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Simultaneous removal of arsenic and nitrate in absence of iron in an attached growth bioreactor to meet drinking water standards: Importance of sulphate and empty bed contact time

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ABSTRACT

There are several reports on simultaneous occurrence of arsenic and nitrate in drinking water sources especially in groundwater at wide range of concentrations. However, there is no report available, so far, on simultaneous successful removal of arsenic and nitrate from contaminated groundwater in absence of iron, and effects of one contaminant on the overall performance of a biological reactor. The present study investigates the roles of sulphate and empty bed contact time (EBCT) on simultaneous removal of nitrate and arsenic to meet the drinking water standards in an attached growth bioreactor in absence of iron.

An attached growth reactor (AGR) was fabricated using Perspex cylinder, inoculated with mixed bacterial culture and operated in downflow mode in absence of oxygen at 30 °C for more than 400 days under varying influent arsenate (200–750 μg/L) and nitrate concentrations (50–200 mg/L), and EBCT of 45–60 min. Acetate was used as external carbon source and electron donor in this study. Complete nitrate removal was observed at all tested concentrations. Arsenic removal was high (up to 99.8%) and was well below drinking water standards from initial concentrations of up to 750 μg/L. The arsenic removal efficiency was found to depend on sulphate reduction and EBCT of the reactor. Results of X-ray diffraction (XRD) and X-ray absorption spectroscopy (XAS) analyses suggested that arsenic precipitation in the form of arsenosulphides (orpiment and realgar) was the removal mechanism.

1. Introduction

Arsenic (As) is one of the hazardous elements often occurs in the form of oxyanions either as arsenite [As(III)] or arsenate [As(V)] along with nitrate in drinking water sources at wide ranges of concentrations (Guha et al., 2005; Mondal et al., 2013; Rezaie-Boroon et al., 2014). Drinking water is the main route through which it enters in to the human body (Chen et al., 2009). Acute and chronic exposures to arsenic led to human diseases related to respiratory system, gastro-intestinal tract, cardiovascular problems and skin cancer etc. (Singh et al., 2015). Ingestion of high nitrate drinking water is associated with “blue-baby syndrome” (methaemoglobinaemia) in infants, and the potential formation of carcinogenic nitrosamines in humans (Majumdar and Gupta, 2000). Due to severe health impacts on humans, most of the regulatory agencies have imposed maximum permissible limits of 10 μg/L and 45 mg/L for arsenic and nitrate, respectively in drinking water (BIS:10500, 2012; WHO, 2011). Australia has instituted a standard of 7 μg/L whereas, the State of New Jersey in the USA adopted more stringent permissible limits of 5 μg/L for arsenic in year 2006 (Barringer and Reilly, 2013).

There are several technologies available for nitrate removal from drinking water. The most common practices are ion-exchange, reverse osmosis or biological de-nitrification in which nitrate is converted to innocuous nitrogen gas (Aslan and Cakici, 2007; Mateju et al., 1992; Pintar and Batista, 2006; Upadhyaya et al., 2010). Arsenic on the other hand is removed through phase transfer by the processes of adsorption, ion-exchange, precipitation and/or co-precipitation (Altun et al., 2014; Mondal et al., 2013). Physico-chemical processes practiced for arsenic and nitrate removal are often associated with high operating costs, regeneration of used matrix, disposal of exhausted resins and generation of large volumes of hazardous waste which, may further become sources of water pollution (Clancy et al., 2015; Stuckman et al., 2011). Contrary to that, biological systems often offers an eco-friendly and sustainable approach leaving no residual or less

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waste and have high potential even on drinking water treatment (Brown, 2008). Although, application of sulfidogenic bioprocess on arsenic removal have been successful to achieve a high percentage removal, most of the available reports are based on arsenic removal from laboratory made heavily contaminated wastewater added with high amount of arsenic and iron. Some reports are on arsenic removal by using mixed bacterial culture from lab made mining and metallurgical industrial effluents added with arsenic of 20–100 mg/L along with iron of 100–200 mg/L (Altun et al., 2014; Battaglia-Brunet et al., 2012) whereas some are on use of pure bacterial culture on simultaneous removal of arsenic and nitrate from synthetic medium in presence of iron (Li et al., 2015, 2016). Li et al. (2015) utilized anaerobic Fe(II) oxidation denitrifiers (AFODN) bacteria and achieved 44% and 28% nitrate and arsenic removal from an initial concentration of 8 mM and 13.65 μM respectively, in presence of iron. Although, Rodriguez-Freire et al. (2014) reported complete arsenic removal after 9 days in presence of 2 mg/L of $FeCl_3$, the paper is completely silent about iron removal efficiency. Addition of iron during arsenic removal by most of the researchers could be due to co-occurrence of the both most often. However, occurrence of a few to several hundreds of μg/L of arsenic alone (without iron) at several parts of India have been reported (CGWB, 2010; Venkataraman and Uddameri, 2012). The major disadvantages of using such systems are use of unrealistic, (with reference to treatment for drinking water purposes) high arsenic concentration and their dependency on presence of iron in contaminated ground water. Additionally, use of pure culture often imposes several limitations in practice to deal with large amount of contaminated water (Kleerebezem and van Loosdrecht, 2007). Upadhyaya et al. (2010) also added 10 mg/L of iron and 22.4 mg/L of sulphate in a sulfidogenic bioreactor consisting of 2 identical columns in a series operated at the maximum of 40 min (20 + 20 min) EBCT to remove 200 μg/L of arsenic but failed to meet drinking water standards for arsenic and iron. This could be due to insufficient amount of sulphate addition and/or EBCT (20 min) maintained in the first column, which resulted in only 1.5 mg/L of sulphate removal. Although, biological arsenic removal in absence of iron as arsenosulphides has been reported (in the form of orpiment and realgar), not more than 80% removal could be achieved from an initial of 100 mg/L in presence of 1800 mg/L of sulphate and 32 h HRT (Battaglia-Brunet et al., 2012). Altun et al. (2014) reported only 8–9% of arsenic removal from an initial arsenic concentration of 0.5–20 mg/L in absence of iron but presence of 2000 mg/L of sulphate at an HRT of 9.6 h. Thus, none of the above two reactors could meet the drinking water standards might be due to presence of very high initial arsenic concentration (100 mg/L) and/or presence of high sulphate concentration (1800–2000 mg/L). Sulphate bioreduction resulted in formation of high bicarbonate alkalinity that can prevent formation of precipitation of arsenosulphide in the form of realgar and/or orpiment (Henke, 2009).

The main objective of this study is simultaneous removal of arsenic and nitrate in absence of iron in a sulphate induced attached growth bioreactor to meet the drinking water standards. Effect of one contaminant on removal of the other at varying EBCT were also assessed.

2. Materials and methods

2.1. Bioreactor set-up

A transparent perspex column of 5 cm internal diameter and 32 cm height was used to fabricate a laboratory scale attached growth reactor (AGR), which was partially filled (17 cm, 333 cm^3) with approximately 203 g of granular waste activated carbon (WAC) of grain size between 1.5 and 2.0 mm as a supporting material for the growth of microbes. Schematic diagram with the details of the reactor AGR is shown in Fig. A.1 and the reactor set up is shown in Fig. 1. The WAC granules were collected from a number of house hold water purifiers in the residential campus of IIT Guwahati, after their useful life. Purposes behind selection of WAC were to reuse a waste material for water purification. The WAC granules were washed by rinsing twice with deionized water and overnight oven drying at 70 °C before its use in AGR. A set of batch experiments on removal of arsenic [As(V)] from simulated groundwater spiked with 500 μg/L of arsenic, through adsorption by WAC granules were conducted before being used in the AGR. Less than 10% arsenic removal was observed after 12 h of agitation at 150 rpm and 30 °C. The active bed volume (333 cm^3) was considered for the calculation of empty bed contact time (EBCT). In addition to feed inlet and treated water outlet, five sampling ports, named P1, P2, P3, P4, and P5 were provided along the depth of the reactor to collect profile samples. The bioreactor was equipped with a feed distribution system connected to peristaltic pump (PP 10EX, Miclins, India) to regulate the feed flow rate in the reactor system. Two nos. of circular screens (about 70% perforation, 1 mm diameter holes) of 5 cm diameter (equal to the internal diameter of the main reactor) were placed, one between P1 and inlet, and the other between P5 and the final outlet of the AGR. The purpose of the top screen was to prevent WAC from being washed out during backwashing as well as to help distribute feed more uniformly throughout the cross section of the reactor whereas, the bottom screen was to support the WAC granules from being washed out along with the treated water (Fig. 1).

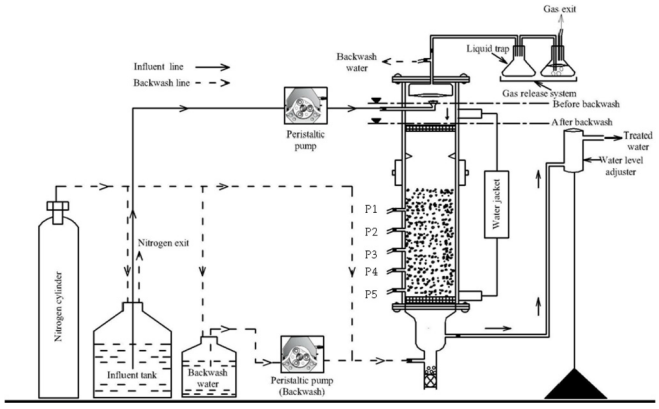

Fig. 1. Lab scale schematic diagram reactor set-up of AGR.

2.2. Bioreactor operation and sampling

The bioreactor was inoculated with diluted seed culture obtained by mixing sludge collected from IIT Guwahati sewage treatment plant (0.75 L, MLSS: 3780 mg/L and MLVSS: 2560 mg/L) with (about 5% of the total MLSS) a bench scale perchlorate and nitrate reducing anoxic bioreactor (Ghosh et al., 2011), and a bench scale sulphate removal anoxic bioreactor (Brahmacharimayum and Ghosh, 2014).

After inoculation with mixed bacterial culture, the AGR was operated for more than 6 months as start-up period mainly to optimise the EBCT and backwash frequency, before being tested for its performance at varying arsenic and nitrate concentrations at 45 min EBCT and 96 h backwash frequency. This paper presents the effects of initial arsenic and nitrate concentration on overall performance of the AGR. Throughout the study, arsenic and nitrate contaminated simulated water was fed to the bioreactor with the composition as given in Table 1. Composition of simulated groundwater was similar to the one prepared by Upadhyaya et al.