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Biological removal of selenate and ammonium by activated sludge in a sequencing batch reactor

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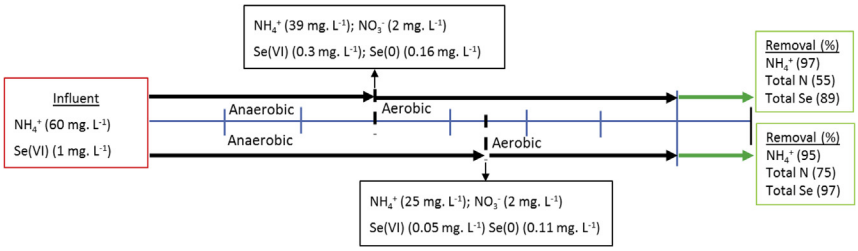
HIGHLIGHTS

- Selenate bioreduction by alternating anaerobic – aerobic conditions in a SBR.
- Ammonium was removed by simultaneous nitrification and denitrification.
- Majority of selenium was entrapped in activated sludge as Se(0).
- First report on the simultaneous removal of Se(VI) and NH_4^+-N by activated sludge.

GRAPHICAL ABSTRACT



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ABSTRACT

Wastewaters contaminated by both selenium and ammonium need to be treated prior to discharge into natural water bodies, but there are no studies on the simultaneous removal of selenium and ammonium. A sequencing batch reactor (SBR) was inoculated with activated sludge and operated for 90 days. The highest ammonium removal efficiency achieved was 98%, while the total nitrogen removal was 75%. Nearly a complete chemical oxygen demand removal efficiency was attained after 16 days of operation, whereas complete selenate removal was achieved only after 66 days. The highest total Se removal efficiency was 97%. Batch experiments showed that the total Se in the aqueous phase decreased by 21% with increasing initial ammonium concentration from 50 to 100 mg L^{-1}. This study showed that SBR can remove both selenate and ammonium via, respectively, bioreduction and partial nitrification-denitrification and thus offer possibilities for treating selenium and ammonium contaminated effluents.

1. Introduction

Many wastewaters such as mining effluents (acid mine drainage, coal mine wastewaters, uranium mine discharge and gold mine wastewaters), agricultural drainage and industrial effluents are contaminated by both selenium oxyanions and ammonium, and thus need to be treated before discharging into natural water bodies (Dale et al., 2015; Kapoor et al., 2003; Tan et al., 2016; Muscatello et al., 2008; Ríos et al., 2008; Uhrie et al., 1996; Zaitsev et al., 2008). Selenium in these wastewaters is mainly present as selenate and selenite at typical concentrations varying between 0.4 and 53 mg L^{-1} and has become a matter of public

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and scientific attention (Mal et al., 2016a; Tan et al., 2016). While the current selenium discharge criteria for aquatic life and the proposed toxicity thresholds are debatable, selenium release and its pollution has become a serious concern in recent years (Tan et al., 2016). Physical and chemical treatment approaches such as adsorption, ion exchange, chemical reduction and reverse osmosis are available for selenium removal from contaminated waters. But microbial technologies are promising for the treatment of selenium-containing wastewaters, mainly due to low cost and robustness of the bioprocess (Nancharaiah and Lens, 2015a,b).

The mining industry uses large amounts of explosives containing ammonium nitrate, which is among the main sources of ammonium in mining effluents (Papirio et al., 2014; Zaitsev et al., 2008). The ammonium concentration in mining wastewaters was reported to range from 2 to 83 mg NH_4^+ L^{-1} (Kapoor et al., 2003; Koren et al., 2000; Ríos et al., 2008; Zaitsev et al., 2008). Agricultural waste, domestic and industrial effluents can also contain large quantities of ammonium (Islam et al., 2009; Ríos et al., 2008). Ammonium and free ammonia can promote corrosion and eutrophication, reducing the oxygen content and increasing toxicity to living organisms in aquatic ecosystems (Paredes et al., 2007). Therefore, wastewaters containing both ammonium and selenium need to be treated before being released into the environment. Sequencing batch reactors (SBR) allow complete nitrogen removal by the simultaneous nitrification and denitrification (SND) process (Wang et al., 2015). The presence of high ammonium concentrations in wastewater can influence the selenium treatment process and vice-versa (Yang et al., 2009). Synchronous removal of ammonium (NH_4^+-N) and sulfate can be achieved by the co-existence of anaerobic ammonium oxidation (ANAMOX) bacteria with sulfate reducing bacteria (SRB) (Yang et al., 2009; Zhao et al., 2006). However, to the best of our knowledge, simultaneous removal of ammonium and selenium has not yet been reported.

In biological treatment of selenium-containing wastewaters, a significant fraction of the biogenic elemental selenium nanoparticles (SeNPs) remains in the bioreactor effluents due to their colloidal nature (Buchs et al., 2013). This not only compromises the discharge criteria of the treatment process, but also raises the concern on the fate of discharged biogenic SeNPs in the environment in terms of their toxicity and potential re-oxidation to soluble forms (Buchs et al., 2013; Mal et al., 2016b). Recently, Jain et al. (2016) have shown that activated sludge has better entrapment abilities for the biogenic SeNPs formed from selenite reduction. The entrapment of biogenic SeNPs in turn improved the settleability and hydrophilicity of the activated sludge. Thus, the activated sludge process could offer a promising alternative treatment, wherein effective total selenium removal can be achieved without the need for a post-treatment step for removing biogenic SeNPs envisaged in the case of an anaerobic treatment system (Park et al., 2013).

There are only a few studies on the use of activated sludge for the treatment of selenium-rich wastewaters. Moreover, the effect of alternating aerobic - anoxic or anaerobic conditions on selenate bioreduction and the fate of biogenic SeNPs in the activated sludge wastewater treatment system are unknown. The objective of this study was, therefore, to investigate the potential of simultaneous removal of NH_4^+ and Se(VI) using activated sludge. A SBR was chosen for the study as it allows maintaining alternating aerobic-anoxic conditions required for oxidation of NH_4^+ and reduction of selenate, respectively. The SBR was inoculated with activated sludge and fed with artificial mining effluent containing ammonium chloride and sodium selenate. Removal of ammonium-N and selenate was monitored during long term SBR operation. Batch experiments were conducted to investigate the effect of different concentrations of ammonium on selenate bioreduction.

2. Materials and methods

2.1. Source of biomass

Activated sludge was collected from a full-scale domestic waste water treatment plant in Harnaschpolder (The Netherlands; Jain et al., 2016) and used for inoculating the SBR. The reactor was inoculated with 400 mL of activated sludge (1.2 g mixed liquor volatile suspended solids (MLVSS) L^{-1}).

2.2. Synthetic wastewater

The synthetic ammonium-selenate wastewater was prepared in deionized water and consisted of the following basic ingredients as described in Nancharaiah et al. (2008) (in g L^{-1}): Na_2SeO_4, 1.6; NH_4Cl, 0.3; K_2HPO_4, 0.06; KH_2PO_4, 0.02; $MgSO_4{\cdot}7H_2O$, 0.06; and $CaCl_2{\cdot}2H_2O$, 0.03. Trace elements were provided by adding 0.1 mL of trace element mix per 1 L of simulated ammonium-selenate wastewater (Nancharaiah et al., 2008). The influent of the SBR was supplemented with 1.0 g L^{-1} sodium acetate as sole carbon source, corresponding to an organic loading rate (OLR) of ~1.8 g chemical oxygen demand (COD) L^{-1} d^{-1}. The pH of the simulated ammonium-selenate rich wastewater was 7.0–7.1.

2.3. Sequencing batch reactor operation

A polyacrylic column with influent, effluent and aeration ports was set-up and operated with a 1.2 L working volume in SBR mode for three months at 30 °C. The schematic of reactor configuration used for the experiments is shown in Fig. S1 (Supplementary data). It was fed with the simulated ammonium-selenate wastewater with 66% volume exchange ratio with three cycles per day.

The SBR operation was divided into two periods. During period – I, the SBR was operated for 60 days with an 8 h cycle period consisting of the following steps: 3 h static fill (anoxic/anaerobic), 4 h aeration, 10 min settle, 5 min decant and 45 min idle period. In period – II, the SBR cycle period was altered to 4.5 h static fill (anoxic/anaerobic), 2.5 h mixing by aeration, 10 min settle, 5 min decant and 45 min idle periods. During the aeration phase, mixing was achieved by bubbled aeration at the reactor bottom using a porous stone connected to an aerator. The dissolved oxygen (DO) in the SBR during the aeration period was >5 mg L^{-1}.

Sampling ports of the SBR were named S1, S2 and S3 from the bottom to the top for studying one typical SBR cycle (Fig. S1, Supplementary data). Samples from the influent and effluent of the SBR were collected daily to monitor the overall performance of the SBR. Occasionally, samples were collected during the SBR cycle period at 1.5 and 3 h during the filling period and at 4, 5, 6 and 7 h during the aerobic period to determine removal profiles.

2.4. Batch experiments – effect of ammonium on selenium removal

Batch tests were conducted to investigate the effect of ammonium on selenate (0.25 mM or 20 mg Se L^{-1}) bioreduction at different initial NH_4^+-N concentrations of 0, 50 and 100 mg L^{-1} at pH 7.0. The tests were performed in 120 mL volume glass serum bottles. The serum bottles were inoculated with 10 mL activated sludge collected from the SBR after the 66th day in 90 mL simulated ammonium-selenate wastewater as described above. The bottles were incubated at 30 °C on an orbital shaker set at 150 rpm for 36 h. All the batch experiments were performed either in duplicate or triplicate. Liquid samples were collected at regular time intervals for analyzing selenate (Se (VI)), total selenium, elemental selenium (Se(0)), ammonium (NH_4^+), nitrite (NO_2^-) and nitrate (NO_3^-). Suitable controls were kept by incubating bottles containing only