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Startup and initial operation of an MLE-MABR treating municipal wastewater

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ABSTRACT

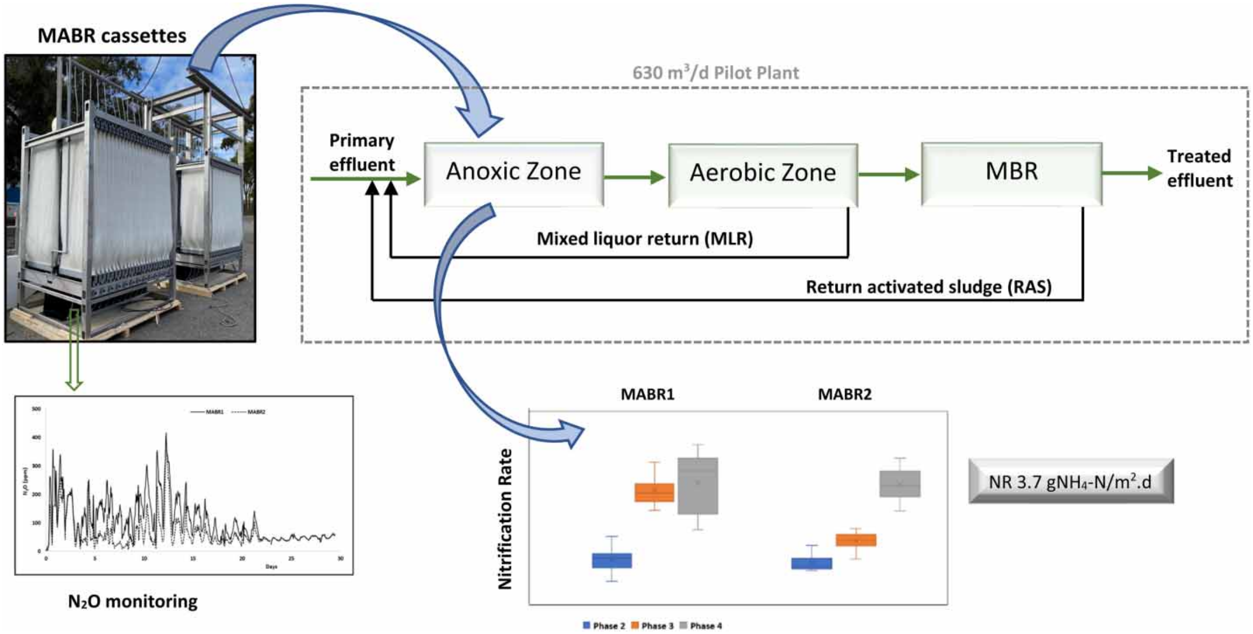
A 630 m^3/d pilot plant was installed at Subiaco WRRF to determine design and operational parameters of a hybrid Modified Ludzack-Ettinger – Membrane Aerated Biofilm Reactor (MLE-MABR) configuration. Two commercial ZeeLung MABR cassettes were installed in series in the anoxic zone and the pilot was fed with primary effluent (averaging COD 601 mg/L, TKN 68.5 mg/L and 17–29 °C). A nitrifying biofilm was developed within 3 weeks and the nitrous oxide (N_2O) gas emissions from the MABR exhaust gas proved to be a reliable parameter to assess biofilm development. Both MABRs achieved the average nitrification rate (NR) of 3.7 gNH_4-N/m^2.d when air flow was 8.6 and 11.2 Nm^3/h to MABR1 and MABR2 respectively, which reached a maximum oxygen transfer rate of 17.4 gO_2/m^2.d. Biofilm thickness was controlled via air scouring and intermittent coarse bubble mixing (90 s on/90 s off). This paper discusses the startup strategy, minimum requirements for process monitoring, impact of different air flow conditions, ORP and mixing patterns on performance efficiency over a 22-week period.

Key words: MABR, N_2O emission, nitrification rate, startup

HIGHLIGHTS

- Nitrifying biofilm is developed in 3 weeks in an MABR.
- N_2O emissions from MABR exhaust gas is a reliable parameter for start-up monitoring.
- Monitoring of O_2% in the MABR exhaust gas coupled with ammonia concentration in the wastewater are satisfactory parameters for process performance evaluation.
- It appears possible to install MABR technology in the anoxic zone of an MLE process with no major process changes.

GRAPHICAL ABSTRACT



INTRODUCTION

MABR is a novel technology which enables process intensification by increasing the nitrifying biomass inventory. The nitrogen removal capacity of a plant improves with the unique benefit of enhancing simultaneous nitrification and denitrification in a single reactor (Hu *et al.* 2009; Houweling & Daigger 2019). A fundamental characteristic of MABRs is the counter-diffusional biofilm process where air is supplied via a hollow-fiber gas-permeable membrane and oxygen diffuses throughout the biofilm while the substrate from the bulk liquid diffuses into the biofilm (Nerenberg 2016). The technology was first commercialized in 2015 (Uri-Carreño *et al.* 2018) and both pilot and full-scale installations have been increasing recently, contributing to the understanding of its challenges and opportunities (He *et al.* 2021).

However, the majority of available design data to date has been obtained from facilities located in cool or temperate regions (Underwood *et al.* 2018; Guglielmi *et al.* 2020; Elsayed *et al.* 2021; Uri-Carreño *et al.* 2021), and not from Mediterranean climates as experienced in Western Australia. It has been demonstrated that MABRs are unaffected by low temperatures (Uri-Carreño *et al.* 2021) but further investigation is required for operation in higher temperature climatic zones. Németh *et al.* (2021) have demonstrated that NR improved from 3.1 to 5.5 g N/m^2.d when temperature increased from 8 to 30 ^{o}C in a lab scale MABR. Temperature affects diffusivity constants, oxygen solubility, membrane permeability, biomass growth rate and, consequently the biofilm thickness. Excessive biofilm thickness raises concern for mass transfer and membrane clogging, which is a frequent operational problem (He *et al.* 2021). Another factor that affects biofilm characteristics is the availability of biodegradable soluble carbon as it promotes the potentially excessive growth of heterotrophs, which may be detrimental to the nitrification process. However, there is inconclusive information from literature to date. A previous study in 2008 (Downing & Nerenberg) predicted that nitrification in the MABR was less sensitive to BOD loadings. They demonstrated that nitrifying bacteria grew preferentially in the deeper aerobic portion of the biofilm while heterotrophs grew in the outer layers. Ten years later, it was noted that soluble COD (CODs) can have an inhibitory effect on nitrification by increasing biofilm thickness and the resistance to ammonia transfer (Nerenberg 2018). More recently, in 2021, a study demonstrated that the carbon-nitrogen ratio (C/N) did not affect the NR in MABR treating municipal wastewater and even thick liquid films created by insufficient mixing conditions hardly disturbed it (Elsayed *et al.* 2021). According to the authors, the ammonia transport was sufficiently fast in both the liquid film and biofilm. Despite the controversial research findings, all concur that biofilm thickness control is crucial. In commercial MABRs, managing biofilm thickness is achieved by scouring air bubbles on the surface of the biofilm to detach excessive growth (Peeters *et al.* 2017). Optimal biofilm thickness depends on the pressure of the air supply to the membrane and the bulk concentration of the substrate to be oxidised (Nerenberg 2018). Therefore, the frequency and intensity of scouring requires optimisation for each specific system. On one hand, excessive scouring promotes excessive biofilm detachment and consequent biomass washout. On the other, ineffective scouring encourages membrane bridging issues with the associated decrease of biofilm surface area (He *et al.* 2021).

It is expected that thinner biofilms develop in hybrid processes as the suspended biomass consumes the soluble carbon and minimizes the growth of heterotrophs in the biofilm (Uri-Carreño *et al.* 2018). From this perspective, the MLE-MABR configuration is a promising application where the majority of soluble carbon is consumed in the early stages of the treatment process. However, limited data available from full-scale plants hinder proper design and much of the understanding is based on simulations (Carlson *et al.* 2021). Modelling studies show that a hybrid MLE-MABR can maintain nitrogen removal similar to an MLE process, but at significantly lower sludge retention time (SRT), and with the additional benefit of reduced process aeration demand.

The monitoring of ammonia removal in the biofilm is fundamental to evaluate the process performance. At full-scale installations, NR can be evaluated by either measuring the ammonia concentration in the bulk liquid or by analyzing the composition of the membrane exhaust air (Houweling & Daigger 2019). The percentage of oxygen transferred to the biofilm gives an estimate of ammonia oxidation, assuming that nitrifying bacteria have grown predominantly in the biofilm. The monitoring of N_2O in the exhaust air is another alternative, as it is a byproduct from the multiple reactions occurring during nitrification and denitrification. Environmental factors such as DO concentration, nitrite accumulation, C/N ratio and sudden changes in process conditions can affect N_2O emissions (Guo *et al.* 2018). Nevertheless, it is probable that N_2O production depends primarily on the activity of the nitrifying bacteria and is triggered by high ammonia concentration (Valkova *et al.* 2021). In the MABR, N_2O is produced closer to the biofilm-membrane interface; that is, where the oxygen availability is higher, and it is captured in the external layers, furthest from the biofilm-membrane interface (Kinh *et al.* 2017). The membrane exhaust air carries the excess of N_2O not captured in the biofilm, therefore enabling the identification of process imbalances.

This research aims to obtain design and operational parameters for full-scale implementation of MLE-MABR hybrid system treating high strength domestic wastewater under high seasonal temperatures, ranging from 17 to 29 °C. The main objectives were to validate: (i) the MABR startup procedure, (ii) the viability of installing the MABR in the full-scale plant maintaining the existing MLE configuration, (iii) the achievable NR of the MABR and (iv) the effective monitoring of process performance.

MATERIALS AND METHODS

The pilot plant

Subiaco WRRF is located in Perth, Western Australia, with average inflow of 60,000 m^3/d. Following the examination of numerous secondary treatment upgrade options, MABR technology was selected. This was because significant process intensification (about 89% increase in capacity) could be achieved via retrofit to the existing MLE process volume. This assessment was based on design parameters available from two full-scale plants operating on weak influent and lower Northern Hemisphere temperatures. The absence of design parameters for higher strength wastewaters and temperatures averaging 27 °C in summer necessitated the construction of a pilot plant, which was commissioned in July 2020.

The pilot plant has a design capacity of 630 m^3/d and is configured in a modular format (Figure 1). Tank 1 is a bioselector and during this study was bypassed. All of the other 24 m^3 tanks may also be bypassed to test different operational arrangements. Tanks 2 and 3 are designed as anoxic zones and house a commercial MABR cassette each. The membrane surface area of each cassette is 1,920 m^2 with a packing density of 80 m^2/m^3. Air is supplied to the membranes at 45 to 55 kPa and flow rate can be adjusted from 7.0 to 11.2 Nm^3/h. Scouring air is provided intermittently by the expelled exhaust air collected in the bottom channel of the cassette. The MABR tanks also receive intermittent aeration through coarse bubble diffusers using air drawn from the fine bubble aeration to ensure proper mixing. Frequency was adjusted during the experimental period and is discussed later.

Tank 4 is equipped with both diffusers and a mixer, which allowed operation under anoxic or aerobic conditions and during this study it was maintained aerobic. Tanks 5 to 7 are aerobic and Tank 8 is a UF membrane bioreactor (MBR). In this study, mixed liquor (MLR) was recycled from Tank 7 to Tank 2 and activated sludge (RAS) returned from Tank 8 to Tank 2, mimicking the Subiaco WRRF MLE process. The treated water is temporarily stored in Tank 9. There are independent aeration systems for the MABR, mixing/aeration and MBR.

Flowmeters have been installed to monitor inflow, RAS, MLR and waste activated sludge. Oxidation-Reduction Potential (ORP) is monitored (SensoLyt 700 IQ, Xylem) in the MABRs and dissolved oxygen (DO) is controlled (FDO 70x IQ, Xylem)

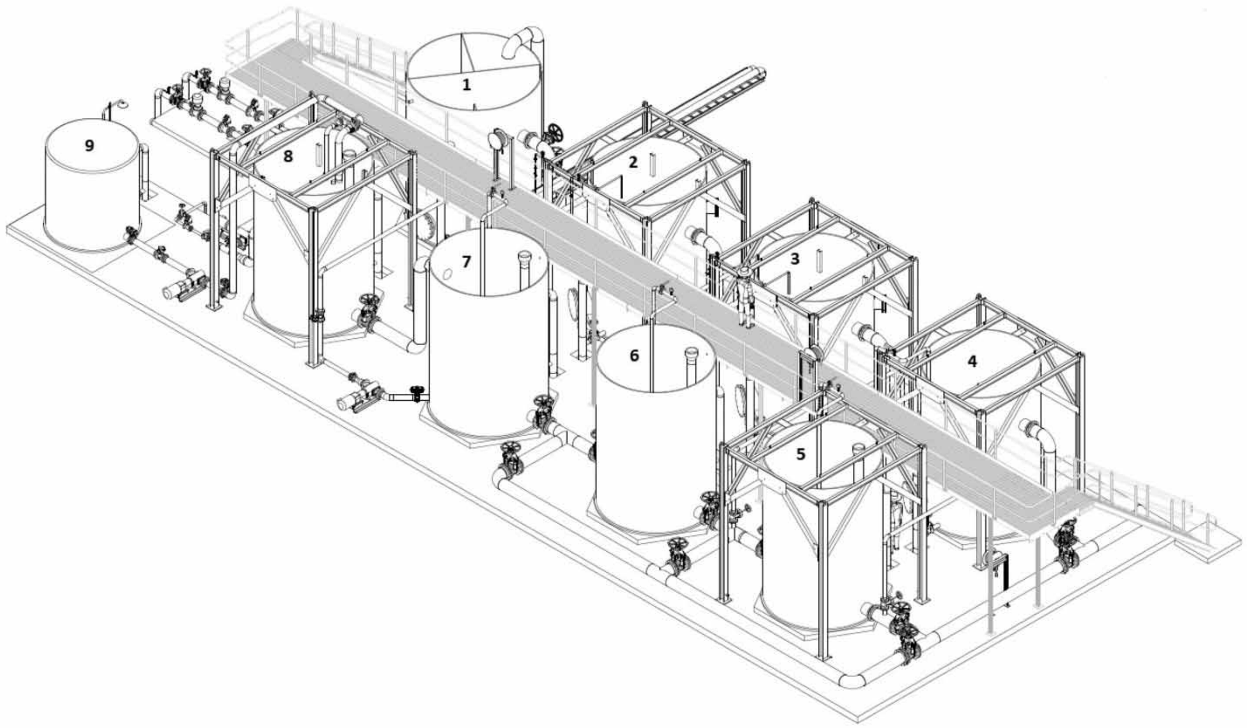


Figure 1 | Pilot plant overview. (1) Bioselector, (2) MABR1, (3) MABR2, (4) Anoxic/aerobic tank, (5) Aerobic tank 1, (6) Aerobic tank 2, (7) Aerobic tank 3, (8) MBR, (9) Treated water tank.

in the aerobic tanks. Ammonia and nitrate levels are continuously measured with ion selective electrode sensors (Varion 700 IQ, Xylem) in the primary effluent inflow, Tanks 2, 3, 7 and 9. COD of primary effluent is also monitored online with a photometric sensor (CarboVis 70x IQ, Xylem). The oxygen percentage and nitrous oxide emissions from the MABR exhaust gas has online continuous monitoring (SmartCEMS, AquaGas). Flowmeters and online sensors are connected to the SCADA system. To validate and calibrate the online monitoring, grab samples were collected three times a week. The parameters nitrite, phosphate, VFA, COD_s, BOD and BOD_s were also analysed.

MABR efficiency

Treatment efficiency was evaluated in terms of ammonia removal, oxygen transfer efficiency (OTE) and oxygen uptake by the biofilm. The NR achieved in the biofilm was calculated in terms of media surface (Equation (1)) where Q_{in} is the feed flow and comprises the primary effluent, the RAS and MLR. NH_4-N is the ammonia concentration in the bulk liquid entering and leaving the MABR tank.

$$NR = \frac{Q_{in} * (NH_4N_{in} - NH_4N_{out})}{1,920\ m^2} \tag{1}$$

The oxygen transfer rate (OTR) is a function of both membrane characteristics, operational parameters (Cote *et al.* 2015) and biofilm activity. It was calculated as per Equation (2) and OTE was calculated using Equations (3) and (4). Q_{air} is the air flow to the MABR and $O_2\%_{exhaust}$ is the percentage of oxygen in the exhaust gas of the MABR.

$$OTR = OTE \frac{32 \frac{g\ O_2}{mol}}{22.414 \frac{L}{mol}} \frac{\left[Q_{air}\left(20.9\% \frac{mol\ O_2}{mol\ air}\right)\right]}{1,920\ m^2} \tag{2}$$

$$OTE = \frac{20.9\% - O2\%_{exhaust} * F_v}{20.9\%} \tag{3}$$

$$F_v = \frac{1 - 20.9\%}{1 - O2\%_{exhaust}} \tag{4}$$

Research approach

This study was performed over a period of almost 6 months. The pilot plant was operated in steady state mode with constant feed flow of 630 m^3/d and a SRT target of 4 days. However, different conditions shaped the operation (Table 1). The startup was concluded in 30 days, but an additional 20 days were required to adjust sensors and aeration control (Phase 1). Subsequently, unexpected problems related to the mixing/aeration system reduced the DO to low levels (Phase 2) and another 15 days were required until the process stabilized, with DO around 1.5 mg/L in the aerobic tanks (Phase 3). The air flow to each MABR cassette was 8.6 Nm^3/h and the intermittent mixing of the MABRs bulk liquid was adjusted to prevent sludge settling. These conditions were maintained until suspected sludge settling/accumulation necessitated more intense mixing (Phase 4). At the same time, the air flow to the MABRs was increased. To compare the results from online monitoring of phases 2 to 4, Welch's t-test (95% confidence) was applied.

RESULTS AND DISCUSSION

Startup

The pilot plant was seeded with nitrifying sludge from Subiaco WRRF (SRT of 9 days) and the feed flow was increased gradually, from 300 to 630 m^3/d. Correspondingly, RAS was recirculated from 390 to 630 m^3/d and MLR was adjusted to 380 m^3/d. The temperature of the wastewater averaged 21.7 °C at the end of the startup phase and represented the minimum operational temperature during this investigation period.

The main difference between the two MABRs concerns the ratio BODs/NH_4-N in the feed water. MABR1 receives the highest ammonia concentration, which benefits the development of the nitrifying biofilm. However, MABR1 also receives the highest BODs and high VFA (Table 2), which potentially contributes to the growth of heterotrophs and excessive increase in biofilm thickness. It appears that the MLE configuration protected MABR1 from excessive concentration of soluble organic matter since it was mostly consumed by the returning sludges for denitrification. According to Daigger (2014), denitrification requires a C/N ratio greater than 3.5 to 4.0 mg of biodegradable COD per mg of ammonia biodegraded via nitrification-denitrification. During the course of this study. BODs was evaluated from grab samples and the ratio BODs/NH_4-N averaged 3.93 (0.25) in the feed of MABR1 and 0.69 (0.23) in MABR2, indicating that the likelihood of heterotrophs outcompeting nitrifiers for DO in MABR2 was low.

The air flow was initially supplied to each MABR at 7.0 m^3/h and the oxygen in the exhaust gas averaged 15.3 and 14.8% at MABR1 and MABR2 respectively. Figure 2(a) shows the trends from the online monitoring of oxygen in the exhaust gas. The inverted peaks tending to zero indicate the periods when aeration was intentionally switched off for biomass selection; that is, controlling of protozoa and red worms as they are obligate aerobes.

Table 1 | Operational conditions during the study period

Phase	Timeline	Inflow (m^3/d)	Ammonia monitoring	Average of aerobic tanks DO (mg/L)	Air flow per cassette (Nm^3/h)	Mixing frequency of MABR bulk liquid
Startup	Day 0 to 29	300–630	Grab samples	5.0	7.0 to 8.6	Mixing 60 s in 300 s cycles
1	Day 30 to day 48	630		2.1	8.6	
2	Day 49 to 64		Online sensors	0.7		
3	Day 65 to 130			1.5		
4	Day 131 to day 174				11.2	Mixing 90 s in 180 s cycles

Table 2 | Average (St. Dev) of historical plant data of primary effluent 24 h composite samples

BOD	BODs	COD	CODs (1.2 μm)	VFA	TKN	NH_4	SS	TP	Alk
286.2 (32.8)	164.2 (9.0)	601.0 (83.2)	328.1 (15.6)	120.2 (32.5)	68.5 (4.8)	52.2 (3.1)	237.5 (50.1)	10.8 (1.4)	341.5 (44.2)

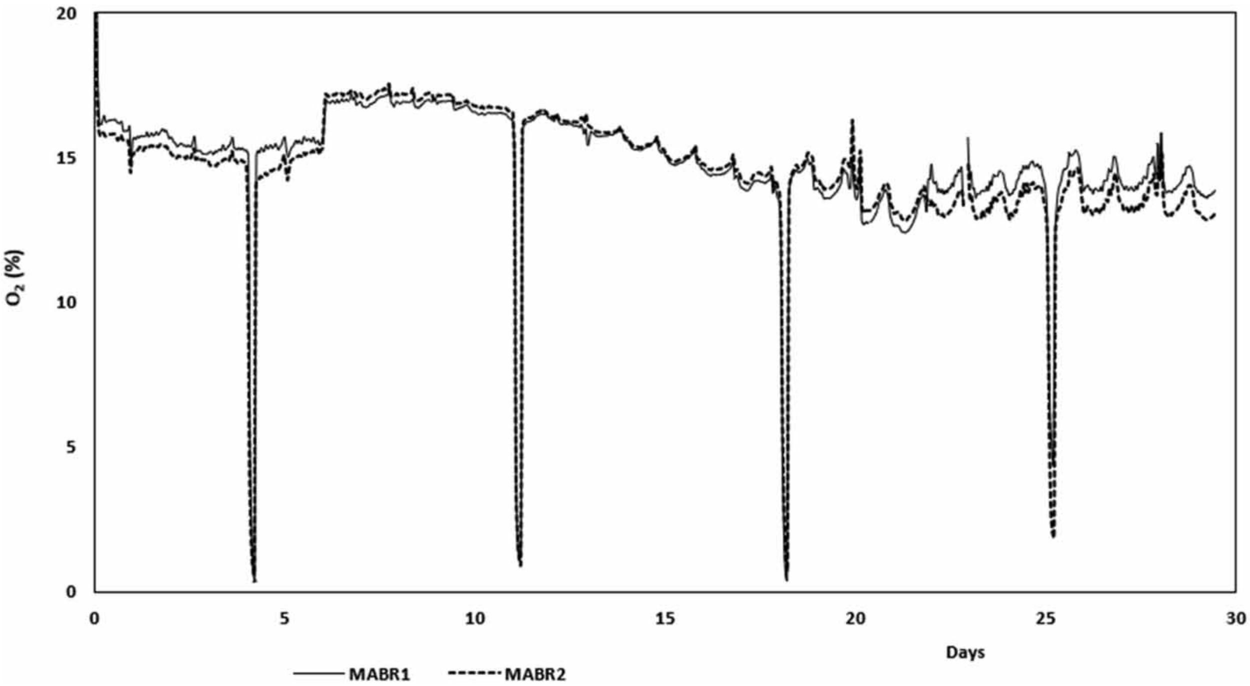


Figure 2 | Oxygen percentage in the exhaust gas of MABR1 and MABR2.

On day 6, the air flow was adjusted to 8.6 m^3/h and this changing of condition was evident in the exhaust gas. The fraction of oxygen increased and, at the same time, became more comparable: 16.9 and 17.2% in MABR1 and MABR2, respectively. After a few days, the exhaust gas of both MABRs contained roughly the same oxygen percentage. This increase in oxygen percentage suggested that an excess of air was supplied, however, since the biofilm was under development, by raising the air flow oxygen limiting conditions were avoided. As explained by Houweling & Daigger (2019), the availability of oxygen to the biofilm is proportional to the partial pressure of the oxygen in the lumen of the membrane and also to the respiration rate of the biofilm. The biofilm amplifies the oxygen diffusion across the membrane.

From day 6 to day 21, the oxygen demand by the biofilm increased progressively and the percentage in the exhaust gas reached minimum values of 12.4% in MABR1 and 12.9% in MABR2. From this point, a shift was observed in MABR1. The oxygen in the offgas increased to 14.8% while no significant changes were observed in MABR2. However, a shift in both MABRs was confirmed by N_2O emissions (Figure 3). Readings oscillated from 20 to more than 400 ppm (v/v) but on day 21 it stabilized around 50 ppm.

Since N_2O is predominantly a product from nitrification and an intermediary of denitrification, it is plausible that the great variability observed was due to establishment of the biofilm. It is expected that the increasing of N_2O occurs during process imbalances such as ammonia shock loading or oxygen deficiency. In the case of MABR, oxygen deficiency during the formation of the biofilm was unlikely and hence N_2O production could be related exclusively to the ammonia loading.

An MABR biofilm can hold a consortium of both nitrifiers and denitrifiers, depending on process conditions. In this trial, the anoxic environment of the bulk liquid appears to have encouraged the grow of denitrifiers. Therefore, the decrease of N_2O in the exhaust gas could also be attributed to the development of the denitrifying layer. The surface of an aerated membrane is an ideal environment for nitrifiers forming a biofilm, but the establishment of denitrifiers will depend on the biofilm thickness and the formation of anoxic layers.

In the present study, the emissions were erratic during the first days. On day 11, both inflow to the plant and RAS increased from 300 to 450 m^3/d and coincided with an increase in the N_2O emission. The increasing inflow not only increased the ammonia load but also increased the mixing in the tank. In fact, the ammonia concentration in the bulk liquid did not change after the increasing of inflow and it is known that biofilms respond to the bulk ammonia concentration, not to the load (Houweling & Daigger 2019). Thus, the increasing of turbulence might have improved the ammonia diffusion into the biofilm. The importance of mass transfer limitations and hydrodynamic conditions for nitrifying MABRs was stressed by Castrillo *et al.* (2019) as solutes must diffuse from the bulk liquid to the biofilm.

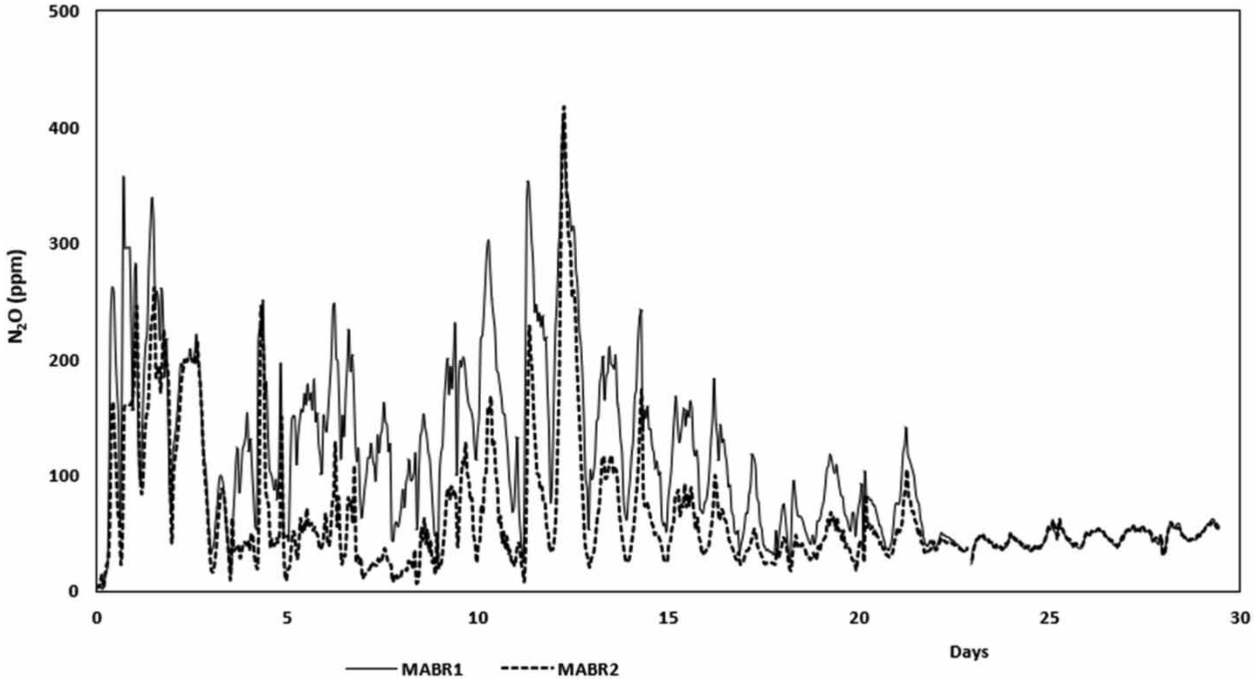


Figure 3 | Nitrous oxide emissions in the exhaust gas of MABR1 and MABR2.

On days 12 and 13 the N_2O emissions peaked, after which there was a continuous and permanent decline. Further inflow increases had no effect, which implies that the maximum ammonia diffusion was already achieved. Note that until the stabilization, MABR1 presented higher N_2O emissions than MABR2. This may be explained by the high ORP detected in the bulk liquid of MABR1, around −100 mV, which might have negatively impacted the development of the denitrifying layer. The high ORP was a consequence of the return of over-aerated sludge to MABR1 as the aeration system for mixing/aeration was being adjusted.

The present study measured only the emissions of gases that were able to back-diffuse through the biofilm and be released with the exhausted air from the membrane lumen. Gases that were eventually released in the opposite direction; that is, into the bulk liquid, were neither captured nor measured. It is planned to measure the N_2O emissions from the liquid phase as part of future studies.

From this perspective, it was possible to assume that the biofilm was completely established in both MABRs after approximately 3 weeks, in agreement with previous studies. A lab-scale gas-permeable membrane seeded with activated sludge from a municipal wastewater plant had the nitrification completely established within 20 days (Semmens *et al.* 2003). The authors used synthetic wastewater with ammonia concentration of 49 mg/L and COD of 218 mg/L during the biofilm formation. A later study, applying a full-scale MABR cassette to the treatment of municipal wastewater, also confirmed the establishment of a nitrifying biofilm in a 3-week period (Uri-Carreño *et al.* 2021).

Since the MABR cassettes were housed in anoxic tanks, no oxidation of ammonia was expected from the mixed liquor in those tanks. Therefore, the ammonia concentration measured in the bulk liquid associated to the oxygen transferred from the membrane can also validate the nitrifying activity in the biofilm. During the startup the NR was estimated from grab samples and ranged from 1.4 to 5.5 g NH_4-N/m^2.d

Steady state operation

The steady state phase progressed for the next 5 months and had the objective of producing baseline evidence of achievable NR. The plant was fed with constant inflow and the variation in the ammonia load were related exclusively to the changes in ammonia concentration during the day. The average concentration in the primary effluent was 54.5 (5.7) mg NH_4-N/L which represents an ammonia load of at least 17.9 g NH_4-N/m^2.d as any non-oxidized ammonia from tank 7 returns in the MLR. Both RAS and MLR contributed to an additional 160% of the primary effluent inflow, making the hydraulic residence time in each MABR tank only 20 min and 2.4 h in the entire treatment train.

Phase 1 lasted for 18 days and ammonia was monitored from grab samples. NR ranged from 4.2 to 5.5 gNH_4-N/m^2.d in MABR1 and from 0.8 to 3.4 gNH_4-N/m^2.d in MABR2. These results are not representative of the daily average of NR; however, they are comparable to a previous pilot investigation with an identical membrane (Nerenberg 2018). Attainable NRs were 1.4 and 3.8 g N/m^2.d for bulk ammonia concentration of 1.8 and 18 mg/L, respectively. No changes were observed in both O_2 and N_2O emissions in the exhaust gas during this period (Table 3), an indicative of a stable process.

Ammonia sensors were installed at the end of phase 1 and NR was evaluated as daily average. As previously mentioned, the challenges associated with the regulation of the aeration system of the aerobic tanks shaped different conditions in the MABR bulk liquid. The aerobic reactors were initially overaerated which caused high ORP in MABR1 bulk liquid. Subsequently, a second adjustment of the blower created a deficit of aeration and MABR1 bulk liquid changed to an anaerobic environment; ORP averaged −395 mV. The oxygen uptake by the biofilm increased in MABR1 however the daily averaged NR was 1.37 gNH_4-N/m^2.d. It is difficult to compare with the results from the previous phase since the monitoring method changed but the maximum NR achieved in phase 2 was only 2.08 gNH_4-N/m^2.d which is significantly lower than the values obtained from grab samples during phase 1. This means that the oxygen was consumed by other bacteria than nitrifiers. From the other side, MABR2 remained stable regarding oxygen consumption and the NR averaged 1.31 gNH_4-N/m^2.d, which was in the range of results obtained from grab samples during phase 1. Considering the theoretical oxygen demand for nitrification of 4.57 gO_2/gNH_4-N, the ratio OTR/4.57 can be compared with NR to evaluate whether oxygen is primarily used by nitrifiers or is being consumed by heterotrophs. During phase 2, the OTR/4.57 was 3.28 in MABR1 while the NR was 1.37 gNH_4-N/m^2.d. This means that only 42% of the oxygen was used for nitrification and a similar condition was found in MABR2.

Phase 2 lasted for two weeks and it was characterized by the lowest NR achieved during this study. The key difference was the ORP and it suggests that the anaerobic environment in the bulk liquid might have favoured reactions other than nitrification. The reasons are not clear and demand further investigation; however, a similar condition was described by Uri-Carreño *et al.* (2021) when ORP of less than −300 mV created problems with nitrification to a point that ORP required controlling.

Finally, during phase 3, a new adjustment of the aeration system was able to maintain the DO at about 1.5 mg/L in the aeration tanks and the ORP averaged −235 mV in MABR1; an ideal anoxic environment. This phase extended for 65 days and the benefits of the redox adjustment became evident. The oxygen in the exhaust gas decreased to 11% and N_2O emissions increased to 115.7 ppm in MABR1, an indication of higher activity of the nitrifiers in the biofilm. This could also be verified by the ratio OTR/4.57 of 3.72; almost 100% of the oxygen was used for nitrification. In MABR2, 62% of the oxygen was used for nitrification. The Welch's t-test result of the comparison between NR from phases 2 and 3 proved that the differences were significant in both MABRs but there was higher improvement in MABR1. For a better understanding, the boxplots provide a summary of the NR behaviour during phases 2 to 4, when ammonia was monitored online (Figure 4).

The main improvement in MABR2 occurred in phase 4, when the mixing frequency of the bulk liquid was drastically changed and the average NR increased to 3.72 gNH_4-N/m^2.d. Reducing short-circuits and improving flow patterns seems to be essential in MABR tanks, in agreement with Castrillo *et al.* (2019). The authors obtained improvements of up to 69% in the volumetric NR by enhancing reactor hydrodynamics. The increasing of the air pressure in the membrane lumen, which caused the air flow to increase from 8.6 to 11.0 Nm^3/h, also contributed to the NR enhancement in MABR2. The ratio OTR/4.57 of 3.52 indicated that 100% of the oxygen was used for nitrification.

When comparing the hourly averaged NR in MABR2 during phases 3 and 4 to the ammonia loading profile (Figure 5), the change of response of the biofilm to new conditions is clear. During phase 3, the NR profile was flatter and less sensitive to changes in the ammonia concentration, typically when the biofilm was either oxygen or biomass-limited. However, during

Table 3 | Daily average (St. dev) from online monitoring of MABR bulk liquid and exhausting gas

		MABR1					MABR2				
Phase	T (°C)	ORP (mV)	O_2 %	N_2O (ppm)	OTR ($g/m^2.d$)	OTE %	ORP (mV)	O_2 %	N_2O (ppm)	OTR ($g/m^2.d$)	OTE %
1	22.5	−274 (123)	14.0 (0.5)	62.6 (14.8)	12.4 (0.8)	38.5 (2.5)	−169 (73)	13.3 (0.4)	62.2 (14.8)	13.4 (0.7)	41.9 (2.1)
2	22.9	−395 (65)	12.3 (0.5)	81.2 (38.9)	15.0 (0.8)	46.7 (2.4)	−221 (31)	13.1 (0.4)	70.0 (26.0)	13.8 (0.7)	42.9 (2.2)
3	23.7	−235 (66)	11.0 (1.0)	115.7 (26.2)	17.0 (1.5)	53.0 (4.8)	−191 (32)	12.7 (0.9)	96.2 (22.7)	14.5 (1.4)	45.1 (4.5)
4	28.3	−214 (89)	13.2 (0.9)	98.3 (13.8)	17.4 (1.8)	42.4 (4.4)	−198 (57)	13.8 (1.0)	94.0 (12.4)	16.1 (2.0)	39.3 (5.0)

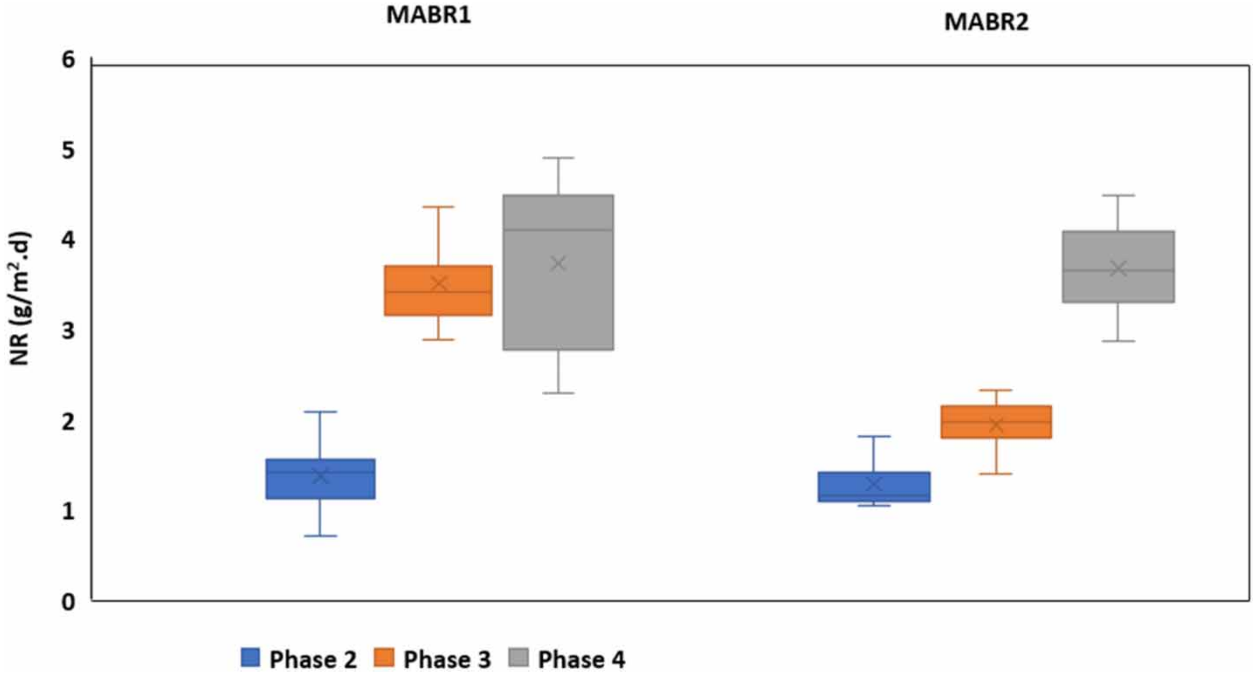


Figure 4 | Boxplots of NR based on hourly averaged online ammonia monitoring.

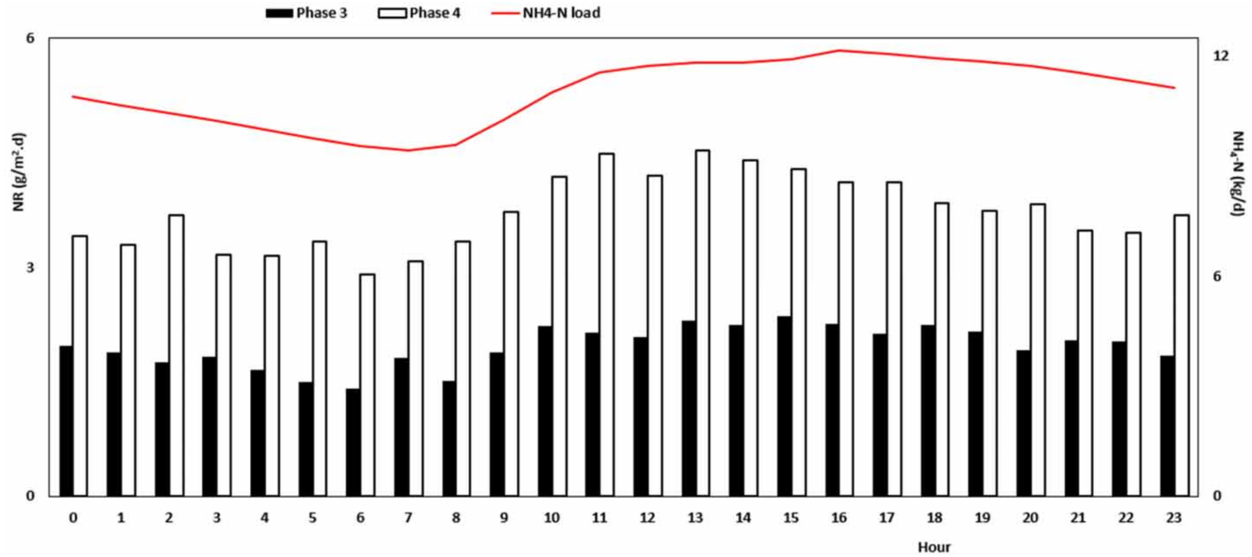


Figure 5 | Hourly averaged of NR in MABR2 during phases 3 and 4 and ammonia load.

phase 4 the NR increased with increased loading, an indication that biofilm had converted to ammonia-limited condition, the expected condition in MABRs. This also explains the substantial increase of NR in phase 4; the ammonia diffusion into the biofilm should have increased. The mixing not only reduced the short-circuits, it might have improved the scouring and promoted the control of biofilm thickness, bringing it to an optimal condition. In biofilms, the nitrification is impacted by the diffusivity of both ammonia and oxygen and fluxes in MABR are extremely dependent on the biofilm thickness. If the thickness is low, NR is biomass limited. If the thickness is excessive, NR is mass transfer limited and from this perspective there always is an optimal thickness (Nerenberg 2018). In addition, the increased air pressure in the lumen may have increased the aerobic layer.

However, there was no significant difference in average NR in MABR1 during phase 4. As previously mentioned, nearly 100% of the oxygen was already used for nitrification and either the air flow or the mixing increasing did not impact the average NR. However, the amplitude between maximum and minimum NR enlarged. A negative impact was observed when ammonia load was minimum, and a positive impact was observed during peak load. It suggests that the biofilm had developed beyond an optimal thickness and when ammonia load was minimum the diffusion into the biofilm was limited. On the other hand, during ammonia peak loads the diffusion increased and the excess of air flow was used for nitrification, increasing the maximum NR achieved. From this perspective, the increasing of air flow was not justifiable for MABR1 during the periods when ammonia concentration is low in the primary effluent; that is, during night-time. This fact can be confirmed by analysing the correlation between OTR and NH_4-N in the bulk liquid (Figure 6). The OTR increased notably from phase 2 to phase 3 and it can be explained by the increased activity of the biofilm since the air flow was identical. This fact also helps proving that the microbial activity in the biofilm augment the oxygen transfer. However, during phase 4 the OTR only increased significantly for higher ammonia concentrations. Similar behaviour was not observed in MABR2 and the best OTR was achieved during phase 4. In this study the OTR ranged from 13.8 to 19.8 in MABR1 and from 12.9 to 19.1 g O_2/m_2.d in MABR2. These values are higher than the ranges mentioned in the literature for municipal wastewater (Uri-Carreño *et al.* 2021).

The excessive air flow caused the increasing of oxygen in the offgas of both MABRs and a negative impact in the OTE was observed, as shown in Table 3. The change of the membrane air pressure changed the relationship between OTR and OTE in both MABRs. From the other side, the N_2O emissions decreased significantly in both MABRs. This fact might be attributed to either the growth of a denitrifying layer or to a reduction of gas back-diffusion due to the higher air pressure in the membrane lumen. A previous study suggested that intra-membrane pressure might be the rate controlling factor for the treatment of high-strength wastewaters and the air pressure must be well-adjusted to sustain the anoxic/anaerobic zone at the biofilm-liquid interface if both nitrification and denitrification are required (Syron & Casey 2008).

Another factor to be further investigated is the temperature as it influences the diffusivity of ammonia and oxygen into the biofilm and affects the bioreaction kinetics. Elsayed *et al.* (2021) reported a seasonal dependency of NR in the MABR and peak was achieved at the end of the summer when water temperature reached approximately 20 °C. During this study, the main temperature change occurred from phase 3 to 4 and the bulk liquid averaged 28.3 °C, which is far higher than the typical temperature found in previous researches. Coincidentally, the highest NR achieved in MABR2 occurred in phase 4; however, other process adjustments happened simultaneously. Thus, any change of performance cannot be attributed exclusively to temperature. At the same time, no significant improvement was observed in MABR1 during phase 4, suggesting that NR in the biofilm is independent of the temperature.

Nitrite and nitrate were not detected in the MABR tanks during this entire investigation, demonstrating that denitrification was taking place simultaneously either in the biofilm or in the mixed liquor. In this study, most of the denitrification occurred in MABR1 tank with the uptake of 87% of the BODs (Table 4 and supplementary material). Part of the VFAs and BODs

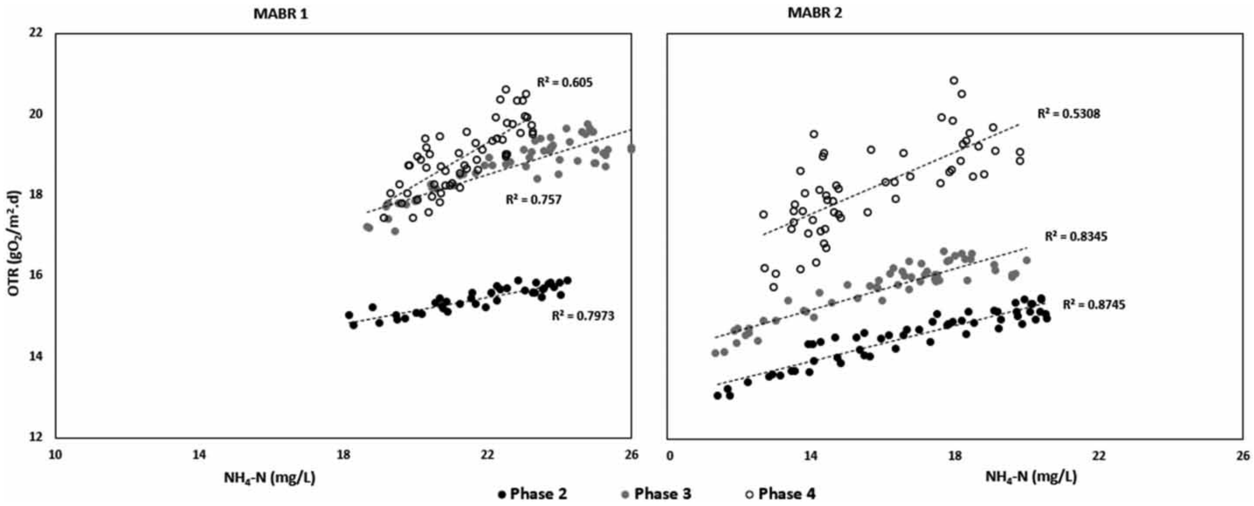


Figure 6 | OTR versus NH_4-N concentration at phases 2 to 4 in MABR 1 and MABR 2 with a linear regression model.

Table 4 | Average (St dev) results from weekly grab samples during phases 3 and 4

Parameter/location	Tank 2	Tank 3	Tank 4	Tank 5	Tank 6	Tank 7	Tank 9
CODs (1.2 μm)	129.3 (23.1)	125.0 (21.0)	117.0 (18.8)	117.5 (18.9)	112.4 (19.0)	115.8 (19.1)	24.4 b(3.0)
BODs (1.2 μm)	8.4 (2.8)	–	–	–	–	–	1.1 (0.3)
NH_4-N	14.1 (2.7)	11.8 (2.7)	7.8 (1.2)	4.8 (6.6)	0.9 (1.0)	0.9 (1.8)	0.1 (0.1)
NO_3-N	0.1 (0.1)	0.1 (0.1)	0.3 (0.5)	2.4 (1.2)	3.9 (1.7)	3.4 (2.0)	7.6 (2.4)
PO_4	24.2 (7.7)	25.3 (6.9)	21.3 (6.1)	16.3 (5.7)	12.9 (5.5)	8.9 (5.4)	6.4 (2.7)

might also be used by PAOs (phosphate accumulating organisms) since partial biological phosphorus removal was detected in the mixed liquor. The low ORP in the MABR tanks encouraged phosphate release; however, the uptake was not completed in the aerobic tanks and the phosphate in the feed wastewater was regularly similar to the phosphate discharged. The SRT or the high temperatures may have restricted the process. CODs was slowly degraded throughout the process and the MBR (Tank 8) was critical for the treatment. It held a significant portion of biomass under vigorous aeration and promoted the oxidization of the remaining biodegradable carbon and ammonia.

CONCLUSIONS

This study presented the initial results of a commercial MABR applied to nitrogen removal of high strength municipal wastewater under high seasonal temperatures. It was demonstrated that a nitrifying biofilm was developed in 3 weeks after seeding the MABR with municipal nitrifying sludge. The nitrous oxide gas emissions from the MABR proved to be a reliable parameter to assess the development of nitrification capabilities of the biofilm over time during the start up.

A baseline rate for nitrification was established and the average of 3.7 gNH_4-N/m^2.d was achieved in both MABRs under high mixing frequency and high air flow to the membranes. At that time the wastewater temperature averaged 28.3 °C but no correlation between NR and temperature could be established.

This research also demonstrated that it is possible to operate an MABR in the anoxic zone of an MLE process with no requirements for process changes. However, the mixing/scouring requirements need to be evaluated to avoid short-circuiting and excessive biofilm growth. The increase of the mixing frequency and the air pressure in the membrane lumen substantially improved the NR of MABR2. This aspect demonstrates that to achieve the maximum treatment efficiency under satisfactory energy requirements, different conditions might be applied to the individual MABR cassettes installed in sequence. It would also be advantageous if the MABR aeration control system allowed the adjustment of different air flows according to the ammonia loads.

Extremely low ORP conditions in the MABR bulk liquid appears to adversely impact the NR and it should be further investigated. Also, biological phosphorus removal is likely to be attainable concomitantly to nitrogen removal in MABRs and additional studies are required.

Literature specifies that measurement and control of biofilm thickness is fundamental to maintain the process efficiency; however, this is not practical for full-scale operations. The monitoring of ammonia concentration in the bulk liquid and the oxygen percentage in the exhausting gas allows the calculation of NR, OTR and OTE. These parameters are satisfactory for process evaluation.

Since the MABR technology is in its early stages of commercialization, this study contributes to the understanding of the design parameters and operational requirements for full-scale installations. However, long-term evaluation is essential to validate the process and its optimisation. The continuity of this research is fundamental to understand the impacts of seasonal changes and the interactions between suspended growth biomass and biofilm. The pilot plant at Subiaco WRRF is a valuable resource to manipulate process conditions, observe the impact of operational changes and gain knowledge for optimisation and troubleshooting of an MABR process.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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