Analysis of nitrous oxide production and system performance for a full-scale anammox plant combining activated sludge and granular activated carbon biofilm: a model-based study

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Abstract

In this work, experimental and long-term one-dimensional mathematical modelling approaches were combined to investigate the mechanisms and the main drivers influencing on emission of nitrous oxide in a full scale anammox plant whereby anaerobic ammonium oxidation (anammox), ammonia oxidizing bacteria, nitrite oxidizing bacteria and heterotrophic denitrifying bacteria enriches in activated sludge treatment process, then grow on granular activated carbon surfaces as biofilms. The model was calibrated and validated using the actual real-time data of nitrogen components concentration in the effluent. The abundancy of various genera of anammox as dominant microbial group was estimated in the model by quantitative fluorescent in situ-hybridization (FISH) and image processing. A parameter identifiability protocol was applied to express the growth kinetic characteristics of microbial groups in the plant. Since nitrous oxide (N_2O) is a greenhouse gas with important impacts on our environment, various pathways of the formation of nitrous oxide were considered and simulated. Finally, the system performance in terms of nitrogen and carbon biological removal efficiency and nitrous oxide production of the plant was optimized and analyzed under different simulation scenarios such as different total nitrogen loading rate, organic substrate rate as well as hydraulic retention times.

Keywords: anammox, granular activated carbon; biofilm, nitrous oxide; system performance optimization; mathematical modeling

1. Introduction

The mathematical modelling of biological nitrogen removal processes in biofilm reactors has been studied to fully understand of the complex dynamic of biofilm structures. Mathematical modeling of N_2O emissions in such systems is also of great importance toward understanding the whole environmental impact of WWTPs, which has been recognized as an appropriate method for study the carbon footprints (Kim *et al.*, 2017; Sabba *et al.*, 2015). Nitrous oxide (N_2O) leads to ozone depletion through photo-chemical nitric oxide (NO) production in the stratosphere. Mitigating its steady increase in atmospheric concentration requires an understanding of the mechanisms that lead to its formation in natural and engineered microbial communities. N_2O is formed biologically from the oxidation of hydroxylamine (NH_2OH) via AOBs or the reduction of nitrite (NO_2) to NO and further to N_2O via ammonia oxidizing bacteria (AOBs) or heterotrophic denitrifiers as described in Fig. 1. (Schreiber, 2009; Schreiber *et al.*, 2012). Recently, mathematical models explaining the two currently known N_2O production pathways by AOB, namely the AOB denitrification and NH_2OH oxidation pathways as well as heterotrophic denitrifiers, were clarified to describe N_2O production (Ali *et al.*, 2016; Ni *et al.*, 2013, 2011). In this study, the anaerobic ammonium oxidation (anammox) and simultaneous nitrification - denitrification (SND) were mathematically simulated within a full-scale activated sludge plant combined with granular biofilm reactor in the anoxic condition. However, in current state of the art, the mathematical modelling of anammox biofilm reactors mainly focus on simulation of the main substrate concentration. But there are limited understanding and researches about how a full-scale anammox plant contributes to N_2O emissions (Meng *et al.*, 2012). The main objective of the research is to develop and apply an integrated one-dimensional mathematical biofilm model by means of calibration and validation. The specific objectives of the research is using this model as well as experimental approaches to (1) forecast fate of biomass in terms of biovolume fraction and substrate in terms of nitrogen and carbon removal, (2) explain various plausible pathways of the formation of nitrous oxide were simulated, (3) predict the time dependent concentration of nitrous oxide under various simulation strategies and (4) optimize the system performance in terms of nitrogen and carbon biological removal efficiency and carbon footprint of plant under different simulation scenarios such as different total nitrogen surface loadings, organic substrate rate as well as hydraulic retention times.

2. Materials and methods

2.1. Reactor operation and biomass

A full scale anammox plant was functioning over a decade for leachate treatment facilities and was divided into the four main stages including feeding, biological treatment with activated sludge, ultrafiltration and activated carbon biofilm process before the main outflow (Fig. 2). The main inflow rate through the plant is approx. 30 [m^3 h-1]. Previous study was found the co-existence of various genera of anammox bacteria as dominant microbial group as well as heterotrophic denitrifiers, ammonium oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB) in the biofilm in different part of this plant (Azari *et al*., 2017; Ke *et al*., 2015). This study focused on the last part of the plant before the outflow consisting of granular activated carbon or GAC modules. GACs serve as biofilm carrier for attachment of microorganisms to increase the biomass retention under nearly anoxic conditions (DO < 0.1 mgO_2 L-1). In this case, the activated carbon acts as a mobilized carrier for microbial attachment and growth and the corresponding bioreactor is as an anaerobic granular biofilm reactor. The granular sludge concentration was 4.15 ± 1.1 g L-1 of mixed liquor volatile suspended solids (MLVSS).

2.2 Model development

Kinetics and stoichiometry of nitrogen removal processes. A model for anaerobic description of anammox-denitrifiers interactions illustrated by (Azari *et al*., 2016) was expanded considering AOBs and NOBs. Nitrous oxide (N2O) emission pathways as an environmentally important atmospheric trace gas leading to ozone depletion through photo-chemical nitric oxide (NO) production in the stratosphere, were incorporated in the model (see Fig. 3). We applied a mathematical model that quantitatively illustrated SMP production, EPS production and EPS hydrolysis rates due to Liu *et al*. (2016). SMP are categorized into two groups based on the type of bacterial metabolism from which they are produced. UAP is assumed to be produced directly from substrate utilization, and biomass- BAP is produced directly from the hydrolysis of a biomass component. Activated Sludge Model No. 1 (ASM1) by the task group of the International Water Association (IWA) was used and the biofilm model in software AQUASIM 2.1 (EAWAG, Switzerland) was applied.

2.3 Chemical analysis

Concentration of NH4-N, NO2-N and NO3-N were measured photo-metrically per the German standards (Dr. Lange, Hach, Germany).

2.4 Molecular cytogenetic analysis

The biomass was enriched from the steady state operation of the anaerobic GAC biofilm reactor. Five monthly timeframes were used for sampling. For each sample, qFISH using five oligonucleotide probe (AMX820, Sca1309, Nso190, Ntspa662 and NIT3) was done in duplicate. 20 FISH images from randomized field of observations (FOVs) were obtained using light and confocal laser scanning microscopy (CLSM) to determine the average biovolume fraction of each microbial group.

2.5 Model calibration

The parameter values were estimated by minimizing the sum of squares of the deviations between the measured data and the model predictions using the secant method. The secant optimization method is well suited for the minimization of numerically integrated equations using linear approximation of the model functions, which can lead to a much faster end convergence being close to a well-defined minimum. The standard errors and 95% confidence intervals of individual parameter estimates were calculated from the mean square fitting errors and the sensitivity of the model to the parameters (Hao *et al*., 2001; Lübken *et al*., 2015; Ni *et al*., 2012).

3. Results And Discussion

3.1 Evaluation of nitrogen removal performance

Nitrogen removal for total duration of the simulation for the last compartment of the anammox plant i.e. anaerobic GAC biofilm reactor is presented in Fig 3. which is almost fitting the observation and it compromises a good agreement between modelled and measured data. The correlation of the observation and simulation showed that good-ness of fitness R-squared index is 0.53 for total nitrogen concentration.

3.2 Prediction of N2O Production Rate.

The model predicted N_2O fluxes during the 345 days' operation of the reactor at different locations showed that the N_2O emissions were highly dynamic but lower than 2.5 gN_2O day^{-1}.

4. Conclusion and outlooks

A mathematical model for nitrogen removal by different microbial groups was verified after a comprehensive sensitivity analysis and parameter estimation. The model was used to explain the kinetics of nitrogen removal via anammox, AOB and denitrifying bacteria. The model performance briefed a good agreement between measured and predicted values.

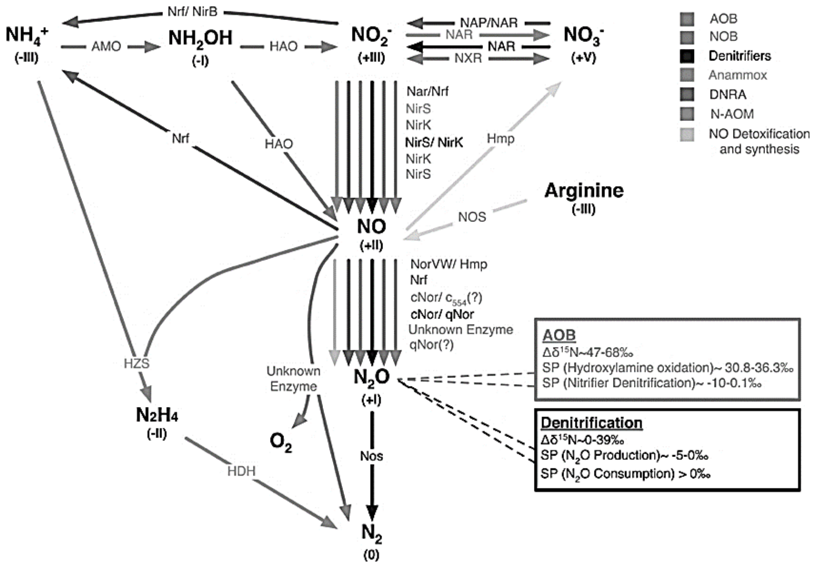

Fig. 1. Biological pathways for NO and N2O turnover in the catabolic branch of the N-cycle

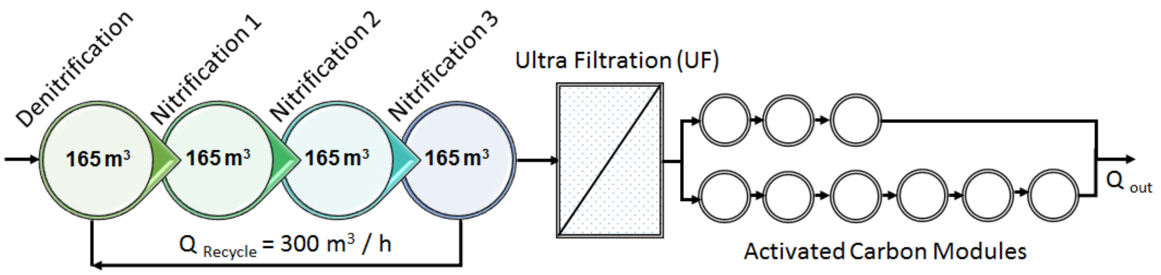

Fig. 2. The illustration of combination of activated sludge and activated carbon biofilm system in the plant

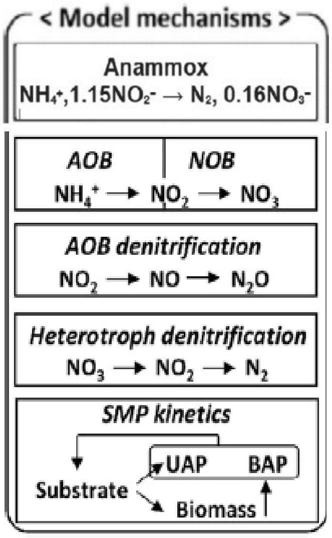

Fig. 3. The demonstration of model structure

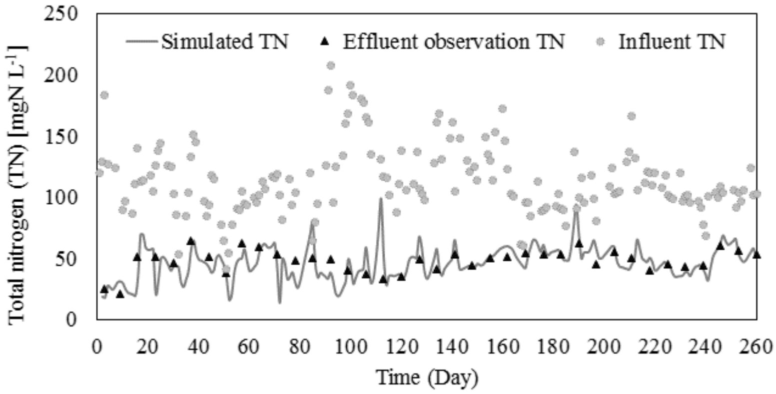

Fig. 4. Model evaluation using the total nitrogen concentration data: simulation and observation

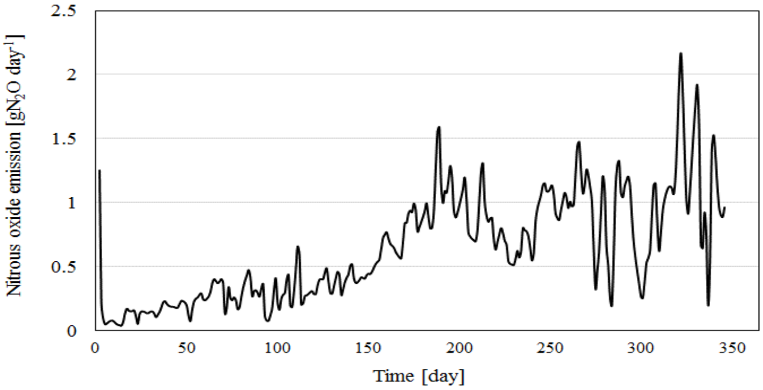

Fig. 5. Model evaluation results of one year N2O production from WWTP

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