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A novel fuzzy-logic control strategy minimizing N₂O emissions

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HIGHLIGHTS

- A fuzzy-logic control strategy for low N₂O emissions is systematically developed,
- The control strategy is simulated for different N₂O formation mechanisms,
- Effluent quality and operation costs are considered while reducing N₂O emissions,
- The control strategy is found to reduce the N₂O produced by AOB denitrification.

ABSTRACT

A novel control strategy for achieving low N₂O emissions and low effluent NH₄⁺ concentration is here proposed. The control strategy uses the measurements of ammonium and nitrate concentrations in inlet and outlet of the aerobic zone of a wastewater treatment plant to calculate a ratio indicating the balance among the microbial groups. More specifically, the ratio will indicate if there is a complete nitrification. In case nitrification is not complete, the controller will adjust the aeration level of the plant in order to inhibit the production of N₂O from AOB and HB denitrification. The controller was implemented using the fuzzy logic approach. It was comprehensively tested for different model structures and different sets of model parameters with regards to its ability of mitigating N₂O emissions for future applications in real wastewater treatment plants. It is concluded that the control strategy is useful for those plants having AOB denitrification

as the main N_2O producing process. However, in treatment plants having incomplete NH_2OH oxidation as the main N_2O producing pathway, a cascade controller configuration adapting the oxygen supply to respect only the effluent ammonium concentration limits was found to be more effective to ensure low N_2O emissions.

Keywords: nitrous oxide, fuzzy-logic, wastewater, control, nitrogen.

List of abbreviations and symbols

AEC	Average aeration energy consumption
AOB	Ammonia-oxidizing bacteria
ASMG1	Activated Sludge Model for Greenhouse gases N°1
BSM2N	Benchmark Simulation Model n°2 for Nitrous oxide
DO_{SP}	Set point for dissolved oxygen concentration
EQI	Effluent Quality Index
FLC	Fuzzy-logic control
η_{TKN}	TKN removal efficiency
η_{TN}	TN removal efficiency
HB	Heterotrophic bacteria
IAE	Integral Absolute Error
ISE	Integral Square Error
k_La	Oxygen mass transfer coefficient
$K_{O_2,AOB}$	Oxygen affinity constant for aerobic AOB activity
$K_{O_2,NOB}$	Oxygen affinity constant for NOB activity
NOB	Nitrite-oxidizing bacteria
$N_{org,inAER}$	Organic Nitrogen in the influent to the aerobic zone
PID	Proportional Integrative Derivative
Q	Volumetric flow rate
RON	Oxygen-to-TKN loading ratio
SF	Scaling Factor

S_I	Soluble inert organic matter
S_S	Readily biodegradable organic matter
T	Temperature
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
TV	Total Variation
X_I	Particulate inert organic matter
X_P	Particulate products from biomass decay
X_S	Particulate biodegradable organic matter
WWTP	Wastewater treatment plant

1. Introduction

On-line process control is a tool widely used to automatically manipulate operating conditions of a process in function of measured disturbances with the aim of achieving predefined process performance targets. In wastewater treatment plants (WWTPs), several objectives need to be satisfied. The legal effluent discharge limits and the minimization of the operational costs are some of the most important objectives. There is however an emerging objective which needs to be tackled through the use of on-line process control, i.e. the minimization of emissions of nitrous oxide (N_2O). It has in fact been found that the amount of greenhouse gases emitted from WWTPs has increased constantly over the last decades (Fischedick et al., 2014) and, in order to limit the carbon footprint of WWTPs, N_2O emissions need to be minimized. There are three biological pathways according to which N_2O is produced in domestic WWTPs: (a) denitrification by ammonia-oxidizing bacteria (AOB), where AOB use nitrites (NO_2^-) and nitric oxides (NO) as electron acceptors for the oxidation of hydroxylamine (NH_2OH), an ammonium-to-nitrite oxidation intermediate; (b) incomplete hydroxylamine oxidation, a process originating from the accumulation of hydroxylamine oxidation intermediates such as nitroxyl radicals or NO; and (c) denitrification by heterotrophs (HB), where N_2O is produced as intermediate of nitrate (NO_3^-) and NO_2^- reduction into dinitrogen gas (N_2) for the oxidation of organic matter.

Several full-scale WWTP experiences have been reported in the literature - such as Foley et al. (2010), Daelman et al. (2015), Aboobakar et al. (2013), Ahn et al. (2010) and Stenström et al. (2014) - having identified AOB denitrification as the main pathway producing most of the N_2O emitted. These results were extensively discussed in Boiocchi et al. (2017). On the other hand, studies such as Ni et al. (2013) and Tumendelger et al. (2014) reported the incomplete NH_2OH oxidation pathway as the main N_2O producer, though pointing out as well that AOB denitrification should not be excluded as potential N_2O producing pathway in other contexts. These differing findings suggest that specific WWTP design (SRT, configuration, etc...), operation (such as aeration strategy) and influent factors (nitrogen loading, temperature, etc...) promotes different N_2O production mechanisms. Although both AOB mechanisms should be considered, AOB denitrification is here chosen as the main process to be slowed down through the control strategy for N_2O emission minimization. This is because relatively more WWTPs have been reported in literature (see e.g. the surveys by Foley et al. (2010) and Ahn et al. (2010)) suggesting the AOB denitrification process as the dominant N_2O producer.

Microbiological and experimental experiences have demonstrated that AOB denitrification is triggered by low availability of dissolved oxygen and high availability of nitrites (Peng et al., 2014, 2015). More specifically, low availability of oxygen forces AOB to use nitrogen oxides as alternative electron acceptors. For example, the experiment by Sun et al. (2014) showed that ensuring oxygen availability in a full-scale WWTP to achieve complete nitrification, i.e. complete conversion of ammonium into nitrate, is the key for minimizing N_2O emissions. However, the N_2O minimizing effect of oxygen is only observed until a certain threshold, above which supplying more oxygen does not further reduce the N_2O production and emissions. On the contrary, excessive oxygen supply may trigger HB-produced nitrite accumulation in the anoxic zone and slow down the HB-mediated N_2O reduction into N_2 . Therefore, it is important to accomplish an optimal oxygen level through automatic regulation of the aeration regime with the aim of reducing the N_2O emissions, which constitutes the objective for this work.

For this reason, the present study focuses on developing an on-line control strategy establishing the optimal oxygen availability in the aerated zone with the aim of preventing N_2O formation in continuously-aerated WWT systems. The control strategy is based on the following principle identified through a comprehensive sensitivity analysis by Boiocchi et al. (2017): (i) the controlled variable is the measured ratio between nitrate

generated and ammonium removed in an aerobic zone, indicating the microbiological imbalance between the different microorganisms, (ii) aeration supply is the variable ultimately manipulated in order to maintain a desired balance among microorganisms.

Several control techniques such as model-predictive (MP), Proportional-Integral-Derivative (PID) and fuzzy-logic (FL), can be used to implement the above control principle. While MP control in theory can provide optimal control actions, its application to the N_2O control problem in full-scale plants is currently not very practical since the present N_2O models are too uncertain to be used for control (Behera et al., 2015). On the other hand, the PID approach is not suitable to deal with multiple control objectives and the highly non-linear dynamic behaviour typical of WWT processes. While the fuzzy-logic (FL) technique has many parameters to be tuned, it provides a flexible tool that can incorporate both the process non-linearity and the multiple objectives of WWT systems (Cosenza et al., 2014). This is why the fuzzy-logic approach is adopted in this study during the control design.

Once designed, the control strategy performance is implemented and tested on three different modified BSM2 simulation platforms describing three different N_2O dynamics. To address the benefits of the novel control strategy to WWTPs, the performance is benchmarked against a typical aeration cascade control technique for the maintenance of low effluent ammonium concentration. The performance of the FL controller is further checked against: a) the sensor and actuator noise commonly occurring in WWTPs, and b) model parameter uncertainties.

The manuscript is structured as follows: in Section 2 the design of the control strategy is described, Section 3 shows the implementation and testing of the control strategy, and the results obtained thereby are presented in Section 4. Conclusions and future perspectives are provided in Section 5.

2. Design of the control strategy

Following the systematic development of the membership functions used by Boiocchi et al. (2016), the approach to develop the fuzzy-logic control strategy involved the following workflow:

- I) Specification of the optimization problem,
- II) Identification of the critical points,

III) Definition of the membership functions for input and output variables,

IV) Definition of additional design parameters.

Each of these workflow steps is explained briefly in the next sections.

2.1. Specification of the optimization problem

This task is split up into the following sub-steps:

- Definition of the control objectives,
- Definition of the control structure,
- Definition of the physical constraints,
- Identification of the key scenarios leading the system away from optimality,
- Identification of the relationship between optimization variables and controlled variables.

Definition of the control objectives

In contrast to what has been done for the controller developed by Boiocchi et al. (2016), the objectives for N_2O emissions cannot be expressed numerically because – differently from the TN removal efficiency – the minimum amount of N_2O possibly emitted varies quite a lot from one plant to another according to many operating and design parameters. Instead, the control objectives are here defined qualitatively as follows: “the controller has to establish those generic environmental conditions which give the least N_2O production and subsequent emission suited for the particular WWTP, while respecting legal effluent NH_4^+ limits”. Thus the strategy can potentially be applied for a large number of WWTPs, without the need for strong adaptations to specific cases. These control objectives will be mathematically translated on the basis of the trends of the steady-state N_2O emission factors and of the effluent ammonium concentration with respect to the oxygen-to-TKN loading ratio (RON) presented in Figure 1. RON represents the ratio between oxygen supplied and TKN in the influent to the aerated zone of the activated sludge unit of a WWTP, as explained in more detail in Boiocchi et al. (2017) . These simulations were performed with the Benchmark Simulation Model n°2 (BSM2) by Jeppsson et al. (2007) extended with the Activate Sludge Model for Greenhouse gases n°1

(ASMG1) by Guo and Vanrolleghem (2014). This model includes only AOB denitrification as the AOB-mediated N_2O producing pathway in line with our objective for the controller development (see introduction). Since the controller is based on the assumption that an effective minimization of N_2O emissions can be achieved by slowing down AOB denitrification, this would enable a better identification of the design parameters of the control technology acting on this process. The testing of the resulting controller will show how this assumption adopted for the control design affects the performance of the controller. For those plants having incomplete NH_2OH oxidation as main N_2O producing pathway, the sensitivity analysis should use the model by Ni *et al.* (2013).

Figure 1 summarizes the so-called “best and worst system operations” identified. The best system operation is found when the minimal N_2O emission factor is achieved and the effluent ammonium is below the typical legal limit of 4 mg N.L^{-1} . As can be seen, at RON equal to zero, the N_2O emission factor is zero. However, this scenario is not considered desirable since the conversion of ammonium is zero as well. On the other hand, for values of RON equal or higher than $5.2 \text{ g (-COD).g}^{-1} \text{ TKN}$, N_2O is minimized while at the same time the highest ammonium conversion occurs. However, N_2O production by HB in the anoxic zone sensibly increases for a load of $5.2 \text{ g (-COD).g}^{-1} \text{ TKN}$, as described in Boiocchi (2016). Although the contribution of HB from the anoxic zone to N_2O emissions is almost negligible according to the model used, it is still relevant to avoid this scenario due to the uncertainty in the model parameters. As a matter of fact, it can be that for some plants this contribution will emerge to be more relevant than predicted. In view of this, the best system operation is identified for RON equal to 5.2. With regard to the “worst system operation”, this is identified to occur according to two different scenarios: (a) excessive oxygen availability, at RON equal to 6 ($\text{RON}_{\text{worst,a}}$), and (b) insufficient oxygen availability, at RON equal to 4.8 ($\text{RON}_{\text{worst,b}}$). In both cases, the control action will have to be the maximum possible in order to re-establish the system operation to its best as soon as possible. The value at $\text{RON}_{\text{worst,a}}$ was chosen since it was the maximum possible indicating the excessive oxygen supply. The value at $\text{RON}_{\text{worst,b}}$ was chosen when the curve for the N_2O emission factor started increasing towards rather high N_2O emissions. In addition to this, regardless the N_2O emission factors, the maximum control action will have to be taken when the effluent ammonium is above the typical legal limit of 4 mg N.L^{-1} .

Figure 1: (a) N_2O emission factors, and (b) effluent NH_4^+ concentration against oxygen-to-nitrogen loading ratio (RON) at a temperature of $15^\circ C$ according to the ASMG1.

Definition of the control structure

The control structure is defined to monitor the degree of complete nitrification as follows: If all the ammonium oxidized by AOB in the aerobic zone is converted into nitrate, this indicates that all the AOB-produced nitrites have been oxidized by NOB and not reduced by AOB to produce N_2O . Therefore, the relative ratio of the ammonium consumed by AOB over the amount of nitrate produced by NOB is used to monitor the extent of nitrite build up which triggers N_2O emissions from continuously-aerated systems. This ratio between overall nitrate produced and overall ammonium nitrogen depleted in an aerobic zone (R_{NatAmm}), expressed in Eqn. (1), is used here as candidate controlled variable.

$$R_{NatAmm} = \frac{NO_3^-|_{OUT,AER} - NO_3^-|_{IN,AER}}{NH_4^+|_{IN,AER} - NH_4^+|_{OUT,AER}} \quad (1)$$

Although theoretically the value of R_{NatAmm} of 1 represents the right balance between AOB and NOB, in practice this ratio is expected to be higher due to additional processes such as biomass decay and additional organic nitrogen release. In control applications, the optimal ratio and therefore set point can be computed from open loop simulation, which is done here.

By setting R_{NatAmm} at its optimal set point, N_2O production by AOB and HB is expected to be automatically minimized while avoiding wasting aeration energy and inhibiting HB denitrification.

Since the conversion of organic nitrogen into nitrate is sped up by an increased operating temperature, the optimal value for R_{NatAmm} is expected to increase in function of operating temperature. To take this into account, temperature adaptation of the set points for R_{NatAmm} is included using a separate fuzzy-logic module, using the measured influent temperature as single input variable.

While keeping R_{NatAmm} at its optimal value and thus reducing N_2O production, it is important to ensure a sustained ammonium conversion with the aim of respecting ammonium effluent limits. For this reason, the

effluent ammonium concentration is used as additional input to the control system to support taking the decision on the proper control action.

With regard to the manipulated variable used to achieve the control objective, either the air supply, here represented by the oxygen mass transfer coefficient (k_{La}), or the oxygen concentration set point (DO_{SP}) of a Proportional Integral (PI) controller, will be adopted to regulate the oxygen availability in the aerobic zone. It will be evaluated which one of the two manipulated variables is more effective in adapting the oxygen demand to achieve the optimal value of R_{NatAmm} and thus reducing N_2O emissions and keeping low $[NH_4^+]_{eff}$. Eqns. (2-5) summarize the control structure in terms of disturbances ($\underline{d}$), states ($\underline{x}$), manipulated ($\underline{u}$) and controlled variables ($\underline{y}$) around an aerated zone.

$$\underline{d} = [NH_{4,in}^+, S_{S,in}, T_{in}, X_{S,in}, X_{P,in}, NO_{2,in}^-, NO_{3,in}^-, Q_{in}, NO_{in}, N_2O_{in}, N_{2,in}, X_{I,in}, S_{I,in}] \quad (2)$$

$$\underline{x} = [NH_4^+, S_S, T, X_S, X_P, NO_2^-, NO_3^-, NO, N_2O, N_2, X_I, S_I] \quad (3)$$

$$\underline{y} = [R_{NatAmm}, NH_{4,eff}^+] \quad (4)$$

$$\underline{u} = [k_{La}] \text{ or } [DO_{SP}] \quad (5)$$

Based on the two manipulated variables shown in Eqn. (5), two generic control structures are implemented in an aerobic zone of a WWTP for the minimization of N_2O emissions, as shown in Figure 2. As can be seen, the measured ammonium and nitrate nitrogen concentration in the inlet and outlet of the aerated zone are used for the calculation of R_{NatAmm} . The influent temperature to the system is used to update the set point for R_{NatAmm} ($R_{NatAmm,SP}$) through a fuzzy-logic module. The difference between $R_{NatAmm,SP}$ and R_{NatAmm} , along with the effluent NH_4^+ concentration, is given as input to the fuzzy-logic control, which infers the unitary variation for the manipulated variable (either $\Delta_U k_{La}$ or $\Delta_U DO_{SP}$). This variation is named unitary because it is comprised between a minimal value of -1 and a maximal value of +1. This quantity therefore forms an indicator of whether the manipulated variable has to be increased or decreased. To attribute a physical dimension to this quantity, a scaling factor (either SF_{kLa} or $SF_{DO_{SP}}$), of the same order of magnitude as the nominal value of the corresponding manipulated variable, is multiplied with this quantity. By summing up these unit variations in time, the variations Δk_{La} or ΔDO_{SP} constitute the difference between the value of the actual manipulated variable and its corresponding nominal value (either $k_{La,nom}$ or $DO_{SP,nom}$). The exact

setting of the manipulated variable is thus known by adding its corresponding nominal value. In the configuration where the set point for dissolved oxygen is used as manipulated variable (Figure 2b), the DO_{SP} inferred through the fuzzy-logic controller is then used in a PI controller exploiting the DO measurements.

Figure 2: Control system implementation using as manipulated variable: **(a)** oxygen mass transfer coefficient, and **(b)** set point of dissolved oxygen concentration.

Considerations about the expected effect of such a control strategy on the N_2O dynamics contribution from heterotrophs and the reasons for not choosing nitrous oxide and/or nitrites as controlled variables are reported in Appendix A1.

Definition of the physical constraints

In line with the definition of the BSM2, the k_La is kept between 0 and 360 d^{-1} . The values for DO are kept between 0.5 and $3\text{ mg (-COD).L}^{-1}$ while the oxygen concentration in the stream recycled to the anoxic zone is kept between 0.5 and $1\text{ mg (-COD).L}^{-1}$. The choice of the minimal oxygen concentration equal to $0.5\text{ mg (-COD).L}^{-1}$ was taken to prevent compromising biomass settleability. The maximum oxygen concentration of $3\text{ mg (-COD).L}^{-1}$ was chosen to avoid wasting aeration energy without improving process performance. It is also well-known that WWTPs working with continuous aeration do not usually surpass this threshold (Holenda et al., 2008; Tallec et al., 2006, 2008). The upper limit in the stream carried to the anoxic zone ($1\text{ mg (-COD).L}^{-1}$) was chosen to avoid direct oxygen inhibition of HB denitrification, which causes a higher amount of non-oxidized organic carbon to be fed in the aerobic zone. This in turn has been found to remove oxygen from NOB (due to competition with heterotrophs), where the NOB play a very important role in avoiding nitrite accumulation.

The physical constraints on the controlled variables R_{NatAmm} and $[\text{NH}_4^+]_{\text{eff}}$ are expressed in Eqn. (6). As can be seen, both of them have a lower limit of zero. The upper limit for R_{NatAmm} is given by the sum of the unit, which is achieved when all the influent ammonium is converted into nitrate, and the ratio between the total amount of influent organic nitrogen ($N_{\text{org,inAER}}$) and the amount of influent ammonium consumed. As a matter of fact, as previously described, the organic nitrogen influent to the aerobic zone can be, under high oxygenation regimes, quickly hydrolysed and ammonified, oxidized into nitrite and finally converted into nitrate by NOB. Thus R_{NatAmm} can have values higher than unity. $\text{TKN}_{\text{in,AER}}$ is the Total Kjeldahl Nitrogen in

the influent to the aerobic zone which is used to represent the maximal amount of effluent ammonium that could potentially be released. As a matter of fact, at rather poor oxygen levels, the influent ammonium is not consumed and, in addition, the influent organic nitrogen is hydrolysed and ammonified.

$$\begin{cases} 0 \leq R_{NatAmm} \leq \left(1 + \frac{N_{org,inAER}}{\Delta NH_4^+}\right) \\ 0 \leq [NH_4^+]_{eff} \leq TKN_{inAER} \end{cases} \quad (6)$$

All disturbances and state variables other than the oxygen concentrations in the aerobic zone can have theoretical values comprised between 0 and $+\infty$.

Identification of the key scenarios leading the system away from the control objectives

The best system operation is achieved when both N_2O emissions are minimized and effluent ammonium concentration respects the legal effluent limits. On the other hand, situations when N_2O emissions are not minimized and/or effluent ammonium concentrations do not respect legal effluent limits are considered to represent a biological system that is away from optimality. Table 1 summarizes all of these conditions. The parameters used as indicators of the proximity of the system to the control objectives are: (a) the net N_2O produced by AOB and HB in the aerobic zone, (b) the net N_2O produced by HB in the anoxic zone, and (c) the effluent ammonium nitrogen concentration.

Table 1: Summary of scenarios leading the biological system away from optimality.

Relationships between optimization and controlled variables

Differently from Boiocchi et al. (2016), in this case study the relationship between control objectives and controlled variables is not calculated on the basis of mathematical expressions but is qualitatively derived from the steady state results achieved during the sensitivity analysis described in Boiocchi et al. (2017). This approach is chosen due to the high degree of uncertainty linked to the calculation of N_2O emissions, where several biological and physical processes are involved.

Figure 3 shows the relationships between N_2O emission factors and RON and the relationships between R_{NatAmm} and RON at three different operating temperatures (10°C, 15°C and 20°C). Figure 3b, representing

the scenario at temperature of 15°C, will be used to derive the critical points for R_{NatAmm} . Figures 3a and 3c will be used to build up the fuzzy-logic module adjusting the set points for R_{NatAmm} in function of influent temperature.

Figure 3: Steady-state N_2O emission factors and R_{NatAmm} in function of R_{ON} at the operating temperatures of (a) 10 °C, (b) 15 °C, and (c) 20°C.

In real applications, historical measurements of nitrogen concentrations and N_2O emissions could be used instead of sensitivity analysis results obtained from model simulations. As an alternative, a model can be calibrated on the full-scale WWTP. This model can then be exploited for sensitivity analyses to achieve the same relationships as the ones used in this paper. In general, it is expected that the minimal value of R_{NatAmm} to be achieved with the aim of having a balanced activity between AOB and NOB should not go below unity in any case. On the other hand, the value of R_{NatAmm} indicating the worst system operation due to excessive oxygen supply could be more subjective and case-specific.

2.2. Identification of the critical points for the controlled variables

Critical points for the controlled variable R_{NatAmm} are retrieved from Figure 3b. As can be seen, the value of R_{NatAmm} minimizing N_2O emissions is equal to 1.2. The reasons for having an optimal point of R_{NatAmm} higher than the theoretical value of 1 are described in Appendix A2.

However, to incorporate potential error in the measurements, negative and positive variations of 0.05 around this set point were considered to be completely acceptable. The value of R_{NatAmm} which describes the so-called “worst system operation” due to insufficient oxygen availability is found to be equal to 1 while the value of R_{NatAmm} which describes the same operation of the system due to excessive oxygen supply is found to be equal to 1.4.

With regards to the effluent ammonium concentration, although a typical legal limit for effluent ammonium concentrations is 4 mg N.L^{-1} , in order to stay on the safe side, it was decided for the controller to identify values higher than 2 mg N.L^{-1} as the worst system operation requiring maximal control action. Optimal values for $[\text{NH}_4^+ - \text{N}]_{\text{eff}}$ are chosen to be equal or lower than 1.5 mg N.L^{-1} .

Eqns. (7-10) express the critical points for the vector of controlled variables identified for each system operation, optimal and worst, for each scenario of Table 1.

$$UB \text{ for } OPTIMAL \text{ SYSTEM OPERATION} \Rightarrow \underline{y}_{UB,OPT} = [1.2 \ 0] \quad (7)$$

$$LB \text{ for } OPTIMAL \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} y_{LB,OPT}^1 = [1.2 \ 1.5] \\ y_{LB,OPT}^2 = [1.25 \ 0] \\ y_{LB,OPT}^3 = [1.15 \ 0] \\ y_{LB,OPT}^4 = [1.25 \ 1.5] \\ y_{LB,OPT}^5 = [1.15 \ 1.5] \end{cases} \quad (8)$$

$$UB \text{ for } WORST \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} y_{UB,WORST}^1 = [1.2 \ 2] \\ y_{LB,OPT}^2 = [1.4 \ 0] \\ y_{LB,OPT}^3 = [1 \ 0] \\ y_{LB,OPT}^4 = [1.4 \ 2] \\ y_{LB,OPT}^5 = [1 \ 2] \end{cases} \quad (9)$$

$$LB \text{ for } WORST \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} y_{UB,WORST}^1 = [1.2 \ TKN_{in,AER}] \\ y_{UB,WORST}^2 = \left[\left(1 + \frac{N_{org,inAER}}{\Delta NH_4^+} \right) 1.5 \right] \\ y_{UB,WORST}^3 = [0 \ 1.5] \\ y_{UB,WORST}^4 = \left[\left(1 + \frac{N_{org,inAER}}{\Delta NH_4^+} \right) TKN_{in,AER} \right] \\ y_{UB,WORST}^5 = [0 \ TKN_{in,AER}] \end{cases} \quad (10)$$

2.3. Definition of the membership functions for input and output variables

The first step in the definition of the membership functions (MFs) for the input variables consists of the identification of the key CVs whose variation clearly indicates the moving away from optimality. In this case, both R_{NatAmm} and effluent NH_4^+-N are key for the identification of the different scenarios presented in Table 1.

287 The following fuzzy sets were able to be identified for R_{NatAmm} and $[NH_4^+-N]_{eff}$:

288

289

290

291

292 With regards to the shape of the membership functions for input and output variables, triangular and
 293 trapezoidal shapes were chosen for the sake of simplicity.

294 The last step consists in the assignment of the degree of membership to fuzzy sets previously identified for
 295 each numerical value of R_{NatAmm} and $[NH_4^+-N]_{eff}$. A degree of membership to the identified fuzzy sets equal
 296 to 1 will be assigned to the critical points defined in subsection 2.2 as expressed in Eqns. (10-13). A degree
 297 of membership equal to 0 to the other fuzzy sets will be assigned to the same critical points.

298

$$UB \text{ for } OPTIMAL \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} (R_{NatAmm})_{GOOD} = 1.2 \\ (NH_{4,eff}^+)_{GOOD} = 0 \end{cases} \quad (10)$$

$$LB \text{ for } OPTIMAL \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} (R_{NatAmm})_{GOOD} = 1.15 \text{ and } 1.25 \\ (NH_{4,eff}^+)_{GOOD} = 1.5 \end{cases} \quad (11)$$

$$UB \text{ for } WORST \text{ SYSTEM OPERATION} \Rightarrow \begin{cases} (R_{NatAmm})_{LOW} = 1 \\ (R_{NatAmm})_{HIGH} = 1.4 \\ (NH_{4,eff}^+)_{HIGH} = 2 \end{cases} \quad (12)$$

$$(13)$$

$$LB \text{ for WORST SYSTEM OPERATION} \Rightarrow \begin{cases} (R_{NatAmm})_{LOW} = 0 \\ (R_{NatAmm})_{HIGH} = \left(1 + \frac{N_{org,inAER}}{\Delta NH_4^+}\right) \\ (NH_{4,eff}^+)_{HIGH} = TKN_{in,AER} \end{cases}$$

299

300 Figure 4 shows the resulting membership functions for R_{NatAmm} and $[NH_4^+ - N]_{eff}$.

301 **Figure 4:** Membership functions for: (a) R_{NatAmm} and (b) $[NH_4^+ - N]_{eff}$.

302

303 As disclosed in the Section “control structure definition”, the difference between the $R_{NatAmm,SP}$ and R_{NatAmm}
 304 ($E_{RNatAmm} = R_{NatAmm,SP} - R_{NatAmm}$), and not R_{NatAmm} , is used as input variable. This makes the external change of
 305 the set point easier, which is in this case needed to incorporate the temperature effect. To obtain the
 306 membership functions for $E_{RNatAmm}$ from the ones for R_{NatAmm} , the numerical values of the membership
 307 function (on the x-axis) in Figure 4a will be multiplied by -1. Then the value of 1.2 will be added, which is
 308 the set point at 15°C for R_{NatAmm} . The membership functions shown in Figure 5 are thereby obtained as a
 309 result.

310

311 **Figure 5:** Membership functions for $E_{RNatAmm}$.

312

313 Similarly to Boiocchi et al. (2016), the membership functions for the unitary variation of $k_L a$ or of DO_{SP} are
 314 defined such that the maximum positive value that this quantity will get is +1 and the maximum negative
 315 value that this quantity will get is -1. Taking into account the Center-of-Area as the chosen defuzzification
 316 method, the membership functions depicted in Figure 6 are defined accordingly.

317 **Figure 6:** Membership functions for the unitary variation of the manipulated variable ($\Delta_U MV$).

318

319 *Fuzzy logic module for temperature adaptation*

320 As previously mentioned, the set point for R_{NatAmm} is adapted in function of the temperature. This is because
 321 temperature increases the amount of influent organic nitrogen overall converted into nitrate and thus it
 322 sensibly increases the optimal set point for R_{NatAmm} at which N_2O emissions are minimized. The present
 323 fuzzy-logic module uses as input the influent temperature and as output $R_{NatAmm,SP}$. The membership

functions were decided on the basis of the values of R_{NatAmm} minimizing N_2O emissions at the three different temperatures from Figure 3. Specifically, the set points in Table 2 were found.

Table 2: Set points for R_{NatAmm} in function of temperature.

Extrapolating, the $R_{\text{NatAmm,SP}}$ for a temperature of 5°C was assumed to be 1 while the set point for a temperature of 25°C was assumed to be 1.6.

Choosing as default defuzzification method the Center-of-Area method, the membership functions for its input and output variables shown in Figure 7 were designed.

Figure 7: (a) Membership functions for the influent temperature, and (b) membership functions for $R_{\text{NatAmm,SP}}$.

2.4. Implementation of the linguistic rules

The following look-up table (Table 3) shows the IF-THEN linguistic rules connecting the inputs (E_{RNatAmm} and the effluent ammonium nitrogen concentration) to the output variable ($\Delta_{\text{U}}k_{\text{LA}}$ or $\Delta_{\text{U}}\text{DO}_{\text{SP}}$). As can be noted, when $[\text{NH}_4^+-\text{N}]_{\text{eff}}$ is found to belong to the fuzzy set “HIGH”, the controller increases the manipulated variable, regardless of the value of E_{RNatAmm} . Otherwise, the controller will: (a) increase the oxygen availability if E_{RNatAmm} belongs to the fuzzy set “HIGH” (i.e. when R_{NatAmm} is lower than the set point), which indicates a suboptimal NOB activity rate, or (b) decrease it if E_{RNatAmm} belongs to the fuzzy set “LOW” (i.e. when R_{NatAmm} is higher than the set point), which indicates waste of oxygen supply, enhancement of N_2O production by HB in the anoxic zone and worsening of effluent quality. If both the effluent NH_4^+ and E_{RNatAmm} belong to the fuzzy set “GOOD”, no control action is taken.

Table 3: Look-up table for the linguistic rules linking E_{RNatAmm} and $(\text{NH}_4^+)_{\text{eff}}$ to $\Delta_{\text{U}}\text{MV}$ (either $\Delta_{\text{U}}k_{\text{LA}}$ or $\Delta_{\text{U}}\text{DO}_{\text{SP}}$).

The linguistic rules for the temperature adaptation fuzzy-logic module are shown in Table 4.

Table 4: Linguistic rules linking the temperature to $R_{\text{NatAmm,SP}}$.

351

352 **2.5 Setting of additional design parameters**

353 The additional design parameters to be decided were the implication and the aggregation method.
 354 Correlation-minimum was chosen as the implication method whereas disjunctive was chosen as the
 355 aggregation method.

356 The scaling factor used for the controller was chosen to be equal to or below the difference between the
 357 saturation limit for the manipulated variable used and its nominal value.

358

359 **3. Controller implementation and performance evaluation**

360 Once designed, the control strategy was implemented and tested in the following three benchmark simulation
 361 models: the Benchmark Simulation Model n°2 for Nitrous oxide a (BSM2Na), b (BSM2Nb) and c
 362 (BSM2Nc). These three models were developed by using respectively the ASMG1, the two-pathway model
 363 by Pocquet et al. (2016) and the two pathway model by Domingo-Felez and Smets (2016) for the description
 364 of the biological processes in the mainstream activated sludge unit of the BSM2. Therefore, the benchmark
 365 simulation models obtained thereby describe different N₂O production dynamics. More specifically, the
 366 BSM2Na describes N₂O production during HB and AOB denitrification, but not during the incomplete
 367 oxidation of hydroxylamine (NH₂OH), while the BSM2Nb and the BSM2Nc incorporate additionally the
 368 N₂O production during the incomplete NH₂OH oxidation but according to different model equations and
 369 assumptions. HB denitrification and NOB activity are modelled the same for all three models. Although the
 370 models by Pocquet et al. (2016) and Domingo-Felez and Smets (2016) were not calibrated with full-scale
 371 plant data, these two are the only models available in the literature including both the two AOB-mediated
 372 N₂O producing pathways. The only two works published regarding N₂O model calibrations with full-scale
 373 WWTP data are the one by Ni et al. (2013) and by Spérandio et al. (2016). None of the models provided in
 374 these works includes both the N₂O production pathways by AOB. This is because identification of the rates
 375 of these two pathways with full-scale plant data is very hard and the hydraulics of full-scale WWTPs is
 376 intrinsically much more difficult to model compared to lab-scale reactors (Sin et al., 2008). Even in case a

model was able to accurately describe full-scale measurements, it would be only valid for the particular WWTP used for the calibration and a lot of uncertainties in the model parameters and in the dominant N_2O producing pathway would still remain when applying the model for other WWTP configurations with different operation and influent composition (Sin et al., 2009). On the contrary, the aim of the control testing performed in this work is to evaluate the controller robustness for a wider range of WWTPs. In fact, as a consequence of the different N_2O modelling approaches by Domingo-Felez and Smets (2016), Guo and Vanrolleghem (2014) and Pocquet et al. (2016), the three benchmark models predict N_2O dynamics differently (see Boiocchi (2016)), which will enable identifying the range of WWTPs where the controller could be successfully applied, and also serves to demonstrating the general applicability of the proposed controller.

A more detailed description of the three benchmark simulation models and how they were developed is given in Boiocchi (2016) while Appendix A3 in Supplementary Material (SI) will provide a description of the structures and default parameter values used for the BSM2Na, BSM2Nb and BSM2Nc. Figure 8 shows the implementation of both the regulatory and the cascade control strategy in the BSM2 layout.

Figure 8: (a) Implementation of the regulatory control strategy, and **(b)** implementation of the cascade control strategy.

The testing and evaluation procedure will be performed as suggested by Jeppsson et al. (2007). More specifically, the performance criteria used to evaluate the control strategies are summarized in Appendix A4 in the SI, including also the algebraic equations used for their calculation.

For each simulation model, the evaluation criteria will be calculated for the novel closed-loop configurations and compared against the ones obtained for a reference scenario. The latter is identified for each of the three benchmark simulation models through the implementation of a cascade controller regulating the set point of the oxygen concentrations in the three aerobic tanks on the basis of the effluent ammonium concentration according to a PI approach (see Appendix A5 in SI for this configuration). In a next instance, sensor and actuator dynamics are introduced using the sensor and actuator models given by Alex et al. (2008). More specifically, the continuous sensors of type A, working without delay, were chosen for temperature and oxygen concentrations while sensors type B1 (photometric with normal filtration) were included for the

measurements of ammonium and nitrate concentrations. Type-A actuators were used for the manipulation of the oxygen mass transfer coefficients. Appendix A6 in SI shows the resulting deviations from the actual values imposed for all measured variables.

Finally, the sensitivity of the controller with respect to model parameter variation is analysed. Therefore, the cascade configuration of the controller is simulated on the BSM2Na by varying the oxygen affinity constants for the aerobic activity of AOB and NOB ($K_{O_2,AOB}$ and $K_{O_2,NOB}$, respectively) according to various values estimated in the literature. The reason for focusing the sensitivity analysis of the controller on these two affinity constants is due to the fact that one of the basic controller ideas consists of triggering the activity of NOB over the one of AOB by increasing oxygen availability. This is because it was found that NOB are more limited by low oxygen concentrations than AOB and increasing the oxygenation would enhance the uptake of AOB-produced nitrite by NOB and thereby prevent N_2O formation. The fact that NOB are more oxygen-limited than AOB translates to a higher value of $K_{O_2,NOB}$ than $K_{O_2,AOB}$ estimated by Hiatt and Grady (2008). Nevertheless, the relationship between $K_{O_2,NOB}$ and $K_{O_2,AOB}$ is still uncertain. In fact, although there is a trend in the literature to estimate $K_{O_2,AOB}$ with lower values than $K_{O_2,NOB}$ as clearly shown in Guisasola et al. (2005), there are a few other studies which have estimated values for $K_{O_2,AOB}$ higher than $K_{O_2,NOB}$ (Manser et al., 2005; Picioreanu et al., 2016). The difference in the respective value of these two constants can be addressed to: different species composition of AOB and NOB, floc size, temperature, size of the colonies of AOB and NOB, spatial distribution of AOB and NOB (Picioreanu et al., 2016). All of these aspects, which are not considered explicitly, may hinder the good performance of the present control strategy.

4. Results

In this section, the simulation results obtained using the BSM2Na, BSM2Nb and BSM2Nc are presented and discussed. The control strategies tested and benchmarked are:

- 1) NH_4^+ cascade controller, using directly the set point for dissolved oxygen (DO_{SP}) as manipulated variable (*CS1*) using 1.5 mg N.L^{-1} as set point for the effluent ammonium nitrogen (i.e. the reference control strategy),
- 2) R_{NatAmm} and NH_4^+ regulatory controller, using directly $k_L a$ as manipulated variable (*CS2*),

- 3) R_{NatAmm} and NH_4^+ cascade controller, using directly the set point for dissolved oxygen (DO_{sp}) as manipulated variable (CS3),
- 4) Control strategy CS3 with sensor and actuator noise added (CS4).

Section 4.1 will be analysing the N_2O dynamics, Section 4.2 will be focusing on the effluent quality, Section 4.3 will be discussing the energy consumption linked to aeration, and finally in Section 4.4 the sensitivity of the controller to specific model parameters will be analysed.

4.1. Control performance on the N_2O emissions

Table 5 show the emission factors for N_2O , R_{NatAmm} , and the N_2O contributions by the different pathways according to the BSM2Na, BSM2Nb and BSM2Nc, respectively. The Integral Absolute Error and the Integral Square Error for R_{NatAmm} are included as well in order to evaluate how the tracking of the set point for R_{NatAmm} affects the N_2O emissions. The total variation of the k_{La} is considered to include the aggressiveness of the control response.

As can be deduced by comparing the results of CS2 and CS3 against the reference configuration CS1 using the BSM2Na and BSM2Nc, the novel controller is able to drastically reduce the average rate of AOB denitrification by increasing the average value of R_{NatAmm} . More specifically, as the tracking of the set point for R_{NatAmm} – indicated by the IAE and the ISE – improves, the reduction of the amount of N_2O produced by AOB increases. A contextual decrease in the average amount of N_2O consumed by HB can also be observed. This can be largely assigned to the overall lower N_2O accumulation in the liquid phase, which in turn slows down the last HB denitrification reduction step. Since the decrease of N_2O produced by AOB is more than the decrease of N_2O consumed by HB, both the total N_2O emitted per unit of influent TKN (N_2O_{efl}) and the total N_2O emissions per unit of TKN removed (N_2O_{ef2}) have been drastically reduced in the BSM2Na and BSM2Nc. The fact that also N_2O emissions per unit of TKN removed diminished demonstrates the fact that N_2O has not been reduced by lowering the overall TKN conversion, but by reducing the rates of those specific processes producing it.

With regard to the BSM2Nb, it has to be noted first that the model has incomplete NH_2OH oxidation as the main N_2O producing pathway. As can be noted, the control strategy is able to decrease the amount of N_2O produced according to the AOB denitrification and incomplete NH_2OH oxidation pathways. The ability of the controller in reducing the amount of N_2O produced by this last pathway can be attributed to the fact that the controller aims at achieving complete nitrification, which means that accumulation of intermediates like NH_2OH can be suppressed as well. Nevertheless, the reduction of N_2O consumption by HB with CS2 and CS3 is larger than the overall reduction of N_2O produced by the AOB pathways. This can be ascribed to the fact that the control strategies CS2 and CS3 need to increase the oxygen supply more to achieve complete nitrification than in the other models, due to the relevance of the incomplete NH_2OH oxidation pathway. By doing so, the stripping of N_2O is triggered and HB have less chance to reduce N_2O into N_2 . As a consequence of this, N_2O emissions are slightly higher when CS2 and CS3 are applied compared to the case when the NH_4^+ cascade controller is applied (CS1). However, it should also be noted that the discrepancies are marginal, which means that the novel controllers do not drastically reduce the performance of the plant. The results also suggest that the present controllers should not be used for those plants where the incomplete NH_2OH oxidation is the main N_2O producing process.

Adaptability to temperature changes

The capability of the controller to adapt to temperature variations is checked by observing Figure 9, showing the dynamics of the N_2O production in the aerobic zone by the different processes and the total amount of N_2O emitted as predicted by the BSM2Nc for strategies CS1, CS2 and CS3. As can be observed from the results with CS1, N_2O emissions (Figure 9a) are in phase with N_2O produced by AOB (Figure 9b). More specifically, as the temperature increases, N_2O production by AOB increases and N_2O emissions follow the same trend. The behaviour of AOB associated N_2O production is to be attributed to the balance between the aerobic growth of AOB and the aerobic growth by NOB. According to Hellinga *et al.* (Hellinga et al., 1999), NOB growth has a disadvantage compared to the growth of AOB as temperature increases. This is in fact one of the reasons why the nitrification systems, like the SHARON, are able to work efficiently. As a matter of fact, by keeping the operating temperatures high (30-35°C), NOB can be washed out in such systems. In activated sludge systems like the one considered here, this behaviour leads to higher nitrite accumulation at

warm temperatures (see Figure 9c) which, in turn, triggers AOB denitrification and, consequently, N_2O emissions. From Figure 9d it can be noted that, similarly to AOB denitrification, HB-mediated N_2O production (i.e. the third HB denitrification step) is high when temperature increases due to higher availability of AOB-produced nitrites. When the novel controllers are implemented, nitrite accumulation is drastically reduced in summer, which in turn slows down AOB denitrification. Similarly, the N_2O produced by HB is reduced drastically by *CS2* and *CS3* (see Figure 9d). Nevertheless, the consumption of N_2O by HB during the last reduction step is reduced (see Figure 9e). Overall, the net N_2O production by HB in the aerobic zone is always negative (see Figure 9f), which means that the amount of N_2O consumed by HB is higher than the amount of N_2O produced by HB, and therefore a fraction of AOB-produced N_2O is consumed by HB. This overall N_2O consumption by HB decreases when the controllers are implemented. This fact can be explained by the lower availability of N_2O in the liquid phase partly due to lower AOB denitrification and partly due to higher N_2O stripping. Nevertheless, the overall N_2O emitted is reduced through implementation of the controllers.

Exceptionally, for cold temperatures the regulatory controller (*CS2*) is not able to optimize the oxygen supply to reduce the nitrite accumulation, compared to the reference scenario given by *CS1*. This led to higher N_2O production by AOB and HB (see Figures 9b and 9d). Nevertheless, also the resulting N_2O consumption by HB was higher, which in turn compensated for the higher N_2O production and smoothed considerably the resulting increase of the amount of N_2O emitted. The cascade controller (*CS3*) was on the contrary able to optimize the oxygen supply better so that nitrite concentration was optimized also for cold temperatures.

Robustness against sensor and actuator noise

As can be noted by comparing the average N_2O emitted per unit of influent TKN and unit of TKN removed with *CS3* and *CS4* according to each model, the novel control strategy *CS3*, and presumably *CS2*, can be considered robust against the sensor and actuator noise. *CS4* is in fact able to achieve a drastic reduction of the N_2O emissions compared to the open-loop. According to all the three models, the IAE and ISE values achieved using *CS4* are slightly higher than the ones achieved with *CS3*, which in turn leads to slightly higher N_2O emissions with *CS4*. Nonetheless, the manipulation of the oxygen mass transfer coefficient

needed to cope with the imposed sensor and actuator noise seems to be much more aggressive, as can be noted from the calculated value of TV.

Table 5: N_2O emission factors, average R_{NatAmm} and average N_2O produced by HB and AOB predicted by the BSM2Na, BSM2Nb and BSM2Nc with the four control strategies (*CS1*, *CS2*, *CS3* and *CS4*).

Figure 9: (a) total N_2O emitted, (b) N_2O production by AOB in the aerobic zone, (c) average NO_2^- concentration in the aerobic zone, (d) N_2O produced by HB in the aerobic zone (third HB denitrification step), (e) N_2O consumed by HB in the aerobic zone (fourth HB denitrification step), and (f) net N_2O produced by HB in the aerobic zone (third plus fourth denitrification steps), predicted by the BSM2Nc with *CS1* (blue line), *CS2* (green line) and *CS3* (red line).

4.2. Control performance on the effluent quality

Within the context of mitigation of N_2O emissions, the effluent quality has to be considered as well. Table 6 show the average TKN removal efficiency, TN removal efficiency, ammonium and total nitrogen violations and effluent quality index with *CS1*, *CS2*, *CS3* and *CS4* according to the BSM2Na, BSM2Nb and BSM2Nc predictions, respectively.

As can be noted, according to all the three models, *CS2* and *CS3* are able to keep the overall TKN removal efficiency at high levels. Furthermore, it can be easily demonstrated that the controllers are able to keep the frequency of ammonium effluent limit violations low (4 mg N.L^{-1}). *CS3* performed considerably better in reducing the ammonium violations than *CS2*.

The effluent ammonium violations obtained by *CS1* are negligible. The cascade PI controller is able to keep respecting the effluent ammonium limits throughout the entire simulation period and performs in this regard better than *CS2* and *CS3*. The reason for this can be due to the fact that *CS2* and *CS3* have also to cope with N_2O emissions, and the control objectives can be sometimes conflicting. In hot seasons aeration has to be increased to work out the lower NOB activity rate compared to AOB activity rate. Contrarily, in winter the controller, while keeping the nitrification complete, attempts to avoid aeration energy waste and high N_2O production by HB denitrifiers. Thus there will be instances in cold temperatures when *CS2* and *CS3* will

infer a decrease of the oxygen supply. As a consequence, this will cause a temporary increase of effluent ammonium, which can become higher than the effluent limit. Anyhow, CS2 and especially CS3 are able to ensure high TKN conversion, which means that they are able to cope with the slightly higher frequency of ammonium violations. To be noted is also that the TKN removal efficiency is higher when using CS3 than when using CS1. As a drawback of CS2 and CS3, the effluent TN violations are variably increased. This has to be attributed to the higher COD demand by heterotrophic denitrifiers to perform the complete reduction of autotrophically-produced nitrogen oxides (such as nitrates and nitrites) into dinitrogen gas (N_2). As a matter of fact, since the controllers achieve a more complete oxidation of ammonium into nitrate and thus avoid nitrite leftover, HB denitrification will receive a higher load of nitrate in the anoxic zone, which requires more biodegradable COD for complete reduction to N_2 (Hellinga et al., 1999). Consequently, due to incomplete denitrification, more nitrate will be in the effluent and a more severe violation of the effluent TN limit will occur, as shown in Table 6. For the same reason, the EQI significantly increases. Since CS3 was more able to achieve complete nitrification than CS2, the EQI is worse when CS3 is used.

Table 6: TKN and TN removal efficiencies, effluent average nitrate, ammonium and TN violations and effluent quality index predicted by the BSM2Na, BSM2Nb and BSM2Nc with the four control strategies (CS1, CS2, CS3 and CS4).

4.3. Control performance on aeration energy consumptions

To evaluate the economic feasibility of the control strategies, the aeration energy consumption (AEC) is considered (Table 7). As can be noted, CS2 and CS3 have led to a variable increment of the average aeration energy consumed according to all the three models. This was expected since more oxygen was needed in these strategies to ensure higher NOB activity. However, the variation is only between 3 and 6 %. More in detail, given the better regulation of the oxygen supply, some aeration energy could be saved by using the cascade configuration (CS3) rather than the regulatory controller (CS2). The energy consumption linked to the electricity consumption needed for the aeration system (surface aerators versus blowers) is another important component of the total carbon footprint of WWTPs. This contribution is proportional to a certain emission factor associated with type/source of electricity production (e.g. coal versus natural gas fired

combustion versus renewable energy from wind, solar, etc...) and with the efficiency of aeration systems themselves. Since the quantification of the total carbon footprint is outside the scope of this contribution, average aeration energy consumption is instead used as an indicator to compare qualitatively the expected contribution by the aeration systems. Since the aeration energy consumption was only slightly affected by the novel control strategies tested in this study, it is expected that the proposed control strategy will lead to significant decrease in the total carbon footprint of the plant thanks to the major reduction on N_2O emissions achieved by the controllers.

Table 7: Average aeration energy consumption ($kWh \cdot d^{-1}$) with the four control strategies (*CS1*, *CS2*, *CS3* and *CS4*) according to the BSM2Na, BSM2Nb and BSM2Nc.

4.4. Sensitivity of the controller against model parameters

Table 8 compares the results achieved by simulating the BSM2Na using different $K_{O_2,AOB}$ - $K_{O_2,NOB}$ couples taken from literature with the cascade control configuration for low N_2O emissions (*CS3*) against the results achieved with the NH_4^+ cascade controller (*CS1*). Two main situations can be identified from the results: (a) $K_{O_2,AOB}$ is lower than $K_{O_2,NOB}$, and (b) $K_{O_2,AOB}$ is larger than $K_{O_2,NOB}$. In the first case, the default controller *CS1* is not able to keep the activities between *NOB* and *AOB* balanced. As a matter of fact, the average values achieved for R_{NatAmm} are rather low. Contrarily, when *CS3* is implemented, the average value for R_{NatAmm} is higher. The N_2O emission factors achieved using *CS3* are therefore much lower than the ones achieved using *CS1*. This improvement becomes larger as the difference between $K_{O_2,NOB}$ and $K_{O_2,AOB}$ increases. This is because *NOB* activity is more limited by low oxygen concentrations than *AOB* activity. On the other hand, when $K_{O_2,NOB}$ is lower than $K_{O_2,AOB}$, the values of R_{NatAmm} achieved with *CS1* are already in the ideal range necessary to minimize N_2O emissions. This is due to the fact that, differently from the previous scenario, *NOB* are less limited by poor oxygen conditions than *AOB*. This means that for the same oxygen concentration, the aerobic activity rate of *AOB* will be lower than the one of *NOB* and no nitrification intermediates will accumulate. When *CS3* is implemented instead, the supply of oxygen tends to be higher, the residence time of N_2O in the liquid phase is reduced and thus the rate of the last *HB* denitrification step gets reduced. Overall, the N_2O emissions are slightly higher. These results suggest

therefore that the novel control strategy proposed would be effective in drastically reducing N_2O emissions only for those plants that have a considerable contribution from AOB denitrification.

Table 8: N_2O emission actors, average R_{NatAmm} , and average aeration energy consumptions for the BSM2Na with different $K_{O_2, \text{AOB}}-K_{O_2, \text{NOB}}$ combinations. The last column is the literature reference where the parameter values were reported.

5. Conclusions and future perspectives

The work presented the development and testing of a novel control idea minimizing N_2O emissions while at the same time keeping low ammonium concentrations in full-scale continuously-aerated WWTPs. The strategy is based on the concept that accumulation of nitrification intermediates like nitrite has to be avoided in order to prevent N_2O production, and its consequent emission. In continuously aerated WWT systems, this can be done by triggering the ratio between NOB and AOB activity rates, which leads to minimization of nitrites produced by ammonia-oxidizing bacteria in aerobic environments.

The testing of the control strategy performed in this work was comprehensive. More specifically, the novel automated control strategy was found to enable achieving a drastic reduction of the total N_2O emissions in those plants having AOB denitrification as main N_2O producer. In plants where the incomplete NH_2OH oxidation pathway was the main N_2O producer, the controller was not as effective and a separate study should be performed to develop a control strategy specifically aimed at slowing down effectively this process.

The proposed control strategy is the first control technology being developed for automatic regulation of WWTP environmental conditions in function of online measurements with the specific aim of N_2O emission mitigation.

The preliminary results presented in this work should be followed up by the implementation of the control strategy in full-scale WWTPs with the aim of drastically reducing their carbon footprint. However, there is a side effect of the controller which may lead to increase in the effluent total nitrogen and especially the nitrate concentrations. Therefore appropriate analysis and corresponding adaptations to plant operations – such as enhancing complete heterotrophic denitrification – are needed.

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APPENDIX A1

Expected effect of controllers on HB denitrification

Since ensuring complete nitrification means that all the NO_2^- produced by AOB is consumed by NOB, this control strategy is expected to slow down not only the N_2O production by AOB denitrifiers but also the gross N_2O production by HB in the aerobic zone. As a matter of fact, it can occur due to low presence of oxygen and nitrite build-up that HB will start using AOB-produced nitrites as electron acceptors for the oxidation of organic carbon and thus produce N_2O , which, contrarily to what would happen in an anoxic zone, is likely to strip fast to the atmosphere given the higher stripping capability of the aerobic zone. This is because typical aeration regimes in the aerobic zone are established with the aim of guaranteeing effluent total ammonia concentrations below legal limits. However, also the last HB denitrification step, consuming N_2O , is slowed down by increased oxygen concentrations. The effect of oxygen on the N_2O net produced by HB and on the total N_2O emissions needs therefore to be properly verified through control testing.

Reasons for not choosing N_2O and NO_2^- as controlled variables

On-line measurements of N_2O concentrations in the liquid phase in the aeration zone are not considered useful for the achievement of the control objectives. As a matter of fact, the N_2O concentrations in the liquid phase can be rather low in an aerated zone, given the high stripping capability. These very low concentrations are much more subject to measurement errors, which in turn would hinder the good performance of a controller using N_2O measurements as an input. Furthermore, mere N_2O measurements do not help the controller to decide whether an increase or a decrease in the control action has to be inferred. On the contrary, measurements of nitrites in the aerobic zone would help detect an incomplete nitrification and, hence, influence the decision on the proper control action to be taken. However, not the entire N_2O is produced as a consequence of nitrite accumulation. There is in fact a high production of N_2O due to establishment of a critical concentration of dissolved oxygen (DO) which promotes NH_2OH build-up while the NO_2^- pick up by AOB is not inhibited yet. Since the nitrite measured would be able to quantify only the nitrite left over from all the reactions consuming it, in this specific case quite low nitrite concentrations can

result while a rather high N_2O production occurs. Nitrite measurements are therefore considered not representative of the entire N_2O production.

APPENDIX A2 – The optimal value for R_{NatAmm}

The difference between ammonium concentrations at the inlet and the outlet of the aerobic zone is supposed to quantify the amount of ammonium consumed by AOB whereas the difference between nitrate in the outlet and in the inlet of the aerobic zone quantifies the amount of nitrate produced by NOB. It is important that the measurements are taken from the inlet and the outlet of an aerobic zone and not of an entire WWTP. This is because the difference between nitrate in the plant effluent and in the plant influent would not represent the NOB activity only, since NO_3^- consumption by heterotrophs would be involved as well as an interfering process. Given these considerations, the theoretical value of R_{NatAmm} indicating complete nitrification should be around 1, which indicates that the ammonium consumed is equal to the amount of nitrate produced and no nitrification intermediate is used to produce N_2O . Nevertheless, the difference between ammonium in and out the aerobic zone does not entirely incorporate all the ammonium consumed by AOB. As a matter of fact, when most of the ammonium is depleted, AOB start using the influent organic nitrogen, which is quickly hydrolysed and ammonified, as electron donor. The nitrite produced thereby is subsequently converted into nitrate by NOB, which is added to the amount of nitrate produced from the oxidation of influent ammonium. The difference between nitrate in the outlet and the inlet to the aerobic zone will as a result be slightly higher than the difference between ammonium in the inlet and the outlet.

As far as the ammonium consumed as nutrient by the biomass, this quantity can be neglected for the following reasons: (a) the autotrophic biomass is only a small percentage of the overall sludge and therefore does not consume a relevant quantity of ammonium for growth; (b) part of the ammonium consumed as nutrient is subsequently released during biomass decay; (c) in a predenitrification configuration, heterotrophic biomass grows very slowly in the aerobic zone as most of the organic carbon is consumed in the anoxic compartment.

This explains why the optimal value of R_{NatAmm} resulting from Figure 3b is higher than 1, i.e. 1.2.

APPENDIX A3

1. Model for mainstream activated sludge processes in the BSM2Na

Table 1: (a) Stoichiometric matrix and (b) Kinetic vector for the AOB-mediated processes.

(a) Stoichiometric matrix						
	S_{NHX}	S_{NO2}	S_{O2}	S_{NO}	S_{N2O}	X_{AOB}
Aerobic AOB growth	$-1/Y_{AOB} \cdot i_{NXB}$	$1/Y_{AOB}$	$-(3.43 - Y_{AOB})/Y_{AOB}$			1
Anoxic AOB growth on NO_2^-	$-1/Y_{AOB} \cdot i_{NXB}$	$-1/Y_{AOB}$	$-(2.29 - Y_{AOB})/Y_{AOB}$	$2/Y_{AOB}$		1
Anoxic AOB growth on NO	$-1/Y_{AOB} \cdot i_{NXB}$	$1/Y_{AOB}$	$-(2.29 - Y_{AOB})/Y_{AOB}$	$-2/Y_{AOB}$	$2/Y_{AOB}$	1

(b) Kinetic vector	
Aerobic AOB growth	$\mu_{AOB,T} \cdot \frac{S_{O2}}{S_{O2} + K_{O2,AOB}} \cdot \frac{S_{FA}}{S_{FA} + K_{FA,AOB} + S_{FA}^2/K_{I9,FA}} \cdot \frac{K_{I9,FNA}}{K_{I9,FNA} + S_{FNA}} \cdot X_{AOB}$
Anoxic AOB growth on NO_2^-	$\mu_{AOB,T} \cdot \eta_{AnoxAOB} \cdot DO_{HaldaneFunc}^* \cdot \frac{S_{FA}}{S_{FA} + K_{FA,AOB}} \cdot \frac{S_{FNA}}{S_{FNA} + K_{FNA,AOB}} \cdot X_{AOB}$
Anoxic AOB growth on NO	$\mu_{AOB,T} \cdot \eta_{AnoxAOB} \cdot DO_{HaldaneFunc}^* \cdot \frac{S_{FA}}{S_{FA} + K_{FA,AOB}} \cdot \frac{S_{NO}}{S_{NO} + K_{NO,AOB}} \cdot X_{AOB}$

$$DO_{HaldaneFunc}^* = \frac{S_{O2}}{K_{SO2,AOBden} + (1 - 2 \cdot \sqrt{K_{SO2,AOBden}/K_{IO,AOBden}}) \cdot S_{O2} + S_{O2}^2/K_{IO,AOBden}}$$

Table 2: (a) stoichiometric matrix and (b) kinetic vector for the NOB activity.

(a) Stoichiometric matrix					
	S_{NHX}	S_{NO2}	S_{O2}	S_{NO3}	X_{NOB}
Aerobic NOB growth	$-i_{NXB}$	$-1/Y_{NOB}$	$-(1.14 - Y_{NOB})/Y_{NOB}$	$1/Y_{NOB}$	1

(b) Kinetic vector	
Aerobic NOB growth	$\mu_{NOB,T} \cdot \frac{S_{FNA}}{S_{FNA} + K_{FNA,NOB} + S_{FNA}^2/K_{I10,FNA}} \cdot \frac{S_{O2}}{S_{O2} + K_{O2,NOB}} \cdot \frac{K_{I10,FA}}{K_{I10,FA} + S_{FA}} \cdot X_{NOB}$

Table 3: (a) stoichiometric matrix and (b) kinetic vector for the HB-mediated processes.

(a)

Stoichiometric matrix

	S_S	S_{O_2}	S_{NO_3}	S_{NO_2}	S_{NO}	S_{N_2O}	S_{N_2}	X_{HB}
Aerobic HB growth	$-\frac{1}{Y_H}$	$\frac{1 - Y_H}{Y_H}$						1
Anoxic HB growth on NO_3^-	$-\frac{1}{n_Y \cdot Y_H}$		-A	A				1
Anoxic HB growth on NO_2^-	$-\frac{1}{n_Y \cdot Y_H}$			-B	B			1
Anoxic HB growth on NO	$-\frac{1}{n_Y \cdot Y_H}$				-B	B		1
Anoxic HB growth on N_2O	$-\frac{1}{n_Y \cdot Y_H}$					-B	B	1

$$A = \frac{1 - Y_H \cdot n_Y}{1.143 \cdot Y_H \cdot n_Y}$$

$$B = \frac{1 - Y_H \cdot n_Y}{0.571 \cdot Y_H \cdot n_Y}$$

(b)

Kinetic vector

Aerobic HB growth	$\mu_{HB,T} \cdot \frac{S_S}{S_S + K_{S1}} \cdot \frac{S_{O_2}}{S_{O_2} + K_{OH1}} \cdot \frac{S_{NH}}{S_{NH} + K_{NH,HB}} \cdot X_{HB}$
Anoxic HB growth on NO_3^-	$\mu_{HB,T} \cdot \eta_{AnoxHB,2} \cdot \frac{S_S}{S_S + K_{S2}} \cdot \frac{K_{OH2}}{S_{O_2} + K_{OH2}} \cdot \frac{S_{NO_3}}{S_{NO_3} + K_{NO_3}} \cdot X_{HB}$
Anoxic HB growth on NO_2^-	$\mu_{HB,T} \cdot \eta_{AnoxHB,3} \cdot \frac{S_S}{S_S + K_{S3}} \cdot \frac{K_{OH3}}{S_{O_2} + K_{OH3}} \cdot \frac{S_{NO_2}}{S_{NO_2} + K_{NO_2}} \cdot \frac{K_{I3,NO}}{K_{I3,NO} + S_{NO}} \cdot X_{HB}$
Anoxic HB growth on NO	$\mu_{HB,T} \cdot \eta_{AnoxHB,4} \cdot \frac{S_S}{S_S + K_{S4}} \cdot \frac{K_{OH4}}{S_{O_2} + K_{OH4}} \cdot \frac{S_{NO}}{S_{NO} + K_{NO} + S_{NO}^2 / K_{I4NO}} \cdot X_{HB}$
Anoxic HB growth on N_2O	$\mu_{HB,T} \cdot \eta_{AnoxHB,5} \cdot \frac{S_S}{S_S + K_{S5}} \cdot \frac{K_{OH5}}{S_{O_2} + K_{OH5}} \cdot \frac{S_{N_2O}}{S_{N_2O} + K_{N_2O}} \cdot \frac{K_{I5,NO}}{K_{I5,NO} + S_{NO}} \cdot X_{HB}$

Table 4: Parameter values used in the BSM2Na for the biological processes.

Parameters	Description	Default value at 15°C	Unit
Aerobic AOB activity			
Y_{AOB}	growth yield of AOB	0.18	g COD _{BIO} ·g ⁻¹ N
μ_{AOB}	maximum specific growth rate of AOB	0.58	d ⁻¹
K_{FA}	NH ₃ half saturation parameter for aerobic AOB activity	0.004	g N·m ⁻³
$K_{O,AOB}$	O ₂ half-saturation constant for AOB activity	0.6	g (-COD)·m ⁻³
$K_{I9,FA}$	NH ₃ inhibition constant for AOB activity	1	g N·m ⁻³
$K_{I9,FNA}$	HNO ₂ inhibition constant for AOB activity	0.1	g N·m ⁻³

b_{A1}	decay coefficient of AOB	0.028	d^{-1}
NOB activity			
Y_{NOB}	growth yield of NOB	0.06	$g\ COD_{BIO} \cdot g^{-1}\ N$
μ_{NOB}	maximum specific growth rate of NOB	0.68	d^{-1}
$K_{O,NOB}$	O_2 half-saturation constant for NOB activity	1.2	$g\ (-COD) \cdot m^{-3}$
$K_{II0,FA}$	NH_3 inhibition constant for NOB activity	0.5	$g\ N \cdot m^{-3}$
$K_{II0,FNA}$	HNO_2 inhibition constant for NOB activity	0.1	$g\ N \cdot m^{-3}$
K_{FNA}	HNO_2 half saturation parameter for NOB activity	10-6	$g\ N \cdot m^{-3}$
b_{A2}	decay coefficient of NOB	0.028	d^{-1}
Aerobic HB activity			
Y_H	growth yield of HB	0.6	$g\ COD_{BIO} \cdot g^{-1}\ COD$
μ_H	maximum specific growth rate of HB	4.78	d^{-1}
K_{S1}	S_S half-saturation coefficient for aerobic HB activity	15	$g\ COD \cdot m^{-3}$
K_{OH1}	O_2 half saturation coefficient for aerobic HB activity	0.2	$g\ (-COD) \cdot m^{-3}$
b_H	decay coefficient of HB	0.3	d^{-1}
HB denitrification			
n_Y	anoxic reduction factor for HB yield	0.9	[-]
$K_{I3,NO}$	NO inhibition constant of for HB-mediated NO_2^- reduction	0.5	$g\ N \cdot m^{-3}$
$K_{I4,NO}$	NO inhibition constant of for HB-mediated NO reduction	0.3	$g\ N \cdot m^{-3}$
$K_{I5,NO}$	NO inhibition constant of for HB-mediated N_2O reduction	0.2	$g\ N \cdot m^{-3}$
K_{N2O}	N_2O half-saturation for HB-mediated N_2O reduction	0.02	$g\ N \cdot m^{-3}$
K_{NO}	NO half-saturation for HB-mediated NO reduction	0.04	$g\ N \cdot m^{-3}$
K_{NO2}	NO_2^- half-saturation for HB-mediated NO_2^- reduction	0.3	$g\ N \cdot m^{-3}$
K_{NO3}	NO_3^- half-saturation for HB-mediated NO_3^- reduction	1.5	$g\ N \cdot m^{-3}$
K_{S2}	S_S inhibition coefficient for HB-mediated NO_3^- reduction	20	$g\ COD \cdot m^{-3}$
K_{S3}	S_S inhibition coefficient for HB-mediated NO_3^- reduction	20	$g\ COD \cdot m^{-3}$
K_{S4}	S_S inhibition coefficient for HB-mediated NO reduction	20	$g\ COD \cdot m^{-3}$
n_{g2}	reduction factor for HB anoxic growth on NO_3^-	0.3	[-]
n_{g3}	reduction factor for HB anoxic growth on NO_2^-	0.3	[-]
n_{g4}	reduction factor for HB anoxic growth on NO	0.6	[-]
n_{g5}	reduction factor for HB anoxic growth on N_2O	0.8	[-]
K_{S5}	S_S inhibition coefficient for HB-mediated N_2O reduction	30	$g\ COD \cdot m^{-3}$
K_{OH2}	O_2 inhibition coefficient for HB-mediated NO_3^- reduction	0.2	$g\ (-COD) \cdot m^{-3}$
K_{OH3}	O_2 inhibition coefficient for HB-mediated NO_2^- reduction	0.2	$g\ (-COD) \cdot m^{-3}$
K_{OH4}	O_2 inhibition coefficient for HB-mediated NO reduction	0.2	$g\ (-COD) \cdot m^{-3}$
K_{OH5}	O_2 inhibition coefficient for HB-mediated N_2O reduction	0.2	$g\ (-COD) \cdot m^{-3}$
Hydrolysis of particulate organics			
k_h	maximum specific hydrolysis rate	2.89	$g\ COD \cdot g^{-1}\ COD_{BIO}$
n_h	reduction factor for hydrolysis	0.8	[-]
K_{OH}	O_2 half-saturation coefficient for hydrolysis	0.2	$g\ (-COD) \cdot m^{-3}$
K_X	Half-saturation coefficient for hydrolysis of X_S	0.1	$g\ COD \cdot m^{-3}$
Ammonification			
k_a	Rate constant for ammonification	0.07	$m^3 \cdot g^{-1}\ COD_{BIO} \cdot d^{-1}$
AOB denitrification			
$K_{SNO,AOB}$	NO half saturation coefficient for AOB-mediated NO reduction	1	$g\ N \cdot m^{-3}$
$K_{SO,AOBden1}$	O_2 half saturation coefficient for AOB-mediated NO_2^- reduction	11.4	$g\ (-COD) \cdot m^{-3}$
$K_{IO,AOBden1}$	O_2 inhibition coefficient for AOB-mediated NO_2^- reduction	0.0351	$g\ (-COD) \cdot m^{-3}$
$K_{SO,AOBden2}$	O_2 half saturation coefficient for AOB-mediated NO reduction	11.4	$g\ (-COD) \cdot m^{-3}$
$K_{IO,AOBden2}$	O_2 inhibition coefficient for AOB-mediated NO reduction	0.0351	$g\ (-COD) \cdot m^{-3}$
n_{AOB}	reduction factor for AOB growth on NO_2^-/NO	0.5	[-]
$K_{FNA,AOB}$	HNO_2 half saturation coefficient for AOB-mediated NO_2^- reduction	6-10-4	$g\ N \cdot m^{-3}$
$K_{FA,AOB}$	NH_3 half saturation coefficient for AOB-mediated NO_2^- and NO reduction	0.0027	$g\ N \cdot m^{-3}$
AnAOB activity			
Y_{AnAOB}	growth yield of AnAOB	0.16	$g\ COD_{BIO} \cdot g^{-1}\ N$
μ_{AnAOB}	maximum specific growth rate of AnAOB	0.0173	d^{-1}
$K_{NH3,AnAOB}$	NH_3 half saturation coefficient for AnAOB activity	0.0012	$g\ N \cdot m^{-3}$
$K_{HNO2,AnAOB}$	HNO_2 half saturation coefficient for AnAOB activity	$2.81 \cdot 10^{-6}$	$g\ N \cdot m^{-3}$

$K_{O_2,AnAOB}$	O_2 inhibition coefficient for AnAOB activity	0.01	$g (-COD).m^{-3}$
b_{AnAOB}	decay coefficient of AnAOB	$6.19 \cdot 10^{-4}$	d^{-1}
Other parameters			
f_P	fraction of X_P generated from biomass decay	0.08	$g COD_{XP}.g^{-1} COD_{BIO}$
i_{XB}	N content in biomass	0.086	$g N.g^{-1} (COD)$
i_{XP}	N content in X_P	0.06	$g N.g^{-1} (COD)$

2. Model for mainstream activated sludge processes in the BSM2Nb

Table 5: (a) Stoichiometric matrix and (b) kinetic vector for AOB-mediated processes.

(a) Stoichiometric matrix							
	S_{NHX}	S_{NH_2OH}	S_{O_2}	S_{NO_2}	S_{NO}	S_{N_2O}	X_{AOB}
Aerobic oxidation of NH_4^+ into NH_2OH	-1	1	-1.14				
Aerobic growth of AOB	$-i_{NXB}$	$-\frac{1}{Y_{AOB}}$	$-\frac{1.71 - Y_{AOB}}{Y_{AOB}}$		$\frac{1}{Y_{AOB}}$		1
Aerobic oxidation of NO into NO_2^-			-0.571	1	-1		
N_2O production by Incomplete NH_2OH oxidation		-1		1	-4	4	
N_2O production due to AOB denitrification		-1		-1		2	

(b) Kinetic vector	
Aerobic oxidation of NH_4^+ into NH_2OH	$\frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \frac{S_{O_2}}{S_{O_2} + K_{O_2,AOBAMO}} \cdot \frac{S_{FA}}{S_{FA} + K_{FA,AOB}} \cdot X_{AOB}$
Aerobic growth of AOB	$\mu_{AOB,HAO} \cdot \frac{S_{O_2}}{S_{O_2} + K_{O_2,AOBHAO}} \cdot \frac{S_{NH_2OH}}{S_{NH_2OH} + K_{NH_2OH,AOB}} \cdot X_{AOB}$
Aerobic oxidation of NO into NO_2^-	$\frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \frac{S_{O_2}}{S_{O_2} + K_{O_2,AOBHAO}} \cdot \frac{S_{NO}}{S_{NO} + K_{NO,AOBHAO}} \cdot X_{AOB}$
N_2O production via Incomplete NH_2OH Oxidation	$\frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \eta_{NN} \cdot \frac{S_{NH_2OH}}{S_{NH_2OH} + K_{NH_2OH,AOB}} \cdot \frac{S_{NO}}{S_{NO} + K_{NO,AOBNN}} \cdot X_{AOB}$
N_2O production via AOB denitrification	$\frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \eta_{ND} \cdot \frac{S_{NH_2OH}}{S_{NH_2OH} + K_{NH_2OH,AOB}} \cdot \frac{S_{FNA}}{S_{FNA} + K_{FNA,AOB}} \cdot DO_{HaldaneFunc}^* \cdot X_{AOB}$

$$DO_{HaldaneFunc}^* = \frac{S_{O_2}}{K_{SO_2,AOBden} + (1 - 2 \cdot \sqrt{K_{SO_2,AOBden}/K_{IO,AOBden}}) \cdot S_{O_2} + S_{O_2}^2/K_{IO,AOBden}}$$

Table 6: Parameter values used in the BSM2Nb for the biological processes.

Y_{AOB}	growth yield of AOB	0.18	$g\ COD_{BIO} \cdot g^{-1}\ N$
$\mu_{AOB,HAO}$	maximum specific growth rate of AOB	0.58	d^{-1}
$i_{N,XB}$	N content of the biomass	0.086	$g\ N \cdot g^{-1}\ COD$
η_{ND}	Reduction factor for AOB denitrification rate	0.25	-
η_{NN}	Reduction factor for the incomplete hydroxylamine oxidation rate	0.0015	-
$K_{FA,AOB}$	Free ammonia affinity constant for aerobic AOB activity	0.003	$g\ N \cdot m^{-3}$
$K_{NH2OH,AOB}$	Hydroxylamine affinity constant	0.9	$g\ N \cdot m^{-3}$
$K_{FNA,AOB}$	Free nitrous acid affinity constant for AOB denitrification	0.178	$g\ N \cdot m^{-3}$
$K_{NO,AOB,HAO}$	Nitric oxide affinity constant for aerobic AOB activity	$3 \cdot 10^{-4}$	$g\ N \cdot m^{-3}$
$K_{NO,AOB,NN}$	Nitric oxide affinity constant for incomplete NH_2OH oxidation	0.008	$g\ N \cdot m^{-3}$
$K_{IO,AOBden}$	Oxygen inhibition constant for AOB denitrification	0.8	$g\ (-COD) \cdot m^{-3}$
$K_{SO2,AOBden}$	Oxygen affinity constant for AOB denitrification	0.5	$g\ (-COD) \cdot m^{-3}$
$K_{O2,AOBAMO}$	Oxygen affinity constant for aerobic oxidation of ammonium to hydroxylamine	1	$g\ (-COD) \cdot m^{-3}$
$K_{O2,AOBAMO}$	Oxygen affinity constant for aerobic oxidation of hydroxylamine to nitrite	0.6	$g\ (-COD) \cdot m^{-3}$

For the NOB-mediated processes and for the HB-mediated processes, the model structure and the parameters are the ones used for the BSM2Na.

3. Model for mainstream activated sludge processes in the BSM2Nc

Table 7: Petersen matrix of the AOB-mediated processes.

<i>Stoichiometric matrix</i>							
	S_{NHx}	S_{NH_2OH}	S_{O_2}	S_{NO_2}	S_{NO}	S_{N_2O}	X_{AOB}
<i>Aerobic oxidation of NH_x into NH_2OH</i>	-1	1	-1.14				
<i>Aerobic growth of AOB from NH_2OH to NO_2^-</i>	$-i_{NXB}$	$-\frac{1}{Y_{AOB}}$	$-\frac{2.29 - Y_{AOB}}{Y_{AOB}}$	$\frac{1}{Y_{AOB}}$			1
<i>Aerobic grow of AOB from NH_2OH to NO</i>	$-i_{NXB}$	$-\frac{1}{Y_{AOB}}$	$-\frac{0.57 - Y_{AOB}}{Y_{AOB}}$		$\frac{1}{Y_{AOB}}$		1
<i>AOB denitrification on NO_2^-</i>		-1		-3	4		
<i>AOB denitrification on NO</i>		-1		1	-4	4	

<i>Kinetic vector</i>	
<i>Aerobic oxidation of NH_x into NH_2OH</i>	$\mu_{AOBAMO} \cdot \frac{S_{O_2}}{S_{O_2} + K_{O2,AOBAMO}} \cdot \frac{S_{NH}}{S_{NH} + K_{NH,AOB}} \cdot X_{AOB}$
<i>Aerobic growth of AOB from NH_2OH to NO_2^-</i>	$\mu_{AOBHAO} \cdot (1 - \eta_{NN}) \cdot \frac{S_{O_2}}{S_{O_2} + K_{O2,AOBHAO}} \cdot \frac{S_{NH_2OH}}{S_{NH_2OH} + K_{NH_2OH,AOB}} \cdot X_{AOB}$
<i>Aerobic grow of AOB from NH_2OH to NO</i>	$\mu_{AOBHAO} \cdot \eta_{NN} \cdot \frac{S_{O_2}}{S_{O_2} + K_{O2,AOBHAO}} \cdot \frac{S_{NH_2OH}}{S_{NH_2OH} + K_{NH_2OH,AOB}} \cdot X_{AOB}$

<i>AOB denitrification on NO₂⁻</i>	$\mu_{\text{AOBHAO}} \cdot \eta_{\text{ND}} \cdot \frac{K_{\text{IO,AOBden}}}{K_{\text{IO,AOBden}} + S_{\text{O}_2}} \cdot \frac{S_{\text{NH}_2\text{OH}}}{S_{\text{NH}_2\text{OH}} + K_{\text{NH}_2\text{OH,AOB}}} \cdot \frac{S_{\text{NO}_2}}{S_{\text{NO}_2} + K_{\text{NO}_2,\text{AOB}}} \cdot X_{\text{AOB}}$
<i>AOB denitrification on NO</i>	$\mu_{\text{AOBHAO}} \cdot \eta_{\text{ND}} \cdot \frac{S_{\text{NH}_2\text{OH}}}{S_{\text{NH}_2\text{OH}} + K_{\text{NH}_2\text{OH,AOB}}} \cdot \frac{S_{\text{NO}}}{S_{\text{NO}} + K_{\text{NO,AOB}}} \cdot X_{\text{AOB}}$

Table 8: Parameter values used in the BSM2Nc for the biological processes.

Y_{AOB}	growth yield of AOB	0.18	$\text{g COD}_{\text{BIO}} \cdot \text{g}^{-1} \text{N}$
μ_{AOBAMO}	Maximal specific oxidation rate for aerobic oxidation of ammonium to hydroxylamine	4.38	d^{-1}
μ_{AOBHAO}	Maximal specific oxidation rate for hydroxylamine oxidation	2.016	d^{-1}
$i_{\text{N,XB}}$	N content of the biomass	0.086	$\text{g N} \cdot \text{g}^{-1} \text{COD}$
η_{ND}	Reduction factor for AOB denitrification rate	0.56	-
η_{NN}	Reduction factor for the incomplete hydroxylamine oxidation rate	0.0013	-
$K_{\text{NH}_4,\text{AOB}}$	Ammonium affinity constant for AOB	0.15	$\text{g N} \cdot \text{m}^{-3}$
$K_{\text{NH}_2\text{OH},\text{AOB}}$	Hydroxylamine affinity constant for AOB	1	$\text{g N} \cdot \text{m}^{-3}$
$K_{\text{NO}_2,\text{AOB}}$	Nitrite affinity constant for AOB denitrification	0.8	$\text{g N} \cdot \text{m}^{-3}$
$K_{\text{NO},\text{AOB}}$	Nitric oxide affinity constant for AOB denitrification	0.02	$\text{g N} \cdot \text{m}^{-3}$
$K_{\text{O}_2,\text{AOBAMO}}$	Oxygen affinity constant for aerobic oxidation of ammonium to hydroxylamine	0.4	$\text{g (-COD)} \cdot \text{m}^{-3}$
$K_{\text{O}_2,\text{AOBHAO}}$	Oxygen affinity constant for aerobic oxidation of hydroxylamine to nitrite	0.073	$\text{g (-COD)} \cdot \text{m}^{-3}$
$K_{\text{IO,AOBden}}$	Oxygen inhibition constant for AOB denitrification	0.15	$\text{g (-COD)} \cdot \text{m}^{-3}$

For the NOB-mediated processes and for the HB-mediated processes, the model structure and the parameters are the ones used for the BSM2Na.

APPENDIX A4

Considering that the sample time used is equal to 15 minutes, and that the evaluation starts from day 244 to 609, the performance criteria used to evaluate the control strategies are calculated in a discretized procedure as follows:

- R_{NatAmm} :

$$R_{NatAmm} = \frac{1}{35041} \cdot \sum_{i=23425}^{58465} \frac{NO_3^-|_{OUT,i} - NO_3^-|_{IN,i}}{NH_4^+|_{IN,i} - NH_4^+|_{OUT,i}}$$

- Integral Absolute Error for R_{NatAmm} :

$$IAE_{R_{NatAmm}} = \sum_{i=23425}^{58465} |R_{NatAmmSP,i} - R_{NatAmm,i}|$$

- Integral Square Error for R_{NatAmm} :

$$ISE_{R_{NatAmm}} = \sum_{i=23425}^{58465} (R_{NatAmmSP,i} - R_{NatAmm,i})^2$$

- Total Variation (TV) of $k_L a$, calculated by summing the TV of the three $k_L a$ in the aerobic zone :

$$TV_{k_L a} = \sum_{i=23425}^{58465} |k_L a_{i+1} - k_L a_i|$$

- N_2O emission factor, calculated per unit of influent TKN [$g N_2O-N \cdot g^{-1} TKN_{in}$]:

$$N_2O_{ef1} = \frac{\sum_{i=23425}^{58465} (N_2O_{ANOX1,i} + N_2O_{ANOX2,i} + N_2O_{AER1,i} + N_2O_{AER2,i} + N_2O_{AER3,i})}{\sum_{i=23425}^{58465} ((S_{NHin,i} + S_{NDin,i} + X_{NDin,i}) \cdot Q_{in,i})}$$

- N_2O emission factor, calculated per unit of removed TKN [$g N_2O-N \cdot g^{-1} TKN_{rem}$],

$$N_2O_{ef2} = \frac{\sum_{i=23425}^{58465} (N_2O_{ANOX1,i} + N_2O_{ANOX2,i} + N_2O_{AER1,i} + N_2O_{AER2,i} + N_2O_{AER3,i})}{\sum_{i=23425}^{58465} ((S_{NHin,i} + S_{NDin,i} + X_{NDin,i}) \cdot Q_{in,i}) - \sum_{i=23425}^{58465} ((S_{NHout,i} + S_{NDout,i} + X_{NDout,i}) \cdot Q_{out,i})}$$

- N_2O produced according to HB denitrification in the aerobic zone (third HB denitrification step) [$g N_2O-N \cdot d^{-1}$]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(\sum_{j=1}^3 \left(\frac{1 - Y_H \cdot n_Y}{0.571 \cdot Y_H \cdot n_Y} \right) \cdot \mu_{HB,T} \cdot \eta_{AnoxHB,4} \cdot \frac{S_{S,j}}{S_{S,j} + K_{S4}} \cdot \frac{K_{OH4}}{S_{O2,j} + K_{OH4}} \cdot \frac{S_{NO,j}}{S_{NO,j} + K_{NO} + S_{NO,j}^2 / K_{I4NO}} \cdot X_{HB,j} \cdot V_j \right)_i$$

- N_2O consumed according to HB denitrification in the aerobic zone (fourth HB denitrification step) [g N_2O -N.d⁻¹]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(\sum_{j=1}^3 \left(\frac{1 - Y_H \cdot n_Y}{0.571 \cdot Y_H \cdot n_Y} \right) \cdot \mu_{HB,T} \cdot \eta_{AnoxHB,5} \cdot \frac{S_{S,j}}{S_{S,j} + K_{S5}} \cdot \frac{K_{OH5}}{S_{O2,j} + K_{OH5}} \cdot \frac{S_{N2O,j}}{S_{N2O,j} + K_{N2O}} \cdot \frac{K_{I5,NO}}{K_{I5,NO} + S_{NO,j}} \cdot X_{HB,j} \cdot V_j \right)_i$$

- N_2O produced according to AOB denitrification in the BSM2Na [g N_2O -N.d⁻¹]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(\sum_{j=1}^3 \frac{2}{Y_{AOB}} \cdot \mu_{AOB,T} \cdot \eta_{AnoxAOB} \cdot DO_{HaldaneFunc,j}^* \cdot \frac{S_{FA,j}}{S_{FA,j} + K_{FA,AOB}} \cdot \frac{S_{NO,j}}{S_{NO,j} + K_{NO,AOB}} \cdot X_{AOB,j} \cdot V_j \right)_i$$

- N_2O produced according to AOB denitrification in the BSM2Nb [g N_2O -N.d⁻¹]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(2 \cdot \frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \eta_{ND} \cdot \frac{S_{NH2OH,j}}{S_{NH2OH,j} + K_{NH2OH,AOB}} \cdot \frac{S_{FNA,j}}{S_{FNA,j} + K_{FNA,AOB}} \cdot DO_{HaldaneFunc,j}^* \cdot X_{AOB,j} \cdot V_j \right)_i$$

- N_2O produced by AOB in the BSM2Nc [g N_2O -N.d⁻¹]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(4 \cdot \mu_{AOBHAO} \cdot \eta_{ND} \cdot \frac{S_{NH2OH,j}}{S_{NH2OH,j} + K_{NH2OH,AOB}} \cdot \frac{S_{NO,j}}{S_{NO,j} + K_{NO,AOB}} \cdot X_{AOB,j} \cdot V_j \right)_i$$

- N_2O produced according to incomplete NH_2OH oxidation in the BSM2Nb [g N_2O -N.d⁻¹]:

$$\frac{1}{35041} \cdot \sum_{i=23425}^{58465} \left(4 \cdot \frac{\mu_{AOB,HAO}}{Y_{AOB}} \cdot \eta_{NN} \cdot \frac{S_{NH2OH}}{S_{NH2OH} + K_{NH2OH,AOB}} \cdot \frac{S_{NO}}{S_{NO} + K_{NO,AOBNN}} \cdot X_{AOB} \cdot V_j \right)_i$$

- TKN removal efficiency :

$$\eta_{TKN} = \frac{\overline{TKN}_{in} - \overline{TKN}_{out}}{\overline{TKN}_{in}}$$

- TN removal efficiency (η_{TN}) :

$$\eta_{TN} = \frac{\overline{TN}_{in} - \overline{TN}_{out}}{\overline{TN}_{in}}$$

- Effluent nitrate [g N.m⁻³]:

$$\overline{[NO_3^-]_{eff}} = \frac{\sum_{i=23425}^{58465} ([NO_3^-]_{eff})_i \cdot Q_{eff,i} \cdot t_i}{\sum_{i=23425}^{58465} Q_{eff,i}}$$

- Percentage of total time when effluent ammonium violations occur :

$$V_{NH} = \frac{\text{number of times}([NH_4^+]_{eff} > [NH_4^+]_{lim}) \cdot \text{sampletime}}{t(i = 58465) - t(i = 23425)}$$

- Percentage of total time when effluent total nitrogen violations occur :

$$V_{TN} = \frac{\text{number of times}([TN_{eff}] > [TN]_{lim}) \cdot \text{sampletime}}{t(i = 58465) - t(i = 23425)}$$

- Effluent quality index [kg pollutant.units.d⁻¹] :

$$EQI = \frac{\sum_{i=23425}^{58465} ((72 \cdot TSS_e(i) + COD_e(i) + 20 \cdot TKN_e(i) + 20 \cdot NO_{3,e}^-(i) + 2 \cdot BOD_{5,e}(i)) \cdot t_i)}{(t(i = 58465) - t(i = 23425)) \cdot 1000}$$

- Average aeration energy consumed [kWh.d⁻¹] :

$$AEC = \frac{\int_{t_{start}}^{t_{end}} \sum_{i=1}^5 V_i \cdot k_L a_i(t) \cdot (S_{O2,sat} - S_{O2,i}(t)) \cdot dt}{1.8 \cdot 1000 \cdot (t_{end} - t_{start})}$$

APPENDIX A5

Proportional integral ammonium cascade controller

APPENDIX A6

Figure A6.1: Ammonium concentration inlet to the first aerobic tank (AER1): actual value (in red) and with measurement noise (in blue).

Figure A6.2: Nitrate concentration inlet to the first aerobic tank (AER1): actual value (in red) and with measurement noise (in blue).

Figure A6.3: Ammonium concentration outlet to the first aerobic tank (AER1): actual value (in red) and with measurement noise (in blue).

Figure A6.4: Nitrate concentration outlet to the first aerobic tank (AER1): actual value (in red) and with measurement noise (in blue).

Figure A6.5: Ratio between nitrate produced and ammonium removed in the aerobic zone: actual value (in red) and with measurement noise (in blue).

Scenario↓	net N ₂ O prod. by AOB and HB in aerobic zone	net N ₂ O prod. by HB in anoxic zone	[NH ₄ ⁺] _{eff}
1	LOW	LOW	HIGH
2	HIGH	LOW	LOW
3	LOW	HIGH	LOW
4	HIGH	LOW	HIGH
5	LOW	HIGH	HIGH

TEMPERATURE [°C]	$R_{\text{NatAmm,SP}}$
10	1.1
15	1.2
20	1.4

		E_RNatAmm		
		LOW	GOOD	HIGH
(NH₄⁺)_{eff}	GOOD	<i>N</i>	<i>Z</i>	<i>P</i>
	HIGH	<i>P</i>	<i>P</i>	<i>P</i>

Legend: P=POSITIVE, N=NEGATIVE, Z= ZERO.

IF		THEN	
TEMPERATURE		$R_{\text{NatAmm,SP}}$	
VERY LOW		VERY LOW	
LOW		LOW	
MEDIUM		MEDIUM	
HIGH		HIGH	
VERY HIGH		VERY HIGH	

		CS1			CS2			CS3			CS4		
BSM2N →		a	b	c	a	b	c	a	b	c	a	b	c
R_{NatAmm}	$\frac{\text{g NO}_3^- - \text{N}}{\text{g NH}_4^+ - \text{N}}$	1.2	1.265	1.17	1.26	1.28	1.24	1.27	1.28	1.26	1.27	1.28	1.256
$IAE_{R\text{NatAmm}}$	[-]	79.3	83.5	92.97	73.84	77.56	84.23	73.62	76.82	83	74.06	77.16	85.1
$ISE_{R\text{NatAmm}}$	[-]	26.84	29.71	35.84	24	26.19	30.74	23.68	25.51	29.78	23.94	25.65	31.21
TV	[d ⁻¹]	2733.4	2909.8	2779.8	1543.2	1685.4	1681.9	3021.2	2985.3	3331.8	5668.5	5688.2	6309.7
N_2O_{eff1}	$\% \frac{\text{g N}_2\text{O} - N_{\text{gas}}}{\text{g TKN} - N_{\text{in}}}$	0.16	0.0176	0.167	0.066	0.0248	0.067	0.045	0.0208	0.047	0.047	0.0226	0.049
N_2O_{eff2}	$\% \frac{\text{g N}_2\text{O} - N_{\text{gas}}}{\text{g TKN} - N_{\text{rem}}}$	0.166	0.0184	0.172	0.068	0.026	0.069	0.047	0.0217	0.048	0.049	0.0234	0.051
N_2O produced by HB	$\frac{\text{g N}_2\text{O} - N_{\text{liq}}}{d}$	-2446	-785.3	-36377	-655.3	-607.5	-10589	-825.4	-640.3	-4408	-659.5	-609.7	-5976
N_2O produced from AOB denitrification	$\frac{\text{g N}_2\text{O} - N_{\text{liq}}}{d}$	3554.2	9.4	36881	725.9	5.7	10945	673.3	4.18	5477.3	522.3	3.36	7528.4
N_2O produced from incomplete NH_2OH oxidation	$\frac{\text{g N}_2\text{O} - N_{\text{liq}}}{d}$	/	371.2		/	269.82		/	252.1		/	236.53	

		CS1			CS2			CS3			CS4		
BSM2N →		a	b	c	a	b	c	a	b	c	a	b	c
η_{TKN}	$\% \frac{\text{g TKN}_{\text{rem}}}{\text{g TKN}_{\text{in}}}$	96.1	95.6	96.96	96.30	95.70	96.80	96.35	95.90	97.14	96.50	96.24	97.00
η_{TN}	$\% \frac{\text{g TN}_{\text{rem}}}{\text{g TN}_{\text{in}}}$	69.8	67.3	76.2	67.30	67.00	74.32	66.76	66.73	73.63	66.64	66.88	74.40
NO_3^-	$\text{g N} \cdot \text{m}^{-3}$	13.07	14.1	10.32	14.42	14.18	11.19	14.72	14.52	11.70	14.84	14.61	11.24
V_{NH}	% of operating time	0.054	0.11	0.077	1.71	4.60	1.54	0.58	2.44	0.38	0.40	0.95	0.44
V_{TN}	% of operating time	22.8	30.76	1.24	29.94	35.70	1.45	35.13	36.84	2.05	36.30	34.63	1.30
EQI	$\text{kg pollutant} \cdot \text{units} \cdot \text{d}^{-1}$	5713.8	6069.1	2864.9	5928.3	6081.4	5087	5967	6060.04	5089.0	5957.2	5979.1	5042.9

AEC [kWh.d ⁻¹]				
	CS1	CS2	CS3	CS4
BSM2Na	2935.1	3276.7	3133.6	3172.6
BSM2Nb	2827.139	3194.62	3068.3	3129.95
BSM2Nc	2930.2025	3209.53	3167.38	3134.72

	Oxygen affinity values for AOB/NOB		Controller output and performance			REFERENCE
scenario	$K_{O_2,AOB}$	$K_{O_2,NOB}$	N_2O_{ef1}	N_2O_{ef2}	R_{NatAmm}	
	$g (-COD).m^{-3}$	$g (-COD).m^{-3}$	$[\% g N_2O-N.g^{-1} N_{in}]$	$[\% g N_2O-N.g^{-1} N_{rem}]$	$[-]$	
<i>CSI</i>	0.25	1.84	7.71	7.96	0.74	Peeters et al. (1969)
<i>CS3</i>			0.569	0.586	1.25	
<i>CSI</i>	0.5	2	6.2	6.42	0.72	Laudelout et al. (1974)
<i>CS3</i>			0.364	0.377	1.25	
<i>CSI</i>	0.18	0.13	0.0131	0.0135	1.28	Manser et al. (2005)
<i>CS3</i>			0.0276	0.0284	1.28	
<i>CSI</i>	0.033	0.43	0.0723	0.0744	1.27	Blackburne et al. (2008)
<i>CS3</i>			0.0379	0.0381	1.29	
<i>CSI</i>	0.75	1.75	0.974	1.017	1.05	Guisasola et al. (2005)
<i>CS3</i>			0.089	0.092	1.26	
<i>CSI</i>	0.6	1.3	0.26	0.27	1.18	Wiesmann (1994)
<i>CS3</i>			0.057	0.06	1.27	
<i>CSI</i>	0.15	0.11	0.0129	0.0133	1.27	Picioreanu et al. (2016)
<i>CS3</i>			0.0274	0.0282	1.28	
<i>CSI</i>	0.36	0.16	0.0124	0.0129	1.27	Picioreanu et al. (2016)
<i>CS3</i>			0.0275	0.0285	1.27