

## Removal of arsenic and iron removal from drinking water using coagulation and biological treatment

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### ABSTRACT

Effects of biological activated carbon (BAC), biological aerated filter (BAF), alum coagulation and *Moringa oleifera* coagulation were investigated to remove iron and arsenic contaminants from drinking water. At an initial dose of 5 mg/L, the removal efficiency for arsenic and iron was 63% and 58% respectively using alum, and 47% and 41% respectively using *Moringa oleifera*. The removal of both contaminants increased with the increase in coagulant dose and decrease in pH. Biological processes were more effective in removing these contaminants than coagulation. Compared to BAF, BAC gave greater removal of both arsenic and iron, removing 85% and 74%, respectively. Longer contact time for both processes could reduce the greater concentration of arsenic and iron contaminants. The addition of coagulation (at 5 mg/L dosage) and a biological process (with 15 or 60 min contact time) could significantly increase removal efficiency, and the maximum removal was observed for the combination of alum and BAC treatment (60 min contact time), with 100% and 98.56% for arsenic and iron respectively. The reduction efficiency of arsenic and iron reduced with the increase in the concentration of dissolved organics in the feedwater due to the adsorption competition between organic molecules and heavy metals.

**Key words** | arsenic, biofilter, coagulation, dissolved organic matter, iron

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### INTRODUCTION

The presence of heavy metals in the water environment has been a major concern for years because of their high acute and chronic toxicity. Approximately 140 million people throughout the world are affected by the consumption of arsenic-contaminated water (Ravenscroft *et al.* 2009). The major means of arsenic entering the body is through drinking water, hence humans may be affected by serious health problems including those of the gastrointestinal tract, cardiac, vascular and central nervous system (Lacasa *et al.* 2011). Therefore, the removal of these contaminants is required to eliminate potential health risks and threats.

Several treatments, such as membrane technology, coagulation and biological processes, have been widely used for water and wastewater treatment. It has been demonstrated that the filtration process can reduce metal contaminants to some extent. Litter *et al.* (2010) found that

low pressure membranes such as microfiltration or ultrafiltration are not very effective for removing arsenic contamination, as the arsenical species are very small and thus traverse the membranes. On the other hand, high pressure membranes can completely reject polyvalent ions in particular, and are suitable for arsenic oxyanions (Ravenscroft *et al.* 2009). However, this process can consume huge amounts of power, hence economically visible, reliable, easy to operate technology is urgently required.

Chemical coagulation treatment is effective and widely used for the removal of organics and particulate matter from water and wastewater because of its availability, low cost and good treatment performance, and it can be used for iron and arsenic removal from water and wastewater. Parga *et al.* (2005) found that ferric coagulant could reduce up to 90% of arsenic. However, chemical coagulation

produces large amounts of chemical sludge. An alternative natural coagulant such as *Moringa oleifera* (*M. oleifera*) seeds seems to be a more cost-effective, viable and environmentally-friendly approach for such an application, because the production of sludge was four to five times less using *M. oleifera* than alum coagulant (Abaliwano *et al.* 2008; Pise & Halkude 2012). Mohamed *et al.* (2014) found that *M. oleifera* seeds reduced the turbidity, COD and phosphorus of wastewater by 90%, 60% and 75%, respectively. Another study by Pramanik *et al.* (2015a) found that *M. oleifera* could reduce 23% of dissolved organic carbon (DOC), 42% of ultraviolet absorbance at 254 nm (UV<sub>254</sub>), 76% of turbidity and 60% of colour from drinking water. The uses of this natural coagulant for arsenic and iron removal from drinking water have not yet been investigated.

Biological processes are also widely used in water and wastewater treatment processes. Biological water treatment processes are better than chemically assisted technologies and are a more cost-effective technology to remove metal content from water and wastewater. It has been suggested that the operational and capital costs of biological treatment processes are 3–10 and 5–20 times lower, respectively, than chemical processes (Marco *et al.* 1997). Lipton *et al.* (1994) found that 99% removal of arsenic from groundwater by combined anaerobic and aerobic bioreactors was observed. Biological activated carbon (BAC) treatment was highly effective in removing both arsenic and iron, with a removal efficiency of 97% and 99.8%, respectively (Pokhrel *et al.* 2005). They also reported that the rapid sand filter removed approximately 50% of arsenic and 99% of iron. Biological aerated filters (BAF) have been studied for water and wastewater treatment due to the large surface area available for the growth of biofilm (Ryu *et al.* 2008; Biplob *et al.* 2011a, 2011b; Pramanik *et al.* 2012). However, the BAF process is mainly used for removing carbonaceous organics and biological nutrients from water and wastewater. However, there is no comparison for the removal of the contaminants by different treatment processes. Moreover, the influence of the presence of organic matter in drinking water during treatment processes needs to be evaluated.

The objective of this study was to compare the removal efficiency of arsenic and iron by alum and BAC treatment processes. The natural coagulant, *M. oleifera*, was compared with conventional alum coagulant, and BAF was compared

with the BAC treatment process. During the coagulation process, the samples were coagulated with different dosages and pH to optimize the process performance for iron and arsenic removal. The effect of contact time on biological treatment for arsenic and iron removal was investigated. An assessment of the effect of the concentration of organics during iron and arsenic removal using biological and coagulation processes was also undertaken.

## MATERIALS AND METHODS

### Sample collection

The samples were collected from a reservoir. Samples were stored at 4 °C and warmed to room temperature (22 ± 2 °C) prior to all experiments. Different concentrations of dissolved organic matter (humic and fulvic acid were used, and supplied by Sigma-Aldrich) were added to feedwater to investigate the impact of the concentration of organics on arsenic and iron removal.

### Treatment processes

#### Coagulation

The chemical coagulant alum (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·18H<sub>2</sub>O) was used in this study. *M. oleifera* was used as a natural coagulant. The seeds of *M. oleifera* were extracted from the dry pods, ground into a fine powder using a blender and then sieved through a 0.8 mm mesh. Afterwards, Milli-Q water was added to the fine powder to make a 1% suspension. The detailed procedure can be found elsewhere (Pramanik *et al.* 2015a).

Both alum and *M. oleifera* coagulation experiments were carried out with 1 L feed water using a jar test apparatus (Phipps and Bird, PB-900). Prior to all types of coagulant experiments, the organics were added into the feed water and then mixed with a stirrer for 30 min. The samples were mixed for 5 minutes at 300 rpm and then mixed for the next 20 minutes at 40 rpm. After 1 hour, the supernatant was tested to determine the amounts of arsenic and iron. This experiment was performed with a range of doses (5–25 mg/L) and pH (4–6) conditions to optimize

the conditions for the target compounds' removal. The pH was adjusted by using 0.1 N HCl or 0.1 N NaOH.

### Biological processes

The BAF was constructed in a PVC chamber with an internal diameter of 8 cm and an effective media height of 60 cm. Plastic media (Kaldness) with a length, diameter, density and an internal surface area of 3 mm, 5 mm, 0.42–0.46 g/cm<sup>3</sup> and 305 m<sup>2</sup>/m<sup>3</sup>, respectively, were used for the BAF.

The BAC column was constructed of glass, with an internal diameter and effective carbon bed height of 4.5 cm and 50 cm, respectively. The surface area, total pore volume and micropore volume of the activated carbon used were 800 m<sup>2</sup>/g, 0.865 cm<sup>3</sup>/g and 0.354 cm<sup>3</sup>/g, respectively.

Prior to packing for both processes, the medium was inoculated with activated sludge, and provided with additional nutrient sources (N, P and C) to promote the rapid growth of biofilm on the media. Afterwards, it was gently washed with tap water to remove excess biofilm and transferred to the system, and feed was commenced. DOC removal was stable after 35 and 40 days of BAC and BAF operation, respectively, indicating that equilibrium had been established.

Both processes were operated in a downflow mode with an empty bed contact time of 15, 30, 45 and 60 min. The processes were backwashed for 20 min every week to avoid physical clogging.

### Analytical methods

DOC concentration was determined using a total organic carbon (TOC) analyser (TOC-5000A, Shimadzu). Potassium hydrogen phthalate (1, 5, 10 and 25 mg C L<sup>-1</sup>) was used for calibration of the TOC analyser. Before these analyses, all samples were filtered through a 0.45 µm filter. The pH of the water samples was measured using a pH meter (Mettler Toledo). The pH meter was calibrated with standard solutions of pH 4, 7 and 10. Zeta potential measurements were measured using a Zetasizer Nano-ZS (model ZEN3500).

The concentration of iron and arsenic was determined using atomic absorption spectrometry (Shimadzu AA6800).

The limit of quantification for arsenic and iron was 1 µg/L and 20 µg/L, respectively. Prior to the analysis, the samples were filtered through 0.45 µm cellulose acetate filters. All filtration analyses were duplicated and reported in terms of mean value.

## RESULTS AND DISCUSSION

### Coagulation process for arsenic and iron removal

#### Effect of coagulant dosages

The effect of coagulant dosages on the removal of arsenic and iron is shown in Figure 1. At the initial dose of 5 mg/L, the average removal efficiency of arsenic and iron was 63% and 58% using alum, respectively. *M. oleifera* led to a reduction in arsenic and iron of 47% and 41%, respectively. A similar result was reported by Pokhrel *et al.* (2005), who found that coagulation was effective in removing an average of 70% of arsenic and 85% of iron.

This was attributed to the adsorption of these contaminants into the flocs, and precipitation of some of these contaminants with precipitates (Edwards 1994; Lytle *et al.* 2005). Moreover, some of the arsenic (particularly As(V)) species are negatively charged (H<sub>2</sub>AsO<sub>4</sub> and HAsO<sub>4</sub>),

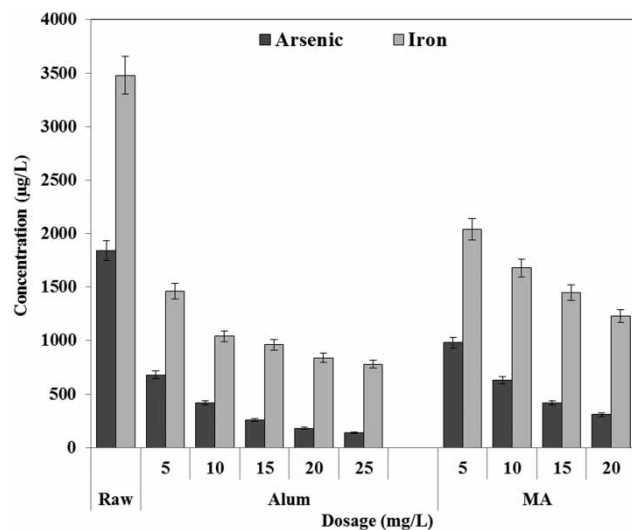


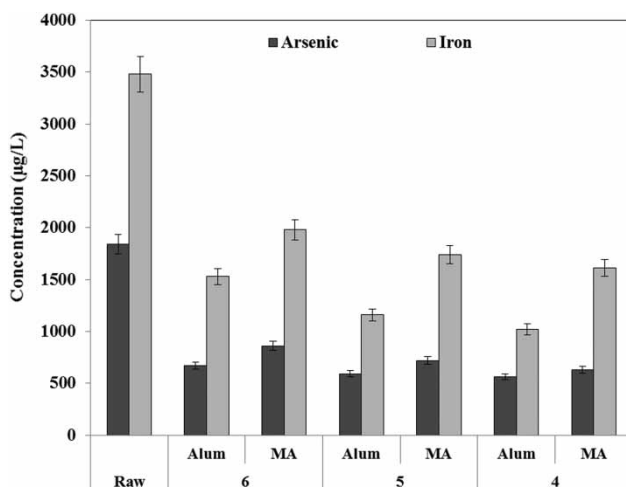
Figure 1 | Effect of the coagulant dosages on the removal of arsenic and iron (at an initial DOC concentration of 5.24 mg/L, pH 6.96).

therefore positively charged coagulant could alter or destabilize negatively charged arsenic content. There was a greater removal of these molecules by chemical coagulant than natural coagulant. This was attributed to the development of a greater settleable floc area, as confirmed by zeta potential measurement, resulting in greater adsorption. All coagulants achieved a considerably higher removal of arsenic than iron. This was possibly due to the molecular size of arsenic, which is larger than iron, therefore leading to a greater adsorption of arsenic to floc than iron.

The concentration of iron and arsenic decreased with increasing coagulant dose. With the increase in coagulation dosage from 5 to 25 mg/L of alum, the removal efficiency increased from 63% to 92% for arsenic and from 58% to 78% for iron. This was attributed to the greater dose allowing the destabilization of the colloidal suspension and the formation of flocs. Similarly to alum, *M. oleifera* had a similar trend in removing arsenic/iron concentrations with increasing dosage, but the removal efficiency was lower compared with alum. This was possibly related to the lower increment of the floc area, suggesting that the removal efficiency was influenced by the size of the flocs.

### Effect of pH

The effect of pH on arsenic and iron removal by coagulation was examined in the pH range of 4 to 6 at dosages of 5 mg/L (Figure 2). Arsenic removal increased when the pH

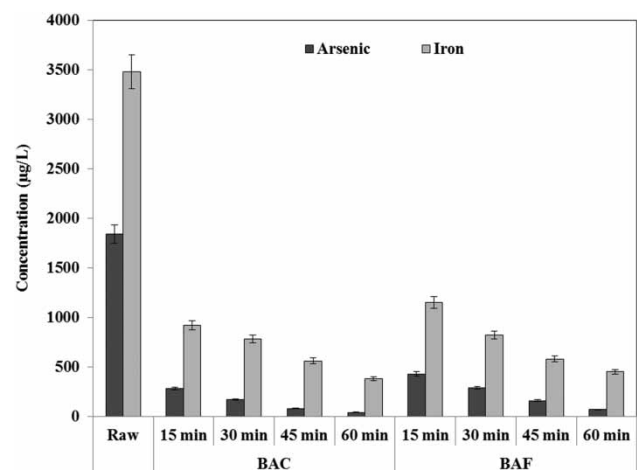


**Figure 2** | Effect of the coagulation pH on the removal of arsenic and iron (at an initial DOC concentration of 5.24 mg/L, and a coagulant dosage of 5 mg/L).

decreased. Arsenic removal was about 63% at pH 6, while arsenic removal efficiency with the same dosage could reach 72% at a pH of 4. The greater removal of arsenic at a lower pH was likely due to the increase in the available surface area for adsorption/enmeshment. Moreover, Ghurye *et al.* (2004) stated that the increase in pH led to a decrease in the number of positively charged hydroxide ferric surfaces and to an increase in the numbers of OH<sup>-</sup> ions, which are a barrier for arsenic adsorption by hydroxide ferric surfaces. Similarly to conventional coagulation processes, the efficiency of this process increased with a decrease in pH. This was likely due to the cationic nature of the *M. oleifera* extracted protein (Kansal & Kumari 2014).

### Effect of biological processes

BAC was effective in reducing the iron by 85% at 15 min contact time (Figure 3). This was attributed to the biological oxidation of some iron content; others were coated to the media and to the biofilm. Similarly to BAC, BAF was also very effective in reducing this contaminant, although the removal efficiency was lower than the BAC process at the same short contact time. Similarly to iron removal, both processes are effective in removing arsenic contamination from water, and the trend of the removal was similar with the treatment processes. A similar observation was made by Lehimas *et al.* (2001), who reported that sand filtration reduced the arsenic content from 400 mg/L to 50 mg/L.



**Figure 3** | Effect of biological processes on arsenic and iron removal (at an initial DOC concentration of 5.24 mg/L, pH 6.96).

With the increase in contact time, both processes led to an increase in the reduction of arsenic and iron concentration, with BAC giving a greater removal than BAF. Both treatments led to a greater reduction of arsenic than iron. This was likely due to the greater solubility of arsenic and the degree of attraction by water molecules, which are prevented from being bound by the carbon surface.

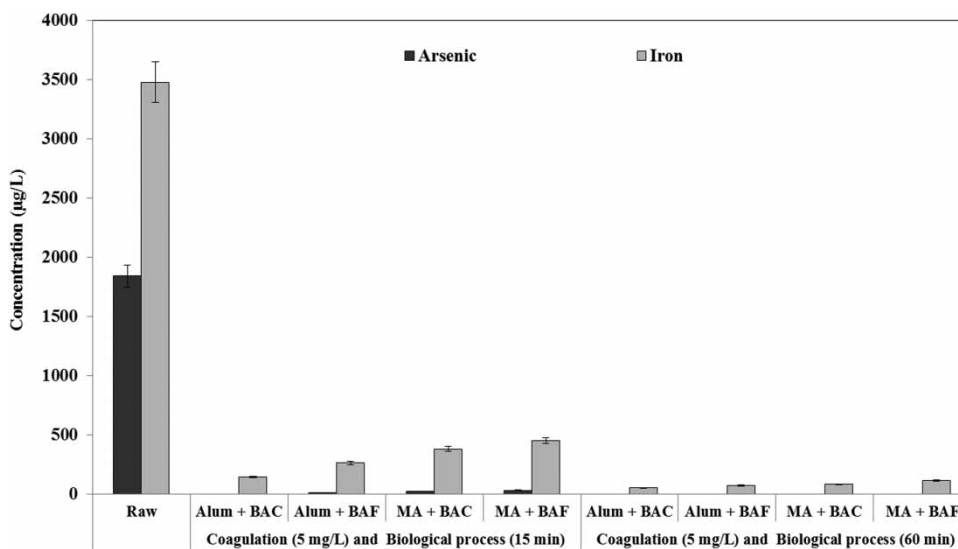
### Combination of coagulation and biological processes

The effect of the combination of coagulation and a biological process was observed to enhance the removal efficiency of arsenic and iron content from drinking water (Figure 4). The addition of the alum coagulation (with an unadjusted pH and a dosage of 5 mg/L) and BAC (15 min contact time) gave a removal efficiency for arsenic and iron of 100% and 96%, respectively. A similar trend was observed in reducing these contaminants with a combination of *M. oleifera* and BAC under the same conditions. This was likely due to the precipitation of this contaminant during the coagulation process and the residual concentration being adsorbed onto the biofilm in the BAC process. Similarly to the condition of BAC and coagulation, the addition of BAF with all coagulants led to the arsenic and iron removal being enhanced compared to the standalone process. Therefore, it can be concluded that the combination of coagulation and a biological process is

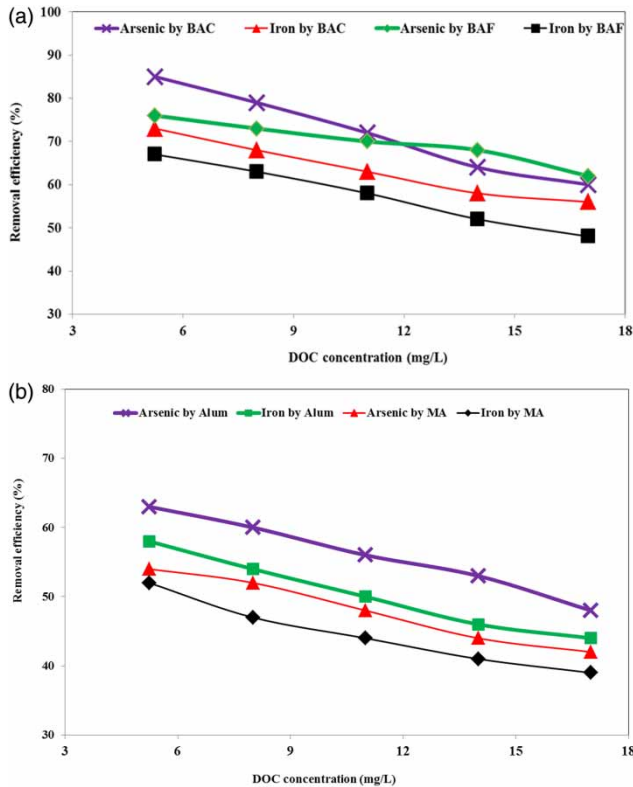
an efficient way to remove these chemicals from drinking water and wastewater.

### Effect of dissolved organic carbon on arsenic and iron removal

The presence of organic matter had a significant effect on arsenic and iron removal during coagulation and the biological process. A greater DOC concentration in the feed water reduced the arsenic and iron removal efficiency. Compared to BAF, the reduction efficiency for arsenic/iron was significantly decreased using BAC treatment when the DOC concentration of the feed was increased (Figure 5(a)). This was likely due to physical blocking of the pores of the activated carbon in the BAC process by organic matter; hence it was unable to sorb other compounds. A similar observation was made by Zeng *et al.* (2008), who found that the presence of silica in the samples had an effect of arsenic removal using iron oxide-based packed columns. A study by Matsui *et al.* (2002) reported that the removal efficiency of pesticides and pharmaceuticals was reduced by activated carbon when organic matter was present in the feed water. Pramanik *et al.* (2015b) also reported the finding that the reduction efficiency of perfluorooctanoic acid and perfluorooctane sulfonate was decreased by activated carbon treatment when the DOC concentration of the feed was



**Figure 4** | Effect of the combination of coagulation and biological process on arsenic and iron removal (at an initial DOC concentration of 5.24 mg/L, a pH 6.96, a coagulant dosage of 5 mg/L and different contact times for BAC/BAF such as 15 min and 60 min).



**Figure 5** | Effect of DOC concentration on arsenic and iron removal efficiency using (a) biological treatment (contact time 15 min) and (b) coagulation (5 mg/L).

increased. This indicates that competition for adsorption between organics and target compounds was involved. Similarly to biological processes, the presence of a greater concentration of organics in the feed water decreased the performance of the coagulation process for arsenic and iron removal (Figure 5(b)). It should also be noted that the addition of DOC had more effect on the removal of iron than arsenic contamination.

### Risk assessment of arsenic and iron

The arsenic and iron content in the untreated water was 1,840  $\mu\text{g/L}$  and 3,480  $\mu\text{g/L}$ , respectively. The World Health Organization (WHO) has set an allowable limit for arsenic in drinking water for human consumption of 10  $\mu\text{g/L}$  (World Health Organization 1993). Canadian Drinking Water Guidelines (1996) also set an allowable limit of 25  $\mu\text{g/L}$  arsenic and 300  $\mu\text{g/L}$  iron for drinking water. Hu *et al.* (2012) reported that China's newly revised drinking water standards for the maximum contaminant level of arsenic have reduced from

50 to 10  $\mu\text{g/L}$ . However, only the combination of coagulation (5 mg/L) and a biological process (60 min contact time) treated water could meet the allowable limit of the arsenic and iron concentration for drinking water.

### CONCLUSIONS

This study compared the performances of BAC, BAF and coagulation processes for removal of arsenic and iron from drinking water treatment in the presence of organic matter. Coagulation could remove arsenic and iron from drinking water, and the removal was achieved by precipitation and adsorption onto the surface of flocs via both electrostatic and hydrophobic interaction. Alum performed better in removing these compounds than *M. oleifera*. This was likely due to the greater floc area, resulting in greater adsorption. Both iron and arsenic removal strongly depended on coagulant dose and to a small extent on coagulation pH. There was a greater reduction of arsenic than iron by the coagulation process as the molecular size of arsenic is larger than iron. Biofilters were more effective for removing these contaminants than coagulation. BAC gave greater removal of arsenic and iron contaminant than BAF. The addition of coagulation before the biological process further reduced the concentration of the target contaminant. When the concentration of DOC increased, the removal efficiency of arsenic and iron decreased because of the steric hindrance effect of organic molecules and competitive adsorption. Overall, this study demonstrated that new BAF and a natural coagulant could reduce iron and arsenic contamination in reservoir water, and the combination of coagulation and biological process at 60 min contact time could meet the allowable limit for drinking water.

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