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Short Communication

Evaluation of micropollutant removal and fouling reduction in a hybrid moving bed biofilm reactor–membrane bioreactor system

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HIGHLIGHTS

- The removals of micropollutants varied significantly (25.5–99.5%) in MBBR–MBR.
- Biodegradation was the primary pathway for micropollutant removal in MBBR–MBR.
- MBBR as pretreatment could reduce the fouling propensity of subsequent MBR.
- MBBR pretreatment could considerably lower the SMP content in MBR unit.

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ABSTRACT

A hybrid moving bed biofilm reactor–membrane bioreactor (MBBR–MBR) system and a conventional membrane bioreactor (CMBR) were compared in terms of micropollutant removal efficiency and membrane fouling propensity. The results show that the hybrid MBBR–MBR system could effectively remove most of the selected micropollutants. By contrast, the CMBR system showed lower removals of ketoprofen, carbamazepine, primidone, bisphenol A and estriol by 16.2%, 30.1%, 31.9%, 34.5%, and 39.9%, respectively. Mass balance calculations suggest that biological degradation was the primary removal mechanism in the MBBR–MBR system. During operation, the MBBR–MBR system exhibited significantly slower fouling development as compared to the CMBR system, which could be ascribed to the wide disparity in the soluble microbial products (SMP) levels between MBBR–MBR (4.02–6.32 mg/L) and CMBR (21.78 and 33.04 mg/L). It is evident that adding an MBBR process prior to MBR treatment can not only enhance micropollutant elimination but also mitigate membrane fouling.

1. Introduction

In recent years, the frequent detection of micropollutants in the aquatic environment has raised specific concerns due to their detrimental effects on aquatic organisms and human health. It has been reported that micropollutants often exhibit incomplete removal during the activated sludge process. As an alternative to the activated sludge process, moving bed biofilm reactor (MBBR) technology has demonstrated its suitability for micropollutant removal (Luo et al., 2014).

While MBBR has become an emerging technology for eliminating micropollutants, a major concern for MBBR applications is the decrease of sludge settleability when treating high strength wastewater, which may lead to severe operational problems when clarifiers are employed for the separation of solids. To counter this problem, various hybrid systems have been developed, which involve modifications of the basic MBBR system by adding coagulants (metal salts or cationic polymers) or applying membrane filtration or floatation as the solid separation process (Leiknes et al., 2006). Among all these modifications, combining membrane technology with MBBR is an established concept with growing popularity, which may also result in better membrane performance (Duan et al., 2013; Yang et al., 2009a). Several studies have demonstrated that hybrid MBBR–membrane bioreactor (MBBR–MBR) systems have the potential to mitigate membrane fouling through either preventing formation of submicron particles caused by aeration or altering the characteristics of EPS and SMP in MBR (Ivanovic

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and Leiknes, 2008; Yang et al., 2009b). However, previous studies have also indicated that hybrid MBBR–MBR systems could experience severe membrane fouling when large amounts of submicron colloidal particles were present in the reactor (Sun et al., 2012).

To date, there is a dearth of knowledge regarding the suitability of hybrid MBBR–MBR system for micropollutant removal. Thus, this study aimed to investigate the performance of a hybrid MBBR–MBR system on micropollutant removal and the effectiveness of the MBBR as a pretreatment option for fouling mitigation in MBR. The fouling propensity was investigated based on mixed liquor characteristics, such as soluble microbial products (SMP), extracellular polymeric substances (EPS), zeta potential, and relative hydrophobicity (RH).

2. Methods

2.1. Synthetic wastewater and sponge carrier

The synthetic wastewater used in the study contained chemical oxygen demand (COD) of 320–360 mg/L, dissolved organic carbon (DOC) of 100–120 mg/L, NH_4–N of 13–16 mg/L, NO_2–N of 0–0.02 mg/L, NO_3–N of 0.4–1.1 mg/L and PO_4–P of 3.0–3.5 mg/L. When necessary, either $NaHCO_3$ (powder, analytical grade) or 2 M H_2SO_4 was used to adjust the pH in the hybrid MBBR–MBR and CMBR to 7. A set of 22 micropollutants that have been frequently detected in municipal wastewater were selected for investigation (Luo et al., 2014). A concentrated stock solution containing 100 mg/L each micropollutant was prepared in pure methanol and kept in a freezer. The stock solution was then added to the synthetic wastewater to obtain an initial concentration of 5 μg/L for each compound. Polyurethane sponge cubes (S28/80R, Joyce Foam Products; dimension of 2 cm × 2 cm × 2 cm) were used as biofilm carriers. The biomass attached to the sponge was 0.41 ± 0.06 g/g sponge.

2.2. Experiment set-up

The hybrid MBBR–MBR system consists of an MBBR unit and a submerged MBR unit. The MBBR unit had a working volume of 40 L and was filled with sponge cubes (20% $v_{sponge}/v_{reactor}$). The MBR unit of the hybrid system and a conventional submerged membrane bioreactor (CMBR) were compared in terms of membrane fouling. Both MBRs used identical hydrophilic polyvinylidene fluoride (PVDF) hollow fibre microfiltration (MF) membrane modules with a pore size of 0.2 μm and surface area of 0.2 m^2. The MBR unit was fed with MBBR effluent (MLSS = 0.06 g/L) through a buffer tank. The CMBR used the same seed sludge as the MBBR unit, and the initial MLSS concentration (2.27 g/L) was similar to the total attached growth concentration of the MBBR unit (2.30 g/L). As no sludge withdrawal was performed except for removing sludge from carrier and mixed liquor for measurement, the SRTs of both systems could be considered infinite. The hybrid system was operated at a constant flow rate of 27.8 mL/min, resulting in HRTs of 24 h in the MBBR and 6 h in the MBR unit. The same HRT of 6 h was applied to the CMBR. Accordingly, the permeate flux of both MBRs was 8.34 L/(m^2 h). Both MBRs were operated in a continuous mode without backwash, relaxation or cleaning and the operation was terminated when the trans-membrane pressure (TMP) exceeded 35 kPa.

2.3. Analytical methods

DOC of the sample was analysed using a DOC analyser (Analytikjena Multi N/C 2000). The measurement of biosolids (indicated as mixed liquor suspended solids, MLSS) and biomass (indicated as mixed liquor volatile suspended solids, MLVSS) concentrations were conducted according to Standard Methods (APHA, 1998). Spectroquant Cell Test (NOVA 60, Merck) was used to measure NH_4–N, NO_2–N, NO_3–N and PO_4–P. The micropollutant concentration was determined by a method involving solid phase extraction and gas chromatography–mass spectrometry quantification as previously described by Hai et al. (2011).

Sludge samples were subjected to an extraction process to separate the SMP and EPS (Deng et al., 2014). Zeta potential of suspended solids of the mixed liquor was measured using a Zetasizer Nano ZS (Malvern Instruments, UK). RH was obtained through an emulsification process previously described by Ji et al. (2010).

3. Results and discussion

3.1. Organic carbon and nutrient removal

The MBBR–MBR system effectively removed DOC (94.7 ± 0.5%) and NH_4–N (84.9 ± 4.5%), which are consistent with a previous study by Duan et al. (2013). However, unstable TN reduction (45.2 ± 8.8%) and low PO_4–P elimination (34.9 ± 4.0%) were observed during the experimental period. Additionally, it was noted that organic carbon and nutrients were principally removed by the MBBR unit, while the subsequent MBR unit offered very limited further elimination. Compared to the MBBR–MBR, the CMBR was less efficient for NH_4–N (56.1 ± 3.9%) and TN (21.9 ± 4.6%) removal, but showed similar DOC removal (94.7 ± 1.6%) and slightly higher PO_4–P elimination (45.1 ± 6.8%) due to the biomass growth.

The MLSS and MLVSS concentrations in the MBBR unit were very low (0.05–0.13 and 0.04–0.11 g/L, respectively), as effluent from MBBR carried suspended solids into the subsequent MBR unit and no sludge was recycled back to the MBBR. Regarding the MBR unit, the initial MLSS and MLVSS were 0.06 and 0.05 g/L, respectively, and both showed gradual growth during the operation, reaching 0.91 and 0.89 g/L at the end of the study. As for the CMBR, the MLSS and MLVSS increased from 2.27 and 2.05 g/L to 7.38 and 7.08 g/L, respectively, during operation period.

3.2. Removal of the selected micropollutants

During the MBBR–MBR treatment, the compound-specific removal efficiencies (shown in Fig. 1) varied significantly, ranging from 25.5% to 99.5%. Although a clear correlation between removal efficiencies and the effective octanol–water partition coefficients ($\log D$) of the selected micropollutants could not be obtained, it was found that all hydrophobic compounds ($\log D > 3.2$) were effectively removed (>80%) (Fig. 2). One possible reason is that the attached growth pattern in the MBBR could enhance the retention of the biomass, thus promoting the enrichment of slow growing microorganisms and the formation of a diverse biocoenosis. In general, micropollutant removal by the MBBR–MBR was higher than that by the CMBR. Moreover, the CMBR was less effective for some micropollutants. Particularly, the removals of carbamazepine, ketoprofen, primidone, estriol and bisphenol A were lowered by 16.2%, 30.1%, 31.9%, 34.5%, and 39.9% respectively during the CMBR treatment.

To gain further insight into the fate of micropollutants during the MBBR–MBR treatment, a mass balance of the investigated compounds was evaluated (Eq. (1)), taking into account the removal pathways of biodegradation and sorption in the MBBR unit, and total removal in the MBR unit.

$$L_{inf} = L_{s,MBBR} + L_{b,MBR} + L_{MBR} + L_{eff} \quad (1)$$