Model-based optimization biofilm based systems performing autotrophic nitrogen removal using the comprehensive NDHA model

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Abstract
Completely autotrophic nitrogen removal (CANR) can be obtained in single stage biofilm-based bioreactors. However, their environmental footprint is compromised due to elevated N_2O emissions. We developed novel spatially explicit biochemical process model of biofilm based CANR systems that predicts N_2O dynamics and stripping, using the biological NDHA model coupled with a simple and roubust pH calculator. In this work we present two case studies: i) membrane aerated biofilm reactor (MABR) with focus on model calibration; and ii) granular system with focus on process optimization.

Keywords
Biofilm; nitrous oxide; scenario analysis; model calibration; optimization

INTRODUCTION

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). 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, such as low and transient dissolved oxygen (DO) and the presence of nitrite, may promote emissions of nitrogen oxides (N_xO), 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. Documented N_2O emissions from single-stage partial nitritation/anammox systems (PN/A; 0.1-10 % of the removed nitrogen ammonium) have been higher than those measured from conventional BNR processes, potentially offsetting the benefit associated with their energy savings. In the present study we aim to use the comprehensive NDHA model to predict performance of biofilm based completely autotrophic nitrogen removal (CANR) systems and optimize the processes by achieving maximum nitrogen removal at lowest N_2O emission rates.

MATERIALS AND 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 ensures convergence to a solution even with poor initial guess.The model is implemented and solved in Simulink environment in MATLAB R2016b (The MathWorks, Natick, MA).

Case studies

Both studies are based on laboratory scale reactors growing autotrophic nitrogen removing biofilms fed with synthetic ammonia rich organic carbon depleted wastewaster.

Membrane Aerated Biofilm Reactor (MABR). 0.8 L MABRs were operated with continuos feeding under intermittent aeration (Ma et al., 2017). Bulk N concentrations, microprofiles (pH, DO and N_2O) within biofilm and N_2O emissions (bulk and offgas phases) were used for model calibration. Calibrated parameters were selected via local sensitivity analysis.

Granular Biofilm Reactor. 4 L continous stirred tank reactor operated with constant aeration. For process optimization, different granule sizes and bulk pH levels were evaluated and the N and O_2 loads were optimized via scenario analysis.

RESULTS

Membrane-aerated Biofilm Reactor. Based on local sensitivity analysis a 2 steps calibration procedure was applied. Kinetic parametes, including $\mu_{AOB,AMO}$ and $K_{I,FA,AOB}$, were first calibrated to fit bulk N concentrations and pH values. Then η_{NIR} (anoxic reduction factor for NO_2^- reduction by AOB) and $K_{NH2OH,AOB}$ were calibrated to predict N_2O production. The model predicts reasonably well the reactor performance (only N_2O profile in biofilm is presented – Fig. 1a). Individual contribution of each N_2O production pathway to total emission in MABRs indicated that heterotrophs could be key contributors to N_2O production within the counter-diffusion biofilm. Thus, process optimization by controlling frequency and lenght of aeration phase should aim at minimizing nitrite concentrations within the biofilm by promoting anammox growth over heterotrophs in the anoxic parts of the biofilm (optimization not shown due to space constrains).

Granular Biofilm Reactor. Simulation results showed close agreement with lab-scale data without further calibration, predicting 87% of nitrogen removal, out of which 2.1% was emitted as N_2O (vs. 1.1% by Blum et al., 2017 under the same operational conditions). Therefore, we used the default parameter set for process model optimization (Fig.2a&b for granule radius 500μm and bulk pH 7.5). Based on scenario analysis it is preferable to operate at low ammonia load, as above 400 mg-N L^{-1} d^{-1} N_2O emissions considerably increase. Furthermore, the O_2 should be supplied at a ratio of 2 mg-O per mg-N for maximum nitrogen removal, but ensuring the system is not inhibited by ammonia, as this could enhance N_2O emissions from the system (Fig. 2b). Similar analysis was performed at different pH and granule sizes, which suggested optimal operational conditions should sustain a granule radius of 500μm, pH controlled at 7.5 and supply of O_2 to N ratio of 1.8 with a relatively low nitrogen load (i.e., 400 mg-N L^{-1} d^{-1}) to ensure mimimal N_2O emissions (Fig. 2c).

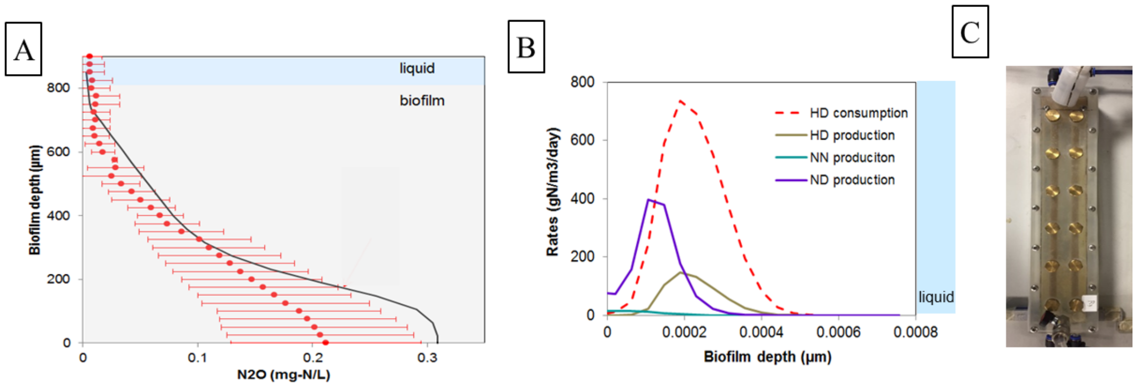

Figure 1. (**a**) Experimental (dots) and predicted (line) N_2O within biofilm during aeration phase; (**b**) predicted contribution to N_2O production/consumption by different pathways; (**c**) lab scale

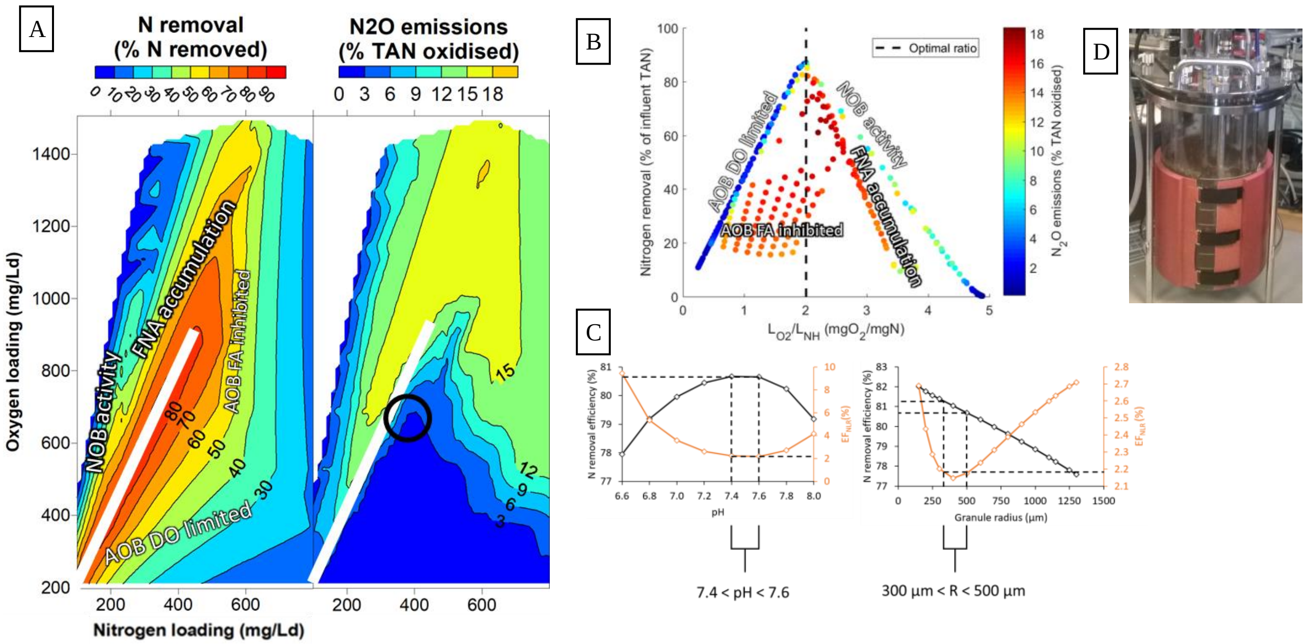

Figure 2. (**a**) Performance optimization for radius 500μm and bulk pH 7.5; (**b**) impact O_2 to N loading ratio on process performance; (**c**) nitrogen removal efficiency and N_2O emission factor as function of bulk pH (left) and granule radius (right); (**d**) laboratory scale reactor.

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