



Enhancement of removal of trace organic contaminants by powdered activated carbon dosing into membrane bioreactors



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ABSTRACT

This study compared the removal efficiency of 22 widespread trace organic contaminants by a laboratory-scale membrane bioreactor (MBR) with and without direct addition of powdered activated carbon (PAC) into the activated sludge reactor over a period of 312 days. The removal of hydrophilic and biologically persistent trace organic contaminants was immediately improved to above 95% after the addition of PAC into MBR. However, a compound-specific gradual decrease in removal underscored the requirement for the addition of fresh PAC. Adsorption onto PAC-added sludge appeared to play a significant role in the relatively more effective aqueous phase removal of a few resistant compounds such as carbamazepine in this study. A slower reduction in removal efficiency of compounds showing extraordinary persistence such as fenprop and diclofenac was observed after raising the PAC concentration in the MBR from 0.1 to 0.5 g/L. Nevertheless, comparison of extent of removal in terms of PAC usage indicated the suitability of more frequent dosing of smaller amounts of PAC.

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1. Introduction

A great number of natural and anthropogenic trace organic contaminants have been detected in raw sewage and sewage-impacted water bodies all over the world at concentrations up to a few $\mu\text{g/L}$ [1]. These trace organic contaminants include pharmaceutically active compounds (PhACs), steroid hormones, pesticides and industrial chemicals and their metabolites. Given their potential adverse effects on human and other biota, their removal is of utmost importance to protect the environment and ensure provision of safe drinking water. However, conventional water and wastewater treatment processes were not specifically designed for the removal of trace organic contaminants and thus their capacity for removing these particular contaminants varies widely [2].

Membrane bioreactors (MBRs) demonstrate superior performance over conventional activated sludge (CAS) processes in terms of basic effluent quality parameters [3]. By compactly combining a bioreactor with membrane filtration units, MBR has the potential to produce effluent suitable for reuse. Thus there has been considerable

research effort on the assessment of trace organic contaminant removal by MBR treatment. Research suggests that compared to CAS, MBR often results in better and/or more stable removal of trace organic contaminants possessing moderate to high biodegradability [2]. However, significant variation in MBR removal performance, particularly for biologically persistent hydrophilic compounds, has been noted in several recent studies [4–6]. Strategies explored in recent studies to enhance the removal of trace organic contaminants include fine tuning of operational parameters such as hydraulic retention time (HRT), sludge retention time (SRT) [7], dissolved oxygen concentration [8], pH [9] and temperature [10]. Adsorption on activated sludge, potentially followed by biodegradation is the main mechanism that accounts for the removal of trace organic contaminants by MBR [2,4,6,11]. A way to increase adsorption of trace organic contaminants is to add powdered activated carbon (PAC) or granular activated carbon (GAC) to the bioreactor. It has been also suggested that a significant increase in the retention of soluble contaminants onto PAC-added sludge may enhance the biodegradation of slowly biodegradable compounds [12].

A number of studies have investigated the application of PAC within an MBR (PAC-MBR) in relation to membrane fouling mitigation [13–15]. However, despite the likelihood of enhanced biodegradation of biologically persistent organic compounds in a

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PAC-MBR system, very few of them have assessed or reported on this aspect. To date, PAC-MBRs have been explored with different sources of wastewater [12,16–19], but only a few studies have specifically explored PAC-MBR for the removal of trace organic contaminants [20–23]. Some studies have also explored adsorption of contaminants on either PAC [24] or GAC [25] as a post treatment following MBR treatment with encouraging results. Previously reported data confirmed enhanced removal of selected biologically persistent trace organic contaminants from aqueous phase by adding PAC into MBR. However, a comprehensive understanding of the phenomena involved, particularly the simultaneous effect of PAC dose and the physicochemical properties of a range of compounds, are yet to be elucidated.

The aim of this study is to compare the long-term removal efficiency of 22 selected trace organic contaminants from a synthetic municipal wastewater by both MBR alone and direct addition of PAC into MBR (PAC-MBR). The addition of PAC into MBR was assessed as a potential tool to provide additional removal capacity for persistent trace organic contaminants. Special focus was given to the effect of the PAC dose and the physicochemical properties of the trace organic contaminants on their removal efficiency by the PAC-MBR system. In addition to information regarding removal from aqueous phase, the extent of adsorption of the target compounds onto sludge is also investigated.

2. Materials and methods

2.1. Model trace organic contaminants and synthetic wastewater

A set of 22 trace organic contaminants (Table 1 and Supplementary Data Table S1) representing four major groups, namely, PhACs, steroid hormones, pesticides and industrial chemicals and their metabolites were selected for investigation based on their widespread occurrence in raw sewage and sewage-impacted water bodies [1,2,4]. These contaminants have

been detected in domestic sewage at a concentration of up to a few $\mu\text{g/L}$ [1,2,4]. In this study, 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. A stock solution of all of the selected compounds was prepared in pure methanol at a concentration of 1 g/L each on a monthly basis and stored at $-18 \text{ }^\circ\text{C}$. This stock solution was then used to prepare the synthetic wastewater.

A synthetic wastewater comprising of glucose (400 mg/L), peptone (100 mg/L), urea (35 mg/L), KH_2PO_4 (17.5 mg/L), MgSO_4 (17.5 mg/L), FeSO_4 (10 mg/L) and sodium acetate (225 mg/L) was utilised [25]. The total organic carbon (TOC), total nitrogen (TN) and chemical oxygen demand (COD) of the wastewater were approximately 180 , 25 and 600 mg/L , respectively.

2.2. Laboratory scale MBR system and experimental protocol

A laboratory scale MBR system (Supplementary Data Fig. S2) consisting of a 4.5 L (active volume) glass reactor was employed. A hydrophilized PVDF hollow fibre membrane module (Mitsubishi Rayon Engineering, Japan) with a membrane nominal pore size of $0.4 \mu\text{m}$ and a total surface area of 0.074 m^2 was submerged into the reactor. As the main focus of this study was on trace organic contaminant removal performance, a stable hydraulic performance under minimised membrane fouling was highly desirable and, owing to the small reactor volume, any periodic in situ membrane backwashing would significantly dilute the reactor media. Therefore, the membrane was operated under a low average membrane flux such that significant membrane fouling, requiring periodic cleaning, could be avoided. The membrane was operated on a 14 min “suction” and 1 min “relaxation” cycle under an average flux of $2.7 \text{ L/m}^2 \text{ h}$, resulting in an HRT of 24 h . Notably, the flux and HRT used in this study are similar to those in the study of Lesage *et al.* [17]. The transmembrane pressure (TMP) was continuously monitored

Table 1
Physicochemical properties of the selected trace organic contaminants.^d

Category	Compound	CAS number	Chemical formula	Molecular weight (g/mol)	Log D (pH 7) ^a	Vapor pressure (mm Hg), at $25 \text{ }^\circ\text{C}$ ^a	Henry's law constant ($\text{atm} \times \text{m}^3/\text{mol}$) at $25 \text{ }^\circ\text{C}$ ^b	Limit of detection (ng/L) ^c
Pharmaceutical and personal care products	Ibuprofen	5687-27-1	$\text{C}_{13}\text{H}_{18}\text{O}_2$	206.28	0.94	$1.39\text{E}-4$	$1.50\text{E}-07$	20
	Acetaminophen	103-90-2	$\text{C}_8\text{H}_9\text{NO}_2$	151.16	0.47	$1.43\text{E}-6$	$6.42\text{E}-13$	20
	Naproxen	22204-53-1	$\text{C}_{14}\text{H}_{14}\text{O}_3$	230.26	0.73	$3.01\text{E}-7$	$3.39\text{E}-10$	1
	Ketoprofen	22071-15-4	$\text{C}_{16}\text{H}_{14}\text{O}_3$	254.28	0.19	$3.32\text{E}-8$	$2.12\text{E}-11$	20
	Diclofenac	15307-86-5	$\text{C}_{14}\text{H}_{11}\text{Cl}_2\text{NO}_2$	296.15	1.77	$1.59\text{E}-7$	$4.73\text{E}-12$	5
	Primidone	125-33-7	$\text{C}_{12}\text{H}_{14}\text{N}_2\text{O}_2$	218.25	0.83	$6.08\text{E}-11$	$1.94\text{E}-10$	10
	Carbamazepine	298-46-4	$\text{C}_{15}\text{H}_{12}\text{N}_2\text{O}$	236.27	1.89	$5.78\text{E}-7$	$1.08\text{E}-10$	10
	Salicylic acid	69-72-7	$\text{C}_7\text{H}_6\text{O}_3$	138.12	-1.13	$4.45\text{E}-5$	$7.34\text{E}-09$	1
	Metronidazole	443-48-1	$\text{C}_6\text{H}_9\text{N}_3\text{O}_3$	171.15	-0.14	$2.67\text{E}-7$	$1.69\text{E}-11$	20
	Gemfibrozil	25812-30-0	$\text{C}_{15}\text{H}_{22}\text{O}_3$	250.33	2.07	$6.13\text{E}-7$	na	1
	Triclosan	3380-34-5	$\text{C}_{12}\text{H}_7\text{Cl}_3\text{O}_2$	289.54	5.28	$3.36\text{E}-5$	$4.99\text{E}-09$	1
Pesticide	Fenoprop	93-72-1	$\text{C}_9\text{H}_7\text{Cl}_3\text{O}_3$	269.51	-0.13	$2.13\text{E}-6$	$9.06\text{E}-09$	20
	Pentachloro-phenol	87-86-5	$\text{C}_6\text{HCl}_5\text{O}$	266.34	2.58	$3.49\text{E}-4$	$2.45\text{E}-08$	1
Industrial chemicals and their metabolites	4-tert-butylphenol	98-54-4	$\text{C}_{10}\text{H}_{14}\text{O}$	150.22	3.40	0.0361	$1.19\text{E}-06$	1
	4-tert-octylphenol	140-66-9	$\text{C}_{14}\text{H}_{22}\text{O}$	206.32	5.18	$1.98\text{E}-3$	$6.89\text{E}-06$	1
	4-n-nonylphenol	104-40-5	$\text{C}_{15}\text{H}_{24}\text{O}$	220.35	6.14	$8.53\text{E}-5$	$1.10\text{E}-06$	10
	Bisphenol A	80-05-7	$\text{C}_{15}\text{H}_{16}\text{O}_2$	228.29	3.64	$5.34\text{E}-7$	$1.00\text{E}-11$	1
Steroid hormones	Estrone	53-16-7	$\text{C}_{18}\text{H}_{22}\text{O}_2$	270.37	3.62	$1.54\text{E}-8$	$3.80\text{E}-10$	5
	17- β -estradiol	50-28-2	$\text{C}_{18}\text{H}_{24}\text{O}_2$	272.38	4.15	$9.82\text{E}-9$	$3.64\text{E}-11$	5
	17- β -estradiol-acetate	1743-60-8	$\text{C}_{20}\text{H}_{26}\text{O}_3$	314.42	5.11	$9.88\text{E}-9$	na	5
	17- α ethinylestradiol	57-63-6	$\text{C}_{20}\text{H}_{24}\text{O}_2$	269.40	4.11	$3.74\text{E}-9$	$7.94\text{E}-12$	10
	Estriol	50-27-1	$\text{C}_{18}\text{H}_{24}\text{O}_3$	288.38	2.53	$1.34\text{E}-9$	$1.33\text{E}-12$	10

na, data not available.

^a Source: SciFinder database (<https://scifinder.cas.org/scifinder/view/scifinder/scifinderExplore.jsf>).

^b Source: <http://chem.sis.nlm.nih.gov/chemidplus/>.

^c Limit of detection (LOD) of the compounds during GC-MS analysis as described in Section 2.3. LOD is defined as the concentration of an analyte giving a signal to noise (S/N) ratio greater than 3. The limit of reporting was determined using an S/N ratio of greater than 10.

^d Molecular structures are shown in Supplementary Data Table S1.

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