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N_2O emissions from a single-stage partial nitritation/anammox granule-based reactor – a model based assessment

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ABSTRACT

Completely autotrophic nitrogen removal (CANR) can be obtained in single stage granule-based bioreactors. However, their environmental footprint is compromised due to elevated N_2O emissions. In this study we develop and present results from a novel spatially explicit biochemical process model of granular CANR systems that predicts N_2O dynamics and stripping. The model is adapted to predict pH gradients within the granule and considers different granule size distributions and their impact on N_2O emissions. The model will serve as a guiding tool to define an operational window, where aeration strategies, granule size and bulk pH are optimized to retain high nitrogen removal whilst minimizing the N_2O production.

INTRODUCTION

Recently, alternatives based on autotrophic ammonium oxidation have been implemented as more energy and resource efficient processes than conventional nitrification/denitrification based BNR (biological nitrogen removal) processes for nitrogen removal from ammonium-rich/organic-carbon-poor residual streams (Lackner et al., 2014). Completely autotrophic systems have a low oxygen demand and no external carbon source requirement, which reduces their sludge production, energy consumption, and carbon footprint. Single-stage nitritation/anammox systems rely on two central microbial functional guilds, aerobic and anaerobic ammonium oxidizers (AOB, AnAOB), but also contain aerobic nitrite oxidizing bacteria (NOB) and heterotrophic bacteria (HB). Optimal conditions for autotrophic nitrogen removal in suspended-growth single-stage systems, such as low and transient dissolved oxygen (DO) and the presence of nitrite, may promote emissions of nitrogen oxides (N_xO). Nitrous oxide (N_2O) and nitric oxide (NO) are atmospheric trace gases, of which N_2O is known as both a stratospheric ozone depleting and a greenhouse gas with 300 times higher radiative forcing than carbon dioxide. The total greenhouse gas (GHG) footprint of a WWTP can be sensitive to its N_2O emissions. Documented N_2O emissions from single-stage partial nitritation/anammox systems (PN/A; 0.1-10 % of the removed nitrogen ammonium; Ali et al., 2016) have been higher than those measured from conventional BNR processes, potentially offsetting the benefit associated with their energy savings and corresponding CO_2 emission reductions.

Therefore, the main goal of this study is to develop a simulation model, which will be used to identify the main pathways producing N_2O emissions in PN/A systems and to develop mitigation strategies to minimize emissions.

METHODS

Modelling approach

The simulation model presented by Vangsgaard et al. (2013), which includes microbial growth of AOB, NOB, AnAOB and HB and pH influence on the microbial activity, is extended to model processes relevant to N_2O production and consumption. Specifically, AOB related processes (two autotrophic pathways) are modelled as suggested by Domingo-Félez and Smets (2016) whilst denitrification by HB is modelled as suggested by Hiat and Grady (2008). Consistent with experimental studies, the biological model considers NO as the direct precursor of N_2O in all three biologically-driven pathways. This model can describe all relevant NO and N_2O production pathways with fewer parameters than other biological models, thereby simplifying the calibration of the biofilm simulation model.

A pH calculation algorithm, which is based on 3 mass balances, 5 acid-base equilibria and a global charge balance, is implemented to estimate pH profiles in the granule as well as pH in the bulk liquid. The resulting system of 9 non-linear algebraic equations are combined and solved using the Brent-Dekker method, which is a more robust method compared to conventional multidimensional Newton-Raphson approach (see, e.g., Vangsgaard et al., 2013). The model is implemented and solved in Simulink environment in MATLAB R2016b (The MathWorks, Natick, MA).

Scenario analysis

Based on our previous experimental studies we have defined several scenarios, whereby we evaluate the impact of aeration strategies, aggregate size distribution and pH on N_2O emissions from a granular PN/A system.

Under oxygen limiting conditions AOB can outcompete NOB, thus the aeration strategy is a key design variable for PN/A systems. Whilst AOB and AnAOB can thrive under continuous and low intensity aeration, intermittent aeration has been shown to hinder NOB proliferation. However, the effects of aeration strategies on N_2O production from single-stage PN/A have not been elucidated. Thus the aeration rate (or DO set-point), and the frequency of aeration will not only affect the N-removing performance of the system but also its N_2O emissions (Domingo-Félez et al. 2014).

NO_2^- has been correlated with increased N_2O emissions from the nitrifier denitrification pathway. To reduce the availability of NO_2^- in PN/A systems, thereby potentially reducing N_2O production by AOB, the NO_2^- removal capacity by AnAOB can be enhanced. This can be accomplished by increasing the fraction of AnAOB relative to AOB. It was shown that different granules sizes host different fractions of AOB vs. AnAOB, where larger granules host relatively more AnAOB than smaller ones (Vlaeminck et al., 2010). Accordingly, larger granules are expected to remove more NO_2^-, thereby accumulating less NO_2^-. Consequently, lower N_2O production is expected with increasing granule size.

pH is another factor influencing N_2O production (Schreiber et al., 2012). Microbial activity is affected by pH due to the direct impact on the enzymatic activity and due to the impact on substrate speciation, which affects the distribution between protonated and un-protonated species (being the uncharged species the real substrates/inhibitors; Vangsgaard et al., 2013).

Based on the experimental observations from our previous work on PN/A (Mutlu 2015; Blum et al., 2017) we have defined different scenarios for simulation (Table1), which will serve to identify the optimal operational conditions.

OUTLOOK

This study will serve to elucidate not only which microbes contribute to increase or reduce N_2O emissions, but also to design mitigation strategies to reduce them. Importantly, the model will help to find trade-offs between operational conditions – e.g. big granules have been suggested to produce little N_2O; however, the aeration strategies that promote large granule size usually also promote N_2O production (Blum et al., 2017). Special attention will be also paid to ammonia removal, so effluent quality is not compromised whilst N_2O emissions are reduced.

Table1. Scenarios for process performance assessment (intermittent and continuous aeration will be assessed):

Dissolved oxygen (mg/L)		
0.1	0.5	1
Granule size (µm)		
Small – 90	Medium – 500	Large – 1000
pH		
6.5	7.5	8

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