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1 **Optimizing resource recovery from wastewater with algae-** 2 **bacteria membrane reactors**

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14 15 **Abstract**

16 Exploiting the combination of algae and bacteria in High Rate Algal/Bacterial Ponds
17 (HRABP) is an emerging approach for wastewater remediation and resource recovery.
18 In this study, the advantage of adding a solid/liquid separation system to uncouple
19 Hydraulic Retention Time (HRT) and Solid Retention Time (SRT) is explored and
20 quantified. A long-term validated model for HRABP was run to simulate and optimize a
21 system at large scale treating digestate. It is shown that by uncoupling HRT and SRT,
22 adapting the liquid depth and the alkalinity content, the algae productivity increases from
23 9.0-14.5 g m⁻² d⁻¹ (for HRT=SRT in the range of 5 to 10 days) to 20.3 g m⁻² d⁻¹ (for HRT
24 =0.2 d and SRT= 2 d). Simulations pointed out that maximizing the algal productivity or
25 the fraction of recovered nitrogen in the algal biomass are conflicting goals that are
26 achieved under different operating conditions. Conditions maximising the algal
27 productivity favour algae and heterotrophic bacteria while algae and nitrifying bacteria

28 dominate the system under those conditions optimizing the efficiency of nitrogen
29 recycling. Finally, increasing the influent alkalinity and adapting the water depth can
30 boost the algal productivity without meeting conditions favourable to N₂O emission,
31 opening new perspectives for resource recovery through algal biomass valorisation.

32

33 **Keywords:** Wastewater remediation, membranes, process optimization, algae-
34 bacteria, nitrous oxide, emerging pollutants.

35

36 **1. Introduction**

37 Exploiting the combination of algae and bacteria in High Rate Algal/Bacterial Ponds
38 (HRABP) is an emerging technology for wastewater treatment addressing the most
39 critical aspects of wastewater treatment [1],[2]. Compared to conventional biological
40 processes, HRABP do not need an external oxygen supply, which is known to account
41 for more than 50% of the energy required for wastewater treatment [3]. Recycling
42 nitrogen and phosphorus into the algal biomass is another remarkable opportunity
43 offered by this process [4], since algae can be used as a source of energy or feedstocks
44 [5],[6], *i.e.* with their lipid fraction for biofuels or proteins for bioplastics [7],[8]. Moreover,
45 HRABP have been highlighted for their ability to remove emerging contaminants. These
46 molecules are persistent in the environment, impact on the reproductive systems of
47 aquatic organisms, and bioaccumulate in the food chain [9]. The main emerging
48 pollutants are pesticides, personal care products, pharmaceuticals and flame retardants
49 [6,7]. Conventional wastewater treatment technologies are not designed to remove or
50 degrade these compounds [10], while microalgae have been proven to be a powerful
51 technology for bioremediation of emerging contaminants [5],[12],[13]. Indeed, in the
52 highly oxidizing environment that is achieved thanks to high oxygen levels, microalgae

53 can catalytically degrade complex compounds, with efficiencies ranging from 30 to 80%
54 for drugs such as ibuprofen, carbamazepine, and caffeine [14],[15],[16],[17]. Combined
55 with bioadsorption and bioaccumulation, the algae process was found to achieve a
56 remarkable removal efficiency for more than 50 emerging contaminants [18].

57 Adding a separation system, like membrane modules, is a technological breakthrough
58 that leads to the uncoupling of Hydraulic Retention Time (HRT) and Solid Retention
59 Time (SRT). On top of enhancing emerging contaminants removal [6], HRT and SRT
60 separation was shown to considerably enhance the overall process efficiency [19],[20]
61 by increasing microalgal productivity by a factor 3.5 compared to a standard
62 photobioreactor [21] (see the synthesis of various literature studies in Table 1). The key
63 idea is that algal biomass productivity results from the product of net growth rate and
64 biomass concentration which is enhanced when increasing the biomass in the reactor
65 while keeping growth rate at high levels. However, a separation system makes the
66 process more flexible but also more complicated to operate and to optimize by
67 increasing the number of parameters that can be tuned. Concentrating the algae in the
68 system leads to a higher light attenuation and therefore, to a lower growth rate. In such
69 a complex situation, a multi-parametric optimization must be carried out and the
70 guidance of mathematical models has already proven very useful [22],[23]. This is
71 especially decisive in the case of complex dynamical bioprocesses involving a large
72 range of interacting species, like for anaerobic digestion [24].

73 Recently, the ALBA model (ALBA standing for ALgae-BActeria) was developed and
74 validated. This mathematical model was built to simulate algal-bacterial interactions and
75 competitions in HRABP, to evaluate bioremediation performances and to provide a
76 better understanding of the interactions among the chemical, physical and biological
77 processes. This model was validated and was proven able to accurately predict, under

78 real outdoor conditions, the complex behaviour of two HRABP pilot-plants operated in
79 two different locations and under different feeding conditions [25],[26]. The model turned
80 out to be accurate in predicting the daily and seasonal responses, even during winter
81 which was never effectively described by previously available models. In total, more
82 than 630 days of pilot-scale experimental data validated the model performance [25],[26]
83 under two different climatic conditions and with the same parameter set. The model
84 confirmed, as experimentally observed [27], that inorganic carbon could become
85 limiting, as a result of the strong competition for inorganic carbon by algae and nitrifiers.
86 It was unexpected that this limitation still happened even under continuous CO₂
87 injection; the ALBA model revealed that this was due to a shortage in alkalinity owing to
88 the limited alkalinity/nitrogen ratio in the feed, insufficient to support full nitrification. At
89 low alkalinity, inorganic carbon can no more be stored in the medium and becomes
90 limiting both for algae and nitrifiers, leading to conditions favourable to N₂O emission.
91 The high-fidelity ALBA model is therefore a powerful validated tool to assess the process
92 productivity, the nitrogen removal rate, but also the risk of highly impacting N₂O
93 emission.

94
95

Table 1: Biomass productivity and nitrogen removal rate reported in literature for various outdoor photobioreactors, different influent, HRT, SRT and weather conditions.

| Wastewater characteristics (Av ± St.Dev) | | | | Experimental set-up | | | | | | | | Performances | | | | Ref | |
|--|------------------------------|---|--|-----------------------------------|------------------------------|----------------------|------------|------------|-----------------|--------------------------------------|----------------------|----------------------|-------------------------------------|--------------------|------------------------------------|------|---------|
| Type | COD [mg L ⁻¹] | N-NH ₄ ⁺ [mg L ⁻¹] | P-PO ₄ ³⁻ [mg L ⁻¹] | Reactor type - | Surface [m ²] | Culture depth [m] | HRT [d] | SRT [d] | Duration [d] | Season [-] | Location [-] | Biomass productivity | Unit | N removal rate | unit | | |
| SWW | 332±55.0 | 17.3±8.1 | 3.9±1.6 | raceway + membrane tank | 56 | 0.3 | 6 | 6 | 20 | Spring | Narbonne | 9.0±0.1 | gVSS m ² d ⁻¹ | 1.2±0.2 | gN m ² d ⁻¹ | [19] | |
| | | | | | | | 4 | | 20 | | | 19.9±0.5 | | 2.3±0.4 | | | |
| | | | | | | | 2.5 | | 20 | | | 28.5±0.5 | | 3.3±0.4 | | | |
| UWW | 577±31.0 | 97.6±4.6 | 17.8±1.1 | raceway + membrane tank | 32 | 0.12 | 4.8 | 4.8 | N.R. | N.R. | Almeria | 15.2±2.1 | gDW m ² d ⁻¹ | 4.1±0.3 | gN m ² d ⁻¹ | [33] | |
| | | | | | | | 3.5 | | | | | 17.0±1.4 | | 5.8±0.3 | | | |
| | | | | | | | 2.6 | | | | | 22.2±1.6 | | 7.0±0.4 | | | |
| | | | | | | | 2 | | | | | 22.6±1.6 | | 8.3±0.3 | | | |
| | | | | | | | 1.8 | | | | | 21.9±1.0 | | 9.1±0.5 | | | |
| AnMBR effluent | 71.0±35.0 | 45.0±9.1 | 4.7±35.0 | flat-panel PBRs + membrane tank | 2.3 | 0.25 0.10 | 1.5 | 4.5 | N.R. 35 | N.R. Winter | Valencia | 15.7±1.4 20.0±2.4 | gVSS m ² d ⁻¹ | 2.2±0.4 2.4±0.5 | gN m ² d ⁻¹ | [39] | |
| AnMBR effluent | 31.0±5.0 | 51.3±9.7 | 6.8±1.6 | 4 flat-panel PBRs + membrane tank | 8.8 | 0.25 | 3-4 | 4.5 9 | 16 25 | N.R. N.R. | Valencia | 12.2±1.4 7.7±1.0 | gVSS m ² d ⁻¹ | 1.7±0.1 1.3±0.5 | gN m ² d ⁻¹ | [40] | |
| Secondary effluent from WWTP | N.R. | 17.8±1.0* | 1.6±0.2* | raceway | 1.93 | 0.3 | 5 | 5 | 18 | Summer | Arcos de la Frontera | 26.2±1.2 | gTSS m ² d ⁻¹ | 0.8±0.1 | gTN m ² d ⁻² | [41] | |
| | | | | | | 0.15 | 4 | 4 | | | | 20.4±1.0 | | 0.6±0.1 | | | |
| Saline SWW | N.R. | N.R. | N.R. | raceway | 9.62 | 0.2 | 13 | 13 | 486 | Spring Summer Autumn Winter | South-West France | 1.9 | gTSS m ² d ⁻¹ | N.R. | N.R. | [42] | |
| | | | | | | | 8 | 8 | | | | 3.6 | | | | | |
| | | | | | | | 17 | 17 | | | | 2.0 | | | | | |
| | | | | | | | 17 | 17 | | | | 1.3 | | | | | |
| UWW | | | | raceway | 8.3 | 0.05 | 2.6 | 2.6 | 46 | Spring Summer Autumn Winter | Almeria | 25.6 | gTSS m ² d ⁻¹ | 96.9 | % | [35] | |
| | | | | | | 0.06 | 2 | 2 | | | | 32.7 | | 98.7 | | | |
| | | | | | | 0.05 | 3.2 | 3.2 | | | | 18.9 | | 98.8 | | | |
| | | | | | | 0.05 | 3.7 | 3.7 | | | | 12.3 | | 97.2 | | | |
| | | | | | | | | | | | | | | | | | |
| UWW | 511±101 | 75.9±17.7 | 12.5±5.1** | raceway + greenhouse | 80 | 0.135 | 10 | 10 | N.R. | Spring Summer Autumn Winter | Almeria | 7.8±4.3 | gDW m ² d ⁻¹ | N.R. | gN m ² d ⁻¹ | [36] | |
| | | | | | | | | | | | | 8.5±3.3 | | | | | 3.5±0.5 |
| | | | | | | | | | | | | 7.8±2.8 | | | | | |
| | | | | | | | | | | | | 7.2±3.8 | | | | | 3.9±0.2 |
| | | | | | | | | | | | | 16.1±3.7 | | | | | |
| | | 25.9±5.1 | | | | | | | | | | | | | | | |
| SWW | 378±57.2 | 8.0±2.1 | 13.0±3.1 | raceway | 56 | 0.28 | 5 | 5 | 443 | Spring Summer Autumn Winter | Narbonne | 18.6±1.7 | gTSS m ² d ⁻¹ | 0.5±0.03 | gN m ² d ⁻¹ | [25] | |
| | | | | | | | | | | | | 16.0±5.6 | | 0.4±0.10 | | | |
| | | | | | | | | | | | | 12.4±3.1 | | 0.4±0.04 | | | |
| | | | | | | | | | | | | 12.8±2.4 | | 0.3±0.18 | | | |
| | | | | | | | | | | | | | | | | | |
| Centrate from UWW | 112±34.0 | 244±79.0 | 5.7±0.8 | raceway | 5.78 | 0.2 | 9 | 9 | 152 | Spring, summer, autumn | Milan | 5.5±7.4 | gTSS m ² d ⁻¹ | 86±7.0 | % | [34] | |
| PWW | 678±284 | 195±55.0 | 19.0±14.0 | raceway | 3.8 | 0.2 | 7-25 | 7-25 | 208 | Spring, summer, autumn | Milan | 10.7±6.5 | gTSS m ² d ⁻¹ | 3.9±1.8 | gN m ² d ⁻¹ | [32] | |
| Centrate from PWW | 514±190 | 310±91.0 | 13.6±4.0 | raceway | 3.8 | 0.2 | 10 | 10 | 189 | Spring Summer | Milan | 10.81±2.16 | gTSS m ² d ⁻¹ | 5.3±0.6 | gN m ² d ⁻¹ | [26] | |
| | | | | | | | 10 | 10 | | | | 12.2±3.5 | | 6.9±1.3 | | | |

| | | | | | | | | | | | | | | | | | |
|-----------------------------------|----------|----------|----------|----------------------|------|------|-----|---|--------|---------------------|--------------------|----------------|--------------------------------------|----------------|------------------------------------|------------|--|
| | | | | | | 20 | 20 | | Autumn | | | 6.1±3.1 | | 4.9±1.5 | | | |
| | | | | | | 0.22 | 0.2 | 2 | | | | 43.1±2.9 | | 22.6±12.2 | | | |
| | | | | | | | 10 | 3 | | | | 19.6±2.1 | | 6.9±0.5 | | | |
| Simulated Centrate from PWW | 514± 190 | 310±91.0 | 13.6±4.0 | Simulated raceway | 2000 | 0.12 | 0.2 | 2 | 50 | Simulated Spring | Simulated Milan | 32.0±3.0 | gVSS m ⁻² d ⁻¹ | 12.8±6.8 | gN m ⁻² d ⁻¹ | This study | |
| | | | | | | | 10 | 3 | | | | 15.4±2.1 | | 3.7±0.2 | | | |
| | | | | | | | 1 | 6 | | | | 16.2±4.6 | | 10.7±3.3 | | | |
| | | | | | | 0.06 | 10 | 3 | | | | 9.8±1.5 | | 1.9±0.1 | | | |

96

97 Note. UWW (urban wastewater); AnMBR (anaerobic membrane bioreactor); WWTP (wastewater treatment plant); PWW (piggery wastewater); SWW (synthetic
98 wastewater).

99 N.R.: Not Reported

100 *Total nitrogen and total phosphorous values.

101 **Total phosphorous value

102 In this work, the ALBA model was upgraded by implementing a solid/liquid separation
103 system to investigate the complex interplay between SRT and HRT. The ALBA model
104 was then validated in this new configuration using the experimental data of Robles et al.
105 [19], carried out in an outdoor pilot reactor. The model was finally used to explore a large
106 range of scenarios, varying HRT, SRT, and liquid depth. The trade-off between algal
107 productivity and nitrogen recycling was explored, while focusing on operating modes
108 that are not susceptible of leading to N₂O emissions.

109 Finally, the simulation results are deeply discussed and compared with the experimental
110 records reported in literature for outdoor algae-bacteria systems equipped with a
111 separation system, comforting the model prediction and further quantifying the potential
112 of separation systems for wastewater remediation, algae production and nitrogen
113 recycling.

114

115 **2. Material and methods**

116

117 **2.1. Brief recall of the ALBA model: structure, validation, and implementation**

118 Simulations were run using the high fidelity ALBA model [25], simulating the dynamics
119 of HRABP. The model considers a mixed culture of photoautotrophic algae,
120 heterotrophic bacteria, Ammonium Oxidizing Bacteria (AOB) and Nitrite Oxidizing
121 Bacteria (NOB). In total, it includes 19 biological processes and involves 17 state
122 variables. The biokinetics equations are based on the Liebig's minimum law [28] for
123 limiting elements (carbon, nitrogen and phosphorus), meaning that the most limiting
124 nutrient drives the overall kinetics. Moreover, biokinetics include dependences from
125 light, temperature and pH, since they are the most influencing parameters on microbial
126 dynamics (see SI.1).

127 The ALBA model embeds an in-depth description of the chemical reactions in the
128 medium resulting in an accurate pH prediction. The pH sub-model is based on
129 dissociation equilibria and mass balances of acids and bases, extending the approach
130 proposed in the ADM1 (Anaerobic Digestion Model n.1 [29],[30]).

131 The Total Alkalinity (TA) turns out to play an important role in the HRABP dynamics,
132 since it determines the potential of inorganic carbon storage in the bulk. Its expression
133 is defined in terms of molar quantities (mol m^{-3}), as reported in Eq. 2.1.2 [26]:

$$\text{TA} = \text{HCO}_3^- + 2\text{CO}_3^{2-} + \text{H}_2\text{PO}_4^- + \text{HPO}_4^{2-} + 2\text{PO}_4^{3-} + \text{OH}^- + \text{NH}_3 - \text{H}^+ - \text{HNO}_2 - \text{HNO}_3 - \text{H}_3\text{PO}_4 \quad 2.1.2$$

134

135 The CO_2 , NH_3 , O_2 gas/liquid transfer is also included, quantifying the rate through the
136 global k_{La} and the diffusivity coefficients.

137 The model was previously calibrated and validated on two long-term datasets from two
138 pilot-plants, operated under different climatic conditions. The first reactor was a
139 demonstrative-scale raceway of 17 m^3 (with a 56 m^2 surface), located in the South of
140 France (Narbonne area), fed on synthetic municipal wastewater [25]. The second
141 reactor was a pilot-scale raceway of 1 m^3 (with a 3.8 m^2 surface), located in the North
142 of Italy (Milan area), fed on the liquid fraction of piggery digestate [26]. In total, 30 days
143 were used for the calibration phase, while 603 days were exploited for the model
144 validation based on on-line measurements (dissolved oxygen, pH, temperature) and off-
145 line measurements (nitrogen compounds, algal biomass, total and volatile suspended
146 solids, chemical oxygen demand). A more detailed description of the ALBA model, its
147 calibration and validation procedure, can be found in Casagli et al. [25], and
148 summarized in Supporting Information (SI.1 and SI.2).

149 Other models for describing algae-bacteria system are available in literature, as
150 reviewed in Casagli et al. [26], they can also be modified similarly by introducing a
151 solid/liquid separation term in the hydraulic balance.

152 The ALBA model was initially developed in AQUASIM and then implemented under
153 MATLAB R2019b. Simulations were run on a PC with 8 i9 vPRO cores. Each simulation
154 took approximately 3 minutes.

155

156 2.2. Case study

157 A theoretical case study was assumed for the optimization exercise, consisting in an
158 industrial HRABP of 2000 m² treating digestate, located in the North of Italy (Milan area).

159 Simulations were run focusing on the spring season, where the most affecting
160 environmental conditions (i.e., light and temperature) are closer to the optima for algal
161 growth. The influent was assumed to be a diluted liquid fraction of digestate with the
162 characteristics reported in Table 2 (taken from Pizzera et al., [31]).

163

164 **Table 2:** Influent characteristics and associated model state variables.

| Influent characteristics used for the simulations: liquid fraction of piggery digestate | | | |
|---|-----------|-------|---------------------------------|
| Description | Symbol | Value | Unit |
| Total ammoniacal nitrogen | S_{NH} | 310.4 | gN m ⁻³ |
| Nitrate | S_{NO3} | 11.7 | gN m ⁻³ |
| Nitrite | S_{NO2} | 0 | gN m ⁻³ |
| Inorganic soluble orthophosphates | S_{PO4} | 13.6 | gP m ⁻³ |
| Inorganic carbon | S_{IC} | 266.1 | gC m ⁻³ |
| Readily biodegradable organic matter | S_S | 133.3 | gCOD m ⁻³ |
| Inert soluble organic matter | S_I | 247.6 | gCOD m ⁻³ |
| Inert particulate organic matter | X_I | 118.9 | gCOD m ⁻³ |
| Slowly biodegradable organic matter | X_S | 13.3 | gCOD m ⁻³ |
| Dissolved oxygen | S_{O2} | 8 | gO ₂ m ⁻³ |

| | | | |
|------------------|-----------|--------|---------------------------------|
| Cations | S_{cat} | 10 | mol m^{-3} |
| Anions | S_{an} | 1e-005 | mol m^{-3} |
| pH | pH | 8.5 | - |
| Total alkalinity | TA | 500 | $\text{mgCaCO}_3 \text{L}^{-1}$ |

Note: all the biomasses (X_{ALG} , X_{AOB} , X_{NOB} , X_H) were considered as zero in the influent.

Validated model parameters were taken from Casagli et al. [25]. Only the k_{LA} value was assumed to be lower (10 day^{-1}), which is more similar to an industrial scale application. In all the simulations, the pH was controlled at 7.5 with CO_2 bubbling (see Supp Info SI.3).

2.3. Implementation of HRT and SRT decoupling

The separation system was implemented by imposing a retention factor on all the particulate variables:

$$\text{Ret} = \frac{\text{SRT} - \text{HRT}}{\text{SRT}} = 1 - \frac{\text{HRT}}{\text{SRT}} = 1 - \alpha \quad 2.3.1$$

This factor can be positive or negative, depending on the value resulting from the ratio $\alpha = \text{HRT}/\text{SRT}$. Consequently, the evolution in time of the particulate variables (X_j) that are retained by the separation system (dX_j/dt [$\text{gX}_j \text{m}^3 \text{d}^{-1}$]) was implemented in the model as follows:

$$\frac{dX_j}{dt} = \frac{Q_{IN}}{V} \cdot X_{j, IN} - \alpha \cdot X_j \cdot \left(\frac{Q_{IN} - Q_{EVAP}}{V} \right) \pm \sum_i v_{ij} \rho_i \quad 2.3.2$$

where Q_{IN} is the imposed inflow rate [$\text{m}^3 \text{d}^{-1}$]; Q_{EVAP} is the outflow leaving the system by evaporation, [$\text{m}^3 \text{d}^{-1}$]; V is the reactor volume, [m^3]; $X_{j, IN}$ is the concentration of X_j entering the system with the influent [g m^{-3}]; X_j is the concentration in the system [g m^{-3}]; v_{ij} is the stoichiometric coefficient associated to the state variable X_j and the process i and ρ_i is the rate of process i [$\text{gX}_j \text{m}^{-3} \text{d}^{-1}$].

184

185 2.4. Optimization study

186 For the optimization study, 19200 simulations were run, testing the simultaneous
187 influence of different factors on the HRABP performances: i) the HRT (0.2 - 10 d); ii) the
188 SRT (0.2 - 10 d); iii) the liquid depth (δ_L 0.06 - 1 m); and finally, the supplementation of
189 Total Alkalinity (TA, 0 – 12.5 mol m⁻³ added) through dosage of CaCO₃.

190 Simulations were run considering that HRT and SRT could be independently tuned,
191 simulating therefore cases with SRT lower than HRT (biomass harvesting with liquid
192 recirculated in the reactor) or, reversely, HRT lower than SRT (biomass retention).

193 Algal productivity [gAlg m⁻² d⁻¹] was defined as:

$$ALG_{productivity} := \frac{X_{ALG} \cdot Q_{out} \cdot \alpha}{S} \cdot \varphi \quad 2.4.1$$

194 Where X_{ALG} is the algae concentration in the pond [gCOD m⁻³]; Q_{OUT} is the outflow,
195 defined as the algebraic sum of $Q_{IN}-Q_{EVAP}$ [m³ d⁻¹]; S is the reactor surface [m²]; $\varphi=0.64$
196 is the conversion factor from COD_{ALG} to dry weight (DW) of algae (gDW-Alg.gCOD⁻¹).

197 The efficiency of nitrogen recycling rate was defined as:

$$Eff_{N_{RECYC}} [\%] = \frac{Flux_{N_{OUT,ALG}}}{Flux_{N_{IN}}} \cdot 100 \quad 2.4.2$$

198 Where: $Flux_{N_{out,ALG}}$ is the mass flow of Nitrogen [gN d⁻¹] in the harvested algal biomass
199 and $Flux_{N_{IN}}$ is the mass flow of Nitrogen [gN d⁻¹] fed to the HRABP:

$$Flux_{N_{OUT,ALG}} \left[\frac{gN}{d} \right] = X_{ALG} \cdot iN_{B_{ALG}} \cdot Q_{OUT} \cdot \alpha \quad 2.4.3$$

$$Flux_{N_{IN}} = Q_{IN} (S_{NH_{IN}} + S_{NO2_{IN}} + S_{NO3_{IN}} + S_{S_{IN}} \cdot iN_{SS} + S_{I_{IN}} \cdot iN_{SI} + X_{S_{IN}} \cdot iN_{XS} + X_{I_{IN}} \cdot iN_{XI}) \quad 2.4.4$$

200

201 where $i_{N_{BM,ALG}}$ is the N content in the algal biomass [$gN\ gCOD^{-1}$], Q_{IN} is the inflow rate
202 [$m^3\ d^{-1}$]; $S_{NH,IN}$, $S_{NO_2,IN}$ and $S_{NO_3,IN}$ are the influent concentrations of ammoniacal
203 nitrogen, nitrite and nitrate, respectively [$gN\ m^{-3}$]; $S_{S,IN}$, $S_{I,IN}$, $X_{S,IN}$ and $X_{I,IN}$ are influent
204 concentration of the state variables representing the organic matter concentration
205 [$gCOD\ m^{-3}$] as soluble degradable, soluble inert, particulate degradable, and particulate
206 inert (I) components, respectively; $i_{N_{SS}}$, $i_{N_{SI}}$, $i_{N_{XS}}$ and $i_{N_{XI}}$, [$gN\ gCOD^{-1}$] quantify their
207 nitrogen content.

208 The N recycling efficiency, and the algal biomass productivity were used as Key
209 Performance Indicators.

210 In addition, the N_2O emission risk-factor was considered to spot unsuitable operation
211 modes. Finally, a closer look at the biomass distribution among algae and bacteria
212 biomasses was helpful to better interpret the evolution of the ecosystem.

213

214 **3. Results and discussion**

215

216 **3.1. Model accuracy in predicting nitrogen removal rate and algal productivity**

217 The model prediction capability for the most relevant monitored variables (pH, S_{O_2} , S_{NH} ,
218 S_{NO_3} , S_{NO_2} and X_{ALG}) can be found in Casagli et al. [25],[26] (see also a summary in
219 Supporting Information SI.1 and SI.2) and it covers an uncommonly broad range of
220 conditions, including all the seasons, and HRT (=SRT) ranging from 2.6 to 20 days (see
221 S.I.7).

222 The total ammoniacal nitrogen (TAN) removal rate ($gN\ m^{-2}\ d^{-1}$) and the Total Suspended
223 Solids (TSS) productivity ($gTSS\ m^{-2}\ d^{-1}$) were also computed from the experimental
224 measurements and compared to the model predictions (Fig. 2 and 3 for summer, autumn
225 and winter, while spring results are shown in Supporting information, SI.4). It is worth

226 mentioning that algal productivity is rarely experimentally measured in algae-bacteria
227 consortia, since only TSS or VSS (Volatile Suspended Solids) are monitored. Both
228 turned out to be accurately predicted, as shown in Figure 1 and 2, with p-values for TSS
229 productivity and TAN removal rate predictions below 1E-6. The highest TSS productivity
230 was reached in summer, for both Narbonne and Milan, with an average value of $16.0 \pm$
231 5.6 [gTSS m⁻² d⁻¹] (Summer 2018, Narbonne), 18.2 ± 2.0 [gTSS m⁻² d⁻¹] (Summer 2019,
232 Narbonne) and 12.2 ± 3.5 [gTSS m⁻² d⁻¹] (Summer 2016, Milan).

233 During autumn and winter, the TSS productivity decreased in both experimentations,
234 with an average value of 12.4 ± 3.1 and 12.7 ± 2.4 [gTSS m⁻² d⁻¹] in autumn and winter,
235 respectively, in Narbonne and of 6.1 ± 3.1 [gTSS m⁻² d⁻¹] in autumn in Milan.

236 The generally higher values obtained in Narbonne along all the seasons are mainly due
237 to the lower HRT, *i.e.* 5 days, (not considering the period where it was varied between
238 2.6 and 10, for days 240-253), compared to Milan (HRT = 10 -20 days), corresponding
239 to a lower biomass harvesting rate. The TSS productivities shown in Figure 1a,b are in
240 agreement with the experimental values reported in other literature works, where the
241 raceways were operated under similar conditions. For instance, 10 ± 6.5 gTSS m⁻² d⁻¹
242 [32]; 15.2 ± 2.1 gTSS m⁻² d⁻¹ [33]; 5.5 ± 7.4 g TSS m⁻² day⁻¹ [34] ; 12.3 gTSS m⁻² d⁻¹ as
243 maximum value during winter and 32.4 gTSS m⁻² d⁻¹ as maximum value during summer
244 [35].

245 The model was further used to estimate the contributions from the different biomasses
246 (X_{ALG} , X_{AOB} , X_{NOB} , X_H) and from inert and biodegradable compounds (X_I and X_S
247 respectively) which all together make up the TSS.

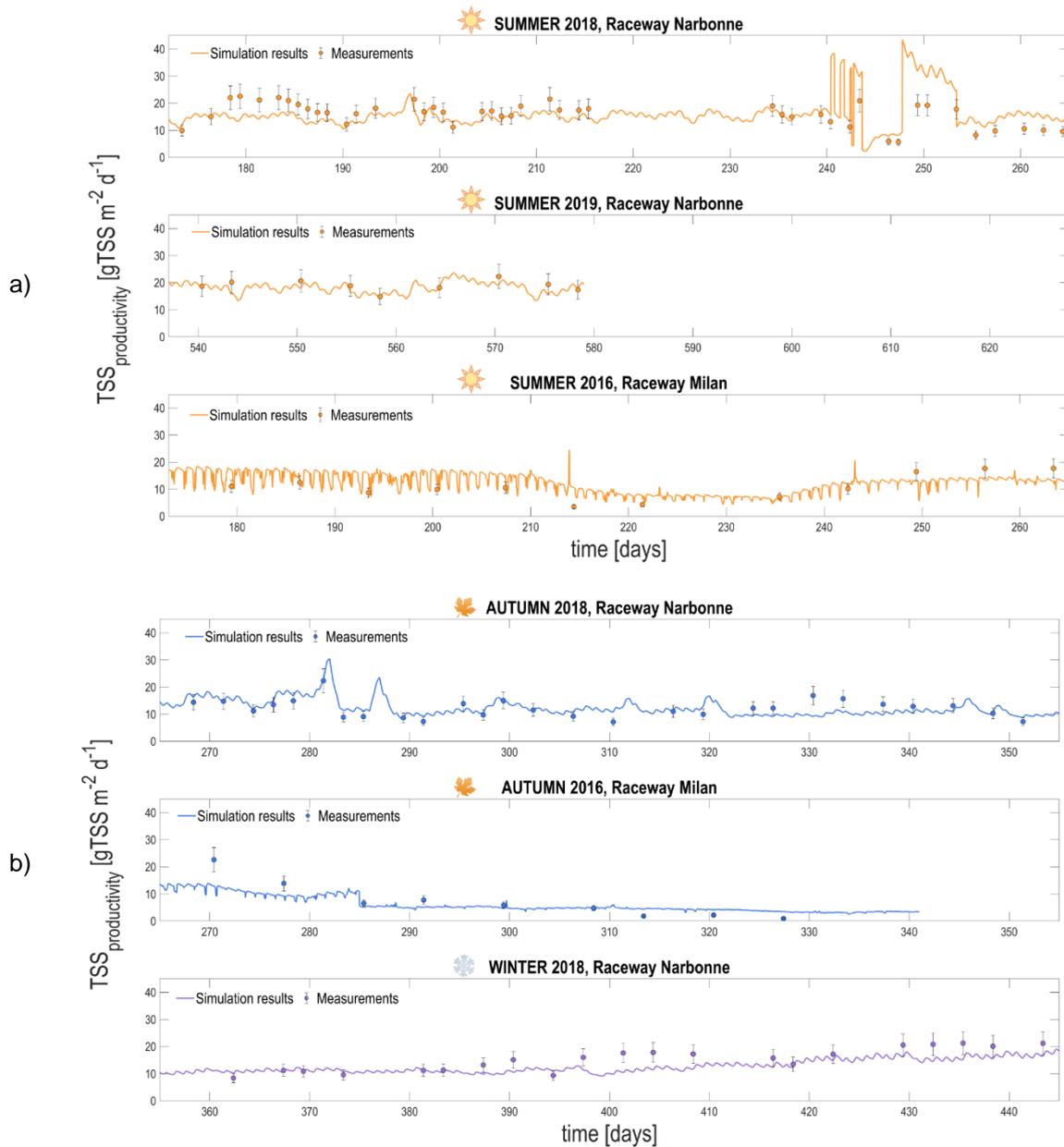
248 Algae productivity in Milan represented 72% of the TSS productivity on a yearly basis
249 (*i.e.* 7.03 gALG m⁻² day⁻¹ compared to 9.8 gTSS m⁻² day⁻¹), corresponding to a
250 microbial biomass ($X_{BIO} = X_{ALG} + X_{AOB} + X_{NOB} + X_H$) composed by 90.4% of algae on yearly

251 average. For the Narbonne case study, algae productivity was 63% of the TSS
252 productivity on a yearly bases (*i.e.* 9.5 gALG m⁻² day⁻¹ compared to 15.1 gTSS m⁻²
253 day⁻¹), corresponding to a microbial biomass where algae represented 76.6% of the
254 community on yearly average.

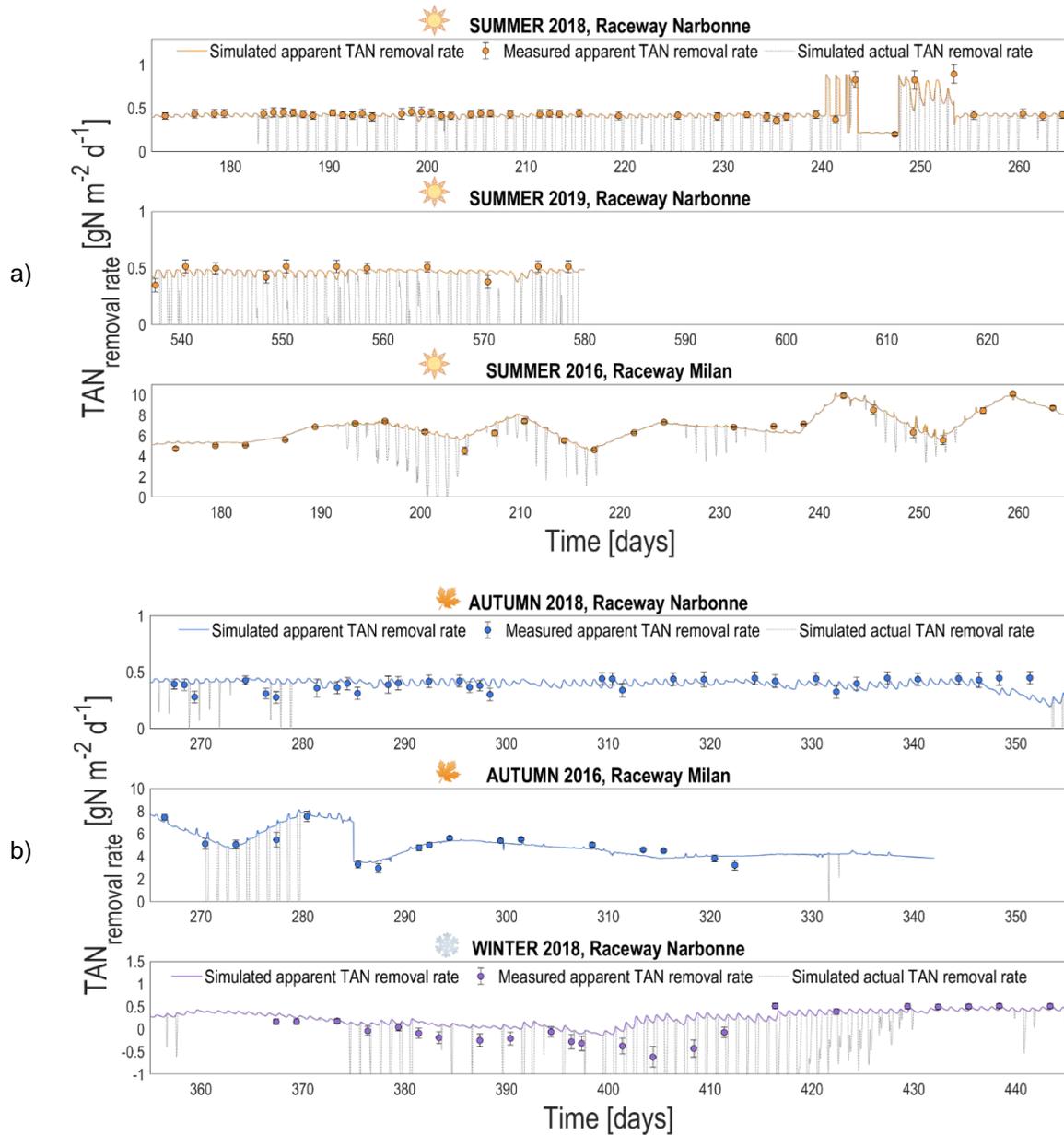
255 Looking at TAN removal rate in Narbonne (Fig. 3), it was on average 0.4 ± 0.1 gN m⁻²
256 d⁻¹ in summer, with HRT of 5 days. Peaks of 0.9 gN m⁻² d⁻¹ were reached at HRT of 10
257 days, while the lowest value was achieved for a HRT of 2.6 days (0.2 gN m⁻² d⁻¹). In
258 autumn and winter, the TAN removal rate was generally lower than 0.5 gN m⁻² d⁻¹, due
259 to the sub-optimal growth conditions for algae and bacteria. For the raceway located in
260 Milan, the TAN removal rate was higher during summer, being in a range of 4.6-10.2 gN
261 m⁻² d⁻¹ (average 6.9 ± 1.3) for a HRT of 10 days. In autumn, these values decreased,
262 varying between 3.5-8.1 gN m⁻² d⁻¹ (average 4.9 ± 1.5), even when the HRT increased
263 up to 20 days. The difference between the TAN removal rate in the two raceways is
264 mainly due to differences in the influent nitrogen, both as concentration (8 gN m⁻³ in
265 Narbonne and 310 gN m⁻³ on average in Milan) and as chemical nature (urea in
266 Narbonne, and TAN in Milan). The TAN removal rates values are in agreement with the
267 ranges reported in the work of Morillas-España et al. [36] (2.7 gN m⁻² d⁻¹ during winter
268 and 4.3 gN m⁻² d⁻¹ during summer, for an inlet N-NH₄⁺ concentration 168-210 g m⁻³), for
269 a raceway operated under similar HRT conditions (3-10 d).

270 In addition, the model allows assessing the fluxes of N-NH₃ stripped out and, therefore,
271 computing the actual TAN removal rate (grey dotted line in Figure 2, a and b) by
272 subtracting the contribution of the stripped N to the apparent TAN removal rate. In
273 Milano, the imperfect pH regulation allowed going from pH 6.5 to 7.8 increasing the free
274 NH₃ level by a factor of 30. Since TAN concentration was high in piggery digestate, this
275 corresponded to large spikes of released NH₃. In Narbonne, the TAN concentration was

276 lower, but no pH regulation was implemented so that pH could even reach values above
277 10, resulting in temporarily high NH₃ fluxes as well.



278 **Figure 1.** Total Suspended Solids productivity experimentally measured and computed with the ALBA model for
279 Milan and Narbonne case studies according to the season (a: summer; b: autumn and winter).
280



281 **Figure 2.** Total ammoniacal nitrogen removal rate, in $\text{gN m}^{-2} \text{d}^{-1}$, for both case studies (Milan and Narbonne) and
 282 according to seasons (a: summer; b: autumn and winter). The term apparent refers to the fact that the flux of N-NH_3
 283 stripped out was considered as untreated nitrogen.

284

285 3.2. Model accuracy when uncoupling HRT and SRT

286 The experimental campaign of Robles et al. [19], was used for validating the ALBA
 287 model when HRT is uncoupled from SRT. The experimental campaign was performed
 288 on the same outdoor pilot-scale raceway located in Narbonne, fed with synthetic
 289 municipal wastewater [37], previously used for the calibration and validation of the ALBA
 290 model [25]. The HRABP was operated with a liquid depth of 0.3 m and connected to a

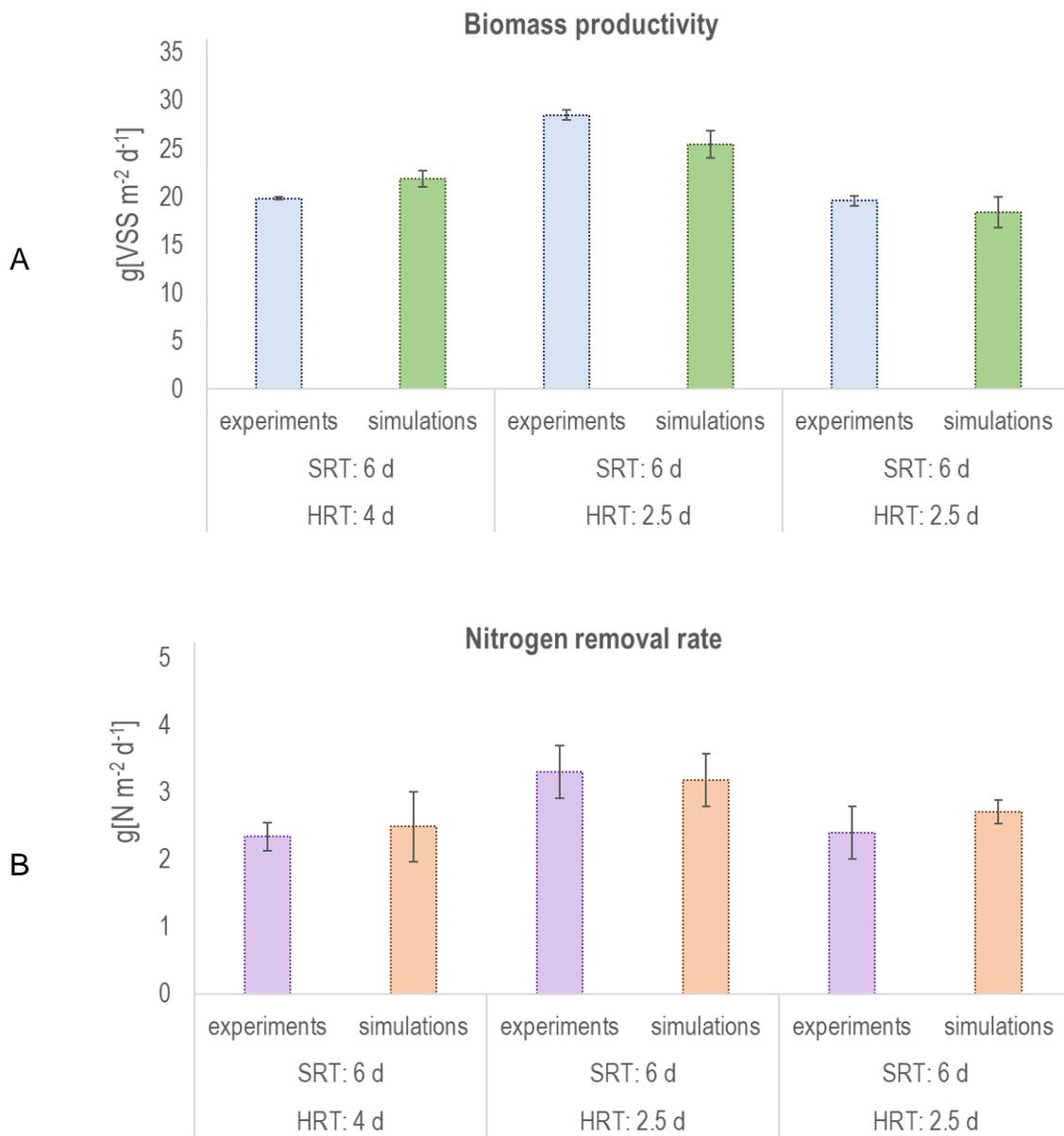
291 high-flow industrial-scale membrane compartment. The authors evaluated the process
292 performances computing the average biomass productivity and nitrogen removal rate
293 over a 20 days experimental period, for each operational condition tested. Uncoupling
294 SRT and HRT by membrane filtration improved process efficiency, with higher biomass
295 throughput and nutrient removal rate at lower HRT operation. At an SRT of 6 days, the
296 biomass productivity increased up to 19.9 ± 0.5 and to 28.5 ± 0.5 gVSS m⁻² d⁻¹ when
297 HRT was set to 4 and 2.5 days, respectively. The corresponding nitrogen removal rates
298 were 2.3 ± 0.4 and 3.3 ± 0.4 gN m⁻² d⁻¹, respectively. The authors ran also an experiment
299 with HRT set to 2.5 and SRT set to 6 days, but under sub-optimal light and temperature
300 conditions, obtaining lower values for both biomass productivity and nitrogen removal
301 rate (19.6 gVSS m⁻² d⁻¹ and 2.4 g N m⁻³ d⁻¹, respectively), compared to the experiment
302 carried out with the same operational conditions and under optimal light and temperature
303 ranges.

304 The ALBA model was run under the same operational conditions, considering the
305 climatology of the year 2018-2019 [25]. Biomass productivity and nitrogen removal rate
306 were computed according to Robles et al., [19] and simulation results were averaged on
307 a seasonal time scale. Spring 2018 presented similar average light and temperature
308 compared to the one reported by Robles et al. [19] as being optimal. The autumn
309 climatology of 2018 turned out to be comparable to the light and temperature conditions
310 reported as suboptimal by Robles et al. [19]. The model predictions, for both biomass
311 productivity and nitrogen removal rate, are perfectly in agreement with the experimental
312 results (Figure 3), with a R² of 0.78 and 0.93, respectively. Discrepancies between
313 simulated and experimental data are mainly due to the dynamic temperature and light
314 dataset used for simulations, that are not belonging to the same year as in Robles et al.
315 [19], since these data were not available. Moreover, while in the work of Robles et al.

316 [19], the measurements were averaged on 20 days of experiments for each tested
317 condition, the simulated data were averaged on the entire season (3 months for spring
318 and 3 months for autumn).

319 These recorded (and simulated) biomass productivity and nitrogen removal rates are in
320 line with other experimental results obtained in different studies testing membrane
321 separation systems uncoupling HRT and SRT in outdoor algae-bacteria systems for
322 wastewater treatment (see Table 1 for a synthetic comparison of literature studies).
323 Morillas-España et al. [33] operated a 4.4 m³ HRABP (surface 30 m², liquid depth 0.12
324 m) fed with primary domestic wastewater, at a constant SRT of 4.8 d and a variable HRT
325 of 4.8, 3.5, 2.6, 2 and 1.8 d, obtaining an algal productivity of 15.2, 17, 22.2, 22.6, 21.9
326 gDW m⁻² d⁻¹, respectively. González-Camejo et al. [38] operated an outdoor membrane
327 photobioreactor (MPBR) of 5 m², treating tertiary sewage effluent, at HRT 1.5 d, SRT
328 4.5 d and liquid depth 0.25 m. They recorded an average biomass productivity of
329 15.7±1.4 gVSS m⁻² d⁻¹. When operating the same system reducing the light path to 0.10
330 m and with HRT 4.5 d and SRT 3 d, the measured biomass productivity was 20±2.4
331 gVSS m⁻² d⁻¹ [39].

332 The simulation results of the ALBA model perfectly fall within these ranges proving its
333 prediction capacity under uncoupled HRT and SRT. Therefore, the model was further
334 used to run extensive simulations for investigating a wide set of scenarios.



335 **Figure 3.** Comparison between experimental [19] and simulation results (this study) for HRT uncoupled from SRT.
 336 A) Biomass productivity, in $\text{g[VSS m}^{-2} \text{d}^{-1}$]; B) Nitrogen removal rate, in $\text{g[N m}^{-2} \text{d}^{-1}$.

3.3. Best strategies to avoid GHG emissions: the key role of alkalinity

337
338 Simulations were run to explore optimal SRT and HRT for treating digestate. The
339 (constant) liquid depth was set to 0.225 m, typical of raceways systems. The TA in the
340 influent was set at its nominal value (10 mol m^{-3}), typical of a diluted digestate.
341 Simulation outcomes are shown in Figure 4, while the 2D version of the graphs are
342 reported in SI.5. As it was shown in Casagli et al. [26], despite pH regulation, a strong
343 competition for inorganic carbon between algae and nitrifiers can take place since
344 alkalinity is strongly reduced by nitrification. The consequence is a risk of N_2O emission
345 that can be very detrimental to the environment. In fact, it turns out that the N_2O risk
346 factor becomes nonzero for couples of $\text{HRT} \geq 4 \text{ d}$ and $\text{SRT} \geq 2 \text{ d}$, reaching the highest
347 values (35-45%) for $\text{HRT} \geq 8 \text{ d}$ and $\text{SRT} \geq 4 \text{ d}$ (Fig.4C).

348 The maximum of algal productivity ($20.18 \text{ gAlg m}^{-2} \text{ d}^{-1}$) was obtained for $\text{HRT}=0.2 \text{ d}$ and
349 $\text{SRT}=2 \text{ d}$ (Fig.4A). This pair of hydraulic and solid retention times is not risky as for N_2O
350 emission (risk factor is 0%). However, looking at the algae concentration and the
351 efficiency in nitrogen recycling, both values are low (280 gCOD m^{-3} and 0.34%
352 respectively, see Fig.4B and 4D). Indeed, for this low HRT, the nitrogen loading rate is
353 high, leading to low efficiency of nitrogen recycling ($\text{Eff}_{\text{N,Recyc}}$). Very low SRT, typically
354 lower than 1 d, whatever the HRT, lead to algal washout, with algal biomass
355 concentration that goes to zero and so does the algal biomass productivity (Fig.4A and
356 4D). The maximum value for the efficiency in nitrogen recycling (10.4%) is obtained for
357 a very different operational regime, i.e. $\text{HRT}=10 \text{ d}$ and $\text{SRT}=3 \text{ d}$ (Fig.4B). However,
358 these conditions are associated to an unsustainable N_2O risk factor of 30% (Fig.4C).
359 The higher algae and nitrifying bacteria concentrations, indeed lead to the exhaustion
360 of the inorganic carbon pool. In addition, the algal productivity related to the highest
361 efficiency in nitrogen recycling is reduced to $12.4 \text{ gAlg m}^{-2} \text{ d}^{-1}$. This opposite trend

362 reflects a classical contrasting tendency between productivity and efficiency in nutrient
363 use, like for the nitrogen removal rate and removal efficiency in wastewater remediation.
364 This explains the conflicting trend for the Key Process Indicators (2.4.1 and 2.4.2).
365 Finally, the best operational conditions in terms of HRT and SRT for reaching a better
366 trade-off between algae productivity and nitrogen recycling, while avoiding the risk of
367 N₂O emission, appear to be when HRT ranges from 2 to 5 d and SRT from 2 to 3 d.
368 However, under these conditions, the efficiency of nitrogen recycling is never higher
369 than 6.6% (see Fig.4A and 4B).

370 In order to provide a better picture of the gain achievable by uncoupling HRT and SRT,
371 Figure 5 reports a comparison between the algal productivity and the efficiency in
372 nitrogen recycling when no solid/liquid separation is applied (*i.e.* HRT=SRT). In this
373 case, HRT is adapted according to SRT, in order to maximize either algal productivity
374 or nitrogen recycling efficiency. Figure 5A clearly shows that higher algae productivity
375 are achieved for all the SRT values between 2 and 10 days, when operating the system
376 with an HRT of 0.2 days. A doubling in algal productivity can be obtained by
377 appropriately modulating HRT compared to the reference case of SRT=HRT. Figure 5B
378 shows that the same is valid for the nitrogen recycling efficiency, that reaches higher
379 values when these two operational parameters are decoupled (imposing HRT 10 days
380 at varying SRT between 2 and 10 days). Nitrogen recycling efficiency could be tripled
381 compared to the standard case when HRT=SRT.

382 Additional simulations were also run, by increasing the influent TA from 10 up to 15,
383 17.5, 20 and 22.5 mol m⁻³ (see Table SI.6.1 in SI.6). For an influent alkalinity increased
384 up to 15 and 17.5 mol m⁻³, inorganic carbon limiting conditions can still be reached when
385 applying HRT ≥ 6 d and SRT ≥ 4 d, with a maximum value of N₂O risk factor of 45% for
386 HRT=10 d and SRT=5, 6 and 7 d. On the contrary, with 20 and 22.5 mol m⁻³ of influent

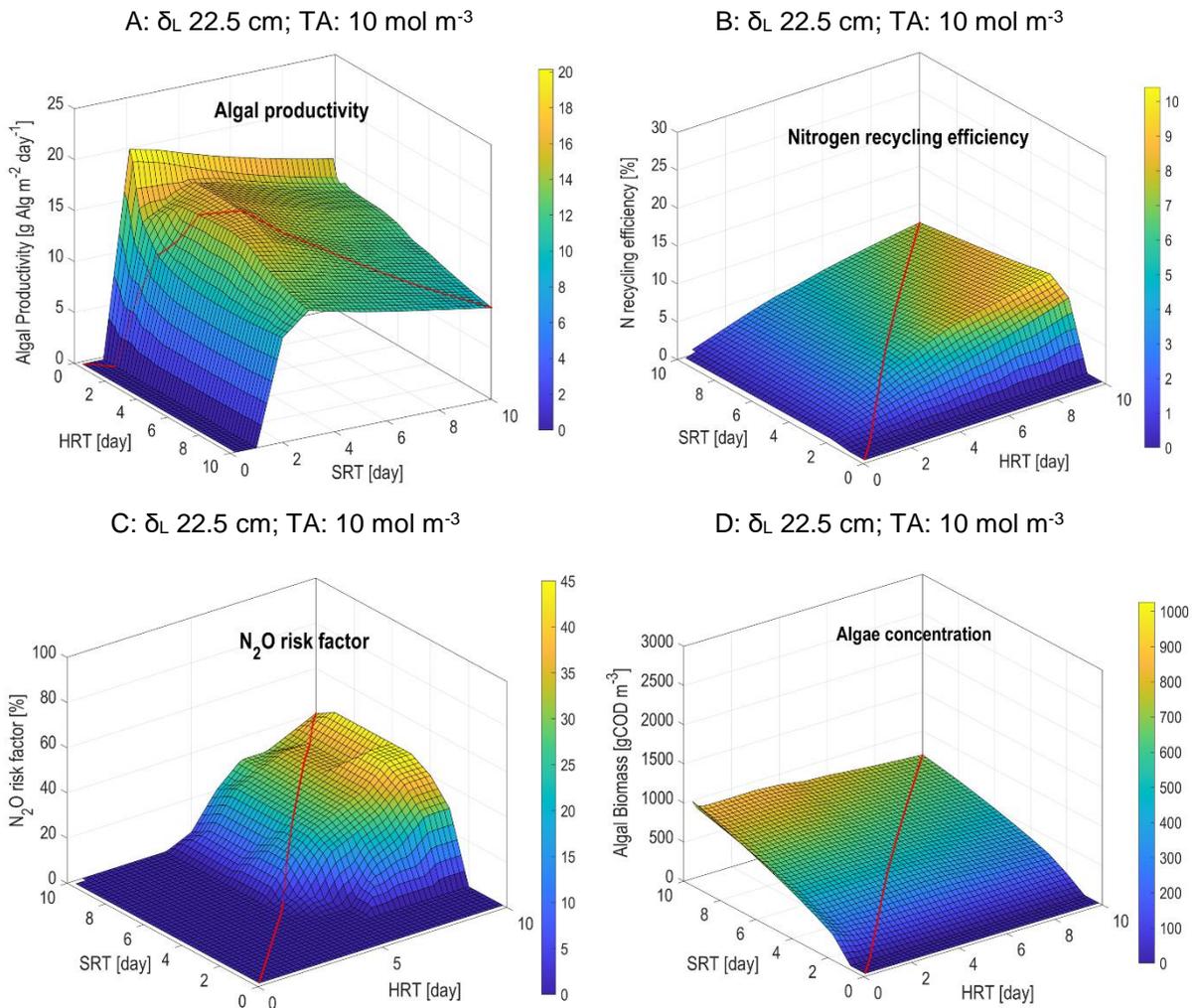
387 TA, the N₂O risk factor is always zero, indicating that this level of alkalinity (0.90 and
388 1.02 mol molNH₄⁺¹ respectively, corresponding to 2.51 and 2.82 gCaCO₃ gNH₄⁺¹) is
389 adequate to fully support both algae and nitrifiers carbon request.

390 TA supplementation appears to be particularly beneficial for algae-bacteria systems
391 when the aim is to treat wastewater, since it allows to set higher HRT and SRT,
392 favouring: i) slow growers, such as AOB and NOB; ii) the inorganic carbon availability
393 to guarantee the full nitrification process, simultaneously avoiding the competition with
394 algae; iii) reduced NH₃ stripping conditions due to the improved TAN removal (see
395 Fig.7D1 and 7D2) and, thus, lower TAN concentration in the reactor. The environmental
396 benefit of this strategy is therefore evident.

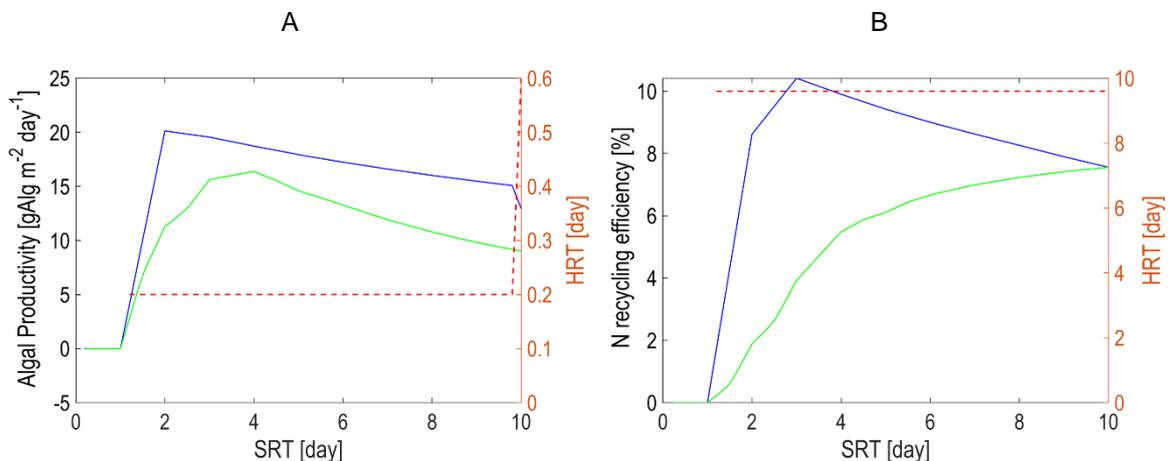
397 It must be highlighted that alkalinity addition should be adjusted carefully to avoid a
398 marked increase in CO₂ emissions due to overloading the system with inorganic carbon,
399 especially when the pH in the raceway is controlled with CO₂ bubbling. Ideally, it should
400 be regulated according to the CO₂ saturation level in the pond, that is strictly related to
401 water temperature. In fact, the driving-force in the gas-liquid exchange is given by the
402 difference between the gas saturation level in the liquid phase and the actual
403 concentration of the dissolved gas. With TA addition, the buffering capacity of the
404 system increases, and so does the level of soluble inorganic carbon. The risk is that
405 when an external source of CO₂ is injected in the system for pH regulation, most of it is
406 lost to the atmosphere.

407 When considering simultaneously the algal biomass productivity and the nitrogen
408 recycling optimization, the situation does not change significantly. Regulating TA avoids
409 N₂O emissions, but it does not allow to simultaneously reach these two conflicting
410 objectives.

411



412 **Figure 4.** Simulation results at varying HRT (0.2-10 d) and SRT (0.2-10 d). Influent TA is 10 mol m⁻³ and depth, δ_L,
 413 is 0.225 m. A: algal biomass productivity [gAlg m⁻² d⁻¹]; B: Nitrogen recycling efficiency in the algal biomass [%]; C:
 414 N₂O risk factor (percentage of time along the day for which N₂O formation conditions occur, *i.e.* inorganic carbon <
 415 0.2 molC m⁻³), [%]; D: Algal biomass concentration in the reactor [gCOD m⁻³]. The red line represents the scenario
 416 with no solid/liquid separation (*i.e.* HRT = SRT).
 417
 418



419 **Figure 5.** Simulation results for TA=10 mol m⁻³ and δ_L=0.225 m, comparing the scenario with no
 420 solid/liquid separation (*i.e.* HRT=SRT, green continuous line) and the scenario where HRT (red dotted
 421 line) is adapted according to the SRT in order to maximize: A) algal productivity [gAlg m⁻² d⁻¹] and B)
 422 nitrogen recycling efficiency [%], (blue continuous line).

423

424 **3.4. Unravelling the influence of liquid depth**

425 Two additional sets of simulations were run, considering the liquid depths of 0.12 and
426 0.06 m, first with the nominal alkalinity in the influent (10 mol m^{-3}). Results are shown in
427 Figure 6.

428 For $\delta_L=0.12 \text{ m}$, the maximum of algal productivity is $19.4 \text{ gAlg m}^{-2} \text{ d}^{-1}$ for $\text{HRT}=0.2 \text{ d}$ and
429 $\text{SRT}=2 \text{ d}$ (Fig. 6A), which is slightly lower than for the nominal depth ($\delta_L=0.225\text{m}$) for
430 the same HRT and SRT. The corresponding concentration of nitrogen recycled in the
431 algal biomass is doubled (0.6%, Fig. 6B). Indeed, at constant HRT, the incoming
432 nitrogen load per surface unit is lower for lower depth, while the incoming nitrogen load
433 per volume unit is constant. The lower areal productivity, even if the algal concentration
434 in the reactor is higher (Fig.7 A1), is due to the lower flowrate Q_{OUT} , necessarily
435 associated to a lower δ_L for the same HRT, eventually resulting in a lower algal effluent
436 flow rate. For this pair of HRT-SRT values, there is no risk of N_2O emission, but the
437 nitrogen recycling efficiency is still very low.

438 The maximum nitrogen recovered as algae biomass (11.8%) is obtained for $\text{HRT}=10 \text{ d}$
439 and $\text{SRT}=3 \text{ d}$, but for this pair of values the N_2O risk factor is 35% (Fig. 6B and 6C). So
440 far, the best working range of both HRT and SRT, for guaranteeing elevated algal
441 biomass productivity and efficiency in nitrogen recycling is 2-5 d, similarly to the case
442 $\delta_L=0.225\text{m}$. In comparison, the nitrogen uptake from the algal biomass is higher and the
443 best trade-off between algal biomass productivity ($15.25 \text{ gAlg m}^{-2} \text{ d}^{-1}$) and nitrogen
444 recycling (7.03%), avoiding N_2O favourable conditions, is obtained for $\text{HRT}=3 \text{ d}$ and
445 $\text{SRT}=5 \text{ d}$.

446 Decreasing δ_L to 0.06 m (assumed to be the lowest operational depth for this kind of
447 systems), the maximum of algal biomass productivity ($15.2 \text{ gAlg m}^{-2} \text{ d}^{-1}$) is obtained for

448 HRT=1 d and SRT=6 d (Fig. 6D), with a corresponding nitrogen recycling of 4.7% (Fig.
449 6E) and algae concentration in the reactor of 2603 gCOD m⁻³ (see Fig. 9A1). Compared
450 to the the two-fold depth ($\delta_L=0.12$ m), the maximum productivity is lower, even if the
451 algal concentration is almost 5 times higher. This is due to a much lower nutrient influent
452 load for 6 cm of liquid depth. Correspondingly, the nitrogen uptake in the algal biomass
453 is higher. In addition, under this condition of HRT and SRT, the N₂O risk factor is 0%
454 (Fig. 6F). The best result for nitrogen recycling efficiency (12.4%) was found for HRT=10
455 d and SRT=3 d, but it corresponds to a N₂O risk factor of 45% (Fig. 6F).

456 Finally, for this low liquid depth competition for inorganic carbon is stronger. It results
457 that the N₂O emission risk appears for a wider range of cases compared to $\delta_L=0.12$ m.
458 In fact, while every value between 2 and 10 d can be applied for the SRT, the HRT
459 should stay lower than 1 d, or being increased: i) up to 7, imposing SRT=2 d; ii) up to 3,
460 imposing SRT=3 d; iii) up to 2, imposing SRT=4-5 d.

461 Additional simulations were run increasing the influent TA (see Figure 7 and 8), in order
462 to see the coupled effect of working at low liquid depth without alkalinity shortage in the
463 reactor.

464 Indeed, the TA influent addition allows to explore a wider range of HRT and SRT
465 (proportionally to the alkalinity added) without operating under inorganic carbon
466 limitation. This allows finding a much better trade-off between algal productivity and
467 nitrogen recycling efficiency. Total alkalinity must be at least 20 mol m⁻³, to guaranty that
468 the N₂O risk factor is zero (Fig. 7C) for all HRT and SRT values.

469 Figure 8A and B show more explicitly which is the advantage for algal production and
470 nitrogen recycling respectively, when decoupling HRT and SRT, compared to the
471 scenario without membrane separation application. Adapting the HRT for each SRT

472 value can lead to a threefold increase in algal productivity or in the nitrogen recycling
473 rate.

474

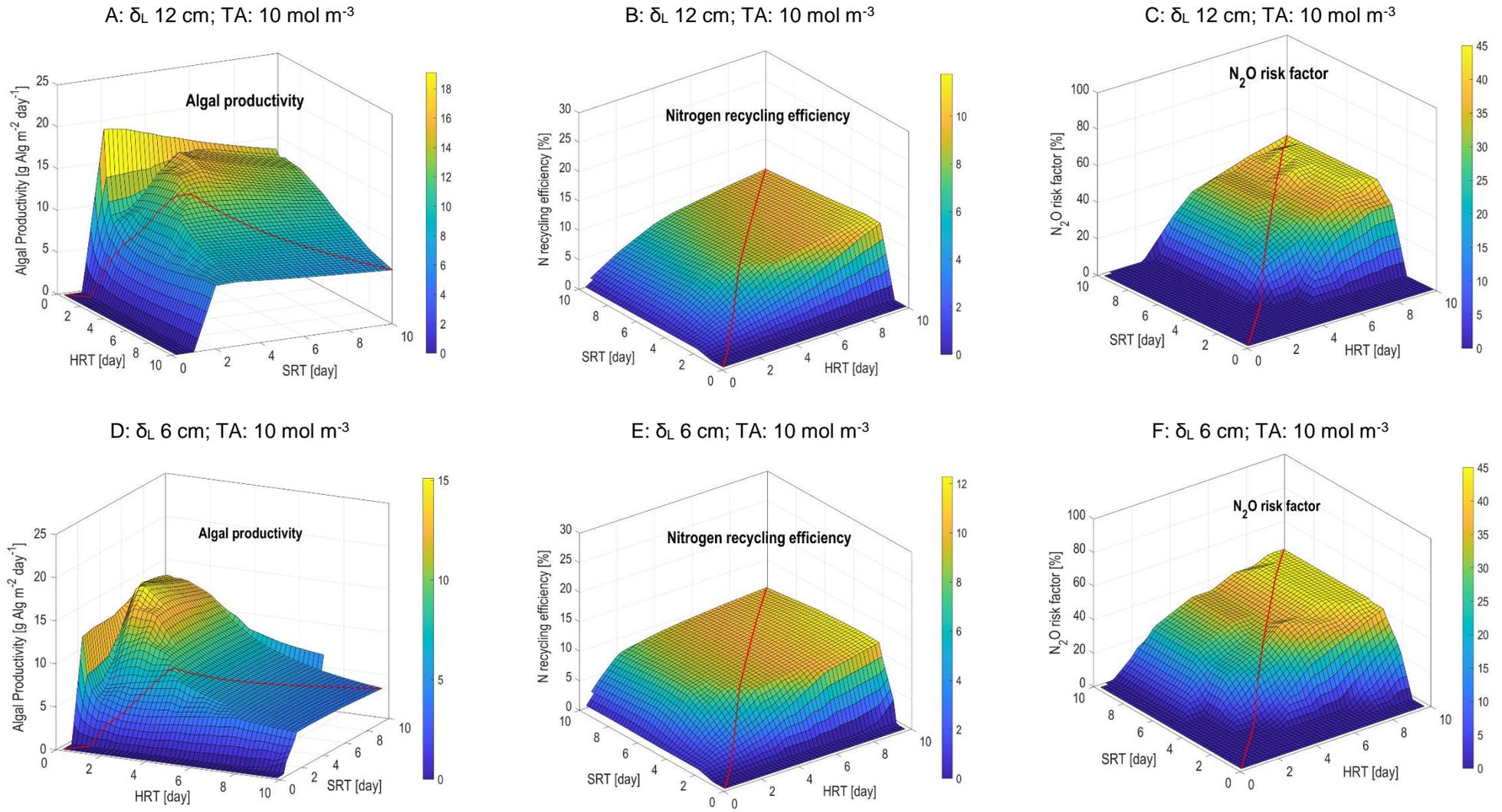
475 **3.5. Analysis of the optimal regimes**

476 Figure 9 summarizes those conditions maximising algae productivity (left column) and
477 nitrogen recycling (right column), for three liquid depths (δ_L : 0.22, 0.12 and 0.06 m).
478 Algae and heterotrophic bacteria are indeed favoured (see Fig.9 A1) for the HRT-SRT
479 pair that maximise the algae productivity (HRT 0.2 d, SRT 2d, δ_L 0.22 and 0.12 m), while
480 nitrifying bacteria are washed-out from the system. Nonetheless, even if algae
481 productivity reaches higher values, between 18 and 21 gAlg m⁻² d⁻¹ (Fig.9 B1), the
482 corresponding nitrogen recycling remains below 1% (Fig.9 C1). The biomass
483 composition drastically changes for HRT 1d, SRT 6 d and δ_L 0.06 m. Algae are more
484 concentrated (with a concentration factor, compared to δ_L 0.12 and 0.22 m, ranging
485 between 5 and 9.5, respectively), and nitrifying bacteria reach concentrations
486 comparable to those of heterotrophic bacteria (Fig.9 A1). In this case, the corresponding
487 algae productivity is slightly lower (15 gAlg m⁻² d⁻¹), due to the decreased harvesting
488 rate. However, the associated nitrogen recycling is higher (4.7%, see Fig.9 C1),
489 suggesting this operational configuration as the best trade-off between algae
490 productivity and nitrogen recycling when the target objective is the algae productivity.
491 The beneficial effect of increasing the influent alkalinity is mostly evident for TAN
492 removal rate (Fig.9 D1), that is always higher than 10 gN m⁻² d⁻¹ (reaching 22.6 gN m⁻²
493 d⁻¹ when operating at δ_L 0.22 m), compared to the cases where alkalinity was not added.
494 Also looking at the TAN removal rate in Fig.9 D1, the best trade-off when maximizing
495 algae productivity remains to operate the system at 0.06 m depth, HRT 1 d and SRT 3

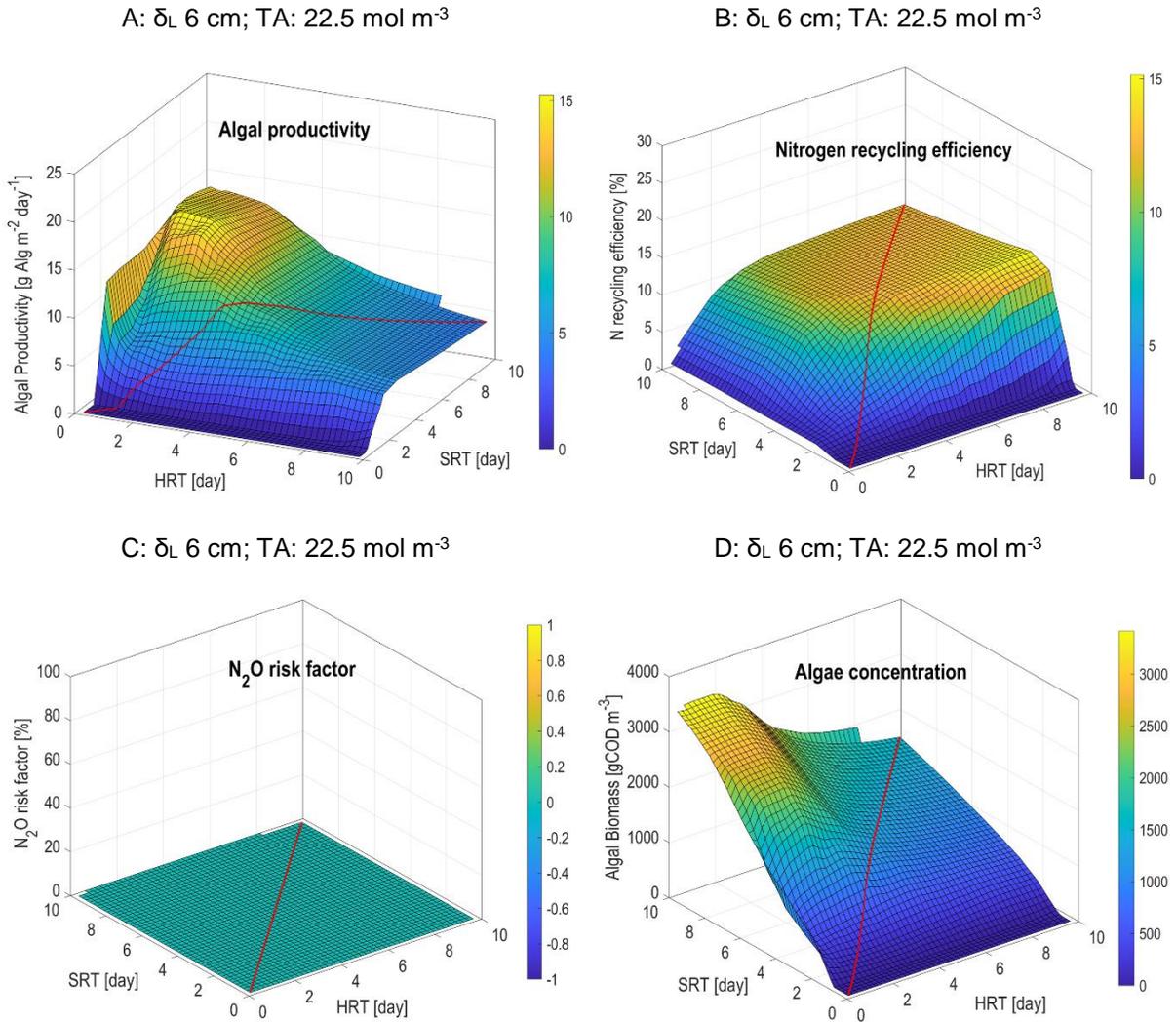
496 d, since there is not a substantial difference comparing the TAN removal rate at 0.12 or
497 0.06 m.

498 The biomass composition reported in Fig.9 A2, shows how the HRT-SRT pair
499 maximizing the nitrogen recycling in the algal biomass (10 and 3 d, respectively) favour
500 algae and nitrifying bacteria, while heterotrophic bacteria concentration remains low.
501 The highest algae productivity ($14.3 \text{ gAlg m}^{-2} \text{ d}^{-1}$, see Fig.9 B2) is obtained with alkalinity
502 addition for δ_L 0.22 m, also corresponding to the highest TAN removal rate (6.9 gN m^{-2}
503 d^{-1} , Fig.9 D2), while the best nitrogen recycling is reached for δ_L 0.06 m and with TA
504 addition (Fig. 9C2). Results reported in Fig.9 A2, B2, C2 and D2 suggest that the best
505 trade-off between algae productivity and nitrogen recycling, when the objective target is
506 the nitrogen recycling, by operating the system at δ_L 0.22 m and with TA influent
507 addition. Therefore, by analysing in detail the cluster of conditions maximizing algae
508 productivity and nitrogen recycling among the tested conditions, we can conclude that
509 operating with $\text{HRT} \leq 1 \text{ d}$ and $\text{SRT} \leq 6 \text{ d}$ allows reaching the highest values of algae
510 production, while operating at HRT 10 d and SRT 3 d allows maximising the nitrogen
511 recovery in the algal biomass (up to 15%). Given the opposite trends of these two
512 targets, the best trade-off is to run the system at lower liquid depth (0.06 m) when the
513 target is maximising the algal productivity, while operating with higher liquid depth (0.12-
514 22 m) when the objective is to maximize the nitrogen recycled in the algae.

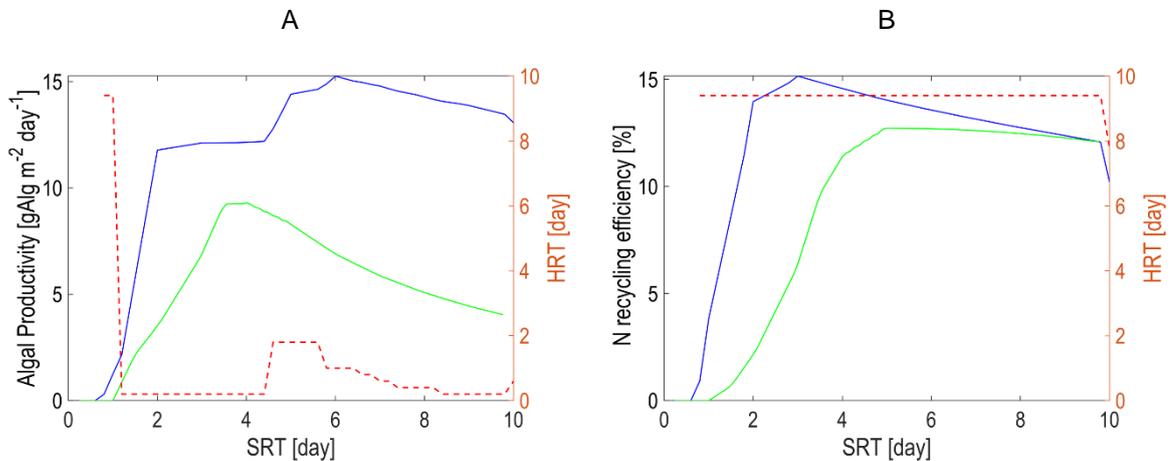
515 Moreover, results highlighted that working at lower liquid depth allows: i) to maintain a
516 higher algal biomass concentration in the reactor (requiring less energy for algae
517 harvesting), with comparable biomass productivity; ii) to prevent massive growth of both
518 heterotrophic and nitrifying bacteria, that can compete with algae for the main nutrients
519 [43].



520 **Figure 6.** Simulation results at varying HRT (0.2-10 d) and SRT (0.2-10 d); influent TA (10 mol m⁻³) and δ_L (0.12 m, A, B and C; 0.06 m, D, E and F). A, D: algal biomass
 521 productivity [gAlg m⁻² d⁻¹]; B, E: Nitrogen recycling efficiency in the algal biomass [%]; C, F: N₂O risk factor (percentage of time along the day for which N₂O formation
 522 conditions occur, *i.e.* inorganic carbon < 0.2 molC m⁻³), [%]. The red line represents the scenario with no solid/liquid separation (*i.e.* HRT = SRT).
 523



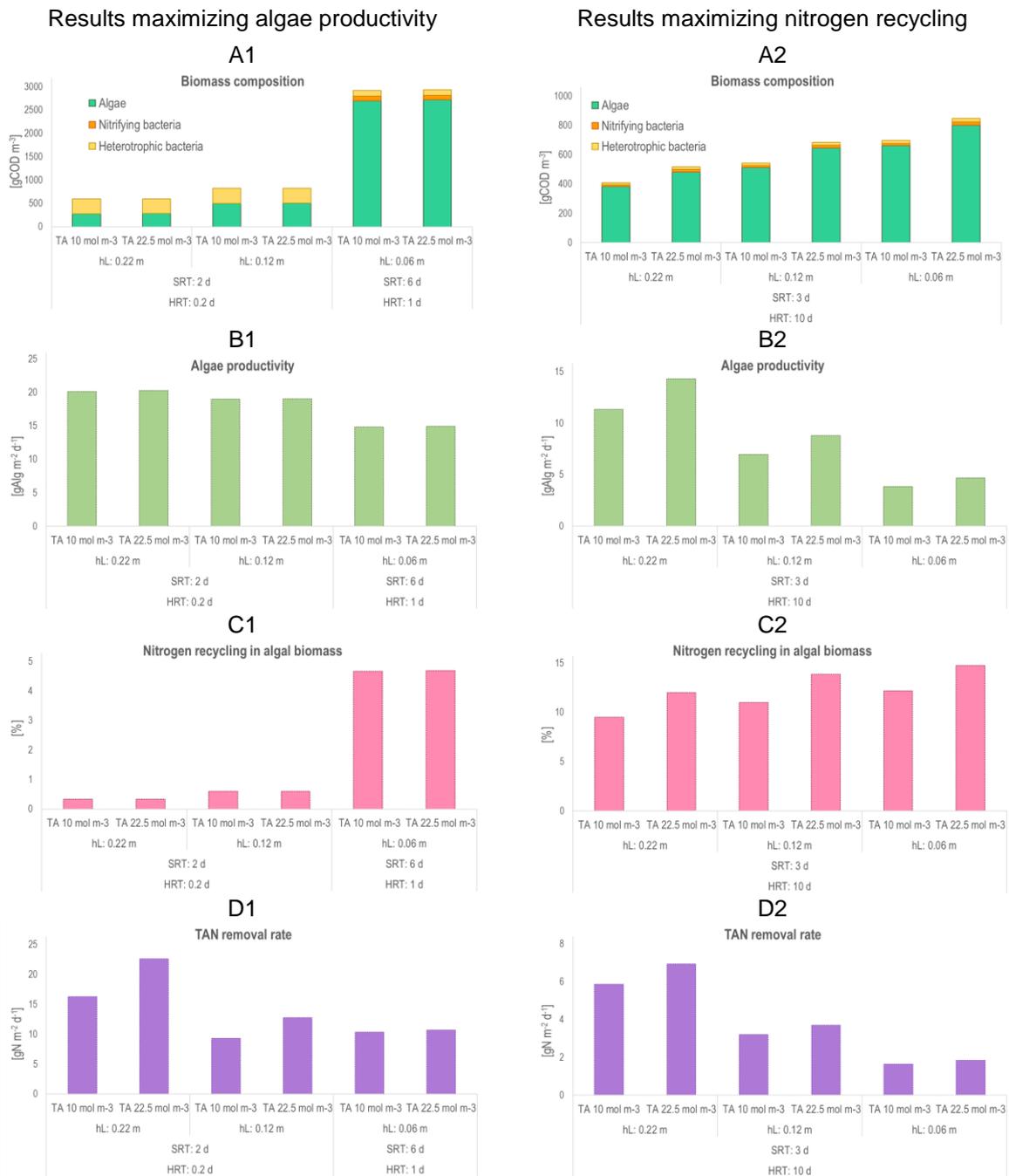
524 **Figure 7.** Simulation results at varying HRT (0.2-10 d) and SRT (0.2-10 d); influent TA (22.5 mol m⁻³) and δL (0.06
 525 m). A: algal biomass productivity [gAlg m⁻² d⁻¹]; B: Nitrogen recycling efficiency in the algal biomass [%]; C: N₂O
 526 risk factor (percentage of time along the day for which N₂O formation conditions occur, *i.e.* inorganic carbon < 0.2
 527 molC m⁻³), [%]; D: Algal biomass concentration in the reactor [gCOD m⁻³]. The red line represents the scenario
 528 with no solid/liquid separation (*i.e.* HRT = SRT).
 529
 530



531 **Figure 8.** Simulation results for influent TA=22.5 mol m⁻³ and δL=0.06 m, comparing the scenario with
 532 no solid/liquid separation (*i.e.* HRT=SRT, green continuous line) and the scenario where the HRT (red

533 dotted line) is adapted according to the SRT in order maximize: A) the algal productivity [$\text{gAlg m}^{-2} \text{d}^{-1}$]
 534 and B) the nitrogen recycling efficiency [%], (blue continuous line).

535



536 **Figure 9.** Simulation results for the conditions (HRT, SRT and δ_L) maximizing algae productivity (left column) and
 537 nitrogen recycling (right column). Results are given for both nominal (10 mol m^{-3}) and increased influent alkalinity
 538 (22.5 mol m^{-3}). A, B: biomass composition [gCOD m^{-3}]; C, D: algal biomass productivity [$\text{gAlg m}^{-2} \text{d}^{-1}$]; E, F: Nitrogen
 539 recycling in the algal biomass [%]; G, H: TAN removal rate [$\text{gN m}^{-2} \text{d}^{-1}$].

540

541 **3.6. Further step to target emerging contaminants**

542 Microalgae can play a role in the treatment of emerging contaminants by
543 bioadsorption, bioaccumulation and metabolic degradation [18]. There is currently
544 no efficient mathematical model validated in outdoor conditions describing such
545 processes. It is even likely that models should be molecule-specific to be accurate.
546 However, the ALBA model predicts the oxygen concentration in the medium, which
547 gives an index of the intensity of the oxidative stress influencing the molecular
548 degradation [17]. Simulation results showed that, under the tested conditions,
549 oxygen concentration could reach pics up to $22 \text{ mgO}_2\cdot\text{L}^{-1}$ during the day. A
550 modelling study can be further carried out, imposing as target objective to maximise
551 the peaks of oxygen concentration in the reactor, also by exploring a wider range
552 of HRT and SRT values. The model can therefore indirectly predict the benefit in
553 terms of emerging contaminant removal due to oxidative stress. Further work must
554 be carried out to quantify these aspects, and further integrate it with an estimation
555 of the gain in emerging contaminants treatment with a separation system. This is
556 an important point when computing the cost-benefit of the separation system.

557 As recently reviewed by You et al. [44], pollutants (including pesticides, metals,
558 engineered nanomaterials, pharmaceutical and personal care products, and
559 aromatic pollutants) affect both bacteria and algae by interfering with their
560 relationships. Cell-to-cell adhesion, substrate exchange and biodegradation of
561 organic pollutants, enhancement of signal transduction, and horizontal transfer of
562 tolerance genes are defence strategies in algal-bacterial systems, which should be
563 further tamed and applied in wastewater treatment systems. Enhancing the
564 treatment capacity by continuous addition of more efficient algae and/or bacteria

565 species is then the next step. This complementary strategy of bioaugmentation
566 could be combined to the separation system to further enhance the process
567 performance. Numeric simulations can guide the process optimization and
568 development for this new and promising strategy.

569

570 **3.7. Long-term membrane application for algae bacteria systems**

571 Membranes allow to decouple HRT and SRT and thus enhance the process
572 performance by more than 20%, with the possibility of recovering the algal biomass
573 for valuable applications [8].

574 However, membrane biofouling due to heavy suspended solids load, the small size
575 of microalgae and their poor sedimentation properties, combined with bacteria and
576 exopolymeric substances can lead to a progressive decline in the permeate flux
577 and an increase in the energy demand for operating the process, thus limiting long
578 term operations [41],[42]. Most of the membrane designs were developed for
579 wastewater treatment [43],[44], with very different objectives, such as removal of
580 pollutants and recovery of clean permeate vs recovery of valuable algal biomass
581 [42],[45]. The traditional fouling control techniques could be inefficient when
582 targeting a by-product recovery [45] and new directions have emerged in recent
583 years, while algal production was coupled to wastewater treatment [46]. Different
584 fouling control techniques have been developed recently, supported by a better
585 understanding of the interactions between the foulants and membrane surfaces.
586 The main strategies consist in changing the operating conditions, in pre-treating the
587 feed (using coagulation, adsorption, or oxidation), in mechanically cleaning of the
588 membranes, and in modifying the membrane surfaces by coatings or blending with
589 polymers and nanoparticles [45]. Choosing the adequate membrane cleaning

590 mechanism is of major importance in terms of energy consumption, permeate
591 production, and overall process performance [46],[47]. The development of
592 membranes for algae-bacteria systems with excellent bioactivity and associated to
593 antifouling strategies is of crucial importance for long term applications.

594

595 **Conclusion**

596 This study showed the complexity and challenges for optimizing outdoor HRABP
597 featured with solid/liquid separations systems, pointing out that the efficiency in
598 nitrogen recycling and the algal productivity cannot be maximized simultaneously.
599 Thus, it is fundamental to first choose the target of the process (e.g. wastewater
600 remediation, algal productivity, emerging contaminant treatment...), which will then
601 be reached by decoupling HRT and SRT: a gain of 23% on algal productivity (up to
602 $20.3 \text{ g.m}^{-2}.\text{day}^{-1}$), and of 35% in nitrogen recycling could be obtained for the specific
603 case study. When combined with depth optimization and alkalinity addition,
604 nitrogen recycling rate can reach 15%, with an algal productivity staying above 16
605 $\text{g.m}^{-2}.\text{day}^{-1}$ thus offering an interesting trade-off. Long-term application of efficient
606 membrane modules, and the characterisation of their efficiency on emerging
607 contaminant treatment, will then be crucial for future implementation of these
608 strategies.

609

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611

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615

616

617 **Author contribution statements**

618

619 FC developed the modified model with feedback from OB. FC and OB implemented

620 the simulation framework. FC wrote the manuscript with permanent feedback from OB.

621 EF, FB and OB guided FC in the definition of the case study framework. FB and OB

622 obtained grants to support the study and supervised the work. In general, all authors

623 provided critical feedback and helped shape the research, analysis and manuscript.

624

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