



Comparison between sequential and simultaneous application of activated carbon with membrane bioreactor for trace organic contaminant removal



Luong N. Nguyen^a, Faisal I. Hai^{a,*}, Jinguo Kang^{a,b}, Long D. Nghiem^a, William E. Price^b, Wenshan Guo^c, Huu H. Ngo^c, Kuo-Lun Tung^{d,e}

^a Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering, University of Wollongong, Wollongong, NSW 2522, Australia

^b Strategic Water Infrastructure Laboratory, School of Chemistry, University of Wollongong, Wollongong, NSW 2522, Australia

^c Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Sydney, Broadway, NSW 2007, Australia

^d Department of Chemical Engineering, National Taiwan University, Roosevelt Rd., Taipei 106, Taiwan

^e R&D Center for Membrane Technology, Chung Yuan University, Chung-Li 320, Taiwan

HIGHLIGHTS

- ▶ >95% removal of all trace organics was achieved by MBR–GAC and PAC–MBR initially.
- ▶ Gradual drop in removal of some hydrophilic compounds occurred in both systems.
- ▶ Known persistent compound carbamazepine showed high removal in both systems.
- ▶ Charged compounds fenoprop and diclofenac showed high persistence in both systems.
- ▶ PAC–MBR outperformed MBR–GAC in terms of adsorbent consumption.

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ABSTRACT

The removal efficiency of 22 selected trace organic contaminants by sequential application of granular activated carbon (GAC) and simultaneous application of powdered activated carbon (PAC) with membrane bioreactor (MBR) was compared in this study. Both sequential application of GAC following MBR treatment (MBR–GAC) and simultaneous application of PAC within MBR (PAC–MBR) achieved improved removal (over 95%) of seven hydrophilic and biologically persistent compounds, which were less efficiently removed by MBR-only treatment (negligible to 70%). However, gradual breakthrough of these compounds occurred over an extended operation period. Charged compounds, particularly, fenoprop and diclofenac, demonstrated the fastest breakthrough (complete and 50–70%, in MBR–GAC and PAC–MBR, respectively). Based on a simple comparison from the long-term performance stability and activated carbon usage points of view, PAC–MBR appears to be a better option than MBR–GAC treatment.

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1. Introduction

In view of the potential adverse effects of trace organic contaminants on human health and that of other biota, numerous studies have been devoted for their removal from wastewater by membrane bioreactors (MBRs). The reported data have demonstrated better and/or more stable removal of trace organic contaminants of moderate to high biodegradability and/or significant hydrophobicity by MBRs as compared to conventional activated sludge (CAS) processes. However, significant variations in removal of hydrophilic and biologically persistent compounds by MBR have been

noted in several recent studies (Bernhard et al., 2006; Visvanathan et al., 2005). Besides biological processes, adsorption onto powdered activated carbon (PAC) or granular activated carbon (GAC) can also be used to efficiently remove trace organic contaminants from water (Ternes et al., 2002). However, limited adsorption of ionic compounds, particularly of those containing electron-withdrawing functional groups, has been reported (Ternes et al., 2002). In this connection, the concept of combined processes such as coupling of MBR with PAC/GAC has been explored (Li et al., 2011; Lipp et al., 2012; Nguyen et al., 2012). Activated carbon adsorption in conjunction with an MBR can be applied in two different configurations: (i) direct addition of PAC into MBR (PAC–MBR), and (ii) post-treatment of MBR permeate by passing it through a GAC column (MBR–GAC) or by dosing of PAC. It is

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

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

envisaged that due to the complete retention of sludge by membrane, the trace organic contaminants adsorbed onto PAC may be efficiently removed by a PAC–MBR (Li et al., 2011). On the other hand, because MBR can produce suspended solids-free permeate with low total organic carbon content, GAC can specifically target the residual trace organic contaminants in MBR permeate with reduced interference from the bulk organics (Nguyen et al., 2012).

A few studies have explored PAC–MBR for the removal of trace organic contaminants (Li et al., 2011; Serrano et al., 2011; Zhang et al., 2008). Several studies have also explored adsorption of trace organic contaminants on either PAC (Lipp et al., 2012) or GAC (Nguyen et al., 2012) as a post-treatment following MBR treatment. Previously reported data have confirmed instant improvement of removal of selected biologically persistent trace organic contaminants from liquid phase by combined MBR–activated carbon systems. However, a comprehensive understanding of the phenomena involved based on long-term operation is yet to be achieved. In particular, no study has specifically compared the trace organic contaminant removal performance of MBR–GAC and PAC–MBR.

This study systematically compared the removal efficiency of 22 selected trace organic contaminants by sequential application of GAC and simultaneous application of PAC with MBR (MBR–GAC and PAC–MBR, respectively). Compounds showing extraordinary resistance towards MBR treatment and their extent of removal by the combined systems were given special focus. The two options were compared on the basis of performance stability and activated carbon consumption.

2. Methods

2.1. Trace organic contaminants

A set of 22 trace organic contaminants were selected for investigation based on two criteria: (i) their representation of four major groups of trace organic contaminants, namely, pharmaceutically active compounds, steroid hormones, pesticides and industrial chemicals and metabolites (Table S1), and, (ii) their widespread occurrence in natural water bodies polluted with wastewater (Kim et al., 2007; Ternes et al., 2002). The compounds were purchased from Sigma–Aldrich (St. Louis, MO, USA). The purity of these chemicals was reported to be 98% or higher. The selected trace organic contaminants were first dissolved in pure methanol to make up stock solutions of 1 g/L. The stock solutions were stored in a freezer at $-18\text{ }^{\circ}\text{C}$ and were used within 1 month. The trace organic contaminants were introduced to the feed solution to achieve a constant concentration of approximately 5 $\mu\text{g/L}$ of each compound.

2.2. Synthetic wastewater

A synthetic wastewater containing glucose (400 mg/L), peptone (100 mg/L), KH_2PO_4 (17.5 mg/L), MgSO_4 (17.5 mg/L), FeSO_4 (10 mg/L), CH_3COONa (225 mg/L) and $\text{CO}(\text{NH}_2)_2$ (35 mg/L) was used. The synthetic wastewater simulated medium strength wastewater with a total organic carbon (TOC), total nitrogen (TN) and chemical oxygen demand (COD) of approximately 180, 25 and 600 mg/L, respectively. The synthetic wastewater was prepared freshly each day from concentrated stock solution and fed into the reactor.

2.3. Activated carbon

In this study, two types of activated carbon namely GAC 1200 and PAC 1000 (Activated Carbon Technologies Pty Ltd, Victoria, Australia), with a specific surface area of 1121 and 1355 m^2/g , respectively were used. The characteristics of each type of acti-

vated carbon are listed in Table S2. Activated carbon was washed with Milli-Q water to remove fine particles, and then dried at $105\text{ }^{\circ}\text{C}$ for 24 h and stored until use.

2.4. MBR–activated carbon systems and experimental protocols

2.4.1. Design of MBR and GAC column

A laboratory scale MBR set-up (Fig. S3) with an active volume of 4.5 L and equipped with a PVDF hollow fibre membrane module (Mitsubishi Rayon Engineering, Japan), was employed in this study. A borosilicate glass column (Omnifit, Danbury, CT, USA) filled with 7.5 g of GAC was used as a post treatment unit for the MBR permeate (Nguyen et al., 2012). The column had an internal diameter of 1 cm and an active length of 22 cm, resulting in a bed volume (BV) of 17 mL.

2.4.2. Experimental protocol

The current study was conducted over total 306 days, with 51 days of initial start-up period, 93 days of operation in MBR–GAC mode, 100 days of operation in PAC–MBR mode, and rest of the period in MBR-only operation mode. The exact sequence of different operations has been listed in Table S4. The addition of the selected trace organic contaminants to the synthetic wastewater was started after the start-up period, when the mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentration were 5 and 4.5 g/L, respectively. The MBR system was operated for further 15 days before adding a GAC column as a post-treatment process for MBR permeate. During operation in MBR–GAC mode the MLSS concentration increased to 9.8 g/L. On day 158, the GAC column was disconnected from the MBR and MBR operation was continued as usual. Sludge was withdrawn on day 197 to reduce the MLSS concentration to 6 g/L prior to addition of PAC. PAC was added into the reactor on day 206 and subsequently on day 243 of continuous operation to obtain PAC concentrations of 0.1 and 0.5 g PAC/L, respectively (Table S4).

The MBR was seeded with activated sludge from another laboratory scale MBR system, which had been used for trace organic contaminant removal for about 3 years. Dissolved oxygen (DO) concentration of the mixed liquor was maintained at above 3 mg/L. The MBR was placed into a water bath to maintain the mixed liquor temperature at $22 \pm 0.1\text{ }^{\circ}\text{C}$. The pH of the mixed liquor remained stable within the range of 7.2–7.5. To ensure that membrane fouling did not interfere in the observation of trace organic contaminant removal, a low average membrane flux of 3.1 $\text{L}/\text{m}^2\text{h}$ was applied in this study. The membrane was operated on a 14 min “suction” and 1 min “relaxation” cycle, resulting in a hydraulic retention time (HRT) of 24 h. However, transmembrane pressure (TMP) was continuously monitored using a high-resolution ($\pm 0.1\text{ kPa}$) pressure sensor (SPER scientific 840064, Exttech equipment Pty. Ltd, Victoria, Australia), and ex situ backwash (NaOCl solution with 500 mg active chlorine per L) of the membrane every 90 days was observed to keep the TMP stable. In the MBR–GAC configuration, the MBR permeate was pumped through the GAC column in an up-flow mode at a flow rate of 2.4 mL/min (equivalent to 8.5 BV/h), resulting in an empty bed contact time (EBCT) of 7 min. The PAC–MBR system was operated in the same fashion as that during the MBR operation.

2.5. Analysis of trace organic contaminants and other basic parameters

The performance of the MBR, MBR–GAC and PAC–MBR systems was compared mainly in terms of trace organic contaminants and TOC/TN removal efficiency. Operating parameters such as MLSS and MLVSS concentration, turbidity, sludge volume index (SVI) and specific oxygen uptake rate (SOUR) were also monitored to

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