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Modelling the N_2O emissions in municipal wastewater treatment plants under dynamic conditions

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Abstract

Nitrous oxide (N_2O), a greenhouse gas with a significant global warming potential, can be produced during the biological nutrient removal in wastewater treatment plants (WWTPs). N_2O modelling under dynamic conditions is of vital importance for its mitigation. Following the activated sludge models (ASM) layout, an ASM-type model was developed considering three biological N_2O production pathways for a municipal anaerobic/anoxic/aerobic (A^2/O) WWTP performing chemical oxygen demand, nitrogen and phosphorus removal. Precisely, the N_2O production pathways included were: nitrifier denitrification, hydroxylamine oxidation, and heterotrophic denitrification, with the first two linked to the ammonia oxidizing bacteria (AOB) activity. A stripping effectivity (SE) factor was used to mark the non-ideality of the stripping modelling. With the dissolved oxygen (DO) in the aerobic compartment ranging from 1.8 to 2.5 mg L^{-1}, partial nitrification and high N_2O production via nitrifier denitrification occurred. Therefore, low aeration strategies can effectively lead to a low overall carbon footprint only if complete nitrification is guaranteed. After suddenly increasing the influent ammonium load, the AOB had a greater growth compared to the NOB. N_2O hotspot was again nitrifier denitrification. Especially under concurring partial nitrification and high stripping (i.e. combination of low DO and high SEs), the highest N_2O emission factors were noted.

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1 Introduction

Emissions of greenhouse gases (GHG) occur in various treatment stages during the biological nutrient removal (BNR) within wastewater treatment plants (WWTPs). Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are amongst these emissions (Massara, Solis, Guisasola, & Baeza, 2017). N_2O is primarily significant due to its significant global warming potential (GWP). Precisely, N_2O has a GWP 265 times higher than CO_2, in contrast to the GWP of CH_4 that is 28 times higher than the CO_2 respective one (IPCC, 2013). In this concept, the development of novel, cost-effective and flexible tools enabling the detection of GHG emissions in real time and their connection with a specific plant activity will facilitate the design of effective mitigation strategies.

The International Water Association (IWA) Activated Sludge Models (ASM) (Henze, Gujer, Mino, & van Loosdrecht, 2000) have constituted a popular mathematical tool for the description of chemical oxygen demand (COD), nitrogen (N) and phosphorus (P) removal during the BNR. Nevertheless, these models do not take account of the N_2O production and quantification.

The microbial pathways for N_2O production during the BNR in WWTPs are activated via the biochemical processes of nitrification and denitrification. The nitrification-related ones occur through the ammonia oxidizing bacteria (AOB) activity (i.e. nitrifier denitrification and hydroxylamine (NH_2OH) oxidation). Moreover, N_2O is an intermediate product of heterotrophic denitrification, which is listed as third biological pathway (Rodriguez-Caballero, Aymerich, Marques, Poch, & Pijuan, 2015; Wunderlin, Mohn, Joss, Emmenegger, & Siegrist, 2012; Wunderlin, et al., 2013; Ni & Yuan, 2015). The major parameters fostering the N_2O production have been summarized as follows: insufficient dissolved oxygen (DO) while nitrification is happening, increased nitrite (NO_2^-) levels during both nitrification and denitrification, and low COD/N during denitrification (Kampschreur, Temmink, Kleerebezem, Jetten, & van Loosdrecht, 2009; Desloover, Vlaeminck, Clauwaert, Verstraete, & Boon, 2012; Massara, et al., 2017).

Hence, this work focused on the development of an ASM-type model that considers N_2O production in WWTPs in the most holistic way. Thus, the suggested model included N and P removal, the three biological pathways for N_2O production/consumption, N_2O stripping, and N_2O emission factor (EF) estimation under dynamic conditions (e.g. changing DO levels).

2 Materials and Methods

The model was indicatively developed for a municipal WWTP with an anaerobic-anoxic-aerobic (A^2/O) configuration. The influent composition was typical of the municipal Manresa WWTP (Catalonia, Spain) (Machado, Lafuente, & Baeza, 2014). The model structure followed the IWA ASM2d principles (Henze, Gujer, Mino, & van Loosdrecht, 2000), while the microbial N_2O production pathways were described by extending and adapting relevant past studies (Pocquet, Wu, Queinnec, & Spérandio, 2016; Hiatt & Grady, 2008). Steady-state was simulated by applying constant influent composition for a period of 200 d. All the kinetic parameter values were normalized for 20 ºC from the ASM2d section of Henze et al. (Henze, Gujer, Mino, & van Loosdrecht, 2000). The N_2O EF was calculated in three ways: i) $N_2O\text{-}EF_{TOTAL}$: considering both the stripped N_2O and the effluent N_2O (i.e. our most conservative approach), ii) $N_2O\text{-}EF_{GAS}$: resulting only from the N_2O stripping, and iii) $N_2O\text{-}EF_{EF}$: considering exclusively the N_2O released in the effluent. The N_2O stripping modelling involved the k_La_{N2O} (i.e. the volumetric mass transfer coefficient for N_2O), as well as a factor in the range 0-1 expressing the non-ideality of the stripping modelling (i.e. stripping effectivity: SE).

3 Results and Discussion

3.1 DO effect on nitrification and N_2O emissions

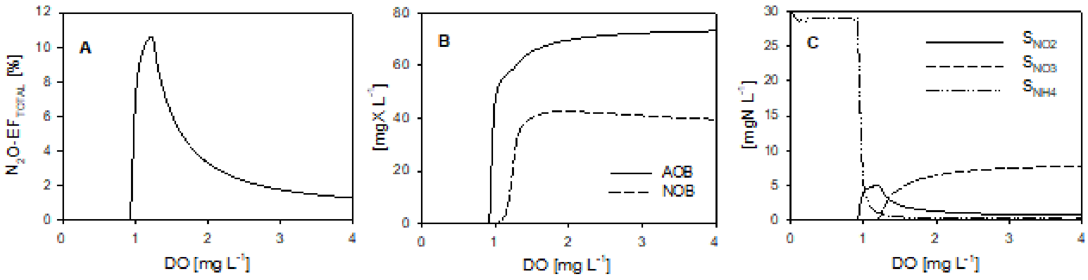


Figure 1: DO effect in the aerobic reactor on the steady state values of (A) N_2O emission factor, (B) AOB and NOB concentration, and (C) NO_2^-, NO_3^- and NH_4^+ concentrations. SE was 1.

DO ranging from 0 to 4 mg L^{-1} in the aerobic reactor was simulated to examine the impact on nitrification and N_2O emissions. Fig. 1B and 1C show that neither AOB/nitrite oxidizing bacteria (NOB) growth, nor NO_2^-/nitrate (NO_3^-) production occurred under low DO (i.e. DO<0.8 mg L^{-1}). The DO increase from 0.8 mg L^{-1} onwards benefited the AOB growth. NOB growth started only as soon as DO rose around 1.1 mg L^{-1} (Fig. 1B). These values (i.e. 0.8 and 1.1 mg L^{-1}) are mainly linked to oxygen affinity constants. The NOB have a lower oxygen affinity constant compared to the AOB (Wiesmann, 1994). Thus, partial nitrification/nitritation (i.e. ammonium (NH_4^+) oxidation to NO_2^-) strategies are designed upon the choice of a suitable DO setpoint (Guisasola, Marcelino, Lemaire, Baeza, & Yuan, 2010). In line with this, the AOB prevailed over the NOB under relatively low DO (i.e. 0.8<DO<1.1 mg L^{-1}) (Fig. 1B). In parallel, the NH_4^+ concentration decreased, while NO_2^- started increasing. The latter can be viewed as a sign of nitritation causing NO_2^- accumulation (Fig. 1C). In this DO range (i.e. 0.8-1.1 mg L^{-1}), the N_2O EF significantly increased up to almost 10.5% (Fig. 1A). Under these oxygen-limiting conditions, the dominant N_2O production pathway is expected to be nitrifier denitrification. NO_2^- replaces oxygen at the role of the final electron acceptor and, hence, the AOB perform nitrifier denitrification (Desloover, Vlaeminck, Clauwaert, Verstraete, & Boon, 2012; Tallec, Garnier, Billen, & Gousailles, 2006; Kampschreur, et al., 2008). At a DO of 1.5 mg L^{-1}, AOB and NOB were stabilized around 70 mg L^{-1} and 40 mg L^{-1}, respectively (Fig. 1B). (Full) nitrification began, thus leading to a continuously decreasing NO_2^- accumulation and a gradual deactivation of the nitrifier denitrification pathway. This can be seen in Fig. 1A by the N_2O-EF_{TOTAL} decrease that starts at a DO≈1.5 mg L^{-1} and persists with the further DO increase. Moreover, NO_3^- production was noted; this can be attributed to the occurrence of full nitrification (Fig. 1C). At high DO (i.e. >3 mg L^{-1}), the N_2O EF was importantly lower (i.e. <2%). Applying high DO (i.e. >3 mg L^{-1}) to mitigate N_2O emissions can be effective, although quite energy-consuming. Optimizing a WWTP's operation requires the testing of various DO intervals inside which both full nitrification and moderate energy consumption are guaranteed; this can be between 1.8 and 2.5 mg L^{-1} for the current study.

3.2 Stripping effectivity (SE) impact on the N_2O emission factor (EF)

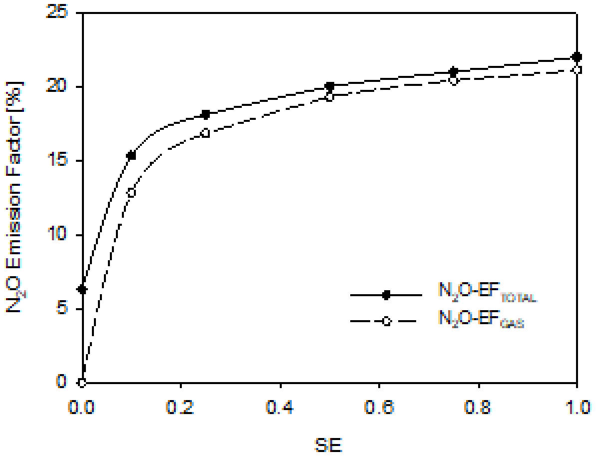
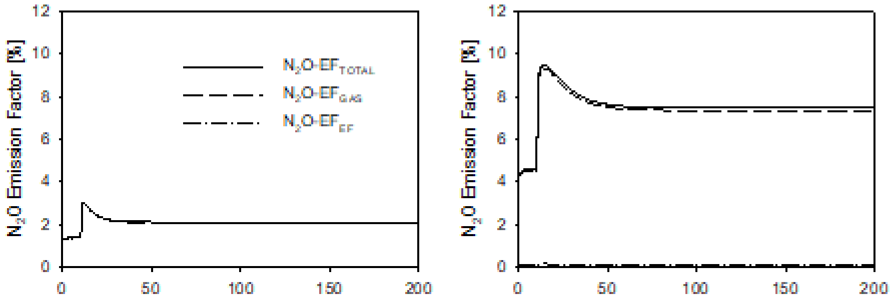


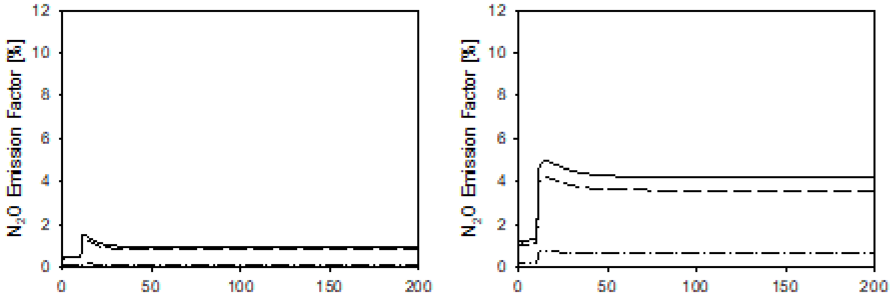
Figure 2: The maximum N_2O EF (N_2O-EF_{TOTAL}: considering both the N_2O stripped and the N_2O released in the effluent; N_2O-EF_{GAS}: referring exclusively to the N_2O stripping contribution) noted for different SE values (0, 0.1, 0.25, 0.5, 0.75, 1). Influent NH_4^+ was 40 mg L^{-1}.

Although N_2O can be an intermediate of heterotrophic denitrification, the aerobic compartments in WWTPs (where nitrification occurs) are considered as the major N_2O hotpots. Aided by aeration, the produced N_2O is stripped and emitted to the atmosphere (Law, Ye, Pan, & Yuan, 2012; Mannina, et al., 2016). The combined effect of different DO setpoints under the highest influent NH_4^+ value tested in this study (i.e. 40 mg L^{-1}) on the N_2O EF under different SEs (i.e. 0, 0.1, 0.25, 0.5, 0.75, 1) was examined. The results are shown in Fig. 2 for the N_2O-EF_{TOTAL} (i.e. EF considering both the stripped N_2O and the N_2O released in the effluent) as well as for the N_2O-EF_{GAS} (i.e. EF considering only the stripping contribution). Similar general trends were always noted: the maximum N_2O EF was observed at a DO≈1.2 mg L^{-1}. However, the maximum absolute values were different. The maximum N_2O-EF_{GAS} ranged from 0% (SE=0) to ~21.1% (SE=1), whereas the maximum N_2O-EF_{TOTAL} from 6.3% (SE=0) to ~22% (SE=1). Thus, it can be deduced that the SE increase generally led to higher EFs (Fig. 2). The observed trend can be explained through the fact that lower SEs render the activation of the heterotrophic denitrification pathway more possible. Hence, N_2O will be rather consumed (via denitrification) than stripped. The N_2O-EF_{TOTAL} was always reported to be higher than the respective N_2O-EF_{GAS} one, although not significantly (Fig. 2). Therefore, it can be alleged that the N_2O stripping majorly contributed to the N_2O EF estimation. More importantly, the results of this study indicated that the SE factor majorly affected the final EF estimation. In this context, a more detailed striping modelling approach is needed in the future, to avoid similar simplifications and potential EF overestimations.

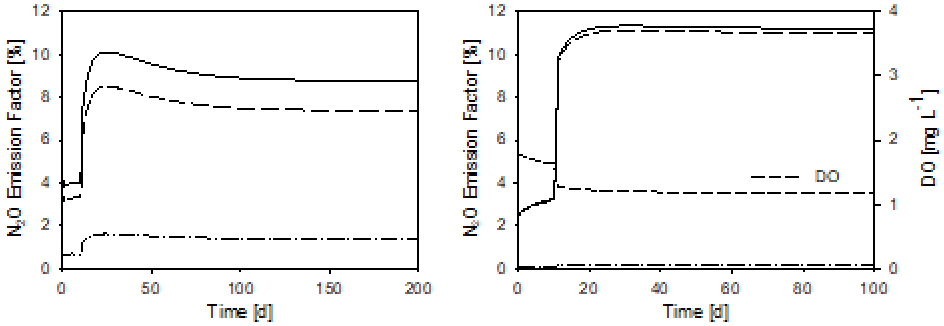
3.3 Disturbing the normal WWTP operation: impact on the N_2O emission factor (EF)



a. SE = 1 & DO setpoint = 3 mg L^{-1} b. SE = 1 & DO setpoint = 1.5 mg L^{-1}



c. SE = 0.1 & DO setpoint = 3 mg L^{-1} d. SE = 0.1 & DO setpoint = 1.5 mg L^{-1}



e. SE = 0.1 & DO setpoint = 1.2 mg L^{-1} f. SE = 1 & no DO control

Figure 3: The effect of increasing the influent NH_4^+ concentration (from 20 to 30 mg L^{-1}) on the 10th day of the plant operation on the N_2O EF. Different SE values (1 and 0.1) and DO setpoints (3 mg L^{-1}, 1.5 mg L^{-1}, 1.2 mg L^{-1} and no DO control) were tested.

Transition from a system shock to normal WWTP operation creates an environment favorable to intermediates accumulation and, thus, prone to higher N_2O emissions. Within this context, the effect of a 'step' increase (from 20 to 30 mg L^{-1}) of the influent NH_4^+ concentration on the 10th day of the plant operation was examined for various scenarios with different combinations of SEs and DO setpoints in the aerobic reactor. Scenarios a and b: SE was 1, thus allowing us to observe the full stripping effect. The abrupt influent NH_4^+ increase caused a sudden increase in the N_2O emissions. The N_2O-EF$_{TOTAL}$ ranged as follows: 1.4→3.1% almost up to the 12th day of operation (scenario a) and 4.5→9.6% until the 17th day (scenario b). Then, the EF reduction started until it was stabilized at lower levels: at ~2.1% after the 30th day (scenario a), and at ~7.5% after the 40th day (scenario b) (Fig. 3). DO in case b was significantly lower than in scenario a; thus, higher EF values were expected. Under such conditions, the AOB perform nitritation, use NO_2^- as terminal electron acceptor and, finally, produce N_2O (nitrifier denitrification pathway) (Kuenen, 2008; Peng, Ni, Ye, & Yuan, 2015; Jin, Wang, & Zhang, 2016). In both scenarios a and b, the decreasing N_2O EF points to the fact that the NOB started growing and oxidizing NO_2^-. Nevertheless, the final N_2O EF never returned to its initial value; it is possible that the NOB were not enough to oxidize all the NO_2^- produced. Under the same DO values, different SEs were applied to examine the effect of different levels of stripping (comparison between scenarios a and c, and comparison between scenarios b and d in Fig. 3). Decreasing the SE (from 1 to 0.1) explains the distance between the $N_2O\text{-}EF_{TOTAL}$ and $N_2O\text{-}EF_{GAS}$ lines. Less N_2O emissions were noted in the SE=0.1 cases (Fig. 3c and 3d); more N_2O was released in the effluent. Lowering the SE value (i.e. 0.1) decreases the stripping importance. Hence, it promotes the existence of a higher N_2O concentration inside the aerobic reactor, and the recycling of more N_2O to the anoxic reactor (where N_2O can be consumed through denitrification). Scenario e investigated the effect of DO conditions clearly harmful to the NOB growth: DO setpoint of 1.2 mg L^{-1} and SE =0.1. N_2O emissions>9% were reported due to partial nitrification and NOB washout. Finally, scenario f indicated how the sudden influent NH_4^+ increase effect can be more important and evident if DO is not controlled. A higher NH_4^+ load decreases the DO concentration. Consequently, the system can shift from full to partial nitrification; the latter can explain the higher EFs noted.

3.4 Sensitivity Analysis (SA)

A local SA was performed to define the model (kinetic, stoichiometric) parameters that are mostly sensitive to the $N_2O\text{-}EF_{TOTAL}$ at steady state. The central difference method was applied to calculate the sensitivity for each parameter. Different perturbation factors were tested within the 0.01-10% range to ensure no interference in the parameter ranking (Reichert & Vanrolleghem, 2001). As discussed in sections 3.1 and 3.3, different DO values in the aerobic reactor (i.e. varying from 1 to 4 mg L^{-1}), resulted in different EFs. In the context of understanding the cause of high N_2O emissions, the SA was conducted under two different steady-state scenarios: the 1st for a high DO setpoint in the aerobic reactor (i.e. 3 mg L^{-1}), and the 2nd for a low DO setpoint equal to 1 mg L^{-1}. The influent NH_4^+ concentration was fixed at 30 mg L^{-1} and the SE at 0.5. Table 1 shows the 20 most sensitive parameters to the $N_2O\text{-}EF_{TOTAL}$ for the two applied scenarios. The values are listed in descending order considering the absolute sensitivity values. A positive sensitivity index indicates that an increase in the parameter results in increasing the $N_2O\text{-}EF_{TOTAL}$, while a negative sensitivity suggests the opposite. The results showed in Table 1 were obtained with a perturbation factor of 0.01% based on the study by De Pauw (De Pauw, 2005) who presented it as a factor producing equal derivative values for forward and backward differences.

The most sensitive parameters to the $N_2O\text{-}EF_{TOTAL}$ factor varied under the two different operational modes. For the DO setpoint of 3 mg L^{-1}, the most sensitive parameters were those related to the NOB, followed by those referring to the AOB and, finally, by those related to the phosphorus accumulating organisms (PAOs). The sensitivity results for the NOB-related parameters enhance the understanding of the NO_2^- dynamics. The NO_2^- accumulation will affect the total N_2O emission factor through the activation/deactivation of the nitrifier denitrification pathway, as discussed in sections 3.1 and 3.3. Under the DO setpoint of 1 mg L^{-1} (i.e. 2nd scenario), the AOB parameters were the most sensitive since limited NOB growth occurs in a low-DO environment (Fig. 1B). Thus, the NOB-related parameters became insensitive. For such a DO setpoint, the WWTP model performs nitritation and increased N_2O production through nitrifier denitrification is expected (section 3.1).

However, it was noted that the SE appeared only in the 17th and 20th place for the DO setpoints of 3 and 1 mg L^{-1}, respectively. The reference value of this parameter (0.5) explains the sensitivity results. According to Fig. 2, the SE parameter has a severe effect on the $N_2O\text{-}EF_{TOTAL}$ while increasing from 0 to 0.25; its further increase from 0.25 to 1 has a lower impact on the N_2O-EF values. If a lower value had been assigned to this parameter (i.e. between 0 and 0.25), its relative sensitivity would have increased. Furthermore, Table 1 was re-examined to detect potential common parameters that appeared in the first ten places for both scenarios. It was observed that n_G (anoxic growth factor), $q_{AOB\_N2O\_ND}$ (maximum N_2O production rate by the nitrifier denitrification pathway), Y_{PAO} (yield coefficient for the PAOs) and Y_H (yield coefficient for the heterotrophs) were amongst the first ten parameters for both DO setpoints; all with positive sensitivity. Hence, it can be assumed that decreasing these values leads to a decrease in the $N_2O\text{-}EF_{TOTAL}$. The n_G, Y_{PAO} and Y_H stoichiometric parameters, in specific, are included in the stoichiometry of the processes related to the anoxic growth of heterotrophs and PAOs. These processes can indeed significantly influence the EF since they occur in an anoxic environment where N_2O can be consumed through denitrification. Finally, the impact of the $q_{AOB\_N2O\_ND}$ kinetic parameter was found to be important for both scenarios. Considering that $q_{AOB\_N2O\_ND}$ expresses the N_2O production rate through nitrifier denitrification, this observation strengthens the view that nitrifier denitrification is possibly the most influential N_2O production pathway.

Table 1: Sensitivity analysis results for the two different scenarios (1st: DO_{AE} =3 mg L^{-1}; 2nd: DO_{AE} =1 mg L^{-1}); both with influent NH_4^+ concentration=30 mg L^{-1} and SE=0.5. DO_{AE} stands for the DO control setpoint in the aerobic reactor.

Order	**DO_{AE}=3 mg L^{-1}**		**DO_{AE}=1 mg L^{-1}**	
	Parameter	**Sensitivity**	**Parameter**	**Sensitivity**
1	Y_{NOB}	-2.138	Y_{AOB}	2.233
2	n_G	1.489	n_G	1.978
3	b_{NOB}	1.059	q_{AOB_AMO}	1.407
4	$q_{AOB\_N2O\_ND}$	0.997	Y_{PAO}	1.108
5	q_{AOB_HAO}	-0.926	b_{AOB}	-1.024
6	$K_{I\_O2\_AOB}$	0.878	n_{G5}	-0.947
7	Y_{AOB}	0.863	K_{OH5}	-0.853
8	K_{HNO2_AOB}	-0.857	$q_{AOB\_N2O\_ND}$	0.841
9	K_{NO2_NOB}	0.851	K_{O2_AOB1}	-0.738
10	Y_{PAO}	0.739	i_{NXS}	0.674
11	K_{O2_NOB}	0.629	Y_H	-0.470
12	n_{G5}	-0.620	Y_{PO4}	-0.435
13	K_{OH5}	-0.470	q_{PP}	0.400
14	K_{N2O_Den}	0.435	Y_{PAO}	-0.386
15	i_{NXS}	0.428	i_{NBM}	-0.375
16	b_{PAO}	-0.408	K_{HNO2_AOB}	-0.360
17	SE	0.375	i_{NSF}	0.338
18	Y_H	-0.364	$K_{I\_O2\_AOB}$	0.299
19	K_{MAX_P}	0.259	K_{MAX_P}	0.292

Order	**DO_{AE}=3 mg L^{-1}**		**DO_{AE}=1 mg L^{-1}**	
	Parameter	**Sensitivity**	**Parameter**	**Sensitivity**
20	i_{NBM}	-0.247	SE	0.223

4 Conclusions

An ASM-N_2O model considering COD, N and P removal, in addition to all the microbial N_2O production pathways was developed for a municipal WWTP, with emphasis on the estimation of the N_2O EF. Main conclusions:

- With the DO in the aerobic compartment ranging from 0.8 to 1.8 mg L^{-1}, high AOB growth was reported. The system moved from full to partial nitrification, thus promoting N_2O production through nitrifier denitrification. Considering the significant N_2O GWP, such operational conditions can lead to a high overall WWTP carbon footprint. Consequently, low aeration strategies can succeed only if nitrification is not disturbed.
- A SE factor (ranging from 0 to 1) was used to describe the non-ideality of the stripping modelling. Decreasing the SE meant higher N_2O concentration in the mixed liquor. The latter was translated into a higher N_2O denitrification rate inducing, subsequently, lower emissions (because of the N_2O consumption via denitrification).
- The impact of a sudden 'step' increase (from 20 to 30 mg L^{-1}) in the influent NH_4^+ on the 10th day of the plant operation was studied. AOB prevailing over the NOB enhanced the NO_2^- accumulation and activated the nitrifier denitrification pathway. Higher emissions occurred under the following conditions: lower DO setpoints (i.e. environment more favorable to nitrifier denitrification) combined with higher SE values (i.e. higher stripping significance).
- Given the limited NOB growth under low-DO conditions (i.e. 1 mg L^{-1}), the SA showed that the NOB-related parameters had minor influence over the N_2O-EF_{TOTAL}. The n_G, $q_{AOB\_N2O\_ND}$, Y_{PAO} and Y_H parameters were amongst the top ten for both DO setpoints tested (i.e. 3 and 1 mg L^{-1}). n_G, Y_{PAO} and Y_H are related to the N_2O consumption through denitrification. $q_{AOB\_N2O\_ND}$ indicates that nitrifier denitrification is possibly the most important pathway to consider for the N_2O mitigation.

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