



Biological groundwater treatment for chromium removal at low hexavalent chromium concentrations



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HIGHLIGHTS

- Anaerobic systems provide for higher Cr(VI) removal rates compared to aerobic systems.
- Complete microbial Cr(VI) reduction is achieved under anaerobic conditions.
- Easily degradable substrates support higher Cr(VI) removal rates than complex ones.
- Cr(VI) removal rates follow the Arrhenius equation for temperatures between 12 °C–33 °C.
- Sand filtration as a polishing step provides for complete Cr removal from groundwater.

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ABSTRACT

The objective of this work is to develop and evaluate biological groundwater treatment systems that will achieve hexavalent chromium reduction and total chromium removal from groundwater at hexavalent chromium (Cr(VI)) groundwater concentrations in the 0–200 µg/L range. Three lab-scale units operated, as sequencing batch reactors (SBR) under aerobic, anaerobic and anaerobic–aerobic conditions. All systems received groundwater with a Cr(VI) content of 200 µg/L. In order to support biological growth, groundwater was supplemented with milk, liquid cheese whey or a mixture of sugar and milk to achieve a COD concentration of 200 mg/L. The results demonstrate that a fully anaerobic system or an anaerobic–aerobic system dosed with simple or complex external organic carbon sources can lead to practically complete Cr(VI) reduction to Cr(III). The temperature dependency of maximum Cr(VI) removal rates can be described by the Arrhenius relationship. Total chromium removal in the biological treatment systems was not complete because a significant portion of Cr(III) remained in solution. An integrated system comprising of an anaerobic SBR followed by a sand filter achieved more than 95% total chromium removal thus resulting in average effluent total and dissolved chromium concentrations of 7 µg/L and 3 µg/L, respectively.

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1. Introduction

The release of chromium (Cr) wastes has in many cases resulted to serious contamination of water bodies. In the environment, Cr is usually encountered in the oxidation states of trivalent Cr (Cr(III)) and hexavalent Cr (Cr(VI)). Each of the above oxidation states has very different biological and chemical properties. Cr(III) is more stable, forms relatively insoluble substances, has relatively low

toxicity and it is not able to cross cell membrane (Gonzalez et al., 2003). On the contrary Cr(VI) is very soluble and thus highly mobile in water and presents high toxicity, been acutely toxic, teratogenic and carcinogenic (Chen and Gu, 2005a). Cr(VI) frequently appears in many wastes from industrial activities such as steel-works, petroleum refining, metal finishing, Cr electroplating, leather tanning, etc (Chen and Gu, 2005a).

Until recently, high levels of Cr(VI) in the environment were always attributed to anthropogenic pollution. However over the last fifteen years there are reports in the literature (Gonzalez et al., 2005; Manning et al., 2015) demonstrating that relatively high levels of Cr(VI) may be due to natural geogenic processes. These

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conditions are met in quite a few populated areas in the Pacific (California (USA), Mexico), in the Mediterranean (Greece, Italy) and other parts of the world. According to these findings, Cr content in groundwater can be as high as 300 ppb in ultramafic rocks and serpentinites of ophiolite complexes.

In order to combat Cr groundwater pollution, USEPA has designated Cr as one of seventeen chemicals causing the greatest threat to human health. Similarly, Canada has included Cr and its compounds in its list of priority pollutants. WHO, 2004 proposes a maximum allowable limit for total Cr in drinking water of 50 µg/L.

Several treatment technologies have been developed to remove chromium from water. The most often used methods are physico-chemical techniques and most specifically: a) chemical oxidation (Barrera-Diaz et al., 2012), b) ion exchange (Ren et al., 2012), c) adsorption via activated carbon (Singh and Singh, 2012; Zhang et al., 2015) and d) membrane separation (Ghosh and Bhattacharya, 2006; Yao et al., 2015). All these methods present several disadvantages such as high capital and operational cost, production of chemical sludge, sludge disposal problems, etc. Despite the extensive literature regarding the physicochemical methods for Cr(VI) removal from water, there is no literature on biological treatment of groundwater for Cr(VI) removal, especially at relatively low Cr(VI) concentrations in the 0–200 µg/L range. However the microbial reduction of Cr(VI) to Cr(III) has been extensively reported in the literature for the treatment of liquid wastes under both aerobic and anaerobic conditions (Shen and Wang, 1993; Stasinakis et al., 2004; Ge et al., 2013). These biological methods offer an efficient, cost effective and environmentally friendly option for Cr removal. The main reasons that biological methods have not been implemented for the treatment of groundwater are: a) the absence of electron donor (organic substrate) in groundwater to provide for the biochemical reduction of Cr(VI) to Cr(III), b) the lack of experience on the operation of biological treatment systems for groundwater purification, c) the relatively high operational cost due to aeration requirements in the case of aerobic treatment systems and d) the questionable effectiveness of anaerobic treatment systems at low water temperatures.

The objective of this work was to develop and evaluate biological groundwater treatment systems that will achieve appreciable Cr(VI) reduction and total chromium removal. Lab-scale units operated, as sequencing batch reactors (SBRs) in order to evaluate the effect of: 1) aerobic or anaerobic conditions, 2) type of electron donor and 3) temperature on the mechanism of biological Cr(VI) removal from groundwater. In addition, the effectiveness of a sand filtration unit as a polishing step for the treatment of biologically treated groundwater was also evaluated.

2. Experimental materials and methods

2.1. Continuous flow and batch experiments

A series of bench scale experimental systems operating as SBRs were employed in order to evaluate the effect of several parameters on biological Cr(VI) removal. The experiments are divided in two phases. During the 1st phase, one system operated as an anaerobic system (ANAER), one as an anaerobic–aerobic system (ANAER-AER) and one as aerobic system (AER). All systems received groundwater from the National Technical University of Athens campus water supply network, supplemented with Cr(VI) to reach a concentration of 200 µg/L. The quality characteristics of groundwater used are summarized in Table S1 in the Supplementary Material. In order to support biological growth, groundwater was supplemented with milk at a chemical oxygen demand (COD) concentration in the water of 200 mg/L, and nutrients. Cr(VI) concentration in influent water was achieved through daily Cr(VI) addition in the form of

potassium dichromate solution ($K_2Cr_2O_7$, Merck), whereas nitrogen and phosphorus were dosed in the form of NH_4Cl and K_2HPO_4 solutions to reach a concentration of nitrogen and phosphorus of 12 and 2 mg/L, respectively. Average temperature of the three bench scale units was equal to 17.5 °C. Sludge age was equal to 3 days for the ANAER-AER system and 10 days for the AER and ANAER systems. All experimental systems were inoculated with a mixture of mesophilic anaerobic digested sludge and aerobic activated sludge from Psytalia Wastewater Treatment Plant (PWTP) at a ratio of 50%/50% on a mass basis. The nominal hydraulic residence time of the three systems was equal to 1.7 d. All SBRs operated with a 24 h cycle consisting of 0.5 h feeding time, 22 h reaction time, 1 h settling time and 0.5 h decanting time. In ANAER-AER system reaction time consisted of 14 h non aerated period and 8 h aerated period. During the 2nd phase of the experiments all systems operated under anaerobic conditions and were immersed in water baths for temperature control. Mixed liquor temperature ranged from 12 °C to 33 °C, while sludge age was equal to 10 days for all the experimental systems. During the second phase of the experiments all systems received groundwater with a Cr(VI) concentration of 200 µg/L, supplemented with milk or a mixture of sugar and milk (80% sugar, 20% milk on a COD basis) or cheese whey with a total COD concentration of 200 mg/L and nutrients. The operational characteristics of all SBRs are shown in Table S2 in the Supplementary Material.

In order to measure maximum total Cr and Cr(VI) removal rates, batch experiments were conducted in triplicates at constant temperature, by submerging batch reactors in temperature controlled water baths. These batch assays were conducted with biomass from the SBR systems that was acclimatized to the substrate used, the Cr(VI) initial concentration and the temperature employed in the batch experiments. Following batch feeding, hourly samples from the batch systems were collected and analyzed for soluble COD, dissolved oxygen (DO), redox (ORP), total and hexavalent chromium for a period of 24 h. Abiotic batch experiments conducted in triplicates with biomass that was inactivated by autoclaving (121 °C, 30 min contact time) were used as a control to evaluate the portion of Cr(VI) removal that could be attributed to chemical reduction or any other abiotic process.

In addition to biological treatment, the effectiveness of a sand filtration unit as a polishing step for chromium removal was also evaluated. More specifically following biological treatment, treated groundwater was transferred to a sand filter through a peristaltic pump at a hydraulic loading of 6 m³/m²/h. Before its use, sand was washed with distilled water and dried at 105 °C. Sand filtration was taking place in a 5 cm plexiglas column filled with silica sand with effective size of 0.3–1.0 mm.

2.2. Analytical methods

2.2.1. Chemicals

The performance of the bench scale units was assessed by routine daily measurements of total and soluble COD, total suspended solids (TSS), volatile suspended solids (VSS), NH_4-N , NO_3-N , DO, total Cr and Cr(VI) chromium throughout the experimental period. COD, NO_3-N and total chromium were determined according to Standard Methods (American Public Health Association, 2005). Mixed liquor volatile suspended solids (MLVSS) were determined by filtering the sample through a glass fiber filter (type Whatman GF/C). Soluble COD and Cr were measured on the filtrate through a 0.45 µm membrane filter (Millipore). In order to further investigate the size distribution of total Cr, fractionation was accomplished by filtration of the samples through polyethersulfone membranes having different molecular weight cutoffs. More specifically the membranes that were employed had a nominal

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