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Biological performance and trace organic contaminant removal by a side-stream ceramic nanofiltration membrane bioreactor

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ABSTRACT

This study evaluated the performance of a side-stream ceramic nanofiltration membrane bioreactor (NF-MBR) system with respect to basic water quality parameters as well as trace organic contaminant (TrOC) removal efficiency. The results show a stable biological performance of the continuous NF-MBR system with high effluent quality (total organic carbon < 4 mg L^{-1} and NH_4^+–N below the detection limit). Significantly higher performance by this NF-MBR in comparison to the conventional microfiltration/ultrafiltration MBR regarding the removal of a large number of TrOCs was observed. TrOC removal efficiency depended on their hydrophobicity and molecular features. All hydrophobic compounds ($LogD_{pH=6} > 3$) were well removed (>85%), except diazinon (59 ± 7%). Hydrophilic compounds containing electron donating groups were also well removed (>90%). By contrast, hydrophilic compounds containing electron withdrawing groups were poorly removed (8–54%). Most of the 40 TrOCs investigated in this study did not accumulate in the sludge. Only three hydrophobic compounds, namely amitriptyline, triclosan and triclocarban showed considerable accumulation in sludge (>500 ng g^{-1}). Mass balance indicated biodegradation/transformation as the most significant TrOC removal mechanism by this NF-MBR.

1. Introduction

Increasingly stringent environmental regulations and freshwater shortage are major drivers for introducing advanced water recycling technologies (Anderson et al., 2014). In recent years, membrane bioreactors have been widely used for wastewater treatment, in most cases, for subsequent water recycling (Anderson et al., 2014; Hai et al., 2014a; Li et al., 2015). Compared to the conventional activated sludge (CAS) process, membrane bioreactor (MBR) can be operated at a longer sludge retention time (SRT), higher mixed liquor suspended solid (MLSS) concentration, and with a much smaller physical footprint (Hai et al., 2014b). Thus, MBRs can offer a high effluent quality, which can be further purified for water reuse applications (Alturki et al., 2010; Pandey et al., 2014; Qin et al., 2006).

A challenging hurdle to water recycling is the widespread occurrence of trace organic contaminants (TrOCs) in municipal wastewater. Conventional wastewater treatment technologies were not designed for the removal of these TrOCs. As a result, effluent discharge is a major pathway for the introduction of TrOCs into the aquatic environment (Luo et al., 2014b). Uncertainty about potential health effects of chronic exposure to these TrOCs even at trace level has triggered the need for their removal during water reuse and wastewater treatment (Schwarzenbach et al., 2006).

MBR with key characteristics such as long SRT, high MLSS concentration has been considered as a promising technology for enhancing TrOC removal (Li et al., 2015; Navaratna et al., 2012). Several previous studies have compared TrOC removal between MBR and CAS. With respect to readily biodegradable TrOCs (*e.g.* caffeine and bezafibrate), MBR showed more stable removal performance than CAS (Sui et al., 2011). MBR also achieves better removal of certain TrOCs (*e.g.* trimethoprim, gemfibrozil, and metoprolol) that are moderately removed by CAS (15–80%).

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However, several persistent TrOCs (*e.g.* carbamazepine and diclofenac) were not sufficiently removed by both CAS and MBR (Bernhard et al., 2006; Sui et al., 2011). In addition, the removal efficiency of TrOCs by MBR can vary widely depending on their physiochemical properties (Tadkaew et al., 2011) as well as operating conditions such as SRT (Boonyaroj et al., 2012; Phan et al., 2014; Weiss and Reemtsma, 2008), temperature (Hai et al., 2011; Sui et al., 2011), hydraulic retention time (Fernandez-Fontaina et al., 2012) and mixed liquor pH (Sanguanpak et al., 2015; Tadkaew et al., 2010). More importantly, MBR alone is not sufficient for adequate removal of TrOCs for water reuse. As a result, MBR effluent is usually further polished by other advanced treatment processes such nanofiltration (NF) or reverse osmosis, UV oxidation, and activated carbon adsorption prior to water reuse applications (Alturki et al., 2010; Nguyen et al., 2013; Qin et al., 2006).

To further enhance TrOC removal by MBR, several new configurations have been explored (Luo et al., 2014a). These include the integration of NF instead of the conventional microfiltration or ultrafiltration membrane with the biological reactor to form the NF-MBR configuration. Choi et al. (2007) reported the first NF-MBR study of municipal wastewater treatment in which they demonstrated excellent effluent quality of less than 4 mg L^{-1} in total organic carbon (TOC) content. However, previous studies have also showed several challenges in NF-MBR operation. They include salt accumulation in the biological reactor (Choi et al., 2007) and low permeate flux (<2.5 L m^{-2} h^{-1}) (Choi et al., 2007; Zaviska et al., 2013). A key driver for developing NF-MBR is the capacity of the NF membrane to directly retain TrOCs or the macromolecules binding TrOCs (Fujioka et al., 2015; Nghiem and Hawkes, 2007), thus prolonging their retention time in the biological reactor for an enhanced removal. With the exception of the study by Zaviska et al. (2013), TrOC removal by NF-MBR has not been studied. Furthermore, Zaviska et al. (2013) investigated the removal of only ciprofloxacin and cyclophosphamide at 100 μg L^{-1} each. Thus a more systematic study covering compounds of diverse chemical compositions is required.

This study aims to evaluate the performance of an NF-MBR system with respect to basic water quality parameters as well as TrOC removal efficiency. The fate of 40 TrOCs during NF-MBR treatment was systematically evaluated and discussed.

2. Materials and methods

2.1. Trace organic contaminants

A set of 40 TrOCs was selected for investigation. These contaminants represent major TrOC groups (*e.g.* pharmaceutically active compounds, pesticides, industrial chemicals, and personal care products) of concern in domestic wastewater and surface water. They also cover a diverse range of physicochemical properties including molecular weight, hydrophobicity and chemical structure that allow for a systematic evaluation of the performance of membrane rejection and bioreactor. The hydrophobicity of the selected TrOCs was categorised according to the Log *D* value at the specific pH of operation. The presence/absence of electron donating group (EDG)/electron withdrawing group (EWG), nitrogen bearing cyclic structure were also examined to describe the biodegradability of TrOCs during biological treatment (Tadkaew et al., 2011). The compounds were purchased from Sigma–Aldrich (Australia) with a purity of 99% or higher. A combined stock solution of TrOCs was prepared in methanol and stored at − 20 °C in the dark. TrOCs were spiked to the synthetic wastewater to achieve a final concentration of approximately 750 ng L^{-1} of each selected compound.

2.2. Laboratory scale NF-MBR set-up

A laboratory scale aerobic NF-MBR system was constructed for this study (Fig. 1). The system consists of an aerobic bioreactor and a side-stream ceramic membrane module (Fraunhofer IKTS, Germany). The membrane module was 0.25 m in length with a total effective membrane area of 0.033 m^2. It has 7 channels with inner diameter of 6 mm. According to the manufacturer, this membrane has a mean pore size of less than 0.9 nm. Two peristaltic pumps (Masterflex L/S, USA) were used for recirculation and effluent extraction. The effluent extraction pump was operated on an 8 min on and 2 min off cycle. The on/off time aimed to reduce the stress of cross-flow intensity on biological flocs and to provide relaxation time to the membrane module. The reactor volume was maintained at 4 L using an automatic floating valve for feeding. An air pump was used to maintain dissolved oxygen content of 7 ± 1 mg L^{-1} in the bioreactor via a diffuser located at the bottom of the tank. Transmembrane pressure was monitored using two pressure gauges. The hydraulic retention time (HRT), temperature and mixed liquor pH were 27 h, 21.0 ± 2.6 °C, and 6.0 ± 0.5, respectively. A long HRT (corresponding to a permeate flux of 4.5 L m^{-2} h^{-1}) was applied in this system to maintain a relatively stable membrane flux and minimize membrane fouling so that the focus of the study could be maintained on the evaluation of the TrOC removal. 70 Kpa cross-flowrate of 1.2 L min^{-1} within the membrane module was maintained for fouling minimisation. The NF-MBR system was operated without sludge withdrawal except sampling for MLSS concentration measurement (approximately 0.5% total mass per week). During the period of TrOC addition, the system was covered with aluminium foil to prevent any photodegradation.

2.3. Experimental protocol

The NF-MBR system was inoculated with activated sludge obtained from the Wollongong Wastewater Treatment Plant (Wollongong, Australia). A medium strength municipal synthetic wastewater (TOC = 124 ± 16 mg L^{-1}, n = 26) was used to provide carbon, nitrogen, phosphorus and trace metal ions for the growths of the microbes (Alturki et al., 2012). The synthetic wastewater was prepared daily by diluting a concentrated stock with deionized water. The concentrated stock solution was prepared weekly and stored at 4 °C. After 25 d of acclimatization, the NF-MBR achieved a stable biological performance as indicated by the stable removal efficiency of TOC and NH_4^+–N as well as MLSS concentration (Section 3.1). TrOCs then were introduced into the synthetic wastewater that was continuously fed to the system. Supernatant samples were collected from the mixed liquor by centrifuging (at 3000 g) then filtering through 1 μm filter paper (Millipore, Australia). Over the last two weeks of the experiment, mixed liquor samples were

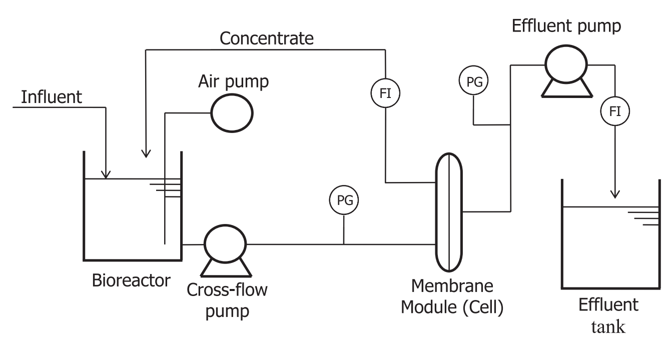

Fig. 1. Schematic diagram of the lab-scale NF-MBR system. PG: pressure gauge; FI: flow indicator.