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Modelling Greenhouse Gas Emissions of a Hybrid Fixed-film Anammox Process Treating Sludge Dewatering Centrate in Wastewater Treatment

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Authors' contributions

This work was carried out in collaboration between both authors. Both authors read and approved the final manuscript.

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ABSTRACT

The aim of this study is to estimate and optimize greenhouse gas (GHG) emissions of a process in wastewater treatment, which utilizes anaerobic ammonium oxidation (Anammox). The single-stage nitritation-Anammox process applies fixed biofilm carriers and treats the centrate of sludge dewatering. GPS-X biokinetic modelling tool was used for quantifying the specific nitrous oxide, carbon dioxide and methane emissions at various operational conditions. In general, the amount of biology related GHG production was estimated to be higher than that of indirect emissions, by three orders of magnitude. Of direct emissions, nitrous oxide gas production should be taken into account primarily. Based on the simulations, feasible options of minimising N_2O emissions include applying an operational temperature of 30-35℃, and increasing airflow to reduce the effect of oxygen limitation. To release less N_2O, the process should also preferably be operated as an IFAS application with a low concentration of suspended solids (1.5-2 g/L), or even without sludge recycle.

Keywords: Anammox; dewatering centrate; greenhouse gases; wastewater treatment.

1. INTRODUCTION

Anaerobic ammonium oxidation (Anammox) is a commonly used process in wastewater treatment, where ammonium nitrogen is converted into gaseous dinitrogen under anoxic conditions with nitrite as the electron acceptor [1]. Prior to the Anammox reaction, ideally a

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mixture of nitrite and ammonia is prepared in a so-called Sharon process, where part of the influent ammonia-N is oxidized to nitrite-N, by ammonia oxidizing bacteria [2]. To avoid the conversion of nitrite-N into nitrate-N by nitrite oxidizers, the Sharon process operates at a high temperature (>30℃) and pH of 7-8, generally without sludge retention [3]. Anammox microorganisms are inhibited reversibly by the presence of oxygen. Research studies have shown that aerobic nitrifiers do not play an important role in the main process itself [4].

Anammox is applied as a side-stream technology to reduce the load of reject water from sludge management to the bioreactors treating municipal wastewater, since 15–20% of the inlet nitrogen load is recycled with the return liquors from sludge dewatering [5]. There are examples of Anammox solutions from the whole spectrum of the leading wastewater treatment technologies; e.g. activated sludge in sequence batch reactors [6], rotating biological contactors [7], moving bed biofilm reactors [8], membrane bioreactors [9] and technologies applying upflow anaerobic sludge blanket [10]. The integrated fixed-film activated sludge (IFAS) construction can also be applied for Anammox treatment, with the use of clarifiers for sludge recycle; allowing suspended biomass to be retained in the bioreactors, apart from the biofilm bound to the carrier media. The slightly aerated suspended phase is ideal for implementing nitritation as part of the Sharon process, while Anammox microorganisms generally reside in the inner, anaerobic biofilm layers [11]. Although many studies describe the design and operation of such a system, during in-depth literature review no specific papers were found investigating GHG emissions of Anammox applications in terms of operational parameters. In this paper the authors' contribution to Anammox technology is to optimize GHG emissions originating from this process.

The commonly discussed greenhouse gas emissions in wastewater treatment are those of carbon dioxide, methane and nitrous oxide. Biology related carbon dioxide emissions derive from the degradation of organic matter and the aerobic respiration of biomass [12]. During anaerobic processes, methane is generally produced concurrently with carbon dioxide. The quantity of CH_4 depends on the amount of organic matter in wastewater; as well as the temperature and the type of treatment system applied [13]. Nitrification involves autotrophic (ammonia-oxidizing) bacteria, which convert the NH_4^+ ions into the intermediate compound of NH_2OH, followed by NO_2^- ions. Because of the latter step, NO and N_2O are released as by-products. Nitrite oxidizers transform NO_2^- ions to NO_3^-. The build-up of NO_2^- ions can lead to an increased production of N_2O gas. Furthermore, due to low dissolved oxygen conditions, ammonia oxidizers can also consume NO_2^- as a source of oxygen, which is then reduced into NO, then N_2O, as a result of the process known as autotrophic denitrification [14]. However, nitrifiers also consume CO_2 as an inorganic carbon source [15]. Denitrification is a heterotrophic process involving four metabolic stages, during which NO_3^- ions are formed into NO_2^- ions, NO, N_2O, and then N_2 gas. Lower C/N ratios of wastewater can cause higher emissions of nitrous oxide [14].

Operation of the Sharon-Anammox process also emit N_2O, especially due to nitrite accumulation, which results in higher N_2O concentrations in the off-gas [16]. The emissions can be reduced by applying a combined single-stage technology for the nitritation and Anammox processes (such as IFAS), partly preventing the accumulation [17]. Efficient control of aeration is required in such systems, since oxygen limitation is generally assumed to cause increased N_2O emissions [16]. It is proposed that N_2O emissions from single-stage nitritation-anammox reactors can also be minimised by operating the technology under conditions where anaerobic activity exceeds aerobic activity [18].

2. MATERIALS AND METHODS

2.1 Model Used for Estimating GHG Emissions – Mantis 3

Mantis 3, a biokinetic model developed for GPS-X by Hydromantis, was used for quantifying GHG productions of the wastewater treatment plant – focusing on the hybrid Anammox reactor –, in CO_2 equivalents. The Petersen matrix and mathematical scheme of Mantis 3 is built based on the development of ASM2d. It covers the biological, physical and chemical processes experienced in wastewater engineering, such as hydrolysis, as well as metabolisms involving heterotrophs, autotrophs and phosphorus accumulating. It interprets nitrification and denitrification as two-step processes. It also incorporates denitrification by autotrophic bacteria, utilizing NO_2^- as the electron acceptor instead of O_2. A major function in terms of GHG emissions is the simulation of gas-liquid transfer processes: apart from the exchange of oxygen between the gas and liquid phase, it also takes into account the absorption and desorption of CO_2, N_2, CH_4, H_2 and N_2O, based on K_La volumetric mass transfer coefficients and their related saturation concentrations [19].

In Mantis 3, the most important feature for estimating GHG emissions is the integrated Carbon Footprint (CF) module. It classifies gas productions into three types. The sources of these emissions are detailed as follows.

Direct emissions related to biology:

- CO_2 discharges from anaerobic, anoxic, and aerobic biological processes;
- N_2O production of nitrification and denitrification
- CH_4 emitted by anaerobic processes.

Indirect emissions related to energy consumption:

- Emissions attributed to pumping energy requirement;
- Emissions caused by the energy demand of aeration;
- Miscellaneous, energy use emissions.

Material emissions:

- Emissions caused by usage of chemicals;
- Emissions brought about by use of materials, such as membranes or media;
- Emissions related to transportation of materials.

The CF module also includes offsets of treatment plants, that help reduce net emissions.

The following offsets can be applied for sequestering direct emissions:

- Biogenic capture of CO_2 (by nitrification, for example);
- Flaring of CH_4;
- Applying CH_4 for heating and energy production.

Practically, there is no way of offsetting the two other types of emissions mentioned.

Eq. (1) is used as a method of estimating direct emissions, at a given time of *t*:

$$E_{scope1,i}(t) = K_L a_i(t)\left(C^*_{\infty,i}(t) - C_{L,i}(t)\right)V(t)f_{GWP,i} \quad (1)$$

Where,

$E_{scope1,\,i}(t)$: direct emissions of gas *i*, interpreted in CO_2 equivalent (gCO_2/d)
$C_{L,\,i}(t)$: concentration of dissolved gas *i* in a reactor (g/m^3);
$K_La_i(t)$: volumetric mass transfer coefficient of gas *i*, at field conditions (d^{-1});
$V(t)$: reactor volume (m);
$C^*_{\infty,\,i}(t)$: equilibrium concentration of dissolved gas *i* (mg/L);
$f_{GWP,\,i}$: global warming potential of gas *i* (-); 25 in case for CH_4, and 298 for N_2O [20].

The model quantifies indirect emissions based on Eq. (2), also at a time of *t*:

$$E_T(t) = \left(P_{pump}(t) + P_{blower}(t) + P_{mis}(t)\right) \cdot 24 \quad (2)$$

Where,

$E_T(t)$: daily electricity consumption (kWh/d);
$P_{pump}(t)$: pump power (kW);
$P_{blower}(t)$: blower power (kW);
$P_{mis}(t)$: miscellaneous power (kW).

The amount of indirect emissions is deduced by using the following formula, Eq. (3):

$$E_{scope2,i}(t) = E_T(t) \cdot f_{elec,i}(t) f_{GWP,i} \quad (3)$$

Where,

$E_{scope2,\,i}(t)$: indirect emissions of gas *i* in CO_2 equivalent (g/d),
$f_{elec,\,i}$: gas *i* emission factor for electricity generation (-).

The applied emission factor is a region specific value found in databases. This presumes that the three examined gases are produced in a set ratio, based on the generally known energy production processes of a given region. Certain regions can be chosen in GPS-X, of which the US national was selected for our simulations, to generalize GHG calculations [19]. Mantis 3 can also be used for estimating emissions by use of materials. In this study however; effects of chemical dosing, or replacement of carriers over time were not included.

2.2 Mathematical Model Setup of the Treatment Plant and Anammox Process

The wastewater plant – utilizing the Anammox process for centrate treatment – uses a fixed bed biofilm reactor cascade for biological wastewater treatment. The process layout was built in GPS-X (Fig. 1). The design capacity of the plant is 76 000 m^3/d, the average influent and effluent water quality parameters are summarized in Table 1.

Table 1. Water characteristics of the sewage treatment plant influent and effluent

Parameter	Influent value	Effluent value
Temperature (℃)	23	23
COD (mg/L)	305	27
$cBOD_5$ (mg/L)	153	5
TSS (mg/L)	142	4
TN (mg N/L)	46	20
NH_4-N (mg N/L)	36.4	4.9
TP (mg P/L)	5.8	3.2

The bioreactor cascade is set up of six biofilm reactor stages, without recycled sludge. It is designed for organic matter removal and nitrification; all reactor units are aerobic, with dissolved oxygen setpoints of 3 mg/L. The cascade incorporates an arranged mesh of fabric threads as a fixed bed biofilm carrier, with a specific surface of 200 1/m and filling ratio of 0.066 m^3/m^3. The total reactor volume applied for sewage treatment is 12920 m^3.

Disc filter units with a pore size of 30 µm are used for phase separation after the biological reactors. They provide solids removal of 97%, and require counter-flow backwashing to remove the residual filter cake. The sludge removed by backwash from the filter units – with a quantity of 2280 m^3/d – is forwarded to a gravity thickener, from which the supernatant is recycled to the bioreactors. Approximately 240 m^3/d thickened sludge is generated, and further treated by an anaerobic digester applying a hydraulic residence time of 30 days. The digested sludge is dewatered by a centrifuge, from which the sludge cake is temporarily contained, then transported away.

Prior to being recycled to the reactor cascade, the centrate from dewatering is treated by a fixed-film Anammox system, to lower its high nitrogen content due to anaerobic digestion. This process is based on an integrated fixed-film activated sludge (IFAS) Anammox reactor, with a volume of 150 m^3, and filling ratio of 0.071 m^3/m^3; utilizing the same fixed bed biofilm carrier as the sewage treatment cascade. A dissolved oxygen concentration of 0.4 mg/L, and a temperature of 35℃ is applied for operation of this single-stage process. A clarifier is used for recycling the suspended biomass into the Anammox reactor. The effluent is forwarded to the bioreactors, and so is the wasted sludge, containing active nitrifying biomass that can be used for continuous inoculation of the cascade. The IFAS Anammox reactor receives – in average – a 210 m^3/d hydraulic inflow of centrate. The main influent and clarifier effluent characteristics of the centrate are revised in Table 2.

Table 2. Main influent and effluent parameters of the hybrid Anammox system

Parameter	Influent value	Effluent value
COD (mg/L)	1252	30
TSS (mg/L)	1368	13
TN (mg N/L)	803	154
NH_4-N (mg N/L)	726	128

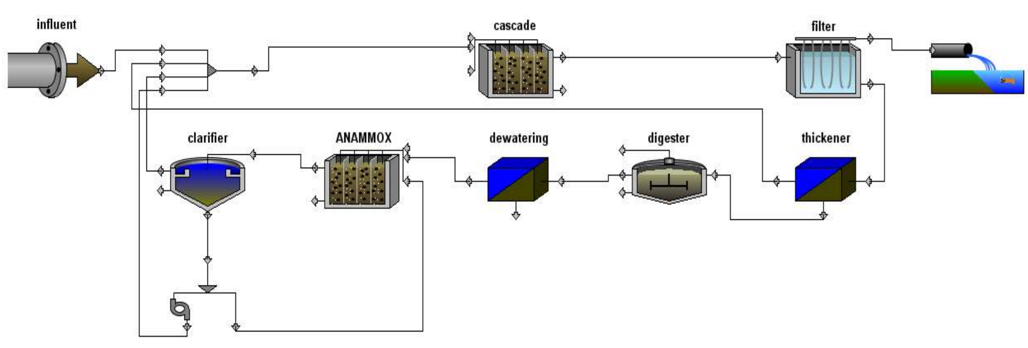

Fig. 1. Layout of the wastewater treatment plant with the hybrid Anammox system

2.3 Modell Calibration and Validation

Based on IWA Good Modelling Practice Guidelines [21] model calibration is a step-wise procedure whereby different aspects of the plant model (in this case: sludge production, nitrification, denitrification, oxygen transfer) are calibrated in sequence. The procedure consists of characterization and fractionation of the influent wastewater, the specification of operational variables (e.g. flow rates), and the adjustment of key model parameters (e.g. target biofilm thickness) in order to minimize the error between measured and calculated data. The changes to the influent fractions were made based on the influent data and effluent soluble COD. The particulate COD/volatile suspended solids ratio in the model was adjusted match the influent VSS. The main calibration parameters of fixed film processes were as follows: anoxic growth reduction factor for ordinary heterotrophic organisms, biofilm mass per surface area ratio, rate of diffusion of pollutants into the biofilm. As the result of the calibration the standardized residuals for the variables were examined and the model results were within two standard deviations of the measured values. The whole procedure and a more detailed evaluation approach is published in the literature [21].

3. RESULTS AND DISCUSSION

3.1 Evaluating Emissions of the Treatment Plant and the Anammox Process

Direct and indirect GHG productions were modelled, interpreting the standard operational conditions of the wastewater treatment plant, to compare the emissions from the Anammox treatment to the other biological treatment units. The summarized results are stated in Table 3, as specific emissions relative to the influent wastewater flow, in CO_2 equivalents.

The majority of GHG emissions originates from the sewage treatment cascade, the Anammox reactor's direct emissions are approximately 10 percent of the cascade's biology related GHG production. Indirect emissions from the cascade are more substantial, only lower by two orders of magnitude than its direct emissions; compared to the Anammox process where the quantity of indirect emissions is lower by three orders of magnitude than the biological emissions. This is mostly due to the fact that the nitritation-Anammox reactor operates at a reasonably lower DO concentration, requiring a lower specific airflow. The anaerobic digester is the unit with the lowest amount of direct emissions, comprising mostly of CO_2, due to the combustion of most of the CH_4 gas. The simulation software implies that in case of the digester, there is no need to account for indirect emissions, as the energy requirement of heating can be covered by burning the CH_4 content of the biogas.

Table 3. Summary of estimated GHG emissions from the main operational units

Parameter	Value
Direct emission from reactor cascade (g CO_2/m^3 water)	1927
Indirect emission from reactor cascade (g CO_2/m^3 water)	45
Direct emission from digester (g CO_2/m^3 water)	105
Indirect emission from digester (g CO_2/m^3 water)	-
Direct emission from Anammox process (g CO_2/m^3 water)	201
Indirect emission from Anammox process (g CO_2/m^3 water)	0.6

The N_2O production of the IFAS Anammox process is much more significant compared to CO_2 and CH_4 emissions, than in the case of the bioreactor cascade, since reactors applying the nitritation and Anammox processes mainly involve nitrogen removal; and are not purposely intended for removal of organic carbon, like sewage treatment operations are.

3.2 Effects of the IFAS Anammox Operational Parameters on GHG Production

Five process variables of the nitritation-Anammox system were analysed regarding CO_2, CH_4 and N_2O production, by running steady-state simulations. This study focuses on biology related emissions since the indirect emissions are negligible in comparison, as mentioned previously.

The specific gas productions – relative to the volumetric flow of centrate – were quantified in CO_2 equivalents and they should be minimized besides meeting the target effluent quality. The characteristics of the centrate given in Table 2 were specified as a baseline for the simulations, varying one IFAS operational parameter at a time. Input parameters related to other units of the wastewater treatment plant have not been modified during the analyses.The ranges of operational parameters were selected based on experiences, the hydraulic residence time is between 10.5 and 22.5 hours, the temperature is between 30 and 50℃, the DO setpoints are between 0.2 and 1.0 mg/l, the filling ratio is between 0.04 and 0.08 mg/l and the mixed liquor suspended solids concentration is between 1400 and 4000 mg/l. The effect of the parameters on GHG emission were analyzed individually, while one was being varied the other remained constant. Complex optimization algorithms with varying all of the parameters simultaneously have not been carried out, since the operators of wastewater treatment plants have limited capability of controlling all of these parameters at the same time. Emission of N_2O was plotted on a secondary axis, because it was, in all cases, higher than CO_2 and CH_4 emissions by at least one order of magnitude.

3.2.1 Effect of hydraulic residence time

Applying a higher reactor volume proved advantageous for increasing the amount of Anammox biomass, lowering the effluent NH_4-N and NO_2-N concentrations. Results of the direct GHG emission estimations – at different residence times – are illustrated on Fig. 2. N_2O production was shown to rise as a result of more intensive Anammox activity, however, the emission appears to stop increasing above a HRT of 17 hours. According to the simulations, altering the residence time apparently has no effect on CH_4 and CO_2 emissions. Thus selecting a higher Anammox reactor volume is rather to be considered economically than in the terms of GHG emissions.

3.2.2 Effect of reactor temperature

Raising the temperature of the water phase was also shown to be beneficial for Anammox bacteria, but disadvantageous for nitrifying biomass. Based on the modelling, the unit needs to be heated to at least 30℃ to provide operational conditions for Anammox metabolism. The effects of centrate temperature on GHG productions are summarized by Fig. 3.

Though higher temperatures provide better nitrogen removal, a gradually higher amount of N_2O is released. CO_2 and CH_4 emissions increase, too, on a smaller scale, due to anaerobic degradation processes promoted by higher temperatures. Considering GHG production, and from a financial point of view, it is safer to operate Anammox reactors at lower temperatures.

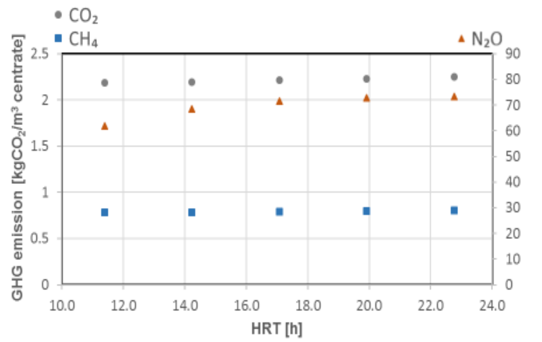

Fig. 2. Direct GHG emissions of the IFAS Anammox process as a function of HRT

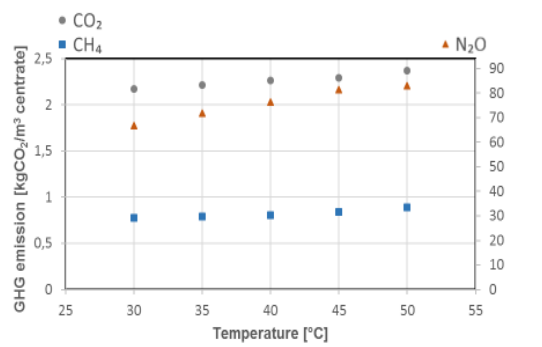

Fig. 3. Direct GHG emissions of the IFAS Anammox process at altered temperatures

3.2.3 Effect of dissolved oxygen

Raising the dissolved oxygen level minimises oxygen-limited zones in the reactor, and helps avoid the autotrophic denitrification process by nitrifiers, that produces N_2O as they consume NO_2-N. As shown by Fig. 4, a large amount of N_2O emission can be spared by increasing DO levels, though this does not sensibly affect nitrogen removal.

Moreover, DO levels above this range can influence Anammox microbes by diffusion into the biofilm, risking the ammonia oxidizers outcompeting them, the accumulation of NO_2-N and worse treatment efficiency. Based on the model, a DO level of at least 0.2 mg/L is needed for suitable nitritation.

3.2.4 Effect of filling ratio

Larger media surfaces intensify biofilm activity, promoting nitrogen removal and – compared to other operational variables – moderately raising N_2O production, as seen on Fig. 5. Increasing the filling grade is beneficial for Anammox organisms, that generally reside in the biofilm; thus selecting a high enough surface area is essential for stable operation of the single-stage process, to remove both nitrite-N and ammonia-N sufficiently. Applying a higher filing ratio virtually does not influence the removal of organic carbon, having no apparent effect on the other two examined GHG emissions.

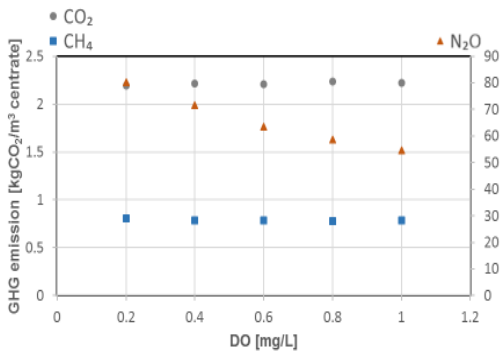

Fig. 4. Direct GHG emissions of the IFAS Anammox process at different DO levels

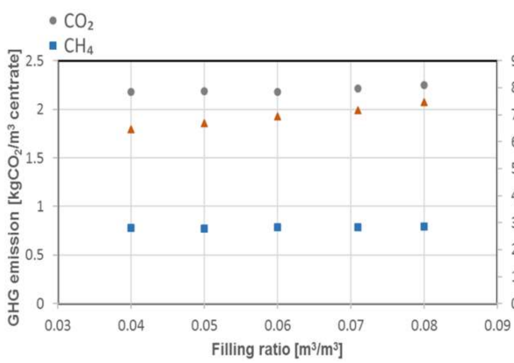

Fig. 5. Direct GHG emissions of the IFAS Anammox process at different filling grades

3.2.5 Effect of mixed liquor suspended solids

Fig. 6 shows GHG emissions estimated by modifying the recycled activated sludge flow.

Increasing the RAS flow mainly promotes nitrification in the suspended phase. The process can be operated quasi as a pure biofilm reactor without sludge retention, with an MLSS concentration of approximately 1400 mg/L. The introduction of recycled sludge provides a steep increase in NH_4-N removal, but also sharply increases N_2O production – as it can be seen on Fig. 6. As higher MLSS concentrations lower the oxygen transfer efficiency, more oxygen limited zones are present, also contributing to the increase in nitrous oxide emission.

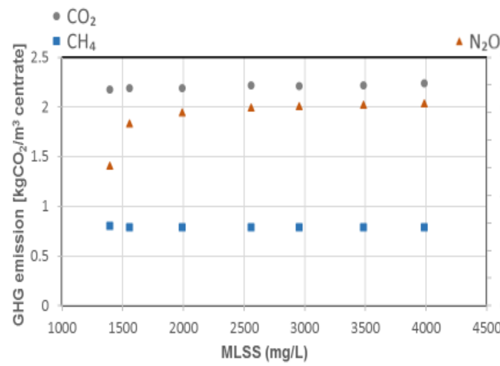

Fig. 6. Direct GHG emissions of the IFAS Anammox process as a function of MLSS

4. CONCLUSION

Regarding the operation of an integrated fixed-film Anammox process, modelled indirect GHG emissions are negligible compared to biology related emissions. Emissions of N_2O are the most considerable, and suggested to be minimized by applying a sensibly low MLSS, and high enough DO concentration to evade oxygen limitation and autotrophic denitrification. Reactor temperature within 30-35℃ also keeps the production of N_2O at a lower level.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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