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Assessing membrane biofouling and its gel layer of anoxic/oxic membrane bioreactor for megacity municipal wastewater treatment during plum rain season in Yangtze River Delta, China

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ABSTRACT

This study assessed membrane biofouling and its gel layer of anoxic/oxic membrane bioreactor (A/O-MBR) for megacity municipal wastewater treatment during plum rain season, which was continuous rainy weather, in Yangtze River Delta, China. A laboratory-scale A/O-MBR was operated to treat the municipal wastewater from Quyang wastewater treatment plant, which located at the typical megacity of Shanghai in Yangtze River Delta, from April to July accompanying with plum rain season. As reactor performance showed, COD_{Cr}, NH_4^+-N, TN, TP of the influent gradually decreased during plum rain season, and inhibited pollutant removal due to organic carbon shortage. However, dissolve inorganic carbon and inorganic components in mixed liquid had an obvious increase under rainy weather. Membrane filtration results indicated that plum rain season enhanced pore blocking behavior, further leading to the serious membrane biofouling but inhibiting gel layer formation. Additionally, gel layer analysis predicted that plum rain season led to plenty of inorganic components and precipitate flew into A/O-MBR reactor. Inorganic components with elements of Ca, Mg Ba, Fe, Al and Si seriously blocked membrane pores. Those components also accumulated into gel layer in the form of SiO_2, $CaCO_3$, $CaSiO_3$, $MgNH_4PO_4$, $BaCO_3$, $AlPO_4$, etc. Consequently, plum rain season enhanced pore blocking behavior and led to severe membrane biofouling but with the inhibition of gel layer formation.

1. Introduction

With over 30-years economic development, megacities, containing over millions population, have been built up all over the China, such as Beijing, Shanghai, Shenzhen, Guangzhou, Chengdu, Wuhan, Changsha, Suzhou, etc. Especially, Yangtze River Delta contains Shanghai and parts of Jiangsu, Zhejiang and Anhui provinces, which is the economic and industrial center of China (Chong et al., 2016; Gao et al., 2017). Thus, dozens of megacities, Shanghai (24 million population), Suzhou (13 million population), Nanjing (8.3 million population), Hangzhou (9.0 million population), Changzhou (4.7 million population), Hefei (7.9 million population), Wuxi (6.5 million population), etc. were located in the region of Yangtze River Delta, promoting the formation of Yangtze River Delta urban agglomerations (Li et al., 2016).

However, huge population of megacities induced the serious pollution pressure to municipal wastewater treatment plant, and environmental risk of megacities, especially Yangtze River Delta, have been attracted more attentions in recent years (An et al., 2015; Kang et al., 2016; Zhuo et al., 2017). Thus high efficiency municipal wastewater treatment technology is required. Membrane bioreactor (MBR), combining physical separation and biological degradation, is a high-efficiency technology for wastewater treatment and water reuse (Aslam et al., 2017; Eyvaz et al., 2016; Navarro et al., 2016; Torretta et al., 2017). In recent decades, because of the outstanding advantages, such as sludge maintainment, low footprint, less sludge production, high quality of effluent, etc., MBR has been widely applied in China (Meng et al., 2017). There are dozens

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of installations of large-scale MBR for >10,000 m^3/d municipal wastewater treatment in China, and parts of them were mainly located at megacities of Yangtze River Delta, China (Huang et al., 2010; Shen et al., 2012). Hu et al. (2014) also reported that over 6 municipal wastewater treatment plants in Wuxi were applied with MBR technology. Consequently, MBR is one of the best effective technology for megacity municipal wastewater treatment, especially space-limiting area.

However, membrane biofouling, as the obstacle for the widespread application of MBR, usually occurs with pore blocking and the gel-like layer formation on membrane surface (Wang et al., 2008; Wang and Wu, 2009). Gel-like layer is normally classified into gel layer (mainly caused by solutes, colloids, extracellular polymeric substances (EPS), etc.) and cake layer (attributed to the adhesion and accumulation of cake sludge, inorganic particles, etc.), and recent study (Hong et al., 2014) reported that formation of both gel layer and cake layer simultaneously occurred on membrane surface during MBR operation. In addition, during MBR operation in practice, membrane was applied under sub-critical flux to mitigate severe cake layer formation on membrane surface, and membrane was generally fouled with a thin gel-like layer and then operated with physical and/or chemical washing to extend the membrane lifetime (Guo et al., 2012; Huang et al., 2010; Wang et al., 2008). This thin gel-like layer, which is attributed to the adhesion of solutes, colloids, EPS (majority) and the accumulation of cake sludge, inorganic particles (minority), is the key contribution for membrane biofouling, and this thin gel-like layer was called as gel layer in this study.

Yangtze River Delta annually has a continuously rainy weather from late spring to early summer, which is commonly called plum rain season. Plum rain season normally begins from the middle of June, and ends at the beginning of July, lasting for approximately 20-plus days. According to previous literature (Sun et al. 2010, 2015), municipal wastewater treatment system of megacities in Yangtze River Delta would show obvious performance variations on pollution removal because of the influent evolution with rainy weather. Tsai et al. (2011) predicted that acid rainfall caused plenty of inorganic components, such as NH_4^+, HCO_3^-, Na^+, Ca^{2+}, etc., dissolved into the influent of wastewater treatment system during plum rain season. Zhou et al. (2015a) also reported that lots of inorganic components from constructions or its waste easily flew into municipal wastewater plants with rainfall in megacity. On the other side, recent researches (Meng et al., 2017; Zhou et al., 2014, 2017c) proved that inorganic components play a significant role in gel layer formation and membrane biofouling. Thus, it could be speculated that plum rain season might have some effects on membrane biofouling and its gel layer of MBR, especially for megacity municipal wastewater treatment. However, this speculation have never been studied.

This study aimed to assess membrane biofouling and its gel layer of anoxic/oxic MBR (A/O-MBR) for megacity municipal wastewater treatment during plum rain season in Yangtze River Delta, China. A laboratory-scale A/O-MBR was set-up and operated to treat the municipal wastewater from Quyang wastewater treatment plant, which located at the typical megacity of Shanghai in Yangtze River Delta, from April to July accompanying with plum rain season. Temperature was in the range of 22–36 °C during the entire operation, and this temperature variation would not induce obvious effects on membrane biofouling based on previous study (Miyoshi et al., 2009). Thus, this study ignored the temperature effects on membrane biofouling. In this study, various measurements, such as XRD (X-ray diffraction), SEM-EDX (scanning electron microscopy-energy dispersive X-ray analyzer), membrane resistance, EPS of gel layer, etc., were carried out to analyze membrane biofouling and identify its gel layer properties.

2. Materials and methods

2.1. Set-up and operation of A/O-MBR

A laboratory-scale A/O-MBR, containing a working volume of 4.5 L (anoxic and oxic zones of 1.5 L and 3.0 L, respectively), was operated from April to July. A PVDF (polyvinylidene fluoride) hollow fiber membrane module (with a total surface area of 260 cm^2; 0.4 μm pore size; Litree Company, China) was installed in the oxic zone. A constant fluid flux was set at 17 L/(m^2 h) with intermittent suction mode (10 min suction and 2 min relaxation per cycle) was applied during operation. Pressure gauge was used for *trans*-membrane pressure (TMP) measurement. The flow rate of recycled mixed liquor from the oxic zone to the anoxic zone was controlled at 200% of the influent flow rate. pH in the reactor was controlled by NaOH and HCl addition within the range of 7.0–7.7. Hydraulic retention time (HRT) and solids retention time (SRT) were maintained at 10.0 h and 30 days, respectively. When TMP reached 40 kPa, the membrane module was removed for physical (washing with tap water) and chemical cleaning (1% NaOCl and 10% citric acid immersion for 6 h, respectively) to recover the membrane permeability.

The specific aeration demand per membrane area (SADm) in some full-scale MBRs is generally ranged 0.2–1 m^3 air/m^2 h. However, according to previous studies (Wang et al. 2008, 2010), pilot-scale MBRs for megacity (Shanghai, China) municipal wastewater treatment in Yangtze River Delta remained a high SADm (approximately 8.6–35 m^3 air/m^2 h), even with sheet membrane module. Hu et al. (2014) and its references showed that large-scale MBRs for megacity (Wuxi, China) municipal wastewater treatment with hollow fiber membrane module also needed high SADm to induce the effective scouring with a cross-flow action for the inhibition of cake sludge accumulation on membrane surface. Especially, Chengbei WWTP, the largest scale MBR (50,000 m^3/d; with hollow fiber membrane module) in Wuxi, maintained approximately 13–17 m^3 air/m^2 h SADm during operation. Thus, MBR operational conditions for megacities in Yangtze River Delta were different from other areas, especially high aeration rate. In addition, because of the small reactor volume and high mixed liquid concentration, high SADm (>13 m^3 air/m^2 h) was necessary for prevention of bubbling pore blocking. Consequently, this study applied a high SADm (approximately 15 m^3 air/m^2 h) to simulate large-scale MBR operation, and the air diffuser was set for supplying continuously air (0.4 m^3/h; controlling with gas flow-meter) to offer oxygen for microbial activity and induce a cross-flow action for effective scouring.

The influent of this A/O-MBR was the effluent from an aerated grit chamber of the Quyang municipal wastewater treatment plant (WWTP) (31°16′ N, 121°28′ E, Shanghai, China; Fig. S1). Quyang WWTP, built up at 1982, locates at the downtown of Shanghai (Hongkou district) with total occupied space of 3.54 ha. Quyang WWTP serves over 650 ha residential area with 200 thousands people, which is in the system of mixture of rainwater and sewerage. To better modify the practical operation, the inoculating biomass was drawn from the return activated sludge stream in Quyang WWTP. The newly inoculated A/O-MBR was initially operated to achieve steady state for the acclimatization of activated sludge. The membrane module was then replaced with a new unit and the A/O-MBR was operated for the experiments from April to July.

2.2. Membrane resistance analysis

Membrane resistance, which is a major characteristic for membrane biofouling, can be calculated as the following equation: