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Removals of pharmaceutical compounds from hospital wastewater in membrane bioreactor operated under short hydraulic retention time

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HIGHLIGHTS

- MBR operated at short HRT of 3 h was effective for pharmaceutical removals.
- Majority of pharmaceuticals was removed by adsorption onto colloidal particles.
- Biodegradation of DCF, IBP and GFZ was significant under short HRT.
- MLSS concentration and compound properties influenced pharmaceutical removals.
- Pharmaceutical degrading microorganisms were developed after short term operation.

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ABSTRACT

Pilot-scale membrane bioreactor (MBR) was operated at a short hydraulic retention time (HRT) of 3 h for the treatment of hospital wastewater. The removals of eleven pharmaceutical compounds in MBR operated at different mixed liquor suspended solids (MLSS) level were investigated during which nitrification degree was differed. The results experiments revealed the importance of immediate adsorption onto the colloidal particles in supernatant of MBR sludge and subsequently removed by membrane filtration for the recalcitrant pharmaceutical compounds. Nevertheless, the removals through biodegradation during short HRT were also found significant for some compounds. DGGE profile revealed the development of pharmaceutical degrading microorganisms in MBR.

1. Introduction

Pharmaceutical and personal care products (PPCPs) are recently identified as important emerging contaminants in wastewater as they can create significant adverse human health effect and environmental impact (Tran et al., 2009; Kasprzyk-Hordern., 2009; Hai et al., 2011; Verlicchi et al., 2012). Although pharmaceutical compounds were mostly designed to use and active at low concentration, their persistent characteristics could prolong their presence in the ecosystems (Grenni et al., 2013). The major sources of pharmaceutical compounds in urban water environment are municipal wastewater, commercial wastewater including those from hospitals and health care institutions and industrial wastewater. Pharmaceutical compounds discharging from hospitals and medical service are posing higher risk to public health (Verlicchi et al., 2012). Conventional wastewater treatment systems have limited capacity to remove majority of those PPCPs. Nevertheless, enhancement of their removals in membrane bioreactor (MBR) has been reported (Sipma et al., 2010). Beneficial effects of membrane separation include complete retention of biomass in the system allowing the operation under high biomass concentration and long sludge age condition. However, the removal efficiencies of PPCPs were also depending on the properties of PPCPs (Tadkaew et al., 2011). Previous researches were conducted to determine the influence of operating conditions such as sludge retention time (Huang and Lee, 2008), pH (Tadkaew et al., 2010) and temperature (Hai et al., 2011). As the MBR can be operated at much shorter hydraulic retention time (HRT) than conventional wastewater treatment system due to

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its higher biomass concentration, the removal efficiencies of PPCPs could deviate from those found in conventional systems. There is still limited information on the performance and mechanisms of PPCPs removal in MBR operated at short HRT. So, the objectives of this study were to investigate the removal efficiencies and fate of pharmaceutical compounds containing in hospital wastewater in MBR operated under real fluctuation of wastewater characteristics. The removal mechanisms of pharmaceutical compounds including adsorption and biodegradation were also assessed. Furthermore, the microbial community of pilot-scale MBR responsible for the biodegradation of those PPCPs was analyzed.

2. Materials and methods

2.1. Pilot-scale MBR set-up and operation

Pilot-scale MBR was installed at a hospital in Bangkok, Thailand. The influent wastewater was primarily treated by screening 10 mm prior to feeding to the MBR with 1.3 m^3 working volume. The MBR utilized 4 hollow fibre membrane modules (PVDF Sterapore SADF™, 0.4 μm pore size, 9.0 m^2 surface area in each). Intermittent suction (7 min on and 1 min off) was performed to maintain permeate flux and total flow rate at about 14 L m^{-2} h^{-1} or 500 L h^{-1} yielding HRT of 3 h in the bioreactor. Aeration was supplied at 340 L min^{-1} (equivalent to specific aeration demand per membrane area of 0.57 m^3 m^{-2} h^{-1}) to support aerobic microbial activities and membrane fouling control. The MBR was operated under 2 different conditions, i.e. 1st period from day 0–42 and 2nd period from day 43–76. The influent wastewater characteristics during both periods are shown in Table 1. During the 1st period, mixed liquor suspended solids (MLSS) in the MBR were allowed to increase freely during the operation. Subsequent operation (2nd period) were kept at a MLSS concentration of less than 13 g L^{-1} to promote nitrification in the bioreactor. Sludge wastage was only performed at the end of 1st period (day 42) and 2nd period (day 76) for MLSS concentration adjustment. According the amount of sludge withdrawn from the MBR at the end of each experimental period, theoretical solid retention time (SRT) of the system was calculated as 27.7 and 27.0 d for the 1st and 2nd periods respectively.

The chemical analysis of biochemical oxygen demand (BOD), chemical oxygen demand (COD), total kjeldahl nitrogen (TKN), ammonia nitrogen (NH_3–N), suspended solids (SS), mixed liquor suspended solids (MLSS) of all samples were analyzed according to Standard Methods for the Examination of Water and Wastewater (APHA, 2005). TOC and TN concentrations were determined using a TOC/TN-V_{CSH} analyser (Shimadzu, Japan). pH was measured by a Metrohm advanced pH/ion meter. DO was measured with a DO meter and particle size analysis were analysed using a Mastersizer 2000E (Malvern, UK).

2.2. Determination of pharmaceuticals compounds and their removal mechanisms

In this study, eleven pharmaceutical compounds presented in actual hospital wastewater were investigated for their removals in the MBR. They were diclofenac (DCF), sulfamethoxazole (SMX), trimethoprim (TMP), carbamazepine (CBZ), tramadol (TMD), naproxen (NPX), propanolol (PPL), ibuprofen (IBP), 17β-Estradiol (E2), triclosan (TCS), and gemfibrozil (GFZ). Physicochemical properties of the compounds are presented in Table 2. Their concentrations in the influent and effluent were regularly monitored. During the MBR operation, the pharmaceutical compounds remaining in the supernatant (separated by 7000 g centrifugation for 10 min) and sludge bounded fractions were analysed to determine the fate of their removals.

Batch experiments were performed to differentiate adsorption and biodegradation mechanisms in the removal of individual pharmaceutical compound by MBR sludge. The experiments were conducted using 250 mL stoppered conical flasks filled with 200 mL of the centrifuged sludge obtained from pilot-scale MBR and adjusted initial MLSS concentration to 1000 mg L^{-1} while the initial concentration of pharmaceutical compounds were set at 500 μg L^{-1}. The batch reactors were wrapped in aluminum foil to prevent possible photo-degradation and put on shaker at 125 rpm. The samples were taken at constant time intervals over 6 h to determine remaining the pharmaceutical compounds in dissolved and particle bound form.

Inactivated sludge was used for determining adsorption capacity of MBR sludge. The sludge samples obtained from pilot-scale MBR were inactivated three times by pasteurization at 121 °C for 15 min in order to terminate microbial activities. The batch experiments were carried out in the same manner with that of active sludge. The pharmaceutical compounds were analyzed in dissolved and particulate forms.

During batch experiment, the amount of pharmaceutical compounds adsorbed on sludge at equilibrium (q_e, mg g^{-1}) was calculated according to the following equation:

$$q_e = \frac{(C_0 - C_e)V}{X} \quad (1)$$

Where C_o and C_e (mg L^{-1}) are the initial and equilibrium concentration of pharmaceutical compounds in the liquid phase, respectively; V (L) is the volume of sludge solution, and X (g) is the mass of sludge.

Actual amount of pharmaceutical compounds adsorbed on MBR sludge during the reactor operation was determined by analysing adsorbed compounds at different time over the entire operation period. The accumulation of pharmaceutical compounds in MBR sludge at any time (q_t, mg g^{-1}) was calculated

Table 1
Influent characteristics and treatment performance of MBR.

Parameter	Day 0–42			Day 43–76		
	Inf.[a]	Eff.[a]	%R	Inf.[a]	Eff.[a]	%R
pH	6.7(0.2)	7.2(0.4)	–	6.8(0.2)	6.7(0.1)	–
EC	1962 (360)	1683 (218)	–	1808(659)	1830 (827)	–
SS (mg L^{-1})	90.3(39.7)	ND	100	68.6(9.7)	ND	100
BOD (mg L^{-1})	98.6(13.7)	5.3(5.6)	94.9	78.1(12.7)	2.1(0.6)	97.24
COD (mg L^{-1})	167.2(23.8)	53.1(29.7)	67.0	121.7(17.2)	35.5(23.9)	72.05
TOC (mg L^{-1})	52.3(7.7)	11.2(3.0)	78.6	30.2(4.9)	5.3(2.8)	82.50
TN (mg L^{-1})	42.7(6.4)	33.6(3.5)	21.3	35.5(2.3)	17.1(12.0)	51.90
TKN (mg L^{-1})	41.2(8.1)	26.8(14.5)	32.5	32.5(2.8)	1.1(1.4)	96.22
NH_3–N (mg L^{-1})	36.0(2.2)	24.1(13.7)	32.9	28.7(1.9)	0.5(0.4)	98.13

[a] Average (SD) values, no. of samples = 11, %R = Removal percentage.

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