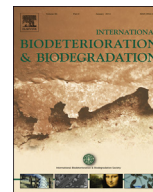




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Nutrient and trace organic contaminant removal from wastewater of a resort town: Comparison between a pilot and a full scale membrane bioreactor



Hop V. Phan^a, Faisal I. Hai^{a,*}, James A. McDonald^b, Stuart J. Khan^b, Ren Zhang^c, William E. Price^d, Andreas Broeckmann^e, Long D. Nghiem^a

^a Strategic Water Infrastructure Lab, School of Civil, Mining and Environmental Engineering, University of Wollongong, Australia

^b School of Civil and Environmental Engineering, University of New South Wales, NSW 2052, Australia

^c Molecular Biology Lab, School of Biological Sciences, University of Wollongong, Australia

^d Strategic Water Infrastructure Lab, School of Chemistry, University of Wollongong, Australia

^e GHD, Australia

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ABSTRACT

The occurrence of a broad spectrum of trace organic contaminants (TrOCs) in raw sewage from a small resort town and their removal by a full- and a pilot-scale membrane bioreactor (MBR) was analysed in this study. The MBR systems demonstrated similar reduction of chemical oxygen demand. However, the full-scale MBR sustained higher and more stable nutrient removal (>95% for both total nitrogen, TN and phosphate, PO₄³⁻-P) than the pilot-scale system (ca. 80% TN and 30% PO₄³⁻-P removal). Of the 45 monitored TrOCs including pharmaceuticals and personal care products (PPCPs), industrial chemicals, steroid hormones, and pesticides, 41 compounds were detected in the raw sewage above detection limits of 5–20 ng L⁻¹. A correlation between the removal of TN and eight TrOCs (atenolol, caffeine, naproxen, ibuprofen, gemfibrozil, DEET, estrone and diuron) was observed. Additionally, the full-scale MBR demonstrated higher and/or more stable removal for sulfamethoxazole, trimethoprim, diclofenac, diuron and amitriptyline. With the exception of caffeine, estrone and triclosan, TrOC concentrations in MBR effluent were lower than the Australian Guidelines for Water Recycling: Augmentation of Drinking Water Supplies.

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Introduction

Increasingly stringent environmental regulations and freshwater shortages are key drivers for a worldwide trend to introduce advanced sewage treatment infrastructure for removing nutrients (i.e., nitrogen and phosphorous) and trace organic contaminants (TrOCs) along with the bulk organics. In particular, due to the ineffectiveness of conventional secondary wastewater treatment processes, TrOCs such as pharmaceuticals and personal care products (PPCPs), industrial chemicals, steroid hormones and pesticides are ubiquitous in wastewater treatment plant (WWTP) effluents. Ineffective wastewater treatment is a major conduit by which TrOCs reach natural water bodies. This raises considerable concern

regarding their detrimental effects on the aquatic organisms and even humans after chronic ingestion (Luo et al., 2014).

Small to medium WWTPs are being progressively implemented in small townships and tourism hot spots around Australia to phase out unreliable septic tank systems. Several studies have assessed the occurrence of TrOCs in wastewater originating from various catchment areas including agricultural, rural, urban and industrial wastewater catchments (Leusch et al., 2014; Scott et al., 2014) and the treated effluent produced by various types and scales of WWTPs (Coleman et al., 2008; Ying et al., 2009; Leusch et al., 2014). However, only a few studies reported the TrOC profile of raw sewage generated from small Australian towns (Braga et al., 2005; Coleman et al., 2008; Leusch et al., 2014), particularly those which are tourist destinations (Le-Minh et al., 2010; Trinh et al., 2012b). Wastewater from small resort towns can have distinct characteristics in terms of volume of wastewater produced and also the frequency and concentration in which TrOCs may occur. A majority

* Corresponding author. Tel.: +61 2 4221 3054.

E-mail address: faisal@uow.edu.au (F.I. Hai).

of the Australian studies investigated the removal efficiency of WWTPs for endocrine disrupting chemicals, particularly the steroid hormones, with fewer studies also focussing on pharmaceuticals, industrial chemicals and pesticides (Le-Minh et al., 2010; Trinh et al., 2012a). Notably most available reports on TrOC removal from real sewage have documented the performance of conventional treatment technologies (e.g., activated sludge process, bio-filters, and lagoons) (Ying et al., 2009).

Membrane bioreactors (MBRs) are an attractive option for decentralised wastewater treatment and reuse due to their ability to produce high quality effluent with a small footprint (Hai et al., 2014). MBRs account for the majority of new sewage treatment infrastructure in Australia. Most of these are small to medium MBR plants for water recycling applications in coastal towns and small cities, and are mostly driven by stringent environmental regulations, particularly targeting nutrient and TrOC removal, and to a lesser extent by freshwater scarcity. To date only a few Australian studies have investigated TrOC removal from real sewage, and these studies have been conducted mostly via pilot-scale MBRs (Coleman et al., 2008; Le-Minh et al., 2010; Trinh et al., 2012b). Available studies provide useful preliminary understanding; however, the performance of current MBR technology as a barrier for a range of TrOCs and specific removal mechanisms involved remains unclear.

Biodegradation can proceed under aerobic, anoxic or anaerobic regimes. Sequential exposure to different redox conditions is a pre-requisite to nutrient (i.e., nitrogen and phosphorus) removal from wastewater. For example, nitrogen removal occurs via nitrification under aerobic conditions and denitrification under anoxic conditions. In order to achieve high nitrogen removal, complete nitrification and effective recirculation of nitrate to the anoxic zones is necessary. A simple two-stage pre-anoxic/aerobic reactor configuration can typically meet the total nitrogen (TN) disposal guideline of 10 mg L^{-1} (Hai et al., 2014). A series of aerobic/anoxic zones with supplemental organic carbon dosing to the anoxic zone may be required to achieve further improved TN removal to comply with a more stringent effluent TN guideline (sensitive areas). This may also facilitate stable removal when TN loading in wastewater fluctuates significantly. Notable in this context is that recent studies demonstrate close relationships between stable $\text{NH}_4^+\text{-N}$ removal and the removal of TrOCs (Helbling et al., 2012). Most of the previous reports have shown the correlation of TrOC removal with the stability of $\text{NH}_4^+\text{-N}$ removal via batch tests conducted with synthetic wastewater. Other than the study by Vader et al. (2000) who showed a noticeable connection between $\text{NH}_4^+\text{-N}$ and 17α -ethinyl estradiol removal, this correlation has not been validated at full scale level. Furthermore, compared with aerobic (nitrifying) conditions, fewer studies have investigated TrOC removal performance of combined anoxic/aerobic reactors (Xue et al., 2010; Phan et al., 2014). It is not clear whether, like TN, a combination of a number of aerobic and anoxic zones with different levels of dissolved oxygen concentration (DO) may be conducive to removal of different TrOC categories.

Given the research gaps discussed above, the aim of this study was to assess the occurrence and removal of a broad spectrum of TrOCs by a full-scale MBR plant serving a small resort town. Performance comparison with a pilot-scale MBR fed with the same sewage was used to clarify important aspects regarding bulk organics, TrOCs and nutrient removal. The potential impact of the application of multiple sequences of anoxic/aerobic regimes on nutrient and TrOC removal is also discussed.

Materials and methods

This study was conducted at a full-scale MBR plant (designed for a maximum capacity of $743 \text{ m}^3 \text{ d}^{-1}$) located in Kangaroo Valley

(New South Wales, Australia), which is a tourist destination known for caravan parks. The Kangaroo Valley township has a permanent population of about 340 people; however, this increases during peak holiday periods to approximately 1400. In addition to influent wastewater sampling over 15 events (from November 2012 to October 2014), a pilot-scale MBR was operated at the site. It was first operated for 11 weeks for acclimatization and performance stabilization. Then, a 10-week sampling campaign was carried out to compare the treatment performance of the pilot- and full-scale MBRs receiving the same sewage.

Description of the full-scale MBR

The MBR received wastewater via a pressurised sewerage network from the Kangaroo Valley township. A schematic diagram of the plant is presented in Supplementary Data Figure 1. The treatment process comprised i) primary treatment, ii) two parallel trains of activated sludge reactors integrated with membrane filtration cells, and iii) a UV disinfection unit (UV dose of 40 mJ cm^{-2}). One of the duplicate process trains was operated in stand-by mode. The activated sludge system consisted of a pre-anoxic zone ($\text{DO} = 0\text{--}0.5 \text{ mg L}^{-1}$), aerobic zone-1 ($\text{DO} = 0.5\text{--}1.0 \text{ mg L}^{-1}$), aerobic zone-2 ($\text{DO} = 2\text{--}2.5 \text{ mg L}^{-1}$), and a post-anoxic zone ($\text{DO} = 0\text{--}0.5 \text{ mg L}^{-1}$) receiving supplemental organic carbon (acetic acid, approximately 40 L d^{-1}) to enhance denitrification. The mixed liquor from the aerobic zone-2 was recycled to the pre-anoxic zone with an internal recirculation ratio of 4. The return activated sludge from the membrane cell was recycled to aerobic zone-2 and the pre-anoxic zone, also with a recirculation ratio of 4. The solids retention time (SRT) of the MBR was 25 d and the mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentration in the aerobic reactors were respectively 8.5 ± 0.7 and $6.2 \pm 0.5 \text{ g L}^{-1}$ ($n = 10$) during the period of performance-comparison with the pilot MBR. The total hydraulic retention time (HRT) was 1.5–1.7 d with approximate HRTs in pre-anoxic zone, aerobic zone-1, aerobic zone-2, post-anoxic zone and the membrane cell of 0.45, 0.45, 0.45, 0.22 and 0.14 d, respectively. PVDF microfiltration membranes (Memcor, Evoqua Water Technologies, Australia) were submerged into two membrane cells to provide a surface area of $2400 \text{ m}^2 \text{ cell}^{-1}$.

During the course of this study, the total sewage flowrate was $146 \pm 76 \text{ m}^3 \text{ d}^{-1}$ ($n = 66$), and the membrane flux was 2.1 ± 1.1 ($n = 66$) and $1.1 \pm 1.1 \text{ L m}^{-2} \text{ h}^{-1}$ ($n = 31$) for the primary and stand-by membrane cells, respectively. The final effluent was directed to a storage dam and then used for irrigation to farms and recreational facilities around the region.

Pilot-scale MBR setup and operation

A pilot-scale anoxic–aerobic MBR (Supplementary Data Figure 1) was operated parallel to the full-scale MBR. The pilot MBR was operated under the same SRT (25 d), total HRT (1.5 d) and with the same internal recirculation ratio (4) between anoxic–aerobic reactors. However, compared to the four reactors ($2 \times$ anoxic and $2 \times$ aerobic) in the full-scale MBR, it contained only one pre-anoxic zone (working volume = 13.8 L, HRT = 0.8 d) and an aerobic zone (working volume = 11.7 L, HRT = 0.7 d). A hollow fibre ultrafiltration membrane (Zeweed-10) supplied by Zenon Environmental (Ontario, Canada) was submerged in the aerobic reactor. This membrane had a nominal pore size of $0.04 \mu\text{m}$ and an effective membrane surface area of 0.93 m^2 , and was operated at a flux of $1.2 \text{ L m}^{-2} \text{ h}^{-1}$. The transmembrane pressure (TMP) was continuously recorded via a high resolution ($\pm 0.1 \text{ kPa}$) pressure sensor connected to a data logging computer. All pumps were controlled

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