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Research article

Sustainable control of microplastics in wastewater using the electrochemically enhanced living membrane bioreactor

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ABSTRACT

Wastewater treatment plant (WWTP) discharges are major contributors to the release of microplastics (MPs) into the environment. This research work aimed to assess the performance of the novel living membrane bioreactor (LMBR), which utilizes a biological layer as a membrane filter for the removal of polyethylene (PE) MPs from wastewater. The impact of an intermittently applied low current density (0.5 mA/cm^2) on the reduction of MPs in the electrochemically enhanced LMBR (e-LMBR) has also been examined. The reactors were also compared to a conventional membrane bioreactor (MBR) and an electro-MBR (e-MBR). ^{1}H nuclear magnetic resonance spectroscopy (^{1}H NMR) was implemented for the MPs detection and quantification in terms of mass per volume of sample.

The LMBR and MBR achieved comparable mean PE MPs reduction at 95% and 96%, respectively. The MPs mass reduction in the e-LMBR slightly decreased by 2% compared to that achieved in the LMBR. This potentially indicated the partial breakdown of the MPs due to electrochemical processes. Decreasing and inconsistent NH_4-N and PO_4-P removal efficiencies were observed over time due to the addition of PE MPs in the MBR and LMBR. In contrast, the integration of electric field in the e-MBR and e-LMBR resulted in consistently high values of conventional contaminant removals of COD (99.72–99.77 %), NH_4-N (97.96–98.67%), and PO_4-P (98.44–100.00%), despite the MPs accumulation.

Integrating electrochemical processes in the e-LMBR led to the development of a stable living membrane (LM) layer, as manifested in the consistently low effluent turbidity 0.49 ± 0.33 NTU. Despite the increasing MPs concentration in the mixed liquor, applying electrochemical processes reduced the fouling rates in the e-LMBR. The e-LMBR achieved comparable efficiencies in contaminant reductions as those observed in the e-MBR, while using a low-cost membrane material.

1. Introduction

A growing global concern is the widespread presence of microplastics (MPs) in the environment. MPs have been considered by previous studies as tiny pieces of synthetic polymeric particles with the dimension of less than 5 mm (Thompson, 2015). A recent standard (ISO 24187:2023) has defined MPs as solid plastic particles that are not soluble in water and are of the size ranging between 1 μm and 1000 μm (International Organization for Standardization, 2024). Those solid plastic particles that are of the dimension between 1 mm and 5 mm are defined by ISO 24187:2023 as large MPs (International Organization for Standardization, 2024). Wastewater treatment plant (WWTP) effluents are among the several conduits that permit MPs to infiltrate the environment. A report on MPs in the aquatic environment revealed that 37% of the MPs entering the global oceans are released through WWTP effluents (Boucher and Friot, 2017). The characteristics of MPs, including

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small dimension, high hydrophobicity, low degradability, and large surface area, make them suitable as adsorbents and, eventually, as carriers for other contaminants, notably pathogens, organic chemicals, and metal nanoparticles (Mozafarjalali et al., 2023; Yi et al., 2022). Additionally, aquatic species may take in MPs, which may move through the food chain, presenting possible health hazards to humans that consume these aquatic organisms (Dey et al., 2021).

Recent investigations found that the various wastewater treatment stages in WWTPs contribute to MPs reduction (Al-Amri et al., 2024; Kwon et al., 2022). Even with the reported removal or retention of MPs by existing wastewater treatment technologies, copious amounts of MPs still enter the environment because of the high daily amounts of released treated wastewater (Talvitie et al., 2015). Aside from the treatment technologies in existing WWTPs, other strategies have also been explored for the separation and/or degradation of MPs from various kinds of wastewaters, notably municipal, laundry, and industrial wastewaters. Among the studied approaches were adsorption (Lv et al., 2025; Spacilova et al., 2023), use of magnetic materials (Sacko et al., 2024; Wei et al., 2024), coagulation and flocculation (He et al., 2024; I. Lee et al., 2024), advanced oxidation processes (Easton et al., 2023; Piazza et al., 2022), micro- and ultrafiltration (Bhuyan et al., 2024; S. Kim et al., 2024; Oluwoye et al., 2024), capture of MPs by micromotors (Hermanová and Pumera, 2022; Ho and Yoo, 2024), interception of MPs by microbubble flotation (Swart et al., 2022; Zhao et al., 2024), electrocoagulation (Gabisa and Ratanatamskul, 2024; Sezer et al., 2024), electrocatalytic oxidation (Zheng et al., 2024), and electrophoretic deposition (Abdeljaoued et al., 2024).

Previous reviews on strategies to control MPs in wastewater have identified membrane bioreactors (MBRs) as one of the efficient technologies for MPs removal (Ahmed et al., 2024; Sun et al., 2019). A pilot-scale MBR for secondary treatment achieved high reduction efficiencies of up to 99%, and full-scale MBRs for tertiary treatment provided 53%–98% MPs reduction (Bayo et al., 2020; Cai et al., 2022; Egea-Corbacho et al., 2023). However, a drawback in the retention of MPs in MBRs is the possible breakthrough of microplastic fibers as reported in a prior study (Cai et al., 2022). Furthermore, the buildup of retained MPs in MBRs have been previously shown to increase the secretion of organic foulants in mixed liquor (Wang et al., 2022). This effect of MPs may hasten membrane fouling, which continues to be a major obstacle in MBR applications (Cairone et al., 2024; Chang et al., 2019; J. S. Kim et al., 2024). An alternative MBR that has not yet been investigated for MPs retention is the self-forming dynamic membrane bioreactor (SFDMBR). This reactor utilizes a self-forming biological filtering material that develops on the surface of a coarse-size mesh during wastewater treatment (Vergine et al., 2021). A novel SFDM technology named Living Membrane® (LM), an encapsulated biological layer membrane filter between two large pore size mesh support materials, physically protecting the layer from process hydrodynamics interferences, has been reported recently (Castrogiovanni et al., 2022; Jallouli et al., 2023; Millanar-Marfa et al., 2022). The LM self-forms during wastewater treatment and results from the deposition of several substances, including biomass and suspended solids between the two mesh support layers (Millanar-Marfa et al., 2022). The use of this technology in the living membrane bioreactor (LMBR) has been shown to provide effective wastewater pollutant removal and membrane fouling alleviation using a lower-cost membrane material and operating at transmembrane pressures (TMPs) much lower than conventional MBRs. In addition, the performance of the LMBR has also been enhanced in terms of wastewater treatment and membrane fouling control through its integration with electrochemical processes in the electrochemically enhanced living membrane bioreactor (e-LMBR) (Corpuz et al., 2024; Millanar-Marfa et al., 2022). However, both the LMBR and e-LMBR have not yet been evaluated in their potential to retain MPs.

This study aimed to assess the performance of the LMBR in the control of MPs in wastewater in comparison with that of the conventional MBR. In addition, the effect of the application of low current density on the control of MPs was investigated through the operations of the electro-MBR (e-MBR) and e-LMBR.

2. Materials and methods

2.1. Wastewater composition and microplastics addition

The study utilized a synthetic solution, simulating municipal wastewater, which was prepared daily according to prior studies (Borea et al., 2018; Cabreros et al., 2023; Castrogiovanni et al., 2022; Corpuz et al., 2024; Millanar-Marfa et al., 2022). The synthetic wastewater had the following composition: $C_6H_{12}O_6$ (200 mg/L); $C_{12}H_{22}O_{11}$ (200 mg/L); Protein (68.33 mg/L); $(NH_4)_2SO_4$ (66.73 mg/L); NH_4Cl (10.91 mg/L); KH_2PO_4 (4.43 mg/L); K_2HPO_4 (9.00 mg/L); $MgSO_4{\cdot}7H_2O$ (21.00 mg/L); $MnSO_4{\cdot}H_2O$ (2.68 mg/L); $NaHCO_3$ (30.00 mg/L); $CaCl_2{\cdot}6H_2O$ (19.74 mg/L); and $FeCl_3{\cdot}6H_2O$ (0.14 mg/L).

The target MPs used were micronized nonpolar polyethylene (PE) wax powder (DEUREX E 0920 M, Germany), with particle size: 98% $\leq$ 20 μm and density of 0.96–0.99 g/cm^3. This type of polymer is among the abundant and frequently detected MPs in WWTPs, as it is a common ingredient of personal care products, coatings, and food processing additives (Egea-Corbacho et al., 2023; Giannattasio et al., 2024; Sun et al., 2019). Recent surveys on municipal and industrial wastewaters have identified PE as one of the frequently detected polymer types of MPs in influents, with a relative abundance ranging from ~6 to ~54% (Egea-Corbacho et al., 2023; Hajji et al., 2024; Koyuncuoğlu and Erden, 2023; J.H. Lee et al., 2024; Magni et al., 2019; Park et al., 2020). Based on previous studies, the MPs were dosed daily into the reactor at a concentration of 10 mg PE/L of influent (Carr et al., 2016).

2.2. Experimental setup

Each of the reactors utilized a cylindrical polymethyl methacrylate (PMMA) tank (operating volume = 19 L). The membrane was submerged vertically within the bioreactors. In the MBR and e-MBR, a PVDF hollow fiber ultrafiltration membrane (pore size = 0.04 μm; active filtration area = 0.047 m^2) was used. In the LMBR and e-LMBR, an LM module described in prior studies by Castrogiovanni et al. (2022) and Millanar-Marfa et al. (2022) was utilized. The LM module was constructed using a PMMA rectangular frame. On each side of the frame were two sheets of 30 μm Dacron® mesh (polyester) that were separated by a poly (vinyl chloride) (PVC) mesh, which served both as a spacer and stiffener. The LM module has an effective filtration area of 0.021 m^2. Side and bottom air diffusers were placed in the bioreactors to supply the required dissolved oxygen, and to mix the bulk liquid.

To convert the MBR and LMBR configurations into e-MBR and e-LMBR, respectively, perforated cylindrical electrodes were added and arranged concentrically (with a distance of 6 cm from each other) in the reactor. The aluminum anode was positioned nearer the wall of the PMMA tank, and the stainless steel (SS) cathode was positioned around the membrane. The configuration of the membranes and electrodes in the e-MBR and e-LMBR were based on those used in previous studies of the authors (Borea et al., 2019; Millanar-Marfa et al., 2022). Fig. 1 depicts the experimental setup applied in this work.

2.3. Operating conditions

The first stage of the experiment evaluated the performances of the MBR and LMBR.

Synthetic wastewater was fed into the MBR in a continuous mode at a flow rate of 9 mL/min. The permeate flux in the MBR was set constant at 15 LMH using a dosing pump (Qdos 30; Watson-Marlow Pump Group). The MBR was operated using a filtration cycle that consisted of 9 min of effluent extraction, and 1 min of backwashing with distilled water (323S/D; Watson-Marlow Pump Group). Synthetic wastewater was also continuously pumped into the LMBR at a flow rate of 6.4 mL/min. The

Fig. 1. Representation of the experimental setup. In the experiment with MBR and LMBR (without applied current density), the power supply was switched off. (1 = MBR/e-MBR; 2 = LMBR/e-LMBR; 3 = aeration line; 4 = aluminum anode; 5 = stainless steel cathode; 6 = PVDF hollow fiber membrane module; 7 = LM module; 8 = backwash tank; 9 = pressure transducer; 10 = air compressor; 11 = synthetic wastewater tank; 12 = effluent tank; 13 = backwash pump; 14 = effluent pump; 15 = influent pump; 16 = DC power supply; 17 = data logger; 18 = computer).

LMBR permeate flux was fixed at 30 LMH. The same cycle (9 min of filtration + 1 min of backwashing) was employed in the LMBR.

Inoculum-activated sludge for the bioreactors was collected from the secondary clarifier of a municipal WWTP in Salerno (Italy). The sludge was retained in the bioreactor for the entire operation period (30 days), with only small sludge amounts regularly extracted for the analyses.

The membrane was pulled out from the bioreactor when the threshold TMP of 50 kPa was reached. The membrane was rinsed with water for 20 min and was allowed to soak in a 1000 mg/L Cl_2 sodium hypochlorite solution for 8 h. The membrane was then thoroughly washed with deionized water (Ensano et al., 2019).

The second stage of the experiment was the implementation of the e-MBR and e-LMBR. The same operating conditions mentioned for MBR and LMBR were tested with the e-MBR and e-LMBR, respectively, with the addition of an intermittently applied current density (0.5 mA/cm^2). Previous studies demonstrated that this low current density ensured the efficient removal of contaminants and alleviation of membrane fouling through the enhancement of biological and physical mechanisms and through electrochemical processes without inducing adverse effects to the activated sludge microorganisms (Borea et al., 2018; Millanar-Marfa et al., 2022; Tafti et al., 2015). The current was applied in cycles of 5 min ON and 20 min OFF through an external power supply (CPX400S 420 W DC power supply, Aim-TTI Instruments, United Kingdom). This "5 min ON/20 min OFF" cycle has been found to minimize the deterioration of the electrodes and to have no detrimental impacts to the microorganisms present in the mixed liquor, which refers to the combined influent and activated sludge in the reactor, while providing efficient pollutant removal and membrane fouling alleviation (Borea et al., 2018, 2019; Hou et al., 2019). A programmable controller was used to set the filtration and electric field application cycles.

2.4. Analytical methods

Samples from the influent, mixed liquor, and effluent were collected twice a week from the bioreactors to monitor contaminant concentrations. COD, NH_4-N, PO_4-P, mixed liquor-total suspended solids (ML-TSS), and mixed liquor-volatile suspended solids (ML-VSS) were measured following standard methods (APAT, 2003). MPs concentrations were measured as outlined in *Section 2.5*. Turbidity of the effluent was measured daily using a HACH 2100N laboratory turbidimeter. Temperature, pH, dissolved oxygen (DO), conductivity, and oxidation-reduction potential (ORP) of the influent, effluent, and mixed liquor were monitored daily using a multiparameter probe (Hanna Instruments, HI769828).

To investigate the membrane fouling control, the TMP was monitored with an in-line pressure transducer (PX409–0-15VI; Omega) located along the effluent line between the membrane and the permeate pump. The signals from the transducer were sent to a datalogger (34972 A LXI Data Acquisition/Switch unit; Agilent). The membrane fouling rate was measured by the difference in TMP over time ($dTMP/dt$).

The soluble microbial products (SMP) and extracellular polymeric substances (EPS), which are fouling precursors, were extracted following the heating method proposed in previously reported protocols (Le-Clech et al., 2006; Morgan et al., 1990). The carbohydrate contents of SMP and EPS (SMPc and EPSc) were determined using a colorimetric method (Dubois et al., 1951). On the other hand, the protein contents (SMPp and EPSp) were measured by the Lowry method (Lowry et al., 1951).

2.5. Microplastics extraction, detection, and quantification

The following procedures utilized to isolate, detect, and quantify the MPs in the permeate samples were based on a recently developed protocol (Giannattasio et al., 2024).

2.5.1. Isolation of microplastics

A known volume (200 mL) of permeate sample collected from the reactor was subjected to density-based separation of floating MPs by adding an aqueous NaCl solution (10 wt%). The resulting mixtures were then left to stand for 48h. The microplastics were separated through flotation to the supernatant while the organic matter was allowed to settle. The sample was then vacuum filtered using a 0.45 μm nitrocellulose (CN) filter (Sartorius, Germany), until the filter was saturated. The CN filter with the residue was transferred to an empty 50.0 mL conical centrifuge tube. Acetone (99.6%, Labkem) was added into the centrifuge tube with the CN filter until the 50.0 mL mark. The tube containing the CN filter with residue, and the acetone were then centrifuged (5000 rpm, 5 min). The purpose of the acetone addition was to dissolve the CN filter, leaving only MPs as solids. After centrifugation, the liquid was carefully decanted to leave the MPs at the tube's conical end. The same tube was refilled with the acetone until the 50.0 mL mark to completely dissolve the CN filter and was centrifuged again (5000 rpm, 5 min). The liquid was then carefully decanted from the tube. The centrifuge tube was allowed to air dry for at least 2 days to completely isolate the MPs in the sample. Aluminum foil was used to loosely cover the tube during the air drying to prevent contamination of the sample.

2.5.2. Detection and quantification of microplastics

The isolated MPs using the procedure in ***Section 2.5.1*** were moved to a 4.0 mL vial, in which 0.6 mL of the solvent 1,1,2,2-tetrachloroethane-d_2 (TCE-d_2) was added. The vial was then equilibrated at 80 °C for 24 h to facilitate the dissolution of PE MPs. The solution was then stirred and transferred to an NMR tube, in which a TCE-d_2 solution of mesitylene as an internal standard (0.1 M) was added. The NMR spectroscopy was conducted at 80 °C on a Bruker AVANCE 600 MHz spectrometer with delay (d1) = 2.0 s and number of scans (ns) = 256. The NMR spectra were elaborated using the MestReNova software (from Mestrelab Research, ver. 14.0). The concentrations of PE MPs in the permeate were calculated with the following equation:

$$PE = \frac{\left(\frac{A_{CH_{2PE}}}{2}\right) n_{IS} \bullet MW_{CH_2}}{\left(\frac{A_{CH_{IS}}}{3}\right) V_s} \quad \text{Equation 1}$$

in which PE is the concentration PE MPs in mg/L, $A_{CH_{2PE}}$ is the area of the ^{1}H NMR signal due to the methylene groups of the PE (signal at 1.12 ppm), $A_{CH_{IS}}$ is the area of the ^{1}H NMR signal due to the methine groups of the internal standard (mesitylene, signal at 6.63 ppm), n_{IS} is the mol content of the internal standard, MW_{CH_2} is the molecular weight of the CH_2 group, and V_s is the volume of permeate sample.

The MPs mass reduction was calculated based on the difference of the PE concentration in the permeate sample (mass/volume) and the known concentration of PE MPs of 10 mg PE/L of influent.

To minimize contamination of samples with MPs from the external environment, the following measures were observed: *a)* cotton laboratory gowns and gloves were utilized throughout the experiments, *b)* samples were collected using glass flasks, *c)* samples were filtered using stainless steel Büchner funnel and support screen (Millipore Analytical Stainless Steel Filter Holder – Kit), and glass flask, and *d)* permeate samples were covered with aluminum foil right after collection and during drying to avoid contamination with MPs from the atmosphere (Arslan et al., 2024; Papini et al., 2024).

3. Results and discussion

3.1. Reduction of microplastics concentrations

The obtained mean % reduction of MPs using the conventional MBR and LMBR were similar at 96.86 ± 6.62 % and 95.49 ± 4.86 %, respectively *(see* Fig. 2*)*. The mean MPs concentrations in the permeate were 0.31 ± 0.66 mg PE/L in the MBR, and 0.45 ± 0.66 mg PE/L in the LMBR. The slightly higher reduction in the conventional MBR may be ascribed to the smaller pore size of the ultrafiltration membrane than the LM layer in the LMBR. In both MBR and LMBR, the main mechanism of retention in the reactors was the adsorption of the PE MPs onto the surface of suspended sludge flocs. Since the simulated MPs used were new PE microparticles with a density of less than 1.0 g/cm^3, the MPs tended to float on the surface of the mixed liquor during the first days right after addition. They also tended to adhere to the walls of the tank near the surface of the bulk liquid. Over time, the PE MPs have been entrapped by the suspended sludge flocs (Kwon et al., 2022). The size of sludge flocs onto which the MPs were adsorbed increased over time, preventing the flocs from penetrating through the LM biological filter. In addition, some of the PE MPs may also have adhered to the LM layer instead of passing through.

The % MPs mass reductions ranged from 98.8 % to 99.5% during the first 21 days of operation in both the MBR and LMBR. However, after this period, the reduction efficiencies started to decrease as seen in Fig. 3*a*. As also shown in Fig. 4, the permeate turbidity in the LMBR rapidly increased after the 15th day and even reached the highest value at 13.45 NTU. The increase in turbidity towards the end of the operation may have resulted from the breakage of some parts of the LM layer due to higher TMP values, leading to the escape of contaminants including MPs to the effluent.

Previous studies reported up to 99% efficiency in MPs removal using a pilot-scale MBR applied for secondary treatment (Egea-Corbacho et al., 2023), and 53–98% using a full-scale MBR used for tertiary treatment (Bayo et al., 2020). The % removals obtained in this current study were comparable to those obtained in the earlier studies. Although, it is

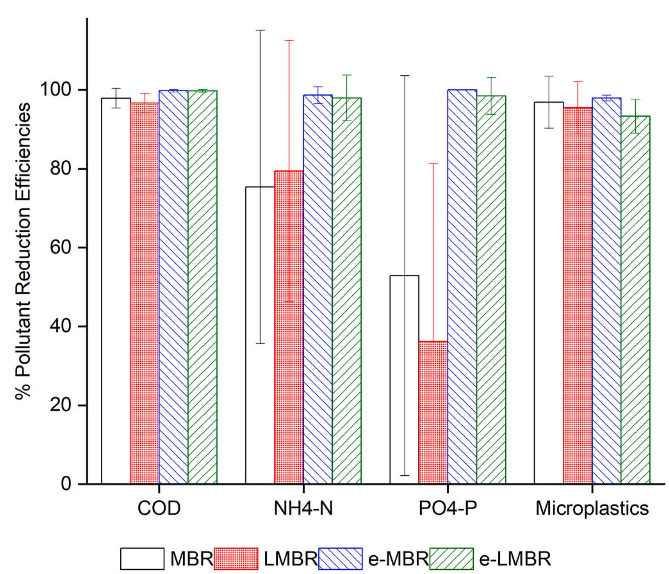

Fig. 2. Comparison of mean % pollutant reduction in the MBR, LMBR, e-MBR, and e-LMBR. The error bars indicate the standard deviations.

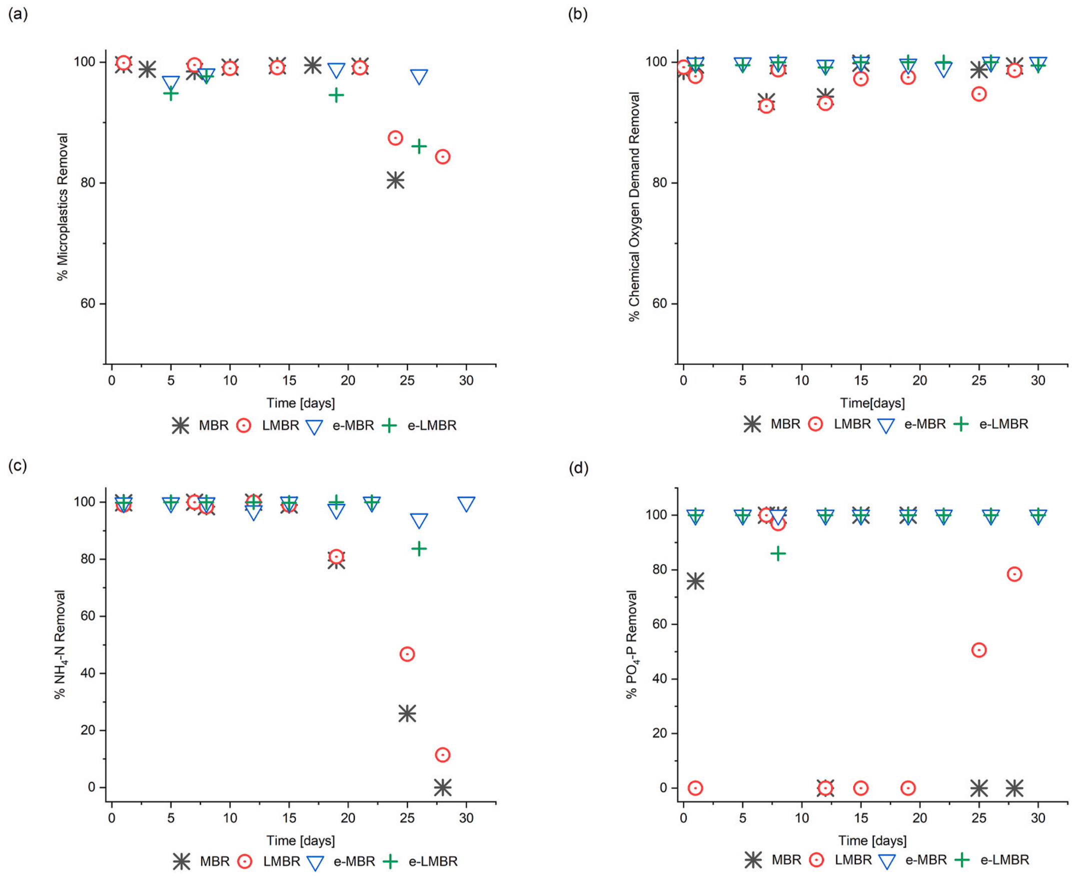

Fig. 3. Trends of efficiencies in the reduction of (a) MPs, (b) COD, (c) NH_4-N and (d) PO_4-P in the MBR, LMBR, e-MBR, and e-LMBR.

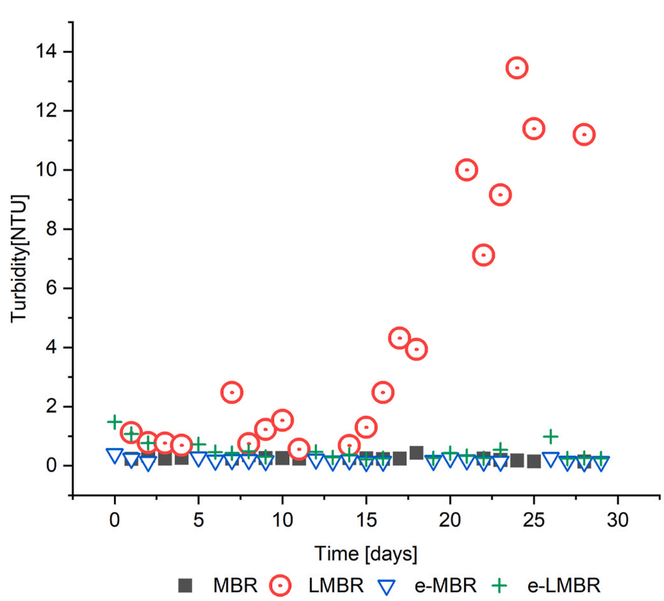

Fig. 4. Monitoring of the effluent turbidity over time in the MBR, LMBR, e-MBR, and e-LMBR.

important to note that the MPs isolation and quantification methods utilized in the former studies were different from the ones applied in the current work. Further, the earlier investigations quantified the MPs as a number of MPs/L of the sample, while the present study expressed the concentrations as a mass of MPs/L of sample. Methods that provide particle number-based concentrations are largely reliant on the size of MPs, in which those of very minute sizes may be difficult to detect. In addition, there may be further size reduction of MPs during the wastewater treatment steps due to physical and chemical mechanisms (Xu et al., 2023). The limitation in the detection of MPs of very small dimensions may lead to the overestimation of the wastewater treatment technologies' MPs removal efficiencies (Wu et al., 2024; Xu et al., 2023). In contrast, the ^{1}H NMR-based procedure of detection and quantification, which provides mass-based concentrations, is not dependent on the size of MPs, but rather relies on the response of the hydrogen nuclei in the polymers to an applied magnetic field and a radiofrequency pulse (Papini et al., 2024).

The cyclic application of electric current slightly increased the mean % MPs reduction to 97.93 ± 0.76% in the e-MBR. A slightly lower mean % MPs removal was observed in the e-LMBR at 93.49 ± 4.33% *(see* Fig. 2*)*. The mean MPs concentrations in the permeate were 0.21 ± 0.088 mg PE/L in the e-MBR, and 0.67 ± 0.50 mg PE/L in the e-LMBR. Among the electrochemical mechanisms in the e-LMBR is electrocoagulation, which potentially contributed to the removal of MPs. Gabisa and Ratanatamskul (2024) proposed that one of the MPs removal mechanisms from wastewater in an electrocoagulation system is the interaction between the surface of the electrogenerated aluminum hydroxide and MPs as represented by **Equation (2)**.

$$MPs + Al(OH)_3 \rightarrow MPs\ complex - Al(OH)_3 \qquad \text{Equation 2}$$

Integrating electrochemical processes in the e-LMBR resulted in slightly lower MPs reduction efficiencies than those achieved in the LMBR *(see* Fig. 3a*)*. This may be attributed to the potential partial breakdown of the PE MPs due to the electrochemical processes in the e-LMBR. A previous study observed partial changes in the morphology of PE MPs, such as visible cracks and pits on the surface, when subjected to electrocoagulation (Senathirajah et al., 2023). The partial degradation of the MPs may have resulted in the generation of MPs with reduced size, and subsequently in more MPs passing through the LM layer in the e-LMBR.

Previous studies attained 93–100% MPs removal by stand-alone electrocoagulation systems with aluminum as anode. These results agreed with the 93% reduction in the e-LMBR. However, the % MPs removals in the previously studied electrocoagulation systems compared to that observed in the e-LMBR may be different due to various factors. One of these is that the e-LMBR used synthetic municipal wastewater, while previous studies used wastewater of different compositions from other sources. These included secondary effluent (Akarsu et al., 2021; Elkhatib et al., 2021), laundry wastewater (Senathirajah et al., 2023), ultra-pure water spiked with MPs (Mateo et al., 2024; Shen et al., 2022), and cooling water used in a food packaging line (Sezer et al., 2024). As discussed above, the differences in MPs isolation and detection methods may also contribute to the differences in the reported % removal efficiencies. As compared to the current densities (1.63–8.07 mA/cm^2) applied in the electrocoagulation systems for the reduction of MPs found in wastewater (Elkhatib et al., 2021; Mateo et al., 2024; Sezer et al., 2024), the e-LMBR intermittently applied a lower magnitude of 0.5 mA/cm^2. It is also worth noting that the e-LMBR was not only studied for the removal of MPs alone but also for the reduction of other contaminants present in municipal wastewater, as will be discussed in the subsequent sections.

3.2. Removal of conventional pollutants

3.2.1. Removal of organic matter

The mean % COD removals in the MBR and LMBR were similar at 97.89 ± 2.53% and 96.63 ± 2.45%, respectively. The trends in Fig. 3*b* show that the % COD reductions in the MBR and LMBR were relatively constant. These results agreed with those of previous studies employing aerobic systems, in which the COD removals were not substantially impacted by the accumulation of MPs (Guo et al., 2024; Yi et al., 2022). Applying an electric field increased the mean % COD removal efficiencies (compared to MBR and LMBR) by 1.88% and 3.09% in the e-MBR and e-LMBR, respectively.

3.2.2. Removal of NH_4-N

The mean efficiencies of removal of NH_4-N in MBR and LMBR were at 75.37 ± 39.73% and 79.46 ± 33.11%, respectively. Compared to those obtained in the authors' previous studies on the LMBR at the same operating conditions without the addition of MPs, the mean NH_4-N reduction efficiencies from the present study were higher by 23.96% (Castrogiovanni et al., 2022; Corpuz et al., 2024; Millanar-Marfa et al., 2022). However, it was observed that the % NH_4-N reductions decreased rapidly starting at the 19th day of operation for both the MBR and LMBR. It was also observed that in the MBR, approximately 0.00% removal was observed towards the end of the operation. This significant drop in the efficiency of NH_4-N removals indicated the effect of the accumulation of the MPs to ammonium oxidation as they were retained in the reactors. These results agreed with those of previous studies, which reported that MPs, including PE, inhibited the activity of ammonium-oxidizing bacteria in activated and aerobic granular sludges (Li et al., 2020; Wang et al., 2021).

Applying electric current increased the NH_4-N reduction efficiencies by 23.3% in the e-MBR and by 18.5% in the e-LMBR. Contrasting with the decreasing trend obtained in the MBR, the removal efficiencies were relatively constant in the e-MBR throughout the operation (Fig. 3*c*). This suggests that the integration of an electric field mitigated the inhibitory effect of accumulated MPs to the ammonium-oxidizing bacteria. In the e-LMBR, the trend of removal efficiencies was also relatively constant and only decreased on the 26th day of operation (Fig. 3*c),* which coincided with the increase in the permeate turbidity. Applying an electric field potentially increased the activity of ammonium-oxidizing bacteria (Battistelli et al., 2019). Additional electrochemical processes, including ammonia stripping at the cathode and electrooxidation of ammonium in the anode, also contributed to the enhanced removal of NH_4-N in the electro bioreactors (Borea et al., 2019; Jing-wei et al., 2007).

3.2.3. Removal of PO_4-P

The mean % PO_4-P removals were 52.88 ± 50.74% in the MBR and 36.21 ± 45.14% in the LMBR *(*Fig. 2*)*. During the operation period, the removal efficiencies were not consistent as seen in the trend in Fig. 3*d*. This also implies the potential effect of the accumulated MPs in the reactors on the biological uptake of PO_4-P. Results of a previous study revealed that PE MPs in an aerobic granular sludge inhibited the growth of bacteria responsible for phosphorus removal (Zheng et al., 2022).

The applied low current density increased the mean PO_4-P removal efficiencies, reaching values of 100% in the e-MBR and 98.44 ± 4.67% in the e-LMBR. The PO_4-P reductions were also maintained at high values throughout the operation period despite the accumulating PE MPs within the bioreactors *(*Fig. 3*d)*. The electric field contributed to the attainment of high PO_4-P reductions through the following mechanisms: *a)* the combination of PO_4^{3-} with the Al^{3+}, which was generated from the sacrificial anode, to form the precipitate $AlPO_4$, and *b)* the adherence of PO_4^{3-} -containing substances to the Al $(OH)_3$ flocs (Belli et al., 2019; Hasan et al., 2014).

3.3. Living membrane formation

The self-formation of the LM, which is crucial in ensuring the quality of treated wastewater, is investigated through the monitoring of the permeate turbidity and TMP. The attainment of effluent turbidity values less than 5 NTU is indicative of the formation of a stable SFDM (Millanar-Marfa et al., 2021). In this study, the LMBR obtained turbidity <5 NTU at the 1st hour of operation, implying the formation of an effective LM layer. Turbidity was maintained at an average of 1.08 ± 0.56 NTU from the 1st to 15th day. However, after this period the turbidity values increased rapidly (Fig. 4). The increase in effluent turbidity coincided with the increase in TMP values in the LMBR, as seen in Fig. 5. The TMP continually increased until the 17th day of operation. External cleaning of the membrane was already conducted even if the

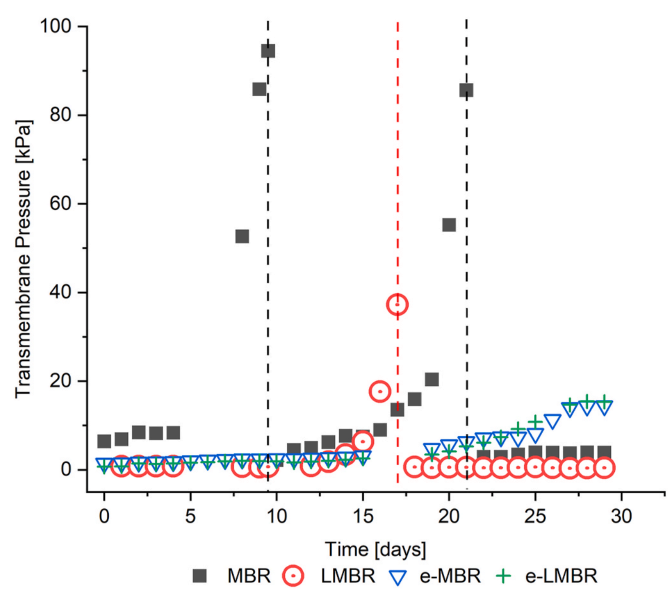

Fig. 5. Variation of transmembrane pressure values during the runs of MBR, LMBR, e-MBR, and e-LMBR. The vertical black dashed lines indicate the pull-out and cleaning of the ultrafilter in the conventional MBR. The vertical red dashed line indicates the external cleaning of the membrane module in the LMBR. No cleaning was done in the e-MBR and e-LMBR due to low TMP values during the operation period (TMP <50 kPa).

maximum TMP was still less than the threshold of 50 kPa (TMP at 37.25 kPa) since the constant permeate flux of 30 LMH was already difficult to achieve. In addition, at this point there was a significant amount of biomass going out of the reactor as also indicated by the very high turbidity. A new module was placed in the reactor to allow the development of a new LM layer. The achieved turbidity was less than 5 NTU in one day, indicating the re-formation of a new LM layer. However, the turbidity rapidly increased again until the end of the operation, which implied that the formed LM layer was not stable. In contrast, in the authors' previous work on LMBR with same operating conditions but without added MPs, the LM layer formation lasted 6–7 days, but the turbidity was maintained below 5 NTU after LM formation stage until the end of operation (at least 30 days), and the TMP did not surpass 2.5 kPa (Castrogiovanni et al., 2022; Corpuz et al., 2024; Millanar-Marfa et al., 2022).

The application of electric current in the e-LMBR resulted in a turbidity value lower than 5 NTU in 1 h. In contrast to the increasing trend obtained in the LMBR, the turbidity values were maintained at values below 5 NTU in the e-LMBR until the end of the operation (mean permeate turbidity: 0.49 ± 0.33 NTU) (Fig. 4). This suggests that the integration of electric field brought about the formation of a stable LM layer until the end of operation, despite the potential effect of accumulated PE MPs in the reactor. Despite the gradual rise in TMP toward the end of the run (Fig. 5), this did not result in the breakage of the LM layer, as manifested in the low turbidity values in the e-LMBR.

Further discussion on the increase in TMP and membrane fouling is presented in the subsequent section ***(Section 3.4)***.

3.4. Membrane fouling

The rate of membrane fouling in the MBR was higher than that observed in the LMBR (Table 1, and Fig. 5). In the MBR, the threshold TMP of 50 kPa was already reached at Day 7 and needed external cleaning. A second cleaning was also necessary since TMP value had reached >50 kPa again after Day 19. On the other hand, the TMP increased slowly in the LMBR compared with the MBR, which agreed with the lower concentrations of the fouling precursors, notably SMPc, EPsc, and EPSp in the LMBR *(*Fig. 6*)*. However, as discussed in ***Section 3.3***, the TMP in the LMBR also increased until 37.25 kPa. In the LMBR with the same operating conditions without dosed MPs in the previous studies of the authors, the TMP values were lower and were maintained to be below 2.5 kPa (Castrogiovanni et al., 2022; Corpuz et al., 2024; Millanar-Marfa et al., 2022). These results highlighted the effect of the accumulated MPs to membrane fouling in the LMBR. Some earlier research have found that the presence of PE MPs has the potential to enhance the secretion of EPS (Jachimowicz et al., 2022). Results of the current work agree with the previous study's findings as seen in the trends in Fig. 7, in which the relative contents of the substances SMP and EPS increased over time in the MBR and LMBR, as the PE MPs in the activated sludge also increased due to the daily dosing. The increase in EPS secretions was the defense mechanism of the microorganisms to the external stress due to the increasing amount of PE MPs (Jachimowicz et al., 2022).

On the contrary, the average SMP and EPS concentrations in the e-MBR and e-LMBR were significantly less than the values measured in the MBR and LMBR *(*Fig. 6*)*. The TMP values and membrane fouling rates were also lower in the bioreactors integrated with electrochemical processes compared to the conventional MBR and LMBR *(*Fig. 5 *and* Table 1*)*. These results emphasized the contribution of the intermittently applied low current density to the reduction of membrane fouling rates in the e-MBR and e-LMBR, despite the accumulation of MPs in the mixed liquor in the reactors. However, in comparison with the authors' prior studies on the e-LMBR at the same operating conditions but without added MPs, the TMP values in the e-LMBR in the present work were higher (maximum of 15 kPa) than those obtained in the previous studies (TMP values < 1.5 kPa) (Corpuz et al., 2024; Millanar-Marfa et al., 2022). This may imply an effect of the accumulation of the MPs to membrane fouling toward the end of operation in the electrochemical bioreactors.

Table 1
Fouling rates in the MBR, LMBR, e-MBR, and e-LMBR.

Bioreactor	Mean ***dTMP/dt*** kPa/day
MBR	
Before 1st cleaning	8.36 ± 12.30
Before 2nd cleaning	7.80 ± 12.42
LMBR	2.73 ± 5.26
e-MBR	0.45 ± 0.79
e-LMBR	0.48 ± 0.62

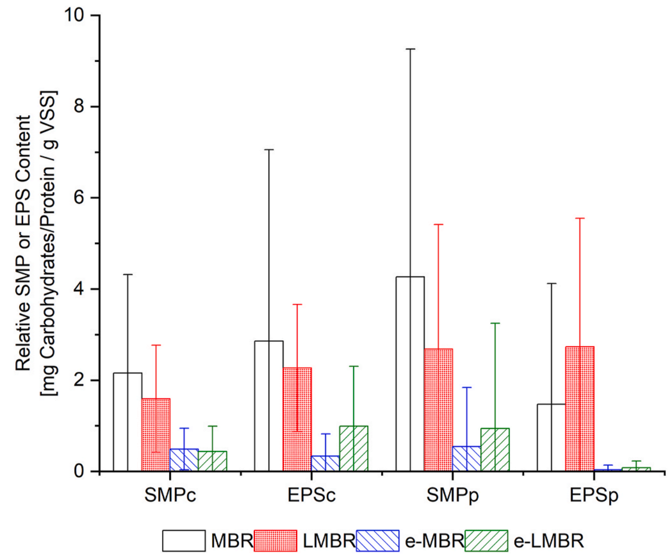

Fig. 6. Comparison of mean concentrations of fouling precursors: relative SMP and EPS contents (carbohydrates: SMPc and EPSc; proteins: SMPp and EPSp) in the MBR, LMBR, e-MBR, and e-LMBR.

4. Conclusions

The LMBR obtained PE MPs reduction efficiency of 95%, which is comparable to that obtained in the conventional MBR. Applying low current density slightly decreased the %PE MPs reduction by 2% in the e-LMBR, compared to that observed in the LMBR.

The accumulation of PE MPs resulted in low % NH_4-N and % PO_4-P reductions in the MBR and LMBR. These results underlined the influence of the accumulated PE MPs in the mixed liquor to the inhibition of the biological NH_4-N and PO_4-P removal processes. In contrast, the application of low current density in the e-MBR and e-LMBR mitigated the impact of the PE MPs, and still obtained consistently high nutrient removal efficiencies (NH_4-N: 97–98%; PO_4-P: 98–100%)

Despite the accumulation of PE MPs, the presence of electric field in the e-LMBR contributed to the development of a more stable LM (compared to the LMBR) as seen in the lower turbidity values (lower than 5 NTU) throughout the operation. Applying a low current density in the e-LMBR alleviated the tendency of the accumulated MPs in the reactor to increase the SMP and EPS concentrations. This mitigation caused slower fouling rates in the e-LMBR than those observed in the LMBR (LMBR: 2.73 ± 5.26 kPa/day; e-LMBR: 0.48 ± 0.62 kPa/day).

The study revealed that the e-LMBR is applicable for municipal wastewater treatment, particularly as a secondary treatment technology, with consistently high pollutants removal and greatly reduced membrane fouling rates. The synergistic effect of the LM technology and the electrochemical processes makes the e-LMBR a sustainable

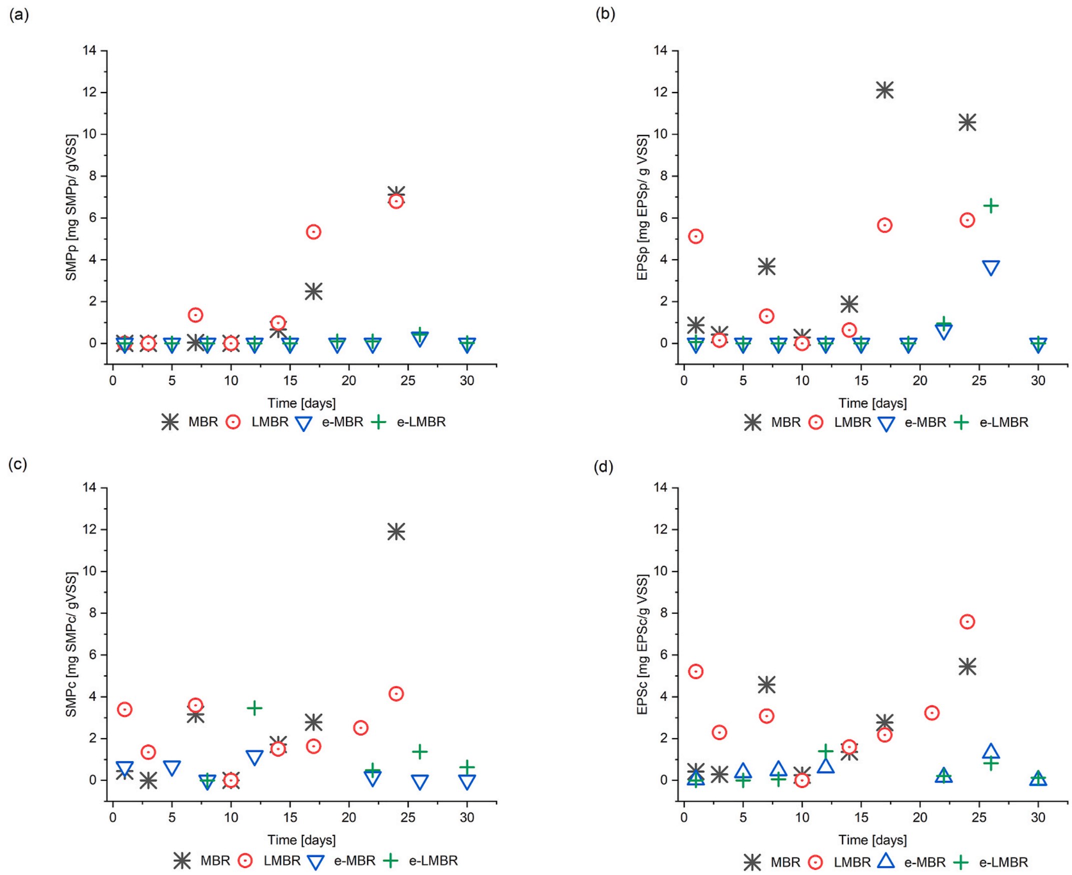

Fig. 7. Time-course monitoring of the relative SMP and EPS contents (carbohydrates: SMPc and EPSc; proteins: SMPp and EPSp) in the MBR, LMBR, e-MBR, and e-LMBR.

technology for advanced wastewater treatment, targeting removal of both conventional pollutants and MPs.

Future studies on the e-LMBR are recommended, particularly on its economic feasibility and on the application with real wastewaters.

CRediT authorship contribution statement

Mary Vermi Aizza Corpuz: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Stefano Cairone:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Mario Natale:** Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Data curation. **Alessia Giannattasio:** Writing – review & editing, Investigation. **Veronica Iuliano:** Writing – review & editing. **Alfonso Grassi:** Writing – review & editing. **Alfieri Pollice:** Writing – review & editing. **Giorgio Mannina:** Writing – review & editing. **Antonio Buonerba:** Writing – review & editing. **Vincenzo Belgiorno:** Writing – review & editing. **Vincenzo Naddeo:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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