



Arsenic removal from simulated groundwater using household filter columns containing iron filings and sand



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ABSTRACT

Recent investigations have found zero-valent iron [Fe(0)] as promising material for removal of arsenic from groundwater. This study aimed at improving the arsenic removal efficiency of a household level filter unit containing iron filings and sand. A novel methodical approach was used to assess the aeration (oxygen transfer) capacity of the filter unit. The calculations suggest that the designed unit possessed sufficient capacity to oxidize ferrous ion to ferric ion and resulted in co-oxidation of arsenite (As^{3+}) to arsenate (As^{5+}). The modifications made to the system reduced the problems of clogging, excessive headloss development and enabled prolonged leaching of iron leading to better arsenic removal efficiency. Under laboratory conditions, the modified unit was able to treat simulated groundwater containing 500 $\mu\text{g/L}$ of As^{3+} , 2 mg/L of P, and 20 mg/L of Si to arsenic levels below WHO standard of 10 $\mu\text{g/L}$ without adding any chemical oxidant. The Fe/As ratio of 88.46 was required to bring the arsenic concentrations below 10 $\mu\text{g/L}$ which is much less than that are achieved by other researchers under similar conditions. The removal of arsenic by zero-valent iron [Fe(0)] is attributed to adsorption by iron hydroxides generated from the oxidic corrosion of Fe(0) and/or precipitation of iron arsenate within the system. Results indicated that a filtration system consisting of iron filings and sand could be effectively used at household level to bring the total arsenic levels below WHO standard of 10 $\mu\text{g/L}$ in drinking water.

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1. Introduction

Arsenic is a toxic compound found ubiquitously in nature. In natural ground water, it occurs mostly as arsenite [As^{3+}] and arsenate [As^{5+}]. As^{3+} is reported to be 25–60 times more toxic than As^{5+} and several hundred times more toxic than methylated arsenicals [1]. Arsenic contamination of groundwater and associated health risks have been reported in many parts of the world such as Bangladesh [2–5]; India [6,7]; China [8]; Taiwan [9]; United States [10]; Japan [11]; Argentina [12]; and Vietnam [13], and thus, its presence in drinking water is a worldwide concern. Arsenic removal is often challenging due to high arsenite, phosphate, and silicate concentrations and low natural iron concentrations.

Various treatment technologies have been developed for arsenic removal from drinking water. Commonly used technologies include coagulation and precipitation with iron and aluminum salts [14], adsorption onto activated alumina and activated carbon, ion

exchange and reverse osmosis [15]. Some other technologies are based on use of iron oxide coated sand [16], manganese dioxide coated sand [17], and zero-valent iron [Fe(0)]. However, these techniques have been found to be not as efficient for As^{3+} removal as for As^{5+} removal. Therefore, for efficient arsenic removal, chemical oxidation of As^{3+} – As^{5+} has often been suggested.

The use of Fe(0) to remove arsenic has been investigated by many researchers [18–22] for which the surface area of iron was found to play a major role in both the adsorption kinetics and removal capacities. Column filtration experiments have been conducted by a few investigators to evaluate the effectiveness of Fe(0) for arsenic removal [20,23,24]. Nikolaidis et al. [21] reported that the removal rates of arsenic were up to 10 times faster near the inlet end of the iron column than near the effluent end. The faster removal was attributed to rapid oxidation of Fe(0) by small amounts of dissolved oxygen (DO) in the influent. On the contrary, Ramaswami et al. [25] reported that the presence of air in batch reactors hindered arsenic removal by Fe(0). Many other column filtration experiments reported in the literature have been conducted under anoxic conditions and at relatively long hydraulic contact time. The effect of DO content of water on arsenic removal by Fe(0) filters has not been investigated systematically.

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Bang et al. [22] utilized zero-valent iron filings for arsenic removal. The removal was dramatically affected by oxygen content, and pH wherein, arsenate removal was faster than arsenite under oxic conditions. Greater than 99.8% of the As^{5+} was removed whereas 82.6% of the As^{3+} was removed at pH 6 after mixing for 9 h. When dissolved oxygen was removed by nitrogen purging, less than 10% of the As^{3+} and As^{5+} was removed. Other studies conducted to evaluate the potential of arsenite oxidation by dissolved oxygen alone were found to be inefficient in the absence of $\text{Fe}(0)$. In the presence of iron though, high dissolved oxygen content and low solution pH increased the iron corrosion rate. Thus, arsenic removal by $\text{Fe}(0)$ in the presence of dissolved oxygen was attributed to adsorption onto iron hydroxides generated from $\text{Fe}(0)$.

Leupin et al. [26] investigated smaller filter columns with 4 filters containing 2.5 g of iron filings and 100–150 g of sand. The filters treated 36 L of synthetic groundwater with initially 500 $\mu\text{g/L}$ of As^{3+} and 2–3 mg/L of P, and 75–90 L of well water containing 440 $\mu\text{g/L}$ of As (tot), 1.8 mg/L of P, 4.7 mg/L of Fe, and 19 mg/L of Si to below 50 $\mu\text{g/L}$ of As (tot), without addition of chemical oxidant. However, both these filter experiments encountered short life time i.e., period of operation. This could be due to fine sand size (0.2–0.8 mm) used for the filter which led to headloss of about 2–20 cm in all the filters eventually leading to operational problems like clogging. The filters were also inefficient in retaining the dissolved iron concentrations within the filter leading to decreased arsenic removal efficiency. Similar observations have also been made for other household filters developed for arsenic removal. However, very few studies have been done to systematically study the effect of various parameters on arsenic removal efficiency. Thus, in a detailed laboratory study, modification of filter column designed by Leupin et al. [26] has been evaluated. The modification is aimed at improving the efficiency of the unit for efficient arsenic removal while addressing the problems of clogging, excessive head loss development, and short-term iron leaching. For the modified system, iron corrosion should precede iron oxidation by the dissolved oxygen present in water before finally forming hydrous ferric oxides. Thereby, the study was initiated by applying a methodical approach to assess aeration capacity of the system which can also be used for systems beyond arsenic removal. The modified system was investigated for various parameters such as sand grain size, flow rate, and sand depth.

2. Material and methods

All chemicals used for the experiments were of analytical grade and were used without purification. Sand used for the study with a grain size of 0.6–1 mm was washed several times before use. Iron filings were freshly produced before every experiment with a “bastard” hand file from cast iron plate. All glassware were cleaned with chromic acid and then rinsed with distilled water. Arsenic standards were prepared from the solid, primary standard arsenic trioxide (As_2O_3) for As^{3+} . Stock solution of 1000 mg/L As^{3+} was prepared by dissolving 1.32 gm of As_2O_3 in 1 L distilled water and preserved by adding 1 mg/mL of Ascorbic acid. As^{5+} stock solution of 1000 mg/L was prepared by dissolving 4.16 gm of sodium arsenate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) in 1 L distilled water and preserved by adding 1 mL HCl/100 mL of solution. Stock solutions were stored in amber bottles from which intermediate standards were prepared and further diluted to make working standards. Stock iron (Fe^{2+}) solution of 200 mg/L was prepared by using ferrous ammonium sulphate [$\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$] which was then diluted further and used for preparing intermediate and working standards.

2.1. Preparation of synthetic/simulated ground water

Synthetic groundwater containing 8.2 mM HCO_3^- , 2.5 mM Ca^{2+} , and 1.6 mM Mg^{2+} was used for all experiments. The water was

prepared by bubbling CO_2 into a vigorously stirred suspension of measured amounts of CaCO_3 and MgCO_3 , resulting in a clear solution with an excess of dissolved CO_2 and a pH of 6.0–6.5, which was measured using Orion model 420A. Spiking with silicate, phosphate, and arsenic (in this order) was carried out as follows: silicate was added from an alkaline stock solution (2 g of Si/L prepared daily from $\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$) and rapidly mixed with the acidic synthetic groundwater. The rapid dilution is intended to prevent the formation of silicate polymers, as the main silicate species in natural aquatic systems is monomeric H_4SiO_4 [27]. After silicate spiking, the pH is raised to near neutral by bubbling compressed air through the solutions to outgas CO_2 . Phosphate was added from a neutral stock solution. Finally, the pH was adjusted to 7 by bubbling air, and As^{3+} or As^{5+} was added from neutral stock solutions to initial concentrations of 500 $\mu\text{g/L}$. The typical composition of synthetic/simulated ground water used for the study is given in Table 1.

2.2. As and P analysis

Arsenic and phosphorus analyses were carried out by molybdenum blue method as developed by Dhar et al. [28]. The method allows for routine analysis of As^{3+} , As^{5+} , and phosphorus by spectroscopic measurement of arsenic–phosphate–molybdenum complexes with detection limits of 3 and 60 ppb for arsenic and phosphorus, respectively. Random samples were selected and analyzed for total arsenic by using Wagtech and Merck field kits to cross validate the values obtained by molybdenum blue method.

2.3. Iron measurement

Dissolved iron (Fe^{2+}) and total iron concentrations were estimated by phenanthroline method as prescribed by standard methods for examination of water and waste water, APHA. The absorbance values were measured at 510 nm in spectrophotometer.

2.4. XRD analysis of solids

Powder X-ray diffractograms were measured with PANalytical X'Pert MPD (Cu anode 40 KV and 30 mA) from 10° to 100° 2θ . The samples obtained for the analysis were collected at the end of the experimental run by separating them from the dried mixture of sand and iron filings. Magnetic separation was used to separate the precipitates from the mixture. The purpose of the analysis was to identify the compounds formed during the experiment in each of the filter.

2.5. Analysis of BGS and field data

British Geological Survey (BGS) is established as National Geological Survey Centre for Earth Science Information and Expertise. A detailed survey of tubewells in Bangladesh region was done by BGS for constituents like arsenic, phosphorus, iron, silica, sulfate, calcium, magnesium, cobalt, copper, chromium, manganese, sodium,

Table 1
Typical composition of synthetic/simulated ground water.

Constituents	Concentrations/values
pH _{initial}	7 ± 0.1
HCO_3^- (mM)	8
Ca (mM)	2.5
Mg (mM)	1.6
Si (mg/L)	20
P (mg/L)	2
$\text{As}^{3+}/\text{As}^{5+}$ ($\mu\text{g/L}$)	500

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