



Effects of salinity build-up on biomass characteristics and trace organic chemical removal: Implications on the development of high retention membrane bioreactors



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HIGHLIGHTS

- The removal of hydrophilic TrOCs by MBR decreased due to salinity build-up.
- Organic carbon and nutrient removals by MBR decreased at elevated salinity.
- High salinity had negligible impact on the removal of most hydrophobic TrOCs by MBR.
- Fate of TrOCs in the sludge phase was not significantly affected by salinity.
- High salinity condition accelerated membrane fouling due to SMP and EPS release.

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ABSTRACT

This study investigated the impact of salinity build-up on the performance of membrane bioreactor (MBR), specifically in terms of the removal and fate of trace organic chemicals (TrOCs), nutrient removal, and biomass characteristics. Stepwise increase of the influent salinity, simulating salinity build-up in high retention MBRs, adversely affected the metabolic activity in the bioreactor, thereby reducing organic and nutrient removal. The removal of hydrophilic TrOCs by MBR decreased due to salinity build-up. By contrast, with the exception of 17 α -ethynylestradiol, the removal of all hydrophobic TrOCs was not affected at high salinity. Moreover, salinity build-up had negligible impact on the residual accumulation of TrOCs in the sludge phase except for a few hydrophilic compounds. Additionally, the response of the biomass to salinity stress also dramatically enhanced the release of both soluble microbial products (SMP) and extracellular polymeric substances (EPS), leading to severe membrane fouling.

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1. Introduction

Fresh water scarcity in many parts of the world is a significant concern (Elimelech and Phillip, 2011). This issue is further exacerbated by population growth, urbanization, environmental pollution, and climate change. As a result, over the last few decades, there have been many dedicated efforts to develop and improve treatment processes that utilize alternative water sources including wastewater in order to augment water supply and alleviate

water stress. A notable treatment process is membrane bioreactor (MBR) which integrates membrane filtration with the conventional activated sludge (CAS) treatment technology. MBRs can offer a better quality effluent and a lower sludge production, but with a much smaller physical footprint in comparison to CAS processes (Hai et al., 2014). Thus, the MBR system has been widely recognized as a preferable alternative, especially in water reuse applications where high effluent quality is required.

The widespread occurrence of trace organic chemicals (TrOCs) in secondary treated effluent, such as pharmaceutically active chemicals and endocrine disrupting compounds, remains a vexing issue associated with wastewater treatment, particularly for potable water reuse applications (Tran et al., 2013; Luo et al., 2014a).

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MBR is typically operated using a longer sludge retention time (SRT). The mixed liquor suspended solids (MLSS) concentration in MBR is typically from 5 to 20 g/L, which is considerably higher than that in CAS (up to 4 g/L) (Hai et al., 2014). Thus, compared to CAS, MBR can provide an appreciable removal of certain TrOCs. However, several previous studies have demonstrated a remarkable variation in the removal of TrOCs, ranging from almost complete removal of certain chemicals (e.g., estradiol and ibuprofen) to negligible removal of several others (e.g., diclofenac and carbamazepine) (Hai et al., 2011; Tadkaew et al., 2011). Such a large variation was possibly due to the diverse physicochemical properties of TrOCs (Tadkaew et al., 2011). Additionally, the removal of TrOCs could also be significantly influenced by the operating conditions of MBRs, such as hydraulic and sludge retention times (Weiss and Reemtsma, 2008), temperature (Hai et al., 2011), and mixed liquor pH (Urase et al., 2005; Tadkaew et al., 2010).

Further developments of MBRs have resulted in the emergence of a novel high retention (HR)-MBR concept. Typical HR-MBRs include osmotic membrane bioreactor (OMBR) (Achilli et al., 2009), membrane distillation bioreactor (Phattaranawik et al., 2008), and nanofiltration membrane bioreactor (Choi et al., 2002). In these systems, forward osmosis, membrane distillation and nanofiltration are employed as the high retention membrane process. These membranes have high rejection capability and can effectively retain small and/or persistent TrOCs, thus prolonging their retention time in the bioreactors for further biodegradation. As a result, HR-MBRs can be considered as a reliable technique to produce high quality effluent for potable water reuse or direct effluent discharge in sensitive areas.

The rejection of colloidal particles and certain dissolved substances by high retention membranes allows HR-MBRs to produce high quality effluent. But, this can also result in the build-up of salinity in the bioreactor (Lay et al., 2010; Luo et al., 2014b). Salinity build-up is expected for all HR-MBR types but can be more severe for OMBRs due to the reverse diffusion of draw solutes. In addition, high and/or varying wastewater salinity also presents a challenge to biological treatment such as coastal sewers subjected to infiltration of seawater or discharges from individual high salinity processes (e.g., seafood and cheese manufactures). Moderate to high salinities can alter sludge characteristics and microbial community, thereby deteriorating MBR performance (Reid et al., 2006; Yogalakshmi and Joseph, 2010). Thus, understanding the impact of salinity build-up on the performance of the biological reactor and membrane fouling is essential for the development of HR-MBRs. It is also noteworthy that, to date, the impact of salinity build-up on the removal of TrOCs during either conventional MBR or HR-MBR treatment remains unclear.

This study aimed to investigate the effects of salinity build-up (up to 16.5 g NaCl/L) on the removal of TrOCs and biomass properties during MBR treatment. These will have important implications for understanding and management of salinity build-up in HR-MBRs during wastewater treatment. The basic performance of MBR at elevated salinity was also examined in terms of organic and nutrient removals as well as membrane fouling propensity.

2. Methods

2.1. Trace organic chemicals

A set of 31 TrOCs was selected in this study based on their widespread occurrence in raw sewage and/or sewage-impacted water bodies as well as their diverse physicochemical properties, such as hydrophobicity and molecular structure (Supplementary Data, Table S1). These compounds represent four major groups of chemicals of emerging concern, including pharmaceutical and personal

care products, endocrine disrupting chemicals, pesticides, and industrial chemicals. A stock solution containing all 31 TrOCs was prepared in pure methanol at a concentration of 25 mg/mL of each chemical and stored at -18°C in the dark. The stock solution was used within a month or else discarded.

2.2. MBR set-up

In this study, two parallel lab-scale MBR systems, namely control- and saline-MBRs (Supplementary Data, Fig. S1), were continuously operated under identical operating conditions. Each system consisted of a glass reactor with effective volume of 5.7 L and housed a submerged hollow fibre microfiltration membrane module (SADF0790M mini module, Mitsubishi Rayon Engineering, Japan). The membrane was made of poly-vinylidene fluoride with a nominal pore size of 0.4 μm and an effective surface area of 740 cm^2 . The reactors were placed in a temperature-controlled water bath which was equipped with an immersion PID regulated heating unit (Julabo, Germany). Two uniform electrical air pumps (Heilea, model ACO 012) were used to aerate the reactors via coarse bubble diffusers (Aqua One, Australia) located at the bottom of the reactors to supply enough dissolved oxygen, prevent sludge settlement and also to scour the membrane. The effluent peristaltic pumps (Masterflex L/S, USA) were controlled by a computer to operate the membrane modules in 14 min on and 1 min off cycles. This on/off cycle aimed to provide a relaxation time to the membrane modules. The flow rate of the influent peristaltic pumps (Masterflex L/S, USA) was matched with that of the effluent pumps to maintain a constant reactor volume. The trans-membrane pressure (TMP) was continuously monitored by a high resolution (± 0.1 kPa) pressure sensor (Extech Equipment, Australia) as an indicator of membrane fouling.

2.3. Experimental protocol

Synthetic wastewater (Supplementary Data, Table S2) was used in this study to simulate medium strength municipal sewage and to maintain a stable influent condition. The MBR systems were seeded with activated sludge from the Wollongong Wastewater Treatment Plant (Wollongong, Australia). They were initially acclimatized for 30 days at a MLSS concentration of 5 g/L, temperature of $26.0 \pm 0.2^{\circ}\text{C}$, dissolved oxygen concentration of 5 ± 1 mg/L, and mixed liquor conductivity and pH of 260 ± 27.9 $\mu\text{S}/\text{cm}$ and 6.7 ± 0.5 , respectively. The hydraulic retention time was maintained at 24 h, corresponding to a constant permeate flux of 3.5 L/m^2 h. Once the steady-state performance of the MBRs had been established, a stock solution containing 31 TrOCs was spiked into the synthetic wastewater every day to obtain a concentration of 5 $\mu\text{g}/\text{L}$ for each compound. Both MBRs were operated for two weeks to achieve stable conditions with regard to TrOC removal and other performance before NaCl was added to the feed of the saline-MBR to simulate salinity build-up.

Salinity build-up was simulated by gradually increasing the concentration of NaCl in the saline-MBR from 0 to 16.5 g/L with a salinity gradient of 0.5 g NaCl/L per day (Supplementary Data, Fig. S1). The maximum salinity was determined by modelling the steady-state salt accumulation in an OMBR system with a commercial cellulose triacetate forward osmosis membrane (Supplementary Data, Appendix A). To investigate the possible microbial adaptation to high salinity, the saline-MBR was operated for two weeks at the salinity loadings of 10 and 16.5 g NaCl/L. Therefore, the saline-MBR was continuously operated for 70 days. By contrast, the control MBR system was operated under the same operating conditions without any NaCl addition. The weekly removal of some sludge for analysis resulted in an operating SRT of approximately 50 days. Membrane cleaning was conducted

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