Comparative study on membrane fouling between membrane-coupled moving bed biofilm reactor and conventional membrane bioreactor for municipal wastewater treatment

W. Yang, W. Syed and H. Zhou

ABSTRACT

This study compared the performance between membrane-coupled moving bed biofilm reactor (M-MBBR) and a conventional membrane bioreactor (MBR) in parallel. Extensive tests were conducted in three pilot-scale experimental units over 6 months. Emphasis was placed on the factors that would affect the performance of membrane filtration. The results showed that the concentrations of soluble microbial product (SMP), colloidal total organic carbon and transparent exopolymer particles in the M-MBBR systems were not significantly different from those in the control MBR system. However, the fouling rates were much higher in the M-MBBR systems as compared to the conventional MBR systems. This indicates membrane fouling potential was related not only to the concentration of SMP, but also to their sources and characteristics. The addition of polyaluminum chloride could reduce the fouling rate of the moving bed biofilm reactor unit by 56.4–84.5% at various membrane fluxes.

Key words | membrane fouling, membrane-coupled moving bed biofilm reactor, membrane bioreactor, wastewater treatment

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INTRODUCTION

Membrane filtration has become more widely used for wastewater treatment and water reuse. A successful application is membrane bioreactor (MBR) technology for wastewater treatment which primarily focuses on the coupling of membrane filtration with activated sludge process (ASP) (Judd 2006; Yang *et al.* 2006). However, combining a submerged membrane filtration with an aerobic biofilm process has not been applied in full scale. The benefits and/or challenges of such a combination are in great need of further investigation.

Biofilm processes have the following advantages over conventional ASP: (1) higher organic loading rates; (2) better oxygen transfer; (3) higher nitrification rates; (4) a larger surface area for mass transfer; and (5) high resistance of attached biomass to overloading and toxic compounds (Sombatsompop *et al.* 2006; Chan *et al.* 2009; Rahimi *et al.* 2011). An attractive biofilm-based process is the moving bed biofilm reactor (MBBR) that would generally upgrade wastewater treatment facilities to increase organic loading and simultaneous nutrient removal. Due to the higher loading rates in MBBR systems compared to conventional ASP systems, smaller footprint bioreactors are often feasible. Unfortunately, it has been reported that settleability of MBBR sludge was less efficient than conventional activated sludge (Lee *et al.* 2006). According to Ødegaard (2000), settleability of biosolids remains the largest challenge in MBBR design. Membrane filtration could be a solution to solve the poor settling problem of MBBR sludge.

A number of studies have evaluated the performance and affecting factors of coupling membrane filtration with MBBR (Melin *et al.* 2005; Lee *et al.* 2006; Rahimi *et al.* 2011). Originally introduced by Leiknes & Ødegaard (2007), the moving bed membrane bioreactor (MBMBR) or the biofilm-MBR (BF-MBR) process has been studied and developed for shipboard wastewater treatment (Ivanovic *et al.* 2008; Sun *et al.* 2010). However, none of the studies mentioned above compared the MBMBR performance with conventional MBRs in pilot scale. Published in 2009,

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a research group did comparison studies between a MBMBR and a conventional MBR (Yang *et al.* 2009a, b). Their research focused on organic carbon and nitrogen removal and only a short communication paper involved the study of membrane fouling. The main objectives of this study were to compare side-by-side the performance between membrane-coupled MBBR (M-MBBR) and a conventional MBR in pilot-scale experimental units. Emphasis was placed on the factors that would affect the performance of membrane filtration, such as colloidal total organic carbon (TOC), soluble microbial products (SMPs) and transparent exopolymer particles (TEP). In addition, a coagulant was added as a pre-treatment for membrane filtration of MBBR effluents, attempting to reduce colloidal TOC in the membrane filtration tank and hence to improve membrane filtration performance. The impact of coagulation in M-MBBR process on membrane filtration performance was studied.

MATERIALS AND METHODS

Experimental set-up

Figure 1 shows the process flow diagram of the three pilot-scale units. The MBBR was filled with plastic carriers at a filling fraction of 70%. For M-MBBR test units, two submerged membrane filtration units were installed and operated in different process configurations (Pilot-A and Pilot-B) to treat effluents from a MBBR. Pilot-B was run at two different modes. At mode I, the membrane tank was installed in the settling tank; whereas the membrane tank was separated with the settling tank at mode II. Also, polyaluminum chloride (PACl) was added into the settling tank with stirring at the dosage of 50 mg/L when Pilot-B was run at mode II. In parallel, a conventional MBR unit (Pilot-C) was run as a comparison to the two M-MBBR units. Hollow fiber membrane modules (GE Water & Process Technologies, Oakville,

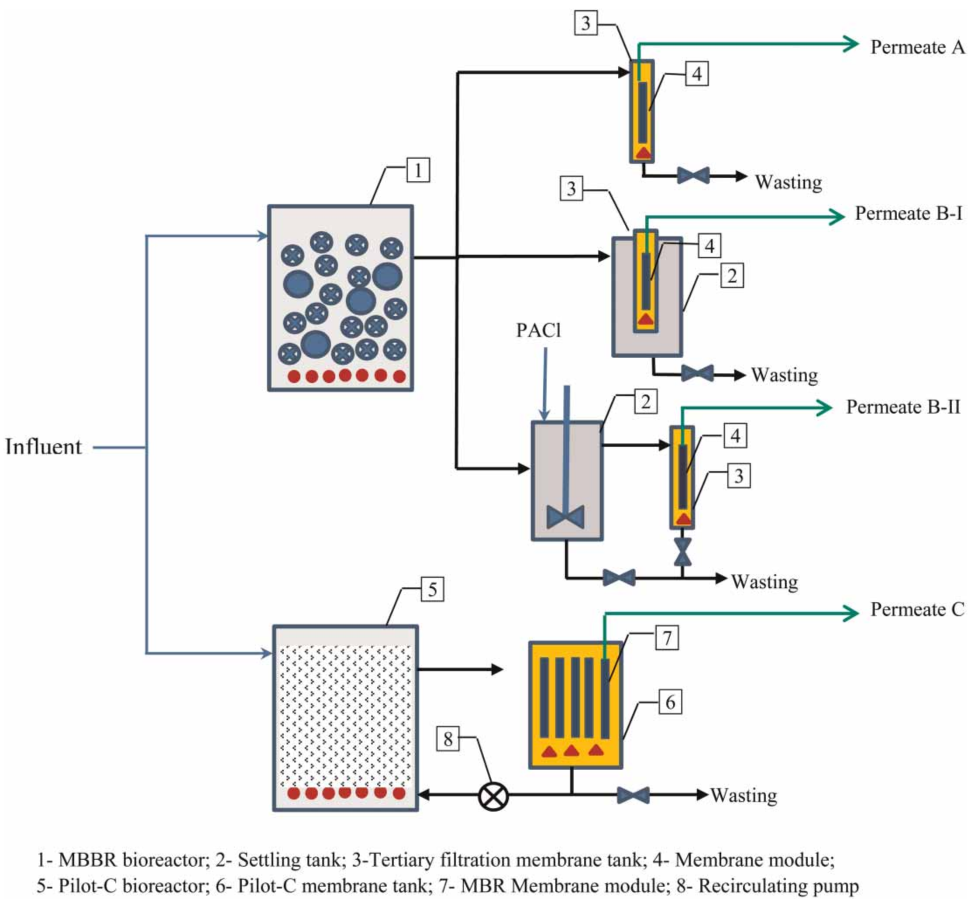

Figure 1 | Schematic diagram of experimental set-up.

Canada) were submerged in membrane tanks. The specifications of the pilot-scale units and membrane modules are summarized in Table 1.

Operation of the pilot units

To start the process, the two bioreactors (MBBR bioreactor and MBR bioreactor) were seeded with activated sludge from a local wastewater treatment plant and fed with municipal wastewater from the City of Burlington, ON, Canada. Fine bubble aeration was provided to the bioreactors for biological respiration and to maintain media in suspension in the MBBR bioreactor. The operational parameters are summarized in Tables 2 and 3. Membrane filtration was achieved by withdrawing permeates through hollow fiber membrane modules at a constant flux. The filtration cycle was a 12 min permeation period followed by 30 s of relaxation. Course bubble cyclic aeration (10 s ON/10 s OFF) was applied in the membrane tanks to mitigate membrane fouling.

Table 1 | Specifications of test units and membrane modules

Parameter	Unit	M-MBBR	MBR
Bioreactor tank volume	L	540	550
Membrane tank volume	L	10	250
Settling tank volume	L	100	–
Membrane nominal pore size	μm	0.4	0.4
Fiber outer diameter	mm	1.9	1.9
Fiber inner diameter	mm	0.9	0.9
Surface area per membrane module	m^2	0.72	0.9
Number of membrane modules		2	5

Table 2 | Operational parameters in the bioreactors of M-MBBR and MBR

	MBBR	MBR
Temperature (°C)	16.77 ± 2.05	16.50 ± 2.39
pH	7.42 ± 0.21	7.05 ± 0.17
DO (mg/L)	6.39 ± 1.17	3.99 ± 2.14
MLSS (mg/L)	123.64 ± 48.20	8,390 ± 1,386
MLVSS (mg/L)	104.51 ± 32.75	6,798 ± 1,126
Attached biomass (mg/L)	1,933 ± 645	–

Analytical methods

For wastewater and sludge samples, mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), chemical oxygen demand (COD), soluble COD (SCOD, filtered through 0.45 μm filters), filtered COD (filtered through 1.5 μm filters), alkalinity and time to filtration (TTF) were measured according to *Standard Methods* (APHA-AWWA-WEF 2005). Dissolved oxygen (DO) and pH were measured using a portable DO meter (Model 52, YSI Incorporated, USA) and a pH/ISE meter (ORION STAR A214, Thermo Scientific, USA). Total phosphorus (TP), ammonium nitrogen (NH_4-N), and nitrate (NO_3-N) were measured according to Hach Method 8190, 10023 and 10020, respectively (Hach, USA). Total nitrogen (TN) was measured by using a total nitrogen measuring unit (TNM-1) attached to a TOC analyzer ($TOCV_{CSH}$, Shimadzu, Japan). Total organic carbon (TOC) was measured using a TOC analyzer (TOC-V_{CPH}, Shimadzu, Japan). Particle size distribution was measured using a laser diffraction particle size analyzer with de-ionized water as the dispersant (Mastersizer 2000, Malvern Instruments Ltd, UK).

For the biomass attached to the media, 10 media were taken from the MBBR and dried to 105 °C overnight. The dried media were weighed by group and then cleaned in a solution of 1,000 mg/L NaOCl to remove biofilm, rinsed with de-ionized water and dried to 105 °C for 4 hours before reweighing. The average mass of biofilm as attached

Table 3 | Operational parameters of the three tested pilot units

	Pilot-A	Pilot-B		Pilot-C
		Mode I	Mode II	
Flux (LMH)	30.6, 37.4	30.6, 37.4	30.6, 37.4, 47.6	30.6, 37.4, 47.6
MLSS in membrane tank (mg/L)	340.0 ± 139.8	2,345.8 ± 1,325.7	727.2 ± 318.1	11,556 ± 2,152
PACl dosage (mg/L)	–	–	50	–
Wastage	Full membrane tank drain every 2 h	30 L/d from settling tank	30 L/d from settling tank, full membrane tank drain every 2 h	0–50 L/d[a]

[a]Maintain 8 g/L MLSS in the bioreactor.

growth was extrapolated to the total number of carriers in the bioreactor to estimate the attached biomass in the MBBR.

Soluble microbial products (SMP) in the mixed liquor were measured as filtrate through Whatman 934-AH Glass Microfiber filters and calculated by summing the contents of the carbohydrate and protein fractions. Carbohydrates were measured according to Dubois *et al.* (1956) and proteins were measured according to modified Lowry's method (Lowry *et al.* 1951). Transparent exopolymer particles (TEP) concentration of filtered sludge and permeates were measured according to the protocol developed by De la Torre *et al.* (2008). In brief, 5 ml of prefiltered sludge sample was mixed with 0.5 ml of 0.55% (m/v) alcian blue solution and 4.5 ml of 0.2 mol/L acetate buffer solution (pH 4.0). The mixture was then stirred for 1 min and centrifuged (HERAEUS MEGAFUGE 16R, Thermo Scientific, USA) at 13,000 rpm for 20 min. TEP react with the alcian blue solution yielding a low solubility dye-TEP complex. The amount of the alcian blue in excess is determined by reading the absorbance of the centrifuged supernatant at 602 nm using a spectrophotometer (DR/4000U, Hach, USA). Xanthan gum was used for generating a calibration curve and the results are expressed in mg/L xanthan gum equivalent.

RESULTS AND DISCUSSION

Organic carbon, nitrogen and phosphorus removal

Table 4 shows water quality of the influents to the pilot units and the permeates from the M-MBBR and MBR during the experimental period. The influents were lower strength than typical municipal wastewater with an average COD of 169.02 mg/L. With various fluxes tested in the three membrane units, the COD loading rates in the MBBR and MBR varied from 0.39 to 0.54 kg COD/m^3d and from 1.21 to 1.32 kg COD/m^3d, respectively.

Table 4 | Water quality of the influent and permeates for the M-MBBR and MBR

Parameter	Influent	M-MBBR permeate	MBR permeate
COD (mg/L)	169.02 ± 60.33	12.58 ± 5.24	10.71 ± 5.10
TP (mg/L)	2.53 ± 0.91	0.44 ± 0.19	1.10 ± 0.22
NH_4-N (mg/L)	16.40 ± 6.42	0.14 ± 0.42	0.02 ± 0.05
NO_3-N (mg/L)	3.11 ± 1.57	20.37 ± 3.23	22.03 ± 5.61
TN (mg/L)	23.69 ± 10.73	20.29 ± 3.25	22.69 ± 6.64

The two systems M-MBBR and MBR both achieved excellent performance in organic carbon removal throughout varying influent COD concentrations with removal efficiencies of 91.56 ± 4.48% and 92.98 ± 3.97%, respectively. The results of organic substrate removal were slightly higher than previous results of 85–90% and 87% reported by Leiknes & Ødegaard (2007) and Ahl *et al.* (2006) in similar operating MBBR systems for treating municipal wastewater.

Removal of nitrogen was classified through measuring ammonia, nitrate and TN concentrations of the filtered influent and the permeate samples. It was shown that the average ammonia concentration of MBR permeates was much lower than that of M-MBBR permeates. The decreased nitrification process in the biofilm reactor (MBBR) is most likely attributed to the lower solids retention time with comparison to the suspended bioreactor (MBR). It was also shown that nitrate and TN concentrations were less in the MBBR reactors as compared to the MBR reactor. The increased nitrate removal (denitrification) in the MBBR might be associated with anoxic microzones that developed in the inner regions of the biofilm attached to carrier material.

Total phosphorus removal was found to be 85.05 ± 8.02% and 59.49 ± 7.74% with TP concentrations in the permeate of 0.44 ± 0.19 mg/L and 1.10 ± 0.22 mg/L for the M-MBBR and MBR, respectively. The lower TP concentrations in the MBBR permeate is associated with the higher volumes of wasting sludge from the M-MBBR units.

Profile of transmembrane pressure (TMP)

Figure 2 presents the profile of TMP changes in the three pilot units during the experimental period. In general, the TMPs in the M-MBBR units increased more quickly and required more frequent membrane cleanings in comparison to the MBR unit. This indicates that conventional MBR coupled with suspended biological process achieved better performance in terms of membrane fouling than M-MBBR system using attached biofilm process. The results are in agreement with a previous study suggesting that the mixed liquor of attached growth would have a higher fouling potential in comparison with that of suspended growth (Lee *et al.* 2001).

Fouling rates

The membrane modules were operated at various fluxes. Figure 3 compares the flux of three pilot units and their corresponding fouling rates. It can be seen that the fouling rates of M-MBBR pilot units without PACl addition were

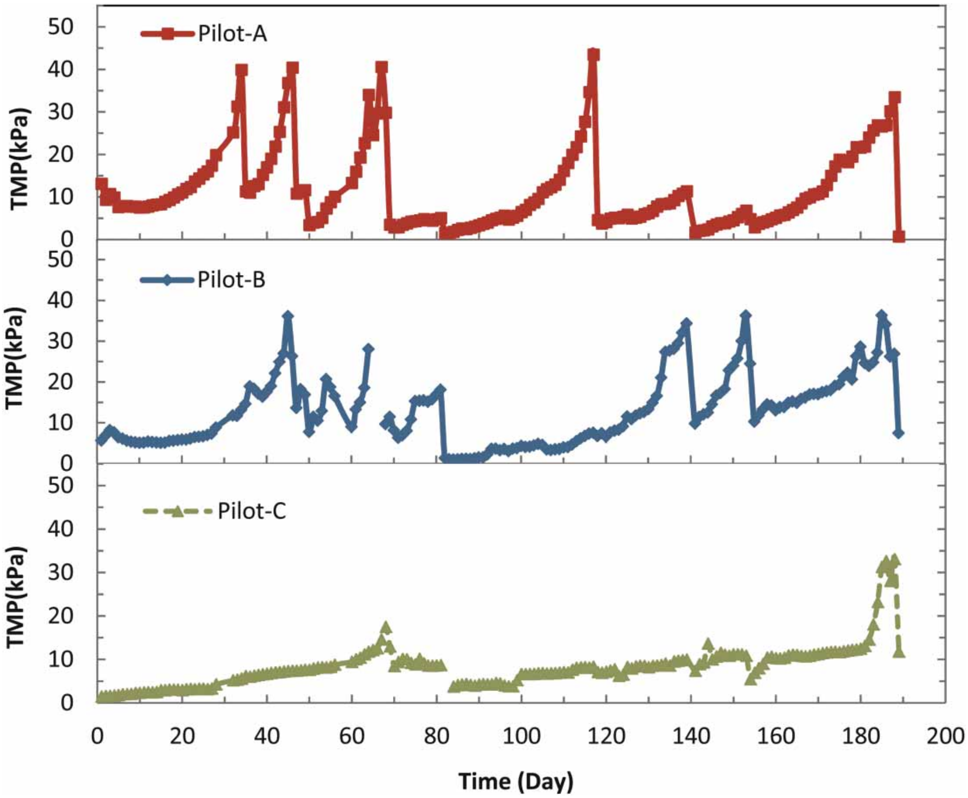

Figure 2 | TMP profile of the three tested pilot units.

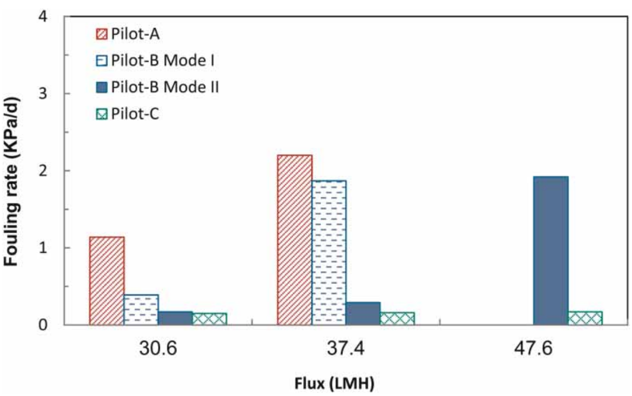

Figure 3 | Comparison of fouling rates between three pilot units.

considerably higher than those of the MBR unit at the tested fluxes. PACl addition could reduce the fouling rate of Pilot-B unit by 56.4% at the flux of 30.6 LMH, and by 84.5% at the flux of 37.4 LMH, respectively. However, the fouling rate of M-MBBR with PACl addition was comparable with the MBR only at the lowest tested flux 30.6 LMH. At the higher fluxes, even with the PACl addition, the fouling rates of M-MBBR were considerably higher than those of MBR.

Particle size distribution

The particle size distributions of suspended solids in the bioreactors and membrane tanks were measured and the resulted are shown in Figure 4. It can be seen from Figure 4(a) that there were more large size of flocs (>160 μm) in the MBBR bioreactor than that in the MBR bioreactor. In the study conducted by Yang *et al*. (2009b), it was claimed that the excess of filamentous bacteria in the MBBR caused the larger flocs in the MBBR in comparison to a conventional MBR. In this study, no filamentous bacteria were observed in the MBBR. The larger flocs in the MBBR likely came from the detachment of the biomass from the carriers. It can also be seen from Figure 4(b) that the pre-treatment by adding coagulant PACl in the settling tank slightly reduced floc size of the suspended solids in the membrane tank of Pilot-B in comparison to Pilot-A. Usually, the sludge suspension with larger floc size would benefit the membrane filtration process (Lim & Bai 2003). Whereas, in this study the MBBR sludge with larger floc size resulted in more severe membrane fouling, which indicated that other factors such as floc (colloidal) structure

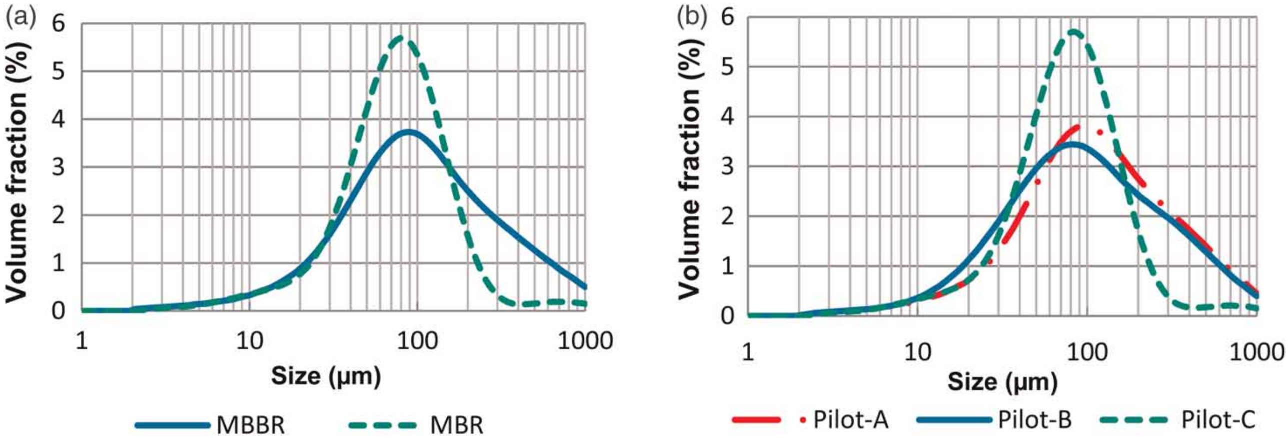

Figure 4 | Particle size distribution of the mixed liquor in (a) bioreactors and (b) membrane tanks.

and/or MLSS concentration might have greater impact on membrane fouling than floc size (Lee *et al.* 2001).

Comparison of SMP, colloidal TOC and TEP between the M-MBBR and MBR units

The concentrations of SMP colloidal TOC and transparent TEP in the mixed liquor from the membrane tanks of three pilot units were measured weekly. Table 5 summarizes the average concentrations. The SMP concentrations in the mixed liquor were measured as filtrate through 1.5 μm filters and calculated by summing the contents of the carbohydrate and protein fractions.

It can be seen that the concentrations of SMP, colloidal TOC and TEP in the M-MBBR systems were not considerably higher than those in the control MBR systems. The addition of PACl greatly reduced the colloidal TOC and SMP concentrations when Pilot-B was operated at mode II, which resulted in lower membrane fouling rates.

As shown in Figure 3, the fouling rates were much higher in the M-MBBR systems as compared to the conventional MBR systems. In contrast, it has been observed in previous studies that attached growth MBRs filtration performance was better than suspended MBRs (Sombatsompop *et al.* 2006; Jamal Khan *et al.* 2012). The previous two studies used synthetic wastewater in their bench-scale reactors; whereas real municipal wastewater was used in this study. Synthetic wastewater might produce different types of microbial products than real municipal wastewater, which could result in different membrane fouling phenomena. This indicates the sources and characteristics of SMP may play an important role in governing membrane fouling potential of the sludge generated from biological wastewater treatment processes. Furthermore, in the study of Jamal Khan *et al.* (2012) the membrane module was directly immersed in the biofilm reactor with media carriers. The collision between circulating media and hollow fibers mitigated cake formation on membrane fibers, which could be the major reason for the better filtration performance observed in the attached growth MBR (Lee *et al.* 2006).

CONCLUSIONS

The following conclusions can be drawn from this study through a side-by-side comparison of membrane performance in three pilot-scale experimental units.

Table 5 | Concentrations of SMP, colloidal TOC and TEP in the three pilot units

	Pilot-A	Pilot-B		Pilot-C
		Mode I	Mode II	
SMP (mg/L)	26.45 ± 5.35	36.22 ± 6.53	20.65 ± 5.44	34.18 ± 12.56
Colloidal TOC (mg/L)	5.75 ± 2.08	15.08 ± 5.73	3.12 ± 0.88	5.63 ± 3.92
TEP (mg/L)	11.37 ± 2.01	N/A	5.24 ± 3.04	16.78 ± 10.17

(1) Both types of membrane-based treatment processes i.e. M-MBBR and MBR, could produce excellent quality of effluents.
(2) More severe membrane fouling in the M-MBBR systems was observed as compared to the conventional MBR, under the experimental conditions of this study.
(3) Membrane fouling potential was related not only to the concentration of SMP, but also to their sources and characteristics.

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