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Understanding ultrafiltration membrane fouling by soluble microbial product and effluent organic matter using fluorescence excitation–emission matrix coupled with parallel factor analysis

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ABSTRACT

Membrane fouling of soluble microbial product (SMP) and effluent organic matter (EfOM) in ultrafiltration (UF) was investigated by using fluorescence excitation–emission matrix coupled with parallel factor analysis (EEM–PARAFAC) as well as dissolved organic carbon (DOC) and polysaccharides measurement. SMP and EfOM samples were generated in bench-scale sequencing batch reactors (SBRs) fed with synthetic wastewater and domestic wastewater under different solid retention times (SRTs). Results showed that EfOM contained more DOC and more sub-fractional components except for tyrosine-like substances compared to SMP, however, SMP caused more severe fouling. The fluorescence intensity of tyrosine-like substances in UF feed showed good correlation with membrane fouling index, suggesting that it can serve as a fouling potential indicator. The mass balance analysis also indicated that the tyrosine-like substances was a major foulant. Membrane fouling was alleviated as SRT increased, which can be associated with the decrease of protein-like (tyrosine-like and tryptophan-like) substances under longer SRT.

Introduction

Ultrafiltration (UF) has been increasingly adopted in wastewater reclamation and reuse schemes to improve the quality of tertiary treated wastewater effluent for use in irrigation, dual-distribution schemes or as a pre-treatment stage prior to reverse osmosis treatment (Wintgens et al., 2005). However, efficient application of UF processes is significantly reduced by membrane fouling, which contributes to increased operational costs and need for frequent chemical cleaning as well as shortened membrane service life (Gillerman et al., 2006). Therefore, understanding of the major foulant in the secondary effluent and the factors that affect the major foulants in the secondary processes is of importance for sustainable application of UF membrane processes in the wastewater reclamation and reuse schemes.

Effluent organic matter (EfOM) is recognized as a critical foulant of UF membrane in the reclamation and reuse schemes (Fan et al., 2008; Henderson et al., 2011). EfOM comprises natural organic matter (NOM), soluble microbial products (SMP) derived from the biological treatment processes, and organic compounds from water use and water disinfection processes (Shon et al., 2006). In order to determine the major foulants in EfOM, advanced organic characterization methods like size exclusion chromatography (SEC) performed with liquid chromatography with organic carbon detector (LC–OCD) and fluorescence excitation–emission matrix (EEM) spectroscopy were utilized to characterize the foulants during UF (Henderson et al., 2011; Tian et al., 2013). With the help of these advanced methods, high-molecular weight organic matters comprised of hydrophilic components such as SMP, and protein-like extracellular matter were widely recognized as the major cause of membrane fouling (Fan et al., 2008; Henderson et al., 2011). However, LC–OCD cannot always successfully identify the fouling fractions in the UF feed (Henderson et al., 2011).

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Recently, increasingly adopted fluorescence spectroscopy enables a rapid and sensitive characterization of DOM. Three-dimensional excitation–emission matrix (EEM) can be generated with samples excited at a range of wavelengths and emission recorded across a range of wavelengths. This EEM can be correlated to the fluorescence components in the DOM and thus gives more insight into DOM fractions and their chemical characteristics (Coble, 1996; Stedmon and Markager, 2005). However, the quantitative interpretation of EEM data remained a problem before the introduction of Parallel Factor Analysis (PARAFAC) (Stedmon et al., 2003). With the PARAFAC analysis, spectrally overlapping EEM data can be mathematically separated into chemically independent fluorescence components. EEM–PARAFAC has proved a suitable approach for monitoring removal of organic matter across drinking water plant and water recycling plant, characterizing DOM in municipal wastewater and extracellular polymeric substance of algae, and charactering the interaction between DOM and trace metals (Yamashita and Jaffé, 2008; Murphy et al., 2011; Xu et al., 2013; Li et al., 2014; Shutova et al., 2014). All of these applications above indicated the robustness of this tool in tracing subtle changes of organic components in aquatic system. It is believed that EEM–PARAFAC is able to illustrate the changes of SMP and EfOM and their fractional component can be discriminated and quantified, and in turn, related to the UF membrane fouling in order to identify major foulants.

With the increasing information of major foulants in the secondary effluent, it is possible to reduce the target fraction in the effluent by adjusting operational parameters of secondary processes, and thus in turn, to alleviate the following UF membrane fouling. Sludge retention time (SRT) is an important parameter in biological wastewater treatment processes. The quantity and quality of SMP and EfOM can be extensively changed at different SRTs. It was reported that more high molecular weight (MW) (>10,000 Da) biopolymers were generated with a longer SRT (Jarusutthirak and Amy, 2007). As the SRT increased, the amount of EfOM reduced, and the aromaticity and reactivity of EfOM increased (Esparza-Soto et al., 2011). However, the variation of EfOM according to the change of SRT has not been related to the UF membrane fouling in the tertiary filtration.

The aim of this research was therefore to investigate the major foulants in SMP/EfOM during tertiary UF and the effect of SRT on the major foulants. SMP and EfOM samples under different solid retention times (SRTs) were generated in bench-scale sequencing batch reactors (SBRs) with different feeds (synthetic wastewater and domestic wastewater). The SMP and EfOM samples obtained as well as the feed, permeate, reversible and irreversible foulant layers during the UF of SMP/EfOM were characterized by using EEM–PARAFAC as well as dissolved organic carbon (DOC) and polysaccharides measurement. With this information, membrane fouling was correlated to these indices of UF feed, and the major foulants were identified. Besides, the effect of SRT on the major foulants, and the UF fouling was illustrated.

Material and methods

Lab-scale SBRs and operation conditions

Six lab-scale SBRs, simulating activated sludge process, were constructed to generate SMP and EfOM samples for characterization. Three of them were fed with synthetic wastewater, which contained easily biodegradable glucose as the sole carbon source. Organic matter in the effluents of these three SBRs was considered to be only SMP, since no NOM was introduced into the reactors and glucose was able to totally transform to other organic products (Jarusutthirak and Amy, 2007). Meanwhile, the other three SBRs, which were designed to generate EfOM, were fed with domestic wastewater obtained from wastewater pipe of a residential community.

SRTs of the six SBRs were 5, 10, and 30 days respectively (denoted as SMP-5, SMP-10, SMP-30, EfOM-5, EfOM-10, and EfOM-30 hereafter). The SRTs of SBRs were maintained by manually wasting mixed liquor at different rates. The wasting volume of mixed liquor was calculated by the following equation (Jarusutthirak and Amy, 2007):

$$F_w = \frac{V}{\theta_c} \quad (1)$$

where F_w is the mixed liquor wasting rate (L/day), θ_c is the solids (sludge) retention time (days), and V is the effective volume of SBR (L). According to Eq. (1), the F_w to maintain the SRT at 5, 10, and 30 days were 0.8, 0.4, and 0.13 L/day, respectively.

Except for feed water and SRT, all six SBRs were operated under identical condition. All SBRs were operated under ambient condition (21 ± 1 °C). Each reactor had a total volume of 5 L (4 L working volume and 1 L head space). Each operational cycle contained filling (2 min), mixing and aeration (10 h), sludge wasting (1 min), sedimentation (1 h 55 min), and decanting (2 min), among which filling, sludge wasting, and decanting were operated manually. 3 L effluent was decanted in each cycle. The hydraulic retention time (HRT) was 12 h. The reactors were aerated by air pumps and stone diffusers, and the dissolved oxygen in each SBR was kept at above 2 mg/L.

All SBRs were initially inoculated with activated sludge from Wenchang WWTP (Harbin, China). The sludge was allowed to acclimate to the feed water for longer than two months (longer than twice the given SRTs) before SMP and EfOM samples were collected (Jarusutthirak and Amy, 2006). The acclimation of biomass was confirmed by stable water quality of the effluent achieved. After the acclimation, mixed liquor suspended solid (MLSS) of SMP-5, SMP-10, SMP-30, EfOM-5, EfOM-10, and EfOM-30 were 994 ± 53 mg/L, 1923 ± 33 mg/L, 3425 ± 68 mg/L, 989 ± 79 mg/L, 2054 ± 38 mg/L, 3679 ± 99 mg/L, respectively. The suspended solids (SS) in the effluent of SMP-5, SMP-10, SMP-30, EfOM-5, EfOM-10, and EfOM-30 were 13 ± 4 mg/L, 11 ± 2 mg/L, 6 ± 3 mg/L, 9 ± 3 mg/L, 10 ± 4 mg/L, 6 ± 3 mg/L. The SRTs of the SBRs were calculated according to the MLSS and SS in the effluent. The real SRTs of SMP-5, SMP-10, SMP-30, EfOM-5, EfOM-10, and EfOM-30 were 4.69 d, 9.46 d, 28.49 d, 4.78 d, 9.54 d, and 28.61 d, respectively. Since the SRTs were not largely changed, the SBRs will be still denoted as SMP-5, SMP-10, SMP-30, EfOM-5, EfOM-10, EfOM-30 in the following parts for a convenient comparison.

Feed water of SBRs

The collected domestic wastewater was sieved with a 1 mm pore size sieve and then added into the EfOM-5, EfOM-10, and EfOM-30 reactor. The characteristics of the influent wastewater are summarized in Table 1.

Table 1
Characteristics of domestic wastewater.

Parameter	Concentration	Unit
COD	286.4 ± 49.3	mg/L
DOC	106.2 ± 21.8	mg/L
$NH_4^+ - N$	36.1 ± 11.8	mg/L
Total P	3.4 ± 1.5	mg/L
UV_{254}	20.4 ± 3.1	m^{-1}
SUVA	0.19 ± 0.05	L/m mg TOC

$n = 64$ (number of different days during which samples were taken for analysis).