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Title: Aeration control in Membrane Bioreactor for sustainable environmental footprint

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Keywords: membrane bioreactor, aeration-based control strategy, proportion-integration control

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Abstract: Aeration in membrane bioreactor systems for wastewater treatment is one of the main source for energy consumption. In this study different scenarios were scrutinized to minimize the energy consumption. Specifically, open-loop and closed-loop scenarios were performed by a two-step cascade control strategies based on dissolved oxygen, ammonia and nitrite concentrations. An integrated MBR model was employed which includes the greenhouse gas formation/emission processes. The air flow in closed-loop control led to a substantial reduction in terms of energy consumption (32% for Scenario 1 and 82% for Scenario 2). The air flow control based on both ammonia and nitrite concentrations within the aerobic reactor (Scenario 2) provided excellent results in terms of operating cost (64% reduction), direct (10% reduction) and indirect (81% reduction) emissions.

Highlights

- Two-step cascade control strategies have been applied to MBR
- An integrated MBR mathematical model has been adopted
- Energy consumption reduces till to 82% controlling the aerobic airflow rate
- Operating cost reduces till 64 % controlling the aerobic airflow rate
- Direct GHG emission reduces from 0.52 to 0.47 kgCO_{2eq} m⁻³ under control condition

15 **Abstract**

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316 In this study different scenarios were scrutinized to minimize the energy consumption of a membrane
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517 bioreactor system for wastewater treatment. Open-loop and closed-loop scenarios were investigated by two-
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718 step cascade control strategies based on dissolved oxygen, ammonia and nitrite concentrations. An integrated
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919 MBR model which includes also the greenhouse gas formation/emission processes was applied. A
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1120 substantial energy consumption reduction was obtained for the closed-loop scenarios (32% for Scenario 1
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1421 and 82% for Scenario 2). The air flow control based on both ammonia and nitrite concentrations within the
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1622 aerobic reactor (Scenario 2) provided excellent results in terms of reduction of operating cost reduction
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1823 (64%), direct (10%) and indirect (81%) emissions.

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1. Introduction

Wastewater treatment plants (WWTPs) can be responsible for both liquid and gaseous pollutants discharge into the environment. WWTPs operation has a constant challenge to provide the excellent effluent quality at the lowest operational costs as possible (Bozkurt et al., 2016). WWTPs are responsible for emitting almost 3% of the main greenhouse gases (GHG) (carbon dioxide - CO₂, methane - CH₄, and nitrous oxide - N₂O) by direct (due to biomass metabolism) and indirect (due to electricity and chemical consumption) sources (Mannina et al., 2016; Polruang et al., 2018; Koutsou et al., 2018; Domingo-Félez and Smets, 2020). Among the most relevant current challenges for WWTPs, GHG emission minimization is one of the utmost (Flores-Alsina et al., 2011).

In view of addressing the aforementioned challenges new operating strategies aimed at improving the overall WWTP performance are required (Wu et al., 2020). With this regard, the use of membrane bioreactors (MBRs) was introduced in the past decade, as a promising alternative to conventional activated systems (CAS) in order to obtain excellent effluent quality (Xiao et al., 2014; Liu et al., 2018). Indeed, MBRs are known due to their ability to provide high effluent quality, to reduce sludge production and to require low space for implementation (Guo et al., 2012). Despite the MBR advantages, their higher energy demand when compared to CAS (for membrane aeration, permeate extraction, among others) coupled with membrane fouling issues still represent serious drawbacks for the technology spread (She et al., 2016). With this regard, several efforts have been performed in literature in order to reduce MBR energy costs and to avoid/reduce/mitigate membrane fouling. Even though, literature is still far from finding a definitive solution for these issues (Krzeminski et al., 2017).

The high energy requirement of MBR represents an environmental issue since electricity is also related to GHG indirect emissions (Mannina et al., 2018a). A great part of the energy consumption in MBRs regards the presence of additional aeration systems for fouling mitigation and the presence of the permeate extraction pumps (Yang et al., 2016; Zheng et al., 2018; Zhang et al., 2019). The aeration systems are responsible for about 70 to 80% of the total energy consumption of a WWTP contributing substantially to the total plant operating costs (Sun et al., 2016; Xiao et al., 2014). Indeed, about 30% of the WWTP budget is related with

53 the aeration systems (Metcalf & Eddy, 2003). For this reason, the optimization of aeration systems is
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254 imperative in view of reducing operating costs.
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55 Aeration-based control strategies are reported in the literature with the attempt to optimize aeration systems
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756 by regulating the air blowers with the use of manual or automatic controllers (Maere et al., 2011; Sun et al.,
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957 2016). Nowadays, manual controllers are hardly implemented because they are susceptible to human errors.
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1158 Thus, automatic controllers are preferable in order to ensure the optimal system response. However, the
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1459 implementation of automatic aeration controllers in real WWTPs requires huge capital investments (Olsson
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1660 and Newell, 1999), which makes the mathematical modelling a recommendable tool prior to the system's on-
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1861 site installation (Rivas et al., 2008; Gabarrón et al., 2015). As a matter of a fact, model simulation enables
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2162 decision-makers to act faster at the smallest disturbance, which constitutes one of the main reason that
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2363 aeration control strategies are very often coupled with modeling systems (González et al., 2018). This
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2564 coupling allows to compare and investigate several operational scenarios that are influenced by changes in
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2765 aeration (Maere et al., 2011).
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3066 Most of the aeration-based control strategies are based on the real-time behavior of key process parameters,
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32
3367 such as dissolved oxygen (DO) and ammonia (NH₄) concentrations. The purpose of a DO-based control
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3568 strategy is to drive the DO concentration within the aerobic tanks towards a stable and optimized condition,
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3769 in which the whole amount of air insufflated is sufficient for maintaining the biomass survival and the
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3970 treatment process (Gabarrón et al., 2015). However, the DO concentration is an operational parameter that
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4171 may influence several processes (e.g., nitrification and denitrification, biomass survival, GHG emissions);
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4472 therefore, establishing a control strategy based only on the above aspect does not guarantee that the effluent
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4673 quality respects the effluent standards (Wahab et al., 2009). For this reason, feedback control, which is based
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4874 on ammonia concentration, is proposed in the literature with the aim of obtaining the optimal trade-off
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5075 between the air supplied and the effluent quality (Wahab et al., 2009; Sun et al., 2016).
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5376 Two main control strategies are reported in the literature with the aim to optimize the air flow rate inside an
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5577 aerobic compartment: i. open-loop control; ii. closed-loop control (Olsson and Newell, 1999). In the open-
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5878 loop control, no automatic feedback derived from the real-time measurement is applied since the control is
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6079 based on a timer and/or a predefined program of actions (e.g., time-set air supply in the aerobic reactor or
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80 MBR) without looking to the effluent quality or gaseous emissions. On the other hand, in the closed-loop
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21 control, the actions (feedback) are automatic and based on real-time measurements (e.g., the control of the
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42 air flow rate inside the aerobic reactor is based on effluent ammonia concentration).
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73 The open-loop control does not guarantee to meet the effluent limits of the discarded pollutants; indeed, not
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934 inter-related changes in the air supply, as a function of the effluent limits, will result in worsening/improving
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11 the WWTP performance in terms of carbon and nutrients removal (Kalboussi et al, 2018). Regarding closed-
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13 loop control, literature reports some applications to MBR mainly focused on the optimization of the
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15 membrane filtration process (Ferrero et al., 2012). Specifically, Ferrero et al. (2011) applied a performance-
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17 based control to optimize aeration in MBR by using permeability as the key parameter. Results demonstrated
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19 that the reduction of the permeate flux can save up to 21% of the energy used for membrane aeration.
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21 Dalmau et al. (2014) applied an experimental approach based on establishing a DO setpoint to maintain
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23 aerobic conditions and lowering fouling in an MBR. Results indicated 75% of energy consumption
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25 reduction, without compromising nutrient removal efficiency. Sun et al. (2016) proposed an in-situ
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27 ammonia-based feedback control strategy to a full-scale MBR obtaining a reduction of the overall energy
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29 specific consumption up to 0.45kWh m⁻³ of treated effluent.
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35 Some authors have also focused the attention on control/optimization strategies aimed at reducing the plant
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37 operational costs in anaerobic membrane bioreactors (AnMBRs), where the closed-loop control strategies are
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39 required for reducing membrane fouling and operating costs (Robles et al., 2018). Specifically, Benyahia et
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41 al. (2013) developed a model applied to an AnMBR with the aim to establish a control tool. In particular,
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43 Benyahia et al. (2013) focused the attention on the reduction of membrane fouling by controlling the soluble
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45 microbial products (SMP) formation/degradation process. Robles et al. (2014) applied an advanced
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47 knowledge-based control system aimed at optimizing the filtration process in an AnMBRs. The authors
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49 obtained substantial saving in energy requirements and operating costs (up to 25% and 53.3%, respectively).
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53 Despite the above referenced literature studies, as far as the authors are aware, there is any study on the
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55 application of aeration/feedback control for MBR systems including multiple output variables: direct and
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57 indirect GHG emissions and effluent quality. This study presents a first attempt to apply a cascade control
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59 for an MBR systems by considering a comprehensive analysis based on the above mentioned multiple
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107 outputs. The final aim of this study is to evaluate the effectiveness of feedback closed-loop strategies applied
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108 to an MBR pilot-plant focusing on system optimization in terms of effluent quality (gaseous and liquid),
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109 operating costs and energy consumption. With this regard, feedback closed-loop strategies were
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110 implemented by adopting an integrated MBR model (Mannina et al., 2018a-b). In particular, three scenarios
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111 are analyzed: i. Scenario 0 – reference scenario with open-loop control – air flow rate was optimized without
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112 considering any real-time measurement (Mannina et al., 2019); ii. Scenario 1 – with a closed-loop control
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113 where the aeration control is based on ammonia concentration inside the aerobic reactor; iii. Scenario 2 – with
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114 a closed-loop control where the aeration control is based on both ammonia and nitrite (NO₂) concentration
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115 inside the aerobic reactor.
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23 2. Material and methods 24

25 2.1 The mathematical model 26

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28 The integrated mathematical model applied here is characterized by two main sub-models: biological and
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30 physical (Mannina et al., 2018b). The biological sub-model is based on the ASM2d algorithms proposed by
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32 Henze et al (2000) modified to include soluble microbial products (SMP) formation/degradation, GHG
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34 production/emission and detailed nitrogen transformation processes. More in detail, the biological sub-model
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36 consists of 116 parameters and 25 state variables. Nitrogen transformation is described as a two-step
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38 nitrification process (Pocquet et al., 2016) and four-step denitrification processes (Hyatt and Grady, 2008).
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42 The two-step nitrification considered by the sub-model is summarized as follows:
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- 45 - First step: (i) NH₄ is oxidized into NO₂ by means of ammonia-oxidizing bacteria (AOB); (ii)
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47 incomplete ammonia oxidation may lead to the formation of intermediate products, such as
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49 hydroxylamine (NH₂OH) and nitric oxide (NO); (iii) oxidation of NH₂OH to NO₂, with the
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51 accumulation of NO; (iv) a reduction of NO may be observed leading to the formation of N₂O.
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- 54 - Second step: the NO₂ is oxidized into nitrate (NO₃) by means of nitrite-oxidizing bacteria (NOB).
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56 The four-step denitrification is assessed taking the contribution of the phosphorus accumulating organisms
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58 (PAOs) and heterotrophic non-PAO biomass (OHO) under anoxic conditions, which includes (i) reduction of
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60 NO₃ to NO₂; (ii) reduction of NO₂ to NO; (iii) reduction of NO to N₂O; (iv) reduction of N₂O to nitrogen gas
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134 (N₂). The incomplete reduction of NO₂ into N₂ may lead to the accumulation of N₂O, which is also included
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135 in the model.

136 The physical sub-model is characterized by 6 parameters and 2 state variables. The physical sub-model
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137 allows to assess the contribution of the membrane module in the organic matter removal by means of the
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138 cake layer formed onto the membrane and the physical separation throughout the membrane (Mannina et al.,
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139 2018a).

140 Biological and physical sub-models are interlinked by means of the total suspended solids (TSS) and SMP
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141 concentration inside the MBR. The model also evaluates the total GHG emissions (both in terms of N₂O and
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142 CO₂) as the sum of direct and indirect emissions of both sub-models. Further details regarding the model can
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143 be found in the literature (Mannina et al., 2018a-b).

2.2 Pilot plant description

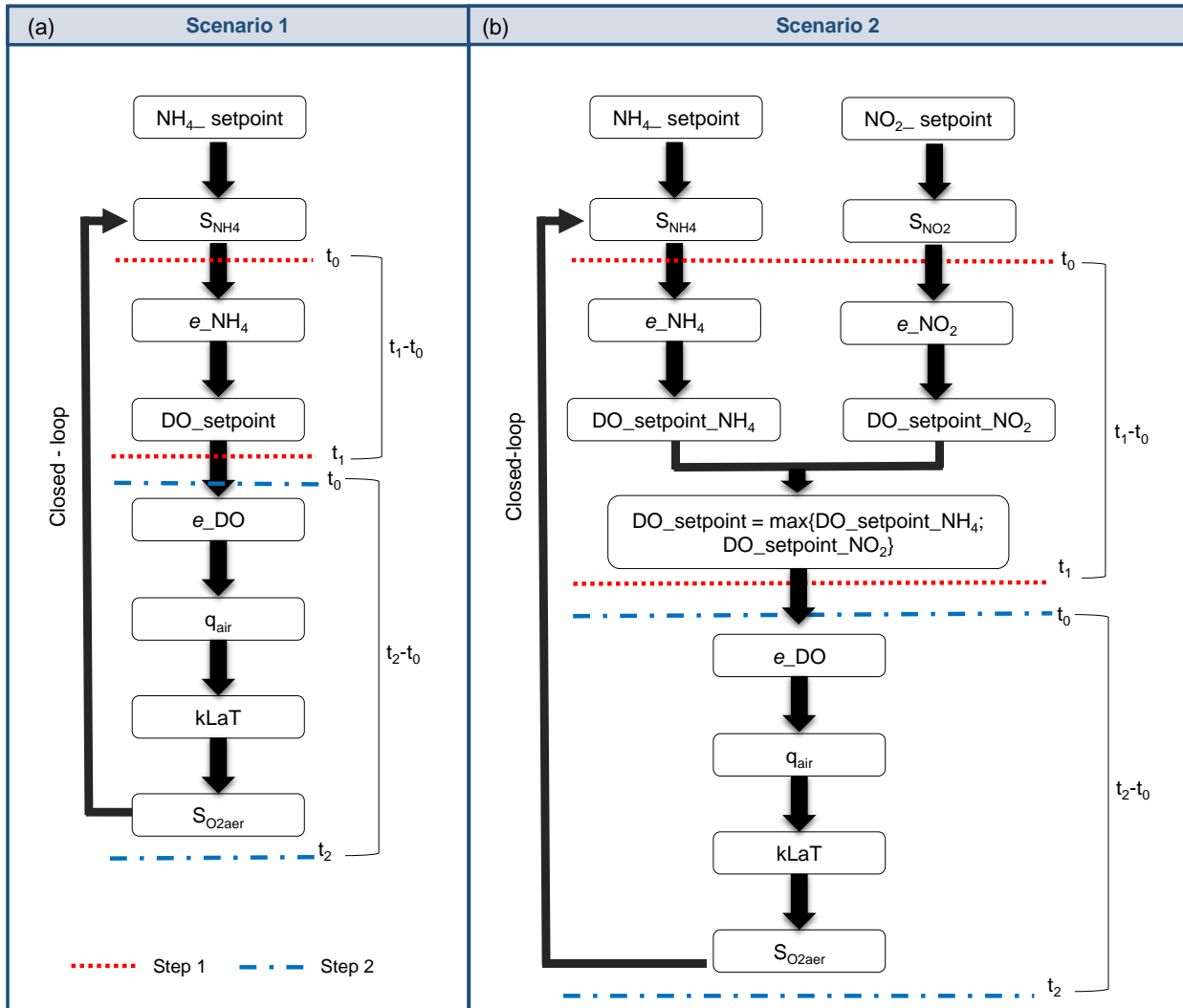
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2745 A University of Cape Town (UCT) MBR pilot plant (composed by anaerobic, anoxic and aerobic reactors in
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2746 series) has been taken as case study (Mannina et al., 2016). The influent wastewater (a mixing between real
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2747 and synthetic wastewater) flow rate was equal to 20 L h⁻¹ with constant carbon-nitrogen ratio features (equal
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2748 to 10 mgCODmgTN⁻¹) (Mannina et al., 2018a). The solid/liquid separation occurred by means of an
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2749 ultrafiltration hollow fiber membrane (PURON® - pore size of 0.03 µm and membrane surface of 1.4 m²)
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2750 located inside the MBR bioreactor (permeate flux of 21 Lm²h⁻¹). For a more detailed description of the pilot
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2751 plant and sampling campaign, the reader is referred to Mannina et al. (2016).

2.3 Scenario analysis

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2752 Three scenarios have been considered in this study: i. Scenario 0 – Benchmark with an open-loop air flow
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2753 control inside the aerobic reactor; ii. Scenario 1 – where a closed-loop cascade ammonia proportional-
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2754 integral (PI) control is applied inside the aerobic reactor to establish the DO setpoint and consequently the air
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2755 flow rate; iii. Scenario 2 – where a closed-loop cascade PI control based on ammonia and nitrite
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2756 concentration inside the aerobic reactor is applied to establish the DO setpoint and consequently the air flow
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2757 rate. Scenario 2 aims at reducing the amount of N₂O emission from the aerobic reactor. The scenario analysis
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2758 has been employed by using the mathematical model described above and considering 42 simulation days.
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160 **2.3.1 Aeration control strategies – scenarios 1 and 2**

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161 Figure 1 shows the closed-loop aeration control strategies applied for scenarios 1 and 2.
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163 **Figure 1.** Schematic representation of two-step cascade control adopted for Scenario 1 (a) and Scenario 2
164 (b). $NH_4_setpoint$ = set point of ammonia concentration inside the aerobic reactor; $NO_2_setpoint$ = set point
165 of nitrite concentration inside the aerobic reactor; S_{NH_4} , S_{NO_2} and S_{O_2aer} = ammonia, nitrite and dissolved
166 oxygen concentration inside the aerobic reactor, respectively; $DO_setpoint$ = set point of the dissolved
167 oxygen concentration inside the aerobic reactor; $DO_setpoint_NH_4$ = set point of the dissolved oxygen
168 concentration inside the aerobic reactor established on the basis of ammonia control (S_{NH_4});
169 $DO_setpoint_NO_2$ = set point of the dissolved oxygen concentration inside the aerobic reactor established on
170 the basis of nitrite control (S_{NO_2}); e_{DO} , e_{NH_4} and e_{NO_2} error of the dissolved oxygen, ammonia and

171 nitrite concentration; q_{air} = air flow rate inside the aerobic reactor; $kLaT$ = oxygen transfer coefficient; t_0 , t_1
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172 and t_2 = time related to the control interval during Step 1 (t_0 and t_1) and Step 2 (t_0 and t_2).
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174 For Scenario 1, a similar approach to previous literature was employed (Sun et al., 2016). In particular, an
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175 aeration control strategy algorithm was implemented as a two-step feedback control based on the NH_4 and
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176 DO concentration (first step), and air flow (second step) (Figure 1a).
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177 The first action in the aeration control strategy is to establish the ammonia set point ($NH_4_setpoint$) inside
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178 the aerobic reactor (Figure 1a). Then, the ammonia error (e_NH_4 , as $mg.L^{-1}$) is calculated as the difference
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179 between $NH_4_setpoint$ and the NH_4 concentration within the aerobic reactor (S_{NH_4} , as $mg.L^{-1}$) (Equation 1).
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$$180 \quad e_NH_4 = NH_4_setpoint - S_{NH_4} \quad (1)$$

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181 The $NH_4_setpoint$ is manually assigned on the basis of the effluent requirements. If the concentration of NH_4
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27 in the aerobic tank is higher than $NH_4_setpoint$ ($e_NH_4 < 0$), the aeration system insufflates more air in order
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29 to increase the nitrification and reduce the ammonia concentration in the bioreactor. Conversely, if $e_NH_4 >$
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31 0, the air flow rate is reduced to ensure that the ammonia concentration in the tank reaches a stable value
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184 with respect to the $NH_4_setpoint$.
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186 The value of e_NH_4 is applied to calculate the DO setpoint ($DO_setpoint$), which represents the DO
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39 concentration of interest that may lead to a NH_4 stable value (Equation 2) (Figure 1a).
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$$188 \quad DO_setpoint = Bias_1 + K_{p1} \cdot e_NH_4 + K_{p1} \cdot \frac{1}{\tau_1} \cdot \int_{t_1-t_0}^{t_0} e_NH_4 \cdot dt \quad (2)$$

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189 where $Bias_1$, K_{p1} and τ_1 are control parameters (Sun et al., 2016), t_0 represents the initial time of the control
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47 (and its equal to zero), t_1-t_0 is the control interval (assumed equal to 30 minutes in this simulation) and
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190 $e_NH_4 \cdot dt$ is the derivate of the NH_4 error during the control interval. Other acronyms were previously
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50 described. In Equation 2 the term $Bias_1$ represents the baseline NH_4 error, while the term $K_{p1} \cdot e_NH_4$ is the
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52 NH_4 real-time error and the term $K_{p1} \cdot \frac{1}{\tau_1} \cdot \int_{t_1-t_0}^{t_0} e_NH_4 \cdot dt$ represents the NH_4 error accumulated during the
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54 control interval.
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195 Therefore, in the first step, the ammonia-based and DO-based control strategies are combined before
 196 applying the cascade control in the second step. At the beginning of the second step, the calculated
 197 DO_setpoint is used to obtain the DO error (e_{DO} , as mg.L^{-1}) related to the DO concentration (S_{O2aer} , as
 198 mg.L^{-1}) inside the aerobic reactor, calculated as shown in Equation 3 (Figure 1a).

$$199 \quad e_{DO} = DO_setpoint - S_{O2aer} \quad (3)$$

200 The error related to the DO concentration is used to obtain the air flow rate that has to be supplied by the
 201 aeration system (Equation 4). If $e_{DO} < 0$, the aeration system reduces the q_{air} value and vice versa.

$$202 \quad q_{air} = Bias_2 + K_{p2} \cdot e_{DO} + K_{p2} \cdot \frac{1}{\tau_2} \cdot \int_{t_3-t_2}^{t_2} e_{DO} \cdot dt \quad (4)$$

203 where $Bias_2$, K_{p2} and τ_2 are controller parameters (Sun et al., 2016), t_2-t_0 is the control interval (assumed as
 204 the 30 minutes that succeed the previous step) and $e_{DO} \cdot dt$ is the derivate of the DO error during the control
 205 interval.

206 The value of q_{air} is used by the model to obtain the oxygen transfer coefficient (kLaT), which is introduced in
 207 the oxygen (namely, S_{O2aer}) mass-balance equation according to the ASM approach (Henze et al., 2000). The
 208 term kLaT is calculated according to Equation 5.

$$209 \quad kLaT = k_1 * (1 - \exp^{(k_2 \cdot q_{air})}) \quad (5)$$

210 where k_1 and k_2 are parameters related to the MBR plant. Table 1 contains the values of the control
 211 parameters mentioned in this section.

212 The control strategy related to Scenario 2 is an extension of Scenario 1. The first phase of control strategy
 213 applied to Scenario 2 includes a cascade PI nitrite controller in the aerobic reactor to calculate the DO
 214 setpoint (Figure 1b). More specifically, during the first step, two DO setpoints are calculated: 1)
 215 $DO_setpoint_NH_4$, evaluated based on the ammonia control analogously to Scenario 1; 2) $DO_setpoint_NO_2$
 216 evaluated on the basis of the nitrite control.

217 $DO_setpoint_NO_2$ is evaluated according to Equation 6.

$$218 \quad DO_setpoint_NO_2 = Bias_{1,NO_2} + K_{p1,NO_2} \cdot e_{NO_2} + K_{p1,NO_2} \cdot \frac{1}{\tau_{1,NO_2}} \cdot \int_{t_1-t_0}^{t_0} e_{NO_2} \cdot dt \quad [6]$$

219 where $Bias_{1,NO_2}$, K_{p1,NO_2} and τ_{1,NO_2} are controller parameters related to nitrite control and e_{NO_2} is the NO_2
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 220 error during the control interval. e_{NO_2} represents the difference between the nitrite set point ($NO_2_setpoint$)
 3
 221 and the actual NO_2 concentration (S_{NO_2}) inside the aerobic reactor.
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222 The maximum value between $DO_setpoint_NO_2$ and $DO_setpoint_NH_4$ is then selected to evaluate the DO
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 223 error (e_{DO}) during the second control step (Figure 2b).
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224 The second control step of Scenario 2 is identical to Scenario 1.
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 14

225 **Table 1.** Summary of the parameters of the control algorithm applied to Scenario 1 and Scenario 2
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Control Parameter	Value	Unit	Reference
$NH_4_setpoint$	10	$mg.L^{-1}$	(this study)
$Bias_1$	1	$mg.L^{-1}$	Sun et al. (2016)
K_{p1}	-1	$mgDO.L^{-1}/ mgN.L^{-1}$	Sun et al. (2016)
τ_1	20	minutes	Sun et al. (2016)
$NO_2_setpoint$	0.5	$mg.L^{-1}$	(Solis et al., 2019)
$Bias_{1,NO_2}$	1	$mg.L^{-1}$	Sun et al. (2016)
K_{p1,NO_2}	-1	$mgDO.L^{-1}/ mgN.L^{-1}$	Sun et al. (2016)
τ_{1,NO_2}	30	minutes	Sun et al. (2016)
$Bias_2$	600	$m^3.d^{-1}$	Sun et al. (2016)
K_{p2}	500	$m^3air.d^{-1}.h^{-1}$	Sun et al. (2016)
τ_2	15	minutes	Sun et al. (2016)
k_1	200	-	Mannina et al. (2018a)
k_2	-0.25	-	Mannina et al. (2018a)

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 227 The control of DO is enhanced by the two-step cascade control leading to to an improvement of the
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 228 nitrification process by acting on the NH_4 oxidation.
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230 2.4 Performance indicators

231 The influence of the open and closed-loop dynamic aeration controls is assessed by the following
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 232 Performance Indicators (PIs): Effluent Quality Index (EQI, kg Pollutant m⁻³) for both liquid (EQI_{LIQ}) and gas
 3
 233 (EQI_{GAS}) flows; oxygen-to-total-Kjeldahl-nitrogen ratio (RON, gO₂ gNH₄⁻¹); ratio nitrate-ammonia (R_{NAT},
 5
 234 gNO₃ gNH₄⁻¹); Operating Costs (OC, as euro m⁻³); Effluent Fine (EF, euro m⁻³); CO₂ and N₂O emissions
 7
 235 (kgCO_{2,eq} m⁻³); direct (DE, kgCO_{2,eq} m⁻³) and indirect (IE, kgCO_{2,eq} m⁻³) GHG emissions.

11
 1236 The EQI quantifies the pollution load discharged into the water body (kg pollution units/day or kg pollution
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 1437 units/treated volume) Equation 7) (Nopens et al., 2010; Mannina et al., 2019).

$$EQI_{LIQ} = \frac{1}{T \cdot 1000} \int_{t_0}^{t_1} [\sum P_k(t)] \cdot Q_{eff} dt \quad (7)$$

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 19
 2039 where t_0 indicates the initial time, t_1 the end of the simulation period, Q_{eff} is the accumulated effluent flow,
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 2240 dt is the simulation period, 1000 is the conversion factor from g m⁻³ to kg m⁻³, P_k in the pollutant load of
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 241 each component in a time t , which is expressed according to Equation 8.

$$P_k = \beta_x \cdot C_k \quad (8)$$

27
 2842 where β_x is the weighting factor of every single pollutant and C_k is the pollutant's concentration (mg·L⁻¹).
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 3143 The following components (k) were considered in this study: chemical oxygen demand (COD_e), ammonia
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 3344 (S_{NH4e}), nitrate (S_{NO3e}), nitrous oxide (S_{N2Oe}) and phosphate (S_{POe}), for which the following weighting factors
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 3545 were used (Mannina & Cosenza, 2015): $\beta_{COD}=1$, $\beta_{NH}=20$, $\beta_{NO3}=20$, $\beta_{N2O}=50$ and $\beta_{PO}=50$.
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 4047 The EQI_{GAS} was also adopted by Mannina et al. (2019) considering the gas flow rate (Q_{offgas}) and the off-gas
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 4348 concentration in terms of CO₂ and N₂O (Offgas_{CO2} and Offgas_{N2O}, respectively). The adopted β_i values for
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 4549 EQI_{GAS}, defined for each GHG are $\beta_{N2O}=50$ and $\beta_{CO2}=50$.

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 4850 RON provides a relationship between the oxygen supplied to the plant and the Total Kjeldahl Nitrogen
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 5051 (TKN) in the influent (Boiocchi et al., 2017a). Considering the main purpose of this work, RON is a key
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 5252 indicator to verify the plant's performance since it allows understanding how much of the oxygen provided
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 5453 to the system was used to oxidize the influent ammonium. RON is calculated according to Equation 9.

$$R_{ON} = \frac{\sum_{i=1}^n k_{LAER,i} \cdot V_{AER,i} (SO_{2,SAT,AER,i} - SO_{2,AER,i})}{Q_{in} \cdot S_{NH,in}} \quad (9)$$

255 where $k_{LaAER,i}$ is the oxygen mass transfer coefficient of the aerated tank i ; $V_{AER,i}$ is the volume of the i -th
 1 aerobic tank; $S_{O_2,SAT,i}$ is the oxygen saturation concentration of i -th aerobic tank; $S_{O_2,AER,i}$ is the oxygen
 256 concentration in the aerobic tank i ; Q_{in} is the inlet flow rate fed to the biological zone; and $S_{NH,in}$ is the inlet
 3 ammonium nitrogen fed to the biological zone.
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9 R_{NAT} is a performance indicator representing the ratio between the nitrate produced and the ammonia
 10 oxidized in the aerobic reactor (Boiocchi et al., 2017b). The results of R_{NAT} can be used as a reference to
 11 understand the emissions of N_2O ; indeed, R_{NAT} indicates the degree of nitrification within the aerobic zone
 1260 and the relation between the autotrophic biomass. For instance, $R_{NAT} = 1 \text{ gNO}_3 \text{ gNH}_4^{-1}$ means that all NO_2
 13 produced by the AOB is converted into NO_3 by the NOB. R_{NAT} is calculated according to equation 10:
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$$21 \quad R_{NAT} = \frac{NO_{3,OUT,AER}^- - NO_{3,IN,AER}^-}{NH_{4,IN,AER}^+ - NO_{4,OUT,AER}^+} \quad (10)$$

22 where $S_{NO_3,IN,AER}$ and $S_{NO_3,OUT,AER}$ represent, the nitrate influent and effluent concentration inside the aerobic
 23 reactor, respectively; $S_{NH_4,IN,AER}$ and $S_{NH_4,OUT,AER}$ are the NH_4 concentrations of the influent and effluent of the
 24 aerobic reactor, respectively.
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31 The operational costs - OC (€/treated volume) represents the sum of three costs (Vanrolleghem and Gillot,
 32 2002; Guerrero et al., 2011): the costs related to the chemical consumption for membrane cleaning (CC, as €/
 33 treated volume), the energy demand (eD , €/ treated volume) and effluent fine (EF) related to pollutants
 34 discharged (in accordance with Italian regulations), according to Equation 11:
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$$41 \quad OC = eD \cdot \gamma_e + CC + EF \quad (11)$$

42 where γ_e represents the cost per kWh. Italian rates are 0.21 € / kWh.
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48 The membrane cleaning cost CC is calculated considering a typical membrane cleaning protocol (i.e.,
 49 including a chemical solution composed of 500 ppm of NaOCl and 2,000 ppm of citric acid, with a cost of
 50 0.48€ per chemical cleaning), which was held only when the transmembrane pressure (TMP) reached a value
 51 higher than 60kPa as suggested by the membrane manufacturer. The EF was assessed in accordance with
 52 Mannina & Cosenza (2015).
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59 The energy demand eD (kWh) is calculated according to Equation 12:
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$$eD = P_w + P_{eff} + P_s + P_m \quad (12)$$

where P_w , P_{eff} , P_s , and P_m represent, respectively, the energy consumption for the air blowers, permeate extraction, recycle pumps and mixers; P_w , P_{eff} , and P_s are calculated according to Mannina et al. (2019). P_{eff} is proportional to the transmembrane pressure (TMP) to be imposed to the membrane to obtain a constant permeate flow rate Mannina et al. (2019). P_w was calculated for both aerobic (P_{w3}) and membrane bioreactor (P_{w4}), while P_m comprised the energy used for constantly mixing the anaerobic and anoxic tanks. It was assumed that both tanks required 0.008 kWh per m^3 tank volume (Tchobanoglous et al., 2003; Maere et al., 2011).

Total direct emissions (DE) represent the sum between CO_2 and N_2O stripped from the liquid to the gas phase (Mannina et al., 2018a), while the indirect emissions (IE) can be evaluated by multiplying eD by γ_{CO} (equal to 0.245 $kgCO_{2,eq} / kWh$); γ_{CO} represents the specific CO_2 emission due to the energy consumption (EIA, 2009). DE and IE are both expressed in terms of carbon equivalent ($kgCO_{2,eq} m^{-3}$) with the aim to obtain comparable units in terms of GHG emissions. For a more detailed description of the performance indicators, the reader is referred to Mannina et al. (2019).

3. Results and discussion

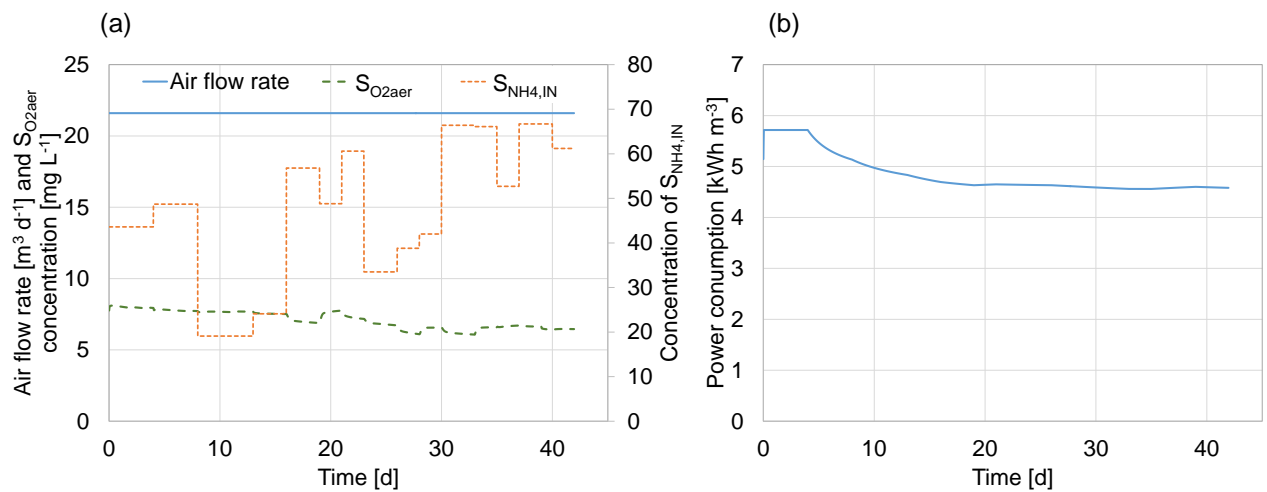
3.1 Scenario 0

Figure 2a reports the patterns of the air flow rate supplied to the aerobic reactor, along with the influent ammonia concentration ($S_{NH_4,IN}$) and of the dissolved oxygen concentration inside the aerobic reactor ($S_{O_{2aer}}$) for Scenario 0. Figure 2b shows the trend of the total power consumption (of the entire plant) inside the pilot plant for Scenario 0.

Data reported in Figure 2a show that, during Scenario 0 no air flow control has been implemented. Indeed, a constant air flow rate ($21.6 m^3 d^{-1}$) was supplied to the aerobic reactor disregarding the amount of influent ammonia to be oxidized and the amount of dissolved oxygen inside the aerobic reactor.

As shown in Figure 2a, the influent ammonia concentration has considerable fluctuation during the 42 days of simulation. Indeed, ammonia ranged between 19 and 67 $mg L^{-1}$. Despite the ammonia variability, the high air flow rate supplied to the aerobic reactor led to a quite high DO concentration inside the aerobic reactor.

306 Indeed, the average $S_{O_{2aer}}$ maintained inside the aerobic reactor was equal to 7.2 mg L^{-1} . This latter value is
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 307 much higher than the dissolved oxygen value suggested in literature for the aerobic processes (i.e., $1.5\text{-}2 \text{ mg}$
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 308 L^{-1}) (Metcalf, & Eddy, 2003). Consequently, an high energy consumption has been observed throughout the
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 309 entire simulation period. On average, 4.8 kWh m^{-3} of energy was consumed by the plant. This latter value is
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 310 almost doubles the average power consumption reported for MBRs treating similar wastewater (Krzeminski
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 311 et al., 2012). Almost 87% of the total power consumption was related to the aeration inside the aerobic
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 312 reactor. This result suggests that the open-loop aeration scenario is highly inefficient and the high energy
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 313 consumption can be translated into potential energy recovery for the plant under study (Solon et al., 2017).



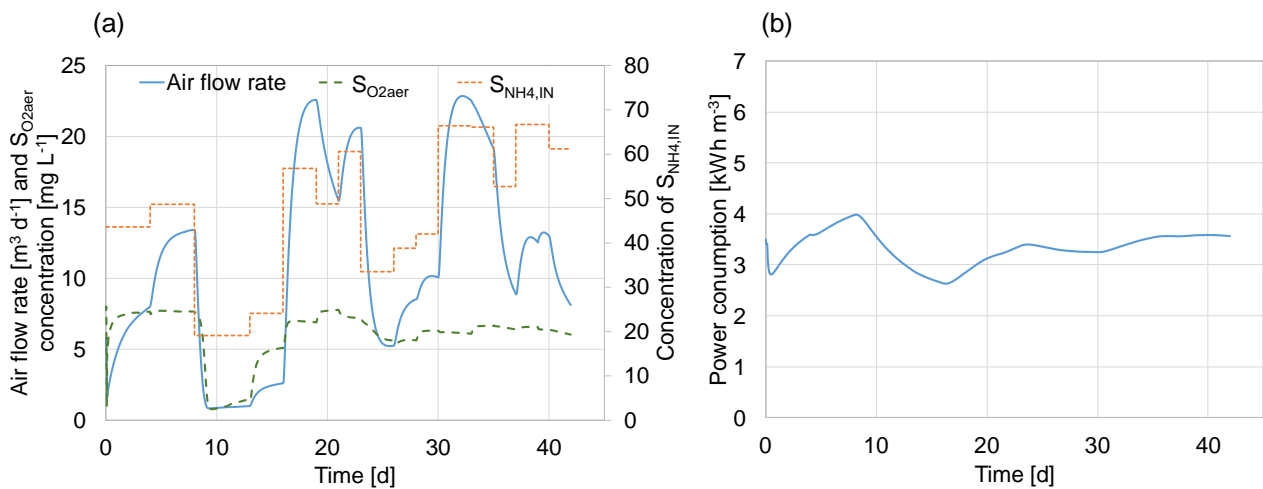
314
 315 **Figure 2.** The pattern of airflow rate and dissolved oxygen ($S_{O_{2ae}}$) within the aerobic reactor and influent
 316 ammonia concentration ($S_{NH_4,IN}$) (a) and power consumption (b) for Scenario 0.

317 318 **3.2 Scenario 1**

319 In Figure 3a, the trends of q_{air} , $S_{O_{2aer}}$ in the aerobic reactor and $S_{NH_4,IN}$ for Scenario 1 are reported. Figure 3b
 320 shows the total power consumption inside the pilot plant for Scenario 1 over the modelling period.

321 As shown in Figure 3a the air flow rate during scenario 1 varies according to $S_{NH_4,IN}$ since the DO setpoint is
 322 controlled on the basis of the ammonia inside the aerobic reactor. Thus, results show a reduction in air flow
 323 rate and DO inside the aerobic reactor. In particular, the average air flow supplied to the aerobic reactor is
 324 equal to $11.5 \text{ m}^3 \text{d}^{-1}$ (almost half of the value reported in Scenario 0). While the dissolved oxygen
 325 concentration inside the aerobic reactor ranges between 0.7 and 7.2 mg L^{-1} . It is important to highlight the

326 beneficial effect of controlling air flow rate in terms of power consumption. Indeed, the average power
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 327 consumption was equal to 3.3 kWh m^{-3} , which is lower than that obtained for Scenario 0. Thus, a substantial
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 328 reduction (namely, 32%) in terms of power consumption occurred during Scenario 1 with respect to Scenario
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 329 0. This value is slightly higher than that obtained by Sun et al. (2016) (from 15 to 20%) for a full-scale MBR
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 330 where the same control strategy of Scenario 2 was applied. The difference between both studies may be
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 331 related to the fact that Sun et al. (2016) presented results considering the whole WWTP, while the current
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 332 work is focused only on the activated sludge process and MBR.



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 335 **Figure 3.** The pattern of air flow rate supplied to the aerobic reactors and dissolved oxygen inside the
 336 aerobic reactor, and influent ammonia concentration (a) and power consumption (b) for Scenario 1.

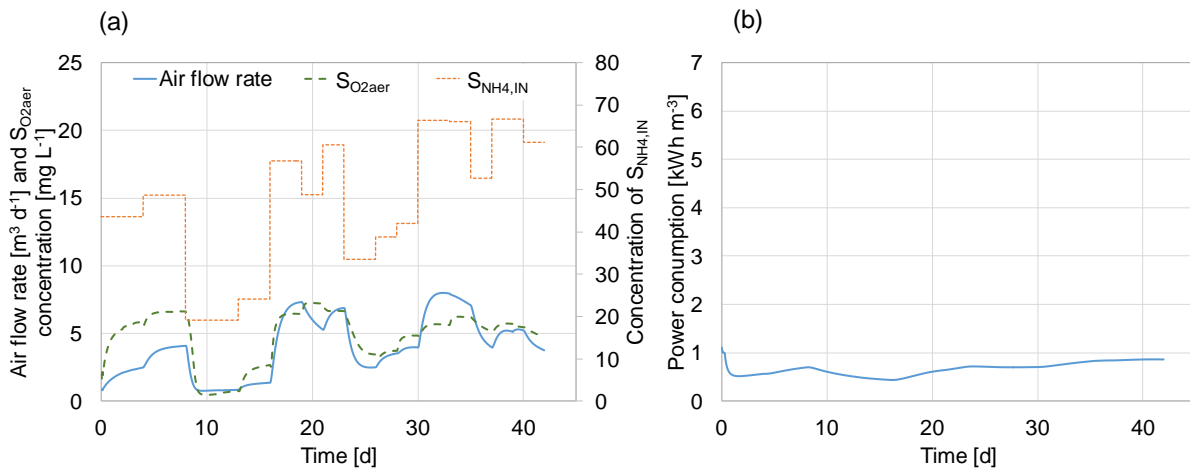
337 3.3 Scenario 2

338 Figure 4a shows the trend of the air flow rate and $S_{O_{2aer}}$ in the aerobic reactor and of $S_{NH_{4,IN}}$ for Scenario 2.

339 Figure 4b shows the total power consumption inside the pilot plant for Scenario 2 throughout the modelling
 340 period.

341 Results reported in Figure 4a show a substantial reduction, respect to previous scenarios, both in terms of air
 342 flow rate and $S_{O_{2aer}}$. Indeed, differently to previous scenarios, the aeration flow rate was adjusted not only
 343 with respect to the ammonia inside the aerobic reactor, but also taking into account the nitrite concentration.
 344 The air flow rate, and consequently $S_{O_{2aer}}$, follows the trend of influent ammonia. In particular, the air flow
 345 rate varied between 0.76 and $21.6 \text{ m}^3 \text{ d}^{-1}$, which are lower respect to previous scenarios. The obtained value

346 of oxygen concentration ($S_{O_{2aer}}$) is able to ensure proper aerobic conditions inside the aerobic reactor, with
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 347 values ranging between 0.45 and 7.2 mg L⁻¹. The substantial decrease of the average air flow rate provided a
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 348 very low power consumption in the plant under study (equal to 0.7 kWh m⁻³). The obtained power
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 349 consumption is in accordance with previous studies related to real MBR plants, which found an energy-
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 350 specific consumption ranging between 0.62 and 0.75 kWh m⁻³ (Giesen et al., 2008; Wallis-Lage and
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 351 Levesque, 2009; Fenu et al., 2010). As discussed above, these results have substantial implications in terms
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 352 of indirect and direct GHG emissions.



353
 354 **Figure 4.** The pattern of airflow rate inserted inside the aerobic reactors, dissolved oxygen inside the
 355 aerobic reactor – $S_{O_{2aer}}$ and influent concentration of ammonia - $S_{NH_4,IN}$ (a) and power consumption (b)
 356 for Scenario 2.

358 3.4 Comparison among scenarios

359 In this section, the comparison of the three analyzed scenarios is presented in terms of PIs. More in detail,
 360 Figure 5 reports the results in terms of average effluent fine (EF), operating costs (OC), R_{NAT} , RON, direct
 361 and indirect emissions, and dissolved N₂O inside the aerobic reactor ($S_{N_2O_{aer}}$) for each analyzed scenario. For
 362 sake of completeness, in Table 2 the values of all PIs obtained for each scenario are also reported.

363 As shown in Figure 5a, the EF value was not affected by the control strategies, ranging between 0.099 and
 364 0.108 € m⁻³ (Table 2). This slight difference is due to twofold reasons: i. the membrane presence, ii. the
 365 sufficient dissolved oxygen for all scenarios in aerobic reactor. Indeed, for all scenarios, an excellent effluent

366 quality has been achieved due to the membrane solid/liquid separation, which guarantees the retaining of all
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367 suspended compounds. Moreover, the dissolved compounds have been adequately removed thanks to the
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368 sufficient dissolved oxygen concentration within the aerobic reactor during the all scenarios. Therefore, the
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369 air flow rate reduction did not affect the biological treatment because even the minimal DO concentration
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370 during the simulations was enough for biomass survival and sufficient for the system adequate performance
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1371 in terms of nutrient removal.

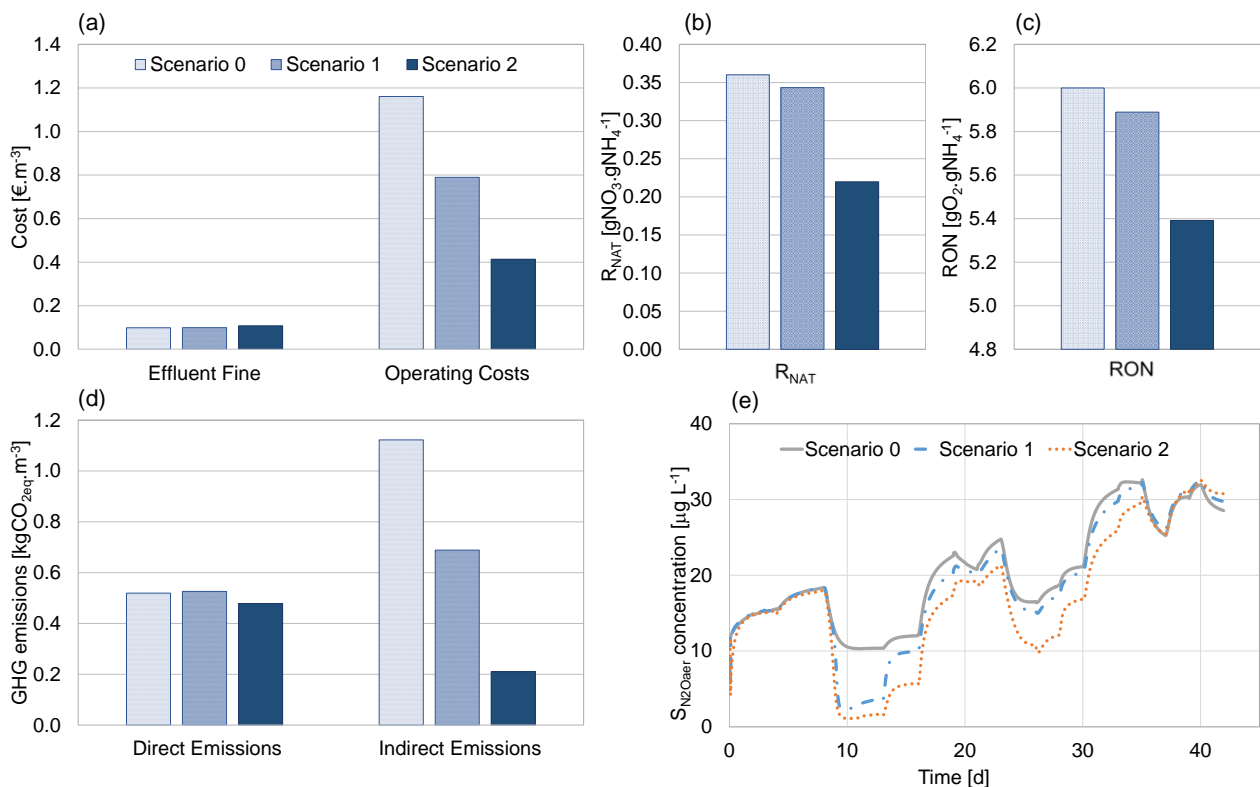
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1372 On the other hand, the reduction of the air flow rate had substantial implications in terms of operating costs.
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1373 As reported in Figure 5a, the obtained average value of operating costs was equal to 1.16, 0.78 and 0.41 € m⁻³
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1374 for Scenarios 0, 1 and 2, respectively, presenting a reduction of 35% of operating costs ranging from
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2175 Scenario 0 to Scenario 1 and of 64% from Scenario 0 to Scenario 2. This latter result is in accordance with
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2376 previous studies stating that aeration has a key role in the operating costs (Xiao et al., 2014; Sun et al., 2016)
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2577 and confirm the great advantage in aeration-based control strategies. Despite the energy demand due to the
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2778 membrane aeration (P_{w4}) was not controlled/varied by means of the controller, the amount of energy required
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3079 for the permeate extraction (P_{eff}) was influenced by the aeration of the aerobic reactor. Indeed, from scenario 0
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3280 to scenario 2, it was obtained a TMP reduction of 30%.

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3581 Both R_{NAT} and RON have been reduced during the closed-loop scenarios with respect to Scenario 0. Indeed,
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3882 as reported in Figure 5b the obtained average value of R_{NAT} was equal to 0.36, 0.34 and 0.22 gNO₃ gNH₄⁻¹
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3883 for Scenarios 0, 1 and 2, respectively. The decrease of R_{NAT} is mainly due to the low dissolved oxygen
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4284 concentration inside the aerobic reactor. However, the R_{NAT} value is always quite high to guarantee the low
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4485 nitrite accumulation inside the system (Boiocchi et al., 2016). In terms of RON a reduction from 6 to 5.4 gO₂
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4686 gNH₄⁻¹ was obtained from Scenario 0 to Scenario 2 (Figure 5c). The RON value obtained for Scenario 2 is in
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4887 accordance with literature (Boiocchi et al., 2017a). Indeed, Boiocchi et al. (2017a) reported that RON equal
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5188 or higher than 5.2 gO₂ gTKN⁻¹ represents the optimal value for obtaining the best trade-off between the
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5389 ammonia conversion rate and the N₂O emission (i.e., the lowest N₂O emission at the highest ammonium
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5590 oxidation). With this regard, Figure 5d reports a reduction of direct GHG emission for Scenario 2 in
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5791 comparison to the other two scenarios. In particular, the direct GHG emission reduced from 0.52 to 0.47
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5992 kgCO_{2eq} m⁻³ (from Scenario 0 to Scenario 2). This reduction is mainly due to the aforementioned N₂O
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393 emissions. The trend of $S_{N_2O_{aer}}$ for all scenarios is reported in Figure 5e and shows a lower concentration for
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 394 Scenario 2. This result is due to two aspects: i. the improvement of biological processes in Scenario 2 thanks
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 395 to the adequate air flow rate; ii. the reduction of the air flow rate in Scenario 2 led to the reduction of the N_2O
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 396 stripped from the soluble form to the off-gas.
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 397 In terms of indirect emission, a substantial reduction (namely, 81%) occurred from Scenario 0 to Scenario 2
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 398 (from $1.12 \text{ kgCO}_{2eq} \text{ m}^{-3}$ for Scenario 0 to $0.21 \text{ kgCO}_{2eq} \text{ m}^{-3}$ for Scenario 2). This reduction is mainly due to
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 399 the lower air flow rate supplied in Scenario 2.
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 400 The results obtained in this study are important to encourage the scattering of the MBR technology because
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 401 demonstrate that the optimization of the membrane systems in terms of their declared major issues (i.e.,
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 402 energy consumption and operating costs) may be achieved by simplified automatic systems. However,
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 403 further studies are recommended to assess the effect of automatic controls and aeration-based control
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 404 systems over membrane fouling issues.
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405
 406 **Figure 5.** Average effluent fine (EF) and operating costs (OC) (a), average value of R_{NAT} (b), average
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 407 value of RON (c), direct and indirect emissions (d), pattern of dissolved N_2O inside the aerobic reactor –
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 408 $S_{N_2O_{aer}}$ for each analyzed scenario.
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Table 2. Summary of the values of PIs obtained for Scenarios 0, 1 and 2)

PIs →	R_{NAT}	RON	$EQI_{LIQ,TOT}$	$EQI_{GAS,TOT}$	Effluent Fine	Operating Costs	Energy Consumption	Indirect Emissions	Direct Emissions
Unit→	$\frac{[gNO_3]}{[gNH_4^-]}$	$\frac{[gO_2]}{[gNH_4^-]}$	$[kg\ m^{-3}]$	$[kg\ m^{-3}]$	$[€\ m^{-3}]$	$[€\ m^{-3}]$	$[kWh\ m^{-3}]$	$[kgCO_{2,eq}\ m^{-3}]$	$[kgCO_{2,eq}\ m^{-3}]$
Scen. 0	0.360	6.0	15.49	55.39	0.099	1.16	4.8	1.12	0.52
Scen. 1	0.343	5.9	15.55	55.47	0.099	0.79	2.8	0.69	0.53
Scen. 2	0.220	5.4	16.65	55.65	0.108	0.41	0.8	0.21	0.48

4. Conclusions

The key findings of the study suggest that it is possible to find a trade-off between effluent quality, GHG emissions, energy consumption and operating costs by applying a closed-loop control system. These findings were achieved by simultaneously controlling ammonia and nitrite concentrations within the aerobic reactor. These results have substantial importance while disseminating the application of aeration-based controls in the MBR field, since the optimization of the MBR major issues may be achieved by the use of simplified automatic systems. Future studies could be performed in order of testing the PI control strategy developed here with other MBR plants configuration.

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