Performance of a membrane-coupled high-rate algal pond for urban wastewater treatment at demonstration scale

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Abstract: The objective of this study was to evaluate the performance of an outdoor membrane-coupled high-rate algal pond equipped with industrial-scale membranes for treating urban wastewater. Decoupling biomass retention time (BRT) and hydraulic retention time (HRT) by membrane filtration resulted in improved process efficiencies, with higher biomass productivities and nutrient removal rates when operating at low HRTs. At 6 days of BRT, biomass productivity increased from 30 to 65 and to 90 g·m-3·d-1 when operating at HRTs of 6, 4 and 2.5 days, respectively. The correspondent nitrogen removal rates were 4, 8 and 11 g N·m-3·d-1 and the phosphorous removal rates were 0.5, 1.3 and 1.6 g P·m-3·d-1. The system was operated keeping moderate specific air demands (0.25 m3·m-2·h-1), resulting in reasonable operating and maintenance costs (€0.04 per m3) and energy requirements (0.287 kWh per m3). The produced water was free of pathogens and could be directly used for reusing purposes.

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Dear Editor,

Attached you will find the manuscript entitled "**Performance of a membrane-coupled high-rate algal pond for urban wastewater treatment at demonstration scale**" submitted for possible publication as an original research article in the Special Issue on Biological Nutrients Removal and Recovery (BNR) in *Bioresource Technology*.

The objective of this study was to evaluate the performance of an outdoor membrane-coupled high-rate algal pond equipped with industrial-scale membranes for treating urban wastewater. The important findings that must be highlighted are:

- Decoupling biomass retention time and hydraulic retention time by membrane filtration resulted in improved process efficiencies.
- Higher biomass productivities and nutrient removal rates were achieved when operating at low hydraulic retention times.
- The system was operated keeping moderate specific air demands, resulting in reasonable operating and maintenance costs and energy requirements.
- The produced water was free of pathogens and could be directly used for reusing

Yours sincerely,

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Highlights

- Decoupling hydraulic and biomass retention times increased the system performance
- N and P removal rates enhanced at lower hydraulic retention times
- Efficient operation achieved at low specific air demands
- Relatively low operational energy requirements

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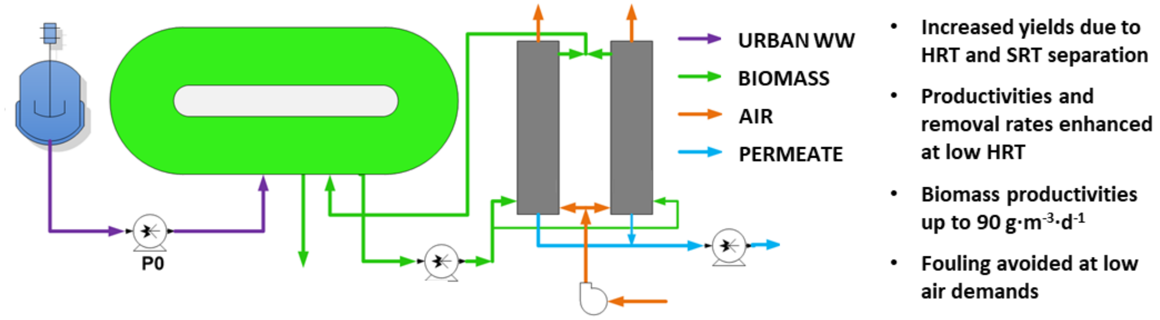
and maintenance costs (€0.04 per m^3) and energy requirements (0.287 kWh per m^3). The produced water was free of pathogens and could be directly used for reusing purposes.

Graphical abstract



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1. Introduction

The concept of circular economy relies on the recovery of valuable compounds from waste streams. To implement this approach, wastewater treatment plants are nowadays being shifted towards modern water resource recovery facilities (WRRFs), where the wastewater is not only treated and disposed, but also transformed into valuable products (*e.g.* energy, nutrients and reclaimed water).

The recovery of nutrients from urban wastewater (UWW) is a key goal to be achieved in future WRRFs due to its essential role in achieving a sustainable food production-consumption network. Microalgae-based processes have a huge potential as main actors for this purpose (Salama et al., 2017). Autotrophic microalgae are organisms able to grow using carbon dioxide as carbon source and light as energy source, assimilating at the same time the nutrients required for their growth (*i.e.* inorganic nitrogen and phosphorous). They convert these materials into biomass and a series of valuables organic compounds which are precursors of different forms of bio-energy (*e.g.* biogas, biodiesel, bio-ethanol, and bio-butanol) and other value-added products (*e.g.* products for livestock or fertilizers) (Wang et al., 2016). The cultivation of microalgae in wastewaters could reduce the production costs, generating at the same time clean water, recovering the nutrients initially present in the wastewater and capturing carbon dioxide during their growth by harvesting solar energy, thus reducing the environmental impact of the process (Wang et al., 2016). However, while these autotrophic microorganisms can efficiently reduce the concentrations of nutrients present in wastewater to very low values (*e.g.* 2.20 mg NH_4-N·L^{-1} and 0.15 mg PO_4-P·L^{-1} (Boelee et al., 2011)), they cannot remove organic matter, thus not being able to provide a complete wastewater treatment. Because of this, microalgae-based treatment systems are generally applied for tertiary wastewater treatment or are often combined with anaerobic pretreatments (Wang et al., 2015).

The utilization of microalgae-bacteria consortia to provide a complete single-stage treatment of wastewater is regarded as a potential solution for this problem. This wastewater treatment approach is based upon a synergetic interaction: the organic matter is degraded by heterotrophic bacteria, producing carbon dioxide, which is consumed by microalgae during photosynthesis, assimilating nutrients during this process and generating the oxygen that bacteria need to carry out aerobic respiration. In addition, other advantages of this mixed-culture systems have been postulated when compared to sole-microalgae cultures: (i) algae and bacteria produce vitamins and other organic compounds which can be beneficial for the growth of the partners, (ii) some microalgae generate a extracellular matrix that can provide attachment sites for bacteria and be used as carbon source, (iii) bacteria have been found to favour the flocculation of algae, enhancing biomass harvesting and (iv) the spatial distance for oxygen and carbon dioxide exchange is decreased (Arbib et al., 2017; Fernández-Sevilla et al., 2018; Galès et al., 2019; Liao et al., 2018; Shoener et al., 2019; Wang et al., 2016). Recent studies have demonstrated the feasibility of microalgae-bacteria consortia for UWW treatment. Removals of 92% of the biological chemical demand (BOD), 75% of the total nitrogen (N_T) and 93% of total phosphorus (P_T) have been reported using offshore photobioreactors (PBRs) (Novoveská et al., 2016). Photo-sequencing batch reactors reached removals of 87% of the chemical oxygen demand (COD) and 98% of the total Kjeldahl nitrogen (TKN), without the need of external aeration (Foladori et al., 2018). In high-rate algal ponds (HRAPs), removal efficiencies of 40-80% of the soluble COD, 80-100% of the NH_4^+ and 30-80% of the PO_4^{3-} have been reported (Galès et al., 2019). Therefore, these systems have appeared as an environmental-friendly wastewater treatment option able to remove both COD and nutrients while avoiding the need of supplying external oxygen or carbon dioxide.

Two key factors are limiting the application of microalgae-bacteria consortia for UWW treatment: (i) high amounts of total suspended solids (TSS) in the effluent (washout of microorganisms) and (ii) expensive biomass harvesting methods (Craggs et al., 2011; Solimeno and García, 2017; Wang et al., 2016). Both of these issues could be overcome by using membranes for biomass retention, enabling the decoupling of the biomass retention time

(BRT) and the hydraulic retention time (HRT) (Bhave et al., 2012; González-Camejo et al., 2019; Liao et al., 2018; Seco et al., 2018; Viruela et al., 2018). The application of membranes also provides an efficient solid-liquid separation, acting as biomass harvesting process and resulting in increased biomass concentrations and higher productivities due to enhanced nutrient removal efficiencies and higher organic loading rates (Bilad et al., 2014b; Honda et al., 2012; Luo et al., 2017). Furthermore, the addition of ultrafiltration membranes allows producing reclaimed water (*i.e.* with negligible levels of pathogens and suspended solids) from wastewater, directly applicable for several purposes (*e.g.* irrigation or fertirrigation, aquifer recharge or urban/industrial uses).

HRAPs have been widely applied for large-scale cultivation of microalgae worldwide, mainly due to their low investment and operational costs, their easy operation and maintenance and their low specific energy demand (Craggs et al., 2011; Kumar et al., 2015). Coupling membranes and HRAP can also help to overcome the low biomass productivities achieved, which has been recognised as a key challenge in HRAPs (Craggs et al., 2011; Dalrymple et al., 2013; Drexler and Yeh, 2014). Nevertheless, before membrane-coupled high-rate algal ponds (M-HRAP) can be applied industrially, research must still be carried out to ensure the feasibility of this technology (Bilad et al., 2014a). A key challenge that this technology may face is membrane fouling (Bilad et al., 2012; Marbelia et al., 2014; Sun et al., 2013; Wicaksana et al., 2012). In addition, the performance of M-HRAPs for wastewater treatment is significantly sensitive to the environmental and operating conditions, which must be optimized for each particular case. The available literature dealing with urban wastewater treatment via microalgae-bacteria consortia using membranes for decoupling BRT and HRT is limited, with no pilot/demonstration-scale studies available using HRAPs. Therefore, the performance and feasibility of this process must be evaluated, determining the achievable biomass productivities and the resulting nutrient recoveries. Potential operational issues must still be identified, setting the baselines for future optimization.

The objective of this study was to evaluate the performance of an outdoor M-HRAP equipped with industrial-scale membranes for treating UWW. The effect of the membrane addition to the HRAP system (decoupling of BRT and HRT) on the treatment performance was assessed. In addition, the effect of naturally varying environmental conditions (*i.e.* temperature and light intensity) on the outdoor M-HRAP performance were also studied. The capability of the process for UWW treatment was evaluated by determining the biomass productivities, the nutrient removal rates and the COD removal efficiencies, all of them being crucial parameters for these systems. After assessing the filtration performance, energetic and economic analyses were carried out to study the potential feasibility on the proposed process.

2. Materials and methods

2.1. Self-inoculation of the HRAP and influent wastewater

The plant was self-inoculated after starting feeding it with UWW. The start-up period for inoculation lasted for 1-2 weeks. An initial natural selection of the predominant microorganisms occurred naturally, facilitating the potential application of this technology. The main characteristics of the synthetic UWW used as substrate for microbial growth are shown in Table 1. This UWW was prepared according to Nopens et al. (2001). It was continuously fed to the HRAP from a refrigerated tank (kept at 4 °C) with a volume of 500 L.

Table 1. Characteristics of the synthetic urban wastewater

Parameter	Units	Mean ± SD
NH_4-N	mg N·L^{-1}	17.3±8.1
N_T	mg N·L^{-1}	45.5±24.2
PO_4-P	mg P·L^{-1}	3.9±1.6
P_T	mg P·L^{-1}	6.1±2.2
COD_T	mg·L^{-1}	332±55
VSS	mg·L^{-1}	89±24

SD stands for standard deviation, NH_4-N for ammonium-N, N_T for total nitrogen, PO_4-P for phosphate-P, P_T for total phosphorous, COD_T for total chemical oxygen demand and VSS for volatile suspended solids.

2.2. Description of the demonstration plant (M-HRAP)

A continuously-operated M-HRAP was used in this study. Its working volume was 22 m^3, with a depth of 0.3 m and a solar irradiance area of approximately 73.4 m^2. The HRAP (located in the south of France, Lat. 43.156711, Long. 2.995075) was continuously mixed by a paddlewheel. As it can be observed in Figure 1, showing a flow diagram of the system, the HRAP was connected to two membrane tanks (MT1 and MT2), each of them including one membrane bundle (with a filtration area of 3.44 m^2) that was obtained from one industrial-scale hollow-fibre ultrafiltration membrane unit (PURON® Koch Membrane Systems (PUR-PSH31), 0.03 μm).

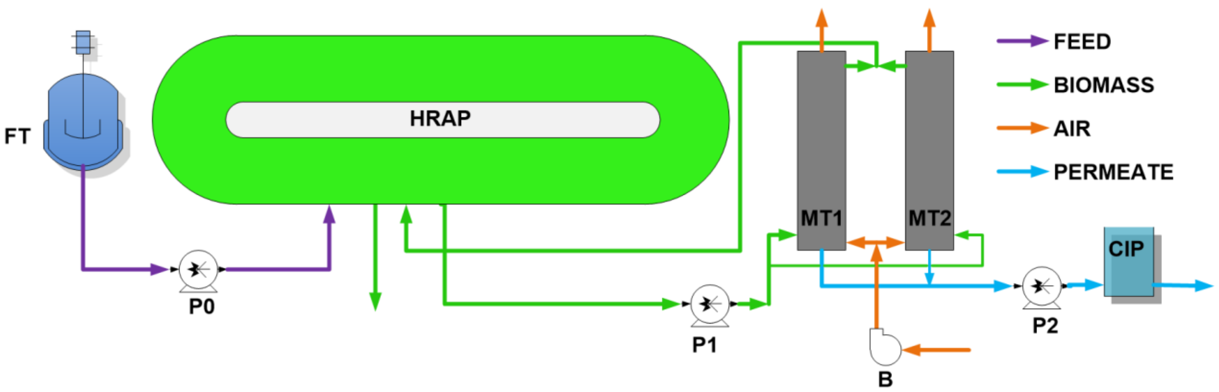


Figure 1. Flow diagram of the membrane-coupled high-rate algal pond. Nomenclature: FT: feeding tank; HRAP: high-rate algal pond; MT: membrane tank; CIP: clean-in-place; P:

pump; and B: blower

2.3. Monitoring of the plant operation

Different on-line sensors were installed in the M-HRAP to obtain real-time information of the state of the process. The on-line sensors placed in the HRAP were: (i) a pH-T transmitter (METTLER TOLEDO InPro® 4260 SG), (ii) a dissolved oxygen probe (METTLER TOLEDO InPro® 6800 G Amperometric Oxygen Sensor), (iii) an ultrasonic flowmeter for determining the influent flowrate (Titan Enterprises Ltd. atrato), and (iv) an irradiation sensor (Skye PAR Quantum Sensor) for measuring the photosynthetic active radiation (PAR). Moreover, several sensors were installed to monitor the membrane performance: two liquid flow-rate transmitters (one after the mixed liquor recycling pump and another after the permeate pump), three level transmitters (one for each membrane tank and another for the clean-in-place unit), one pressure transmitter for monitoring the transmembrane pressure in the membrane tanks, one air pressure transmitter (in the blower outlet) and one air flowmeter for measuring the air sparging for membrane scouring. The T and PAR values provided in this work refer to daily averages of the continuous PAR measurements, considering both daylight and night-time hours.

In addition to the on-line process monitoring, samples were taken three times per week from the influent, the mixed liquor and the effluent streams to evaluate the performance of the biological processes. The concentrations of the total and soluble COD (COD_T and COD_S, respectively), N_T, P_T, inorganic nutrients (NH_4^+, NO_2^-, NO_3^- and PO_4^{-3}), total suspended solids (TSS) and volatile suspended solids (VSS) were periodically measured. In addition, the optical density at 680 nm (OD_{680}) was used for VSS estimation (VSS_{680}). The structure of the microbial community was studied via quantitative polymerase chain reaction (qPCR), estimating the copies per liter of 18S rDNA (from chlorophyte) and bacterial 16S rDNA.

2.4. Operation of the plant

The M-HRAP was operated outdoors (*i.e.* under ambient temperature and solar irradiance conditions) at a constant BRT of 6 days and three different HRTs: 6 days (run I; no membrane operation), 4 days (run II) and 2.5 days (run III). As the temperature and the light irradiation are known to affect significantly the performance of microalgae-based wastewater treatment processes (Perin et al., 2016; Ras et al., 2013), the influence of these variables on the M-HRAP was studied during run IV (at equivalent BRT and HRT as run III). Table 2 shows the particular objective of each run period, as well as the applied working conditions and the daily average solar irradiances and culture temperatures.

Table 2. Average operating conditions and objectives of the different run periods

Run	Objective	BRT (d)	HRT (d)	J ($L\cdot m^{-2}\cdot h^{-1}$)	SAD_m ($m^3\cdot m^{-2}\cdot h^{-1}$)	Solar irradiance ($\mu E\cdot m^{-2}\cdot s^{-1}$)	Temperature (°C)
I	Evaluate effect of BRT and HRT decoupling	6	6	NA	NA	433±113	22.0±3.1
II		6	4	28, 14	0.3, 0.6	395±72	21.2±2.0
III		6	2.5	28	0.12 – 1.0	420±90	24.5±1.8
IV	Evaluate effect of light and temperature changes	6	2.5	27-31	0.6 – 1.2	253±195	14.1±1.1

BRT stands for biological retention time, HRT for hydraulic retention time, J for transmembrane flux, SAD_m for the specific air demand per membrane unit and NA not applicable

The membrane was operated with a gross transmembrane flux (J) of 28 $L\cdot m^{-2}\cdot h^{-1}$ (LMH) at the beginning of run II, lowering its value to 14 LMH afterwards. The value of J was fixed at 28 LMH during run III, varying between 27-31 LMH in run IV. During run II, the average specific air demand per square meter of membrane area (SAD_m) was set to 0.3 $m^3\cdot m^{-2}\cdot h^{-1}$ and then increased to 0.6 $m^3·m^{-2}·h^{-1}$ to maintain the desired J. The SAD_m varied from 0.12-1.0 $m^3·m^{-2}·h^{-1}$ and 0.6-1.2 $m^3·m^{-2}·h^{-1}$ during run III and IV, respectively. The pH varied freely according to variations in the carbon dioxide concentrations, related to the activity of microorganisms.

2.5. Analytical methods and microbial analysis

The concentrations of COD_T, COD_S, N_T, P_T and VSS were measured according to the Standard Methods (APHA, 2005). The concentrations of nutrients, *i.e.* NH_4^+, NO_2^-, NO_3^- and PO_4^{3-}, were determined by ion chromatography, according to Capson-Tojo et al. (2017).

2.6. Data treatment and calculations

To evaluate the performance of the M-HRAP treatment process, the nitrogen removal rate (NRR), the phosphorus removal rate (PRR) and the biomass productivity were calculated according to Equations 1 to 3:

$$NRR = \frac{Q \cdot (N_i - N_e)}{V_{MHRAP}} \quad \text{Eq. 1}$$

$$PRR = \frac{Q \cdot (P_i - P_e)}{V_{MHRAP}} \quad \text{Eq. 2}$$

$$Biomass\ productivity = \frac{Q_W \cdot X_{VSS}}{V_{MHRAP}} \quad \text{Eq. 3}$$

Where, Q is the treatment flow rate ($m^3·d^{-1}$), N_i is the concentration of nitrogen in the influent ($g\ N·m^{-3}$), N_e is the concentration of nitrogen in the effluent ($g\ N·m^{-3}$), V_{MHRAP} (m^3) is the total volume of the M-HRAP, P_i is the concentration of phosphorus in the influent ($g\ P·m^{-3}$), P_e is the concentration of phosphorus in the effluent ($g\ P·m^{-3}$), Q_w ($m^3·d^{-1}$) is the flow rate of wasted biomass and X_{VSS} ($g\ VSS·m^{-3}$) is the VSS concentration in the HRAP.

The photosynthetic efficiency (PE), and the carbon dioxide biofixation (CO_{2BF}) (kg CO_2 per m^3 of treated water) were also used as indicator of the biological activity. They were calculated according to Eq. 4, and Eq. 5, respectively.

$$PE\ (\%) = \frac{r_G \cdot H_B}{I \cdot S \cdot f} \cdot 100 \quad \text{Eq. 4}$$

$$CO_{2BF} = \frac{rG}{Y_{CO2} \cdot Q} \quad \text{Eq. 5}$$

Where r_G is the daily microalgae growth (kg VSS·d^{-1}), H_B is the lower heating value of dry biomass (22,900 kJ·kg VSS^{-1}), I is the PAR (μmol photons·m^{-2}·s^{-1}), f is a conversion factor (18.78 kJ·s·μmol $photons^{-1}$·d^{-1}), S is the surface of the open pond (m^2) and Y_{CO2} is the stoichiometric CO_2 capture for microalgae growth (0.52 kg VSS·kg CO_2^{-1}). For stoichiometric calculations of microalgae biomass composition, the chemical formula used in Viruela et al. (2018) was applied in this study (*i.e.* $C_{106}H_{181}O_{45}N_{16}P$).

The measured J values were standardized to 20 ºC, according to Eq. 6:

$$J_{20} = J \cdot\ e^{-0.0239 \cdot (T-20)} \quad \text{Eq. 6}$$

Where, J_{20} is the 20 ºC-standardized gross flux, J is the gross flux and T is the temperature in degrees Celsius.

2.7. Energy and economic analysis

2.7.1. Power requirements

The energy consumption of the M-HRAP unit was assumed to be mainly related to blowers (air sparging), pumps (culture media and permeate), and paddlewheel. The power requirements for pumps and blower were calculated as (Pretel et al., 2016). On the other hand, an energy demand of 0.4 W/m^2 was set for the paddlewheel.

2.7.2. Estimation of the operational and maintenance costs

The energy requirements of the blower, sludge recycling pump and permeate pump for filtration or back-flushing were calculated as explained in Robles et al. (2014). The costs related to energy consumption assumed an energy cost of 0.07 € per kWh, similarly to average electricity prices for industrial installations in Spain.

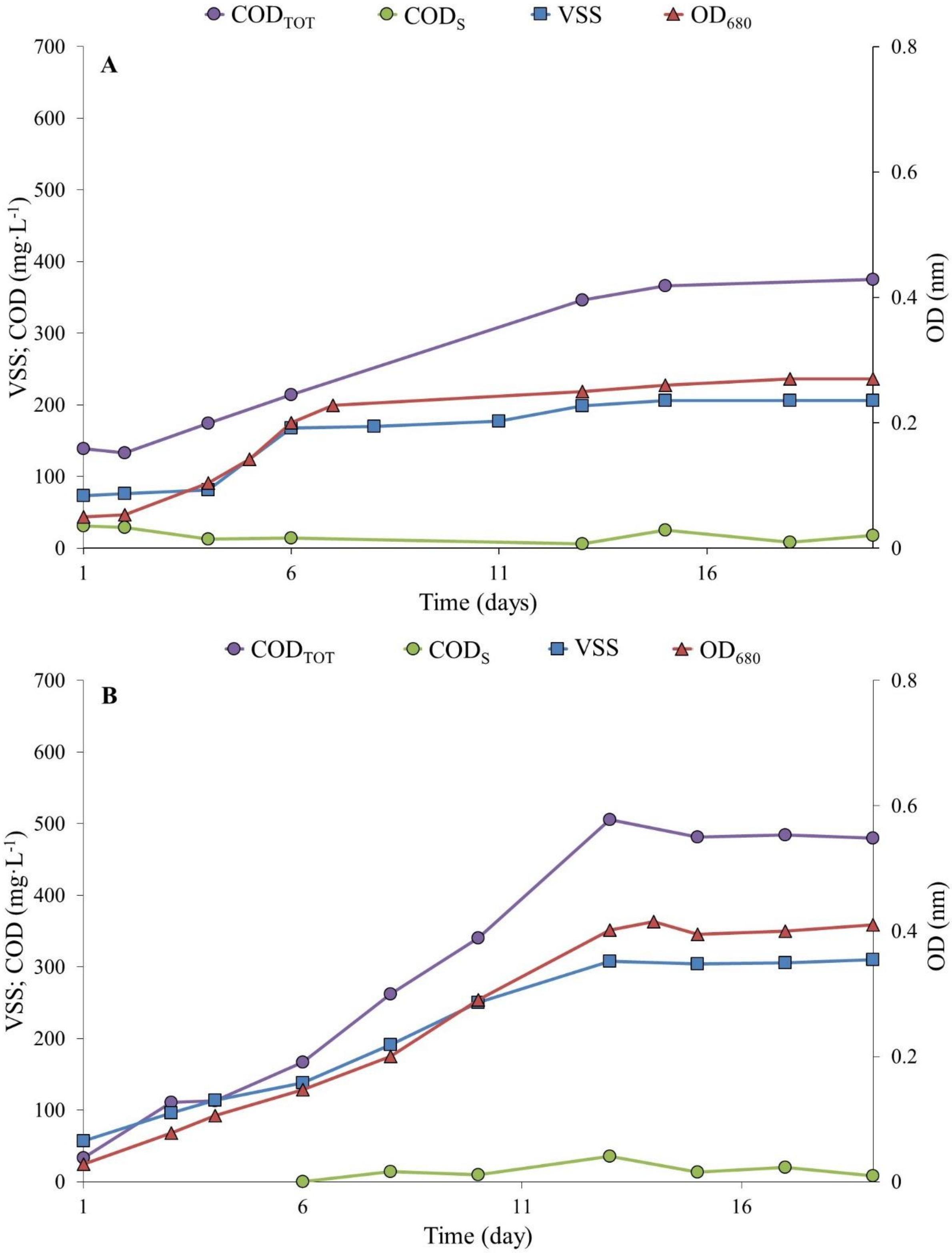
Other than the energy consumption due to air sparging and permeate and culture pumping, the costs related to membrane replacement and membrane chemical cleaning were considered. The useful membrane lifetime was estimated from the total chlorine contact specified by the manufacturer and the recommended membrane chemical cleaning frequency.

A more precise description of the costing methodology can be found in Robles et al. (2018).

3. Results and discussion

3.1. Influence of the BRT and HRT decoupling on the M-HRAP performance

As aforementioned, runs I-III were dedicated to study the influence of decoupling the HRT and the BRT, i.e. testing different HRTs for a given BRT. Starting with runs I to III, the corresponding evolutions of the concentration of COD_T, COD_S, VSS and the OD measurements (together with the estimated VSS) are presented in Figure 2.


COD_TOT
COD_S
VSS
OD_680
A
VSS; COD (mg·L-1)
OD (nm)
Time (days)
B
Time (day)

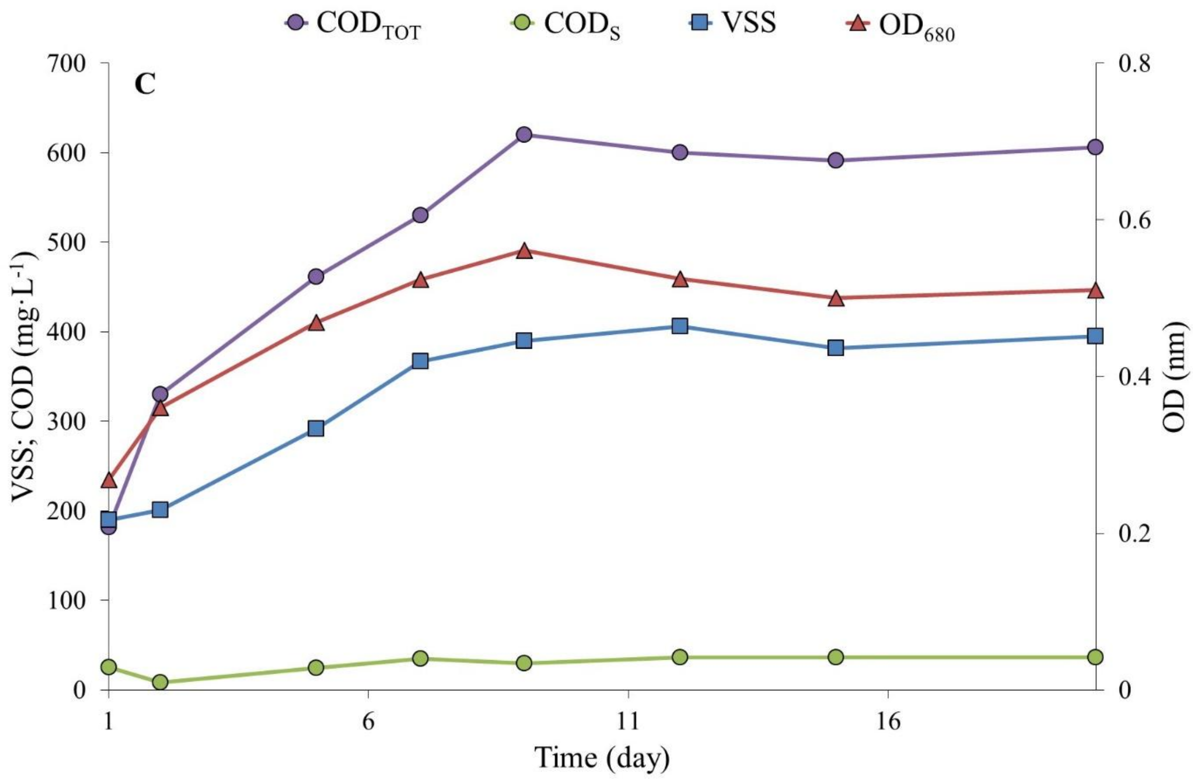


Figure 2. M-HRAP performance when operating at a BRT of 6 days and HRTs of (A) 6, (B) 4, and (C) 2.5 days. COD_T: total chemical oxygen demand; COD_S: soluble chemical oxygen demand; VSS: volatile suspended solids; OD_{680}: optical density at 680 nm.

The first observation to point out is the negligible values of the COD_S that existed in all the conditions after the incubation period (always below 50 mg $COD·L^{-1}$). This indicates that heterotrophic bacteria grew very rapidly initially, without any limitation for their growth in the applied working conditions. Nevertheless, the increasing VSS and COD_T concentrations that can be observed in all the figures suggest that the biomass concentration augmented in the M-HRAP (considering stable COD_S in the influent). This suggests the further development of an adapted microbial population, mainly due to growth of microalgae at this point. Although the raising COD_T concentrations occurred in the three run periods studied, the behaviours were clearly different. When comparing the biomass concentrations in the reactors at pseudo-steady state, it can be observed higher values when lowering HRTs (*i.e.* around 400, 500 and 600 mg VSS·L^{-1} at HRTs of 6, 4 and 2.5 days, respectively). This resulted in an enhanced general performance of the biochemical system, decreasing also the time to reach a stable community. The favoured biomass growth when decoupling the BRT and HRT can be attributed to two main factors: (i) the membrane avoided the wash-out of microorganisms, which otherwise would have left the reactor (improving the start-up process) and (ii) the increased mass flow rate of both COD and nutrients at lower HRTs allowed a faster development of the microorganisms.

This improvement can be easily appreciated in the results presented in Table 3, where the NRRs, PRRs and biomass productivities are given for each run period.

Table 3. Average results in runs I to III at pseudo-steady state for: nitrogen and phosphorous removal rates, biomass productivities, photosynthetic efficiency, and carbon dioxide biofixation

Run	BRT (d)	HRT (d)	NRR (g N·m^{-3}·d^{-1})	PRR (g P·m^{-3}·d^{-1})	Biomass productivity (g VSS·m^{-3}·d^{-1})	PE (%)	CO_{2BF} (kg CO_2·m^{-3})
I	6	6	4	0.5	30	1.0	0.2
II	6	4	8	1.3	65	4.0	0.3
III	6	2.5	11	1.6	95	3.5	0.4

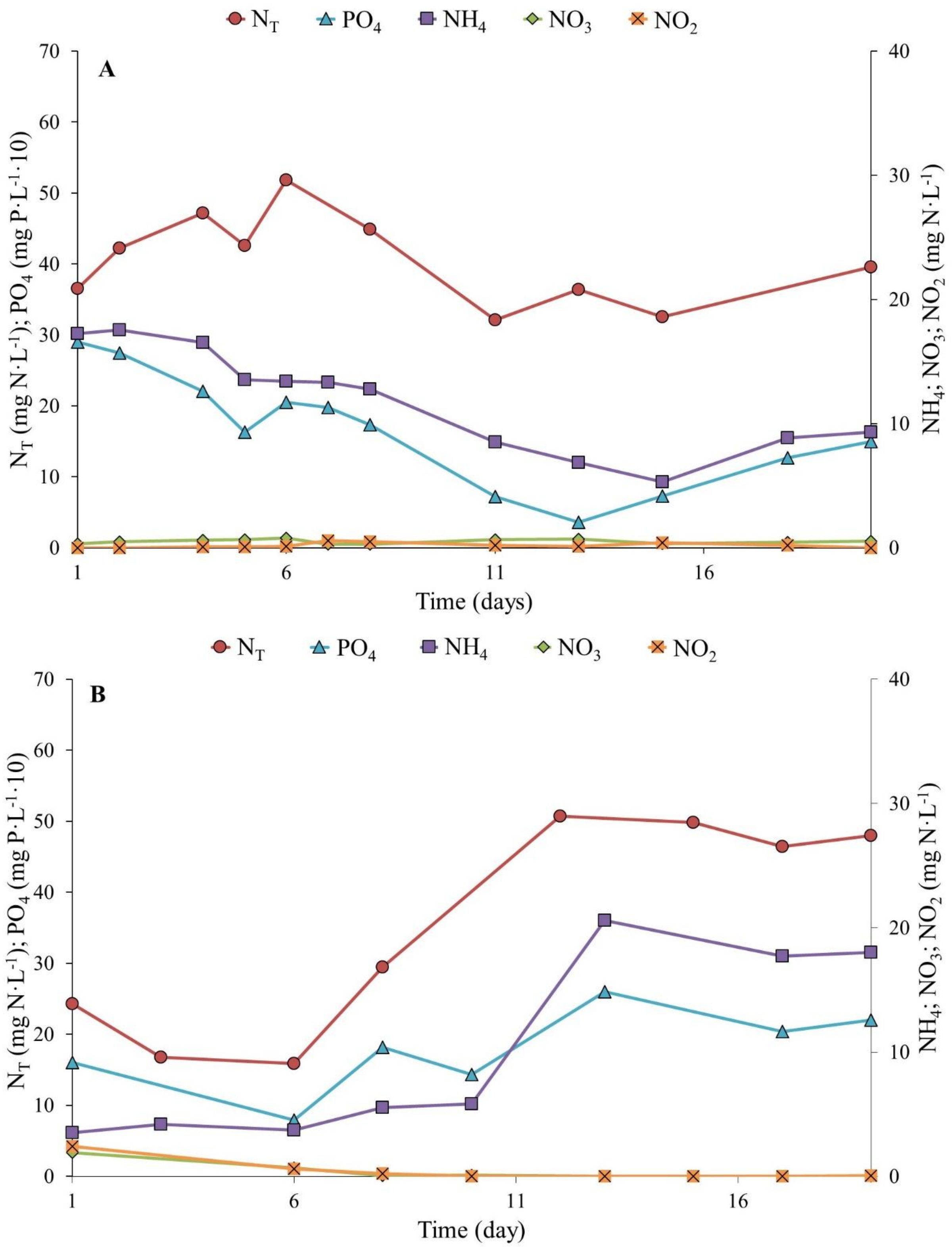
BRT stands for biological retention time, HRT for hydraulic retention time, NRR for nitrogen removal rate, PRR for phosphorous removal rate, PE for photosynthetic efficiency, CO_{2BF} for carbon dioxide biofixation, and VSS for volatile suspended solids

As this table illustrates, decoupling the BRT and the HRT increased significantly the nutrient removal rates and the biomass productivities. Decreasing the HRT by a factor of 2.4 (*i.e.* from 6 to 2.5 days) resulted in 3-folded NRRs and PRRs when comparing runs I and III. In addition, the biomass productivity increased from 30 to 65 and to 90 g VSS·m^{-3}·d^{-1} at decreasing HRTs of 6, 4 and 2.5 days, respectively. The increased biological activity due to

BRT/HRT decoupling can also be appreciated when looking at the average pseudo-steady state values of PE and CO_{2BF} during the different run periods, of 1% and 0.2 kg $CO_2 \cdot m^{-3}$ in run I, 4% and 0.3 kg $CO_2 \cdot m^{-3}$ in run II and 3.5 % and 0.4 kg $CO_2 \cdot m^{-3}$ in run III. Both the PE and CO_{2BF} increased during the run periods following an asymptotic pattern until reaching a maximum value, corresponding to the presence of a well-stablished microalgal community in the HRAP. Interestingly, when comparing these maximum values, the PE was 4-folded and the CO_{2BF} increased by 50 % between runs I and II, confirming the positive effect of biomass retention. The maximum CO_{2BF} was even further increased in run III. However, the PE was lower during run III when compared to run II. This is very likely caused by a shading effect related to the higher biomass concentrations, decreasing the light uptake efficiency. Similar phenomena have been reported previously at high HRTs (Viruela et al., 2018). This suggests that an optimum combination of BRT and HRT exists, allowing to optimize the performance of the system establishing efficient light uptake rates.

The results presented above are in agreement with previous studies dealing with membrane filtration coupled to outdoors microalgae-based treatment systems. Using a PBR for tertiary sewage treatment at BRT of 4.5 days and HRT of 3.5 days, optimum conditions were achieved, with a CO_{2BF} of 0.55 kg $CO_2 \cdot m^{-3}$ and a PE of 2.7 % (González-Camejo et al., 2019). Viruela *et al.* (2018) also achieved maximum biomass productivities, NRR and PRR (66 mg $VSS \cdot L^{-1} \cdot d^{-1}$, 7.7 mg $N \cdot L^{-1} \cdot d^{-1}$ and 1.2 mg $P \cdot L^{-1} \cdot d^{-1}$, respectively) at a BRT of 4.5 days. These results suggest that M-HRAPs can achieve similar (or even higher) productivities and nutrient removal rates than MPBRs with lower power requirements.

Despite the positive effect of HRT reduction, the high nutrient loading rates into the system resulted in higher concentrations of nutrients in the effluent. This fact can be appreciated in Figure 3, where the evolutions of the concentrations of N_T, NH_4-N, PO_4-P, NO_3-N and NO_2-N during runs I to III are given. Nevertheless, it must be pointed out that this result was obtained at a fixed BRT of 6 days. By optimizing this parameter, discharge limits could be obtained by favoring a faster algae growth (González-Camejo et al., 2019).



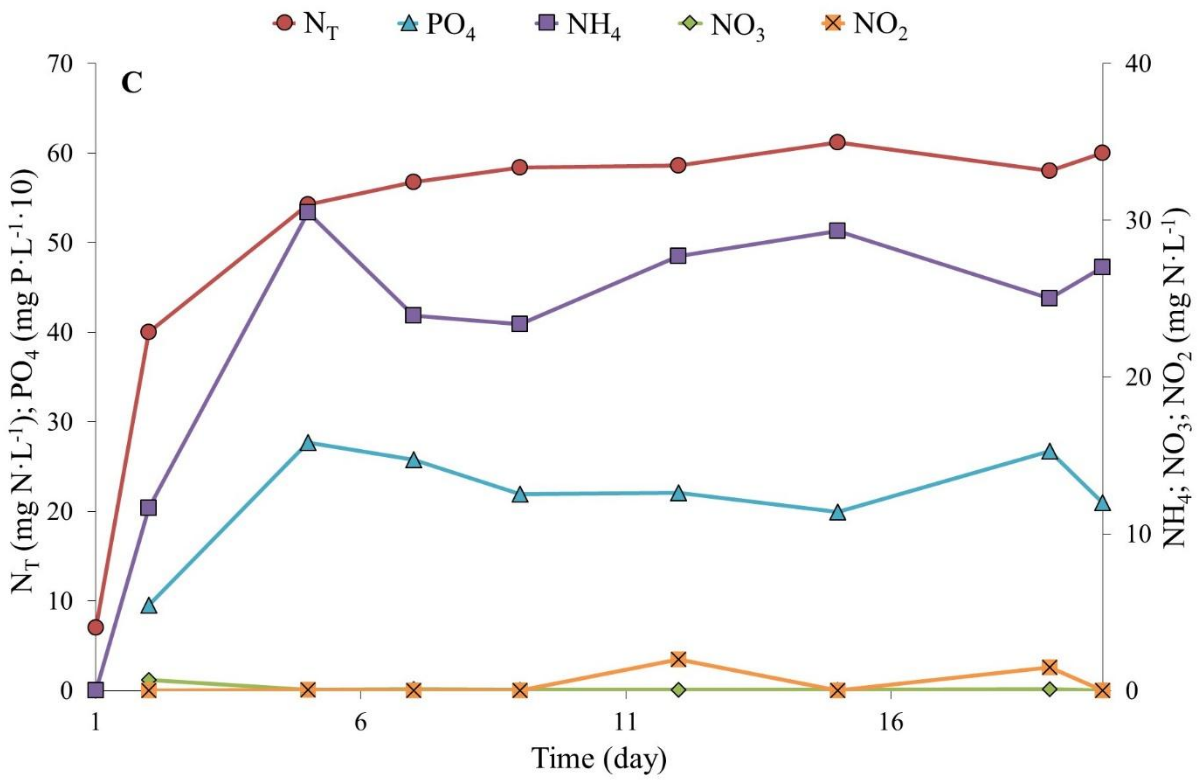


Figure 3. M-HRAP performance when operating at a BRT of 6 days and HRTs of (A) 6, (B) 4, and (C) 2.5 days. The evolutions of the concentrations of total nitrogen (N_T) in the mixed liquor and the inorganic nutrients (NH_4-N, PO_4-P, NO_3-N and NO_2-N) in the effluent are given

As it can be observed, while the concentrations of NH_4-N and PO_4-P were far below 15 mg $N{\cdot}L^{-1}$ and 2 mg $P{\cdot}L^{-1}$, respectively, at the end of run I, the concentrations of these species were 18 mg $N{\cdot}L^{-1}$ and 1.2 mg $P{\cdot}L^{-1}$ at the end of run II and of 28 mg $N{\cdot}L^{-1}$ and over 2 mg $P{\cdot}L^{-1}$ at the end of run III (N_T concentrations over 40 $mg{\cdot}L^{-1}$). This was simply related to the higher nutrient loading rates caused by the lower HRTs. Besides these higher nutrient concentrations in the effluent in run II, the achieved values were nearby the limits imposed by European effluent nutrient standards (European directive 91/271/CEE). Considering this and the negligible amounts of solids and microorganisms in the effluent from the M-HRAP, it is important to highlight that this high-quality effluent is suitable for its application in multiple reuse purposes, such as irrigation, fertigation, urban utilization, etc.

Although it can be concluded that the M-HRAP (at the conditions applied) treated the UWW successfully, it is clear that there is a great room for improvement. Control strategies aiming at optimizing the BRT and HRT for the given operating conditions (*i.e.* environmental conditions and influent characteristics) have a huge potential for improving the process performance (*e.g.* by minimizing the values of the BRT required to maximize the biomass productivities and nutrient removal rates while fulfilling the nutrients limits in the effluent).

Run IV served for evaluating the influence of the temperature and the light irradiation on the M-HRAP performance. The weather conditions (mainly T and solar irradiance) are known to have a significant effect on the performance of outdoors algae-based treatment systems, with open ponds being particularly affected by seasonal variations (Mata et al., 2010). The M-HRAP used in this study was run outdoors for several months, which allowed to obtain results at different ambient temperatures and natural irradiances. The results presented in Table 4 correspond to the plant performance during runs III and IV, with equivalent working conditions but under different meteorological conditions.

Table 4. Average nitrogen and phosphorous removal rates and biomass productivities in runs III and IV at pseudo-steady state

run	Solar irradiance ($\mu E \cdot m^{-2} \cdot s^{-1}$)	Temperature (℃)	NRR ($g\ N \cdot m^{-3} \cdot d^{-1}$)	PRR ($g\ P \cdot m^{-3} \cdot d^{-1}$)	Biomass productivity ($g\ VSS \cdot m^{-3} \cdot d^{-1}$)
III	420±90	24.5±1.8	11	1.6	95
IV	253±195	14.1±1.1	8	1.1	65

BRT stands for biological retention time, HRT for hydraulic retention time, NRR for nitrogen removal rate, PRR for phosphorous removal rate and VSS for volatile suspended solids

As expected, the lower solar irradiances and temperatures during run IV (average values of 253 $\mu E \cdot m^{-2} \cdot s^{-1}$ and 14.1 ºC, respectively) when compared with run III (420 $\mu E \cdot m^{-2} \cdot s^{-1}$ and 24.5 ºC) reduced nutrients removal rates and biomass productivities. The reduction on light lowered ATP production via photophosphorylation by algae. The lower temperatures are known to affect the algae growth rates (Ras et al., 2013). The combined effects of these parameters led to the reduction in the evaluated yields.

Although the effect of weather patterns on the performance of the M-HRAP cannot be neglected, it is interesting to consider that all the parameters used to evaluate the plant performance were higher during run IV when compared to run I (see Table 3 and Table 4). Therefore, the enhanced behaviour related to BRT/HRT decoupling (avoiding biomass wash-out) was able to overcome the negative effect of lower temperatures and light availabilities. This implies that M-HRAP technology can be applied in temperate climates to maintain acceptable performances during low temperature seasons.

3.2. Membrane filtration performance: energy and economic analysis

To assess the energy performance of the system (and thus its economic feasibility), it is essential to study the membrane filtration performance. The values of J, J_{20}, SAD_m, the specific air demand per permeate volume (SAD_P), the TMP and the VSS concentrations during runs II and III are presented in Figure 4.

Low SAD_m values were maintained at the beginning of run II (0.3 $Nm^3 \cdot h^{-1} \cdot m^{-2}$), aiming at keeping low energy requirements. However, the increasing VSS concentrations and membrane fouling led to a TMP peak around day 10. To keep the TMP below 0.4-0.5 bar and avoid membrane damage, J was lowered and the SAD_m was increased to 0.6 $Nm^3 \cdot h^{-1} \cdot m^{-2}$, which led to stable TMP values, but increasing the SAD_P due to the reduced J up to unsustainable values (see Figure 4A and Figure 4C). The relatively small reduction in the

TMP after increasing the SAD_m suggests that the membrane fouling responsible for the TMP peak was not caused by the formation of an easily-removable cake layer. Observations of the membrane showed that, although reversible, the fouling layer consisted of a remnant viscous layer, covering the surface of the membrane. This also suggests that further increasing the SAD_m would not improve the membrane performance and that back-flushing was more effective to clean the membrane than relaxation with air. Despite this issue, the last days of operation during this run period show that the membrane can be efficiently operated at relatively low SAD_m values, keeping the TMP within acceptable limits.

The same issue was observed during run III (Figure 4B and Figure 4D). The higher VSS concentrations led to a TMP peak earlier (days 6-9), which was corrected by further increasing the SAD_m, keeping the same J. Nevertheless, after a momentary drop, the TMP continued to increase, even when the SAD_m was raised up to unsuitable values of around 1.2 $Nm^3 \cdot h^{-1} \cdot m^{-2}$, confirming that increasing the SAD_m above 0.5 $Nm^3 \cdot h^{-1} \cdot m^{-2}$ did not improve the filtration performance. Because of this continuous TMP raise, the membrane was manually washed with water (no chemical regeneration occurred) on day 13. The instantaneous TMP drop confirmed the reversible nature of the fouling layer. After membrane cleaning, it was possible to keep the TMP below 0.1 bar with a SAD_m of 0.25 $Nm^3 \cdot h^{-1} \cdot m^{-2}$. This operation was maintained for over a week, without significant TMP increases. This suggests that it is possible to operate the system with low SAD_m without applying any chemical recovery to the membranes, simply by sporadically cleaning them with water.

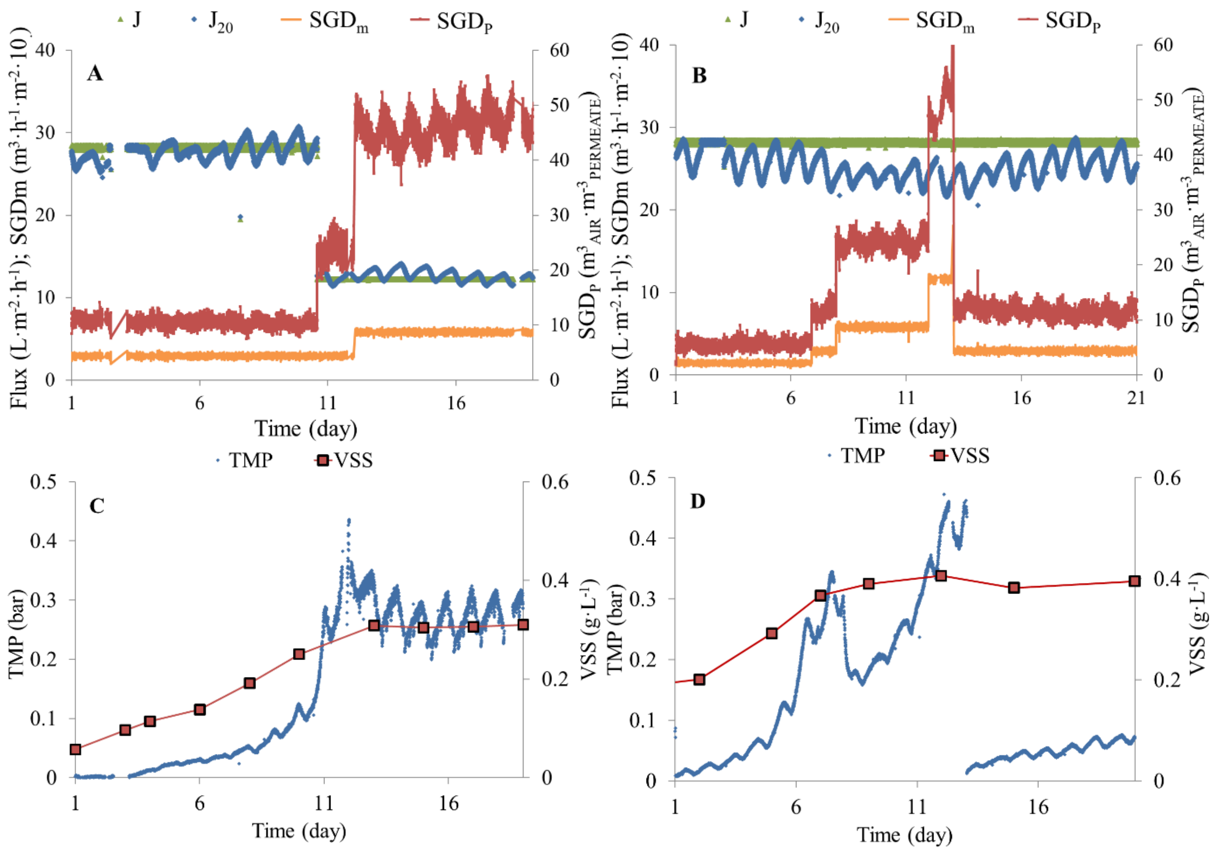


Figure 4. Evolution of J, J_{20}, SAD_m and SAD_P during (A) run II and (B) run III. The TMP and the VSS concentration (C) run II and (D) run III are also presented

As representative example, the values of J_{20} (28 $L \cdot m^{-3} \cdot h^{-1}$) and SAD_m (0.25 $m^3 \cdot m^{-2} \cdot h^{-1}$) achieved during the last section of run III were used to calculate the power requirements and the operational and maintenance costs of the M-HRAP. The results are presented in Figure 5.

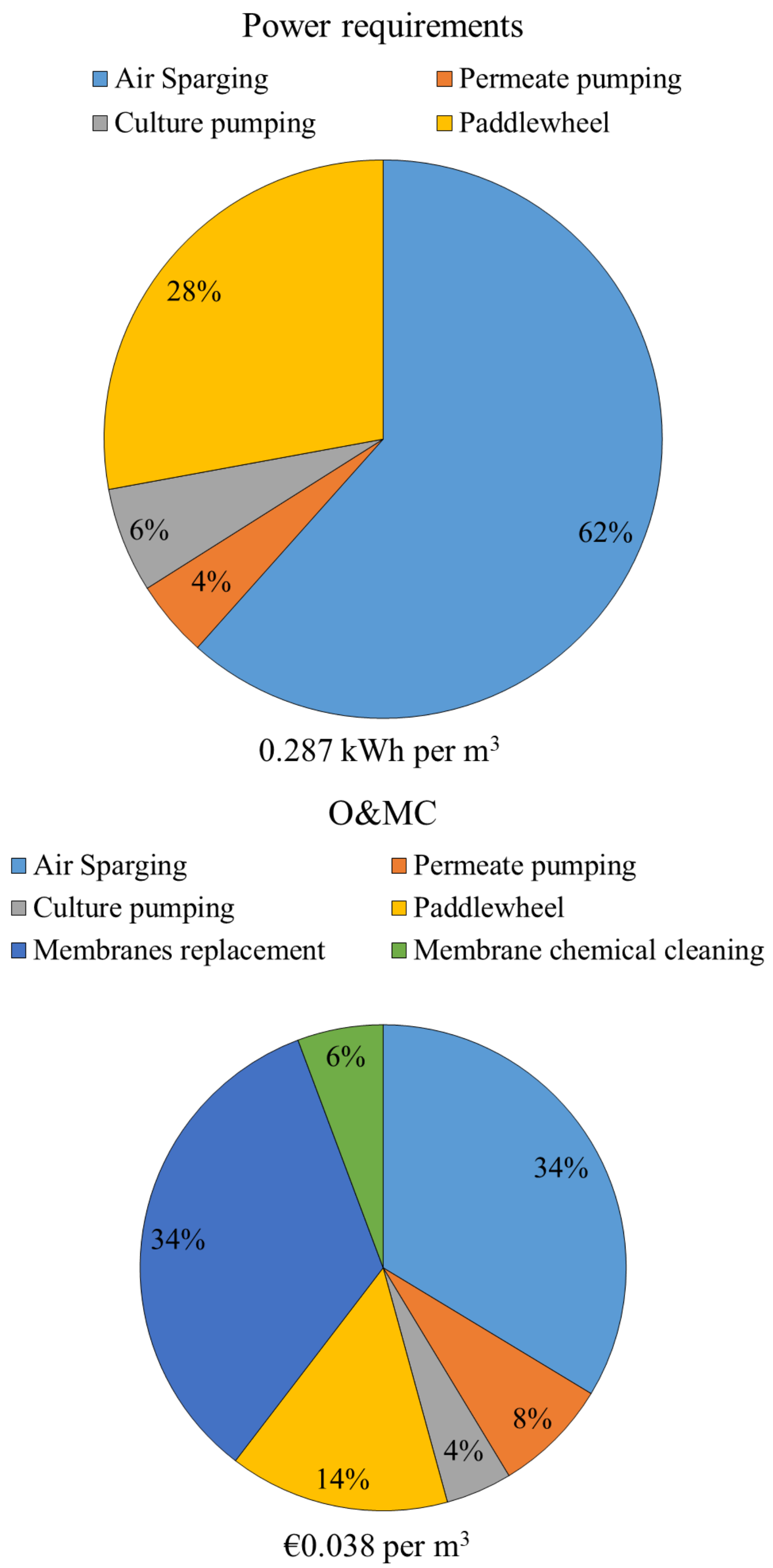


Figure 5. (A) Power requirements and (B) operational and maintenance costs (O&MC) for a full-scale plant design with a treatment capacity of 1,000 $m^3 \cdot d^{-1}$. J_{20} = 28 $L \cdot m^{-3} \cdot h^{-1}$; SAD_m = 0.25 $m^3 \cdot m^{-2} \cdot h^{-1}$

The low SAD_m resulted in energy requirements for the M-HRAP of around 0.29 kWh per m^3 of treated water. These values are lower than those achievable for other wastewater treatment methods, such as conventional activated sludge systems (0.25-0.6 kWh per m^3 or aerobic membrane bioreactors (0.50-2.5 kWh per m^3), pointing out the energetic feasibility of proposed M-HRAP system (Lazarova et al., 2012). Despite the low SAD_m applied, air sparging still accounted for almost 62% of the total energy requirements of the system, indicating that there is a clear room of improvement to further reduce this cost. Control strategies aimed at optimising the working conditions for given situations (*i.e.* HRTs, T and light intensity) have a great potential for further improving the energetic costs of these systems.

The operational and maintenance costs (O&MCs) further reinforce the importance of reducing the air sparging frequency, representing 34% of the total O&MCs. The results of the economic analysis also point out that, together with air sparging, the membrane replacement and its chemical cleaning account for most of the O&MC, representing 34% and 6% of the total, respectively. The frequency of membrane replacement and chemical cleaning depend greatly on how the plant is operated (*e.g.* the working TMP, the applied J, the VSS concentrations and the BRT). Therefore, control strategies optimising the working conditions can also help to reduce these costs. In addition, water could be effectively used for cleaning the membranes, applying an expert control system to optimise the back-flushing effect.

Finally, it is worth to point out that the water produced in the the M-HRAP was free of pathogens and could be directly used for reusing purposes (*i.e.* irrigation or fertirrigation). Therefore, the disinfecting cost needed for ad equating the effluent from other systems (*e.g.*

conventional activated sludge systems) is avoided.

4. Conclusions

Decoupling BRT and HRT enhanced biomass productivities (BPs), NRR and PRR. BP increased from 30 to 95 $g \cdot m^{-3} \cdot d^{-1}$ when lowering the HRTs from 6 to 2.5 days (at 6 days of BRT). NRR and PPR also increased from 4 to 11 $g\ N \cdot m^{-3} \cdot d^{-1}$ and 0.5 to 1.6 $g\ P \cdot m^{-3} \cdot d^{-1}$, respectively. The system kept high BPs, NRR and PRR at lower temperatures and solar irradiances. The membrane was efficiently operated at low SAD_m (around 0.25 $m^3 \cdot m^{-2} \cdot h^{-1}$), resulting in adequate energy requirements (0.287 $kWh \cdot m^{-3}$) and treatment costs (0.04 €$\cdot m^{-3}$). The produced water could be directly used for reusing purposes (*i.e.* irrigation).

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Figure and table captions

Figure 1. Flow diagram of the membrane-coupled high-rate algal pond. Nomenclature: FT: feeding tank; HRAP: high-rate algal pond; MT: membrane tank; CIP: clean-in-place; P:

pump; and B: blower

Figure 2. M-HRAP performance when operating at a BRT of 6 days and HRTs of (A) 6, (B) 4, and (C) 2.5 days. The evolutions of the total and soluble chemical oxygen demand (CODT and CODS, respectively), and the measured volatile suspended solids (VSS) are presented, together with the optical density values used for VSS estimation

Figure 3. M-HRAP performance when operating at a BRT of 6 days and HRTs of (A) 6, (B) 4, and (C) 2.5 days. The evolutions of the concentrations of total nitrogen (NT) in the mixed liquor and the inorganic nutrients (NH4-N, PO4-P, NO3-N and NO2-N) in the effluent are given

Figure 4. Evolution of J, J20, SADm and SADP during (A) run II and (B) run III. The TMP and the VSS concentration (C) run II and (D) run III are also presented

Figure 5. (A) Power requirements and (B) operational and maintenance costs (O&MC) for a full-scale plant design with a treatment capacity of 1,000 $m^3 \cdot d^{-1}$. J_{20} = 28 $L \cdot m^{-3} \cdot h^{-1}$; SAD_m = 0.25 $m^3 \cdot m^{-2} \cdot h^{-1}$

Table 1. Characteristics of the synthetic urban wastewater

Table 2. Average operating conditions and objectives of the different run periods

Table 3. Average results in runs I to III at pseudo-steady state for: nitrogen and phosphorous removal rates, biomass productivities, photosynthetic efficiency, and carbon dioxide biofixation

Table 4. Average nitrogen and phosphorous removal rates and biomass productivities in runs III and IV at pseudo-steady state

*Declaration of Interest Statement

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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