Minimizing membrane bioreactor environmental footprint by multiple objective optimization

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Abstract

An integrated model for membrane bioreactors (MBR) was employed in view of the management optimization of an MBR biological nutrient removal (BNR) pilot plant in terms of operational costs and direct greenhouse gases emissions. The influence of the operational parameters (OPs) on performance indicators (PIs) was investigated by adopting the Extended-FAST sensitivity analysis method. Further, a multi-objective analysis was performed by applying the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS). The results show-up that the sludge retention time is the OP mostly affecting all the investigated PIs. By applying the set of optimal OPs, there was a reduction of 48% and 10% of the operational costs and direct emissions, respectively.

Keywords: Mathematical modelling optimization, multi-objective analysis, wastewater treatment plant, greenhouse gases, membrane fouling.

1. Introduction

The aim of this paper is to describe methods to minimize the environmental footprint for membrane bioreactors (MBR). Several parameters will influence the footprint, such as effluent quality, operational cost, energy consumption and greenhouse gas (GHG) emissions. Consequently, the minimization has to be addressed by using multicriteria optimization, where the various influencing factors can be weighted in different ways. The aim of the study is to obtain operating strategies that will reduce the environmental footprint.

Wastewater treatment plants (WWTP) are focusing new challenges and are moving towards new frontiers which include complying with increased wastewater discharge standards, reducing greenhouse gas (GHG) emissions, minimizing operational and capital cost for the treatment facilities, increasing effective energy management, using more compact systems and reducing the WWTP footprint (Sweetapple et al., 2014; Bozkurt et al., 2016). Indeed, WWTPs are shifting from being ”end-of-the-pipe” solutions to resource recovery sites (Puyol et al., 2016). GHG emissions are mainly generated in the biological processes, some of them occurring from the process reactions (direct emissions – DE), and others from electricity consumption (indirect emissions - IE) (IPCC, 2013). The main emitted GHGs are carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) (Mannina et al., 2016). Among the GHGs, N_2O is of special interest due to its great global warming potential (GWP) and the high capacity to deplete the ozone layer (IPCC et al., 2007; Mannina et al., 2018). An accurate quantification of GHG emissions is an important step to reduce process footprint. Plant land occupation is an essential factor to consider, and it is suggested that membrane bioreactors (MBR) adoption as a viable solution to meet lower effluent demands and reduced space requirements (Judd, 2010; Atasanova et al., 2017). Mathematical models are powerful tools to quantify GHG emissions, comparing different WWTP design and operational strategies (Mannina et al., 2016). Several mathematical models have been proposed in literature for accounting GHG emissions ranging from empirical simplified to mechanistic approaches (Pocquet et al., 2016; Spérandio et al., 2016).

However, only few mathematical models quantify GHG emission from MBRs (Mannina et al., 2018). MBRs differ from conventional activated sludge systems (CASs) and CAS results cannot easily be translated to MBR operations (Judd, 2010; Mannina et al., 2018). Consequently, MBR footprint optimization requires dedicated studies. As an illustration of the multiple criteria problem, a reduction of the airflow rate in the aerobic reactor, for minimizing the energy consumption, may increase the N_2O emissions because of incomplete nitrification (Flores-Alsina et al., 2014). The identification of the interrelationship between operational conditions and direct (i.e., N_2O, CH_4 and CO_2) and indirect (i.e., energy consumption) GHG emissions represents a key issue in reducing the environmental footprint (Mannina et al., 2017). Multi-objective optimization is aiming to cope with competing criteria that will influence the footprint. Such a tool can help decision makers on obtaining a deeper perception of necessary trade-offs between conflicting operational strategies (Sweetapple et al., 2014; Wang and Rangaiah, 2017). Maere et al. (2011) compared several control and operational strategies to optimize MBR operation. Authors found effective results for the MBR operation optimization by employing closed loop aeration (based on a fixed dissolved oxygen concentration inside the aerobic reactor) rather than open loop. Indeed, by implementing the closed loop aeration, a reduction of the operational costs by 13-17% was obtained (Maere et al., 2011). Despite useful insights gained by Maere et al. (2011), the results were obtained using an ideal membrane (i.e., neglecting the interplay between physical and biological processes). Therefore, the results may not be directly applicable to full scale MBR systems. Sweetapple et al. (2014) presented a study on a multi-objective optimisation, for a CAS system, taking into account GHGs, effluent quality and operational costs. Different problem formulations were explored to identify the most effective approach and the optimal set of parameters for plant operation. Main conclusions were that GHG emissions could be substantially reduced without increasing operational costs (Sweetapple et al., 2014). Another multi-objective optimization for a CAS system was carried out by Long et al. (2019). The authors applied Monte Carlo simulations to optimize costs and reduce pollution from an industrial WWTP. Their study was applied for pre-treatment, centralized and reclaimed wastewater facilities and the results showed how

pollutant level and operational costs were related. Their results confirmed the importance to apply multiple objectives to balance costs and pollution. The fact that many criteria, such as energy reduction, membrane fouling, and GHG emission, are influenced in different directions, which motivate the use of multiple criteria optimization. This has been clearly demonstrated for CAS systems (see e.g., Flores-Alsina et al., 2014). Even if multi-objective optimization has been applied for CAS system, there are no studies presented for MBR system, to the authors' knowledge, whereas minimizing MBR environmental footprint considering multiple objectives is highly desired.

In this paper an integrated MBR mathematical model was adapted to a University of Cape Town (UCT)-MBR pilot-plant (Mannina et al., 2018). The influence of five operational parameters on ten performance indicators has been explored. Multi-objective optimization analysis has been used to find the trade-off between plant performance and cost.

2. Material and methods

2.1 Mathematical model description and application

The MBR integrated model described in (Mannina et al., 2018) is applied here. The model consists of biological (Mannina et al., 2018) and physical (Mannina et al., 2011) sub-models. The biological sub-model is described by 116 parameters and 25 state variables. The model includes nitrogen transformation considering two- step nitrification and four-step denitrification processes (Pocquet et al., 2016; Hiatt and Grady, 2008).

In the first nitrification step, the model considers the ammonia (NH_4) oxidation into nitrite (NO_2) by means of ammonia-oxidizing bacteria (AOB). The second step describes oxidation of NO_2 into NO_3 by means of nitrite-oxidizing bacteria (NOB). In the first step incomplete ammonia oxidation is incorporated. This may lead to the formation of intermediate products, such as hydroxylamine

(NH_2OH) and nitric oxide (NO). Furthermore, incomplete oxidation of NH_2OH into NO_2 with the accumulation of NO, and further reduction into N_2O is also included in the model.

The model takes into account that phosphorus accumulating organisms (PAOs) and heterotrophic non-PAO biomass (OHO) contribute under anoxic conditions to the four-step denitrification. This includes: (i) reduction of NO_3 to NO_2; (ii) reduction of NO_2 to NO; (iii) reduction of NO to N_2O; and (iv) reduction of N_2O to N_2. The incomplete reduction of N_2O into N_2 leading to N_2O accumulation and emission (Mannina et al., 2018) is part of the model.

The biological sub-model evaluates the total GHG emissions (both in terms of N_2O and CO_2) as the sum of direct and indirect emissions.

The physical sub-model is characterized by 6 parameters and 2 state variables. Overall, four key processes occurring during the membrane physical filtration are taken into account (Mannina et al., 2011): (i) cake layer formation during the filtration and backwashing phases; (ii) partial organic matter removal in the cake layer; (iii) chemical oxygen demand (COD) removal due to the physical retention effect of the membrane as a barrier (pre-filter effect); and (iv) membrane fouling.

Biological and physical sub-models are highly interrelated as a result of total suspended solid (TSS) and soluble microbial products (SMP) interactions. Further details regarding the MBR integrated model can be found in Mannina et al., (2011, 2018).

The model has been applied to a UCT- MBR pilot plant, consisting of anaerobic (62 L), anoxic (102 L) and aerobic (221 L) reactors in series. The solid-liquid separation phase was accomplished by an ultrafiltration hollow fiber membrane module (PURON® Triple Bundle Demo Module with a nominal pore size of 0.03 µm and a membrane area of 1.4 m^2) located inside the aerated MBR reactor (Mannina et al., 2018). An oxygen depletion reactor (ODR) was installed between the MBR and the anoxic reactors to reduce the amount of oxygen recycled with the flow rate (Q_{RAS} = 80 L h^{-1}). For a more detailed description of the pilot plant we refer to Mannina et al. (2018).

2.2 Sensitivity Analysis

Sensitivity analysis has been applied to evaluate the model accuracy and calibration. The Extended-FAST (E-FAST) method (Saltelli et al., 2004), a widespread method based on the variance decomposition theorem, has been applied. In accordance to the method, two sensitivity indices for each i-th model factor must be calculated: the first-order effect index (S_i) and the total-effect index (S_{Ti}). S_i assesses the contribution of the i-th parameter to the variance of the model output [Var(Y)] without considering the interaction among the model parameters. S_{Ti} is calculated to evaluate the contributions from high order interactions (Jing et al., 2018). Thus, the difference between S_{Ti} and S_i represents the interaction among the model parameters.

The E-FAST method requires n x N_R simulations, where n is the number of parameters and N_R is the number of runs per model parameter and varies from 500 to 1000 (Saltelli et al., 2004).

2.3 Multi-objective optimization method

A major challenge in multi-objective optimization is to find the weights of the various components of the multi-criteria. To define what is "best" is a subjective decision, made by the modeller. For the model optimization, the TOPSIS method has been adopted (Wang and Rangaiah, 2017). This method will select as the optimal solution (among *m* solutions), the one having the smallest Euclidean distance from the ideal solution among *m* solutions (A^+, known as positive - ideal solution) and the largest Euclidean distance from the negative – ideal solution among *m* solutions (A^-).

By selecting the A^+ and A^- solutions the modeller will define the performance indicators adopted as objective function (OF) for the system under study. The ideal solution represents the combination of the best value of OFs. Conversely, the negative - ideal solution represents the combination of the worst value of OFs.

The TOPSIS method is based on the evaluation of a normalized OF (f_{ij}) matrix (F_{ij}), computed by dividing each (f_{ij}) to the square root of the squared sum of all the f_{ij}. Then the F_{ij} is weighed depending on the influence of each OF (f_{ij}).

$$F_{ij} = \frac{f_{ij}}{\sqrt{\sum_{i=1}^{m} f_{ij}^2}} \qquad [1]$$

where *m* represents the number of solutions for each OF and *n* the number of the OFs.

The TOPSIS procedure consists on 5 -steps (Hwang and Yoon, 1981). In the first step, the normalized objective matrix (F_{ij}) (*m* rows X *n* columns) related to each solution (*i*) of each OF (*j*) (f_{ij}) is composed according to Equation 1. The *m* solutions represent the non-dominated solutions.

In the second step, the normalized objective matrix (v_{ij}) is calculated by multiplying each column of objective matrix (F_{ij}) with its weight (w_j), in accordance to Equation 2.

$$v_{ij} = F_{ij} \, X \, w_j \qquad [2]$$

In the third step, the best and the worst values of each OF (*j*) is evaluated.

Considering the objectives that require to be maximized, the best value (v_j^+) is the largest value within the related columns of matrix v_{ij}. Conversely, for the OFs that have to be minimized, the best value (v_j^+) is the smallest value within the related columns of matrix v_{ij}.

The worst objective value that requires maximization (v_j^-) is the smallest value within the related columns of matrix v_{ij}.For the OFs that have to be minimized, the worst value (i.e., v_j^-) is the largest value within the related columns of matrix v_{ij}.

In the fourth step, the Euclidean distance between each solution and the ideal and negative -ideal solution is calculated.

The distance to positive ideal solution (S_{i+}) is calculated according to Equation 3.

$$S_{i+} = \sqrt{\sum_{j=1}^{n}(v_{ij} - v_j^+)^2} \quad i = 1, 2, 3, \dots, m \qquad [3]$$

Similarly (S_{i-}) is evaluated by Equation 4:

$$S_{i-} = \sqrt{\sum_{j=1}^{n}(v_{ij} - v_j^-)^2} \quad i = 1, 2, 3, \dots, m \qquad [4]$$

In the final step, the closeness of each optimal solution is calculated according to Equation 5.

$$C_i = \frac{S_{i-}}{S_{i-} + S_{i+}} \qquad [5]$$

The solution having the largest C_i represents the optimal solution.

Further applications of the TOPSIS method can be found in (Wang and Rangaiah, 2017).

2.4 Performance Indicators

Ten Performance Indicators (PIs) were considered for the sensitivity and the multi-objective optimization analysis: Operational Costs (OC); Effluent Fine (EF); Effluent Quality Index (EQI) for

both liquid (EQI_{LIQ}) and gas (EQI_{GAS}) flows; oxygen-to-total-Kjeldahl-nitrogen ratio (RON); ratio nitrate-ammonia (R_{NAT}); CO_2 and N_2O emissions; and direct (DE) and indirect (IE) GHG emissions.

The OC (€/treated volume) is calculated by adapting the cost function proposed by Vanrolleghem and Gillot (2002) to the case of MBR. Specifically, the cost is calculated as the sum of three terms (Guerrero et al., 2011): costs related to the chemical consumption for membrane cleaning (CC, as €/ treated volume), energy demand (*e*D, €/) and effluent fine (EF) related to pollutants discharge (in accordance with Italian regulations), as expressed in Equation (6):

$$OC = eD \cdot \gamma_e + CC + EF \qquad [6]$$

where γ_e represents the cost per kWh. Italian rates are 0.21 € / kWh.

The energy demand *e*D (kWh) is calculated as:

$$eD = P_w + P_{eff} + P_s \qquad [7]$$

where P_w, P_{eff} and P_s represent the energy consumption for the air blowers, permeate extraction and the recycle pumps, respectively. P_w and P_{eff} have been calculated according to literature (Mannina & Cosenza, 2013; 2015):

$$P_w = \frac{\mathrm{wRT}}{29.7(0.283)e}\left[\left(\frac{p_2}{p_1}\right)^{0.283} - 1\right] \qquad [8]$$

where P_w [kW] is the power requirement for each blower, w is the mass flow of air [kg s^{-1}], R is the gas constant for air [8.314 kJ $kmol^{-1}$ K^{-1}], T is the absolute temperature [K], p_1 and p_2 are the absolute

inlet and outlet pressures [atm], respectively. The constant 29.7 is a conversion to metric units, 0.283 is a constant for air, *e* is the blower efficiency (common range 0.7–0.9).

The power requirement (in kW) for the permeate extraction pump is

$$P_{eff} = \frac{1}{t_1 - t_0} \int_{t_0}^{t_1} \frac{TMP \cdot Q_{eff}(t)}{3600 \cdot \eta} dt \qquad [9]$$

where, TMP [kPa] is the trans-membrane pressure, Q_{eff} [m^3 h^{-1}] is the effluent flow rate, t_0 and t_1 are the initial and the final times, respectively, of pump operation, and η is the permeate pump efficiency.

The energy consumption for the recycle pumps (P_s) has been calculated as (Metcalf & Eddy, 2003):

$$P_s = \frac{1}{t_1 - t_0} \int 0.04 \cdot (Q_{R1} \cdot 0.06 + Q_{R2} \cdot 0.06 + Q_{WAS} \cdot 0.06) \cdot dt \qquad [10]$$

Where Q_{R1} is the recycled flow rate from the anoxic to anaerobic tank, Q_{R2} the flow rate from the aerobic to MBR tank, and Q_{WAS} the waste sludge flow rate, respectively.

The effluent fine EF has been evaluated according to Mannina & Cosenza (2013; 2015). The membrane cleaning cost CC has been calculated considering a typical membrane cleaning protocol that includes a chemical solution composed of 500 ppm of NaOCl and 2,000 ppm of citric acid, with a cost of 0.48€ per chemical cleaning. For the pilot-plant considered in this work, the CC were activated only when the transmembrane pressure (TMP) reached a value higher than 60kPa. The threshold value of 60kPa is suggested by the membrane manufacturer.

The EQI (kg/treated volume) represents the mass of pollutants discharged throughout the evaluation period (Mannina & Cosenza, 2015). EQI_{LIQ} has been calculated according to:

$$EQI_{LIQ} = \frac{1}{T \cdot * 1000} \int_{t0}^{t1} (\beta_{COD} \cdot COD_e + \beta_{SNH4} \cdot S_{NH4e} + \beta_{SNO3} \cdot S_{NO3e} + \beta_{SN2O} \cdot S_{N2Oe} + \beta_{SPO} \cdot S_{POe}) \cdot Q_{eff}\, dt \qquad [11]$$

where β_i are the weighting factors of the effluent concentrations and are attributed for each single soluble component of the effluent (i = chemical oxygen demand - COD_e, ammonia - S_{NH4e}, nitrate - S_{NO3e}, nitrous oxide - S_{N2Oe} and phosphate - S_{POe}). The following weighting factors were used (Mannina & Cosenza, 2013): β_{COD}=1, β_{NH}=20, β_{NO3}=20, β_{N2O}=50 and β_{PO}=50. *Qeff* is the effluent flow rate, *T* is the simulation period, 1000 is the conversion factor from g m^{-3} to kg m^{-3}, t_0 and t_1 represent the initial and the final simulation time, respectively.

The calculation of EQI was updated to consider gaseous emissions (EQI_{GAS}). Applying the same concept of the EQI_{LIQ}, the EQI_{GAS} has been calculated as:

$$EQI_{GAS} = \frac{1}{T \cdot * 1000} \int_{t0}^{t1} (\beta_{CO2} \cdot Offgas_{CO2} + \beta_{SN2O} \cdot Offgas_{N2O}) \cdot Q_{Offgas}\, dt \qquad [12]$$

where the terms are similar to Equation 11. However, $Offgas_{,CO2}$ and $Offgas_{,N2O}$ describe the gas emitted as CO_2 and N_2O. The β_i values are defined for each GHG (β_{N2O}=50 and β_{CO2}=50), and Q_{offgas} is the gas flow rate. RON indicates the amount of oxygen supplied by the aeration system (i.e., within the aerobic reactor) versus the influent Total Kjeldahl Nitrogen (TKN). The PI allows to quantify the rate of oxygen consumed to oxidize the influent TKN. The PI indicates the aeration regime of the treatment plant consisting on the amount of air supplied to the aerobic reactor (Vangsgaard et al., 2012). RON has been calculated according to (Boiocchi et al., 2017a):

$$RON = \frac{\sum_{i=1}^{n} k_L a_{AER,i} \cdot V_{AER,i} \cdot (SO_{2,SAT,AER,i} - SO_{2,AER,i})}{Q_{in} \cdot S_{NH,in}} \qquad [13]$$

where $k_La_{AER,i}$ is the oxygen mass transfer coefficient of the aerated tank *i*; $V_{AER,i}$ is the volume of the *i*-th aerated tank; $S_{O2,SAT}$ is the oxygen saturation concentration; $S_{O2,AER,i}$ is the DO concentration

inside the *i*-th aerated tank; Q_{IN} is the inlet flow rate fed to the biological zone; and $TKN_{,in}$ is the inlet TKN fed to the biological zone.

R_{NAT} is the ratio between the nitrate produced and ammonia depleted in an aerobic zone and is an indicator of the degree of complete nitrification (Boiocchi et al., 2017b):

$$R_{NAT} = \frac{NO^{-}_{3,OUT,AER} - NO^{-}_{3,IN,AER}}{NH^{+}_{4,IN,AER} - NO^{+}_{4,OUT,AER}} \quad [14]$$

where $S_{NO3,IN,AER}$ and $S_{NO3,OUT,AER}$ represent the influent and effluent NO_3 concentration of the aerobic tank, respectively. $S_{NH4,IN,AER}$ and $S_{NH4,OUT,AER}$ denote the influent and effluent NH_4 concentrations of the aerobic tank, respectively. R_{NAT} indicates the amount of ammonia being oxidized by the AOB and converted into nitrate in the aerobic zone. When all the AOB-produced NO_2 are oxidized by NOB (i.e., forming NO_3) (complete nitrification), R_{NAT} will be equal to one. However, R_{NAT} larger than one is expected as there are additional processes (e.g., biomass decay and additional organic nitrogen release through ammonification) contributing to enhance the organic nitrogen concentrations within the aerobic tank. R_{NAT} indicates if the N_2O production is due to the low AOB activity (Boiocchi et al., 2017b).

The emissions of CO_2 ($kgCO_2 \cdot m^{-3}$) and N_2O ($kgN_2O \cdot m^{-3}$) are evaluated by considering their stripping from the liquid phase to the gas phase according to (Mannina et al., 2018). The total direct emissions (DE, $kgCO_{2,eq}$ m^{-3}) are calculated as the sum of the N_2O and CO_2 emissions. Since N_2O has a GWP 265 times higher than that of CO_2, N_2O emission is multiplied by 265.

Indirect emissions (IE, $kgCO_{2,eq}$ m^{-3}) are calculated multiplying eD by $\gamma_{CO,}$ (equal to 0.245 $kgCO_{2eq}$ /kWh) representing the specific CO_2 emission due to the energy consumption (EIA, 2009).

2.5 Operational parameter values

The E-FAST method (Saltelli et al., 2004) has been applied to assess the influence of the following operational parameters on the PI, with respect to the benchmark scenario: sludge retention time - SRT, air flow rate in the aerobic reactor - $Q_{air,AER}$, air flow rate in the MBR - $Q_{air,MBR}$, the recycle ratio from the anoxic to the anaerobic reactor - R_{QR1}, and the recycle ratio from the aerobic to the anoxic reactor - R_{QR2}.

Table 1 summarizes the value of each operational parameter, its investigated variation range and the references. $Q_{air,MBR}$ was changed according to the manufacturer's suggestion. The minimum value of $Q_{air,AER}$ should allow a dissolved oxygen concentration to exceed 0.5 mg L^{-1} (Metcalf, & Eddy (2003)). The maximum value of $Q_{air,AER}$ has been twice the benchmark scenario one.

<Here Table 1>

The E-FAST method was applied with N_R (=5) simulations per parameter value, and consequently 5,000 model simulations were executed. The purpose was to evaluate the outputs of the modelling application and sensitivity analysis over ten performance indicators, related to the operational costs, energy demand, oxygen consumption, nitrification efficiency, effluent and gas quality and GHG emissions.

The TOPSIS method has been applied considering the ten aforementioned performance indicators as the objective function (OF) to be optimized. All the performance indicators, except R_{NAT} (that was maximized), have been minimized during the TOPSIS method application. The same weight (w_j) (equal to 0.1) has been adopted for all the OFs.

3. Results and Discussion

3.1 Sensitivity Analysis

The values of the first-order effect index (S_i) and the total-effect index (S_{Ti}) and the difference between them are summarized in Table 2.

<Here Table 2>

The sum of each S_i for all ten performance indicators ranged between 0.91 and 0.99. Since the sum of S_i is close to 1, it is reasonable to conclude that the investigated parameters are non-correlated and performance indicators are not additive (Saltelli et al., 2004). Therefore, a few interactions among the investigated parameters are expected. This statement is also confirmed by the sum of S_{Ti}, which is always close to 1.0. This latter result suggests that there is a very low interaction between the parameters.

Figure 1 presents the Extended-FAST results for each performance indicator. The results related to $Q_{air,MBR}$ are not reported in the figure, as it was shown that this parameter has a negligible influence on the PIs (i.e., S_i, S_{Ti} and S_{Ti}-S_i are $<10^{-2}$). Further details regarding $Q_{air,MBR}$ are reported in the following sections.

<Here Figure 1>

Figure 1 demonstrates that SRT has the highest influence on the PIs, with S_i close to 1.0 for $Offgas_{N2O}$, $Offgas_{CO2}$, DE, EQI_{GAS}, EQI_{LIQ} e EF (Figure 1a). SRT influences the results of R_{NAT} and RON, but with a minor intensity with respect to the other indicators (for R_{NAT}, S_i = 0.60 and S_{Ti} = 0.68; for RON, S_i = 0.73 and S_{Ti} = 0.75). $Offgas_{N2O}$ increases with SRT (up to 0.66 10^{-2} $kgCO_{2,eq}$ m^{-3}) due to the increase of the autotrophic bacteria activities. At high SRTs biomass endogenous decay rate dominates since most carbon has been oxidized. This will limit the denitrification rate, thus contributing to N_2O emissions (Boiocchi et al., 2017b). $Offgas_{CO2}$ increases with an increase of SRT.

This increase is most evident for SRT values ranging between 10 and 25 days ($Offgas_{CO2}$ increases from 0.35 to 0.50 10^{-2} $kgCO_2$ m^{-3}) due to the increase of the biomass activity. Further increase of the SRT leads to the inert biomass accumulation inside the system (Judd, 2010). Therefore, for SRT values higher than 25 days, the $Offgas_{CO2}$ is quite stable and equal to 0.55 10^{-2} $kgCO_2$ m^{-3}. The DE and EQI_{GAS} follow the same trend of the individual GHG emissions previously presented, being more influenced by the N_2O emissions due to its higher GWP. The difference between both PIs in this case is that DE represents the amount of GHG emitted, while EQI_{GAS} represents the potential of the WWTP to emit GHG. SRT also influences the results of EF and EQI_{LIQ}, due to the higher capability of the system to support nitrification. The $Q_{air,AER}$ variation strongly influences IE and OC (Figure 1b). Specifically, the variation of $Q_{air,AER}$ influences the energy consumption, which is the main contributor for both IE and OC.

R_{QR1} (Figure 1c) exerts a smaller influence over the PIs when compared to SRT and $Q_{air,AER}$. A similar result was obtained for R_{QR2} (Figure 2d), which slightly influences RON, EF and EQI_{LIQ}. Figure 2 shows the variation of RON and R_{NAT} with SRT, R_{QR1} and R_{QR2}.

<Here Figure 2>

RON increases (from 4.04 to 5.90 gO_2 gNH_4^{-1}) with the increase of SRT and R_{QR2} (Figure 2a, Figure 2b). The increase of the SRT leads to the increase of nitrification with the consequent rise of the amount of oxygen consumed and RON. The increase of R_{QR2} reduces the oxygen concentration within the aerobic reactor, thus causing an increase of RON.

The R_{NAT} will increase together with the SRT (Figure 2c). The reason is that it allows an increase of the nitrification rate, i.e. a higher amount of nitrate has been produced. The increase of R_{QR1} leads to the decrease of R_{NAT} (Figure 2d) since the inlet nitrate load to the aerobic reactor decreases. It is

caused by the increase of the combined oxygen concentration recycled from the anoxic to the anaerobic reactor. Therefore, most of the PAOs activity in the anaerobic reactor (turned anoxic) is as denitrifiers. Consequently, the nitrate concentration in the following reactors will be reduced.

To understand the role of SRT on GHG emissions, the spatial distribution of $Offgas_{N2O}$ within each reactor of the investigated MBR plant is shown in Figure 3. Data of Figure 2 consider three values of SRT (10, 25 and 50 days). Furthermore, $Q_{air,AER}$ = 35 $m^3.d^{-1}$, $Q_{air,MBR}$ = 15 $m^3.d^{-1}$, R_{QR1} = 0.8, and R_{QR2} = 6.2.

<Here Figure 3>

From Figure 3 it is noted that the N_2O emissions from the anaerobic (Figure 3a) and anoxic (Figure 3b) reactors are lower than that of the other reactors. These emissions are related to the heterotrophic activities (PAO and heterotrophic non-PAO) while incomplete denitrification takes place.

The AOB and NOB are the major contributors to N_2O emissions (Boiocchi et al., 2017a), which can be observed by the increase of the $Offgas_{N2O}$ from the aerobic reactor (Figure 3c). The aerated reactors are the major contributors of $Offgas_{N2O}$ within the MBR plant (Ribera-Guardia et al., 2019), followed by the MBR reactor (Figure 3d). The emissions from the MBR reactor are mostly due to the stripping of N_2O in gas form from its related dissolved component (Massara et al., 2018).

The emissions for SRT equal to 10 days were negligible, mainly due to the lower AOB and NOB activities at low SRT values. The N_2O emissions are related to both the nitrification and denitrification processes, which are less pronounced (especially the nitrification) at low SRT values. For SRT values higher than 10 days allows a more complete nitrification, enhancing the probability of N_2O formation by the AOB. As mentioned before, the high SRT also favours the processes related to the heterotrophic microorganisms (e.g., phosphorus removal and denitrification), which also contributes to the N_2O formation pathways.

The influence of SRT on the growth of AOB and the NO_2 production within the aerobic reactor reported in Figure 4.

Figure 4a shows that AOB concentrations will decrease due to the low AOB growth rate at low SRT. For higher SRT values the Figure 4b, 4c), the AOB growth increases so that the AOB concentrations will increase. This will cause the NO_2 concentration to accumulate in the aerobic reactor. Similar results were obtained by Massara et al. (2017).

<Here Figure 4>

Figure 3 and Figure 4 suggest that a low concentration of AOB biomass leads to a low dissolved concentration of NH_2OH or NO and, consequently, the N_2O emission is negligible. This also means that the growth of NOB, heterotrophic non-PAOs and PAOs is compromised, negatively affecting nutrient removal. This emphasizes that the SRT is the model parameter having the largest influence on the most performance indicators.

3.2 Multi-objective optimization and performance assessment

In Table 3 the results of the five investigated operational parameters for the optimal and benchmark solution are displayed. The optimal solution shows an increase of the SRT value (from 35 to 49 days), a decrease of R_{QR1} (from 1 to 0.54), an increase of R_{QR2} (from 5 to 6.4), a substantial decrease of $Q_{air,AER}$ (from 22 to 11 m^3 d^{-1}), a slight decrease of $Q_{air,MBR}$ (from 14.4 to 14.2 m^3 d^{-1}).

<Here Table 3>

Table 4 summarizes the results for each OF related to the benchmark and the optimal solution. The TOPSIS application allowed to optimize seven of the ten OFs (in grey in Table 4). The substantial reduction of the $Q_{air,AER}$ value contributed to reducing the optimal OC to almost half compared to the benchmark solution (from 1.05 to 0.59 € m^{-3}), due to the reduced energy consumption. Since the IE are mainly related to energy consumption, a substantial reduction of IE occurred as well (from 1.12 to 0.57 $kgCO_{2,eq}$ m^{-3}). Energy savings of this magnitude, 48% of the OCs due to the aeration and IE, is naturally of major interest. Note that the dissolved oxygen concentration in the aerobic reactor is not limiting the nitrification process (always >1.5 mgO_2 L^{-1}), despite the low $Q_{air,AER}$ value.

<Here Table 4>

The optimal solution achieved a 10% reduction of DE (Table 4), mainly caused by the reduction of $Offgas_{N2O}$ compared to the benchmark (from 0.57 to 0.50 $kgCO_{2,eq}$ m^{-3}). This result seems to contradict the trend shown in Figure 3 where the $Offgas_{N2O}$ concentration increases with the increase of SRT. However, the results in Figure 3 have been obtained for a higher $Q_{air,AER}$ value (around 30 m^3 d^{-1}) than that of the optimal solution. Consequently, since the $Offgas_{N2O}$ concentration depends on $Q_{air,AER}$ value (lower $Q_{air,AER}$ reduce the stripping effect) the results of the optimal solution have been influenced by the lower $Q_{air,AER}$ value.

Table 4 illustrates that a slight increase of EF (from 0.09 to 0.1 € m^{-3}) and EQI_{LIQ} (from 14.7 to 15.6 kg m^{-3}) occurred for the optimal solution. The increase of EQI_{LIQ} is caused by a slight increase (around 10%) of effluent ammonia concentration; conversely, a substantial decrease in terms of S_{PO} concentration in the effluent occurred for the optimal solution (from 1.5 mg L^{-1} to 0.4 mg L^{-1}). The reason is the increased hydraulic retention time (HRT) inside the anaerobic reactor, due to the

decreased R_{QR1} (from 1 to 0.54). A lower HRT of the anaerobic reactor allowed a major anaerobic phosphate release and a subsequently uptake from PAOs in the aerobic and anoxic conditions.

A value of R_{NAT} less than 1.0 gNO_3 gNH_4^{-1} represents a good balance between AOB and NOB. This value was obtained for the optimal solution, confirming the low nitrification efficiency for the system under study (Boiocchi et al., 2017b), which has also influenced the N_2O formation during the nitrification/denitrification processes.

RON increased from 3.50 to 5.79 gO_2 gNH_4^{-1} mainly due to reduction of the oxidized ammonia in the optimal solution (Table 4). The higher RON (5.79 gO_2 gNH_4^{-1}) of the optimal solution is in agreement with the related lower $Offgas_{N2O}$ concentration. Indeed, according to Boiocchi et al. (2017a), for high value of RON (>5.0 gO_2 gNH_4^{-1}), the NOB activity increases as well; the AOB and the heterotrophic biomass denitrification producing N_2O decreases due to the low NO_2 availability, consequently N_2O decreases. The decrease of $Offgas_{N2O}$ also caused up to 8% reduction in EQI_{GAS} compared to the benchmark solution (from 60.1 to 55.60 kg m^{-3} Figure 5 displays the average gaseous $Offgas_{N2O}$ concentration per reactor and the average total DE.

Figure 5 shows that the major $Offgas_{N2O}$ concentration occurred in the MBR in both solutions. This is due to the higher aeration rate of the MBR compared to aerobic reactors. The result is in agreement with Mannina et al (2017) finding the highest N_2O flux from the MBR reactor. The second major emitter is the aerobic reactor, emphasizing the role of the aeration in the $Offgas_{N2O}$ concentration. Only negligible $Offgas_{N2O}$ and DE emission were found from the anaerobic and anoxic reactors, since the greater part of N_2O has been produced during the nitrification (Figure 5).

Figure 5a demonstrates the average $Offgas_{N2O}$ concentration and DE for all the plant reactors is smaller for the optimal solution than for the benchmark solution. The $Offgas_{N2O}$ concentration emitted from the MBR reduced from 0.45 to 0.38 10^{-2} $kgCO_{2eq}$ m^{-3}, and from the aerobic reactor from 0.075 to 0.06 10^{-2} $kgCO_{2eq}$ m^{-3}. Similar results were obtained for the DE, since it is mainly related to $Offgas_{N2O}$. DE for to the MBR decreased from 0.48 to 0.4 10^{-2} $kgCO_{2eq}$ m^{-3}, and for the aerobic reactor from 0.095 to 0.86 10^{-2} $kgCO_{2eq}$ m^{-3} (Figure 5b).

<Here Figure 5>

Figure 6 illustrates results obtained from the TOPSIS application for some OF. The trend of the non-dominated solutions, representing all the solutions obtained for the 5,000 simulations, has been reported for R_{NAT} *vs* $Offgas_{N2O}$, $Offgas_{N2O}$ *vs* $Offgas_{CO2}$, and DE *vs* R_{NAT}. Optimal and benchmark solutions, (Table 3) are also indicated in Figure 6.

Data of Figure 6a show that few solutions enable the increase of R_{NAT} (which should be maximized) at low total $Offgas_{N2O}$. This result is mainly related to the R_{NAT} value that is lower than 0.25 gNO_3 gNH_4^{-1}; this value corresponds to an $Offgas_{N2O}$ concentration lower than 0.37 10^{-2} $kgCO_{2eq}$ m^{-3} that is typical of the AOB inhibition condition (Baiocchi et al., 2017a-b). The corresponding $Offgas_{N2O}$ concentration value is low due to the negligible nitrification and consequently to the denitrification. Since there is a direct relationship between N_2O emission and DE, the increase of R_{NAT} leads to the increase of DE (Figure 6c). However, since no R_{NAT} value close to 1.0 gNO_3 gNH_4 has been obtained,

it can be confirmed that insufficient nitrification occurred inside the system for the all solutions. Further investigations with the use of a wider range for the assessed operational conditions may obtain better nitrification results and, consequently, lower $Offgas_{N2O}$ and DE at the highest $R_{NAT.}$

<Here figure 6>

4. Conclusions

The sludge retention time is the key operational parameter affecting mainly the direct emissions; the results show that direct emissions increase with sludge retention time mainly due to the nitrous oxide concentration in the off-gas increases (up to 0.66 10^{-2} $kgCO_{2,eq}$ m^{-3}). Further, increasing sludge retention time (from 10 to 50 days) enhances the nitrification thanks to a higher concentration of autotrophic microorganisms. The multi-objective optimization approach is practical and feasible to be adopted both by modelers and by operators even for complex integrated membrane bioreactor models.

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List of Tables and Figures