



Characteristics and trihalomethane formation reactivity of dissolved organic matter in effluents from membrane bioreactors with and without filamentous bulking



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HIGHLIGHTS

- Bulking sludge reduced chlorine reactivity, but increased bromine-containing THMs.
- Bulking sludge increased EfOM and the content of aromatic moieties DOM.
- Bulking sludge led to a higher proportion of high molecular weight EfOM.
- Bulking sludge led to a relatively high hydrophobicity.
- Bulking sludge increased simple aromatic protein, fulvic and humic acid-like.

ARTICLE INFO

Article history:

Received 30 December 2015
Received in revised form 1 March 2016
Accepted 3 March 2016
Available online 18 March 2016

Keywords:

MBR
Filamentous bulking
Dissolved organic matter
Chlorination
Trihalomethanes

ABSTRACT

In this study, synthetic wastewater was treated by two identical membrane bioreactors (MBRs): the normal sludge MBR (NS-MBR) and the bulking sludge MBR (BS-MBR). Effects of filamentous bulking on the characteristics and trihalomethane (THM) formation reactivity of MBR effluent dissolved organic matter (EfOM) were investigated. Filamentous sludge bulking had no significant influence on the regulated MBR effluent water quality except NO₂-N and NO₃-N. NS-MBR effluent had more low molecular weight (LMW) (<5 kDa) EfOM (92.43%) than BS-MBR (75.18%). About two-thirds of EfOM from BS-MBR were hydrophilic substances. On the contrary, EfOM from NS-MBR exhibited higher hydrophobicity. The ratio of polysaccharides and proteins in MBR effluents increased after filamentous bulking. There were more protein-like materials, fulvic acid-like and humic acid-like in BS-MBR EfOM. The THM formation reactivity of BS-MBR EfOM was 30.15% of NS-MBR EfOM, whereas BS-MBR EfOM exhibited higher formation reactivity of bromine containing species.

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1. Introduction

The conventional activated sludge (CAS) process is the most commonly used wastewater treatment technology, which is composed of a biochemical stage (aeration tank) and a physical settling stage (secondary clarifier). However, bulking sludge (BS), as a common problem in CAS systems, results in biomass loss and poor effluent quality (Wang et al., 2010). The overgrowth of filamentous bacteria is usually considered as the main reason of sludge bulking (Martins et al., 2004). In order to relieve global water scarcity (Chen et al., 2003), membrane bioreactor (MBR) technology has been applied for municipal wastewater treatment and reclamation

(Francy et al., 2012). MBR technology is the combination of membrane separation and activated sludge processes. Stable and high quality reclaimed water can be produced from municipal wastewater through a MBR system (Hirani et al., 2010). However, due to the membrane fouling, the rapid decline of membrane flux is a major obstacle for the applications of MBRs (Wang et al., 2010). Previous studies showed that filamentous bulking sludge could reduce the sustainable operation time of MBRs and had significant negative effects on membrane fouling because that the excess growth of filamentous bacteria resulted in much more release of extracellular polymeric substances (EPS), lower zeta potential, higher hydrophobicity of sludge flocs and more irregularly shaped flocs, which reduced the membrane filtration efficiency (Meng et al., 2006). Sludge properties are highly related to the physiological behaviors of microorganisms. If the species and/or dominant

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colony changed, it will lead to different physiochemical characteristics and abnormal conditions of mixed liquor, which can accordingly influence the property of MBR effluent dissolved organic matter (EfOM). It was found in previous research that a filamentous sludge bulking condition led to the larger PSD, lower hydrophobic contents in soluble metabolic products (SMP), lower value of weight-average molecular weight (MW) of EfOM (Wang et al., 2010). Filamentous sludge bulking condition was proven to have great influences on the physiochemical characteristics of MBR EfOM (Choi et al., 2002).

Generally, to prevent the transmission of pathogens in reclaimed water, adequate disinfection of MBR effluents is required (USEPA, 2004). Chlorine is an efficient, low-cost and widely used disinfectant. Sufficient residual chlorine can prevent the bacteria regrowth in treated water. However, when chlorine reacts with EfOM, a variety of genotoxic, mutagenic and/or carcinogenic disinfection by-products (DBPs) will be formed (Chellam and Krasner, 2001). Total trihalomethane formation potential (THMFP) of the MBR treated municipal wastewater was found to be as high as 665 µg/L (Ma et al., 2013a). During chlorination, trihalomethane (THM) formation and speciation are dependent on water quality, especially on EfOM characteristics (Gang et al., 2003; Zhang et al., 2009).

Since filamentous sludge bulking has great influences on the physiochemical characteristics of MBR EfOM, it is expected to affect the formation and speciation of DBPs during chlorination of MBR effluents (Meng and Yang, 2007). Filamentous sludge bulking is a common problem in MBR process. Therefore clearly understanding the DBP formation characteristics of MBR with filamentous sludge bulking is important considering the ecological safety of wastewater recycling. However, the related information is not available.

Therefore, the primary objective of this study is to investigate the influences of filamentous bulking on MBR EfOM characteristics regarding THM formation. Two identical submerged MBRs with and without filamentous bulking were operated in parallel for more than 3 months. Dissolved organic carbon (DOC), specific UV absorbance at 254 nm (SUVA), MW distribution, polarity properties and chemical compositions of EfOM were studied. The relationships between characteristics of EfOM and the formation and speciation of THMs during chlorine disinfection were also analyzed.

2. Materials and methods

2.1. Lab-scale MBR

Two identical 18 L submerged aerobic MBRs equipped with a PVDF hollow fiber membrane module that was produced by Xiamen Kymem Technology Co., Ltd (normalized pore size, 0.02 µm; surface area, 0.175 m²) were constructed. Activated sludge seeded in the reactors was obtained from an aeration basin of the sewage treatment plant (Jinan, China). According to previous research (Liang et al., 2007), synthetic municipal wastewater, which was used to feed the reactors, was prepared. Sodium acetate produced the carbon in the synthetic wastewater. Since sodium acetate can be completely biodegraded in the activated sludge process, SMP was the only dissolved organic matter (DOM) in supernatants and effluents were without any refractory organic matters. MBR was manipulated at 20 ± 1 °C with the hydraulic retention time (HRT) of 12 h, which was automatically controlled by a programmable logic controller (PLC). The MBR systems' sludge retention times (SRTs) were maintained at 10 days by sludge discharging every day. The concentration of dissolved oxygen (DO) in the sludge mixture was maintained at 3.0 ± 1.0 mg/L, and pH

was 7.0–8.0. Please refer to the previous study for detailed characterization of the bioreactors system with the flux diagram (Ma et al., 2013a). The MBRs' performance-related information can be discovered in Tables 1 and 2. The DO concentration in the sludge mixture of one MBR that was intended to represent filamentous bulking MBR was reduced to 0.5 mg/L and continued for 5 days. When the above process was finished, the two MBRs exhibited different sludge characteristics. The filamentous bulking occurred in the MBR that had reduced DO concentration. The DO concentration was recovered after 5 days to simulate the condition in reality. But it was useless in improving the situation of filamentous bulking. Within 27 days the transmembrane pressure of ultrafiltration (UF) membrane module in the normal sludge MBR (NS-MBR) increased from 0 to 0.05 MPa; however, it only took 11 days to increase from 0 to 0.05 MPa in the bulking sludge MBR (BS-MBR).

2.2. EfOM fractionation

A MinimTM II Tangential Flow Filtration (TFF) system (PALL, USA) consisting of a series of OmegaTM UF membranes with MW cut-off of 1, 5, 10, 30 and 100 kDa (Ma et al., 2013b) was used to fractionate NS-MBR and BS-MBR EfOM into six fractions. Before applying samples, a blank sample with ultrapure water was prepared for each membrane module. The previous study (Ma et al., 2013b) recorded the detailed UF fractionation procedure.

Nonionic Amberlite XAD-8 resin (20–60 mm) was applied to fractionate EfOM from two MBRs into hydrophilic substances (HiS), hydrophobic acids (HoA), hydrophobic bases (HoB) and hydrophobic neutrals (HoN). The XAD-8 resins were cleaned before application (Leenheer, 1981). EfOM fractionation was referred to the previous report (Ma et al., 2014b).

The pH of MBR effluents and EfOM fractions were adjusted to pH 7.0 after fractionation. Ultraviolet absorbance at 254 nm (UV₂₅₄) and DOC of each sample were measured. After both UF separation and XAD-8 resin adsorption processes, EfOM recoveries (as %UV₂₅₄ and %DOC) were computed to assess any loss or contamination (Gang et al., 2003).

2.3. Excitation–emission matrix (EEM) fluorescence spectroscopy analysis

An F-4500 FL spectrophotometer (Hitachi, Japan) was applied in the process of the EEM fluorescence spectroscopy measurements. The EEM spectra were collected with relevant scanning emission spectra from 250 to 550 nm at 5 nm increments by varying the excitation wavelength from 200 to 400 nm at 5 nm sampling intervals. The EEM spectra normalized to 1 mg/L DOC were plotted as contours.

The specific components of organic matters were represented by three regions that EEM spectra were separated into: Region I (EX230 to 300/EM250 to 390, protein-like materials: microbial byproducts, proteins, polypeptides and amino acid-like), Region II (EX230 to 300/EM390 to 550, fulvic acid-like) and Region III

Table 1
Processing index when running NS-MBR and BS-MBR.^a

Items	NS-MBR	N	BS-MBR	N
MLSS (g/L)	1.71 ± 0.04	24	1.46 ± 0.03	17
MLVSS (g/L)	1.42 ± 0.01	24	0.89 ± 0.01	17
DO (mg/L)	3.48 ± 0.31	55	3.36 ± 0.23	22
T (°C)	20.44 ± 0.62	55	20.40 ± 0.55	22
pH	8.08 ± 0.12	18	7.78 ± 0.10	13

^a Values are given as mean ± standard deviation, N means the number of measurements.

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