

Optimizing sulfur-driven mixotrophic denitrification process: System performance and nitrous oxide emission



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HIGHLIGHTS

- Mitigating N₂O from S⁰-driven mixotrophic denitrification was studied with a model.
- The model is successfully validated using data from two experimental systems.
- Increasing SRT and influent COD/N ratio substantially reduces N₂O emission.

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ABSTRACT

Nitrate contamination of groundwater has been recognized as a significant environmental problem world widely. Sulfur-driven mixotrophic denitrification has been demonstrated as a promising groundwater treatment process, which though plays an important role in nitrous oxide (N₂O) emissions, significantly contributing to the overall carbon footprint of the system. However, the current process optimizations only focus on nitrate removal and excess sulfate control, with the N₂O emission being ignored. In this work, an integrated mathematical model was proposed to evaluate the N₂O emission as well as the excess sulfate production and carbon source utilization in sulfur-driven mixotrophic denitrification process. In this model, autotrophic and heterotrophic denitrifiers use their corresponding electron donors (sulfur and organic matter, respectively) to reduce nitrate to nitrogen gas, with each modeled as three-step denitrification (NO₃⁻ to N₂ via NO₂⁻ and N₂O) driven by sulfur or organic matter to describe all potential N₂O accumulation steps. The developed model, employing model parameters previously reported in literature, was successfully validated using N₂O and sulfate data from two mixotrophic denitrification systems with different initial conditions. Modeling results revealed substantial N₂O accumulation due to the relatively low autotrophic N₂O reduction activity as compared to heterotrophic N₂O reduction activity, explaining the observation that higher carbon source addition resulted in lower N₂O accumulation in sulfur-driven mixotrophic denitrifying system. Based on the validated model, optimizations of the overall system performance were carried out. Application of the model to simulate long-term operations of sulfur-driven mixotrophic denitrification process indicates that longer sludge retention time reduces N₂O emission due to better retention of active biomass. High-level total nitrogen removal with significant N₂O emission mitigation, appropriate excess sulfate control and maximized COD utilization can be achieved simultaneously through controlling the influent nitrate and COD concentrations.

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1. Introduction

Due to the intensive utilization of nitrogenous fertilizers and inappropriate discharge of wastewaters and solid wastes (Chen et al., 2016; Wakida and Lerner, 2005), nitrate contamination of groundwater has become a significant environmental issue

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throughout the world (Rivett et al., 2008). It has been reported that 10–25% of groundwater used for drinking water supply in the US contains nitrate concentrations over the maximum allowable contaminant level of 10 mg-N/L (Lee and Rittmann, 2002). Such elevated nitrate concentrations in groundwater can cause serious human health problems (i.e., methaemoglobinaemia and cancer) (Peng et al., 2016a) and ecological disturbances (i.e., eutrophication of water bodies) (Roy and Bickerton, 2010).

Traditional nitrate treatment approaches include ion exchange, reverse osmosis, electrodialysis and distillation, which are expensive in terms of operation cost and also not suitable for in-situ applications (Peng et al., 2015). For this reason, biological denitrification (heterotrophic and autotrophic) is considered as an alternative process. Typically, groundwater contaminated with nitrate contains no organic matter, therefore sulfur-driven autotrophic denitrification has attracted more attentions compared to heterotrophic denitrification which requires massive external organic carbon (Chen et al., 2015a; Di Capua et al., 2015; Koenig and Liu, 2001; Liu et al., 2009; Sahinkaya et al., 2014; Sahinkaya et al., 2015; Sierra-Alvarez et al., 2007).

The main disadvantage of sulfur-driven autotrophic denitrification process is the inevitable generation of a large amount of sulfate. Theoretically, ca. 33 mg-N/L nitrate in groundwater can be removed via sulfur-driven autotrophic denitrification, without exceeding the US allowable sulfate limit in drinking water (i.e., 250 mg/L) (Oh et al., 2001). Therefore, for treating groundwater with high nitrate concentrations, a more effective strategy to control sulfate formation is to combine autotrophic and heterotrophic denitrification processes (Sahinkaya et al., 2011). A number of studies have been carried out on this promising mixotrophic denitrification process, focusing on the selection of carbon source, effects of C/N ratio, kinetics analysis, microbial community structure, reactor types and other operation parameters (Chen et al., 2014; Chen et al., 2009; Garcia-de-Lomas et al., 2007; Liu et al., 2016a; Mora et al., 2015; Qian et al., 2015; Sahinkaya and Dursun, 2012; Sahinkaya and Kilic, 2014; Xu et al., 2015a; Xu et al., 2015 b; Xu et al., 2013; Xu et al., 2014; Yu et al., 2016).

However, recent studies have revealed that nitrous oxide (N_2O) can be produced and accumulated as a significant intermediate product (i.e., maximum accumulation concentration of ca. 24% of the nitrogen load) during this mixotrophic denitrification process with both sulfur and organic matter as electron donors (Zhang et al., 2015a), which has raised increasing concerns owing to its potent greenhouse gas effect and its ability to deplete stratospheric ozone (Chen et al., 2012; Ravishankara et al., 2009). For example, it has been showed that 1% increase in N_2O emission would induce a 30% increase in the carbon footprint during the wastewater treatment due to the high global warming potential of N_2O (Law et al., 2012). Therefore, understanding and reducing N_2O production during this mixotrophic denitrification process is of great importance to optimize the application of such system.

Mathematical modeling is important toward a full understanding of biological nitrogen removal process, which has been applied to predict and mitigate N_2O emission during wastewater treatment processes (Liu et al., 2015b, 2015c; Ni et al., 2011; Peng et al., 2016b). However, little effort has been dedicated to modeling the N_2O dynamics during mixotrophic denitrification process for high-concentration nitrate removal despite of considerable N_2O production. Under such circumstances, the competition for nitrogen compound between heterotrophic and autotrophic denitrifiers could induce a different scenario on N_2O emission, which is not accounted for in previous studies. In addition, the previous process optimizations only focus on nitrate removal and sulfate control in mixotrophic denitrification, with the N_2O emission being not considered (Sahinkaya et al., 2011).

This study aims to propose a model for the prediction of N_2O production during sulfur-driven mixotrophic denitrification and to optimize the process performance taking N_2O emission into consideration. The model is validated using experimental data under two different conditions. The validated model is applied to investigate the optimal conditions for achieving high level of nitrate removal with relative low N_2O emission and sulfate production during mixotrophic denitrification, which is expected to provide first insights into the improvement of the design and operation of such systems for future applications.

2. Materials and methods

2.1. Model development

In mixotrophic denitrification process, autotrophic and heterotrophic denitrifiers use their corresponding electron donors (sulfur and organic matter, respectively) to reduce nitrate to nitrogen gas. The biological reactions in this model were based on Liu et al. (2016c) and Schulthess and Gujer (1996), considering the three-step denitrification (NO_3^- to N_2 via NO_2^- and N_2O) driven by both sulfur and organic matter to describe all potential N_2O accumulation steps (Fig. 1). Nitric oxide (NO) is not taken into account in our model since the NO reduction related parameters are beyond the ability of measurement. Indeed, NO reduction is usually prioritized by bacteria to avoid its toxicity and thus ensure no accumulation of NO as intermediate (Liu et al., 2015c). Also, thiosulfate and sulfite are not considered in the biological model since they are the intermediate products of chemical oxidation rather than biological oxidation (Liu et al., 2015a; Xu et al., 2016).

The model describes the relationships among seven compounds involved in autotrophic denitrifiers (X_{SOB}) and heterotrophic denitrifiers (X_{HB}), namely NO_3^- (S_{NO3}), NO_2^- (S_{NO2}), N_2O (S_{N2O}), N_2 (S_{N2}), S^0 (S_{S0}), SO_4^{2-} (S_{SO4}) and soluble chemical oxygen demand (S_{COD}). The units are g-N m^{-3} for all nitrogenous species, g-S m^{-3} for sulfurous species and g-COD m^{-3} for other compounds (Table S1 in SI). Two groups of biological processes (Table S2 and S3 in SI) were considered, namely, sulfur-driven autotrophic denitrification processes (Process 1–3) and heterotrophic denitrification processes (Process 5–7), each modeled as three sequential denitrification processes from NO_3^- to N_2 via NO_2^- and N_2O with individual reaction-specific Monod-type kinetics. In addition, biomass decay of autotrophic denitrifiers (Process 4) and heterotrophic denitrifiers (Process 8) was also included. All model parameter values were obtained from literature, as presented in Table S4 in SI (including definitions, values, units, and sources).

2.2. Model evaluation with experimental data

Experimental data previously reported by Zhang et al. (2015a) was used to evaluate the proposed mixotrophic denitrification model. An enriched autotrophic denitrifying culture was employed as the inoculum and further developed in two 2.3-L lab-scale continuous-flow anaerobic fluidized bed membrane bioreactors. Initially, 200 g of sulfur were added to each reactor as electron

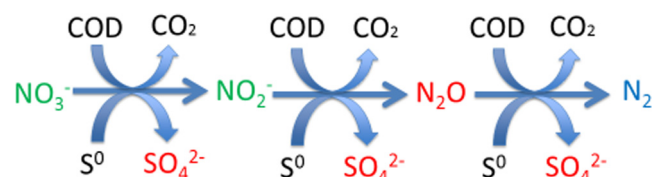


Fig. 1. Schematic representation of the proposed N_2O model concept in mixotrophic denitrification processes.

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