Effect of hydraulic retention time on the performance of membrane bioreactor for pharmaceutical compound removal from hospital wastewater

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Abstract

Pharmaceutical compounds are a concern, as they are one of the most serious emerging environmental contaminants and hospital waste water is a major contributor as it discharges pharmaceutical compounds, via its liquid discharges, into municipal wastewater. Conventional waste water treatment systems are not designed to remove these compounds. Consequently, membrane bioreactors (MBRs) have been widely used to remove pharmaceutical compounds from hospital wastewater. This study investigated the pharmaceutical compound removal of a pilot-scale membrane bioreactor system operated under different hydraulic retention times (HRTs 3 and 6 h) for hospital wastewater treatment. Two pharmaceutical compounds, gemfibrozil (GFZ) and trimethoprim (TMP) were selected to be monitored in this study. The MBRs performance provided better removal of organic matters (BOD and COD > 90%) under operating conditions with lower hydraulic retention time. The results showed that prolonged HRT conditions, operated with complete nitrification, achieved greater pharmaceutical compound removal. Removal efficiencies of GFZ, when the HRT was increased from 3 to 6 h, were 10.80%, and 90.10% respectively, whereas the removal efficiency of TMP was found to be slightly increased from 40.79% (HRT 3 h) to 50.23% (HRT 6 h). GFZ tended to be removed by degradation, this was in fact confirmed by using the batch experiment. TMP was found to be adsorbed under an HRT of 3 h and had improved biodegradation after increasing the HRT to 6 h. Optimization of the operating conditions could increase the removal efficiency of pharmaceutical compounds.

Keywords: pharmaceutical compounds, hydraulic retention time (HRT), membrane bioreactor

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Introduction

Pharmaceutical and personal care products are of concern as they are an emerging contaminant due to their high usage and continuous discharge into water bodies and the environment as parental compounds and metabolites (Barretto et al., 2018). These products have been extensively reported as potentially harmful to the living ecosystem due to the effect of PPCPs on aquatic organisms and human health. In many countries, wastewater from domestic, industry and hospitals are directly discharged into water bodies without any prior treatment which can generate pollution in the environment (Oros-Ruiz, Zanella, & Prado, 2013). Hospital wastewater represents a significant source of pollution, discharging considerable amounts of pharmaceutical compounds into wastewater treatment plants (WWTPs) (Verlicchi, Galletti, Petrovic, & Barceló, 2010).

Gemfibrozil (GFZ) is a lipid regulator that belongs to the class of fibrate drugs. Due to its widely used, this compound is incompletely removed from wastewater during treatment and it can be found in the influent, effluent and freshwater at concentration ranging from 0.07 to 0.51 µg/L (Zhao et al., 2010; Fang, Karnjanapiboonwong, Chase, Wang, Morse, & Anderson, 2012), as well as in surface waters ranging from 11 to 187 ng/L (Patrolecco, Capri, & Ademollo, 2015; Huerta et al., 2016; Osorio, Larrañaga, Aceña, Pérez, & Barceló, 2016; Andres-Costa, Carmona, & Picó, 2016; Tlili et al., 2016). Hence the presence of GFZ may pose potential adverse effects on non-target organisms (Fang, Karnjanapiboonwong, Chase, Wang, Morse, & Anderson, 2012). Moreover, GFZ was the main contributor to toxic units (TU, calculated as the ratio between the detected concentration and the EC50 value) for fish and the second largest contributor to crustaceans in Iberian rivers (Osorio, Larrañaga, Aceña, Pérez, & Barceló, 2016). It is necessary to improve the elimination efficiency of GBZ by using water treatment techniques.

Trimethoprim is an antibiotic, usually combined with sulfamethoxazole, used to treat various types of bacterial infections, including urinary tract infections, acute worsening of chronic bronchitis and ear infections (Oros-Ruiz, Zanella, & Prado, 2013). As reported by the World Health Organization, TMP is one of the most important medicines for basic health and can effectively treat and prevent respiratory or gastrointestinal tract infections in cattle, swine and poultry (de Paula, de Pietro, & Cass, 2008; Banrnes et al., 2013). Because TMP is highly effective and cheap, an abundance of TMP is produced and used every year. Trimethoprim was detected in surface water at 17 ng/L in the Mekong Delta in Vietnam (Nguyen Dang Giang, Sebesvari, Renaud, Rosendahl, Hoang Minh, & Amelung, 2015) whereas a high level at 28,000 ng/L of trimethoprim, was measured

in a river downstream of a pharmaceutical formulation facility in Pakistan (Khan, Berglund, Khan, Lindgren, & Fick, 2013). Because only 40% of a given dose can be digested by humans (Pérez, Eichhorn, & Aga, 2005), the residual is discharged into sewage (Siemens, Huschek, Siebe, & Kaupenjohann, 2008). However, the removal of TMP by wastewater treatment plants has been reported as less than 10% (Luo, Zheng, Greaves, Cooper, & Song, 2012). After long-term exposure to TMP, bacteria may generate a resistance resulting in TMP becoming ineffective in treating illnesses (Wang & Wang, 2015). TMP has been detected in surface water and effluents of wastewater treatment plants at nano and microgram per litre levels, this poses a potential threat to the aquatic environment (Batt, Kim, & Aga, 2006). Therefore, removal of TMP from water and wastewater is an essential problem that is required to be managed. Therefore, it is necessary to improve the elimination efficiency of GBZ and TMP using water treatment techniques.

Li, Cabassud, & Guigui (2015a); Huang et al. (2018) reported that pharmaceutical compounds have been detected in domestic wastewater treatment plants around the world. Conventional wastewater treatment systems are not designed to treat these compounds, only organic matters (Kimura, Hara, & Watanabe, 2007). Thus, effluent from wastewater treatment plants has been specified as a major source of discharge of pharmaceutical compounds into the environment (Kimura, Hara, & Watanabe, 2007; Ba, Jones, & Cabana, 2014). Various physico-chemical and biological treatment technologies were performed for the removal of micropollutants from wastewater. Physico-chemical methods including coagulation-flocculation, adsorption, and advanced oxidation processes are reported to perform sufficiently well in treating micropollutants. Wang, & Wang (2016) described that the combination of advanced oxidation processes and biological treatment technology has shown good removal efficiency for pharmaceutical compounds and their by-products formed during the advanced oxidation process. Therefore, the combined process of advanced oxidation and biological methods could be an option for replacement of the disadvantaged single biological treatment or advanced oxidation treatment. More studies are needed to determine the optimum conditions for each unit. For different compositions of pharmaceutical compounds, different parameters such as hydraulic retention time, pH, reactor configuration, temperature, redox conditions and the concentration of the catalyst are required to set the combination treatment process to achieve optimal performance, in terms of both removal efficiency and economical cost. Membrane bioreactors (MBRs) comprise of biodegradation and membrane filtration, which have demonstrated an improvement over conventional activated sludge

(CAS) processes for wastewater treatment (Kim, Guerra, Shah, Parsa, Alaee, & Smyth, 2014; Phan et al., 2014; Shi et al., 2018). MBRs have several advantages including a small footprint, high sludge retention time (SRT), lower sludge production, high effluent quality, flexible process design and higher removal of nutrients, organic and emerging contaminants (Nguyen, Hai, Kang, Price, Nghiem, 2013; Phan et al., 2014; Ganiyu, van Hullebusch, Cretin, Esposito, & Oturan, 2015; Li, Cabassud, & Guig, 2015b). The high sludge concentration in an MBR is not only beneficial for the biodegradation of pharmaceuticals but is also presumed to have a beneficial effect on the removal efficiency of micro-pollutants that tend to accumulate in the sludge, either due to their intrinsic hydrophobicity or via electrostatic interactions with the biomass (Sipma et al., 2010). In Thailand, have been used MBRs for treating domestic wastewater (Tiranuntakul, 2012) and seafood industry (Choksuchart Sridang, Lobos, & Kaiman, 2012). However, the application of membrane bioreactors in hospital wastewater treatment has not been reported in literature. So, the objective of this study was to investigate the removal efficiencies and fate of pharmaceutical compounds contained in hospital wastewater in an MBR operated under real fluctuating wastewater characteristics with lower hydraulic retention times. The removal mechanisms of the pharmaceutical compounds, including adsorption and biodegradation, were also assessed.

Methodology

1. The pilot-scale MBR set-up and operation

A pilot-scale MBR (Figure 1) was installed at a hospital in Bangkok, Thailand. The influent wastewater was primarily treated by screening, using a 10 mm screen, prior to feeding it into the MBR having a 1.3 m^3 working volume. The MBR utilized four hollow fibre membrane modules (PVDF Sterapore SADF^{TM}, 0.4 µm pore size, each having a 9.0 m^2 surface area). Intermittent suction, 7 mins on and 1 min off, was performed to maintain the permeate flux and a total flow rate of approximately 14 L/m^2/h or 500 L/h and 7 L/m^2/h or 250 L/h, yielding HRTs of 3 h and 6 h respectively in the bioreactor. Aeration was supplied at 20.4 m^3/h to support aerobic microbial activities and membrane fouling control. Operation of the MBR was performed over two periods, i.e. 1st period (HRT 3 h) from day 0-76 and a 2nd period (HRT 6 h) from day 77-126. During the first period, mixed liquor suspended solids (MLSS) in the MBR could increase freely during the operation without any sludge wastage. Subsequent operations were operated at a controlled MLSS concentration of less than 12 g/L to promote nitrification in the bioreactor. Throughout the second period, mixed liquor suspended solids (MLSS) in the MBR were controlled to maintain the MLSS concentration at about 12 g/L during the operation. Chemical analysis of the 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 the samples was carried out in accordance with the Standard Methods for the Examination of Water and Wastewater (APHA, 2005), pH was measured by using a Metrohm Advanced pH/Ion meter and DO was measured by using a DO meter.

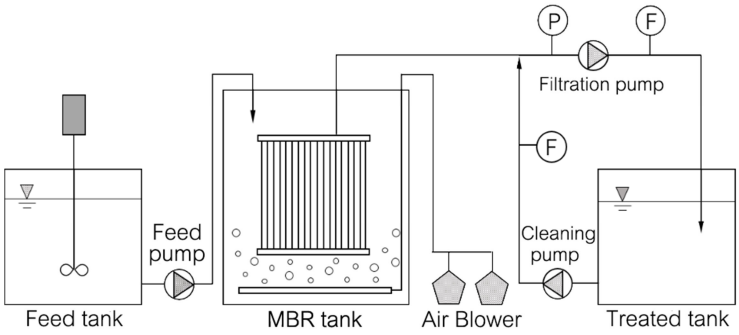


Figure 1 Schematic diagram of the MBR system.

2. Determination of pharmaceuticals compounds and their removal mechanisms

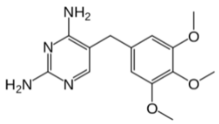
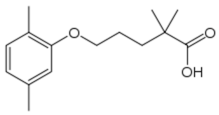
In this study, two pharmaceutical compounds were selected to represent the major compounds detected in hospital wastewater, namely Trimethoprim (TMP) and Gemfibrozil (GFZ). The selection of these pharmaceutical compounds was based on their occurrence and their physico-chemical properties, presented in (Table 1). This study focused on their removal mechanisms, by using batch experiments performed to determine the combined removal of pharmaceutical compounds via adsorption and biodegradation mechanisms using MBR sludge. The solubility of the pharmaceutical pollutants was determined by their octanol-water partition coefficient (Log K_{ow}), which is a measure of hydrophobicity. Rule of thumb on Log K_{ow} values of pharmaceutical pollutant was applied for estimating the sorption of pharmaceutical pollutant in the sludge. Compounds with high log K_{ow} > 4 and high molecular weight tend to be more sorbed than compounds with low log K_{ow} < 2.5. Sorption of most of the pharmaceutical compounds is on sludge. Moderate hydrophilic compounds with 2.5 <Log K_{ow}< 4 were found to have variable mechanisms dependent on the physicochemical properties of the compounds.

2.1 Biodegradation experiment

The biodegradation batch experiment arrangement consisted of two batch reactors (250 ml stoppered conical flasks) filled with 200 ml (1000

mg/L) of the pilot-scale MBR sludge and the initial concentration of pharmaceutical compounds was set at 500 µg/L. The batch reactors were wrapped in aluminium foil, to prevent possible photodegradation, and put on a shaker at 125 rpm. Samples were taken at uniform time intervals, over six hours, to determine the remaining pharmaceutical compounds in dissolved and particle-bound form.

Table 1 Chemical properties of the major pharmaceuticals found in hospital wastewater.

compounds	CAS no.	formula	molecular weight	Log K_{ow}
Trimethoprim 	738-70-5	$C_{14}H_{18}N_4O_3$	290. 30	0.91
Gemfibrozil 	25812-30-0	$C_{15}H_{22}O_3$	250.34	4.77

2.2 Adsorption experiment

To determine the adsorption removal mechanisms alone, inactivated sludge was used for determining the adsorption capacity of the MBR sludge. The sludge samples obtained from the pilot-scale MBR were inactivated by sterilization at 121 °C for 15 min over 3 cycles in order to terminate the microbial activity using the methods described in Boonyaroj, Chiemchaisri, Chiemchaisri, & Yamamoto (2017). Procedures were similar to the biodegradation experiment, which was performed using inactivated sludge and selected pharmaceutical compounds were analyzed in dissolved and particulate forms. Adsorption experiments were performed comparably to the biodegradation experiments.

3. Pharmaceutical compounds analyses

Influent, effluent and mixed liquor suspended solids in the bioreactor were collected and transferred to the laboratory. The concentration of the pharmaceutical compounds was analysed separately, in solid and soluble forms. The analytical methods described in Tadkaew, Hai, MaDonald, Khan, & Nghiem (2011) were used with some modification. Samples were extracted by solid phase extraction by using an Oasis HLB 3 cc cartridge (Waters, Millford, MA, USA). The cartridge was preconditioned with 3 mL of MTBE, 3 mL of methanol and 3 mL of Milli Q water, then the sample was loaded into the cartridge at 15 mL/min, followed by rinsing with 3 mL of Milli Q water and finally dried with a stream of nitrogen

for 30 min. The samples, for analyses, were eluted from the cartridges with 3 mL of methanol followed by 3 mL of 1:9 (v/v) methanol: MTBE and loaded into centrifuge tubes. The extracts were evaporated with nitrogen to approximately 100 µL and adjust to a final volume of 1.5 mL with methanol. Analyses were conducted by using LC-MS/MS (Varian, Inc.) equipped with a Pursuit XRs C18 column (diameter, length and pore size of 4.6 mm, 150 mm and 5 µm, respectively) During the MBR operation, the remaining pharmaceutical compounds in the supernatant (separated by 3000g centrifugation), sludge bound and effluent of the MBR were analyzed to determine the fate of their removal. HPLC (Shimadzu, Japan) with UV detection was used to determine the pharmaceutical compounds in all the batch experiment samples. A GL science Inc. C-18 column (with diameter, length and pore size of 4.6 mm, 250 mm and 5 µm, respectively) was used for separation. The mobile phase was Milli Q grade deionised water, buffered with 25 mM KH_2PO_4, and acetonitrile which was delivered at 1.0 mL/min. Detection wavelengths for this study were set at 230 nm, except for Sulfamethoxazole which was set at 280 nm. Calibration generally yielded standard curves with coefficients of determination (R^2) greater than 0.98 within the range of experimental concentrations used. The analysis was carried out immediately upon the conclusion of each experiment. A sample injection volume of 20 µL was used, and the quantification limit for all the analytes under investigation using these conditions was approximately 10 µg/L.

Results and discussion

1. Treatment performance of MBR

This research utilized the application of MBR technology for the treatment of hospital wastewater, which was operated continuously for 126 days, to investigate the characteristics of hospital wastewater and determine appropriate operating conditions for removing the toxic and emerging pollutants. The performance of the pilot-scale MBR during operation at different HRT is shown in (Table 2). The characteristics of the wastewater were similar to municipal wastewater, but also contained some toxic compounds released from hospital activities. The 1^{st} period was operated under an HRT of 3 h for 76 days, after that the 2^{nd} period was operated at an HRT of 6 h to investigate the appropriate operating conditions for the treatment of hospital wastewater. At an HRT of 3 h, the average BOD and COD removal efficiencies 3 h were 96.06% and 69.27%, respectively. Influent concentrations of NH_3-N and TKN were found to be 32.69 mg/L and 37.23 mg/L, whereas high ammonia concentrations of 21.19 mg/L and 21.34 mg/L were detected in the effluent. Thus, the average NH_3-N and TKN during the operation were 37.50% and 44.53% respectively. Therefore, operating

under lower HRT could not provide better removal efficiencies of nitrogen. The 2nd period was operated under an HRT of 6 h, day 77-126. The average incoming BOD and COD concentrations were 76.16 mg/L and 133.09 mg/L respectively, while the removal efficiencies of BOD and COD were found to be 97.75% and 97.10% respectively. Meanwhile, NH_3-N and TKN removal efficiencies were higher than 96% during stable operation, which indicates complete nitrification since nitrate was found to be the major nitrogen form in the effluent. The main difference in the performance between 3 h and 6 h was the degree of nitrification as indicated by the NH_3-N and TKN concentrations.

Table 2 Wastewater characteristics and removal efficiency of the MBRs during operation.

parameter	HRT 3 h			HRT 6 h		
	influent	effluent	removal efficiency (%)	influent	effluent	removal efficiency (%)
pH	6.84 (0.22)	6.99 (0.15)	-	7.33 (0.16)	7.19 (0.50)	-
SS (mg/L)	80.45 (30.91)	N.D.	100.00	97.86 (46.44)	N.D.	100.00
BOD (mg/L)	98.57 (16.45)	3.69 (4.09)	96.06	76.16 (19.20)	1.41 (1.72)	97.75
COD (mg/L)	167.16 (31.10)	45.12 (27.43)	69.27	133.09 (17.18)	5.01 (7.85)	97.10
NH_3-N (mg/L)	32.69 (4.31)	21.19 (4.31)	37.50	26.94 (4.85)	0.56 (0.87)	97.93
TKN (mg/L)	37.23 (7.52)	21.34 (12.58)	44.53	27.68 (6.26)	0.67 (0.84)	96.65
NO_2-N (mg/L)	0.41 (0.23)	2.04 (0.10)	-	0.04 (0.03)	1.34 (1.09)	-
NO_3-N (mg/L)	0.38 (0.47)	0.52 (0.71)	-	0.58 (0.30)	19.89 (3.09)	-

ND: Not detected; *Average (SD) values, no. of samples = 17

(Figure 2) shows the variation in BOD, COD, SS, and NH_3-N removal by the pilot-scale MBR during the operation. During the operation, the wastewater characteristics varied depending on water consumption and personnel used. The BOD/COD ratio was found to be between 0.41-0.78, this could be explained by Typical values for the ratio of BOD/COD for untreated municipal wastewater which are in the range of 0.30 to 0.80. If the BOD/COD ratio for untreated wastewater is 0.50 or greater, the wastewater is considered to be easily treatable by biological means. If the ratio is below about 0.30, either the wastewater may have some toxic components or acclimated micro-organisms may be required in its stabilization. Hence, this hospital wastewater characteristics demonstrated it could be treated by MBR. During the 1st period of operation, the DO concentration in the bioreactor was an important parameter in the aerobic treatment process. Initially, the DO was maintained at higher than 4 mg/L but became lower when the MLSS concentration was increased during the operation

without sludge wastage. After MLSS concentration adjustment in the 2nd period, the DO level could be maintained for satisfactory microbial activities. The DO concentration in the bioreactor represents the amount of residual oxygen available to the microbial community (Tadkaew, Sivakumar, Khan, Mcdonal, & Nghiem, 2010). The pH level of both HRTs showed slightly different pH levels of the influent in the 1st period and 2nd period, which was found to be 6.84 and 7.33, respectively, whereas the pH levels were 6.99 and 7.19, respectively, in the effluent, which indicated that the microbial could be active in the bioreactor.

At an HRT of 3 h, the MLSS was increased from 9.1 to 18 g/L (Figure 3) during the 42 days without sludge wastage. As a result of the MLSS increase, the DO level in the MBR decreased from 4-5 mg/L to <1 mg/L resulting in high NH_3-N and TKN concentrations in the effluent. Subsequent operation was performed keeping the MLSS between 6 and 13 g/L during day 43-76, thus the DO level was improved to 4-5 mg/L. From these results, the MBR operated under an HRT of 3 h could provide high organic removal efficiencies of hospital wastewater by controlling the MLSS concentration in the range of 6 to 13 g/L to promote aerobic condition in the MBR. When increasing the HRT to 6 h, the MLSS concentration was controlled to be less than 12 g/L, which provided a nitrogen level in the treated water to be lower than the previous operation. This operation helped improve nitrification in the MBR resulting in low effluent NH_3-N and TKN concentrations. The result indicated that the MBR could be effective for organics and nitrogen control at short HRT provided that the MLSS was properly controlled.

In order to investigate the fouling behaviour, the change of transmembrane pressure (TMP) was monitored at HRT 3 h and 6 h. The TMP profiles during the operation are shown in (Figure 3). The TMP variations in the pilot-scale MBR during the operation are described into three step fouling phenomenon i.e. gradual and slow rises in TMP, then an initial short-term rapid rise followed by a rapidly TMP jump to 30 kPa. The 1st period (HRT 3h), fouling period, was 42 days after cleaning the membrane, fouling was again found after 76 days. When the MLSS concentration was set at 6 g/L, the TMP slightly increased in the initial stage. The TMP increased rapidly from 23 kPa to 32 kPa when the MLSS concentration was higher than 13 g/L, which explained that when the MLSS concentration was higher 13 mg/L, it could be a factor causing the TMP jump. After operating at an HRT of 3 h, the HRT experiment was changed from 3 to 6 h to observe the pattern of membrane fouling in the 2nd period, by controlling the MLSS concentration not to exceed 13 g/L. It was found that the membrane was fouled after 126 days.

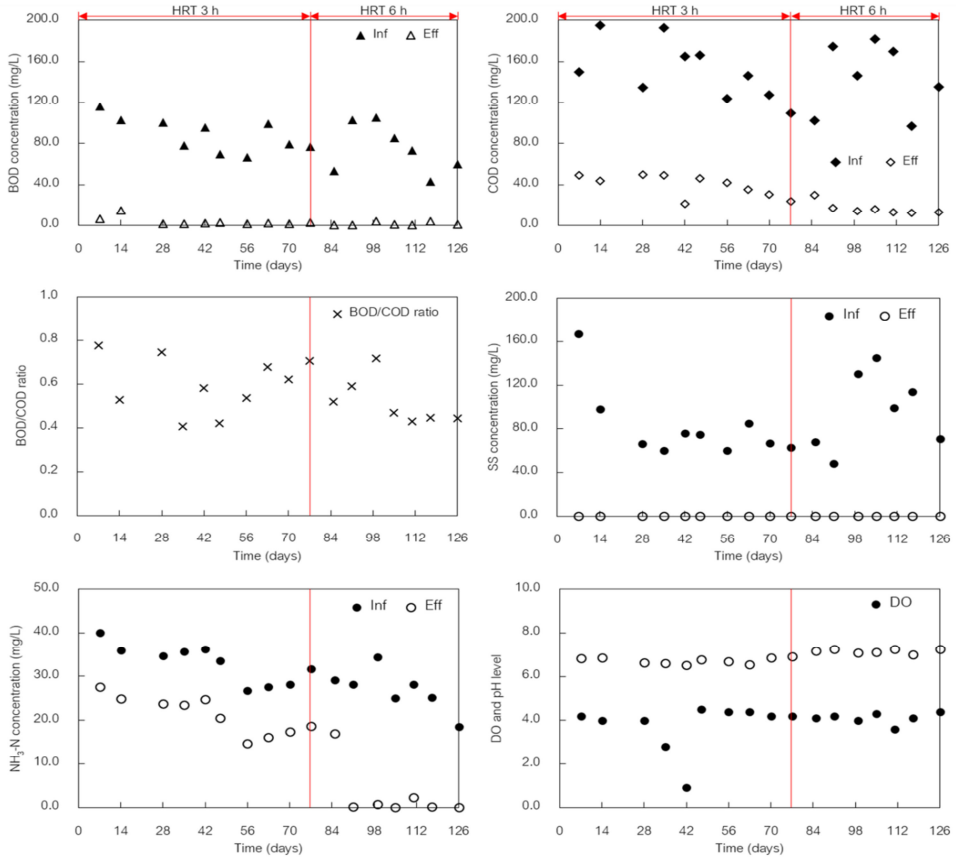


Figure 2 shows the variation in BOD, COD, SS, and NH_3-N removal by the pilot-scale MBR during operation.

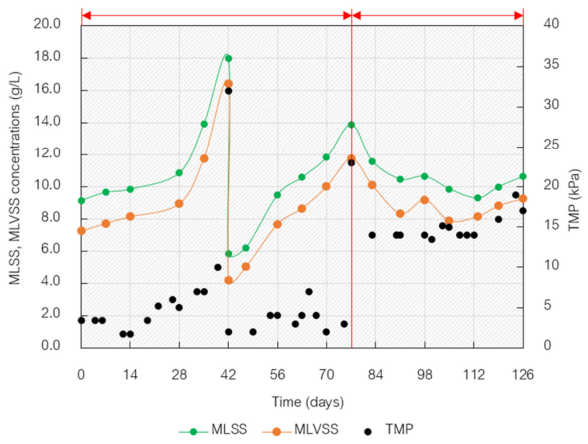


Figure 3 Sludge concentrations and transmembrane pressure (TMP) profile.

2. Removal mechanisms of pharmaceutical compounds

The pharmaceutical compound concentrations and removal efficiency of the pilot-scale MBR during operation are shown in (Table 3). The concentration of the influent pharmaceutical compounds was detected at a few µg/L up to nearly mg/L level. TMP was found to be moderately removed in the water phase during both operations (40.79% and 50.23% respectively), whereas GFZ was poorly removed during operation at an HRT of 3 h (10.80%), after increasing the HRT to 6 h, the removal efficiency of GFZ was found to be improved up to 90.10%. This may demonstrate that the removal efficiency of TMP and GFZ can be improved by extending their contact time with the sludge. Tambosi et al. (2010) reported that the removal efficiency of trimethoprim under operating an HRT at 13 h (94%) was higher than operating under an HRT of 9 h (84%). In a previous study, long term operation of an MBR under prolonged SRT conditions was reported to be associated with some operational problems, including inefficient mixing and increased aeration demand for the biological metabolism and membrane cleaning.

Table 3 Removal of pharmaceuticals during MBR operations.

compounds	HRT 3 h			HRT 6 h		
	influent (µg/L)	effluent (µg/L)	removal (%)	influent (µg/L)	effluent (µg/L)	removal (%)
TMP	N.D.-265.59	4.25-198.00	40.79	23.60-68.58	18.22-31.96	50.23
GFZ	6.09-88.01	N.D.-107.00	10.80	87.71-844.86	17.87-37.76	90.10

Batch experiments were carried out, using mixed liquor obtained from the MBR, to investigate their adsorption capacities, and the biodegradation of individual pharmaceutical compounds. As demonstrated in (Figure 4), the removal of TMP and GFZ via adsorption and biodegradation were clearly different. While the removal of TMP was mainly due to adsorption, the removal of GFZ was mainly from biodegradation. In this study, GFZ was found to be removed via adsorption (34.11%) and biodegradation (32.77%), these mechanisms accounted for 66.88% removal in the batch experiment. Whereas, TMP was found to be removed via adsorption (7.29%) and biodegradation (19.19%) giving a total removal of 26.48%. After changing the HRT to 6 h, GFZ was found to be removed via adsorption (25.52%) and biodegradation (33.60%) giving a total removal of 59.12%. TMP was found to be removed via adsorption (38.95%) and biodegradation (19.17%) with total removal of 58.12%. Nevertheless, observed removal of TMP was found to be better than GFZ during operation at an HRT of 3 h, after increasing the HRT to 6 h GFZ showed better removed than TMP. Essentially,

the high removal efficiencies of GFZ in biological wastewater treatment processes could also be explained by the sole presence of electron donating groups in their molecules, e.g. methyl (— CH_3) and alkoxy (—O—CH_2—CH_2—CH_2—C[CH_3]$_2$—COOH) groups in GFZ. Related with Kjeldal, Zhou, Wissenbach, von Bergen, Gough, & Nielsen (2016), who found that biodegradation of GFZ started at the methyl group on its aromatic ring via the hydroxylation route and biodegradation of GFZ in activated sludge processes could also be contributed by hydrophobic interactions between these micropollutants with biomass/suspended solids in the sludge system because these compounds have a relatively high octanol-water distribution coefficient (log K_{ow} 4.77) under the environmental pH range of 6-8. Tiwari, Sellamuthu, Ouarda, Drogui, Tyagi, & Buelna (2017) reported that pharmaceutical pollutants with high sorption potential have a higher removal rate than the compounds with low sorption potential. Prasertkulsak, Chiemchaisri, Chiemchaisri, Itonaga, & Yamamoto (2016) discovered that the electrical charge of compounds was found to affect their removal efficiencies. The negatively-charged compounds such as diclofenac, naproxen, ibuprofen, and gemfibrozil were found to be removed better than neutral compounds, i.e. sulfamethoxazole and trimethoprim. Hence, in this study, it could be explained that the removal mechanisms of TMP and GFZ by TMP was hydrophilic, in which the compounds could not be well adsorbed onto sludge. These hydrophilicity compounds were found to have moderated removal when operating at short MBR HRTs. Meanwhile, GFZ, a hydrophobicity compound, could be adsorbed onto sludge better than TMP, then it could have greater adsorption and better biodegradation in the system. Nevertheless, when considering the removal of pharmaceutical compounds together with their chemical properties, it was generally revealed that most of the hydrophobic compounds were initially removed through adsorption into the sludge or colloidal particles in relatively short contact time (less than 3 h). For the latter case, their removal from the system through a sludge withdrawal strategy to control appropriate solid retention time (SRT) in the system would be necessary.

Conclusion

Pharmaceutical compounds contained in hospital wastewater could be effectively removed in MBRs operated at HRTs of 3 h and 6 h. Accumulation of compounds via adsorption onto colloidal in the supernatant which are subsequently separated by membrane filtration were significant. Their removal via adsorption and biodegradation were varied depending on the compound's properties. The sludge in the MBR could increase the adsorption capacity by increasing the MLSS concentration. HRTs of 3 h and 6 h could be operated efficiently to prevent membrane fouling by controlling the MLSS concentration properly and maintaining nitrification in the system.

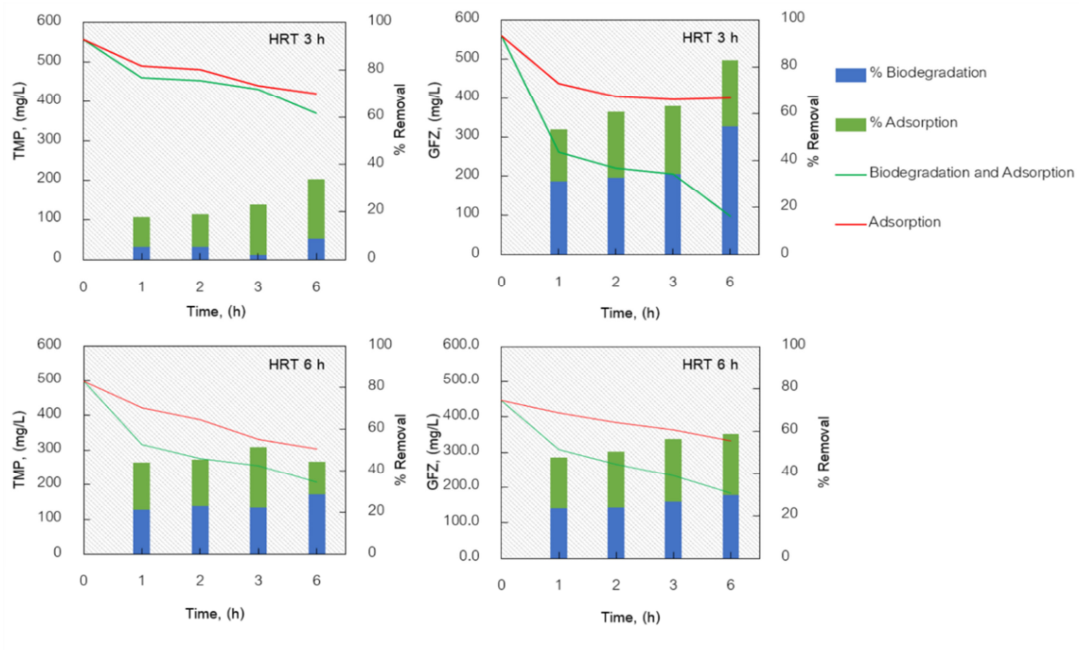


Figure 4 Removal mechanisms of pharmaceutical compounds: Trimethoprim and Gemfibrozil.

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