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Short Communication

Effect of hydraulic retention time on the performance of a hybrid moving bed biofilm reactor-membrane bioreactor system for micropollutants removal from municipal wastewater

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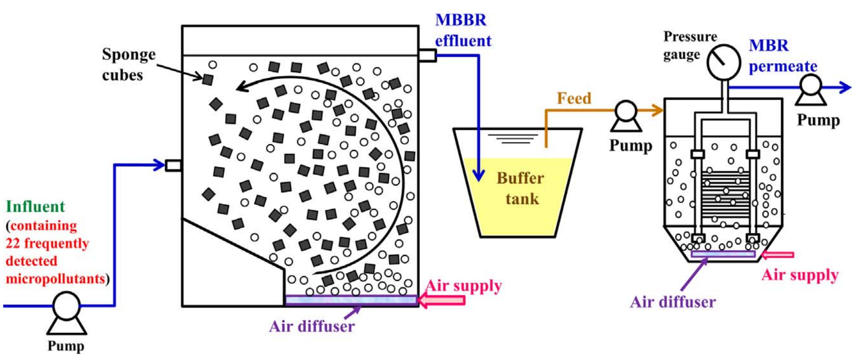
GRAPHICAL ABSTRACT



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ABSTRACT

This study evaluated micropollutants removal and membrane fouling behaviour of a hybrid moving bed biofilm reactor-membrane bioreactor (MBBR-MBR) system at four different hydraulic retention times (HRTs) (24, 18, 12 and 6 h). The results revealed that HRT of 18 h was the optimal condition regarding the removal of most selected micropollutants. As the primary removal mechanism in the hybrid system was biodegradation, the attached growth pattern was desirable for enriching slow growing bacteria and developing a diversity of biocoenosis. Thus, the efficient removal of micropollutants was obtained. In terms of membrane fouling propensity analysis, a longer HRT (e.g. HRTs of 24 and 18 h) could significantly mitigate membrane fouling when compared with the shortest HRT of 6 h. Hence, enhanced system performance could be achieved when the MBBR-MBR system was operated at HRT of 18 h.

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1. Introduction

Although the frequently detected micropollutants are at trace concentrations in our aquatic environment, they may cause long-term adverse effects, such as bioaccumulation and carcinogenicity (Klavarioti et al., 2009; Soares et al., 2008). Membrane bioreactors (MBRs), as a promising biological treatment technology, have been widely used to remove micropollutants (Tadkaew et al., 2011; Trinh et al., 2012; Wijekoon et al., 2013). However, two major concerns remain during the application of MBR systems, including membrane fouling, which could be alleviated by frequent physical and/or chemical backwashing or relaxation, and inconsistent removal of some polar and persistent hydrophilic micropollutants even at long hydraulic retention time (HRT).

Combination of moving bed biofilm reactor (MBBR) with MBR in series (MBBR-MBR) is one of recent advances in improving MBR performance, due to its potential for reducing membrane fouling and prolonging filtration duration as well as the possibility of prompting microbial degradation of certain organic compounds (Luo et al., 2015; Sombatsompop et al., 2006). It has been shown that the prolonged HRT decreased membrane fouling propensity and enhanced the formation of a diverse biocoenosis and biodegradation efficiency for certain micropollutants (De Gusseme et al., 2009; Schröder et al., 2012). Nevertheless, a previous research (Chen et al., 2008) concluded that the changes in HRT (HRTs of 8, 6, and 4 h) had little effect on removal efficiency of Bisphenol A in a lab-scale submerged MBR, probably due to its rapid biodegradation and low potential for bioaccumulation.

Up to now, there has been no comprehensive study regarding the impact of HRT on micropollutants removal. Our previous investigation indicated that MBBR as pretreatment to MBR exhibited better performance in removing micropollutants than single conventional MBR (CMBR), and more importantly MBBR could mitigate membrane fouling of MBR to a great extent (Luo et al., 2015). Therefore, in this study, the performance of MBBR-MBR hybrid system was investigated at four different HRTs (24 h, 18 h, 12 h, and 6 h) to determine the optimal HRT in terms of micropollutants removal and membrane fouling control. Soluble microbial products (SMP), extracellular polymeric substances (EPS), zeta potential, and relative hydrophobicity (RH) were used to examine the membrane fouling propensity.

2. Materials and methods

In this study, a set of 22 frequently detected micropollutants were selected to represent 11 pharmaceutical and personal care products (ibuprofen, acetaminophen, naproxen, ketoprofen, diclofenac, primidone, carbamazepine, salicylic acid, metronidazole, gemfibrozil and triclosan), 5 steroid hormones (estrone, 17-β-estradiol, β-estradiol 17-acetate, 17-α ethinylestradiol and estriol), 4 industrial chemicals (4-*tert*-butylphenol, bisphenol A, 4-*tert*-octylphenol and 4-n-nonylphenol) and 2 pesticides (fenoprop and pentachloro-phenol). The preparation of synthetic wastewater (containing micropollutants), characteristics of sponge carrier and analysis methods (e.g. concentrations of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS)) could refer to our previous study (Luo et al., 2014, 2015). Influent and effluent samples were taken for analyses of dissolved organic carbon (DOC), chemical oxygen demand (COD), ammonium nitrogen (NH_4-N), nitrate (NO_3-N), nitrite (NO_2-N), and orthophosphate (PO_4-P). The hybrid MBBR-MBR system was composed of a 40 L MBBR unit and a 10 L submerged MBR unit. Four different HRTs of 24 h, 18 h, 12 h and 6 h were applied to the operation of MBBR unit, and the HRT of the MBR unit was kept at 6 h (i.e. constant flux of 8.33 L/m^2 h) throughout the experimental period. The filling ratio of sponge cubes in the MBBR unit was 20% ($v_{sponge}/v_{reactor}$). For MBR unit, a hydrophilic polyvinylidene fluoride (PVDF) hollow fibre microfiltration (MF) membrane module was used with a pore size of 0.2 μm and surface area of 0.2 m^2. The operation conditions of the MBR unit were described in previous study by Luo et al. (2014). Chemical cleaning and fouling resistance analyses were performed when transmembrane pressure (TMP) reached 35 kPa (Deng et al., 2014). Measurements of zeta potential, RH, particle size, EPS and SMP were carried out on sludge samples which were directly collected from the MBR unit. The analytical methods and the process for determining micropollutant concentration using solid phase extraction (SPE) and gas chromatography–mass spectrometry (GC–MS) quantification were reported in Luo et al. (2014).

3. Results and discussion

3.1. The performance of MBBR at different HRTs

3.1.1. Overall removal of organic carbon and nutrients in MBBR unit

At HRTs of 24, 18, 12 and 6 h, the MBBR unit was able to achieve effective removals of DOC (> 94%), COD (> 94%) and NH_4-N (> 82%), but unstable total nitrogen (T-N) (45.2–72.3%) and PO_4-P elimination (26.4–49.9%). It was noteworthy that the MBBR showed the highest performance efficiency for removing DOC, COD, NH_4-N, PO_4-P and T-N at HRT of 18 h, which were 96.1 ± 0.4%, 97.4 ± 0.8%, 91.1 ± 1.6%, 49.9 ± 7.2%, and 72.3 ± 6.9%, respectively. This could be explained by the food to microorganisms (F/M) ratio in the MBBR unit. The F/M ratios were 0.72, 1.83, and 3.06 g COD/g MLVSS d at HRTs of 24, 12 and 6 h, respectively, while at HRT of 18 h, the feed supplied for microorganisms was adequate for their growth with F/M ratio of 0.91 g COD/g MLVSS d, which was very close to the value (1.05 g COD/g VSS d) as mentioned by Pozo et al. (2012) when treating kraft mill wastewater using a MBBR. The increased population of ammonium oxidation bacteria and utilization of phosphate by microorganisms for their growth and phosphorus uptake by phosphate accumulating organisms (PAOs) could contribute to high NH_4-N and PO_4-P removals at HRT of 18 h, respectively. In the MBBR unit, MLSS concentrations maintained at very low levels (0.13 ± 0.05, 0.09 ± 0.02, 0.11 ± 0.02 and 0.13 ± 0.02 g/L at HRTs of 24, 18, 12 and 6 h, respectively). The attached biomass concentrations were 0.41 ± 0.06 gMLVSS/g sponge (HRT of 24 h), 0.47 ± 0.03 gMLVSS/g sponge (HRT of 18 h), 0.40 ± 0.03 gMLVSS/g sponge (HRT of 12 h) and 0.37 ± 0.03 gMLVSS/g sponge (HRT of 6 h), respectively.

3.1.2. Removal and fate of the selected micropollutants in MBBR unit

Significant variations on micropollutants removal among each individual compound were observed over the study period (Fig. 1). At HRT of 18 h, the removals of most micropollutants remained constantly high over the experimental period except for carbamazepine, fenoprop, and metronidazole, because these three compounds are more recalcitrant due to the possession of electron withdrawing groups such as amide ($CONH_2$) carboxylic acid (COOH) and nitro groups (NO_2). Particularly, the removal of diclofenac increased by at least 30% along with better removals of ketoprofen, gemfibrozil, acetaminophen, bisphenol A, and pentachlorophenol as compared with those at other HRTs. Overall, the highest removal of most micropollutants could be accomplished at HRT of 18 h. The selected pharmaceutical and personal care products (PPCPs) in this study were mainly removed by biodegradation since they possessed low hydrophobicity (solid-water distribution coefficient, log D < 2.5) and their removal efficiencies were notably influenced by their intrinsic biodegradability (Luo et al., 2014; Tadkaew et al., 2011). The MBBR systems at all HRTs could effectively remove four of the investigated PPCPs, including ibuprofen (> 92%), salicylic acid (> 90%), primidone (> 81%), triclosan (> 80%), while carbamazepine showed particularly low removals (< 27%). Especially at HRT of 18 h, the highest removal efficiencies of ibuprofen (98.4 ± 1.2%), salicylic acid (98.1 ± 0.5%), primidone (90.9 ± 1.2%), carbamazepine (26.2 ± 5.7%) and relatively higher triclosan removal efficiencies (90.6 ± 1.0%) were achieved. Additionally, a significant increase was observed in the removals of metronidazole and diclofenac from 27.4 ± 10.8% and 33.3 ± 11.0%