Nitrate and Arsenic Removal From Drinking Water With a Fixed-Bed Bioreactor [Project #4293]

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OBJECTIVES

The research team developed a fixed-bed biologically active carbon (BAC) system to simultaneously remove arsenic and nitrate from drinking water sources. Removal mechanisms were elucidated through the use of microbial community analyses and solids characterization techniques. A variety of operating conditions were evaluated and optimized at the bench-scale level, keeping in mind the future goals of demonstrating this technology at pilot-scale and its eventual integration in full-scale drinking water treatment schemes.

BACKGROUND

Contamination of natural water sources with multiple contaminants, including arsenic and nitrate, has been a major concern in the context of providing safe drinking water throughout the world. There is a need for new technologies capable of removing these contaminants simultaneously. In past studies, fixed-bed BAC bioreactor systems have been demonstrated to successfully remove the oxy-anionic contaminants nitrate and perchlorate at the lab-, pilot-, and full-scale levels. Based on preliminary batch reactor studies, the research team hypothesized that a fixed-bed BAC reactor system could be developed to simultaneously remove arsenic and oxy-anionic contaminants such as nitrate, in the presence of sulfate and iron naturally occurring in groundwater.

APPROACH

A system consisting of two fixed-bed BAC bioreactors in series was designed and seeded with BAC obtained from lab- and pilot-scale nitrate and perchlorate removal systems. These systems were originally seeded with a mixed community of microbes indigenous to natural groundwater. Using a simulated groundwater containing nitrate, arsenic, sulfate, and iron, and feeding acetic acid as the sole electron donor, we operated this system for over 1,000 days. During operation, the empty bed contact time (EBCT), backwashing strategy, and influent iron and sulfate...
concentrations were varied. System performance was monitored by measuring concentrations of total arsenic, iron, acetate, chloride, sulfate, and nitrate. Additional studies of microbial community composition and activity were performed using molecular biology methods. Solids deposited in the reactors were characterized using X-ray absorption spectroscopy (XAS) and X-ray diffraction (XRD).

RESULTS/CONCLUSIONS

Simultaneous removal of nitrate and arsenic was achieved in the fixed-bed BAC bioreactor system. Complete denitrification occurred, removing 50 mg/L (as NO$_3^-$) to below the detection limit of 0.2 mg/L. Initially, arsenic was reduced from 300 µg/L to below 20 µg/L, although performance varied during subsequent experiments and effluent concentrations ranged from 10 µg/L (the U.S. EPA and World Health Organization standard) to 50 µg/L.

Bacteria present in the reactor system were identified by sequencing their 16S ribosomal RNA genes to be closely related to known nitrate-, sulfate-, and iron-reducing bacteria. Analysis targeting specific functional genes coding for enzymes in sulfate- and arsenate-reduction pathways ([bi]sulfite reductase and arsenate reductase) indicated the presence of sulfate- and arsenate-reducing bacteria throughout the depth of the BAC bioreactors. Additionally, specific functional activities measured through messenger RNA abundance profiles were correlated with terminal electron accepting process (TEAP) zones as indicated by concentration profiles of sulfate and arsenic along the depth of the BAC bioreactors.

Iron sulfides, mackinawite and greigite, were found to be the dominant crystal phases in the system, suggesting that arsenic removal through adsorption to these solids was an important removal mechanism. It was further determined that sulfur was the nearest neighboring atom to arsenic, indicating that arsenic sulfides were present in the system, and were partially responsible for arsenic removal.

By varying the EBCT between 40 and 27 minutes, it was determined that nitrate and arsenic removals were achievable at these EBCTs. However, other factors, such as iron and sulfate concentrations may determine the EBCT needed for optimal arsenic removal. Experiments varying iron and sulfate concentrations found complete nitrate removal under all conditions. Arsenic removal varied depending on the amount of iron and location of its addition (1st reactor or 2nd reactor). Influent sulfate concentrations tested included 22 and 50 mg/L, demonstrating the flexibility of this system for treating source waters with variable sulfate concentrations.

The first BAC bioreactor was backwashed every 48 hours, whereas the second BAC bioreactor was only backwashed every 3-4 months. Initially, nitrogen gas was used during backwashing. Considering the practical advantages of air-assisted backwashing, including simpler operation, improved safety, and lower cost, the impact of air-assisted backwashing on system performance was evaluated. It was determined that overall nitrate and arsenic removals with long term use of either gas were comparable, although a slight impact on arsenic removal in the first BAC bioreactor was observed. It was determined that the two-column system helped keep arsenic concentrations in the final effluent low, even immediately after an air-assisted backwashing event.

This study demonstrated that a fixed-bed BAC bioreactor system can simultaneously remove arsenic and nitrate from a simulated groundwater utilizing an inoculum originating from a mixed community of microbes indigenous to groundwater. Microbial and chemical analyses indicated that the microorganisms utilized dissolved oxygen, nitrate, sulfate, and arsenate as the electron acceptors in a sequential manner in the presence of acetic acid as the electron donor.
Biologically produced sulfides effectively removed arsenic from the water through the formation of arsenic sulfides, and/or surface precipitation and adsorption on iron sulfides. The optimization of EBCT, iron and sulfate concentrations, and backwashing strategy resulted in a set of operational characteristics that can be used during pilot-scale evaluation of this technology.

APPLICATIONS/RECOMMENDATIONS

An anaerobic fixed-bed bioreactor system was developed for the simultaneous removal of arsenic, nitrate, sulfate, and iron from contaminated drinking water sources. This work highlights the potential of biological drinking water treatment processes to simultaneously remove multiple contaminants.

It was shown that a natural consortium of microorganisms can be used to seed a bioreactor and remove multiple contaminants. The inoculum for this system was taken from lab- and pilot-scale perchlorate and nitrate removing bioreactors, which had originally been seeded with the natural microbial communities present in ground waters. These microbial communities had not been exposed to water containing detectable levels of arsenic previously, but were able to reduce arsenate to arsenite, showing that an inoculum from a water containing arsenic was not required.

The removal of arsenic under anaerobic conditions has advantages compared to more commonly used oxidative removal strategies for arsenic. First, anaerobically produced arsenic bearing solids are more stable under long-term disposal conditions, such as in landfills. Reductive dissolution of arsenic waste generated under aerobic conditions has been demonstrated and raises the concern that arsenic can be re-introduced to the environment. In addition, several contaminants have the potential to be removed under anaerobic conditions in the same system, including nitrate, perchlorate, uranium, and chlorinated organics.

Fixed-bed bioreactor systems need to be backwashed regularly to remove excess biomass. Initially, the system was backwashed using water and nitrogen gas to maintain reducing conditions during backwashing. We subsequently demonstrated that air-assisted backwashing can be a feasible option for anaerobic biological systems, which increases the flexibility and applicability of this type of drinking water treatment system. Using air for backwashing can significantly lower the operational costs compared to the use of compressed nitrogen gas for backwashing.

This system removes arsenic through precipitation of arsenic sulfide and adsorption of arsenic on iron sulfide precipitates. Therefore, the concentrations of iron and sulfate naturally present in the contaminated water source are important parameters. Prior to implementation of this system, pilot studies should be conducted to determine optimal operating conditions for each contaminated water. In the current study, we operated the laboratory system with total influent iron concentrations between 2 and 10 mg/L and influent sulfate concentrations of 22 and 50 mg/L. The optimal combination of influent iron and sulfate concentrations will vary depending on the concentration of arsenic and other compounds present in each contaminated water.

Future work will evaluate the requirements for post-treatment needed to remove excess carbon, aerate the finished water, and provide disinfection. Additionally, characterization of arsenic containing solid waste generated in the system must be performed to determine the long-term stability under disposal conditions.
RESEARCH PARTNERS

The current research was conducted at the University of Michigan with Carollo Engineers, Inc. as a collaborative partner. The U.S. National Science Foundation provided support for a related project that focused on (1) the characterization of the arsenic containing solids produced by the technology described in the current study, (2) the evaluation of household and community-based drinking water treatment systems employing the described removal mechanisms, and (3) the microbial characterization of system effluent with the goal of developing appropriate post-treatment strategies (project CBET 0967707).

PARTICIPANTS

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