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ultrafiltration-osmotic membrane bioreactor**


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Abstract

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Keywords

osmotic, ultrafiltration, hybrid, novel, performance, chemicals, organic, trace, bioreactor, removal, membrane

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Removal of Trace Organic Chemicals and Performance of a Novel Hybrid Ultrafiltration-Osmotic Membrane Bioreactor

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Abstract

A hybrid ultrafiltration-osmotic membrane bioreactor (UFO-MBR) was investigated for over 35 days for nutrient and trace organic chemical (TOrC) removal from municipal wastewater. The UFO-MBR system uses both ultrafiltration (UF) and forward osmosis (FO) membranes in parallel to simultaneously extract clean water from an activated sludge reactor for non-potable (or environmental discharge) and potable reuse, respectively. In the FO stream, water is drawn by osmosis from activated sludge through an FO membrane into a draw solution (DS), which becomes diluted during the process. A reverse osmosis (RO) system is then used to reconcentrate the diluted DS and produce clean water suitable for direct potable reuse. The UF membrane extracts water, dissolved salts, and some nutrients from the system to prevent their accumulation in the activated sludge of the osmotic MBR. The UF permeate can be used for non-potable reuse purposes (e.g., irrigation and toilet flushing). Results from UFO-MBR investigation illustrated that the chemical oxygen demand, total nitrogen, and total phosphorus removals were greater than 99%, 82%, and 99%, respectively. Twenty TOrCs were detected in the municipal wastewater that was used as feed to the UFO-MBR system. Among these 20 TOrCs, 15 were completely removed by the hybrid UFO-MBR system. High FO membrane rejection was observed for all ionic and nonionic hydrophilic TOrCs and lower rejection was observed for nonionic hydrophobic TOrCs. With the exceptions of bisphenol A and DEET, all TOrCs that were detected in the DS were almost completely rejected by the RO membrane. Overall, the UFO-MBR can operate sustainably and has the potential to be utilized for direct potable reuse applications.

Keywords: trace organic chemicals (TOrCs), membrane bioreactor, forward osmosis, osmotic MBR (OMBR), nutrient rejection, salt rejection

1. Introduction

The need to develop alternative sources of water has increased as climate change and growing populations further exacerbate the pressure on existing fresh water supplies. Reclaimed water is one of such alternative sources for augmenting fresh water supplies through water reuse. The challenge for water reuse applications is to efficiently employ a combination of biological, chemical, and physical treatment barriers to provide adequate removal of pathogenic microbial agents, nutrients, and especially trace organic chemicals (TOrcs) for public and environmental health protection.¹

Wastewater treatment facilities using either conventional activated sludge (CAS) or membrane bioreactor (MBR) were designed to remove organic carbon, nutrients such as nitrogen and phosphorus, and pathogenic microbial agents; however, they have not been specifically designed to remove TOrcs (e.g., pharmaceuticals, steroid hormones, and industrial chemicals) that could pose a potential risk to humans and the environment.² The removal of TOrcs by CAS treatment processes is highly variable and generally low. Thus, additional treatment processes such as nanofiltration (NF) or reverse osmosis (RO),³⁻⁷ activated carbon adsorption,⁸ advanced oxidation,^{9, 10} and ozonation^{11, 12} are required subsequent to activated sludge treatment to ensure adequate TOrc removal.

Highly selective membrane separation technologies such as NF and RO have an advantage over most other processes for TOrc removal because TOrcs can be physically removed from the product water instead of transformed to unidentified compounds by for example oxidative techniques. High TOrc rejection (>85%) by NF and RO has been demonstrated in bench-^{6, 7}, pilot-⁵, and full-scale studies^{3, 4} treating tertiary treated effluent for water reuse. The removal of TOrcs through semipermeable membranes used for these processes is a function of the physicochemical properties of the compound such as molecular weight, charge, and hydrophilicity, as well as of the membrane, such as charge, hydrophilicity, and salt permeability.¹³

Similar to NF and RO, excellent TOrc rejection has been demonstrated using the osmotically driven forward osmosis (FO) membrane process.¹⁴⁻²⁸ In particular, Xie et al.²³ reported that due to the retarded forward diffusion phenomenon, using the same membrane material, FO could

outperform RO for rejection of hydrophobic TOCs. Xie et al.²³ described the retarded forward diffusion phenomenon as the hindrance of TrOC transport from feed to draw solution in the membrane pores caused by the diffusion of draw solute in the opposite direction. FO differs from RO and NF in that the driving force for FO is the difference in osmotic pressure (salt concentration) between the low salinity feed stream and high salinity draw solution (DS) compared to hydraulic pressure difference between the feed and product water in RO and NF. In FO, water diffuses through the semipermeable FO membrane from the feed solution to the DS; thus, the DS is diluted during FO and the feed becomes more concentrated. To recovery fresh water and maintain a constant DS concentration, FO is commonly coupled with a DS recovery process such as membrane distillation^{29, 30} or RO.³¹⁻³³ In addition to reconcentrating the DS, a major benefit of using a hybrid FO-MD or FO-RO process is that two highly selective barriers are employed in series for removal of almost all dissolved feed constituents, including TOCs.³⁴

A further implementation of FO in a multi-barrier treatment approach is the osmotic membrane bioreactor (OMBR).³⁵⁻⁴⁰ Instead of a microfiltration or ultrafiltration membrane, an FO membrane is used in OMBR to extract water from activated sludge into a concentrated DS. The OMBR has several advantages over conventional MBR technologies including low membrane fouling propensity and superior rejection of suspended and dissolved contaminants.^{21, 31} One shortcoming of the OMBR is that dissolved constituents accumulate in the bioreactor due to high rejection of the FO membrane and reverse salt diffusion of draw solution into the feed, resulting in decreased driving force across the FO membrane and biological activity in the bioreactor, both of which reduce the efficiency of the system in terms of water recovery and carbon and nitrogen oxidation and removal.^{41, 42}

In a previous study, we have demonstrated that the accumulation of salinity in the bioreactor could be mitigated by integrating a UF membrane to the OMBR system to bleed out dissolved salts from the activated sludge.⁴³ This resulted in a novel hybrid process called UFO-MBR that can simultaneously produce one effluent stream through the UF membrane, suitable for non-potable reuse, and high quality stream through the hybrid FO-RO dual-barrier system suitable for potable reuse. In this study, we aim to extend our previous work to evaluate the performance of a

UFO-MBR system with respect to water flux and nutrient and TOrC removal. The obtained results are used to determine the applicability of this UFO-MBR system for water reuse.

2. Materials and Methods

A pilot-scale UFO-MBR was operated continuously from October 2012 to May 2013, treating municipal wastewater at the Water Reclamation Research Facility of the Colorado School of Mines in Golden, Colorado. The system was evaluated for water flux, salt accumulation, and nutrient removal (biologically) and rejection (by membranes) over the full testing period and investigated for TOrC removal and rejection for 35 days. The TOrC evaluation was conducted towards the end of the full testing period (March 2013 to April 2013) to ensure that the microbial community was well established and the system operating under steady-state conditions.

2.1. UFO-MBR pilot system

A schematic drawing of the UFO-MBR process is illustrated in Figure 1. The UFO-MBR system comprises a 105-L anoxic bioreactor and a 235-L aerobic bioreactor containing a submerged FO plate-and-frame cassette (HTI, Albany, OR) and a UF hollow-fiber PVDF membrane module (Koch Membrane Systems Puron[®], Wilmington, MA). The FO plate-and-frame cassette has five plates, each with two cellulose triacetate (CTA) membranes (one on each side) with a total system membrane area of 1.2 m². The active layer of the FO membrane faces the activated sludge in the aerobic bioreactor and the porous support layer faces the draw solution channels inside the FO plates. The UF module has a membrane surface area of approximately 0.44 m², and the nominal pore size of the membrane is 0.03 μm. A computer controlled peristaltic pump was used to draw permeate through the membrane at 90 mL min⁻¹ for 3 minutes and backwash the membrane at the end of each permeate cycle at 180 mL min⁻¹ for 30 seconds.

The anoxic bioreactor was fed with pre-screened (2 mm) municipal wastewater from a student residential complex. Sludge from the anoxic bioreactor was transferred to the aerobic bioreactor by gravity, and sludge from the aerobic bioreactor was recirculated back to the anoxic bioreactor by a pump at a flowrate of 0.25 L min⁻¹. A detailed description of the UFO-MBR system and operating conditions are provided elsewhere.⁴³ The UFO-MBR system was also integrated with a pilot RO system for DS recovery.

The pilot-scale RO system is fully automated and uses a programmable logic controller (UE9-Pro, LabJack Corp., Lakewood, CO) and a data acquisition and instrument control software (LabVIEW, National Instruments Corp., Austin, TX) to maintain a constant DS concentration by varying the hydraulic pressure through an array of three spiral-wound thin film composite membranes (SW30 2540, Dow Filmtec, Edina, MN). A detailed description of the pilot system is available elsewhere.³¹

2.2. Analytical methods for water chemistry

Samples from the influent, anoxic and aerobic bioreactors, DS, and RO permeate were collected weekly and analyzed for ammonia, nitrate, total nitrogen, total phosphorus, chemical oxygen demand (COD), and mixed liquor suspended solids (MLSS). The MLSS concentration in the anoxic and aerobic bioreactors was quantified according to Standard Methods 2540. Samples were diluted as necessary to avoid sodium and chloride interferences and measured using Hach TNT-plus™ (Loveland, CO) reagent vials and a Hach DR 5000™ spectrophotometer.

Conductivity, pH, and temperature of the anoxic and aerobic MLSS, and of the UF permeate were measured daily. The RO subsystem performance was continuously monitored using online meters that recorded the conductivity, temperature, and pH of the DS and RO permeate.

2.3. TOrC analysis

TOrC concentrations were determined using an analytical method modified from Vanderford and Snyder⁴⁴ as reported in Teerlink et al.⁴⁵ The method includes isotope dilution, solid phase extraction, followed by quantification by high performance liquid chromatography coupled with tandem mass spectrometry (LC/MS-MS). A description of the procedure for sample preservation, solid phase extraction, and analysis is provided in the Supporting Information section. Key physicochemical properties of TOrCs that were detected in the municipal wastewater and their average concentrations are summarized in Table S1 of the Supporting Information.

2.4. Rejection calculations and TOrC mass balance

The concentration of TOrCs in the UFO-MBR influent, UF permeate, DS, RO permeate, and anoxic and aerobic bioreactors were measured every seven days over the 35 day testing period to determine the removal and rejection of TOrCs in the bioreactors and across the FO and RO membranes. The TOrC rejection by the RO membrane could be directly calculated using measured concentrations; however, TOrC removal in the bioreactors and rejection by the FO membranes had to be estimated using a mass balance approach. The process flows, bioreactor volumes, and TOrC concentrations used to calculate the rejection and removal, and develop the mass balance equations are illustrated in Figure 1.

FIGURE 1

The percent rejection of TOrCs by the RO membrane (R_{RO}) is calculated using the measured TOrC concentration (C_{DS}) in the DS and in the RO permeate (C_{RO}) as described by Equation 1:

$$R_{RO} = \left(1 - \frac{C_{RO}}{C_{DS}}\right) \cdot 100 \quad (1)$$

The percent TOrC rejection by the FO (R_{FO}) membrane is calculated using a mass balance that accounts for the accumulation of TOrCs in the DS due to higher TOrC rejection by the RO membrane compared to the rejection by the FO membrane. Thus, R_{FO} is calculated using the calculated TOrCs concentration in the water crossing the FO membrane (C_{DS}^*) and the measured TOrC concentration in the aerobic bioreactor (C_{AR}) as determined using Equations 2 through 6:

$$R_{FO} = \left(1 - \frac{C_{DS}^*}{C_{AR}}\right) \cdot 100 \quad (2)$$

$$C_{DS}^* = \frac{\dot{M}_{DS}}{Q_{FO}} \quad (3)$$

$$\dot{M}_{DS} = \frac{(M_{DS} - M_{DS(t-1)})}{(t - t_{t-1})} + \frac{M_{RO}}{(t - t_{t-1})} \quad (4)$$

$$M_{DS} = V_{DS} \cdot C_{DS} \quad (5)$$

$$M_{RO} = \frac{(C_{RO} + C_{RO(t-1)})}{2} \cdot Q_{RO}(t - t_{t-1}) \quad (6)$$

C_{DS}^* is calculated from the mass flowrate of TOrCs that crossed the membrane (\dot{M}_{DS}), which is a function of the change in the mass of TOrCs in the DS (M_{DS}) over time and the mass of TOrCs that left the DS over the same period with the RO permeate (M_{RO}). The M_{DS} and M_{RO} are calculated using the measured TOrC concentration in the DS (C_{DS}) and DS tank volume (V_{DS}) and TOrC concentration in the RO permeate (C_{RO}) and RO permeate flowrate (Q_{RO}), respectively. All mass balance concentration units are in ng L^{-1} and time unit is hr.

Similar to estimating the FO rejection, percent TOrC removal in the bioreactors (R_{BIO}) is determined using a mass balance approach. The mass balance is used to calculate the modeled mass of TOrCs in the bioreactors (M_{MBIO}) assuming that no TOrCs were removed from the system through biological degradation or adsorption. The M_{MBIO} is subtracted from the measured (actual) TOrC mass in the bioreactor (M_{ABIO}) to estimate R_{BIO} as described by Equations 7 through 12:

$$R_{BIO} = \left(\frac{M_{MBIO} - M_{ABIO}}{M_{MBIO}} \right) \cdot 100 \quad (7)$$

$$M_{ABIO} = M_{AN} + M_{AR} = C_{AN} \cdot V_{AN} + C_{AR} \cdot V_{AR} \quad (8)$$

$$M_{MBIO} = \dot{M}_{MBIO}(t - t_{t-1}) + M_{AN(t-1)} + M_{AR(t-1)} \quad (9)$$

$$\dot{M}_{MBIO} = \dot{M}_{IN} - \dot{M}_{UF} - \dot{M}_{DS} - Q_W \left(\frac{C_{AR} + C_{AR(t-1)}}{2} \right) \quad (10)$$

$$\dot{M}_{IN} = \frac{(C_{IN} + C_{IN(t-1)})}{2} \cdot Q_{IN} \quad (11)$$

$$Q_{IN} = Q_{UF} + Q_{FO} + Q_W \quad (12)$$

M_{ABIO} is calculated using the measured TO_rC concentration in the anoxic bioreactor (C_{AN}) and aerobic bioreactor (C_{AR}) and corresponding bioreactor volumes (V_{AN} and V_{AR}). M_{MBIO} is determined using a mass balance that included the mass entering the system (\dot{M}_{IN}), the mass exiting with the UF permeate (\dot{M}_{UF}), the mass diffusing through the FO membrane to the DS (\dot{M}_{DS}), and the mass wasted which is a function of the wasting rate (Q_W). The mass of TO_rC for the respective inputs and outputs is calculated at each time step (time between measurements) using the measured concentrations and flowrates (Q_{IN} , Q_{UF} , Q_{FO} , and Q_W).

3. Results and Discussion

3.1. Water flux and bioreactor salinity

Throughout the investigation the UFO-MBR was operated with a NaCl DS having a concentration of 42 g L⁻¹. The UF subsystem was operated at an average flux of 7 L m⁻² h⁻¹ over the first 14 days of experimentation to maintain a steady-state salinity in the anoxic and aerobic bioreactors. Between day 14 and 28 the UF system was turned off to evaluate the effects of solute accumulation on system performance, and after day 28 the UF system was turned on again. The FO water flux, DS temperature, aerobic bioreactor salinity, and DS salinity over the testing period are shown in Figure 2.

FIGURE 2

The FO water flux shown in Figure 2a was steady over the study period apart from some variations due to changes in DS temperature and aerobic bioreactor salinity. The FO water flux increased from approximately 4 to 4.5 L m⁻² h⁻¹ over the first 14 days of the TO_rC removal evaluation as a result of an increase in the DS temperature from 13.8 to 17.7 °C. Temperature affects FO flux due to changes in solution viscosity at the feed-membrane and DS-membrane boundary layers.^{26, 46-48}

After the UF system was turned off at day 14, the FO water flux declined marginally by about 0.6 L m⁻² h⁻¹ primarily due to an increase in salinity in the aerobic bioreactor (Figure 2b) from 1.6 g L⁻¹ to 3.6 g L⁻¹ NaCl, which diminished the osmotic pressure driving force between the feed and DS. The water flux recovered to approximately 4.7 L m⁻² h⁻¹ once the UF system was turned on and the salinity in the aerobic bioreactor decreased to lower than 2 g L⁻¹ NaCl.

3.2. *Bioreactor solids, SRT, and nutrient removal*

Samples from the UFO-MBR were analyzed weekly for MLSS and nutrient concentrations as part of monitoring system stability and performance (nutrient removal). The MLSS data from the anoxic and aerobic bioreactors were used to calculate the SRT in the bioreactors. MLSS concentrations in the bioreactors and the corresponding SRT over the testing period are shown in Figure 3.

FIGURE 3

The MLSS in the aerobic and anoxic bioreactor remained relatively constant, between 1.6 and 3.6 g L⁻¹, throughout the study. The MLSS in the bioreactor fluctuated but were very similar, indicating that the sludge was well mixed and circulated between the two bioreactors. The TSS concentration in the aerobic bioreactor was consistently higher than in the anoxic bioreactor because water was constantly removed from it through the UF and FO membranes, leaving behind the suspended solids.

The SRT during testing ranged from 63 to 68 days and varied with changes in the TSS concentrations in the bioreactors. Operating at long SRTs (> 20 days) is typical of bench-scale OMBR^{36, 43} and full-scale MBR processes.^{49, 50} The advantages of operating MBRs at long SRTs include lower sludge production/wasting,⁵¹ reduced membrane fouling potential,^{50, 52} and increased removal of recalcitrant compounds and TOxCs.⁵³⁻⁵⁵

In addition to monitoring the solids concentrations in the bioreactors, ammonia, nitrate, total phosphorus, and COD of the influent and effluent were also measured on a weekly basis (Figure 4). The average influent ammonia concentration over the testing period was approximately 44 mg L⁻¹ and it was completely oxidized to nitrate in the aerobic bioreactor and remained at an average concentration of approximately 5 mg L⁻¹ in the anoxic bioreactor. As a result of low ammonia concentrations in the aerobic bioreactor, the concentration of ammonia in the DS and RO permeate were below 0.08 mg L⁻¹ (detection limit) for all measurements.

FIGURE 4

Elevated concentrations of nitrate were detected in the anoxic bioreactor, aerobic bioreactor, and DS during the first four weeks of testing. High nitrate concentrations in the anoxic bioreactor may have been due to a carbon limitation, an insufficient HRT in the anoxic zone for complete nitrate reduction (denitrification), or a reduction in the denitrification rate due to the denitrifying microorganisms being negatively affected by the elevated TDS concentration in the anoxic bioreactor.⁵⁶ The substantial decrease in nitrate concentration in the anoxic tank with increasing influent COD concentration at the end of the evaluation might support the former hypothesis.

Nitrate concentrations in the aerobic bioreactor and DS were elevated throughout the study. The high nitrate concentration in the aerobic bioreactor is due to the oxidation of ammonia (nitrification) to nitrate under the aerobic conditions in the bioreactor, and the elevated concentration in the DS is due to low nitrate rejection by the FO CTA membranes.^{31, 39, 57} The nitrate concentration in the RO permeate was consistently less than 10 mg L^{-1} prior to stopping the UF system; after which, the nitrate concentration in the RO permeate increased as a result of the higher levels of nitrate in the DS.

The influent phosphorus concentration remained consistent over all measurements with an average concentration of approximately 16.6 mg L^{-1} . The minimum measured concentration of phosphorus in the bioreactors was 44 mg L^{-1} and increased to a maximum concentration of 110 mg L^{-1} in the aerobic bioreactor when the UF system was not operating. The high concentration of phosphorus in the bioreactors is a result of phosphorus being well rejected by the FO membrane, as evident by the low measured phosphorus concentrations in the DS, and to the lack of environmental conditions in the bioreactors that would support biological phosphorus removal.

Similar to phosphorus, COD was well rejected by the FO membrane. The concentration of COD in the DS was less than 3 mg L^{-1} for all but two measurements. The COD did not accumulate in the bioreactors, as observed with phosphorus, because the carbon was biologically removed in the bioreactors. Overall, the COD was completely removed from the influent to the RO permeate.

3.3. *Bioreactor TOrC removal*

There are two pathways regularly observed for TOrC removal in activated sludge processes: microbial transformation (biodegradation) and physical separation (adsorption).^{55, 58-63} Biodegradation is governed by the bioreactor configuration because TOrCs are metabolized in the aerobic and anoxic bioreactors, and can be further degraded when the sludge is recirculated between these two unique environments.⁶⁴ Operating conditions such as SRT and MLSS concentration can also affect TOrC removal by biodegradation due to increased contact with the biologically active solids. The adsorption of TOrCs to activated sludge varies depending on the hydrophobicity, charge, and liquid phase concentration of the compound, and on the characteristics of the sludge, including the fraction of organic carbon, MLSS concentration, and food-to-microorganism ratio.^{58-60, 63} To describe the adsorption of TOrCs in activated sludge, Freundlich and Langmuir isotherm models can be used to calculate adsorption coefficients. However, these calculated coefficients must be applied with caution because they can vary among different biological systems.

Due to the complex mechanisms responsible for TOrC removal in activated sludge processes, the percent removal of TOrCs by biodegradation and adsorption in the UFO-MBR aerobic and anoxic bioreactors was calculated as a single value using a mass balance that only included the measured liquid-phase TOrC concentrations of the influent, aerobic and anoxic bioreactors, and DS as described by Equations 7 through 12. Solid phase TOrC concentrations were not considered in this evaluation because only TOrCs in the aqueous phase will potentially transport through the FO and RO membranes, impacting the final product water quality. The aqueous phase removal of 20 TOrCs and associated Log D are shown in Figure 5.

FIGURE 5

Removal of TOrCs in the bioreactor ranged from 15% to 100%, and can be grouped into two categories: low and moderately (removal <80%) and highly (removal >90%) removed compounds. All readily biodegradable compounds were well removed by the biological reactor. Aside from sulfamethoxazole (Log D = 2.43), the removal efficiency of the remaining compounds generally increased with increasing Log D (Figure 5). The lack of correlation indicates

that adsorption was likely a less relevant removal pathway for TOrCs that were highly removed during UFO-MBR operation. BIOWIN (v4.10, United States Environmental Protection Agency), a model widely used to predict biodegradability of organic compounds, could not effectively simulate the biodegradation and adsorption processes in the UFO-MBR. Limitations of the model are discussed elsewhere.^{65, 66} Hence, removal of most of the analyzed TOrCs in this study could not be explained by their BIOWIN indices (Table S2, Supporting Information).

TOrCs that were poorly or moderately removed by the bioreactor include sucralose (15%), sulfamethoxazole (64%), TCEP (57%), TCPP (70%), fluoxetine (55%), TDCP (67%), bisphenol A (74%), and diclofenac (75%). Sucralose was detected in the influent at concentrations exceeding $40 \mu\text{g L}^{-1}$ and was not degraded or adsorbed in the bioreactors due to its poor biodegradability and low log D.⁶⁷ Sulfamethoxazole, fluoxetine, and diclofenac are pharmaceutical compounds with reported removal of less than 20% in nitrifying and nutrient removing wastewater treatment plants.^{62, 68} The chlorinated flame retardants TCEP, TCPP, and TDCP are recalcitrant in the aqueous environment^{69, 70} and resistant to biological degradation in municipal wastewater treatment plants.⁵⁵ Bisphenol A, a plasticizer, is highly biodegradable in municipal wastewater treatment plants;^{71, 72} however, removal and rejection results for bisphenol A can be skewed for experiments conducted with plastic appurtenances because this compound will leach from these appurtenances during operation and testing.^{73, 74}

Twelve TOrCs were highly degraded by the bioreactors: four (acetaminophen, caffeine, ibuprofen, propylparaben) are considered to be highly degradable,^{62, 75} five (naproxen, diphenhydramine, atenolol, oxybenzone, triclocarban) reported to be moderately degradable,^{61, 62, 75, 76} and three (acesulfame, DEET, trimethoprim) would have low removal expectancies in municipal wastewater treatment systems.^{55, 67, 77} Lange et al.⁶⁷ surveyed the influent and effluent of seventeen municipal wastewater treatment plants for artificial sweeteners concentration; results from this survey revealed that acesulfame was not removed through any of the traditional treatment systems. Similarly, low removal of DEET and trimethoprim through CAS plants was reported by Bernhard et al.⁵⁵ and Göbel et al.,⁷⁷ respectively. However, in MBR systems operated at long SRTs, removal of DEET and trimethoprim increased substantially with increasing SRT.^{55, 77}

3.3.1. *FO and RO TOrC rejection*

Of the 20 TOrCs detected in the UFO-MBR influent, 18 compounds were detected in the DS and 12 in the RO permeate. Rejection of TOrCs by the FO membrane was calculated using a mass balance (Equations 1 through 6) and rejection by the RO membrane was calculated using the measured TOrC concentrations in the DS and RO permeate. TOrC rejection by the FO and RO membranes, ordered by charge and increasing molecular weight, are shown in Figure 6.

FIGURE 6

Rejection was greater than 99% by the FO and RO membranes for all measured negatively and positively charged TOrCs. Nonionic (neutral at pH values around 7) and hydrophobic ($\log D > 2$ at pH 8) TOrC rejection by the FO membrane was lower compared to the charged species and the rejection tended to increase with increasing molecular weight. Similar to the rejection of charged TOrCs, the RO membrane demonstrated near complete rejection of nonionic compounds; however, lower rejection was observed through the membrane for the hydrophobic nonionic TOrCs.

High rejection of charged TOrCs by the CTA FO membrane used for the UFO-MBR evaluation has been previously documented in bench and pilot-scale studies treating synthetic wastewater¹⁸ and secondary effluents.^{19, 21} The mechanism for the rejection of negatively charged species has been associated with electrostatic repulsion by the slightly negatively charged CTA membrane and may also be in part due to the formation of a larger hydrated layer around ionic species. Indeed, high rejection of positively charged TOrCs can be explained by the large hydrated radius of ionic species in an aqueous solution. Alturki et al.¹⁴ reported that TOrC rejection exceeded 85% for all measured TOrCs with a molecular weight greater than 266 g mol^{-1} in a short-term OMBR evaluation; this result is in general agreement with similar FO research. Furthermore, FO TOrC removal experiments conducted with secondary effluent have demonstrated higher rejection by fouled FO membranes compared to virgin membranes for positively charged and some nonionic compounds.^{19, 21} It has been hypothesized that the fouling layer may prevent partitioning of these compounds through the membrane because of pore blocking and adsorption to the organic cake layer. The CTA FO membranes were removed at the conclusion of the evaluation

and inspected for membrane fouling. Although the fouling was not severe, there was evidence of organic, inorganic, and biological fouling on the membrane surface that would have influenced TOrC rejection while operating in the activated sludge.

The rejection of TOrCs by the RO membrane was similar to that of the FO membrane for all charged species but exhibited higher rejection for the nonionic and hydrophobic nonionic TOrCs. High rejection of the hydrophilic nonionic TOrCs (acetaminophen, caffeine, acesulfame, TCEP, TCPP, and TDCP) by the SW30 RO membrane compared to the CTA FO membrane may be due to the higher selectivity of the SW30 RO polyamide active layer.⁷⁸ The rejection of the nonionic hydrophobic compounds with low molecular weight (DEET and bisphenol A) was the lowest for the FO and RO membranes; however, the RO membrane rejection was approximately three times greater for DEET and 30% greater for bisphenol A compared to the FO membrane. This result differs from those reported by Hancock et al.¹⁹ in which DEET and bisphenol A were better rejected by the CTA FO membrane than the SW30 RO membrane. Similarly, Xie et al.²³ reported that bisphenol A rejection in FO mode was higher than that in RO mode. However, these studies were conducted with deionized water and secondary effluent as the feed solutions, not activated sludge. The chemical and physical properties of the fouling layer affect convective and diffusive transport of TOrCs through the membrane in both FO and RO mode; therefore, the difference in results of the current study and previous works may be associated with properties of the fouling layer.^{18, 78}

The total UFO-MBR system removal was calculated using the influent and RO permeate concentrations. The removal included degradation in the anoxic and aerobic bioreactors and rejection by the FO and RO membranes. The total average system removal and removal over time for five TOrCs that were not completely removed during the tested period are shown in Figure 7.

FIGURE 7

The UFO-MBR removed 15 of the 20 measured TOrCs to a concentration below quantification limits from the influent to the RO permeate when the bioreactor was operated at an HRT of 30 h. Five TOrCs were detected in the RO permeate at quantifiable concentrations in the low ng L⁻¹

range (20 to 190 ng L⁻¹); of the five, four compounds (sulfamethoxazole, TCEP, TCPP, and DEET) were removed on average by greater than 95%. The low removal efficiency for sulfamethoxazole may be a consequence of the analytical technique not being sensitive enough to detect changes in the sulfamethoxazole concentration at the very low measured concentrations (15 ng L⁻¹). Bisphenol A (87% removal) was the only compound to be removed on average by less than 90%. The removal of bisphenol A was not only the lowest but the removal efficiency also appeared to be affected by the operation of the UF system (Figure 7b). Furthermore, as discussed above, leaching of bisphenol A from plastic materials during testing can lead to lower than expected rejection/removal.

The UF system was offline for 14 days (between days 14 and 28) during which the total system removal (calculated from the influent to the RO permeate) of bisphenol A, DEET, TCEP, and sulfamethoxazole decreased. The most significant decrease was observed for bisphenol A (20%) and DEET (12%), both of which increased in concentration in the bioreactors (bisphenol A from 135 ng L⁻¹ to 268 ng L⁻¹ and DEET from 62 ng L⁻¹ to 94 ng L⁻¹) while the UF system was offline. As a result of increasing bioreactor concentrations, the concentrations of bisphenol A and DEET increased in the DS from 67 ng L⁻¹ to 132 ng L⁻¹ and 146 ng L⁻¹ to 404 ng L⁻¹, respectively. Consequently the concentrations of both compounds increased in the RO permeate from 50 ng L⁻¹ to 60 ng L⁻¹ and 53 to 110 ng L⁻¹ for bisphenol A and DEET, respectively. The change in system removal over this period demonstrates the dynamic relationship between the operation of the UF membrane and FO-RO hybrid membrane system on UFO-MBR treatment performance.

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Supporting Information Available

Details on the TOrcs physical and chemical properties and analytical methods used for quantifying TOrc concentrations as well as a list of variables used in mass balance equations are provided. This information is available free of charge via the Internet at <http://pubs.acs.org/>.

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