



Carbon footprint reduction by coupling intermittent aeration with submerged MBR: A pilot plant study

Paulo Marcelo Bosco Mofatto^{a,*}, Alida Cosenza^a, Daniele Di Trapani^a, Lan Wu^b, Bing-Jie Ni^c, Giorgio Mannina^a

^a Engineering Department, Palermo University, Viale delle Scienze, Ed. 8, Palermo 90128, Italy

^b Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, NSW 2007, Australia

^c Water Research Centre, School of Civil and Environmental Engineering, The University New South Wales, Sydney, NSW 2052, Australia

ARTICLE INFO

Keywords:

Wastewater treatment
Sludge minimisation
Simultaneous nitrification and denitrification process
Membrane bioreactor

ABSTRACT

To find a possible trade-off between effluent quality, sludge production, energy consumption and GHG emissions, this study monitored the carbon and nutrient removal and greenhouse gas (GHG) emissions in a membrane bioreactor (MBR) pilot plant with intermittent aeration (IA). The pilot plant was operated by alternating aerobic and anoxic conditions inside the biological reactor. Up to 98.2 % of carbon and 76.4 % of nitrogen were respectively removed through this MBR pilot plant with IA. The carbon footprint was equal to 2.4 kgCO_{2eq} m⁻³. Indirect emissions contributed the most to the carbon footprint (55.3 %), mainly due to energy consumption, despite the alterations in aeration during the pilot plant operation. The result of this study provides theoretical guidance for building the wastewater treatment plant in a sustainable way.

1. Introduction

The key challenges in wastewater treatment are reducing energy consumption and waste sludge. It is therefore necessary to reduce operating costs and containing the carbon footprint (CF) derived from wastewater treatment plants (WWTPs) [1]. The adoption of intermittent aeration (IA) in wastewater treatment plants (WWTPs) for nitrification/denitrification has been identified in the literature as a cost-effective strategy [2]. IA can be adopted under several operation modes, including batch feeding and continuous feeding. Systems such as Sequential Batch Reactors (SBR) and biological reactors cyclically operated under aerated and non-aerated conditions are where IA is typically applied [3]. The application of IA in WWTPs has several advantages in improving the nitrogen removal process, lowering the N₂O emissions, and reducing operational costs by limiting energy consumption [3]. In the recent decade, IA operation has been widely adopted in units used for the secondary treatment in WWTPs, such as oxidation ditch and SBRs [4,5]. Zhan et al. [4] found an improvement in the nitrogen removal process when the IA was implemented in a WWTP with the oxidation ditch. The nitrogen effluent in the system with IA was around 50 % lower than the system with continuous aeration. Li et al. [5] found an improvement in nitrogen removal from 50.5 % to 72.8 %

when IA was applied in an oxidation ditch pilot plant. A substantial reduction of energy consumption required for aeration when intermittent aeration was used has been widely reported by previous researchers [6, 7, among others]. Zhan et al. [6] and Elkaramany et al. [7] found a reduction of more than 60 % of the energy required for aeration in an SBR by applying intermittent aeration. Nitrous oxide (N₂O) is an important greenhouse gas (GHG) emitted from WWTPs, since it serves as an intermediate of both nitrification and denitrification processes, with a global warming potential 298 times higher than that of carbon dioxide [8]. Studies on intermittent aeration on Conventional Activated System (CAS) have revealed that alternating aerated and non-aerated conditions favours the reduction of N₂O production. However, the achievement of this result strongly depends on the average dissolved oxygen concentration during aerobic conditions [3]. Therefore, further studies are required to interpret the influence of aerobic/anoxic ratio on N₂O emissions.

Applying IA as a feasible strategy to improve membrane bioreactors (MBR) performance remains debatable [3]. The energy consumption of an MBR could account to more than 1/3 than that of CAS (i.e., 0.4 kWh m⁻³ and 0.67 kWh m⁻³ for CAS and MBR, respectively) [3,9,10]. Therefore, the adoption of membrane technology strongly increases the energy consumption of the WWTP and accelerates the operational cost. In

* Corresponding author.

E-mail address: paulomarcelo.boscomofatto@unipa.it (P.M. Bosco Mofatto).

<https://doi.org/10.1016/j.jece.2024.113115>

Received 4 January 2024; Received in revised form 5 April 2024; Accepted 18 May 2024

Available online 21 May 2024

2213-3437/© 2024 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

recent years, studies have been performed to reduce the energy requirement of MBR by optimizing membrane aeration systems, such as implementing on/off aeration cycles [11]. Lorain et al. [12] demonstrated a reduction of 20 % in energy consumption in a full-scale MBR operated with 10-sec-on and 30-sec-off aeration cycles. Yang et al. [13] obtained a 50 % reduction of the aeration energy by operating aeration cycles with 5-sec-on and 5-sec-off paradigm. Intermittent aeration in an MBR was proved recently to be more conducive to achieve high TN removal, low sludge production, and lower energy requirements for aeration than continuous aeration when treating textile wastewater [14]. Literature suggests the adoption of IA operation applied to the membrane tank in view of reducing aeration energy demand [12–14]. However, as authors are aware the influence of IA operation of the main biological reactor (e.g. aerobic reactor) in an MBR plant has never been tested. The IA operation of the main biological reactor can have strong influence on the biomass features that coupled with the specific conditions of MBR (e.g. high sludge retention time) could also indirectly affect the energy demand (e.g. influencing membrane fouling).

Previous literature mainly focused the influence of IA on energy demand and membrane fouling. However, no study has yet been available to investigate how intermittent aeration governs the carbon footprint in terms of direct, indirect, and derived emissions. Therefore, the novelty of this study is to operate an MBR pilot plant fed with real wastewater by using an MBR-IA configuration with two reactors: Intermittent aeration (anoxic/aerobic) and MBR.

2. Material and methods

2.1. Pilot plant configuration

The pilot plant was built at the Water Resource Recovery Facility of Palermo University [15,16]. The pilot plant was designed to treat 20 L h⁻¹ of real wastewater produced inside the University of Palermo Campus via MBR-IA process (Fig. 1). Briefly, the wastewater used in the present study derived from both the University canteen and an area of the university residence.

The pilot plant was consisted with the following units: one intermittent aeration reactor (225 L) with working mode of 40 minutes aeration and 20 minutes of anoxic conditions, followed by a membrane bioreactor (MBR) with an ultrafiltration hollow fibers membrane module (48 L). The membrane (PURON® Triple bundle Demo Module, nominal pore size 0.03 μm, membrane area 1.4 m²) was operated under filtration cycles (7 min filtration and 1 min backwashing) by using peristaltic pumps (Watson Marlow Qdos 30 Universal pumps, 30 L h⁻¹). The membrane reactor had a clean-in-place (CIP) tank for ordinary backwashing. An oxygen depletion reactor (ODR, 53 L) was inserted in the internal recycling line between the MBR and IA reactor, depleting

the dissolved oxygen concentration before entering the IA reactor. It is worth noting that the MBR compartment was continuously aerated, to control the fouling development.

2.2. Wastewater features and operation

Table 1 summarizes the average and standard deviation (SD) pilot plant operational conditions and the influent wastewater characteristics in terms of nutrients. Although the plant was designed for treating 20 L h⁻¹ of wastewater, the average flow rate was lower (16.4 ± 0.6 L h⁻¹) due to the membrane fouling.

2.3. Analytical methods

Chemical oxygen demand (COD), ammonia nitrogen (NH₄⁺-N), nitrate (NO₃⁻-N), nitrite (NO₂⁻-N), orthophosphate (PO₄³⁻-P), total suspended solid (TSS) concentrations, biological oxygen demand (BOD) and Total Nitrogen (TN) were measured according to Standard Methods [17], twice a week. Respirometric tests have been performed to analyse the kinetic parameters according to the literature [18].

The intermittent aeration mixed liquor evaluated the sludge volume index (SVI). Furthermore, extra polymeric substances (EPS) and soluble microbial substances (SMP) both in terms of carbohydrates and proteins (namely, EPS_p, EPS_c, SMP_p, SMP_c) have been analysed according to Mannina et al. [19] once week in the mixed liquor contained inside the intermittent aeration and MBR tanks.

The observed yield coefficient (Y_{obs}) was calculated based on the ratio between the cumulative mass of TSS produced and the cumulative mass of COD removed [20]. TCOD_{in} and TCOD_{out} were the concentrations in the influent and effluent, respectively. Q_i was the daily influent flow rate. ΔX was the daily excess sludge production.

$$Y_{obs} = \frac{\Delta X}{Q_i \bullet (TCOD_{in} - TCOD_{out})} \quad (gSSTgCOD^{-1}) \quad (1)$$

Dissolved and gaseous N₂O concentration was measured according

Table 1

Average values of the main influent and operational features for each experimental period; SD = Standard Deviation.

Parameter	Symbol	Units	Average	SD
Total COD	TCOD	[mg L ⁻¹]	1534	493.5
Soluble COD	sCOD	[mg L ⁻¹]	208	80.5
Total Nitrogen	TN	[mg L ⁻¹]	32	4.8
Ammonium	NH ₄ ⁺ -N	[mg L ⁻¹]	28	2.8
Phosphate	PO ₄ ⁻ -P	[mg L ⁻¹]	6	1.6
Flow Rate	Q _{IN}	[L h ⁻¹]	16	0.6

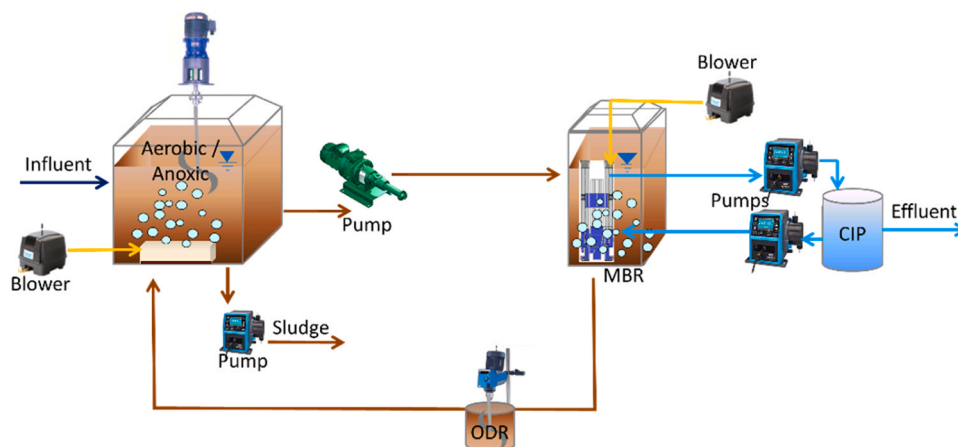


Fig. 1. Representation of the pilot plant.

to the procedure described by Mannina et al. [21] by using a Gas Chromatograph (GC) (Thermo Scientific™ TRACE GC) equipped with an Electron Capture Detector (ECD). Moreover, the N₂O–N was also quantified using each reactor's off-gas flow rate measurement. The N₂O emission factor (EF_{N2O}) has been calculated according to Eq. 2 [22]. N₂O–N_g and N₂O–N_d represented the nitrous oxide gaseous and dissolved concentration, respectively. HRT was the plant hydraulic retention time. HRT_{hs} was the tank headspace hydraulic retention time. TN was the influent total nitrogen concentration.

$$EF_{N2O} = \frac{N_2O - N_g/HRT_{hs} + N_2O - N_d/HRT}{TN/HRT} \quad (2)$$

Moreover, the membrane fouling was quantified by monitoring the transmembrane pressure (TMP) (kPa) and the permeate flux (J) (m³ m⁻² s⁻¹) every day to calculate the total membrane resistance (RT) according to the general form of the Darcy's Law (Eq. 3). ΔP [kPa] is the TMP variation, and μ [Pa.s] is the permeate viscosity.

$$R_T = \frac{\Delta P}{\mu \bullet J} \quad (m^{-1}) \quad (3)$$

Membrane fouling was controlled by physical and chemical cleanings requiring the membrane to get off from the MBR tank. According to the manufacturer's suggestions for modules to be used for scientific scope, physical cleanings were done by first removing the membrane from the tank and then manually eliminating the solids from the membrane surface and flushing them with clean tap water. Chemical cleanings were performed by using a 4 % sodium hypochlorite (NaClO) solution according to the manufacturer's suggestion. In detail, chemical cleanings were carried out to maintain the transmembrane pressure (TMP) below 0.7 bar, the acceptable range suggested by the manufacturer. Specifically, in view of performing chemical cleaning the membrane was submerged for 6 hours (after a physical cleaning) in a tank containing hot (35°C) 4 % NaClO solution. **2.4 Carbon footprint calculation**

Direct, indirect, and derived emissions have been quantified to assess the pilot plant's carbon footprint (CF) under study. Direct emissions (DE) have been quantified by considering the equivalent CO₂ (CO_{2,eq}) due to the organic carbon oxidation (CO_{2,OrgOx}, Eq. 4), due to the endogenous respiration (CO_{2,Endog}, Eq. 6) and the equivalent CO₂ due to the N₂O emission (CO_{2,eq,N2O}). CO_{2,OrgOx}, CO₂, and Endog have been quantified according to Boiocchi et al. [23].

$$CO_{2,OrgOx} = FC_S \bullet r_{o2} \quad (kgCO_2 \quad d^{-1}) \quad (4)$$

FC_S is the conversion factor describing the amount of CO₂ emitted per kg of consumed O₂ (equal to 1.1 kg CO₂ kg⁻¹ O₂ according to Boiocchi et al. [23]), and r_{o2} is the amount of oxygen consumed per day calculated according to Eq. 5.

$$r_{o2} = V_{rs} \bullet \left(\frac{1}{f - 1.42 \bullet Y} \right) (kgO_2 d^{-1}) \quad (5)$$

Where V_{rs} is the BOD₅ removed [in kgBOD₅ d⁻¹] calculated based on the measured data, f equals 0.68, and Y is the cell growth rate [in kgVSS kgBOD₅⁻¹]. In this study, the average calculated Y_{obs} value was adopted.

$$CO_{2,Endog} = FC_{End} \bullet m_{VSS} \quad (kgCO_2 d^{-1}) \quad (6)$$

Where FC_{End} is the conversion factor describing the amount of CO₂ emitted per kg of produced VSS (equal to 1.947 kg CO₂ kgVSS⁻¹ according to Boiocchi et al. [23]), and m_{VSS} is the mass of VSS evaluated based on the mass balance.

The CO_{2,eq,N2O} has been quantified based on the measured data according to Eq. 7.

$$CO_{2,N2O} = Q_g \bullet C_{g,N2O} \bullet GWP_{N2O} \quad (eq.7) \quad (kgCO_{2eq} d^{-1}) \quad (7)$$

Where Q_g [m³ d⁻¹] is the average gas flow, C_{g,N2O} [kgN₂O m⁻³] is the average gaseous measured N₂O concentration emitted and GWP_{N2O} [kgCO_{2eq} kgN₂O⁻¹] is the N₂O global warming potential (equal to 298 according to IPCC [24]).

Indirect emissions (IE) count the equivalent CO₂ due to energy consumption (CO_{2,eq,En}) (Eq. 8) and to the sludge treatment, transportation and landfill disposal (CO_{2,eq,Sludge}) (Eq. 9).

$$CO_{2,eq,En} = E_n \bullet FC_{En} \quad (kgCO_2 kWh^{-1}) \quad (8)$$

Where E_n [kWh d⁻¹] is the total energy consumption of the water line and FC_{En} [kgCO_{2eq} kWh⁻¹] is the conversion factor of the energy (equal to 0.252 kgCO_{2eq} kWh⁻¹ according to EEA [25]).

$$CO_{2,eq,Sludge} = M_{sludge} \bullet FC_{Sludge} \quad (kgCO_{2eq} d^{-1}) \quad (9)$$

Where M_{sludge} [ton d⁻¹] is the mass of wasted sludge per day and FC_{Sludge} [kgCO_{2eq} ton⁻¹] is the emission factor due to the sludge treatment, transportation and landfill disposal (equal to 714.74 kgCO_{2eq} ton⁻¹ according to Zhao et al. [26]).

Finally, the derivative emissions (DerE) have been quantified as that originated from the pollutants discharged into receiving water bodies (Eq. 10).

$$DerE = CO_{2,eq,effBOD} + CO_{2,eq,effN2O} \quad (kgCO_{2eq} d^{-1}) \quad (10)$$

Specifically, the contribution of BOD (CO_{2,eq,effBOD}) and dissolved N₂O (CO_{2,eq,effN2O}) have been calculated according to Eqs. 11 and 12, respectively.

$$CO_{2,eq,effBOD} = M_{BOD} \bullet FC_{BOD} \quad (kgCO_{2eq} d^{-1}) \quad (11)$$

Where M_{BOD} [kgBOD d⁻¹] is the mass of discharged BOD per day and FC_{BOD} [kgCO_{2eq} kgBOD⁻¹] is the conversion factor due to the BOD discharge (equal to 0.96 kgCO_{2eq} kgBOD⁻¹ according to Boiocchi et al. [23]).

$$CO_{2,eq,effN2O} = Q_w \bullet C_{l,N2O} \bullet GWP_{N2O} \quad (kgCO_2 d^{-1}) \quad (12)$$

Where Q_w [m³ d⁻¹] is the average treated flow rate, C_{l,N2O} [kgN₂O m⁻³] is the average liquid measured N₂O concentration discharged into the water body, and GWP_{N2O} [kgCO_{2eq} kgN₂O⁻¹] is the N₂O global warming potential (equal to 298 according to IPCC [24]).

3. Results and discussion

3.1. Pilot plant performance

Fig. 2 shows the nutrients concentration (TCOD, sCOD, N-NH₄, TN and PO₄-P) in influent wastewater and effluent and their removal efficiency during the experimental campaign.

The average removal efficiency of TCOD was equal to 98.2 ± 1.1 % (Fig. 2a). This result is in line with the literature. In the study by Yilmaz et al. [14] a slightly lower COD removal efficiency (84–91 %) was noticed by using intermittent aeration of 90 min on /360 min off in an MBR pilot plant (only MBR reactor); nevertheless, this lower result can likely be due to the different wastewater features of textile origin. The TCOD removal efficiency observed in the study carried out by Kim et al. [27], higher than 95 %, was more in line with that achieved in the present study. Regarding the sCOD removal efficiency (Fig. 2b), the average sCOD removal efficiency was calculated based on the value of influent and permeate and was equal to 86.0 % (±7.1). The sCOD removal was mainly due to the solid/liquid separation by the membrane. Indeed, around 59.7 % (±19.1) of sCOD before membrane filtration was removed due to the biological process (removal performance between MBR and permeate). Moreover, between the 3rd and 18th days of the experimental campaign, the worsening of sCOD

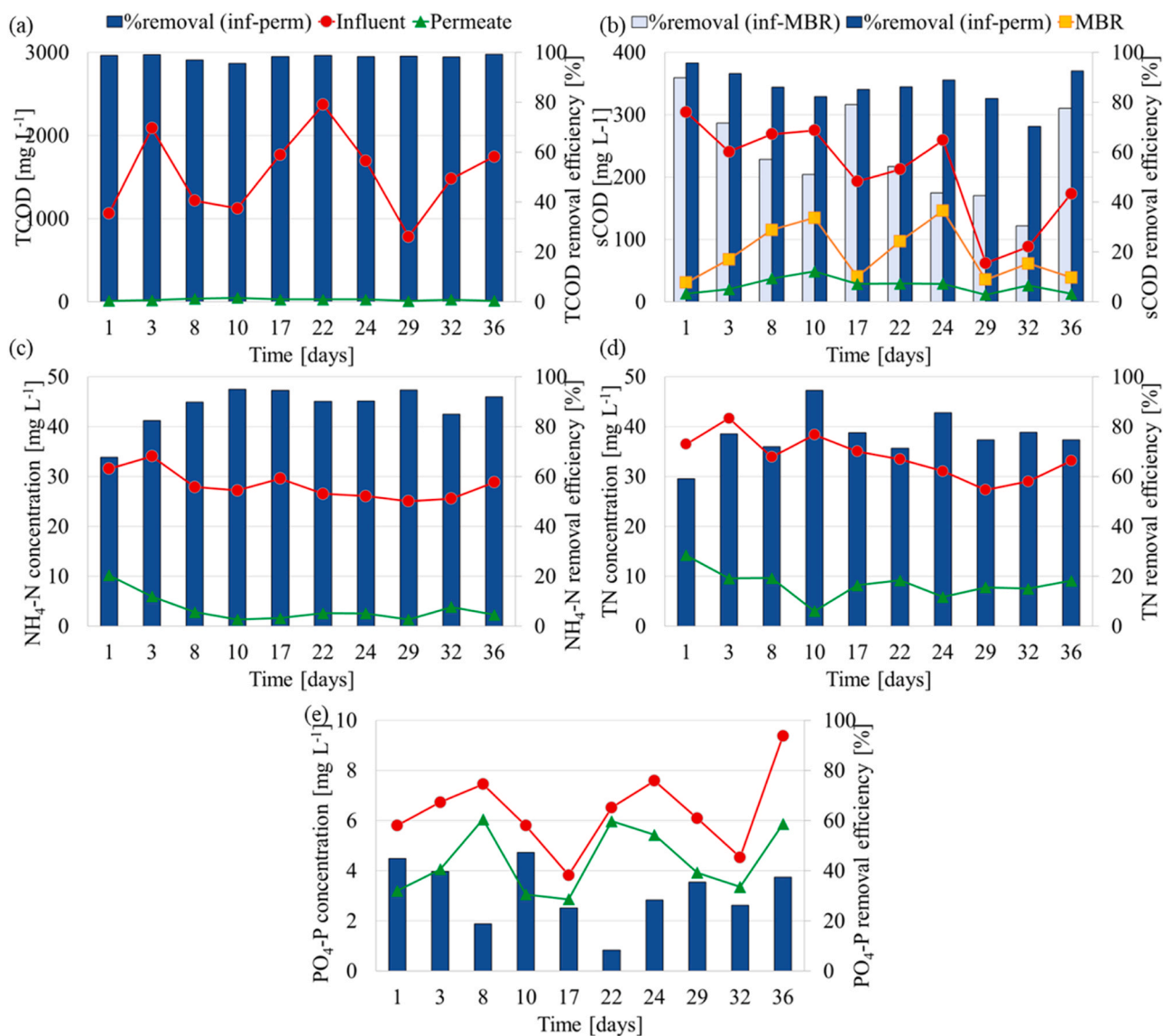


Fig. 2. Pattern of influent, effluent, and removal efficiency for total TCOD (a), sCOD (b), NH₄-N (c), Total Nitrogen (d) and PO₄³⁻⁻P (e).

removal efficiency (between MBR and permeate) was noticed, likely due to the higher EPS concentration in the MBR during these days. According to Kim et al. [28], EPS can cause operational problems, such as the production of biofilm that causes fouling in MBRs, thus worsening the membrane performances. The further worsening of the sCOD removal efficiency (occurred during days 29 and 32) can be related to the decrease of the influent sCOD concentration.

The removal efficiencies of N-NH₄ and TN were 88.1 % (±8.3) and 76.4 % (±7.8) (Fig. 2c & d), respectively. These results are in line with the studies of Yilmaz et al. [14] and Kim et al. [27] who reported an average of 74 % and 72.7 % TN removal efficiency, respectively. The high nitrogen removal efficiency attained by Yilmaz [14] was attained in the system without using external COD addition. However, Lim et al. [29] noticed a TN removal performance between 35 % and 70 % using an MBR operated at intermittent aeration (90 min on / 60 min off). Indeed, the authors suggest that this period with anoxic conditions was not sufficient for a complete denitrification.

The average removal efficiency of PO₄³⁻⁻P over the experimental period was around 31.2 % (±12.1). This value is lower than that reported by other researchers who also adopted IA or MBR as a strategy to control the pollutant removal efficiency during wastewater treatment. For instance, Mannina et al. [30] successfully removed 37 % of PO₄³⁻⁻P

removal via MBR-IFAS system. More than 83 % of PO₄³⁻⁻P was removed using an IA system [31]. Biological P removal depends on the selection and proliferation of phosphorus-accumulating organisms (PAOs). The survival of these organisms is controlled by the rapid substrate uptake during anaerobic phase, which allows PAO growth under anoxic and subsequent aerobic conditions. The slightly higher removal efficiency in the continuous aerobic phases is attributed to the increasing phosphorus uptake. Since the anoxic period applied herewith was lasted for 20 min and only consisted of 1/3 of the total cycle in the reactor, the anoxic time might not be sufficient for the growth of PAO, thus obtaining a low P-PO₄ removal efficiency.

3.2. Operational parameters and sludge properties

Table 2 summarized the average and standard deviation (SD) values of Y_{obs}, F/M, SRT ratio and total suspended solids (TSS) of the experimental campaign. In terms of sludge production, an average value of 0.12 g TSS g⁻¹ COD for Y_{obs} was obtained. This value is in line with previous MBR studies [32]. Indeed, Ferrentino et al. [33] obtained an average value of 0.12 g TSS g⁻¹ COD for Y_{obs} by using MBR systems.

During the experimental campaign, the sludge wasting was carried out when the TSS in the intermittent aeration reactor was higher than

Table 2

Food / Microorganism Ratio (F/M), observed biomass yield coefficient (Y_{obs}) and sludge retention time (SRT) for the experimental period.

Parameter	Symbol	Units	Average	SD
Observed yield coefficient	Y_{obs}	[gTSS/gCOD]	0.12	0.06
Food / Microorganism Ratio	F/M	[gCOD/gTSS d]	0.09	0.04
Sludge Retention Time	SRT	[d]	176.7	49.9
Total suspended solids IA reactor	TSS_{IA}	[g/L]	3.65	0.45
Total suspended solids MBR	TSS_{MBR}	[g/L]	7.09	3.52

4 g/L. The cumulative sludge production obtained during the entire duration of the experimental was 396 gTSS, resulting in 9.9 gTSS d^{-1} . Fig. 3 shown the SMP and EPS-specific concentrations measured during experimental campaign in the IA reactor and the MBR. The average specific EPS concentration during the experimental campaign was $174.29 \text{ mg g}^{-1}\text{TSS}$ (± 109) and $167.26 \text{ mg g}^{-1}\text{TSS}$ (± 105) for IA reactor and MBR, respectively.

3.3. Membrane fouling

The total membrane resistance (R_T) is reported in Fig. 4. R_T rapidly increased between the 12th and 15th days of the experiment, with peak value attained at $1.8 \cdot 10^{13} \text{ m}^{-1}$. This result was mainly attributed to increased TSS concentration in the MBR during this period. Thus, considering the reducing R_T value during the 15th day of the experiment, a chemical cleaning of the membrane was necessary. Bound EPSs is another factor that can cause a higher fouling rate in an MBR system [34]. When more cleanings were required, the specific EPS bound was higher than 150 mg gTSS^{-1} in both reactors.

Table 2 showed the total SRT calculated for the whole experiment period (40 days). Although it is a very high value compared to Yilmaz

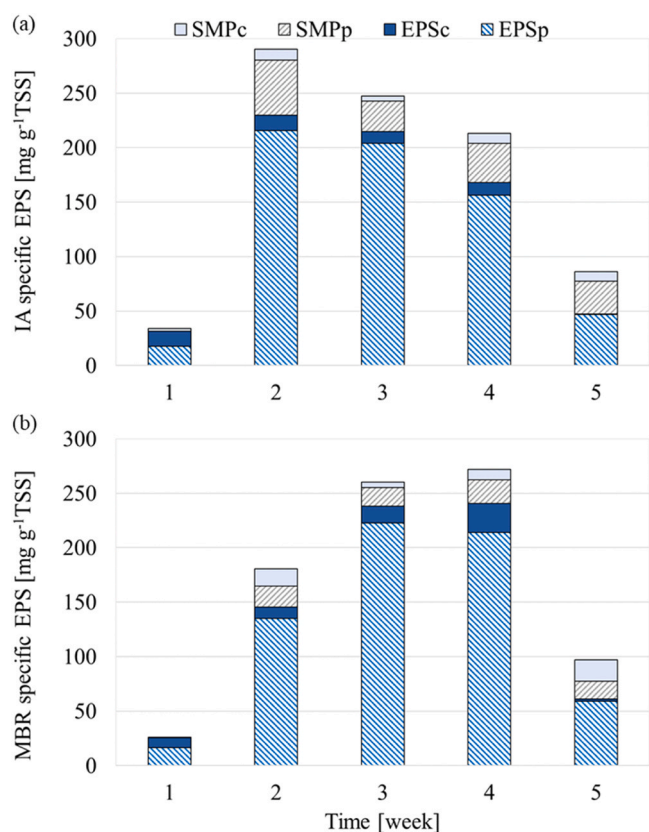


Fig. 3. Specific SMP and EPS concentration in IA reactor (a) and MBR (b).

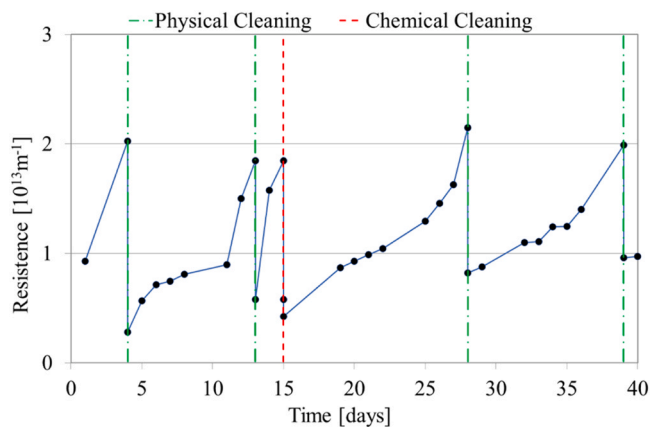


Fig. 4. Total resistance of the membrane.

et al. [14], who investigated SRTs of 10, 20 and 30 days, this result does not seem to have a negative influence on the membrane fouling. This result might be probably due to the continuous aeration in the MBR.

3.4. Greenhouse gases emission

The emission factor (a), gaseous (b) and dissolved $\text{N}_2\text{O-N}$ concentration (c) for each reactor were shown in Fig. 5. While the reactors exhibited similar average concentrations of gaseous and dissolved $\text{N}_2\text{O-N}$, it is noteworthy that the majority of the emission factor was observed in the IA reactor during aerobic conditions.

The whole pilot plant emission factor was, on average, equal to 1.73 % (± 0.83) of the influent nitrogen, which agrees with Massara et al. [35]. Indeed, Massara et al. [35] obtained a value of N_2O emission equal to 0.13–2.69 % of the influent nitrogen. Regarding the N_2O concentrations, as mentioned above, all the reactors had averaged a similar concentration: $0.18 \text{ mg N}_2\text{O-N L}^{-1}$ (± 0.06) and $0.19 \text{ mg N}_2\text{O-N L}^{-1}$ (± 0.06) for gaseous and liquid concentrations, respectively. These results are slightly lower than those of Cosenza et al. [36], who studied the same pilot plant at UNIPA but with two reactors (anoxic and aerobic) in place of the intermittent aeration reactor.

Moreover, during the experimental campaign, the pilot plant emission factor was below 2 %, except for the first operation days (Fig. 6).

3.5. Heterotrophic and autotrophic biomass kinetics

Table 3 summarised the average values of the heterotrophic and autotrophic kinetic parameters obtained during experiments. From the observation of data reported in Table 3, it can be noticed that the heterotrophic biomass showed a lower value compared to literature values for MBR systems characterized by continuous aeration [37]. This result confirms the synergistic effect of membrane configuration and intermittent aeration strategy towards reducing biomass growth ability. The heterotrophic decay rate was significantly high, thus suggesting that endogenous decay could promote the overall sludge reduction. Concerning autotrophic species, the respirometry results revealed the system's excellent development of nitrification, corroborating the good ammonium removal performance outlined above. The results were well aligned with previous values observed in MBR processes [38].

3.6. Carbon footprint

Fig. 7 shows the total emissions divided into direct, indirect and derivative.

Direct emissions are equal to $0.43 \text{ kg CO}_2 \text{ m}^{-3}$, which aligns with the literature. Mannina et al. [39] obtained values from 0.49 to $0.63 \text{ kgCO}_{2\text{eq}} \text{ m}^{-3}$. However, direct emissions represent 18.3 % of the carbon footprint in this study. This result differs from Delre et al. [40], who

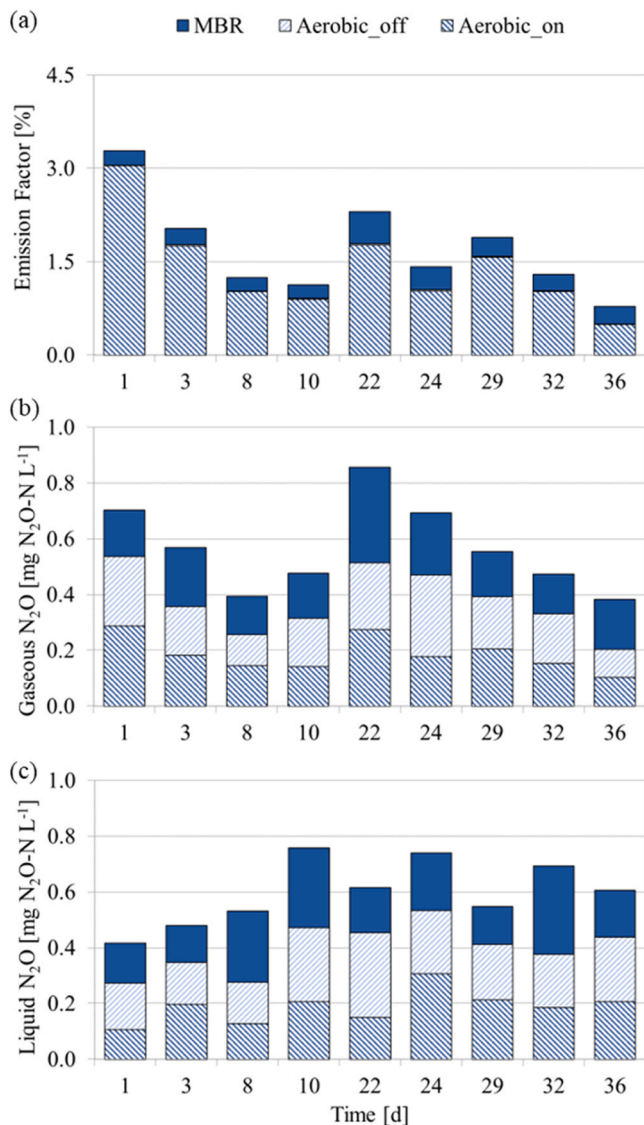


Fig. 5. Emission factor (a), Gaseous (b) and Liquid (c) N_2O concentration measured in the reactors.

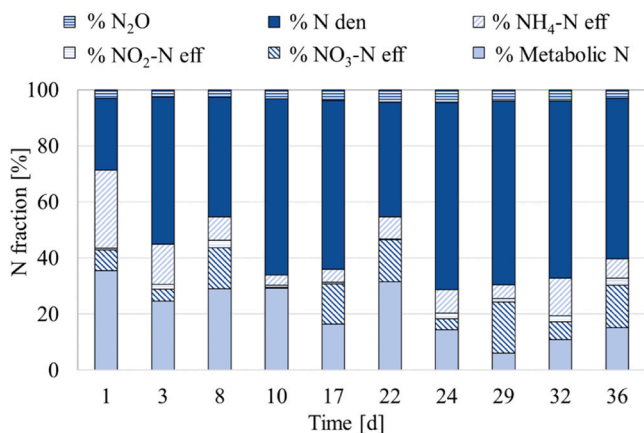


Fig. 6. Nitrogen balance.

suggested that direct emissions are between 44 % and 71 % of the total GHG emissions in a WWTP. In this article, the main contribution to direct emissions comes from $CO_{2,OrgOx}$ (equal to $0.21 \text{ kg CO}_2 \text{ m}^{-3}$),

Table 3

Summary of the main heterotrophic and autotrophic kinetic and stoichiometric parameters as average values (in brackets the standard deviation).

Parameter	Symbol	Units	Heterotrophic
Max. growth yield	Y_H	$[\text{gVSS g}^{-1}\text{COD}]$	$0.37 (\pm 0.04)$
Growth on storage	Y_{STO}	$[\text{gVSS g}^{-1}\text{COD}]$	$0.48 (\pm 0.04)$
Decay rate	b_H	$[\text{d}^{-1}]$	$1.10 (\pm 0.27)$
Max. growth rate	μ_H	$[\text{d}^{-1}]$	$1.12 (\pm 3.72)$
Max. removal rate	ν_H	$[\text{d}^{-1}]$	$3.01 (\pm 11.02)$
Net growth rate	$\mu_H - b_H$	$[\text{d}^{-1}]$	$0.01 (\pm 3.45)$
Active fraction	f_x	$[\%]$	$22.2 (\pm 12.64)$
Parameter	Symbol	Units	Autotrophic
Max. growth yield	Y_A	$[\text{gVSS g}^{-1}\text{NH}_4\text{-N}]$	$0.15 (\pm 0.03)$
Decay rate	b_A	$[\text{d}^{-1}]$	$0.15 (\pm 0.05)$
Max. growth rate	μ_A	$[\text{d}^{-1}]$	$0.17 (\pm 0.09)$
Max. removal rate	ν_A	$[\text{d}^{-1}]$	$1.17 (\pm 0.57)$
Nitrification rate	N_R	$[\text{mgNH}_4 \text{ L}^{-1} \text{ h}^{-1}]$	$4.39 (\pm 2.03)$

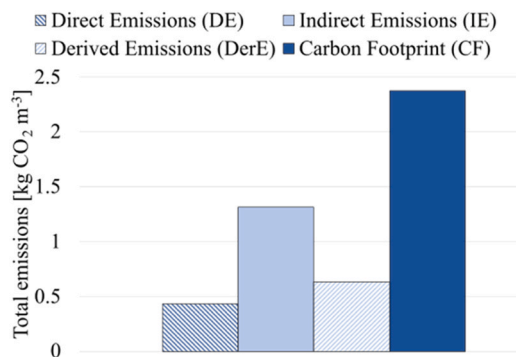


Fig. 7. Total emissions are divided into direct, indirect, and derivative.

correlated with the organic carbon oxidation in the plant. CO_2 , N_2O (equivalent CO_2 due to the N_2O emission) equals $0.18 \text{ kg CO}_{2eq} \text{ m}^{-3}$.

Derived emissions represent only 26.4 % of the carbon footprint calculated in the experimental campaign. Most of it is due to $CO_{2eq, effN_2O}$, equal to $0.59 \text{ kgCO}_{2eq} \text{ m}^{-3}$ average discharged by the plant. As mentioned above, liquid N_2O concentrations aligned with the recent literature, such as Cosenza et al. [36].

Indirect emissions represent 55.3 % of the calculated carbon footprint. This result is mainly due to the energy consumption, which equals $1.31 \text{ kg CO}_{2eq} \text{ m}^{-3}$. This value is higher with respect to the literature. Yilmaz et al. [14] obtained values from 0.24 to $0.94 \text{ kgCO}_{2eq} \text{ m}^{-3}$. $CO_{2eq, sludge}$ (the equivalent CO_2 due to the sludge treatment, transportation and landfill disposal) was equal to $0.01 \text{ kg CO}_2 \text{ m}^{-3}$. The $CO_{2,En}$ (emissions due to energy consumption) represents the main source of emissions in the plant equal to $1.29 \text{ kgCO}_{2eq} \text{ m}^{-3}$. Most energy consumption is due to mixers (in the influent tank and the IA reactor) and the recirculation pump. The energy consumption in the MBR by itself (0.41 kWh m^{-3}) was in line in comparison with Cosenza et al. [36], who obtained an energy consumption of $0.21\text{--}0.37 \text{ kWh m}^{-3}$. Moreover, Xiao et al. [41] affirmed that MBR in full-scale treatment plants is equivalent to 0.23 kWh m^{-3} .

4. Conclusion

An MBR pilot plant was monitored to evaluate the removal of carbon, nitrogen and phosphorus in the systems under intermittent aeration conditions. The study also analysed sewage sludge production and assessed the carbon footprint, encompassing direct, indirect, and derived emissions, within the MBR pilot plant. Results showed excellent removal of TCO_D (98.2 %), SCOD (86.0 %), and TN (76.4 %). The total carbon footprint of the plant was equal to $2.4 \text{ kgCO}_{2eq} \text{ m}^{-3}$. Such excellent performance then lead to low GHG production. The direct emissions only represented 18.3 % of the total carbon footprint.

Conversely, indirect emissions contributed the highest (55.3 %) to the carbon footprint, mainly due to the energy consumption. Therefore, the insertion of a mixer in the tank, where intermittent aeration is applied, has the potential to influence energy consumption, potentially offsetting the energy savings resulting from the alternation of aeration. It is crucial to emphasize that the results presented here stem from pilot plant investigations, and as such, there is the possibility of variations when compared to outcomes obtained at a full-scale level.

CRedit authorship contribution statement

Paulo Marcelo Bosco Mofatto: Writing – review & editing. **Alida Cosenza:** Writing – review & editing. **Daniele Di Trapani:** Writing – review & editing. **Lan Wu:** Writing – review & editing. **Bing-Jie Ni:** Writing – review & editing. **Giorgio Mannina:** Writing – review & editing.

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.

Acknowledgement

This work was funded by the project “Achieving wider uptake of water-smart solutions— WIDER UPTAKE” (grant agreement number: 869283) financed by the European Union’s Horizon 2020 Research and Innovation Programme, in which the author of this paper, Giorgio Mannina, is the principal investigator for the University of Palermo. The UNIPA project website can be found at: <https://wideruptake.unipa.it/>.

References

- M. Faisal, K.M. Muttaqi, D. Sutanto, A.Q. Al-Shetwi, P.J. Ker, M.A. Hannan, Control technologies of wastewater treatment plants: the state-of-the-art, current challenges, and future directions, *Renew. Sustain. Energy Rev.* 181 (2023) 113324, <https://doi.org/10.1016/j.rser.2023.113324>.
- Y. Gu, Y. Li, F. Yuan, Q. Yang, Optimization and control strategies of aeration in WWTPs: a review, *J. Clean. Prod.* 418 (2023) 138008.
- Y. Miao, L. Zhang, D. Yu, J. Zhang, W. Zhang, G. Ma, X. Zhao, Y. Peng, Application of intermittent aeration in nitrogen removal process: development, advantages and mechanisms, *Chem. Eng. J.* 430 (2022).
- J.X. Zhan, M. Ikehata, M. Mayuzumi, E. Koizumi, Y. Kawaguchi, T. Hashimoto, An aeration control strategy for oxidation ditch processes based on online oxygen requirement estimation, *Water Sci. Technol.* 68 (1) (2013) 76–82, <https://doi.org/10.2166/wst.2013.226>.
- F. Li, L. Lu, X. Zheng, H.H. Ngo, S. Liang, W. Guo, X. Zhang, Enhanced nitrogen removal in constructed wetlands: effects of dissolved oxygen and step-feeding, *Bioresour. Technol.* 169 (2014) 395–402.
- X.M. Zhan, M.G. Healy, J.P. Li, Nitrogen removal from slaughterhouse wastewater in a sequencing batch reactor under controlled low DO conditions, *Bioprocess, Biosyst. Eng.* 32 (2009) (2009) 607–614.
- H.M. Elkaramany, A.A. Elbaz, A.N. Mohamed, Alhassan H. Sakr, Study the effect of recycled pressurized air on oxygen transfer design parameters in sequencing batch reactor technology, *Water Environ. J.* 31 (2017) 90–96.
- Intergovernmental Panel on Climate Change (IPCC), *Fourth Assessment Report, Intergovernmental Panel on Climate Change (IPCC), Geneva, Switzerland* (2007).
- X.D. Hao, J. Li, M.C.M. van Loosdrecht, T.Y. Li, A sustainability-based evaluation of membrane bioreactors over conventional activated sludge processes, *J. Environ. Chem. Eng.* 6 (2) (2018) 2597–2605, <https://doi.org/10.1016/j.jece.2018.03.050>.
- T. Gao, K. Xiao, J. Zhang, W. Xue, C. Wei, X. Zhang, S. Liang, X. Wang, X. Huang, Techno-economic characteristics of wastewater treatment plants retrofitted from the conventional activated sludge process to the membrane bioreactor process, *Front. Environ. Sci. Eng.* 16 (4) (2021) 1, <https://doi.org/10.1007/s11783-021-1483-6>.
- G. Mannina, B.J. Ni, T.F. Rebouças, A. Cosenza, G. Olsson, Minimizing membrane bioreactor environmental footprint by multiple objective optimization, *Bioresour. Technol.* 302 (2020) 122824.
- O. Lorain, P.-E. Dufaye, W. Bosq, J.-M. Espenan, A new membrane bioreactor generation for wastewater treatment application: strategy of membrane aeration management by sequencing aeration cycles, *Desalination* 250 (2) (2010) 639–643, <https://doi.org/10.1016/j.desal.2009.09.040>.
- M. Yang, M. Liu, D. Yu, J. Zheng, Z. Wu, S. Zhao, J. Chang, Y. Wei, Numerical simulation of scaling-up for AEC-MBRs regarding membrane module configurations and cyclic aeration modes, *Bioresour. Technol.* 245 (2017) (2017) 933–943, <https://doi.org/10.1016/j.biortech.2017.09.027>.
- T. Yilmaz, E.K. Demir, S.T. Başaran, E.U. Çoğgör, E. Sahinkaya, Impact of aeration on/off duration on the performance of an intermittently aerated MBR treating real textile wastewater, *J. Water Process Eng.* 54 (2023) 103886.
- G. Mannina, R. Alduina, L. Badalucco, L. Barbara, F.C. Capri, A. Cosenza, D. Di Trapani, G. Gallo, V.A. Laudicina, S.M. Muscarella, D. Presti, Water resource recovery facilities (Wrrfs): the case study of Palermo university (Italy), *Water* 13 (2021) 3413 (Article number).
- G. Mannina, L. Badalucco, L. Barbara, A. Cosenza, D. Di Trapani, G. Gallo, V. A. Laudicina, G. Marino, S.M. Muscarella, D. Presti, H. Helness, Enhancing a Transition to a Circular Economy in the Water Sector: The EU Project WIDER UPTAKE, *Water* 13 (2021) 946.
- APHA, *Standard methods for the examination of water and wastewater, Standard Methods* (2012).
- D. Di Trapani, G. Di Bella, G. Mannina, M. Torregrossa, G. Viviani, Comparison between moving bed-membrane bioreactor (MB-MBR) and membrane bioreactor (MBR) systems: influence of wastewater salinity variation, *Bioresour. Technol.* 162 (2014) 60–69.
- G. Mannina, M. Capodici, A. Cosenza, P. Cinà, D. Di Trapani, A.M. Puglia, G. A. Ekama, Bacterial community structure and removal performances in IFAS-MBRs: a pilot plant case study, *J. Environ. Manag.* 198 (2017) 122–131.
- D. Gardoni, E. Ficara, D. Fornarelli, M. Parolini, R. Canziani, Long term effects of the ozonation of the sludge recycling stream on excess sludge reduction and biomass activity at full-scale, *Water Sci. Technol.* 63 (9) (2011) 2032–2038, 2011.
- G. Mannina, M. Capodici, A. Cosenza, D. Di Trapani, Nitrous oxide from integrated fixed-film activated sludge membrane bioreactor: assessing the influence of operational variables, *Bioresour. Technol.* 247 (2018) 1221–1227.
- S. Tsuneda, M. Mikami, Y. Kimochi, A. Hirata, Effect of salinity on nitrous oxide emission in the biological nitrogen removal process for industrial wastewater, *J. Hazard. Mater.* 119 (1-3) (2005) 93–98.
- R. Boiocchi, P. Viotti, D. Lancione, N. Stracqualursi, V. Torretta, M. Ragazzi, G. Ionescu, E.C. Rada, A study on the carbon footprint contributions from a large wastewater treatment plant, *Energy Rep.* 9 (2023) 274–286.
- IPCC (2022). Intergovernmental Panel on Climate Change. <https://www.ipcc.ch/>.
- European Environment Agency – EEA, 2016. Database available on line <https://www.eea.europa.eu/data>.
- Y. Zhao, Z. Yang, J. Niu, Z. Du, F. Conti, Z. Zhu, K. Yang, Y. Li, B. Zhao, H. T. Pedersen, C. Liu, E. Mutabazi, Systematical analysis of sludge treatment and disposal technologies for carbon footprint reduction, *J. Environ. Sci.* 128 (2023) 224–249.
- H.S. Kim, I.S. Seo, Y.K. Kim, J.Y. Kim, H.W. Ahn, I.S. Kim, Full-scale study on dynamic state membrane bio-reactor with modified intermittent aeration, *Desalination* 202 (2007) 99–105, <https://doi.org/10.1016/j.desal.2005.12.044>.
- B. Kim, C.S. Madukoma, J.D. Shrout, R. Nerenberg, Effect of EPS production on the performance of membrane-based biofilm reactors, *Water Res.* 240 (2023) 120101.
- B.R. Lim, K.H. Ahn, P. Songprasert, S.H. Lee, M.J. Kim, Microbial community structure in an intermittently aerated submerged membrane bioreactor treating domestic wastewater, *Desalination* 161 (2) (2004) 145–153.
- G. Mannina, M. Capodici, A. Cosenza, V.A. Laudicina, D. Di Trapani, The influence of solid retention time on IFAS-MBR systems: assessment of nitrous oxide emission, *J. Environ. Manag.* 203 (2017) 391–399.
- J. Huang, L. Xu, Y. Guo, D. Liu, S. Chen, Q. Tang, F. Peng, Intermittent aeration improving activated granular sludge granulation for nitrogen and phosphorus removal from domestic wastewater, *Bioresour. Technol. Rep.* 15 (2021) 100739.
- Z. Wang, H. Yu, J. Ma, X. Zheng, Z. Wu, Recent advances in membrane biotechnologies for sludge reduction and treatment, *Biotechnol. Adv.* 31 (8) (2013) 1187–1199.
- R. Ferrentino, M. Langone, R. Villa, G. Andreottola, Strict anaerobic side-stream reactor: effect of the sludge interchange ratio on sludge reduction in a biological nutrient removal process, *Environ. Sci. Pollut. Res.* 25 (2018) 1243–1256, <https://doi.org/10.1007/s11356-017-0448-6>.
- A. Cosenza, G. Di Bella, G. Mannina, M. Torregrossa, The role of EPS in fouling and foaming phenomena for a membrane bioreactor, *Bioresour. Technol.* 147 (2013) 184–192.
- T.M. Massara, S. Malamis, A. Guisasola, J.A. Baeza, C. Noutsopoulos, E. Katsou, A review on nitrous oxide (N₂O) emissions during biological nutrient removal from municipal wastewater and sludge reject water, *Sci. Total Environ.* 596–597 (2017) 106–123.
- A. Cosenza, H. Gulhan, G. Mannina, Trading-off greenhouse gas emissions and 741/2020 European Union water reuse legislation: an experimental MBR study, *Bioresour. Technol.* 388 (2023) 129794.
- D. Di Trapani, M. Capodici, A. Cosenza, G. Di Bella, G. Mannina, M. Torregrossa, G. Viviani, Evaluation of biomass activity and wastewater characterization in a UCT-MBR pilot plant by means of respirometric techniques, *Desalination* 269 (2011) 190–197, <https://doi.org/10.1016/j.desal.2010.10.061>.
- D. Di Trapani, G. Mannina, G. Viviani, Membrane Bioreactors for wastewater reuse: respirometric assessment of biomass activity during a two year survey, *J. Clean. Prod.* 202 (2018) 311–320, <https://doi.org/10.1016/j.jclepro.2018.08.01>.

- [39] G. Mannina, T.F. Rebouças, A. Cosenza, K. Chandran, A plant-wide wastewater treatment plant model for carbon and energy footprint: model application and scenario analysis, *J. Clean. Prod.* 217 (2019) 244–256.
- [40] A. Delre, M. ten Hoeve, C. Scheutz, Site-specific carbon footprints of Scandinavian wastewater treatment plants, using the life cycle assessment approach, *J. Clean. Prod.* 211 (2019) 1001–1014.
- [41] K. Xiao, S. Liang, X. Wang, C. Chen, X. Huang, Current state and challenges of full-scale membrane bioreactor applications: a critical review, *Bioresour. Technol.* 271 (2019) 473–481.