Sustainable removal of contaminants of emerging concern from wastewater by the living membrane bioreactor: effect of the co-occurrence of microplastics and antibiotics

CORPUZ M.V.A.[1,*], GIANNATTASIO A.[2], BUONERBA A.[2,*], SANNINO F.[3], VIGLIOTTA G.[4], CHOO. K.[5], BELGIORNO V.[1], NADDEO V[1].

[1] Sanitary and Environmental Engineering Division (SEED), Department of Civil Engineering, University of Salerno, Fisciano 84084(SA), Italy, mcorpuz@unisa.it

[2] Department of Chemistry and Biology "Adolfo Zambelli", University of Salerno, 84084, via Giovanni Paolo II, Fisciano, Italy

[3] Department of Agricultural Sciences, University of Naples "Federico II", Piazza Carlo di Borbone I, 80055 Naples, Italy

[4] Laboratory of Microbiology, Department of Chemistry and Biology, University of Salerno, 84084 via Giovanni Paolo II, Fisciano, Italy

[5] Department of Environmental Engineering, Kyungpook National University, 80 Daehak-ro, Buk-gu, Daegu 41566, Republic of Korea

*corresponding author:
e-mail: mcorpuz@unisa.it

Abstract. Wastewater treatment plants (WWTPs) have been considered as sinks of contaminants of emerging concern (CECs) including antibiotics and microplastics. If not well-retained in the WWTPs, these CECs are discharged to the environment. The co-presence of these CECs in wastewater lead to further environmental impacts since MPs can act as adsorbents and carriers of antibiotics. The study aims to investigate the efficiency of a living membrane bioreactor (LMBR) in the treatment of urban wastewater containing ofloxacin and oxidized polyethylene microplastics (PE MPs). The living membrane (LM), in which a series of biological layers (an external sludge cake and an encapsulated biofilm between two coarse-size mesh sheets) function as a membrane filter, is utilized. This study aimed to investigate the effects of the presence of both ofloxacin and PE MPs to the contaminant removal and to the alleviation of membrane fouling. The influence of the presence of the PE MPs on the prevalence of antibiotic-resistant genes in the reactor will also be examined. The experiment was conducted using two parallel LMBRs, which were fed with synthetic urban wastewater with the antibiotic Ofloxacin at a concentration of 0.2 mg L influent^{-1}. One reactor was dosed with 15 µm PE MPs (10 mg L influent $^{-1}$) daily. The other reactor without added PE MPs acted as the control. Results showed that the permeate turbidity of the reactor with PE MPs was slightly higher at 1.14 NTU. This implied that the presence of PE MPs potentially affected the formation of the LM layer. Despite this, the average permeate turbidities in both reactors were lower than 5 NTU. Consistently high removals of COD and NH_4-N were obtained in both reactors. However, the removal of Ofloxacin decreased over time in both cases. The removal of Ofloxacin was slightly higher in the reactor without PE MPs (77%). This ongoing work will contribute to the development of membrane technologies for the sustainable treatment of wastewater containing micropollutants including antibiotics and microplastics.

Keywords: microplastics, antibiotic resistance, dynamic membrane, membrane fouling, wastewater treatment

1. Introduction

Wastewater treatment plant (WWTP) discharges have been shown to serve as channels for the entry of contaminants of emerging concern (CECs) to the environment. Antibiotics and microplastics are among those CECs detected in wastewaters (B. Liu et al., 2023). The co-existence of microplastics and antibiotics in wastewaters potentially results in the increased prevalence of antibiotic-resistant bacteria and genes. In addition, microplastics can serve as vectors of other contaminants such as antibiotic-resistant bacteria, viruses, heavy metals, and per- and polyfluoroalkyl substances (Jia et al., 2024; Q. Liu et al., 2022; Lu et al., 2022; Parashar et al., 2023). This study aims to examine the performance of the living membrane bioreactor in the treatment of urban wastewater containing both microplastics and antibiotics. It also aims to investigate the impact of the presence of microplastics on the levels of antibiotic-resistant bacteria and genes in the activated sludge, sludge cake layer, and encapsulated biofilm in the reactor.

2. Materials and Methods

Two parallel LMBRs are fed with synthetic urban wastewater with an antibiotic (ofloxacin at 0.2 mg $L_{influent}^{-1}$). One of the reactors is dosed with 15 µm PE MPs at 10 mg $L_{influent}^{-1}$. The other reactor without PE MPs acts as the control LMBR. The LMBR consists of a poly (methyl methacrylate) (PMMA) cylindrical tank, with a working volume of 19 L, and a vertically positioned LM module in the tank's center. The LM module is constructed with two 30 µm polyester (Dacron®) mesh sheets (Saati SpA) (Castrogiovanni et al., 2022). The inoculum biomass is an activated sludge collected from the recirculation line of the secondary clarifier of a municipal wastewater treatment plant in Salerno. Synthetic municipal wastewater is continuously fed at 6.4 mL min^{-1} to the reactors. Intermittent permeate extraction is applied according to the following cycle: 9 minutes filtration, and 1 min backwashing. The permeate flux is kept constant at 30 L m^{-2} h^{-1}. Transmembrane pressure (TMP) is monitored using a pressure sensor (PCE 932 P, Model: 100-2bar) connected to a pressure manometer with datalogger (Pressure Manometer PCE-932 Model: PS-9303SD). Permeate turbidity is monitored using a turbidimeter (Hach 2100N). The PE MPs from the permeate are extracted using a MPs extraction protocol based on sample filtration with 0.45 µm cellulose nitrate filters, dissolution of filter with acetone, and centrifugation. ^{1}HNMR spectroscopy was used for the MPs quantification. The antibiotics concentrations were determined using an ultra-high-performance liquid chromatography. Next generation sequencing (NGS) will be conducted on the inoculum sludge, and on the different layers of the LM, to examine the microbial communities and antibiotic resistance levels in the reactors.

3. Results and Discussion

3.1 **Living Membrane Formation.** Initial results revealed that in both reactors an effective living membrane formed within 1 day, as manifested by the permeate turbidity below 5 NTU. The average turbidity was lower at 0.85± 0.17 NTU in the control reactor. The permeate of the reactor with the dosed PE MPs had a slightly higehr average turbidity of 1.14 ± 0.23 NTU.

3.2 Pollutant Removals. Results showed that the average COD and NH_4-N removal efficiencies in both reactors were consistently high during the period of operation of 30 days. This implied that despite the co-presence of PE MPs and antibiotics, efficient removals of COD and NH_4-N were achieved using the LMBR. However, it is worth noting that slightly more variable results were obtained for the Total Nitrogen (TN) removals. The TN removal in the control LMBR achieved 67.62 ± 35.83%, while the LMBR fed with both PE MPs and Ofloxacin obtained 73. 24 ± 22.93%. Although the removal was higher in the reactor with PE MPs, it is worth noting that the removal was inconsistent and it decreased over time.During the first week of operation, the ofloxacin removal efficiencies were high in both reactors (96-95%).

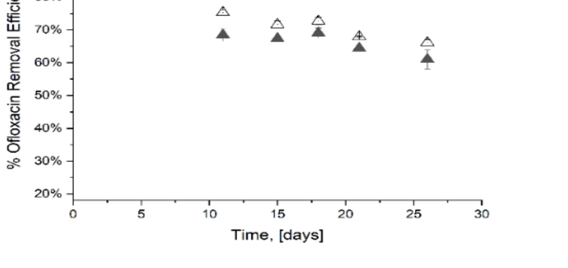

Figure 1. *Trends of efficiencies of removal of ofloxacin by the LMBR fed with urban synthetic wastewtater containing ofloxacin with antibiotics, in the presence or absence of polyethylene microplastics.*

However, efficiencies dropped up to 61 – 66% over time ***(Fig. 1)***. The LMBR without added PE MPs achieved a slightly higher removal of Ofloxacin (by 2%) as compared to that observed in the LMBR dosed with the PE MPs. This may be attributed to the slightly higher permeate turbidity in the LMBR with both PE MPs and antibiotics. This implied that the presence of PE MPs had a slight effect on the stability of the membrane, and thus on the contaminant removals.

3.3 Membrane Fouling Mitigation. The monitoring of TMP showed that in both reactors, TMP values only gradually increased and reached only up to 1.2 kPa. This is way below the treshold of 50 kPa, implying the mitigation of membrane foulign in both reactors despite the presence of Ofloxacin and antibiotics.

3.4 Microbial Community and Antibiotic Resistance. The effect of the presence of PE MPs on the microbial communities and antibiotic resistance levels in the sludge, external sludge cake layer, and encapsulated biofilm will be determined using NGS.

4. Conclusions

Results of the study show that the presence of both ofloxacin (0.2 mg $L_{influent}^{-1}$) and PE MPs slightly affects the stability of the living membrane, which consequently affected the removal of antibiotics. However, it is worth noting that the LMBR achieved consistently high COD and NH_4-N removals and low membrane fouling rates. The analysis of the microbial community will elucidate the influence of the co-presence of ofloxacin and PE MPs on the level of antibiotic resistance in the reactor.

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