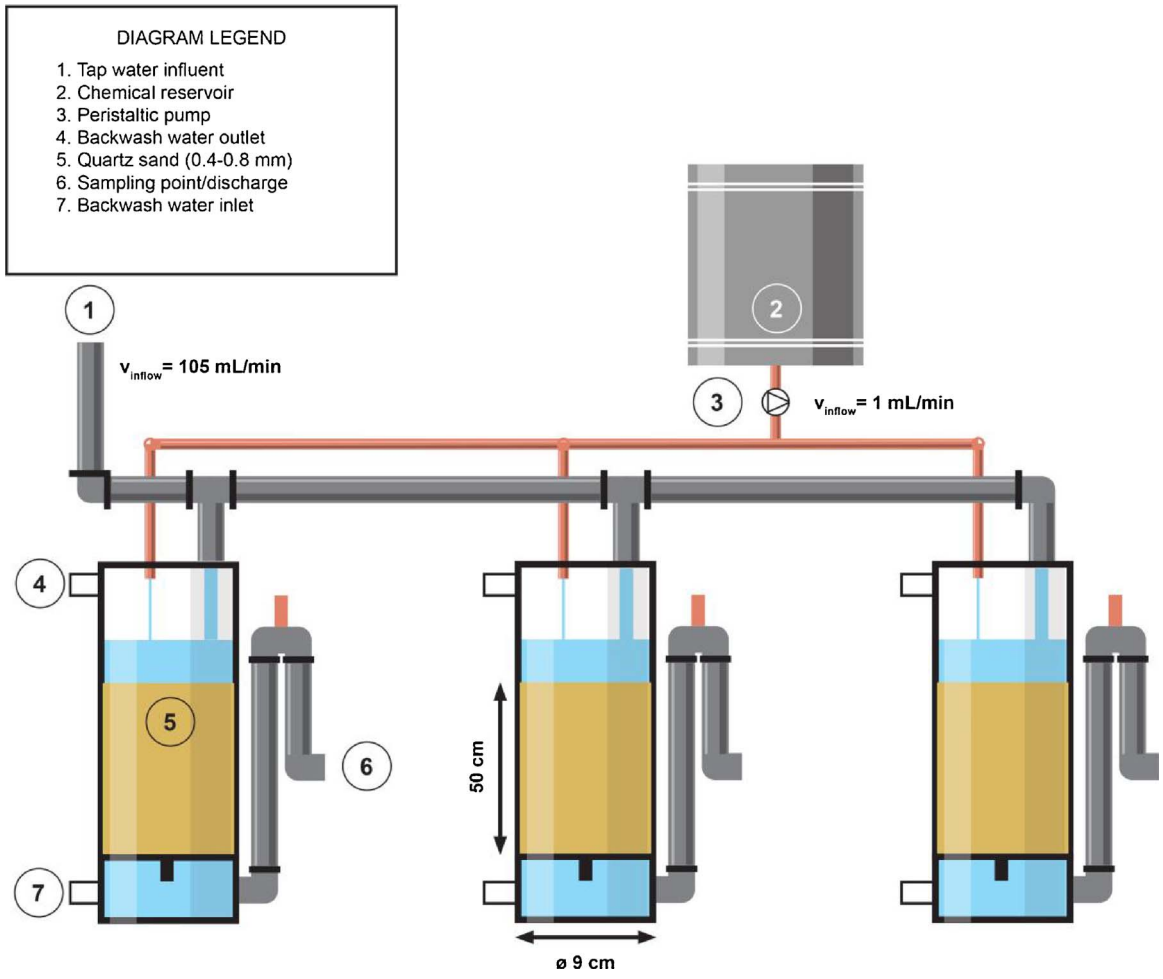

Fig. 1. Schematic overview of sand filter set-up as triplicates (valves not shown).

2.6. Biomass characterization and profiling

The biomass accumulation stages were finalised by removing the filter sand from the columns and analysed on adenine triphosphate (ATP), which serves as an indicator of active biomass [39,40], for the purpose of quantifying the biomass after the different experiments. From a completely filled sampling bottle (100 mL) 1 g of sand was weighted and 9 mL of ATP free water was added. This sample was shaken shortly where after it was ultrasonically vibrated for 10 min in an ultrasonic bath (Bransson). Subsequently, the sample was rested for 5 min before ATP measurement. Samples were processed by the ATP meter "Centro XS3 LB960" (Berthold) using a chemical kit from BioTHema. In addition to the ATP analysis, a microbial community analysis was performed for all columns experiments. 100 mL filter sand was harvested and stored at −80 °C. Of these samples around 0.5 g was used for DNA extraction with the DNeasy UltraClean microbial kit (Qiagen). The DNA was subjected to quality checks that consisted of agarose-gel electrophoresis aiming to verify DNA integrity and a QuBit fluorometer and the ds DNA HS assay (Life technologies) analysis to determine the concentration of DNA that was obtained from the sand samples. The BioAnalyzer (Agilent) was used to perform an additional integrity and concentration check after which bacterial (V3-V4) 16S rRNA genes were amplified and subjected to high throughput sequencing using the MiSeq platform (from Illumina and at BaseClear, Leiden, the Netherlands). Reads were generated using the Illumina Casava pipeline (version 1.8.3), checked using Illumina Chastity filtering plus an in-house protocol (Baseclear) and final assessment was made using the FASTQC quality control tool (version 0.10.0). QIIME workflows were used to generate taxonomic summaries [41].

Table 1
Drinking water and groundwater quality used in column experiments.

Water quality parameters	Units	Raw groundwater	Drinking water
pH	[−]	7.54[a]	7.69
Temperature	°C	15.5	16.5
O_2	mg/L	< 0.01[a]	9.07
HCO_3	mg/L	259	238
Conductivity	mS/m	40.9	39.1
As(tot)	µg/L	13.2	< 1.0
As(III)	µg/L	12.7	< 1.0
Fe	mg/L	1.4	< 0.01
Mn	mg/L	0.04	< 0.01
NH_4	mg/L	0.62	< 0.03
NO_3	mg/L	< 0.03	1.86
PO_4	mg/L	0.45	0.078
TOC	mg/L	2.1	2.1
SO_4	mg/L	< 0.1	0.37
ATP	ng/L	1.1	3.2

[a] The pH and O_2 in the supernatant level of the pilot columns was, depending on the supernatant level, between 7.6–7.7 and 4–4.5 mg/L respectively.

3. Results and discussion

3.1. As(III) oxidation in various preloaded sand columns

Sand columns were preloaded with drinking water ("blank"), drinking water with added NH_4 ("NH_4"), drinking water with added Mn

Table 2
Water quality parameters before and after preloading. (For interpretation of the references to colour in this table legend, the reader is referred to the web version of this article.)

Parameter	unit	Supernatant / Influent while preloading				Filtrate / Effluent after preloading per column			
		n	Average	min	max	Drinking water	+NH_4	+Mn	+As
Duration preloading	[days]					80	80	80	38
Water temperature	[°C]	12	16.7	15.9	18.2				
O_2	[mg/L]	12	9.03	8.89	9.25	9.1	8.07	9.08	9.1
pH		12	7.81	7.76	7.87	7.91	7.73	7.88	7.8
NH_4	[mg/L]	12	0.95	0.87	1.07		0.01		
Mn	[mg/L]	8	1.61	1.47	1.8			1.61	
As	[µg/L]	42	111.3	93.9	135.3				106

(II) ("Mn") and drinking water with added As(III) ("As"). During preloading the water quality was monitored to determine if bacteria had grown to convert NH_4, Mn and As in the columns, the moment of conversion determined the minimum duration of pre-loading. Table 2 summarises the in- and effluent water quality at the end of preloading, as well as the duration of preloading per column.

The water quality parameters in Table 2 primarily shows an effect from preloading in the NH_4 columns, preloading did not affect the water quality over time in the other columns. Initially part of the As was adsorbed to the filter sand, however over the course of the experiment As adsorption decreased, resulting in an average removal of 4.5% during preloading. The preloading with NH_4 resulted in measureable changes in O_2 and pH caused by its oxidation. After the preloading period, the effluent of the columns no longer contained NH_4 nor NO_2, so nitrification to NO_3 was found to be complete. Surprisingly, Mn oxidising bacteria, did not seem to have grown in the Mn(II) preloaded columns, because Mn(II) removal as MnO_2 was not observed, additionally Mn(II) oxidation would have resulted in changing O_2 and pH values during the experiment and this was not observed.

The column preloaded with drinking water, did not show apparent water quality changes, however, the post-experiment analyses of ATP on the filter sand showed that indeed drinking water native bacteria had grown in the column (Fig. 2). Note that the ATP value on the virgin filter sand was only 0.71 ng/g prior to preloading.

The drinking water used for these experiments contained 3.7 ng/L ATP. The ATP values in the columns preloaded with drinking water-containing Mn or As, showed similar values and, although modest, showed more bacterial activity in the top layer than the blank columns, with 73% and 45% higher ATP concentrations respectively. The largest ATP concentrations on sand were achieved by preloading with drinking water and NH_4. In the top of the sand bed this led to more than 15 times higher ATP concentrations than the preloaded columns with drinking water only. Additionally it was observed that the biomass was stratified over the filter, from the highest concentration of biomass in the top of the sand filter to the lowest biomass concentration at the bottom. This is in line with findings of Lee et al. [8], where NH_4 oxidising bacteria in a sand filter showed a similar vertical distribution.

After the preloading stage, all columns were loaded with 20 µg/L As (III) for 24 h to investigate whether the bacteria in the various columns were capable of oxidising As(III) to As(V). About 10% of the dosed As

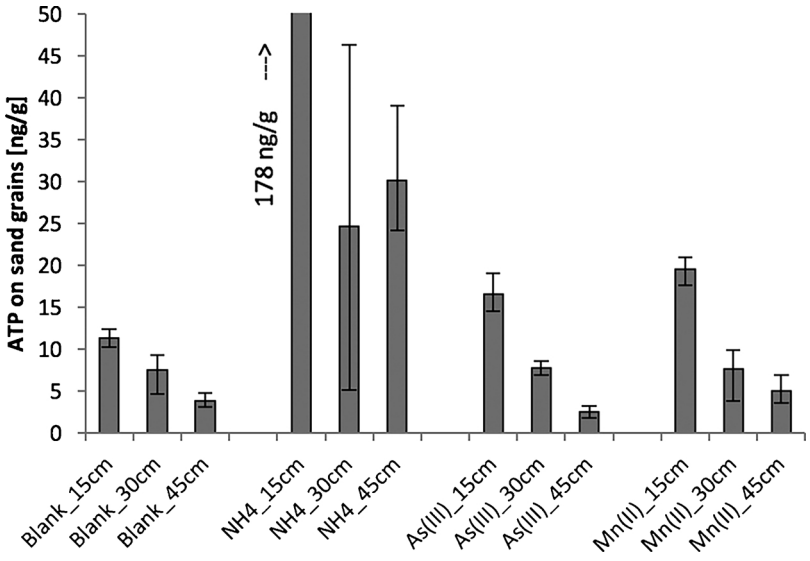

Fig. 2. ATP concentration on the filter sand at 15, 30 and 45 cm from top of the filter after preloading with drinking water (blank), NH4, As(III) or Mn(II). Two outliers removed with 95% certainty interval with Dixon's Q-test.

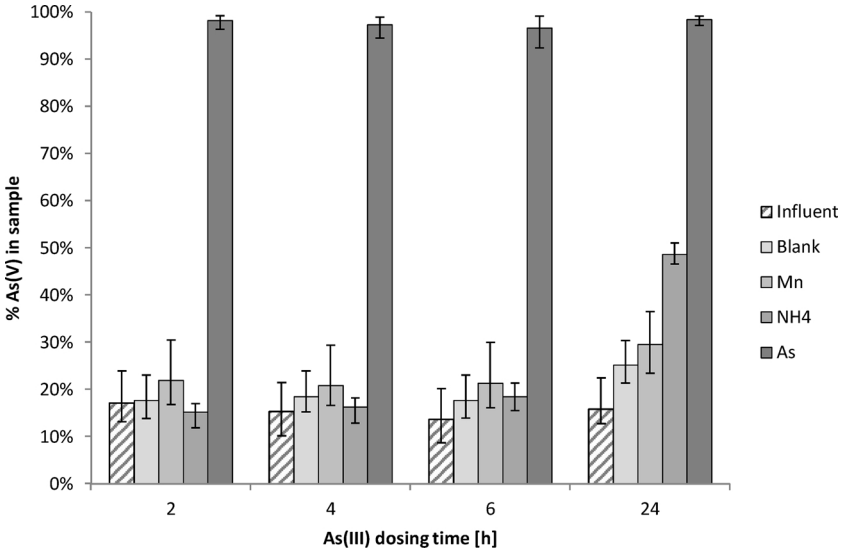

Fig. 3. Percentage of As(V) of total As in water sample during 24 h in the preloaded sand columns. Samples obtained from influent and filtrate of the filter sand columns after preloading with drinking water, NH_4, As(III) or Mn(II).

was retained in the sand columns, therefore the percentage of oxidised As in the filtrate is depicted in Fig. 3.

During the first 6 h no As(III) was oxidised in the columns preloaded with NH_4, Mn(II) or drinking water alone. Thereby establishing that no significant abiotic As(III) oxidation occurred in the sand columns and the biomasses that had not, in advance, been exposed to As(III) could not instantly oxidise the dosed As(III). This was in contrast to the columns preloaded with 100 µg/L As(III), the filtrate of these columns contained solely oxidised As. Apparently, within 38 days, AsOB had grown in these columns during preloading.

After 24 h, it seems that oxidation of As(III) started to occur in all columns, particularly in the NH_4 preloaded column, where 40% of the As in the filtrate was As(V). Apparently an adaptation phase of the well-developed biofilm started and resulted in the oxidation of 9 µg/L As (III).

3.2. Bacterial growth profile of As(III) oxidation on virgin filter sand

As(III) and As(V) in the filtrate during this 38 days of pre-loading with As(III)-spiked drinking water are depicted in Fig. 4. During the entire experiment, the average As(III) concentration in the supernatant water was 116 µg/L (min 98, max 135 µg/L), but due to (slow) homogeneous oxidation in the dosing vessel the As(III) concentration at the end of the experiment decreased to 95 µg/L.

The results in Fig. 4 show that initially the sand columns did not oxidise As(III), but with a daily influx of 150 L As(III)-containing drinking water, the columns gradually started oxidising As(III). After 38 days, 98% of As(III) was oxidized to As(V) in the sand columns. The process started after approximately one week, however, the oxidation rate accelerated after 14 days. This is typical for a bacterial process, where after a lag phase of limited bacterial growth a log phase follows of rapid bacterial growth [42]. Additionally, the ATP concentration of the filter sand increased from 0.71 ng/g to 16 ng/g (in the top of the sand filter) indicating an increased bacterial activity within the 38 days experiment [39]. Therefore it may be concluded that the gradual As(III) oxidation observed in the columns was caused by AsOB. Given that the experiment with drinking water without As(III) or Mn(II) and NH_4 initially showed no As(III) oxidation (Fig. 3), it is suggested that a specific AsOB population was grown by preloading filter sand with As(III) substrate. Additionally, microbial community profiling of the biomass on the filter sand after preloading with As(III), showed a biomass with a high relative abundance of α- and β-Proteobacteria (Fig. 5). Within these two classes of Proteobacteria most AsOB are phylogenetically placed [31,32].

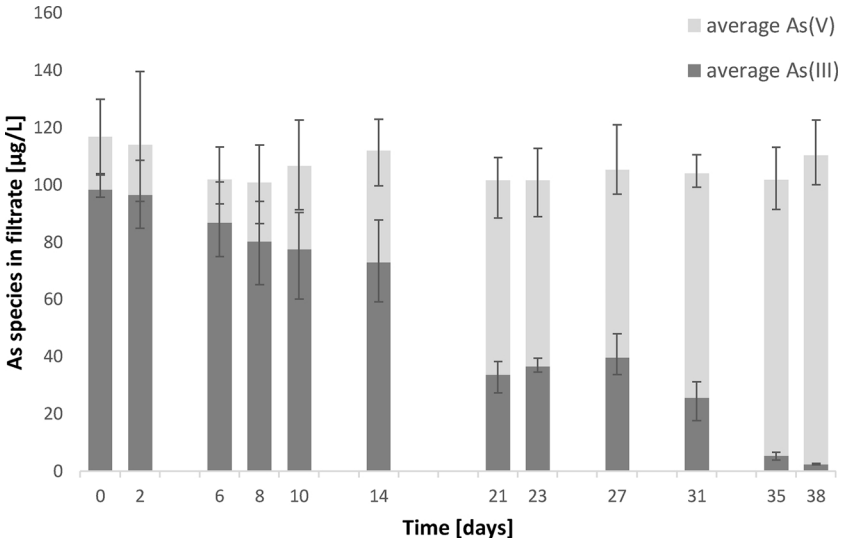

Fig. 4. As(III) and As(V) species in the filtrate of sand columns over time (38 days) by feeding drinking water added with approximately 100 µg/L As(III).

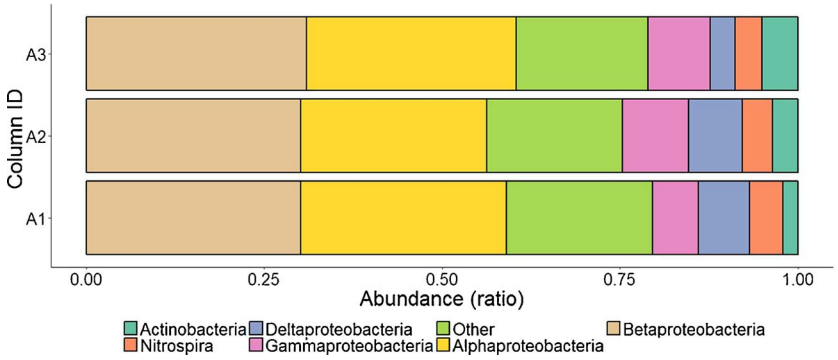

Fig. 5. Results of microbial community analysis after feeding drinking water with 100 µg/L added As(III) for 38 days. Relative abundance of bacteria is shown at the taxonomic rank Class (L3) of the filter sand columns ID's A1, A2 and A3 (triplicates).

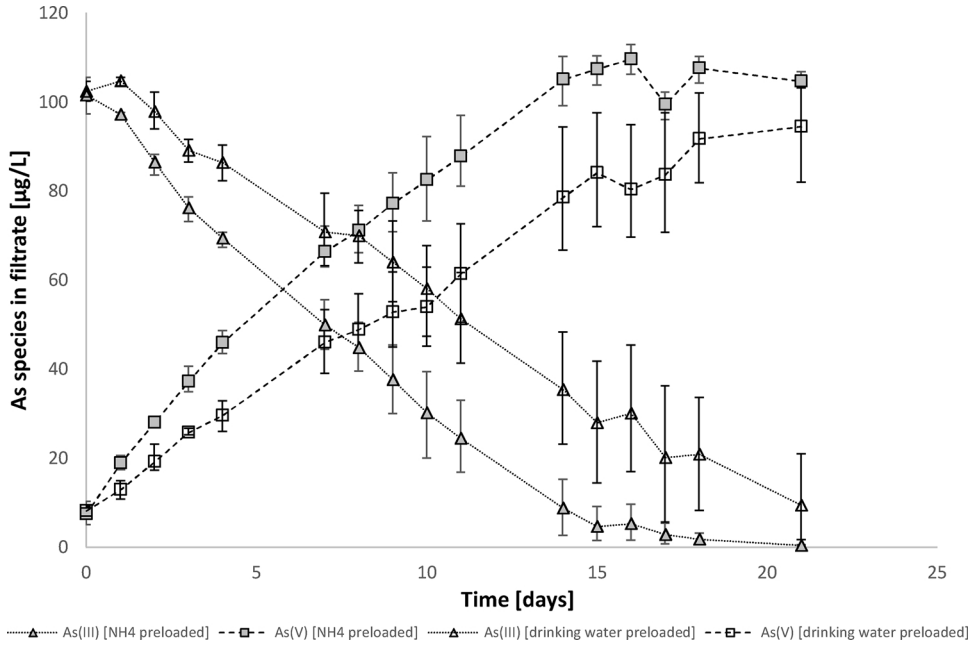

Fig. 6. As(III) and As(V) in the filtrate of drinking water pre-loaded columns spiked with and without 0.79 mg/L-N NH_4. Influent As(III) concentration was 98 µg/L min 83 max 118 (n = 16) As(III) spiked in drinking water.

3.3. As(III) oxidising biomass growth in existing biomass

To investigate whether AsOB would grow in an already accumulated biomass, 100 µg/L As(III) was dosed to preloaded columns for 22 days. Prior to As(III) dosage the columns were loaded with drinking water and drinking water with additional NH_4 for 50 days. The experiment started when the latter biomass was capable of completely converting 1 mg/L NH_4 to NO_3. The resulting As(III) and As(V) concentrations in the filtrate are depicted in Fig. 6.

Results show that independent of the type of preloading, both columns successfully oxidised the added As(III) within three weeks and, in addition, compared to the As(III) oxidation experiment on virgin sand (Fig. 4), both columns already containing a biomass reached complete oxidation in a shorter time. In the NH_4 preloaded sand columns As(III) was completely oxidised within 21 days without retarding or inhibiting nitrification. The experiment without NH_4 addition, using the same drinking water, showed a similar As(III) oxidation pattern, only initially deviating from the oxidation pattern in the NH_4 preloaded columns. Starting with a more gentle slope than the NH_4 preloaded columns in the first week, after 7 days and onwards these columns followed the same As(III) concentration gradient.

The above suggests that established biological processes prior to As (III) exposure do not retard or prevent biological As(III) oxidation but accelerate them. A comparable study performed by Lami et al. [43] showed that when As(III) was dosed to a mixed biomass soil culture, analogue to Fig. 6, alteration and adaptation of the biomass was observed within weeks, whereby some As(III)-oxidizing bacterial groups had increased up to 20-fold compared to a control experiment. The overall conclusion here is that NH_4 oxidising biomass (nitrification) had no detrimental effect on the development of biological As(III) oxidation in groundwater filters, but the additional NH_4 or NO_3 (nutrients) rather stimulated the AsOB accumulation.

3.4. Biological As(III) oxidation in natural, aerated groundwater

In natural groundwater, apart from NH_4 and Mn(II), also other constituents, such as Fe(II), PO_4, CH_4 and H_2S may enter the filter bed together with As(III). Therefore, the sand column experiments were repeated with natural, aerated groundwater with virgin sand columns and As(III) preloaded (35 days) sand columns capable of oxidising 100 µg/L As(III). In contrast to prior experiments, oxidising Fe(II) caused the filter bed to clog with HFO and a 2–3 day backwash cycle had to be applied. The run time of this experiment was 50 days and during this period the supernatant water level, pH and O_2 were 2–15 cm, 7.47–7.78 and 3.56–4.58 mg/L, respectively. Fig. 7 depicts removal percentages for Fe, NH_4 and PO_4 over the run time of the virgin sand columns (left) and the columns preloaded with As(III) (right).

Fe was instantly and completely removed in all columns. It is expected that initially the removal process was due to abiotic, homogeneous and heterogeneous oxidation [2]. It is possible that during the experimental period this shifted to a partially biological Fe oxidation process [44]. The 150 µg/L PO_4 was consistently removed between 80

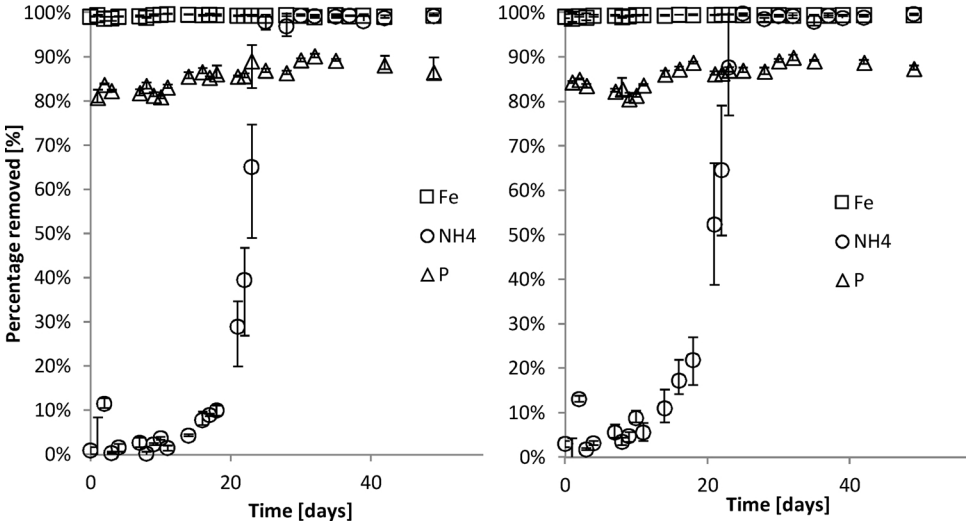

Fig. 7. Removal percentage of Fe, NH_4 and P over time in virgin sand columns (Left) and As(III) preloaded sand columns (Right). Filtration velocity 1 m/h; bed height 0.5 m. Initial concentration of the groundwater: Fe 1.4 mg/L; As 13 µg/L (as As(III)); PO_4 0.45 mg/L; Mn 0.04 mg/L; NH_4 0.62 mg/L; pH 7.54; Water temperature 14 − 18 °C. After aeration/supernatant level: O_2 4.4 mg/L; pH 7.65.

and 90% in both columns, presumably by (co-)precipitation with oxidised Fe(II) and adsorption to HFO [45]. The increased PO_4 removal (± 10 µg/L) after the lag and log phase of the ammonium removal process could indicate the utilisation of PO_4 for the growth of biomass responsible for nitrification [46]. The difference in ripening time between the column types was about 2 days which is consistent with the previous findings where it was observed that preloading had a generic accelerating effect on biological processes (Fig. 6). However both column types consistently converted the major part of the NH_4 within 30 days. Mn removal did not start within this 50 day experiment, both in the virgin sand columns and in the preloaded columns. At the end of the experiment, the various oxidation processes in both the virgin sand and As(III) preloaded columns consumed about 1.6 mg/L O_2.

From the results depicted in Fig. 7 it can thus be concluded that preloading sand columns with As(III)-containing drinking water did not influence Fe, Mn, and PO_4 removal, and only mildly accelerated start-up of NH_4 removal. This suggests that the As(III) oxidising biomass did not accelerate NH_4 and Mn(II) removal.

Fig. 8 depicts the As concentration in the raw groundwater, more than 90% present as As(III), and in the filtrate for both the As(III) preloaded sand columns (top) and virgin sand columns (bottom) over time. As concentration in filtrate is depicted as the sum of the As(III) and As(V) species.

The sand columns that were preloaded with As(III), oxidised the 13 µg/L As(III) in the groundwater immediately and consistently during the 50 days experiment. The increased biological activity caused by conversion of other groundwater compounds during the course of the experiment did not hinder the As(III) oxidation in the 0.5 m sand bed (Fig. 8). The virgin sand columns, which did not contain an As(III) oxidising biomass at the start of the experiment, initially did not show As(III) oxidation. However, complete oxidation of the 13 µg/L As(III) developed within 23 days (Fig. 8), suggesting that, even though As(III) concentrations were low compared to the other groundwater constituents, the AsOB were able to maintain their population on the filter sand and oxidise the As(III) in the natural groundwater.

Concerning the actual As removal, three separate stages can be differentiated for both the virgin sand columns and the As(III) preloaded sand columns. The first five days of filtration As(III) oxidation in the virgin sand columns was low and the removal efficiency of As was steady at around 30%. Although complete oxidation in the As(III) preloaded sand columns was achieved throughout the experiment, initially slightly less As was removed compared to the virgin sand columns; potentially caused by As(III) saturation of the sand grain surface during preloading and desorbing the first days of feeding aerated groundwater. From day 5 until 28, biological As(III) oxidation was developed in the virgin sand columns and subsequently increased the As removal efficiency from 30% to 60%, because the produced As(V) has a higher affinity to HFO than As(III) [12,13]. After 28 days the As removal became stable at 60% for both columns until the end of the experiment. The similarity of both the As(III) preloaded sand columns and the virgin sand after this period suggests that the biological oxidation potential was fully utilised and a higher As removal, due to adsorption on HFO, could not be expected in the current column design and operation. It is concluded that preloading of As(III) to establish an As(III) oxidising biomass provided only a beneficial effect in the initial weeks of operation, as this positive effect got neutralised by the growth of AsOB on the sand grains in columns without any preloading. Additionally, regarding the gradually increasing removal efficiency after complete oxidation was achieved in the filtrate of virgin sand columns, it is hypothesised that as a consequence, the As(III) concentration profile over the height of the column moved upward to the Fe removal zone in the top of the filter bed, subsequently resulting in a higher As removal.

4. Conclusions

It was observed in this study that biological As(III) oxidation quickly developed in rapid sand filter columns fed by drinking water spiked with As(III). With a typical lag and log phase, decreasing As(III) and increasing As(V) concentrations in the effluent of the sand columns were measured in a timeframe of weeks. The growth of biomass in the sand columns was confirmed with ATP analysis. ATP concentrations on the sand grains increased from 0.7 ng/g to 16, 8 and 2 ng/g filter sand stratified from the top of the sand filter to the bottom, respectively. Therefore it was concluded that AsOB can develop on filter sand in absence of other chemical and biological oxidation processes (e.g. as a results of Fe, NH_4 and Mn presence).

Other experiments, performed with natural groundwater, showed that AsOB were able to grow and survive amidst Fe(II) and NH_4 oxidising processes in filters and prior to Mn(II) removal; complete As(III) oxidation was achieved within 22 days on virgin sand. Additionally, AsOB accumulated in filters by preloading with As(III)-containing drinking water were instantly and consistently capable of oxidising As(III) present in natural groundwater. The overall conclusion of this study is that AsOB are able to grow and maintain their population in rapid sand filters at low As(III) concentrations, either in presence or absence, of other common groundwater bacteria and naturally formed mineral precipitates (e.g. HFO and MnO_2).

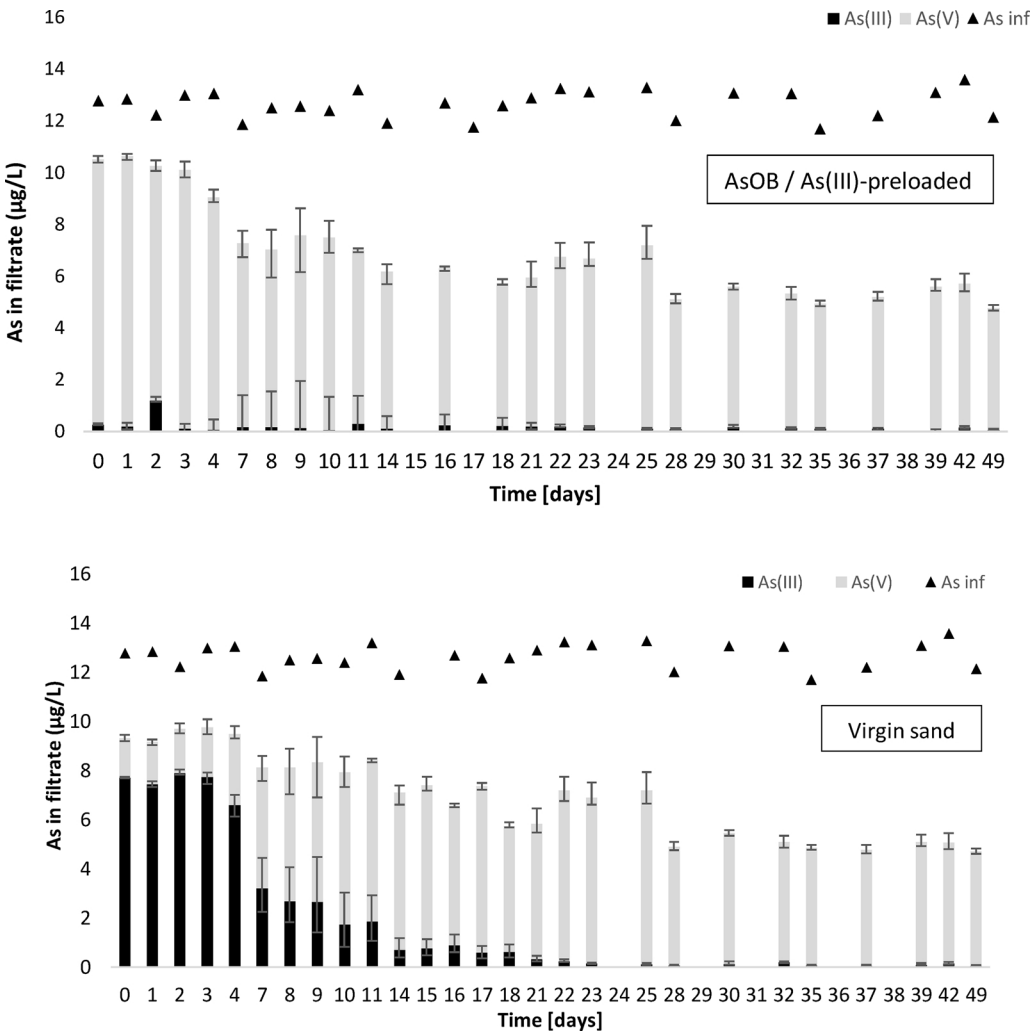

Fig. 8. As(III) and As(V) in the filtrate of As (III) preloaded (top) and virgin sand columns (bottom) over time. Filtration velocity 1 m/h and bed height 0.5 m. Initial concentration of the groundwater: Fe 1.4 mg/L; As 13 µg/L (as As(III)) also depicted < • > ; PO_4 0.45 mg/L; Mn 0.04 mg/L; NH_4 0.62 mg/L. As concentration in the filtrate is depicted as the sum of the As(III) and As(V) species.

Acknowledgements

This research is supported by the Dutch Technology Foundation STW, part of the Netherlands Organisation for Scientific Research and is partly funded by the Ministry of Economic Affairs Project code: 13343 (FixAs). The authors want to thank Timon Huijzendveld for his valuable and precise practical work on the column experiments and Marjet Oosterkamp for performing the microbial community analysis. In addition, the authors are grateful for the hosting of Brabant Water for the on-site experiments at Water Treatment Plant Dorst.

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