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Arsenic removal from iron-containing groundwater by delayed aeration in dual-media sand filters

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ABSTRACT

Generally, abstracted groundwater is aerated, leading to iron (Fe^{2+}) oxidation to Fe^{3+} and precipitation as Fe^{3+}-(hydr)oxide (HFO) flocs. This practice of passive groundwater treatment, however, is not considered a barrier for arsenic (As), as removal efficiencies vary widely (15–95%), depending on Fe/As ratio. This study hypothesizes that full utilization of the adsorption capacity of groundwater native-Fe^{2+} based HFO flocs is hampered by rapid Fe^{2+} oxidation-precipitation during aeration before or after storage. Therefore, delaying Fe^{2+} oxidation by the introduction of an anoxic storage step before aeration-filtration was investigated for As(III) oxidation and removal in Rajshahi (Bangladesh) with natural groundwater containing 329(±0.05) μgAs/L. The results indicated that As(III) oxidation in the oxic storage was higher with complete and rapid Fe^{2+} oxidation (2±0.01 mg/L) than in the anoxic storage system, where Fe^{2+} oxidation was partial (1.03±0.32 mg/L), but the oxidized As(V)/Fe removal ratio was comparatively higher for the anoxic storage system. The low pH (6.9) and dissolved oxygen (DO) concentration (0.24 mg/L) in the anoxic storage limited the rapid oxidation of Fe^{2+} and facilitated more As(V) removal. The groundwater native-Fe^{2+} (2.33±0.03 mg/L) removed 61% of As in the oxic system (storage-aeration-filtration), whereas 92% As removal was achieved in the anoxic system.

1. Introduction

Groundwater arsenic (As) contamination is a severe drinking water quality problem and a threat to human health in Bangladesh and other countries (Smedley and Kinniburgh, 2002; Chakraborti et al., 2013). Chronic exposure to As-contaminated drinking water has resulted in tens of millions of people suffering from skin lesions, hyperkeratosis, melanosis, skin cancer, and cancer of internal organs (Luzi et al., 2004; Li et al., 2012; Guo et al., 2013). The World Health Organization (WHO) recommends that As concentrations in drinking water should not be more than 10 μg/L (Smith et al., 2000). However, according to Bangladesh Drinking Water Standard (BDWS), the recommended value for As in drinking water is 50 μg/L. Nevertheless, groundwater in an extended area of Bangladesh contains As concentrations higher than those recommended values (Rosso et al., 2011; Perez and Francisca, 2013; Haque et al., 2018), and sometimes, it even exceeds 1500 μgAs/L (Nordstrom, 2002; Cavalca et al., 2019). In reducing groundwater, at near-neutral pH, As exists in the thermodynamically stable form as arsenite or As(III) (H_3AsO_3) (Cullen and Reimer, 1989; Smedley and Kinniburgh, 2002), while, in oxidizing conditions, arsenate or As(V) is the predominant species ($H_2AsO_4^-$, $HAsO_4^{2-}$) (Lafferty et al., 2010; Villalobos et al., 2014). Various treatment technologies, including adsorption, coagulation/flocculation, chemical precipitation, lime softening, ion-exchange, and membrane filtration have been studied to remove As from water (Meng et al., 2001; Su and Puls, 2001; Roberts et al., 2004; Zhang et al., 2010; Pio et al., 2015; Niazi et al., 2018; Shakoor et al., 2019; Amen et al., 2020). However, these options are energy-intensive, and/or consume chemicals that make the treatment methods expensive (Hoque et al., 2004, 2006; Hossain et al., 2014). In addition, commonly available treatment systems are complicated to operate and maintain, needing regular parts' replacement and after-market services that are expensive and skilled personnel who may not be locally available (Delaire et al., 2017). As a consequence, these technologies are not sustainable in the long run in vulnerable communities (Hossain et al., 2014, 2015).

Therefore, there is a need for simple, economical, and energy-

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efficient alternatives, utilizing locally available materials and less or no use of additional chemicals (Katsoyiannis et al., 2015; Hering et al., 2017; Senn et al., 2018). Passive groundwater treatment consists of aeration, followed by single or multiple filtration steps (Morrison et al., 2002; Roberts et al., 2004) without using chemicals, as shown in Fig. 1. Aeration frequently consists of cascades or spray aerators, followed by gravitational filtration through a submerged filter bed. Traditionally, passive groundwater treatment is used for the removal of Fe^{2+}, ammonium (NH_4^+), and manganese (Mn^{2+}) (Katsoyiannis et al., 2008a, 2008b; Bruins et al., 2014; Vries et al., 2017). However, this treatment method is not considered a robust barrier for As, as its removal efficiencies typically vary widely between 15% and 95% (Lowry and Lowry, 2002; Sorlini and Gialdini, 2010; Gude et al., 2016), depending on the water chemistry (Sorlini and Gialdini, 2014). Co-removal of As with groundwater native-Fe is reported to be only possible if As concentration is low (<50 μg/L) (Katsoyiannis et al., 2015; Gude et al., 2018a) and the Fe to As ratio is sufficiently high (Biswas et al., 2012; Sharma et al., 2016; Annaduzzaman et al., 2018). Furthermore, studies by Holm and Wilson (2006) showed that despite having groundwater native-Fe^{2+} of >1.5 mg/L, only 20–25% (8–10 μg/L) of As could be removed. This was also the case in Lowry and Lowry's (2002) study that showed aeration and storage of Fe^{2+} containing groundwater, facilitated only partial As removal with hydrous ferric oxide (HFO) flocs. Roberts et al. (2004) also revealed that 50–55 mg/L of Fe^{2+} was required to achieve <50 μg/L As in the treated solution from 500 μg/L of As in a single-step Fe^{2+} oxidation system, while in a step-wise Fe^{2+} oxidation system, only 20–25 mg/L of Fe^{2+} was sufficient to have an efficiency of over 90% As removal. All of these studies underline that the ratio of Fe to As played a major role in the As removal process.

Equilibrium adsorption studies of As on Fe oxides show that although both As(III) and As(V) have an affinity for Fe oxides (Dixit and Hering, 2003; Hug and Leupin, 2003; Voegelin and Hug, 2003; Luzi et al., 2004; Han et al., 2016), during Fe^{2+} oxidation and subsequent fresh HFO formation, the removal capacity for As(V) is much higher (Manning et al., 2002; Mercer and Tobiason, 2008; Hering et al., 2017). The removal of As(III) is more efficient through co-precipitation, while As(V) removal is related to surface complexation/precipitation with HFO flocs (Tian et al., 2017). Therefore, in order to develop passive groundwater treatment into an efficient system for As removal by HFO flocs, it is critical to promote the presence of As in the oxidized As(V) state during onset Fe^{2+} oxidation.

In this study, it was hypothesized that by delaying the oxidation of groundwater native-Fe^{2+}, the available adsorption capacity of the freshly formed HFO flocs can be utilized better for sufficient As removal. Therefore, anoxic storage before aeration and filtration was applied to allow step-wise Fe^{2+} oxidation. This novel oxidation sequence was compared against a conventional oxic storage system, to study the influence on As removal efficiency by delayed/step-wise oxidation followed by aeration and dual-media sand filtration. Moreover, in this treatment system, locally available filter materials were used without additional adsorbents/chemicals. The study was conducted in Rajshahi (Bangladesh), using naturally As-contaminated groundwater (329±0.05 μg/L) in the presence of other inorganic groundwater contaminants (e.g., Fe^{2+}, PO_4^{3-}, NH_4^+).

2. Materials and methods

2.1. Water quality

The pilot-scale experiments were conducted using As contaminated groundwater in Uttar Kazirpara village in Paba Upazila of Rajshahi district, Bangladesh. Table 1 provides the relevant water quality parameters of used natural groundwater. The groundwater was in a reducing/anoxic state and abstracted from 50(±1) m depth using an electric submersible pump (GAZI, Bangladesh).

Table 1
Raw groundwater quality in the shallow well that used as an influent for the pilot experiments.

Water Quality Parameters	Unit	Raw Groundwater
pH	[-]	6.94±0.08(SD)
Dissolved Oxygen (DO)	mg/L	0.07±0.06(SD)
Oxygen Reduction Potential (ORP)	mV	-110±4(SD)
Electrical Conductivity (EC)	μS/cm	675±6(SD)
Temperature	°C	26.7±1(SD)
As(total)	μg/L	329±0.05(SD)
As(V)	μg/L	39±0.02(SD)
As(III)	μg/L	290±0.02(SD)
Iron (Fe^{2+})	mg/L	2.33±0.03(SD)
Manganese (Mn)	μg/L	600±0.04(SD)
Magnesium (Mg)	mg/L	13.93±0.22(SD)
Calcium (Ca)	mg/L	69.96±1.07(SD)
Ammonium (NH_4^+)	mg/L	0.96±0.02(SD)
Nitrate (NO_3^-)	mg/L	0.39±0.02(SD)
Silicate (SiO_4^{4-})	mg/L	28.5±2.04(SD)
Phosphate (PO_4^{3-})	mg/L	2.15±0.03 (SD)

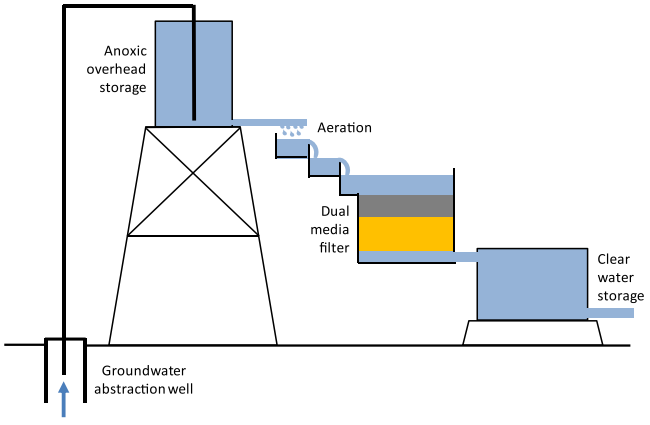

Fig. 1. The concept of delayed aeration by application of an anoxic storage container before aeration and dual-bed filtration.

2.2. Experimental set-up

The experimental set-up consisted of four identical dual-media filter columns to run the two parallel experiments in duplicate (Fig. 2). The As (III) oxidation and removal were investigated with two different storage conditions from the same source water: (1) conventional oxic storage and (2) anoxic storage. The oxic storage container was equipped with a spray aerator placed 35 cm above the container, whereas the anoxic storage container had an inlet at the bottom of the container – preventing any atmospheric contact. On the first day, the systems were allowed to overflow for 5 min to remove atmospheric oxygen. On the subsequent days, groundwater was added to the remaining stored water (about one-third of the volume), to refill the storage systems.

After storage, the water was aerated by letting it drip into the column from the pipeline, which was placed 35 cm above the top of the supernatant level of the column (Fig. 2). Each column has a diameter of 10 cm and a height of 120 cm. The columns were filled with 40(±2%) cm anthracite (0.6–0.9 mm) on top followed by 50(±2%) cm of quartz sand (0.3–0.75 mm) at the bottom. The quartz sand (known as Domar sand) is commonly used in construction, and gravel packing of the screen during tubewell installation. The Domar sand was bought from locally available shops. Before starting the experiment, the filter columns were extensively backwashed until the supernatant was visually clear. The experimental flow rate was set to 9 L/h to achieve a filtration velocity of 1 (±10%) m/h. The supernatant water level was kept at 15–20 cm above the filter bed, when the level rose beyond 20 cm due to filter clogging, backwashing was done.

Backwashing was executed with a 20% expansion of the filter bed using stored water and continued until the supernatant water was visually clear (±20 min). No chemicals were used during the experiments. Throughout the experimental period of 30 days, the column was continuously fed with water and covered to protect the water from direct sunlight exposure. The water quality of the different storage containers, supernatant, anthracite filtrate and column filtrate were compared for 30 days, and all experiments were performed in duplicates.

2.3. Sampling and analytical procedure

The parameters pH, dissolved oxygen (DO), oxidation-reduction potential (ORP), and temperature (T) were directly measured on-site during sample collections using WTW electrodes (SenTix 940, FDO®925, SenTix ORP 900, and Terracon 925, respectively). Samples were collected on days 1, 5, 10, 20, and 30. The filtered and unfiltered 15 ml water samples were collected in polypropylene transparent 15 ml centrifuge tubes (Sigma Aldrich) and the relevant samples were acidified immediately in the field for preservation until further analysis. The samples were acidified with ultrapure HNO_3 acid (ACS reagent, 70%; Formula weight 60.01 g/mol; Sigma Aldrich) that made up for 1.5% of the total solution. Furthermore, 250 ml filtered (0.45 μm) and non-acidified water samples were collected in 250 ml polypropylene laboratory-grade water vials for determining ammonium (NH_4^+), nitrate (NO_3^-), and phosphate (PO_4^{3-}) concentrations. All filtered samples were filtered using a polyether-sulfone 0.45 μm filter (25 mm, VWR). Arsenic and Fe were analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) in the water laboratory at the Delft University of Technology, the Netherlands. The concentration of NH_4^+, NO_3^-, and PO_4^{3-} were determined from 250 ml filtered samples at Rajshahi Regional Laboratory, Department of Public Health Engineering (DPHE), Bangladesh.

The speciation of As(III) was conducted using the ion-exchange resin Amberlite® IRA-400 chlorite (Sigma Aldrich), by pushing 100 ml filtered (0.45 μm filter) water through a 60 ml syringe which contained 30 ml ion-exchange resin. After resin filtration, the remaining As concentrations were considered to be uncharged As(III) (Karori et al., 2006; Gude et al., 2016). The concentration of As(V) was calculated by deducting As(III) from the total As concentration obtained from the 0.45 μm filtered sample as mobile, dissolved As.

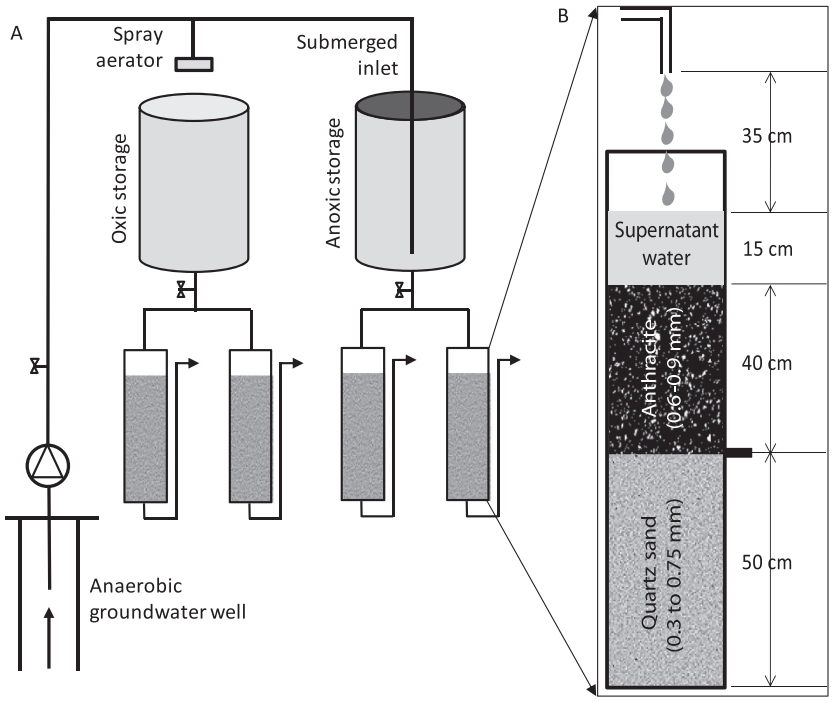

Fig. 2. Schematic overview of the dual-media sand filtration set-up. The anaerobic groundwater was exposed to atmospheric oxygen by a spray aerator before the oxic storage, whereas, the submerged inlet was used to fill the anoxic storage to avoid aeration. The pre-stored water was aerated by letting it drip into the column from the pipeline from 35 cm above the column top. The dual-media column experiments were performed in duplicates for each storage system.

2.4. Data analysis

The data analyses were conducted using one-way analysis of variance (ANOVA) with a confidence level of 95% (α=0.05) for the statistical validation of the removal efficiency of As, Fe, NH_4^+, and PO_4^{3-} during different filtration steps. The obtained duplicate data from each step and condition (oxic and anoxic) were used in duplicate assays from each sampling day (n=5) over the experimental 30 days. The data were presented in the form of mean with their standard deviations. The p-value (probability value) from the ANOVA test was used to determine the significant difference between the duplicate results from the two different operational conditions (oxic and anoxic storage) followed by aeration and RSF steps. The analyzed p-value was consistently below 0.05 for As, Fe, NH_4^+, and PO_4^{3-} removal at different operational conditions and filtration steps, meaning the removal was statistically significant.

3. Results and discussion

3.1. Fe^{2+} oxidation in the storage systems

The pH, DO, ORP, and Fe^{2+} concentrations in the oxic and anoxic storage systems are presented in Fig. 3. The natural groundwater pH, DO, and ORP were stable at 6.94(±0.2), 0.07(±0.06) mg/L, and −110 (±4) mV, respectively (Table 1). Due to aeration before storage in the oxic system, the pH, DO, and ORP increased to 7.5(±0.1), 6.21(±0.2) mg/L, and 50(±14) mV, correspondingly, since CO_2 was stripped (i.e., pH increase) and O_2 was added during aeration (Rahman, 2017). Furthermore, the aeration facilitated rapid oxidation of Fe^{2+} to Fe^{3+} and form HFO flocs. This was to be expected as the kinetics of homogeneous Fe^{2+} oxidation at pH 7.5 and high DO is fast ($t_{1/2}$: roughly 2–3 min) (Morgan and Lahav, 2007; Katsoyiannis et al., 2008a, 2008b). The total Fe concentration in the oxic storage system dropped slightly in the course of experiments and varied between 1.97 and 2.19 mg/L (Fig. 3d), which can be explained by the settling of HFO flocs in this non-stirred storage system. The 2.08 mg/L oxidized Fe^{3+} roughly corresponded to the consumed DO of 0.30 mg/L, which was calculated based on the 1:4 stoichiometry of the Fe^{2+} oxidation reaction with DO (Stumm and Lee, 1961), illustrating that DO was the electron acceptor under the oxic conditions.

In the anoxic storage, the continuously low pH, DO, and ORP resulted in limited Fe^{2+} oxidation. However, the pH and DO remained low over days, whereas ORP increased slowly with increasing Fe^{2+} oxidation to Fe^{3+}. The pH and DO were found to be 6.86(±0.04), 0.24 (±0.1) mg/L, respectively, but ORP drifted from −96.7 mV on the first day to −45.7 mV, −15.8 mV, 21.7 mV, and 48.6 mV on days 5, 10, 20 and 30, correspondingly (Fig. 3c). From the graphs, the observed increase in pH, DO, and ORP on day 5 compared to day 1 could have resulted from the dilution effect of newly added water with remaining stored water (one-third of the volume). Due to the stable operation of the systems over days, the measurements stabilized from day 5–10 onwards (Fig. 3a,c). The oxidation of Fe^{2+} also increased over time from 0.64 mg/L of oxidized Fe^{3+} on the first day to 0.9 mg/L on day 30, where the Fe^{2+} oxidation rate in the anoxic storage was 0.8 mg/Lh and 0.12 mg/Lh on day 1 and 30 correspondingly.

During the filling of the anoxic storage, the introduction of DO into the abstracted groundwater could not be fully avoided, which probably led to partial oxidation of Fe^{2+}. However, the acceleration of Fe^{2+} oxidation over days in the anoxic storage is likely linked to either accumulation of Fe^{3+} hydroxides that catalyze the oxidation reaction (heterogeneous Fe^{2+} oxidation; van Beek et al., 2015) or development of Fe^{2+} oxidizing biofilm (biological Fe^{2+} oxidation) (de Vet et al., 2011; Lin et al., 2012; Uddin et al., 2019). Nevertheless, Fe^{2+} oxidation in the anoxic system was indeed suppressed throughout the experimental period, allowing for delayed/step-wise oxidation.

3.2. As(III) oxidation and removal in the storage systems

Arsenic concentration and speciation in the oxic and anoxic storage systems are presented in Fig. 4. Partial As(III) oxidation and As removal was observed in both storage systems with slightly more oxidation and removal in the oxic storage. The groundwater contained 329(±0.05) μg/

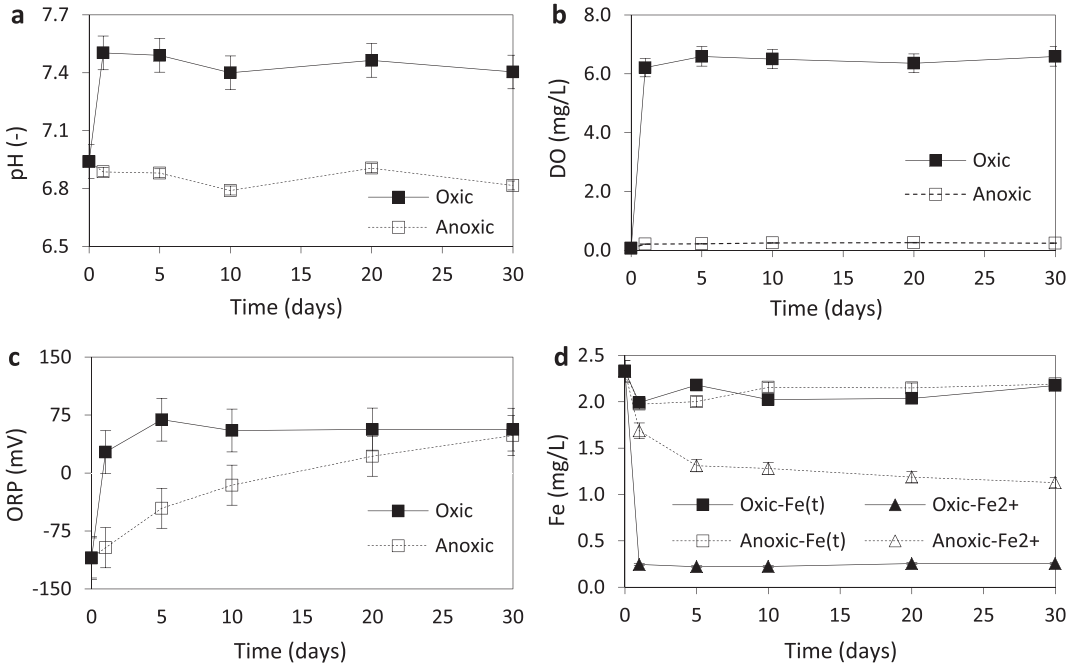

Fig. 3. (a) pH, (b) DO (c) ORP changes, and (d) the average concentration of Fe^{2+} and Fe(total) in the oxic and anoxic storage systems during the 30-day experimental period. The error bar represents the standard deviation.

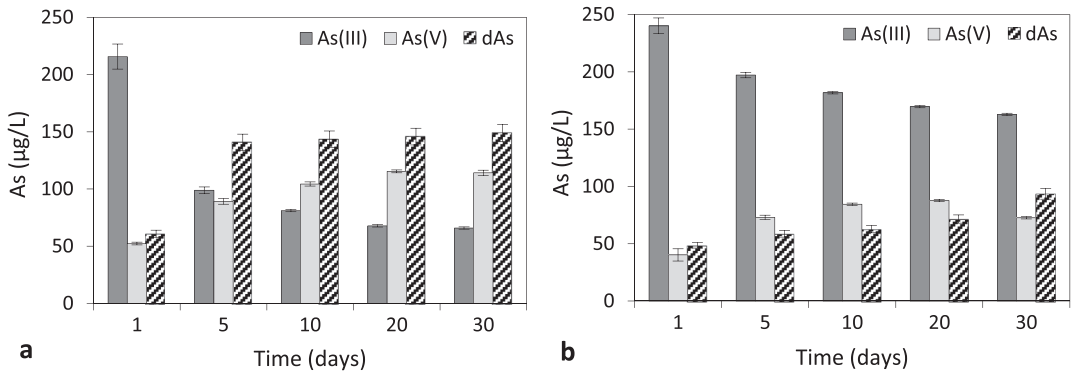

Fig. 4. Arsenic species (As(III), As(V), and removed As) and their average concentration in the duplicate (a) oxic and (b) anoxic storage systems. The error bar represents the standard deviation.

L of total As, with 290(±0.02) µg/L being As(III). On the first day of operation, the oxic storage system contained 267 µg/L of total As, in which As(III) was 216 µg/L (Fig. 4a); the anoxic system contained 280 µg/L of total As and 240 µg/L of As(III) (Fig. 4b). After 5–10 days, the operational mode was considered to be stable (see the previous section), also reflected in the stabilization in As(III) concentrations after 10 days, with an average As(III) concentration of 72(±7) µg/L and 171 (±8) µg/L in the oxic and anoxic storage respectively (Fig. 4).

The elevated average As(V) concentration of 95 µg/L (min. 52 µg/L to the max. 115 µg/L) in the oxic storage indicates the high level of As (III) oxidation under aerated conditions, which may be due to various oxidation processes, such as enhanced homogeneous oxidation (Bissen and Frimmel, 2003; Shumlas et al., 2016), biological As^{3+} oxidation (Quéméneur et al., 2008; Cavalca et al., 2013), oxidation by reactive intermediates ($^{\cdot}OH$, H_2O_2, and $O_2^{\cdot}$) formed during Fe^{2+} oxidation and/or Fenton-like (Fe(IV) based) reactions (Hug et al., 2001; Hug and Leupin, 2003; Roberts et al., 2004; Sahai et al., 2007; Ciardelli et al., 2008). The higher As removal in the oxic storage, compared to the anoxic storage, is likely to be a consequence of produced HFO flocs that bind both As(III) and As(V). The findings are in agreement with studies by Holm (2002), and Mercer and Tobiason (2008), who found that higher in-situ HFO flocs formation in the oxic storage during aeration resulted in higher As removal. After the first 5 days, the As removal in the oxic storage was nearly constant at 145(±2) µg/L, probably because an equilibrium was established between suspended and settled HFO flocs, while the latter did not further contribute to As removal. Over the experimental 30 days, it was observed that both Fe^{2+} and As(III) oxidation increased slightly, which could be induced by microbial activities (Shafiquzzaman et al., 2008; van Beek et al., 2012, 2015; Gude et al., 2018b).

In the case of the anoxic storage system, the low DO level (<0.25 mg/L) limited homogeneous As(III) oxidation (Bissen and Frimmel, 2003; Shumlas et al., 2016), rapid Fe^{2+} oxidation and formation of abundant reactive intermediates (e.g., $^{\cdot}OH$, H_2O_2, and $O_2^{\cdot}$), thus hampering As(V) formation. However, the observed As(III) concentration seemed to be slightly dropping over 30 days, likely due to the increase in Fe^{2+} oxidation over time (Fig. 4b). When calculating the ratio between oxidized Fe^{3+} and removed As on day 30, the amount of removed As per gram of Fe was higher in the system with anoxic storage (80(±4) µgAs/mgFe^{3+}) than in the conventional oxic storage (60(±5) µgAs/mgFe^{3+}), with a p-value of less than 0.05. The lower removal ratio in the oxic storage was in agreement with previous findings, where it has been stated that at pH above 7.0, As(V) removal with HFO flocs decreased considerably, due to lowering the positive surface charge, compared to systems with low pH (Wilkie and Hering, 1996; Katsoyiannis and Zouboulis, 2002; Dixit and Hering, 2003; Senn et al., 2018). Other studies also revealed that freshly formed HFO flocs in anoxic systems are more efficient for As removal as opposed to pre-formed HFO flocs in the traditional oxic storage systems (Kim and Nriagu, 2000; Senn et al., 2018).

3.3. Fe and As oxidation-removal after aeration

Over the experimental period of 30 days, after both the oxic and the anoxic storage, the groundwater was aerated by dipping from 35 cm above the filter bed. The dissolved Fe^{2+}, Fe^{3+}, As(III), and As(V) concentrations before and after aeration is shown in Fig. 5. After aeration before feeding the filtration units, the pH, DO and ORP for the oxic storage system was 7.7(±0.07), 6.7(±0.1) mg/L, and 61(±0.6) mV respectively and for the anoxic storage system, the pH, DO, and ORP was 7.4(±0.04), 5.83(±0.02) mg/L, and 45.7(±0.6) mV, correspondingly. The oxic storage water contained 2.08(±0.07) mg/L of total Fe, wherein 89% was HFO flocs (>0.45 µm), indicating no/limited Fe^{2+} was

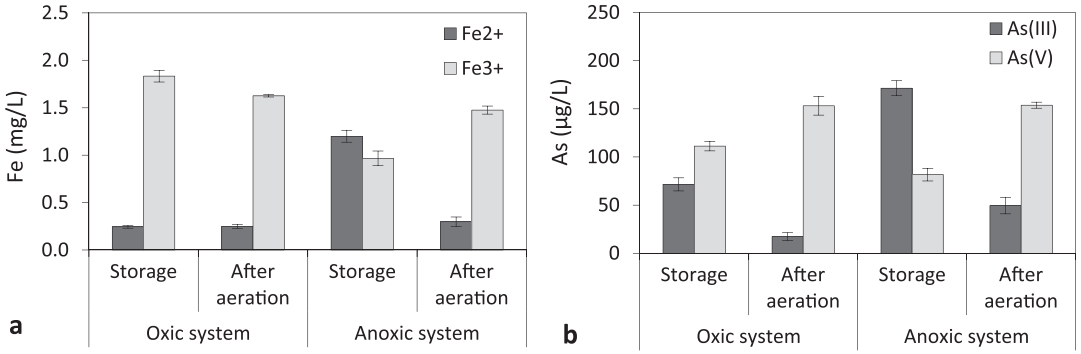

Fig. 5. The (a) Fe^{2+} and Fe^{3+}; and (b) As(III) and As(V) species (average) concentration in the oxic and anoxic storage system and after aeration before the filter bed. The error bar represents the standard deviation of duplicate column experiments.

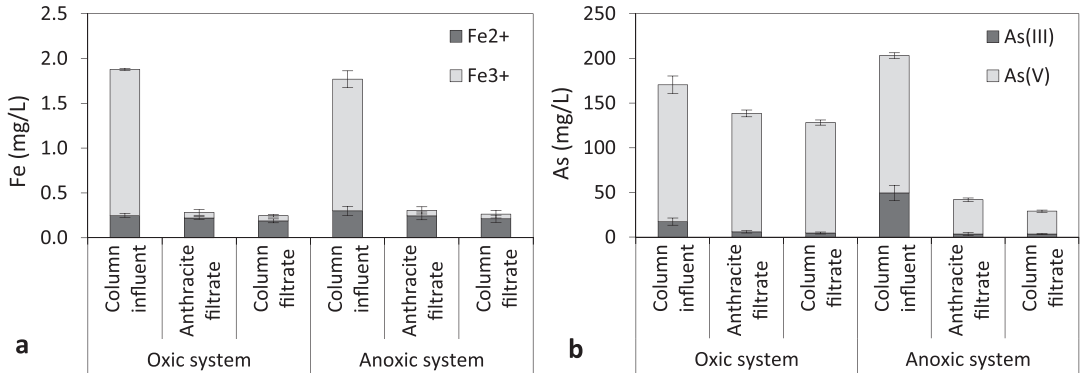

Fig. 6. The column influent, anthracite filtrate, and column filtrate (a) Fe^{2+} and Fe^{3+}; and (b) A(III) and As(V) species (average) concentration for the system with oxic and anoxic storage. The error bar represents the standard deviation of duplicate column experiments.

available for oxidation during the aeration before column feeding.

In the anoxic system, however, the introduction of O_2 during aeration before column feeding resulted in oxidation of the 1.20 mg/L of residual Fe^{2+} from anoxic storage (Fig. 5a). Subsequently, Fe^{3+} hydrolyzed to filterable/insoluble (0.45 μm membrane filter) Fe^{3+} (HFO) flocs. Along with Fe^{2+} oxidation, 120 μg/L of additional As(III) oxidation was also observed after this aeration step (Fig. 5b). In the oxic system, 55 μg/L of As(III) was also oxidized during the second aeration step, while Fe^{2+} was not present anymore, probably as a result of homogeneous As(III) oxidation by DO or bacterial processes that could be developed over time (Shumlas et al., 2016; Gude et al., 2018b). During filtration, a supernatant water level of 15–20 cm was maintained to provide a hydraulic head and a saturated filter bed. The water sample was collected from the supernatant water, which was in contact with the top layer of the filter bed, providing a potential carrier for bacteria and promoting biological oxidation of As(III), as earlier reported by Katsoyiannis and Zouboulis (2004) and Gude et al. (2018b). The aeration step prior to filtration, thus aided in partial As(III) oxidation and As removal in both oxic and anoxic systems. However, the system with anoxic storage showed a higher As removal than the oxic storage system ($p<0.05$), probably due to the presence of newly, in-situ formed active HFO flocs (Wilson et al., 2004; Mercer and Tobiason, 2008; Senn et al., 2018). The removed As after aeration (supernatant) of the oxic and the anoxic system was 12 μg/L (7%) and 50 μg/L (25%), respectively. This also confirmed the findings of the work of Roberts et al. (2004), who stated that multiple additions of Fe^{2+} lead to more As(III) oxidation and removal compared to a higher and single Fe^{2+} dose.

3.4. Fe and As removal in the filter bed

The dissolved Fe, As(III), and As(V) concentrations in the filtrate is shown in Fig. 6. For both the systems with oxic and anoxic storage, Fe removal was quick and efficient to result in 97% removal in the top anthracite layer. Independent of the storage systems before aeration-filtration, the filtrate Fe concentration was consistently lower than 0.3 mg/L (Fig. 6a), and thus below the WHO drinking water standard. Although the column influent concentrations of Fe^{2+}/Fe^{3+} and As(III)/As(V) were similar for both systems, As removal was considerably more effective in the columns that followed anoxic storage. The filtrate As concentration in the anoxic system was 28 μg/L, whereas in the oxic system it was 128 μg/L ($p<0.05$) (Fig. 6b). Hering et al. (1996), proposed that the varied As removal with in-situ and pre-formed HFO flocs results from their surface charge differences. According to model predictions by Holm (2002), the removal capacity of As by in-situ HFO flocs can be 3.8 times higher than that of preformed HFO flocs. Other studies also indicate that the increase in surface site density of in-situ formed HFO flocs (0.7 mol site/molFe) is higher than the pre-formed HFO flocs (0.205 mol site/molFe) (Fuller et al., 1993; Mercer and Tobiason, 2008). Furthermore, the lower filtrate pH of 7.1 in the anoxic system compared to the oxic system pH of 7.7 could be another reason for more As removal with adsorptive HFO flocs. The lower pH in the anoxic system slowed the HFO flocs formation (Katsoyiannis et al., 2008a; Gude et al., 2016; Zhang et al., 2019), and probably helped to increase the positive surface charge of HFO flocs, and thereby improved As removal (Wilkie and Hering, 1996). According to Mercer and Tobiason (2008), for 90% As removal in an ideal scenario, the required Fe:As ratio is about 9 and 6 at pH 7.3 and 6.2 respectively, where Holm's (2002) study mentioned that the pH difference by two units can increase or decrease As removal from 10% to 90%.

Apart from HFO floc filtration, the filter bed probably also functioned as a bio-filter for As(III) oxidation by AsOB (Shafiquzzaman et al., 2008; Gude et al., 2018b). In both storage systems, the residual As(III) that enters the filter bed was fully oxidized to As(V) in the anthracite layer perhaps by the presence of AsOB in the filter bed (Gude et al., 2018b). They also indicated that the penetration of freshly or in-situ formed HFO flocs into the filter bed for the anoxic system facilitates more As(V) adsorption compared to pre-formed HFO flocs penetration for the oxic system (Wang et al., 2008; Amstaetter et al., 2010; Hohmann et al., 2010). Similar to Gude et al. (2016), the rapid removal of HFO flocs in the first layer resulted in the low As removal in the bottom sand layer (3–7 μg/L).

3.5. Ammonium, nitrate, and phosphate removal

Over the experimental period of 30-day, the concentrations of NH_4^+ and NO_3^- in different treatment steps of the system with oxic and anoxic storage are shown in Fig. 7. In groundwater, the NH_4^+ and NO_3^- concentrations were, on average, 0.96(±0.02) mg/L and 0.39(±0.02) mg/L, respectively (Table 1). After the first five days, the concentration of NH_4^+ decreased in the oxic system due to the onset of biological nitrification, resulting in NO_3^- concentration increase by aerobic oxidation of NH_4^+ (Koch et al., 2019). The lack of DO in the anoxic storage system limited the NH_4^+ oxidation process, consequently, 0.79(±0.05) mg/L of NH_4^+ remained in the storage system. After 5–10 days, the oxidation of NH_4^+ was observed to commence in the filter bed for both the oxic and anoxic systems, as can be observed from the lowering of NH_4^+ and increase in NO_3^- concentration in the filter bed (anthracite) (de Vet et al., 2011; van Kessel et al., 2015; Koch et al., 2019). However, the results did not imply a 1:1 NH_4^+ conversion to NO_3^- (N-Balance), probably meaning that the nitrification process was not complete, resulting in (intermediate) nitrite (NO_2^-) production. However, it may be concluded that the nitrification process, although delayed in start-up, was not hampered by the step-wise Fe^{2+} oxidation sequence in the anoxic system.

The PO_4^{3-} concentrations in the stored water and different treatment

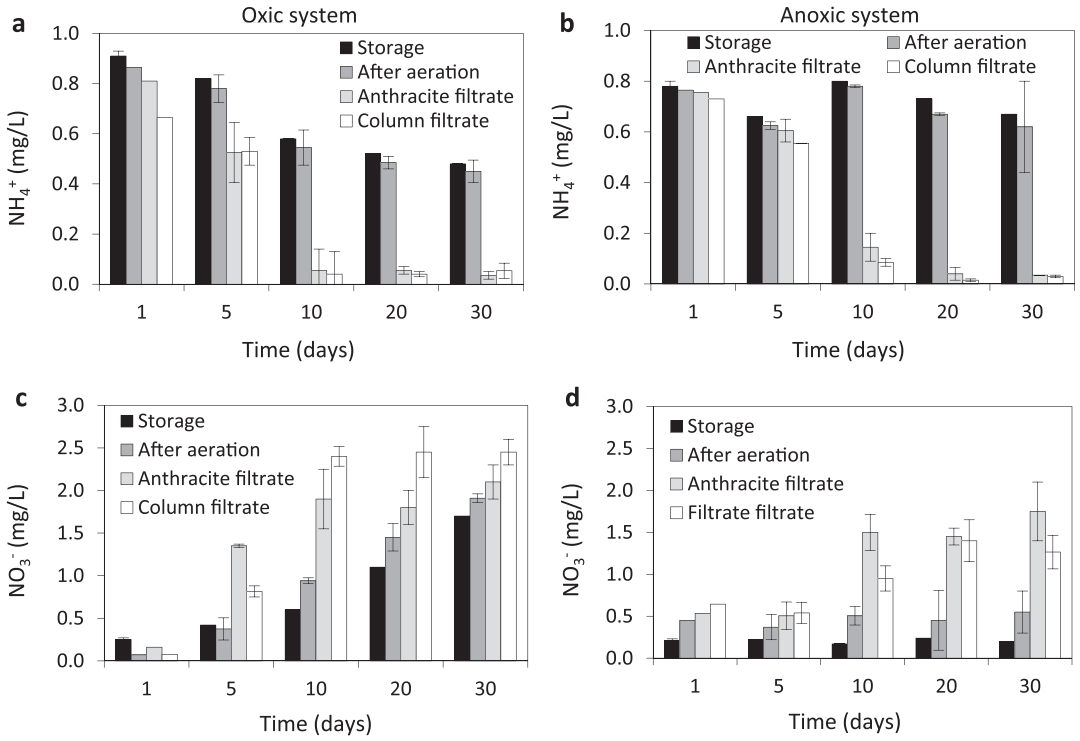

Fig. 7. The average concentration of (a-b) NH_4^+ and (c-d) NO_3 at different treatment steps of the system with oxic (a, c) and anoxic (b, d) storage respectively. The error bar represents the standard deviation of duplicate dual-media sand filtration column experiments.

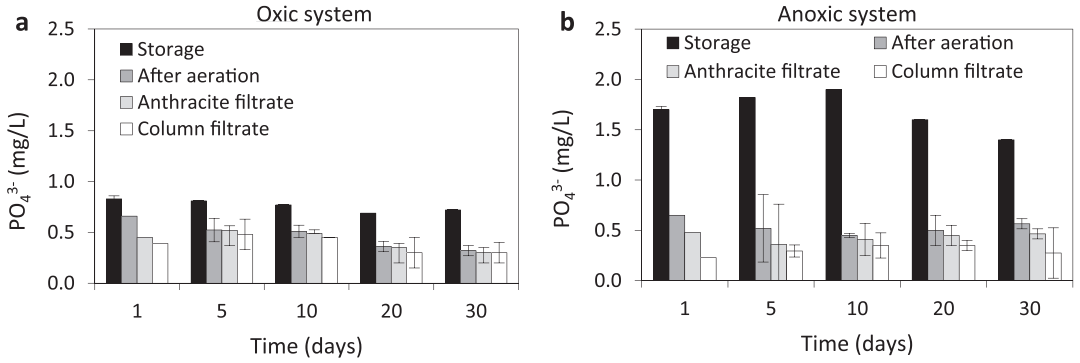

Fig. 8. The average PO_4^{3-} concentration at different treatment steps based on (a) oxic and (b) anoxic storage system. The error bar represents the standard deviation of duplicate column experiments.

steps over time are depicted in Fig. 8. In the groundwater, the PO_4^{3-} concentration was 2.15(±0.03) mg/L, which dropped to an average of 0.76(±0.05) mg/L in the oxic storage system (Fig. 8a). However, in the anoxic storage system, PO_4^{3-} concentration remained high at 1.68 (±0.22) mg/L (Fig. 8b). The large decrease of PO_4^{3-} concentration in the oxic storage system, compared to the anoxic storage system ($p<0.05$), can be explained by co-precipitation with HFO flocs (Guan et al., 2009; Voegelin et al., 2010) originating from rapid Fe^{2+} oxidation. In the anoxic storage followed by aeration-filtration, the PO_4^{3-} removal follows the step-wise Fe^{2+} oxidation. PO_4^{3-} was partially removed during anoxic storage (0.47±0.18 mg/L) and aeration (1.14±0.06 mg/L) before filtration (Fig. 8b). The removal of PO_4^{3-} was also observed at different layers of the filter bed (aerated water vs. anthracite filtrate vs. column filtrate), indicating ongoing adsorption of PO_4^{3-} onto HFO flocs. The concentration of PO_4^{3-} further decreased in the column filtrate to 0.38 (±0.07) mg/L and 0.30(±0.05) mg/L from anthracite filtrate concentration of 0.45(±0.06) mg/L and 0.43(±0.04) mg/L respectively for the oxic and anoxic systems.

4. Considerations for application

The suitability of the As co-precipitation with naturally present Fe^{2+} by anoxic storage followed by aeration-sand filtration system was evaluated in Bangladesh based on the five criteria proposed by WHO (2013), which are effectiveness, appropriateness, acceptability, cost, and implementations.

In Bangladesh, people prefer using tubewells to other drinking water options due to low operation and maintenance costs, and water accessibility around the year (Hossain et al., 2014). With the proposed treatment method, it is possible to achieve As removal without using any additional chemicals. Moreover, it uses only locally available materials and works with an existing groundwater source. Previous studies suggested Fe/As ratios over 40 (mg/mg) would be required to reduce As levels below 50 μg/L (Meng et al., 2002; Ware, 2013) with passive

treatment. The present study indicates that delayed aeration-oxidation of Fe^{2+} could be a promising method for As removal with Fe/As ratios as low as 10 (mg/mg). However, from a waste management perspective, it is recommended to apply this treatment scheme on a larger scale. Although the combination of anoxic storage with aeration–filtration requires a low As/Fe ratio, still As-containing sludge is being produced which needs to be disposed of. At the concentrations found during this study, As-containing Fe sludge can be used in brick industries or concrete masonry work (Rouf and Hossain, 2003). Prior to the application of this novel technology, it is recommended to further investigate the influence of groundwater matrices and varying operational parameters, which might impact As removal efficiency.

5. Conclusion

This study aimed to investigate the influence on As removal efficiency by delayed aeration using an anoxic pre-storage system followed by aeration and dual-media sand filtration. This novel treatment sequence was compared against conventional aeration-filtration for groundwater containing total As of 329 μg/L, with 290 μg/L being As (III) and 2.33 mgFe^{2+}/L. The obtained step-wise Fe^{2+} oxidation with anoxic pre-storage enhanced As removal to 92%, compared to only 61% in the conventional oxic system. It is suggested that this was due to the formation of fresh HFO flocs during step-wise oxidation in the anoxic system that removed more As than the pre-formed HFO flocs in the oxic system. Moreover, Fe^{2+} oxidation did not compromise the removal of other groundwater contaminants like NH_4^+, and PO_4^{3-}. Therefore, this study demonstrated that with anoxic storage, passive treatment is an effective barrier against As(III) with Fe/As ratios much lower (<10 mg/mg) than previously determined (>40 mg/mg). Consequently, the combination of anoxic storage followed by aeration and sand filtration can be used as a cost-effective and chemical-free alternative for removing As(III) from groundwater under field-relevant conditions.

CRediT authorship contribution statement

Md Annaduzzaman: Conceptualization; Methodology; Investigation; Data curation; Analysis; Data visualization; Validation; Software; Writing- Original Draft Preparation, **Luuk C. Rietveld:** Supervision; Writing- Reviewing and Editing, **Bilqis Amin Hoque:** Supervision; Writing- Reviewing and Editing, **Md Niamul Bari:** Writing-Reviewing and editing; Experiments- Fieldwork supervision and organization, **Doris van Halem:** Supervision; Project leader; Experiments- Conceptualization and guidance on design; Data visualization; Writing-Reviewing and Editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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