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Nitrous oxide production in completely autotrophic nitrogen removal biofilm process: A simulation study

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HIGHLIGHTS

- A previous N_2O model is applied in partial nitritation and Anammox biofilm system.
- Considerable amount of N_2O is produced at low DO level for high N removal.
- Lowering ammonium concentration could reduce N_2O production.
- Biofilm thickness control is critical for high TN removal and low N_2O production.

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ABSTRACT

The dissolved oxygen (DO) concentration is known to be one of the most important factors affecting nitrous oxide (N_2O) production, which might weaken the advantages of nitrogen removal in completely autotrophic nitrogen removal biofilm process. In this work, a mathematical model is applied to study the N_2O production in a biofilm reactor performing nitritation followed by anaerobic ammonium oxidation (Anammox) for nitrogen removal. The nitrifier denitrification pathway through utilization of nitrite as the terminal electron acceptor under oxygen limiting conditions is used to predict N_2O production. Simulations explicitly show that a large number of N_2O is produced under conditions of low DO concentration for high nitrogen removal. A low ammonium concentration (<50 mg N L^{-1}) and a suitable DO level (at around 0.5 mg O_2 L^{-1}) could lead to high total nitrogen (TN) removal with a low N_2O production. Biofilm has to be controlled to be in the optimal thickness (1000 μm under the simulating conditions of this study), which allows relatively high TN removal, avoiding higher thickness that favors N_2O production.

1. Introduction

Considering the negative influence of nitrogen compounds (ammonium, NH_4^+-N; nitrite, NO_2^--N; nitrate, NO_3^--N) on the aquatic environment, more stringent legislation on wastewater discharges has been enacted. Hence, nitrogen removal has attracted special attentions and is of great significance during wastewater treatment. Biological nitrogen removal comprising both aerobic nitrification and anoxic denitrification has been widely practiced in wastewater treatment plants (WWTPs). However, a considerable amount of nitrous oxide (N_2O) could be generated during these processes [1]. Since nitrous oxide is not only a potent greenhouse gas with approximately 300 times stronger greenhouse effect than carbon dioxide, but also leads to the depletion of stratospheric ozone layer [2], even a small amount of emissions is environmentally hazardous. A better understanding of N_2O production would help to develop mitigation strategies in WWTPs.

N_2O can be produced by both autotrophic ammonia oxidizing bacteria (AOB) [1,3–5] and heterotrophic denitrifiers [6,7]. Increasing evidence shows that AOB are major contributors to N_2O production from WWTPs as well as from other ecosystems [4,5,8–10].

Complete autotrophic nitrogen removal over nitrite has been developed to promote nitrogen removal economically from high nitrogen strength wastewater [11]. The combination of partial nitrification and anaerobic ammonium oxidation (Anammox) in complete autotrophic nitrogen removal has a high potential for more sustainable ammonium removal processes [12]. A one-stage nitrogen removal process with partial nitritation coupled to Anammox in a biofilm system has been studied for nitrogen

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removal [11]. Ammonia oxidation to nitrite by AOB takes place under aerobic conditions with oxygen as the terminal electron acceptor. Subsequently, the produced nitrite is anaerobically converted to dinitrogen gas with ammonium as the electron donor by Anammox bacteria. The advantage of this process involves low sludge yield, no requirement of organic carbon (COD) supplement and reduced aeration energy consumption (only 40% of a conventional nitrogen removal process) [11,13].

In complete autotrophic nitrogen removal biofilm process, nitrite is accumulated as an intermediate product. Nitrogen removal via nitrite can be achieved through appropriate DO control at low concentration. However, these conditions for obtaining nitritation by AOB are usually key factors leading to N_2O production as well. N_2O production by AOB may occur in two ways (Fig. S1): incomplete hydroxylamine (NH_2OH) oxidation to nitrite [14,15] and nitrite reduction to N_2O in a process termed as nitrifier denitrification [4,5,16]. The nitrifier denitrification pathway has though been considered as a key pathway under the conditions of low DO (<1.0 mg O_2 L^{-1}) [17,18] and favored by nitrite accumulation [5,19], while the NH_2OH oxidation pathway was promoted by increasing DO [14,18].

Mathematical modeling has proven to be essential for comprehensively understanding N_2O production processes by AOB [19–21]. Peng et al. [18] and [19] utilized a N_2O model incorporating both nitrifier denitrification pathway and NH_2OH oxidation pathway to interpret experimental data based on a nitrifying culture and established the dependency of the contributions of N_2O production pathways on DO and nitrite concentrations in a two-dimensional space.

For improving the design and operation of complete autotrophic nitrogen removal, a variety of systems such as one-stage and two-stage partial nitritation and Anammox granular sludge reactors, biofilm reactors as well as membrane aerated biofilm reactors were studied at varying operational conditions (i.e. aeration and influent nitrogen) [22–25]. It is also critical to take into account N_2O emissions from such systems in order for a sustainable nitrogen removal [26]. Recently, Castro-Barros et al. [27] evaluated the effect of aeration regime on N_2O emission from full-scale granular sludge reactor for autotrophic nitrogen removal. However, there is still a lack of systematical assessment on the DO dependency and effect of ammonium concentration on N_2O production in biofilm reactor performing partial nitritation and Anammox. We perform the model-based simulation due to the fact that the extremely slow growth rate of Anammox organism and slow response of stratified microbial abundance to the changing operational conditions make experimental assessment very time consuming. Model-based predictions for reactor design and operation will support the operation of complete autotrophic nitrogen removal biofilm reactor under steady-state or dynamic conditions regarding N_2O production [21,28,29].

In this study, an existing N_2O model [16] was applied to describe the N_2O production in nitritation and Anammox processes in complete autotrophic nitrogen removal biofilm reactor. This biofilm-modeling study focuses on evaluating the effect of DO, ammonium, and biofilm characteristics on N_2O production and nitrogen removal. The results of this work could potentially provide a better understanding of the N_2O production in completely autotrophic nitrogen removal biofilm process and hence improving the design and operation of the system for nitrogen removal.

2. Materials and methods

2.1. Mathematical model

The interactions between microbes and substrates were simulated by a one-dimensional biofilm model, constructed using software AQUASIM V2.1 [30]. A wide range of operational conditions were then tested for completely autotrophic nitrogen removal of the system. The biofilm reactor was modeled through consisting of two different compartments: a completely mixed gas compartment and a biofilm compartment, containing the biofilm and bulk liquid. Both oxygen and ammonium were supplied from the bulk liquid. The gas compartment was linked to the bulk liquid of the biofilm compartment to model the aeration of the liquid phase. The biofilm structures were represented as a continuum and the biofilm matrix contained no diffusive mass transport of biomass. The related parameters are referred to Hao et al. [11]. All simulations were initiated under the following premier conditions: a biofilm thickness of 20 μm; fractional composition of the biofilm solid phase of AOB (0.72), nitrite oxidizing bacteria (NOB) (0.20), and Anammox bacteria (0.08); zero concentration for all soluble components in both the biofilm and bulk liquid. The biofilm was discretized in 20 grid points. The ratio of biofilm surface area to bulk volume of the biofilm reactor was set as 245 m^2 m^{-3}. An unconfined biofilm compartment with a constant bulk volume was constructed to maintain a constant specific loading rate and the same surface loading of NH_4^+-N.

A mathematical model incorporating both nitrifier denitrification pathway and NH_2OH oxidation pathway has been developed and validated using suspended-growth cultures by Ni et al. [31]. However, the DO concentrations in completely autotrophic nitrogen removal systems are controlled to be low, nitrifier denitrification could likely be the dominant contributor to N_2O generation by AOB [1,32,33]. The subsequent application of this two-pathway model in biofilm system revealed that the diffusion of NH_2OH from aerobic to anoxic regions of the biofilm significantly enhanced N_2O production via nitrifier denitrification pathway [34]. As a result, the kinetics for N_2O production through nitrifier denitrification by AOB is applied in the reaction model [16]. Ammonium is oxidized to nitrite via NH_2OH. Nitrite and NO could act as the electron acceptor under oxygen limiting conditions, utilizing electrons generated from NH_2OH oxidation and producing N_2O as the end product. Other bacterial metabolisms for AOB, NOB and Anammox bacteria in our model are as described by Hao et al. [11], which considers both the growth and endogenous respiration processes. The inhibitory effects of free nitrous acid (FNA) and free ammonia (FA) on microbes are expected to be minimal in the biofilm system due to the low accumulation of ammonium and nitrite (mostly converted to N_2) and neutral pH level (buffering capacity in wastewater). The kinetics and parameter values for N_2O production, nitrification and Anammox processes are based on literature knowledge, experimental data and calculations, which were evaluated in different systems. Therefore, we directly adapt them into the model and evaluate the substrate and microbial interactions in the completely autotrophic nitrogen removal biofilm reactor, which has been demonstrated to be a valid method in previous simulation studies [21,34,35].

Overall, there are 4 COD components identified for carbonaceous material and 7 components for nitrogenous material. The active biomass is divided into AOB performing nitritation, NOB performing nitritation, and Anammox organisms performing Anammox process. The model describes the relationships among the four particulate species: AOB (X_{AOB}), NOB (X_{NOB}), Anammox organisms (X_{AMX}), and residual inert biomass (X_I); eight soluble species: ammonia (S_{NH4}), NH_2OH (S_{NH2OH}), nitrite (S_{NO2}), nitrate (S_{NO3}), nitric oxide (NO, S_{NO}), N_2O (S_{N2O}), N_2 (S_{N2}); and DO (S_O). The units for all non-nitrogen species are oxygen demand or oxygen, which is directly proportional to electron equivalents (8 g O_2 per e^- equivalent). The kinetics and stoichiometry describing the conversions and interactions among model components are adapted from previous studies and expressed in Peterson-matrices (Tables S1 and S2 in Supplementary material). Table S3