Research article

Vinegar-amended anaerobic biosand filter for the removal of arsenic and nitrate from groundwater

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ABSTRACT

The performance of a vinegar-amended anaerobic biosand filter was evaluated for future application as point-of-use water treatment in rural areas for the removal of arsenic and nitrate from groundwater containing common ions. Due to the importance of sulfate and iron in arsenic removal and their variable concentrations in groundwater, influent sulfate and iron concentrations were varied. Complete removal of influent nitrate (50 mg/L) and over 50% removal of influent arsenic (200 μg/L) occurred. Of all conditions tested, the lowest median effluent arsenic concentration was 88 μg/L. Iron removal occurred completely when 4 mg/L was added, and sulfate concentrations were lowered to a median concentration <2 mg/L from influent concentrations of 22 and 50 mg/L. Despite iron and sulfate removal and the establishment of reducing conditions, arsenic concentrations remained above the World Health Organization's arsenic drinking water standard. Further research is necessary to determine if anaerobic biosand filters can be improved to meet the arsenic drinking water standard and to evaluate practical implementation challenges.

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1. Introduction

Arsenic is a naturally-occurring groundwater contaminant around the world, including in the United States, Mexico, Peru, Chile, India, Bangladesh, Nepal, and Vietnam (Ravenscroft et al., 2009; Sharma et al., 2014). Inorganic arsenic can be reduced arsenite (As(III)) or oxidized arsenate (As(V)). Arsenite is most common in groundwater in South Asia (Kinniburgh and Smedley, 2001; Ravenscroft et al., 2009). Above the World Health Organization's (WHO) arsenic drinking water standard of 10 μg/L (World Health Organization, 2008), exposure to arsenic through drinking water can cause increased risk for multiple health problems including skin lesions and cancer (Berg et al., 2001; McClintock et al., 2012). Commonly used arsenic removal technologies rely on arsenic adsorption to ferric oxyhydroxide solids and subsequent filtration (Bissen and Frimmel, 2003; Mohan and Pittman, 2007). While these systems effectively remove arsenic, other co-contaminants, including nitrate, can be found in arsenic-contaminated groundwater (Berg et al., 2001; Cole et al., 2004; Fytianos and Christophoridis, 2004). In many arsenic-affected areas, including West Bengal, India and Bangladesh, arsenic contamination has been extensively characterized, but co-occurring contaminants are less well studied. In arsenic-contaminated groundwater in India, nitrate concentrations ~20 mg/L have been measured (Kundu et al., 2008; Rahman et al., 2011). In these areas, technologies capable of removing multiple contaminants simultaneously are needed. Furthermore, arsenic-bearing wastes disposal often involves anaerobic environments, such as landfills and ponds (Badruzzaman, 2003; Clancy et al., 2013; Ghosh et al., 2006). However, wastes produced by aerobic treatment processes can be unstable in anaerobic environments, as demonstrated by the release of arsenic adsorbed to ferric oxyhydroxides under reducing landfill conditions (Ghosh et al., 2006). Anaerobic disposal environments instead favor the stability of reduced arsenic and iron solids (Burton et al., 2011; Jong and Parry, 2005; O'Day et al., 2004). Therefore, for anaerobic disposal environments, anaerobic arsenic removal may produce wastes that are less susceptible to arsenic

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release. Other benefits of anaerobic biological contaminant removal when compared to reverse osmosis and ion exchange include lower energy costs, fewer chemical inputs (no membrane cleaning or resin regeneration), and lower production of concentrated waste streams.

Upadhyaya et al. (Upadhyaya et al., 2010) previously reported the performance of an acetate-fed anaerobic continuous-flow biologically active carbon (CF-BAC) filter for arsenic and nitrate removal from groundwater. Anaerobic biological arsenic removal depends on microbial reduction of arsenate, sulfate, and ferric iron, the precipitation of arsenic sulfide solids, and the adsorption to and/or co-precipitation of arsenic with iron sulfides solids (Gallegos et al., 2007; Kirk et al., 2010; Luo et al., 2008; O'Day et al., 2004; Upadhyaya et al., 2010). These removal mechanisms are highly dependent on pH and relative arsenic, sulfide, and iron concentrations (Burton et al., 2011; Gallegos et al., 2007; Kirk et al., 2010; O'Day et al., 2004). In the CF-BAC system, arsenic was reduced from 200 μg/L to <20 μg/L, and nitrate was reduced from 50 mg/L to <0.2 mg/L (Upadhyaya et al., 2012, 2010). This system, however, would not be applicable for decentralized water treatment. Rather, a "household-scale" biosand filter may be more readily accepted and adapted for such use, given its lower cost, simple construction from local materials, and ease of operation and maintenance (Sobsey et al., 2008).

Biosand filters are point-of-use treatment systems that work by slow, intermittent water filtration through a sand bed, resulting in microbial growth (Elliott et al., 2006; Stauber et al., 2006). Once sufficient microbial biomass accumulation occurs, microbial contaminant removal takes place through mechanisms similar to traditional slow sand filters with a "schmutzdecke" (Bauer et al., 2011; Haig et al., 2011). In contrast to typical biosand filters, sand filtration for arsenic has primarily focused on the removal of arsenic via sorption to iron either present in the groundwater (Berg et al., 2006; Leupin and Hug, 2005; Leupin et al., 2005; Nitzsche et al., 2015) or iron amendments (Hussam and Munir, 2007; Neumann et al., 2013). In these arsenic-removing sand filters, microbial growth was typically avoided and filters were often drained in between uses to maintain aerobic conditions. Only one other study reported the use of a biosand filter for arsenic removal, whereby standing water was maintained to support microbial growth and rusted nails were added as an iron amendment, but found arsenic removal to be limited due to competition with phosphate and low iron concentrations in the groundwater (Chiew et al., 2009).

To leverage the benefits of anaerobic treatment for simultaneous removal of nitrate and arsenic (Upadhyaya et al., 2010), we developed and tested a vinegar-amended anaerobic biosand filter to treat groundwater containing arsenic, nitrate, sulfate, and iron. Influent sulfate and iron concentrations were varied to reflect natural differences in contaminated groundwater and were hypothesized to be the variables controlling arsenic removal. Water quality was monitored over time and spatially within the filter to investigate potential mechanisms for contaminant removal.

2. Materials and methods

2.1. Synthetic groundwater

Synthetic groundwater contained sodium, calcium, chloride, magnesium, potassium, carbonate, and bicarbonate, as previously described (Upadhyaya et al., 2010), along with 200 μg/L arsenate, 50 mg/L nitrate, 22 or 50 mg/L sulfate, 0 or 4 mg/L ferrous iron, 50 mg C/L vinegar (~5% acetic acid), and 100 μg P/L phosphoric acid. Synthetic groundwater was prepared in batches with all constituents except vinegar and iron, stored in a tank with a floating cover, and purged daily for 20 min with nitrogen gas to remove dissolved

oxygen (DO). The pH of the groundwater was 8.7 ± 0.4 (average ± standard deviation) before vinegar addition and was measured using a Mettler Toledo pH meter (Columbus, OH). Vinegar was added as an electron donor in excess of the stoichiometric requirement for reduction of nitrate and sulfate present in the influent. When iron was added, ferrous iron was mixed with oxygen-free vinegar in an anaerobic glove box (Coy, Grass Lake, MI). This mixture (or vinegar only) was added to the nitrogen-purged groundwater just before addition to the filter.

2.2. Biosand filter construction

The biosand filter was constructed in a 23 L plastic bucket with a non-airtight lid. Four sampling ports were installed along the depth of the filter (Fig. 1). The bucket contained 5 cm (3 kg) of gravel (grain size 6–12.5 mm) topped with 12.5 cm (10 kg) of sand (grain size ≤ 4.38 mm). The potential for the sand and gravel to act as a source of sulfate through the dissolution of minerals was determined through a short-term leaching experiment under anaerobic conditions, in the presence of deionized water and filter leachate (Supporting Information (SI)). Water drained through an outlet pipe located in the gravel bed slightly below port 4, and the volume was controlled by a stand pipe. The filter was inoculated with biomass collected from the backwash of the previously described anaerobic arsenic- and nitrate-removing CF-BAC filter (Upadhyaya et al., 2010).

2.3. Filter operation and maintenance

An extended start-up period (days 0–153) was required to adjust operational parameters (e.g., frequency and volume of water drained, electron donor concentration required due to the presence of electron acceptors in the sand and gravel, as described in the SI). After this period, the filter was tested with three different groundwater compositions with varying sulfate and iron concentrations. Groundwater compositions were 22 mg/L sulfate and 0 mg/L iron ("S22," days 154–232), 50 mg/L sulfate and 0 mg/L iron ("S50," days 233–381), and 50 mg/L sulfate and 4 mg/L iron ("S50Fe4," days 382–420).

The filter was operated at room temperature (23.2 ± 1.2 °C) for 420 days. Every day, 3 L of treated water were drained from the filter and replaced with 3 L of synthetic groundwater, poured through a diffuser plate to prevent disturbance of the sand and biofilm. The total volume of water in the filter was 13.3 L, resulting in an average residence time of 4.4 days. Filter cleaning occurred approximately every two weeks to remove excess biomass. Cleaning was performed after draining 3 L of treated water, using a brush to remove the biofilm along the inside wall. The top 2.5 cm of sand was agitated, and the remaining water above the sand bed was scooped out. Finally, the filter was refilled with groundwater. The solids removed during cleaning were the only waste regularly produced by this system.

2.4. Sample collection

Liquid samples were collected from ports 1 and 4 approximately every other day. All samples were collected before draining the treated water. Sample pH was measured immediately. Samples were filtered through a 0.2 μm filter (Fisher, Pittsburg, PA) and stored at 4 °C until analysis for arsenic, iron, nitrate, sulfate, and acetate as described below. Before storage, samples for arsenic and iron analyses were preserved with hydrochloric acid at a final concentration of 5 mM. The DO concentration was measured occasionally during filter start-up using a WTW Multi 340 DO meter (Weilheim, Germany) by submerging the tip of the probe in the