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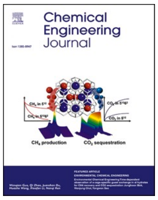

Optimizing resource recovery from wastewater with algae-bacteria membrane reactors

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ABSTRACT

Exploiting the combination of algae and bacteria in High Rate Algal/Bacterial Ponds (HRABP) is an emerging approach for wastewater remediation and resource recovery. In this study, the advantage of adding a solid/liquid separation system to uncouple Hydraulic Retention Time (HRT) and Solid Retention Time (SRT) is explored and quantified. A long-term validated model for HRABP was run to simulate and optimize a system at large scale treating digestate. It is shown that by uncoupling HRT and SRT, adapting the liquid depth and the alkalinity content, the algae productivity increases from 9.0 to 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 = 0.2 d and SRT = 2 d). Simulations pointed out that maximizing the algal productivity or the fraction of recovered nitrogen in the algal biomass are conflicting goals that are achieved under different operating conditions. Conditions maximising the algal productivity favour algae and heterotrophic bacteria while algae and nitrifying bacteria dominate the system under those conditions optimizing the efficiency of nitrogen recycling. Finally, increasing the influent alkalinity and adapting the water depth can boost the algal productivity without meeting conditions favourable to N_2O emission, opening new perspectives for resource recovery through algal biomass valorisation.

1. Introduction

Exploiting the combination of algae and bacteria in High Rate Algal/Bacterial Ponds (HRABP) is an emerging technology for wastewater treatment addressing the most critical aspects of wastewater treatment [1,2]. Compared to conventional biological processes, HRABP do not need an external oxygen supply, which is known to account for more than 50% of the energy required for wastewater treatment [3]. Recycling nitrogen and phosphorus into the algal biomass is another remarkable opportunity offered by this process [4], since algae can be used as a source of energy or feedstocks [5,6], *i.e.* with their lipid fraction for biofuels or proteins for bioplastics [7,8]. Moreover, HRABP have been highlighted for their ability to remove emerging contaminants. These molecules are persistent in the environment, impact on the reproductive systems of aquatic organisms, and bioaccumulate in the food chain [9]. The main emerging pollutants are pesticides, personal care products, pharmaceuticals and flame retardants [6,7]. Conventional wastewater treatment technologies are not designed to remove or degrade these compounds [10], while microalgae have been proven to be a powerful technology for bioremediation of emerging contaminants [5,12,13]. Indeed, in the highly oxidizing environment that is achieved thanks to high oxygen levels, microalgae can catalytically degrade complex compounds, with efficiencies ranging from 30 to 80% for drugs such as ibuprofen, carbamazepine, and caffeine [14–17]. Combined with bioadsorption and bioaccumulation, the algae process was found to achieve a remarkable removal efficiency for more than 50 emerging contaminants [18].

Adding a separation system, like membrane modules, is a technological breakthrough that leads to the uncoupling of Hydraulic Retention Time (HRT) and Solid Retention Time (SRT). On top of enhancing emerging contaminants removal [6], HRT and SRT separation was shown to considerably enhance the overall process efficiency [19,20] by increasing microalgal productivity by a factor 3.5 compared to a standard photobioreactor [21] (see the synthesis of various literature studies in Table 1). The key idea is that algal biomass productivity results from the product of net growth rate and biomass concentration which is enhanced when increasing the biomass in the reactor while keeping growth rate at high levels. However, a separation system makes the

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Table 1

Biomass productivity and nitrogen removarate 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	Spring		19.9 ± 0.5		2.3 ± 0.4		
							2.5		20	Autumn		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**	gTSS 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	1.5	4.5	N.R.	N.R	Valencia	15.7 ± 1.4	gVSS m^{-2} d^{-1}	2.2 ± 0.4	gN m^{-2} d^{-1}	[39]
						0.10			35	Winter		20.0 ± 2.4		2.4 ± 0.5		
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	16	N.R.	Valencia	12.2 ± 1.4	gVSS m^{-2} d^{-1}	1.7 ± 0.1	gN m^{-2} d^{-1}	[40]
								9	25	N.R.		7.7 ± 1.0		1.3 ± 0.5		
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	South-West France	1.9	gTSS m^{-2} d^{-1}	N.R.	N.R.	[42]
							8	8		Summer		3.6				
							17	17		Autumn		2.0				
							17	17		Winter		1.3				
UWW				raceway	8.3	0.05	2.6	2.6	46	Spring	Almeria	25.6	gTSS m^{-2} d^{-1}	96.9	%	[35]
						0.06	2	2	48	Summer		32.7		98.7		
						0.05	3.2	3.2	46	Autumn		18.9		98.8		
						0.05	3.7	3.7	41	Winter		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	Almeria	7.8 ± 4.3	gTSS m^{-2} d^{-1}	N.R.	gN m^{-2} d^{-1}	[36]
										Summer		8.5 ± 3.3				
										Autumn		7.8 ± 2.8				
										Winter		7.2 ± 3.8				
							5	5	N.R.	Spring		16.1 ± 3.7		3.5 ± 0.5		
										Summer		25.9 ± 5.1		4.3 ± 0.5		
										Autumn		20.0 ± 4.9		3.9 ± 0.2		
										Winter		14.2 ± 3.3		2.7 ± 0.4		
							3.3	3.3	N.R.	Spring		13.3 ± 3.9		N.R.		
										Summer		14.5 ± 3.3				
										Autumn		12.5 ± 5.1				
										Winter		11.2 ± 4.9				
SWW	378 ± 57.2	8.0 ± 2.1	13.0 ± 3.1	raceway	56	0.28	5	5	443	Spring	Narbonne	18.6 ± 1.7	gTSS m^{-2} d^{-1}	0.5 ± 0.03	gN m^{-2} d^{-1}	[25]
										Summer		16.0 ± 5.6		0.4 ± 0.10		
										Autumn		12.4 ± 3.1		0.4 ± 0.04		
										Winter		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]

(*continued on next page*)

Table 1 (*continued*)

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	
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	Milan	**10.81 ± 2.16**	gTSS m^{-2} d^{-1}	**5.3 ± 0.6**	gN m^{-2} d^{-1}	[26]
							10	10		Summer		**12.2 ± 3.5**		**6.9 ± 1.3**		
							20	20		Autumn		**6.1 ± 3.1**		**4.9 ± 1.5**		
Simulated Centrate from PWW	514 ± 190	310 ± 91.0	13.6 ± 4.0	Simulated raceway	2000	0.22	0.2	2	50	Simulated Spring	Simulated Milan	43.1 ± 2.9	gVSS m^{-2} d^{-1}	22.6 ± 12.2	gN m^{-2} d^{-1}	This study
							10	3				19.6 ± 2.1		6.9 ± 0.5		
						0.12	0.2	2				32.0 ± 3.0		12.8 ± 6.8		
							10	3				15.4 ± 2.1		3.7 ± 0.2		
						0.06	1	6				16.2 ± 4.6		10.7 ± 3.3		
							10	3				9.8 ± 1.5		1.9 ± 0.1		

Abbreviations. UWW (urban wastewater); AnMBR (anaerobic membrane bioreactor); WWTP (wastewater treatment plant); PWW (piggery wastewater); SWW (synthetic wastewater).
N.R.: Not Reported.
Notes.
*Total nitrogen and total phosphorous values.
**Total phosphorous value.
Data reported in bold identify the cases without solid-liquid separation in studies comparing raceway performances with and without membranes.

process more flexible but also more complicated to operate and to optimize by increasing the number of parameters that can be tuned. Concentrating the algae in the system leads to a higher light attenuation and therefore, to a lower growth rate. In such a complex situation, a multi-parametric optimization must be carried out and the guidance of mathematical models has already proven very useful [22,23]. This is especially decisive in the case of complex dynamical bioprocesses involving a large range of interacting species, like for anaerobic digestion [24].

Recently, the ALBA model (ALBA standing for ALgae-BActeria) was developed and validated. This mathematical model was built to simulate algal-bacterial interactions and competitions in HRABP, to evaluate bioremediation performances and to provide a better understanding of the interactions among the chemical, physical and biological processes. This model was validated and was proven able to accurately predict, under real outdoor conditions, the complex behaviour of two HRABP pilot-plants operated in two different locations and under different feeding conditions [25,26]. The model turned out to be accurate in predicting the daily and seasonal responses, even during winter which was never effectively described by previously available models. In total, more than 630 days of pilot-scale experimental data validated the model performance [25,26] under two different climatic conditions and with the same parameter set. The model confirmed, as experimentally observed [27], that inorganic carbon could become limiting, as a result of the strong competition for inorganic carbon by algae and nitrifiers. It was unexpected that this limitation still happened even under continuous CO_2 injection; the ALBA model revealed that this was due to a shortage in alkalinity owing to the limited alkalinity/nitrogen ratio in the feed, insufficient to support full nitrification. At low alkalinity, inorganic carbon can no more be stored in the medium and becomes limiting both for algae and nitrifiers, leading to conditions favourable to N_2O emission. The high-fidelity ALBA model is therefore a powerful validated tool to assess the process productivity, the nitrogen removal rate, but also the risk of highly impacting N_2O emission.

In this work, the ALBA model was upgraded by implementing a solid/liquid separation system to investigate the complex interplay between SRT and HRT. The ALBA model was then validated in this new configuration using the experimental data of Robles et al. [19], carried out in an outdoor pilot reactor. The model was finally used to explore a large range of scenarios, varying HRT, SRT, and liquid depth. The trade-off between algal productivity and nitrogen recycling was explored, while focusing on operating modes that are not susceptible of leading to N_2O emissions.

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

2. Material and methods

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

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

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

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

$$\begin{aligned} TA = \; & HCO_3^- + 2CO_3^{2-} + H_2PO_4^- + HPO_4^{2-} + 2PO_4^{3-} + OH^- + NH_3 \\ & - H^+ - HNO_2 - HNO_3 - H_3PO_4 \end{aligned} \tag{2.1.2}$$

The CO_2, NH_3, O_2 gas/liquid transfer is also included, quantifying the rate through the global k_La and the diffusivity coefficients.

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

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

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

2.2. Case study

A theoretical case study was assumed for the optimization exercise, consisting in an industrial HRABP of 2000 m^2 treating digestate, located in the North of Italy (Milan area). Simulations were run focusing on the spring season, where the most affecting environmental conditions (*i.e.*, light and temperature) are closer to the optima for algal growth. The influent was assumed to be a diluted liquid fraction of digestate with the characteristics reported in Table 2 (taken from Pizzera et al. [31]).

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).

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	$mgCaCO_3$ L^{-1}

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

2.3. Implementation of HRT and SRT decoupling

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

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

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

$$\frac{dX_j}{dt} = \frac{Q_{IN}}{V} X_{j,\,IN} - \alpha X_j \left(\frac{Q_{IN} - Q_{EVAP}}{V} \right) \pm \sum_i \nu_{i,j} \, \rho_i \tag{2.3.2}$$

where Q_{IN} is the imposed inflow rate [m^3 d^{-1}]; Q_{EVAP} is the outflow leaving the system by evaporation, [m^3 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}]; $\nu_{i,j}$ is the stoichiometric coefficient associated to the state variable X_j and the process i and ρ_i is the rate of process i [gX_j m^{-3} d^{-1}].

2.4. Optimization study

For the optimization study, 19200 simulations were run, testing the simultaneous influence of different factors on the HRABP performances: i) the HRT (0.2–10 d); ii) the SRT (0.2–10 d); iii) the liquid depth (δ_L 0.06 –1 m); and finally, the supplementation of Total Alkalinity (TA, 0–12.5 mol m^{-3} added) through dosage of $CaCO_3$.

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

Algal productivity [gAlg $m^{-2}d^{-1}$] was defined as:

$$ALG_{productivity} = \frac{X_{ALG} \;\; Q_{out} \;\; \alpha}{S} \; \varphi \tag{2.4.1}$$

where X_{ALG} is the algae concentration in the pond [gCOD m^{-3}]; Q_{OUT} is the outflow, defined as the algebraic sum Q_{IN}-Q_{EVAP} [m^3 d^{-1}]; S is the reactor surface [m^2]; $\varphi = 0.64$ is the conversion factor from COD_{ALG} to dry weight (DW) of algae (gDW_{ALG} $gCOD^{-1}$).

The efficiency of nitrogen recycling [%] was defined as:

$$Eff_{N,RECYC} = \frac{Flux_{Nout,ALG}}{Flux_{Nin}} 100 \tag{2.4.2}$$

where: $Flux_{N_{out},ALG}$ is the mass flow of Nitrogen [gN d^{-1}] in the harvested algal biomass and $Flux_{Nin}$ is the mass flow of Nitrogen [gN d^{-1}] fed to the HRABP:

$$Flux_{Nout,ALG} = X_{ALG} \; iN_{BM,ALG} \; Q_{OUT} \; \alpha \tag{2.4.3}$$

$$\begin{aligned} Flux_{Nin} = \; & Q_{IN} \big(S_{NH,IN} + S_{NO2,IN} + S_{NO3,IN} + S_{S,IN} \; iN_{SS} + S_{I,IN} \; iN_{SI} \\ & + X_{S,IN} \; iN_{XS} + X_{I,IN} \; iN_{XI} \big) \end{aligned} \tag{2.4.4}$$

where $iN_{BM,ALG}$ is the N content in the algal biomass [gN $gCOD^{-1}$], Q_{IN} is the inflow rate [m^3d^{-1}]; $S_{NH,IN}$, $S_{NO2,IN}$ and $S_{NO3,IN}$ are the influent concentrations of ammoniacal nitrogen, nitrite and nitrate, respectively [gN m^{-3}]; $S_{S,IN}$, $S_{I,IN}$, $X_{S,IN}$ and $X_{I,IN}$ are influent concentration of the state variables representing the organic matter concentration [gCOD m^{-3}] as soluble degradable, soluble inert, particulate degradable, and particulate inert (I) components, respectively; iN_{SS}, iN_{SI}, iN_{XS} and iN_{XI}, [gN

$gCOD^{-1}$] quantify their nitrogen content.

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

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

3. Results and discussion

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

The model prediction capability for the most relevant monitored variables (pH, S_{O2}, S_{NH}, S_{NO3}, S_{NO2} and X_{ALG}) can be found in Casagli et al. [25,26] (see also a summary in Supporting Information SI.1 and SI.2) and it covers an uncommonly broad range of conditions, including all the seasons, and HRT (=SRT) ranging from 2.6 to 20 days (see S.I.7).

The total ammoniacal nitrogen (TAN) removal rate (gN m^{-2} d^{-1}) and the Total Suspended Solids (TSS) productivity (gTSS m^{-2} d^{-1}) were also computed from the experimental measurements and compared to the model predictions (Figs. 2 and 3 for summer, autumn and winter, while spring results are shown in Supporting information, SI.4). It is worth mentioning that algal productivity is rarely experimentally measured in algae-bacteria consortia, since only TSS or VSS (Volatile Suspended Solids) are monitored. Both turned out to be accurately predicted, as shown in Figs. 1 and 2, with p-values for TSS productivity and TAN removal rate predictions below 1E-6. The highest TSS productivity was

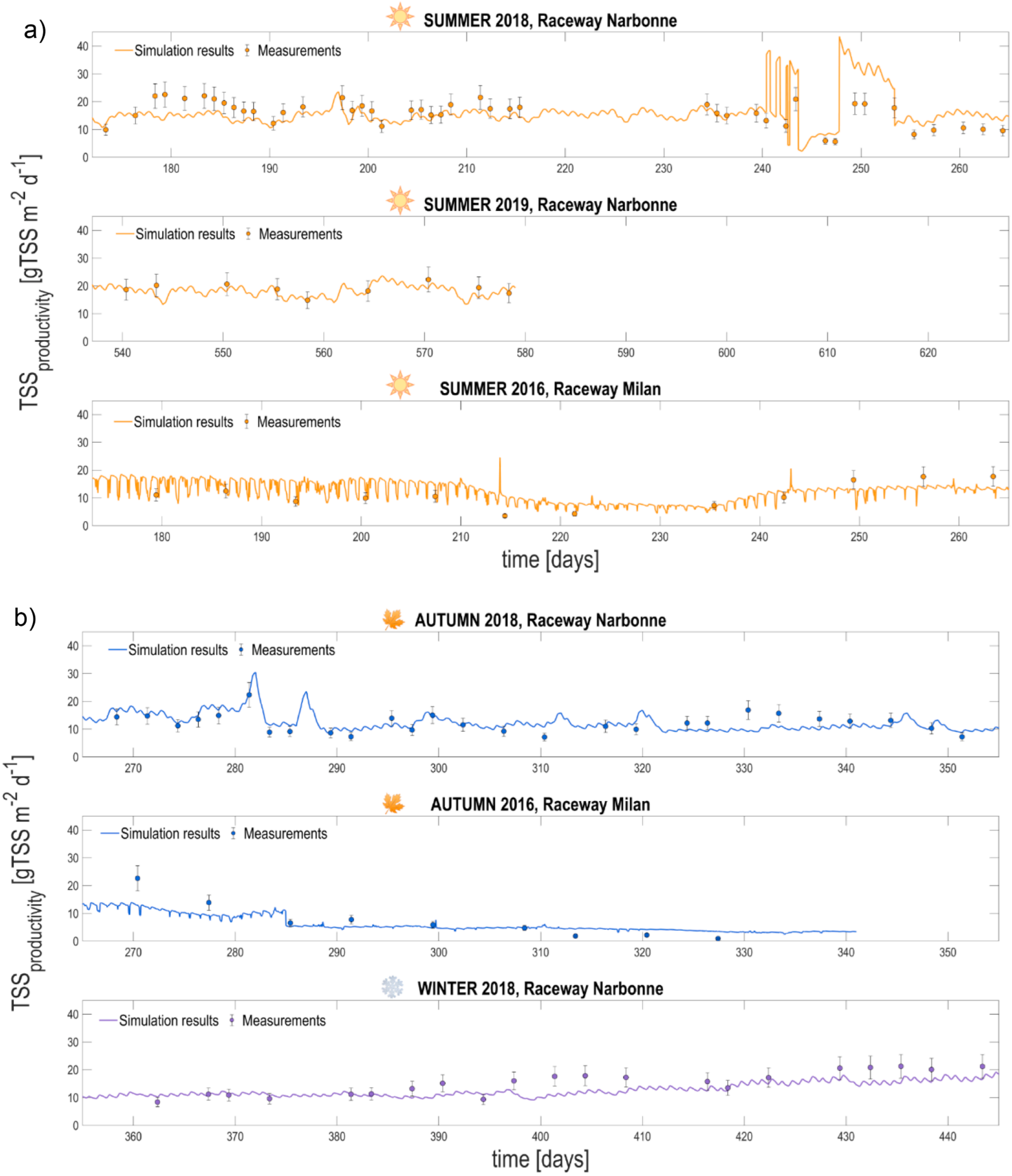

Fig. 1. Total Suspended Solids productivity experimentally measured and computed with the ALBA model for Milan and Narbonne case studies according to the season (a: summer; b: autumn and winter).

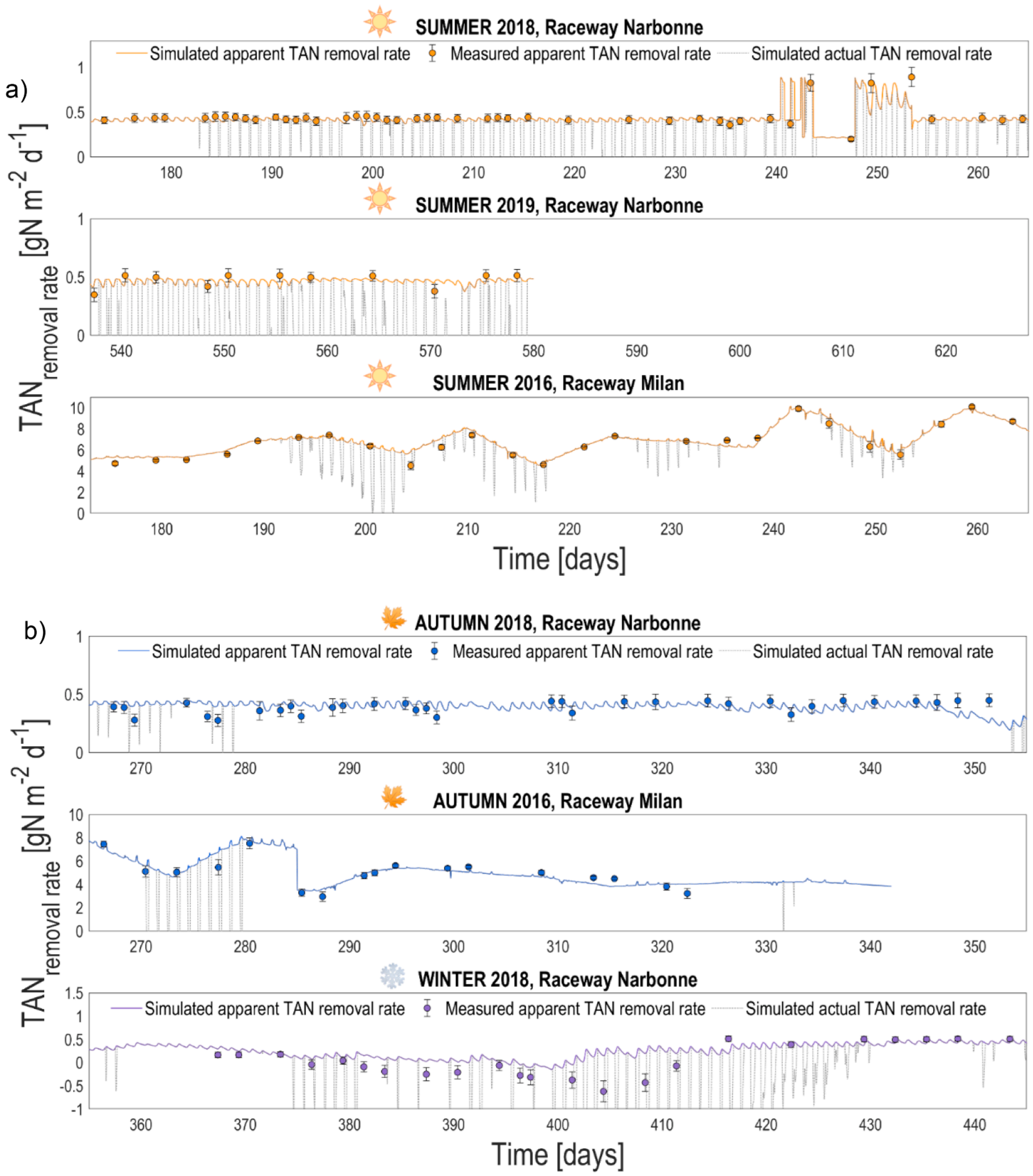

Fig. 2. Total ammoniacal nitrogen removal rate, in gN m^{-2} d^{-1}, for both case studies (Milan and Narbonne) and according to seasons (a: summer; b: autumn and winter). The term apparent refers to the fact that the flux of N-NH_3 stripped out was considered as untreated nitrogen.

reached in summer, for both Narbonne and Milan, with an average value of 16.0 ± 5.6 gTSS m^{-2} d^{-1} (Summer 2018, Narbonne), 18.2 ± 2.0 gTSS m^{-2} d^{-1} (Summer 2019, Narbonne) and 12.2 ± 3.5 gTSS m^{-2} d^{-1} (Summer 2016, Milan).

During autumn and winter, the TSS productivity decreased in both experimentations, with an average value of 12.4 ± 3.1 and 12.7 ± 2.4 gTSS m^{-2} d^{-1} in autumn and winter, respectively, in Narbonne and of 6.1 ± 3.1 gTSS m^{-2} d^{-1} in autumn in Milan.

The generally higher values obtained in Narbonne along all the seasons are mainly due to the lower HRT, *i.e.* 5 days, (not considering the period where it was varied between 2.6 and 10, for days 240–253), compared to Milan (HRT = 10–20 days), corresponding to a lower biomass harvesting rate. The TSS productivities shown in Fig. 1a, b are in agreement with the experimental values reported in other literature works, where the raceways were operated under similar conditions. For instance, 10.7 ± 6.5 gTSS m^{-2} d^{-1} [32]; 15.2 ± 2.1 gTSS m^{-2} d^{-1} [33]; 5.5 ± 7.4 gTSS m^{-2} day^{-1} [34]; 12.3 gTSS m^{-2} d^{-1} as maximum value during winter and 32.4 gTSS m^{-2} d^{-1} as maximum value during summer [35].

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

Algae productivity in Milan represented 72% of the TSS productivity on a yearly basis (*i.e.* 7.0 gALG m^{-2} day^{-1} compared to 9.8 gTSS m^{-2} day^{-1}), corresponding to a microbial biomass ($X_{BIO} = X_{ALG} + X_{AOB} + X_{NOB} + X_H$) composed by 90.4% of algae on yearly average. For the Narbonne case study, algae productivity was 63% of the TSS productivity on a yearly bases (*i.e.* 9.5 gALG m^{-2} day^{-1} compared to 15.1 gTSS m^{-2} day^{-1}), corresponding to a microbial biomass where algae represented 76.6% of the community on yearly average.

Looking at TAN removal rate in Narbonne (Fig. 3), it was on average 0.4 ± 0.1 gN m^{-2} d^{-1} in summer, with HRT of 5 days. Peaks of 0.9 gN m^{-2} d^{-1} were reached at HRT of 10 days, while the lowest value was achieved for a HRT of 2.6 days (0.2 gN m^{-2} d^{-1}). In autumn and winter, the TAN removal rate was generally lower than 0.5 gN m^{-2} d^{-1}, due to

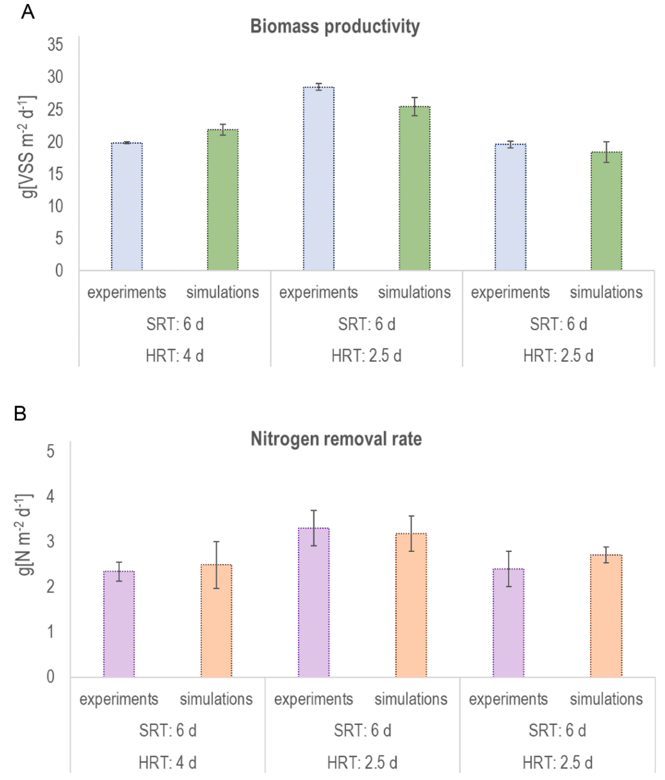

Fig. 3. Comparison between experimental [19] and simulation results (this study) for HRT uncoupled from SRT. A) Biomass productivity, in gVSS m^{-2} d^{-1}; B) Nitrogen removal rate, in gN m^{-2} d^{-1}.

the sub-optimal growth conditions for algae and bacteria. For the raceway located in Milan, the TAN removal rate was higher during summer, being in a range of 4.6–10.2 gN m^{-2} d^{-1} (average 6.9 ± 1.3) for a HRT of 10 days. In autumn, these values decreased, varying between 3.5 and 8.1 gN m^{-2} d^{-1} (average 4.9 ± 1.5), even when the HRT increased up to 20 days. The difference between the TAN removal rate in the two raceways is mainly due to differences in the influent nitrogen, both as concentration (8 gN m^{-3} in Narbonne and 310 gN m^{-3} on average in Milan) and as chemical nature (urea in Narbonne, and TAN in Milan). The TAN removal rates values are in agreement with the ranges reported in the work of Morillas-España et al. [36] (2.7 gN m^{-2} d^{-1} during winter and 4.3 gN m^{-2} d^{-1} during summer, for an inlet N-NH_4^+ concentration 168–210 g m^{-3}), for a raceway operated under similar HRT conditions (3–10 d).

In addition, the model allows assessing the fluxes of N-NH_3 stripped out and, therefore, computing the actual TAN removal rate (grey dotted line in Fig. 2, a and b) by subtracting the contribution of the stripped N to the apparent TAN removal rate. In Milano, the imperfect pH regulation allowed going from pH 6.5 to 7.8 increasing the free NH_3 level by a factor of 30. Since TAN concentration was high in piggery digestate, this corresponded to large spikes of released NH_3. In Narbonne, the TAN concentration was lower, but no pH regulation was implemented so that pH could even reach values above 10, resulting in temporarily high NH_3 fluxes as well.

3.2. Model accuracy when uncoupling HRT and SRT

The experimental campaign of Robles et al. [19], was used for validating the ALBA model when HRT is uncoupled from SRT. The experimental campaign was performed on the same outdoor pilot-scale raceway located in Narbonne, fed with synthetic municipal wastewater [37], previously used for the calibration and validation of the ALBA model [25]. The HRABP was operated with a liquid depth of 0.3 m and connected to a high-flow industrial-scale membrane compartment. The authors evaluated the process performances computing the average biomass productivity and nitrogen removal rate over a 20 days experimental period, for each operational condition tested. Uncoupling SRT and HRT by membrane filtration improved process efficiency, with higher biomass throughput and nutrient removal rate at lower HRT operation. At an SRT of 6 days, the biomass productivity increased up to 19.9 ± 0.5 and to 28.5 ± 0.5 gVSS m^{-2} d^{-1} when HRT was set to 4 and 2.5 days, respectively. The corresponding nitrogen removal rates were 2.3 ± 0.4 and 3.3 ± 0.4 gN m^{-2} d^{-1}, respectively. The authors ran also an experiment with HRT set to 2.5 and SRT set to 6 days, but under sub-optimal light and temperature conditions, obtaining lower values for both biomass productivity and nitrogen removal rate (19.6 gVSS m^{-2} d^{-1} and 2.4 g N m^{-3} d^{-1}, respectively), compared to the experiment carried out with the same operational conditions and under optimal light and temperature ranges.

The ALBA model was run under the same operational conditions, considering the climatology of the year 2018–2019 [25]. Biomass productivity and nitrogen removal rate were computed according to Robles et al. [19] and simulation results were averaged on a seasonal time scale. Spring 2018 presented similar average light and temperature compared to the one reported by Robles et al. [19] as being optimal. The autumn climatology of 2018 turned out to be comparable to the light and temperature conditions reported as suboptimal by Robles et al. [19]. The model predictions, for both biomass productivity and nitrogen removal rate, are perfectly in agreement with the experimental results (Fig. 3), with a R^2 of 0.78 and 0.93, respectively. Discrepancies between simulated and experimental data are mainly due to the dynamic temperature and light dataset used for simulations, that are not belonging to the same year as in Robles et al. [19], since these data were not available. Moreover, while in the work of Robles et al. [19], the measurements were averaged on 20 days of experiments for each tested condition, the simulated data were averaged on the entire season (3 months for spring and 3 months for autumn).

These recorded (and simulated) biomass productivity and nitrogen removal rates are in line with other experimental results obtained in different studies testing membrane separation systems uncoupling HRT and SRT in outdoor algae-bacteria systems for wastewater treatment (see Table 1 for a synthetic comparison of literature studies). Morillas-España et al. [33] operated a 4.4 m^3 HRABP (surface 30 m^2, liquid depth 0.12 m) fed with primary domestic wastewater, at a constant SRT of 4.8 d and a variable HRT 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 gDW m^{-2} d^{-1}, respectively. González-Camejo et al. [38] operated an outdoor membrane photobioreactor (MPBR) of 5 m^2, treating tertiary sewage effluent, at HRT 1.5 d, SRT 4.5 d and liquid depth 0.25 m. They recorded an average biomass productivity of 15.7 ± 1.4 gVSS m^{-2} d^{-1}. When operating the same system reducing the light path to 0.10 m and with HRT 4.5 d and SRT 3 d, the measured biomass productivity was 20 ± 2.4 gVSS m^{-2} d^{-1} [39].

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

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

Simulations were run to explore optimal SRT and HRT for treating digestate. The (constant) liquid depth was set to 0.225 m, typical of raceways systems. The TA in the influent was set at its nominal value (10 mol m^{-3}), typical of a diluted digestate. Simulation outcomes are shown in Fig. 4, while the 2D version of the graphs are reported in SI.5. As it was shown in Casagli et al. [26], despite pH regulation, a strong competition for inorganic carbon between algae and nitrifiers can take place since alkalinity is strongly reduced by nitrification. The consequence is a risk of N_2O emission that can be very detrimental to the environment. In fact, it turns out that the N_2O risk factor becomes

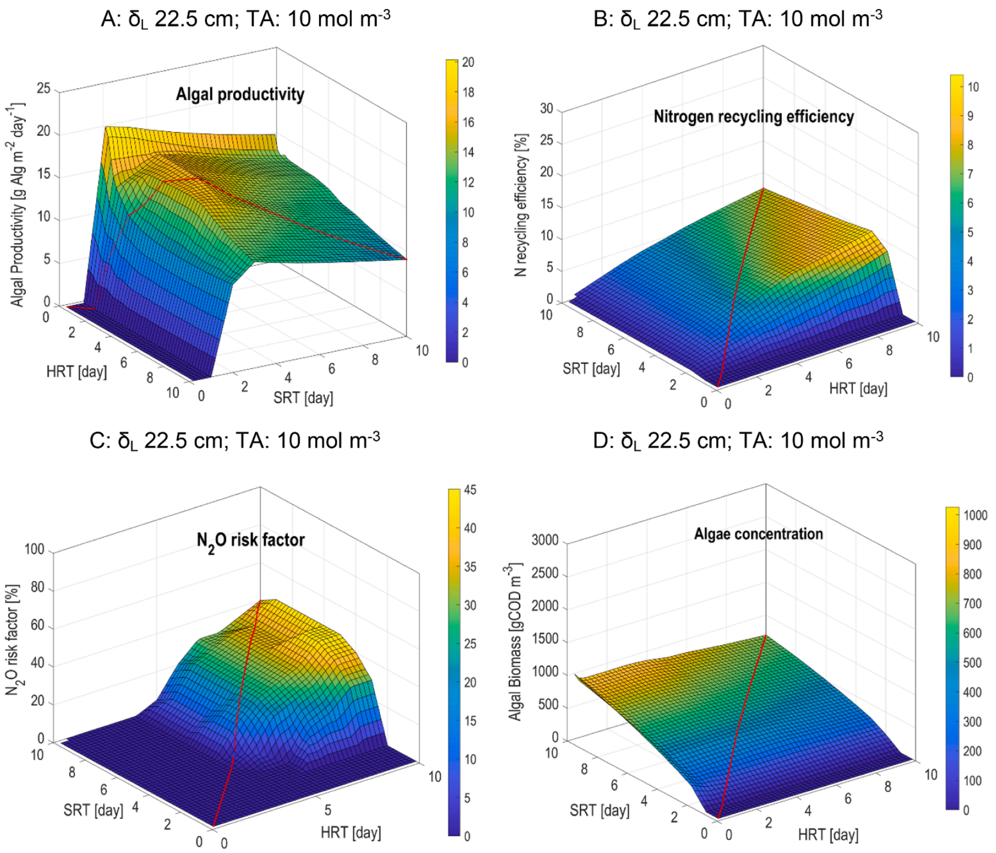

Fig. 4. Simulation results at varying HRT (0.2–10 d) and SRT (0.2–10 d). Influent TA is 10 mol m^{-3} and depth, δ_L, is 0.225 m. A: algal biomass productivity [gAlg m^{-2} d^{-1}]; B: Nitrogen recycling efficiency in the algal biomass [%]; C: N_2O risk factor (percentage of time along the day for which N_2O formation conditions occur, *i.e.* inorganic carbon $<$ 0.2 molC m^{-3}), [%]; D: Algal biomass concentration in the reactor [gCOD m^{-3}]. The red line represents the scenario with no solid/liquid separation (*i.e.* HRT = SRT).

nonzero for couples of HRT ≥ 4 d and SRT ≥ 2 d, reaching the highest values (35–45%) for HRT ≥ 8 d and SRT ≥ 4 d (Fig. 4C).

The maximum of algal productivity (20.2 gAlg m^{-2}d^{-1}) was obtained for HRT = 0.2 d and SRT = 2 d (Fig. 4A). This pair of hydraulic and solid retention times is not risky as for N_2O emission (risk factor is 0%). However, looking at the algae concentration and the efficiency in nitrogen recycling, both values are low (280 gCOD m^{-3} and 0.34% respectively, see Fig. 4B and D). Indeed, for this low HRT, the nitrogen loading rate is high, leading to low efficiency of nitrogen recycling ($Eff_{N,Recyc}$). Very low SRT, typically lower than 1 d, whatever the HRT, lead to algal washout, with algal biomass concentration that goes to zero and so does the algal biomass productivity (Fig. 4A and D). The maximum value for the efficiency in nitrogen recycling (10.4%) is obtained for a very different operational regime, *i.e.* HRT = 10 d and SRT = 3 d (Fig. 4B). However, these conditions are associated to an unsustainable N_2O risk factor of 30% (Fig. 4C). The higher algae and nitrifying bacteria concentrations, indeed lead to the exhaustion of the inorganic carbon pool. In addition, the algal productivity related to the highest efficiency in nitrogen recycling is reduced to 12.4 gAlg m^{-2} d^{-1}. This opposite trend reflects a classical contrasting tendency between productivity and efficiency in nutrient use, like for the nitrogen removal rate and removal efficiency in wastewater remediation. This explains the conflicting trend for the Key Process Indicators (2.4.1 and 2.4.2).

Finally, the best operational conditions in terms of HRT and SRT for reaching a better trade-off between algae productivity and nitrogen recycling, while avoiding the risk of N_2O emission, appear to be when HRT ranges from 2 to 5 d and SRT from 2 to 3 d. However, under these conditions, the efficiency of nitrogen recycling is never higher than 6.6% (see Fig. 4A and B).

In order to provide a better picture of the gain achievable by uncoupling HRT and SRT, Fig. 5 reports a comparison between the algal productivity and the efficiency in nitrogen recycling when no solid/

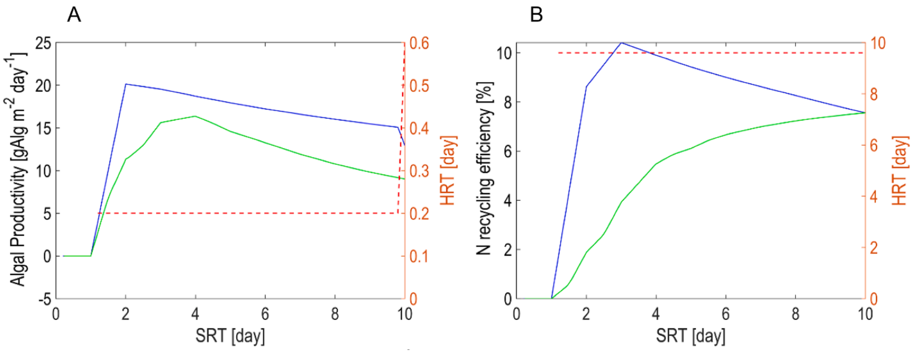

Fig. 5. Simulation results for TA = 10 mol m^{-3} and δ_L = 0.225 m, comparing the scenario with no solid/liquid separation (*i.e.* HRT = SRT, green continuous line) and the scenario where HRT (red dotted line) is adapted according to the SRT in order to maximize: A) algal productivity [gAlg m^{-2} d^{-1}]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

liquid separation is applied (*i.e.* HRT = SRT). In this case, HRT is adapted according to SRT, in order to maximize either algal productivity or nitrogen recycling efficiency. Fig. 5A clearly shows that higher algae productivity are achieved for all the SRT values between 2 and 10 days, when operating the system with an HRT of 0.2 days. A doubling in algal productivity can be obtained by appropriately modulating HRT compared to the reference case of SRT = HRT. Fig. 5B shows that the same is valid for the nitrogen recycling efficiency, that reaches higher values when these two operational parameters are decoupled (imposing HRT 10 days at varying SRT between 2 and 10 days). Nitrogen recycling efficiency could be tripled compared to the standard case when HRT = SRT.

Additional simulations were also run, by increasing the influent TA from 10 up to 15, 17.5, 20 and 22.5 mol m^{-3} (see Table SI.6.1 in SI.6). For an influent alkalinity increased up to 15 and 17.5 mol m^{-3}, inorganic carbon limiting conditions can still be reached when applying HRT $\geq$ 6 d and SRT $\geq$ 4 d, with a maximum value of N_2O risk factor of 45% for HRT = 10 d and SRT = 5, 6 and 7 d. On the contrary, with 20 and 22.5 mol m^{-3} of influent TA, the N_2O risk factor is always zero, indicating that this level of alkalinity (0.90 and 1.02 mol $mol(NH4^{+})^{-1}$ respectively, corresponding to 2.51 and 2.82 $gCaCO_3$ $g(NH4^{+})^{-1}$) is adequate to fully support both algae and nitrifiers carbon request.

TA supplementation appears to be particularly beneficial for algae-bacteria systems when the aim is to treat wastewater, since it allows to set higher HRT and SRT, favouring: i) slow growers, such as AOB and NOB; ii) the inorganic carbon availability to guarantee the full nitrification process, simultaneously avoiding the competition with algae; iii) reduced NH_3 stripping conditions due to the improved TAN removal (see Fig. 7D1 and 7D2) and, thus, lower TAN concentration in the reactor. The environmental benefit of this strategy is therefore evident.

It must be highlighted that alkalinity addition should be adjusted carefully to avoid a marked increase in CO_2 emissions due to overloading the system with inorganic carbon, especially when the pH in the raceway is controlled with CO_2 bubbling. Ideally, it should be regulated according to the CO_2 saturation level in the pond, that is strictly related to water temperature. In fact, the driving-force in the gas–liquid exchange is given by the difference between the gas saturation level in the liquid phase and the actual concentration of the dissolved gas. With TA addition, the buffering capacity of the system increases, and so does the level of soluble inorganic carbon. The risk is that when an external source of CO_2 is injected in the system for pH regulation, most of it is lost to the atmosphere.

When considering simultaneously the algal biomass productivity and the nitrogen recycling optimization, the situation does not change significantly. Regulating TA avoids N_2O emissions, but it does not allow to simultaneously reach these two conflicting objectives.

3.4. Unravelling the influence of liquid depth

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

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

The maximum nitrogen recovered as algae biomass (11.8%) is obtained for HRT = 10 d and SRT = 3 d, but for this pair of values the N_2O risk factor is 35% (Fig. 6B and C). So far, the best working range of both HRT and SRT, for guarantying elevated algal biomass productivity and efficiency in nitrogen recycling is 2–5 d, similarly to the case δ_L = 0.225 m. In comparison, the nitrogen uptake from the algal biomass is higher and the best trade-off between algal biomass productivity (15.3 gAlg m^{-2} d^{-1}) and nitrogen recycling (7.0%), avoiding N_2O favourable

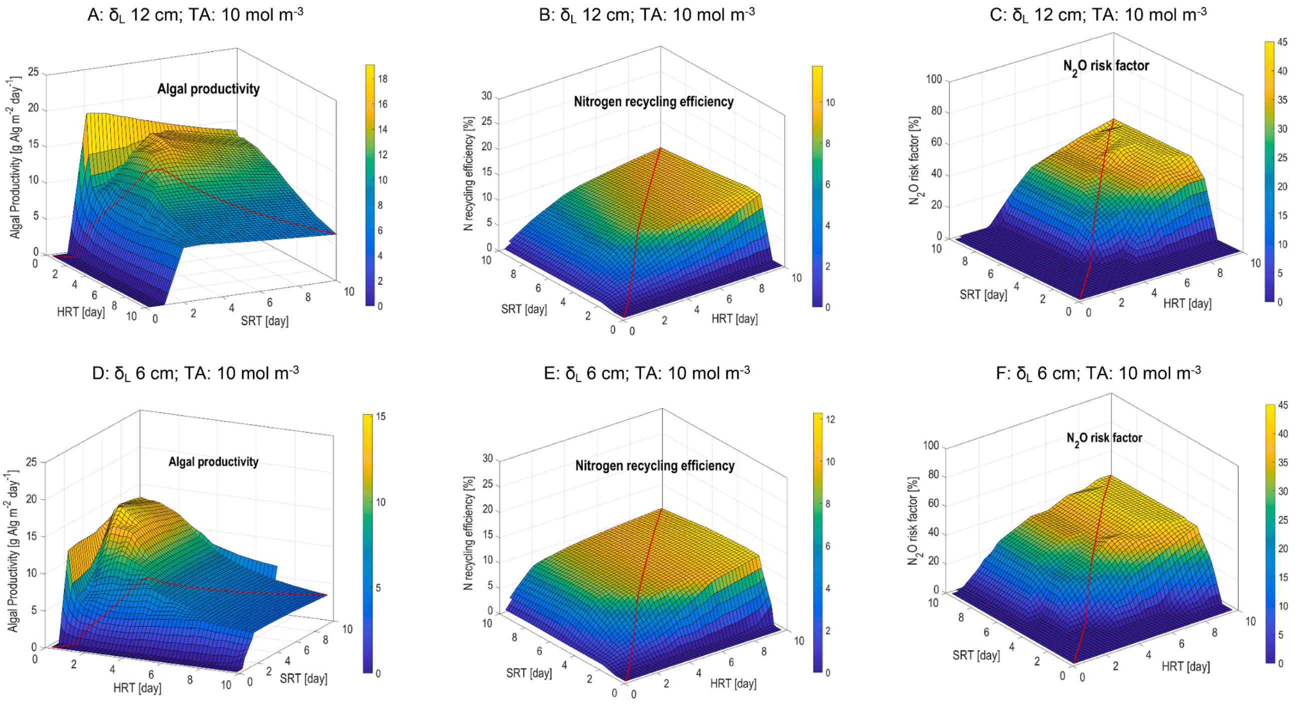

Fig. 6. Simulation results at varying HRT (0.2–10 d) and SRT (0.2–10 d); influent TA (10 mol m^{-3}) and δ_L (0.12 m, A, B and C; 0.06 m, D, E and F). A, D: algal biomass productivity [gAlg m^{-2} d^{-1}]; B, E: Nitrogen recycling efficiency in the algal biomass [%]; C, F: N_2O risk factor (percentage of time along the day for which N_2O formation 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).

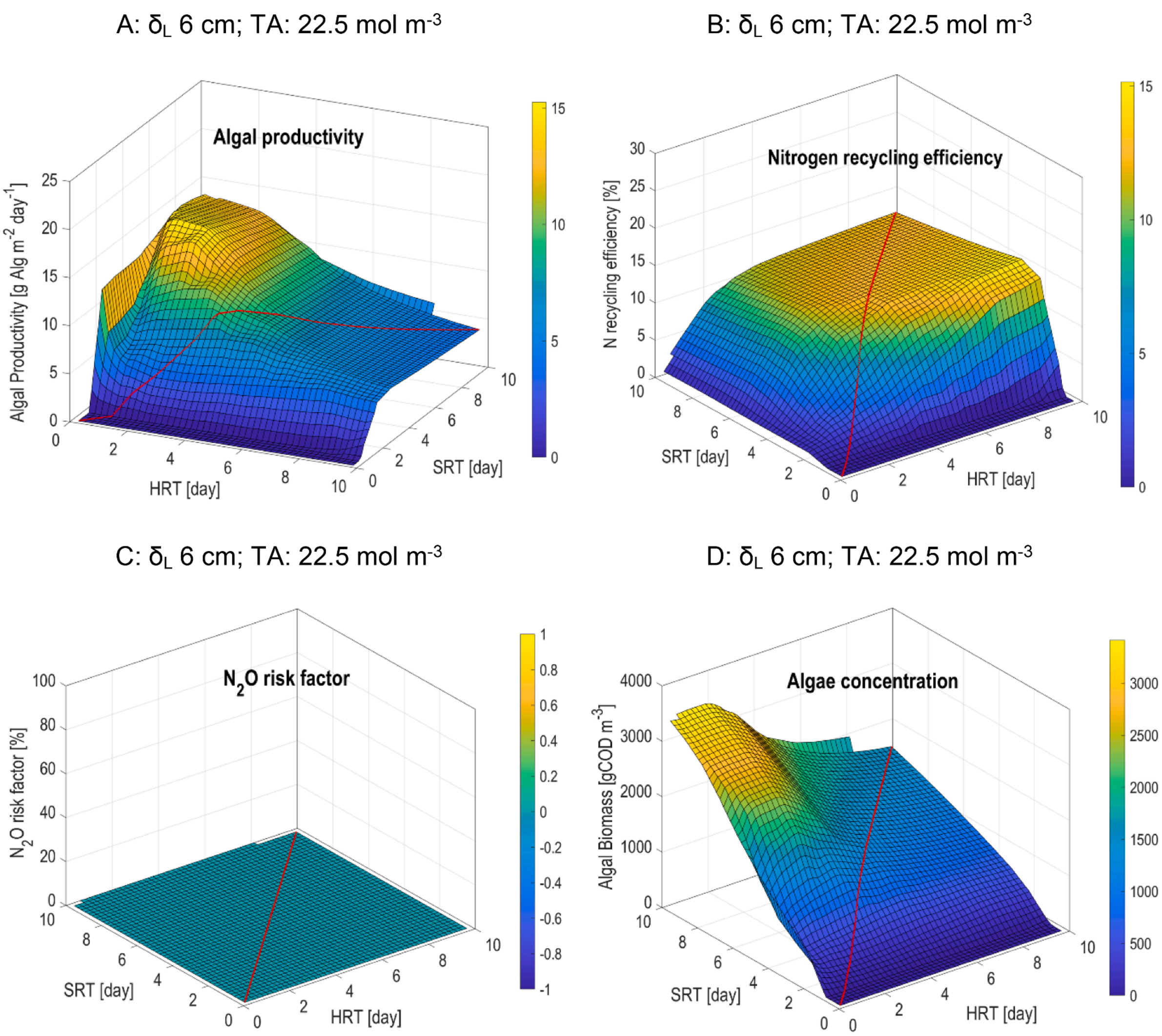

Fig. 7. Simulation results at varying HRT (0.2–10 d) and SRT (0.2–10 d); influent TA (22.5 mol m^{-3}) and δ_L (0.06 m). A: algal biomass productivity [gAlg m^{-2} d^{-1}]; B: Nitrogen recycling efficiency in the algal biomass [%]; C: N_2O risk factor (percentage of time along the day for which N_2O formation conditions occur, *i.e.* inorganic carbon < 0.2 molC m^{-3}), [%]; D: Algal biomass concentration in the reactor [gCOD m^{-3}]. The red line represents the scenario with no solid/liquid separation (*i.e.* HRT = SRT).

conditions, is obtained for HRT = 3 d and SRT = 5 d.

Decreasing δ_L to 0.06 m (assumed to be the lowest operational depth for this kind of systems), the maximum of algal biomass productivity (15.2 gAlg m^{-2} d^{-1}) is obtained for HRT = 1 d and SRT = 6 d (Fig. 6D), with a corresponding nitrogen recycling of 4.7% (Fig. 6E) and algae concentration in the reactor of 2603 gCOD m^{-3} (see Fig. 9A1). Compared to the the twofold depth (δ_L = 0.12 m), the maximum productivity is lower, even if the algal concentration is almost 5 times higher. This is due to a much lower nutrient influent load for 6 cm of liquid depth. Correspondingly, the nitrogen uptake in the algal biomass

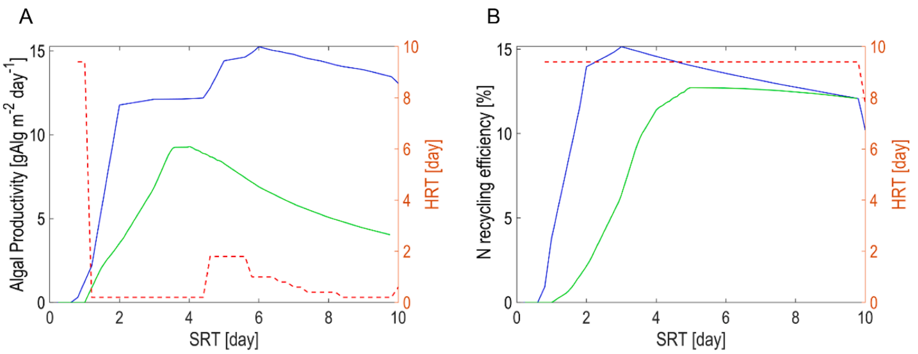

Fig. 8. Simulation results for influent TA = 22.5 mol m^{-3} and δ_L = 0.06 m, comparing the scenario with no solid/liquid separation (*i.e.* HRT = SRT, green continuous line) and the scenario where the HRT (red dotted line) is adapted according to the SRT in order maximize: A) the algal productivity [gAlg m^{-2} d^{-1}] and B) the nitrogen recycling efficiency [%] (blue continuous line).

Fig. 9. Simulation results for the conditions (HRT, SRT and δ_L) maximizing algae productivity (left column) and nitrogen recycling (right column). Results are given for both nominal (10 mol m^{-3}) and increased influent alkalinity (22.5 mol m^{-3}). A1, A2: biomass composition [gCOD m^{-3}]; B1,B2: algal biomass productivity [gAlg m^{-2} d^{-1}]; C1,C2: Nitrogen recycling in the algal biomass [%]; D1,D2: TAN removal rate [gN m^{-2} d^{-1}].

is higher. In addition, under this condition of HRT and SRT, the N_2O risk factor is 0% (Fig. 6F). The best result for nitrogen recycling efficiency (12.4%) was found for HRT = 10 d and SRT = 3 d, but it corresponds to a N_2O risk factor of 45% (Fig. 6F).

Finally, for this low liquid depth competition for inorganic carbon is stronger. It results that the N_2O emission risk appears for a wider range of cases compared to $\delta_L = 0.12$ m. In fact, while every value between 2 and 10 d can be applied for the SRT, the HRT should stay lower than 1 d, or being increased: i) up to 7, imposing SRT = 2 d; ii) up to 3, imposing SRT = 3 d; iii) up to 2, imposing SRT = 4–5 d.

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

Indeed, the TA influent addition allows to explore a wider range of HRT and SRT (proportionally to the alkalinity added) without operating under inorganic carbon limitation. This allows finding a much better trade-off between algal productivity and nitrogen recycling efficiency. Total alkalinity must be at least 20 mol m^{-3}, to guaranty that the N_2O risk factor is zero (Fig. 7C) for all HRT and SRT values.

Fig. 8A and B show more explicitly which is the advantage for algal production and nitrogen recycling respectively, when decoupling HRT and SRT, compared to the scenario without membrane separation application. Adapting the HRT for each SRT value can lead to a threefold increase in algal productivity or in the nitrogen recycling rate.

3.5. Analysis of the optimal regimes

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

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

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

3.6. Further step to target emerging contaminants

Microalgae can play a role in the treatment of emerging contaminants by bioadsorption, bioaccumulation and metabolic degradation [18]. There is currently no efficient mathematical model validated in outdoor conditions describing such processes. It is even likely that models should be molecule-specific to be accurate. However, the ALBA model predicts the oxygen concentration in the medium, which gives an index of the intensity of the oxidative stress influencing the molecular degradation [17]. Simulation results showed that, under the tested conditions, oxygen concentration could reach peaks up to 22 mgO_2 L^{-1} during the day. A modelling study can be further carried out, imposing as target objective to maximise the peaks of oxygen concentration in the reactor, also by exploring a wider range of HRT and SRT values. The model can therefore indirectly predict the benefit in terms of emerging contaminant removal due to oxidative stress. Further work must be carried out to quantify these aspects, and further integrate it with an estimation of the gain in emerging contaminants treatment with a separation system. This is an important point when computing the cost-benefit of the separation system.

As recently reviewed by You et al. [44], pollutants (including pesticides, metals, engineered nanomaterials, pharmaceutical and personal care products, and aromatic pollutants) affect both bacteria and algae by interfering with their relationships. Cell-to-cell adhesion, substrate exchange and biodegradation of organic pollutants, enhancement of signal transduction, and horizontal transfer of tolerance genes are defence strategies in algal-bacterial systems, which should be further tamed and applied in wastewater treatment systems. Enhancing the treatment capacity by continuous addition of more efficient algae and/or bacteria species is then the next step. This complementary strategy of bioaugmentation could be combined to the separation system to further enhance the process performance. Numeric simulations can guide the process optimization and development for this new and promising strategy.

3.7. Long-term membrane application for algae bacteria systems

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

However, membrane biofouling due to heavy suspended solids load, the small size of microalgae and their poor sedimentation properties, combined with bacteria and exopolymeric substances can lead to a progressive decline in the permeate flux and an increase in the energy demand for operating the process, thus limiting long term operations [41,42]. Most of the membrane designs were developed for wastewater treatment [43,44], with very different objectives, such as removal of pollutants and recovery of clean permeate vs recovery of valuable algal biomass [42,45]. The traditional fouling control techniques could be inefficient when targeting a by-product recovery [45] and new directions have emerged in recent years, while algal production was coupled to wastewater treatment [46]. Different fouling control techniques have been developed recently, supported by a better understanding of the interactions between the foulants and membrane surfaces. The main strategies consist in changing the operating conditions, in pre-treating the feed (using coagulation, adsorption, or oxidation), in mechanically cleaning of the membranes, and in modifying the membrane surfaces by coatings or blending with polymers and nanoparticles [45]. Choosing the adequate membrane cleaning mechanism is of major importance in terms of energy consumption, permeate production, and overall process performance [46,47]. The development of membranes for algae-bacteria systems with excellent bioactivity and associated to antifouling strategies is of crucial importance for long term applications.

4. Conclusion

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

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2022.138488.

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