

# A feasibility study of disinfection by-product precursor removal from surface water by a membrane bioreactor acclimatized with wastewater organic matter

Stephen M. Long and Hao L. Tang

## ABSTRACT

A membrane bioreactor (MBR) acclimatized with wastewater organic matter was employed for surface water treatment, and the feasibility of the MBR for the removal of disinfection by-product (DBP) precursors was studied. With a low pressure microfiltration hollow-fiber membrane module, a hydraulic retention time of 1.5 hours and a solids retention time of 180 days, the MBR was able to achieve 35% removal of trichloroacetic acid precursors and 21% removal of dichloroacetic acid precursors. The removal of trichloromethane (TCM) and brominated DBP precursors was unsatisfactory. The TCM yield and bromine substitution factors for trihalomethanes and dihaloacetic acids increased. The phenomena could be attributed to an extended treatment time for hydrophobic dissolved organic matter (DOM), production of soluble microbial products from biomass activities, and an increased Br/DOM ratio by the MBR. Since the MBR treatment would lead to the production of some new DBP precursors and a change of DOM composition, the toxic potency of the DBPs formed needs to be taken into consideration if this process is employed for surface water treatment.

**Key words** | biodegradation, brominated DBPs, DBP formation potential, dissolved organic matter, microfiltration, soluble microbial products

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## INTRODUCTION

Surface water pollution has become a more and more pronounced environmental and human health concern. Compared to groundwater, which is relatively free of organic loading from runoff, wastewater discharge and algal blooms, surface water contains much higher levels of dissolved organic matter (DOM), which has been associated with disinfection by-product (DBP) formation after chlorination in publicly owned treatment works (POTWs). Therefore, it is a great challenge for POTWs drawing surface water with high levels of DOM to meet the US EPA Stage 2 DBP requirements (US EPA 2006). Advances in removing DBP precursors such as DOM are needed.

Generally the DOM in surface waters contains about 50% humic substances (humic and fulvic acids) (Metsamuuronen *et al.* 2014) and possible wastewater effluent organic matter (EfOM) (Chen *et al.* 2009), both of which have been understood as causes of DBP formation after chlorination. DBP precursors are mostly associated with small compounds with molecular weights of less than 1 kDa (Chang *et al.* 2001). Conventional treatment processes

(e.g. coagulation and enhanced coagulation) at POTWs are effective in removing large and hydrophobic compounds, but are limited in eliminating small and hydrophilic organics that contribute more to DBP formation (Liu & Li 2015). The capability of POTWs employing conventional processes in relation to DBP precursor removal is thus questionable. Membrane-based processes such as microfiltration (MF), ultrafiltration, (UF), nanofiltration (NF), and reverse osmosis (RO) have shown to be effective in rejecting small organic molecules as a result of using a membrane as a physical barrier. Among them, the low pressure MF stands out as the most economic approach due to high fluxes, low pressures, and low energy consumption. With typical pore sizes from 0.1 to 1  $\mu\text{m}$ , MF membranes could theoretically reject bacteria (0.5–10  $\mu\text{m}$ ), cysts and oocytes (3–15  $\mu\text{m}$ ), and achieve complete retention of biosolids for adsorption and degradation of DOM (Bodzek *et al.* 2011; Luo *et al.* 2014). During filtration, a bio-layer can form on the membrane surface, functioning as a secondary barrier ahead of the membrane for the rejection of DOM. Thus, coupling MF

membranes with a bioreactor setting, i.e. a membrane bioreactor (MBR), allows the removal of DOM in both the liquid phase and the biofilm phase (Tsai *et al.* 2004; Williams *et al.* 2012), and has gained unprecedented popularity not only in wastewater treatment but also in drinking water production (Tian *et al.* 2008).

Superior effluent quality, small footprint, and reduced sludge production are commonly known advantages for an MBR employed for wastewater treatment. While these aspects might still be sound for drinking water production, there is one more important and beneficial perspective in this application: wash-out of bacteria can be avoided, allowing enrichment of promising bacteria for the biodegradation of humic substances and possible EfOM in surface water. Thus, DBP precursors might also be impacted by MBR treatment. Li & Chu (2003) operated a surface-water-fed-MBR for more than 500 days and achieved a certain degree of biological degradation on trihalomethane (THM) precursors. Since the DOM in surface water is at low levels and mostly refractory, Williams & Pirbazari (2007) used ozone and powder-activated carbon (PAC) in their MBR for enhanced THM precursor removal. However, when PAC addition was discontinued, the removal efficiency substantially declined. Ma *et al.* (2014a) further explored the characteristics of an MBR in surface water treatment and found refractory DOM could be degraded.

Nevertheless, there is inadequate information on operation of MBRs regarding acclimatization of slow-growing bacteria for satisfactory degradation of the refractory DOM in surface water. The removal profiles of DBP precursors by MBRs are also unclear. Since dichloroacetic acid (DCAA) and trichloroacetic acid (TCAA) have different formation pathways and precursors (Liu & Reckhow 2013), their precursors should be separately discussed. Considering that EfOM may contribute to a significant portion of DOM in surface water (Chen *et al.* 2009), the operation of an MBR acclimatized with wastewater organic matter might be helpful in enriching the bacteria capable of degrading refractory DOM originating from EfOM in surface water, and such impacts on DBP precursors need to be evaluated.

The objectives of this research are, therefore, (1) to explore the feasibility of operating an MBR acclimatized with wastewater organic matter for surface water treatment; and (2) to investigate the profiles of DBP precursors during such treatment. The study is beneficial for advancing DBP precursor control technologies in surface water treatment, and is helpful for POTWs that are subject to surface water pollution and struggling to meet the Stage 2 DBP requirements.

## MATERIALS AND METHODS

### MBR setup

An MBR (Figure 1) with a dimension of 27.7 cm × 7.0 cm × 29.0 cm (L × W × H) and a 1.8 L effective volume was built with acrylic plastic sheets. The membrane module in the MBR was assembled with hollow-fiber MF membranes with 0.4 μm pore size and a total surface area of 0.03 m<sup>2</sup>. A 1.5 L working volume maintained by a water level regulator led to a membrane surface/bioreactor volume ratio of 20 m<sup>2</sup>/m<sup>3</sup>. The reactor was placed in a water bath to control the temperature at 25 °C. Influent to the MBR and its effluent were facilitated by two peristaltic pumps. The effluent pump was operated on programmed cycles of 8 min filtration and 2 min relaxation. The pumping flow rate was adjustable, resulting in desired hydraulic retention times (HRTs) for different applications. The trans-membrane pressure (TMP) was monitored by a pressure gauge on the effluent tube between the membrane module and the effluent pump. Aeration was provided through nozzles beneath the membrane module to satisfy oxygen demand during biodegradation and to mix the liquid phase in the reactor. The air flow also provided a shear force through the membrane module to control the thickness of biofilm.

### MBR start-up

The start-up period of the MBR was essentially a 220-day period of the MBR operation for the treatment of primary effluent from a domestic wastewater treatment plant. Periodic sludge wastage was conducted on weekly basis to maintain a solids retention time (SRT) of 180 days. Regular non-chemical backwashes with membrane permeate were

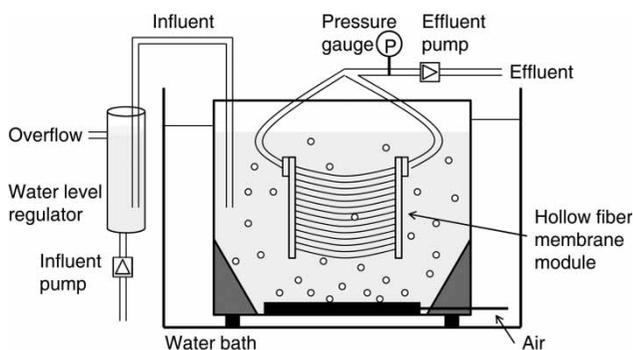


Figure 1 | Schematic diagram of the membrane bioreactor.

performed on a daily basis for TMP control, while off-line chemical backwashes with 1.5% sodium hypochlorite solution were only performed when recovery of TMP by regular backwashes was difficult. It was expected that the gel structure of the biofilm on the membrane could protect bacteria from chlorine attack (Metsamuuronen *et al.* 2014), and that bacteria could repopulate soon after backwashes. With an average HRT of 22.6 hours, the MBR had achieved steady removal of DOM from the wastewater. The start-up conditions of the MBR acclimatized with wastewater organic matter are summarized in Table 1.

### MBR treatment of surface water

Considering surface water contains low DOM for biodegradation compared to wastewater, a high flow rate was used for surface water treatment. When the MBR influent was switched to surface water, the average flow rate was adjusted to 33 L/m<sup>2</sup> h, resulting in an HRT of 1.5 hours. The surface water was obtained from a local reservoir and contained 4.78 mg/L dissolved organic carbon (DOC). The water quality parameters of the reservoir water are summarized in Table 2. Backwashes with membrane permeate were performed every 12 hours. Off-line chemical backwashes and sludge wasting were not performed during the 168 hour experimental period.

**Table 1** | Start-up conditions of the MBR acclimatized with wastewater organic matter

	Unit	Value <sup>a</sup>
Mixed liquor suspended solids	mg/L	2,180
Hydraulic retention time	hours	22.6
Solids retention time	days	180
Influent turbidity	NTU	45.9
Effluent turbidity	NTU	0.22
Influent chemical oxygen demand	mg/L	256
Effluent chemical oxygen demand	mg/L	17
Effluent dissolved organic carbon	mg/L	4.02

<sup>a</sup>Data presented were the averages of 77 samplings during a 220-day period of MBR operation treating domestic wastewater.

**Table 2** | Water quality parameters of the reservoir water

Parameters	pH	Temperature (°C)	Turbidity (NTU)	Fe (mg/L)	Mn (mg/L)	DOC (mg/L)
Reservoir water	7.94	18.7	19.4	0.52	0.154	4.78

### Chlorination and DBP analyses

Both influent and effluent during MBR treatment of the surface water were sampled for characterization of DBP precursors. The samples were filtered with 0.45- $\mu$ m membrane filters for quantification of DOC with a Total Organic Carbon Analyzer (Model 1010, O.I. Analytical, USA). A DBP formation potential (DBPFP) test was initiated by chlorinating 250 mL filtered samples at a Cl<sub>2</sub> dose of 20 mg/L, buffering at pH 7, and incubating in amber borosilicate bottles in the absence of light for 3 days at 20 °C. No headspace was allowed in the bottles, to minimize volatilization of the formed DBPs. After the chlorine residuals were measured by a DPD colorimetric method with a colorimeter (Model DR/890, HACH Co., USA) and quenched with ammonium chloride, the samples were extracted for DBP analyses following USEPA Methods 551.1 and 552.3. Details of sample extraction procedures and program settings for gas chromatographs can be found elsewhere (Tang *et al.* 2012). The analyzed THMs included trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and tribromomethane (TBM). The analyzed haloacetic acids (HAAs) included monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), DCAA, TCAA, bromochloroacetic acid (BCAA), and dibromoacetic acid (DBAA). The DBPFP test could maximally drive reactive DOM to form DBPs under controlled conditions, and the results were used to represent the amounts of precursors for different classes of DBPs.

## RESULTS AND DISCUSSION

### Formation of DBPs in chlorinated MBR influent and effluent

Concentrations of major DBP species after chlorination of MBR influent and effluent are shown in Figure 2. The results are the arithmetic means and standard deviations of 12 samplings throughout a 168 hour experimental period. TCM was found to be the most abundant species followed by TCAA and DCAA, and the relatively higher levels of TCAA

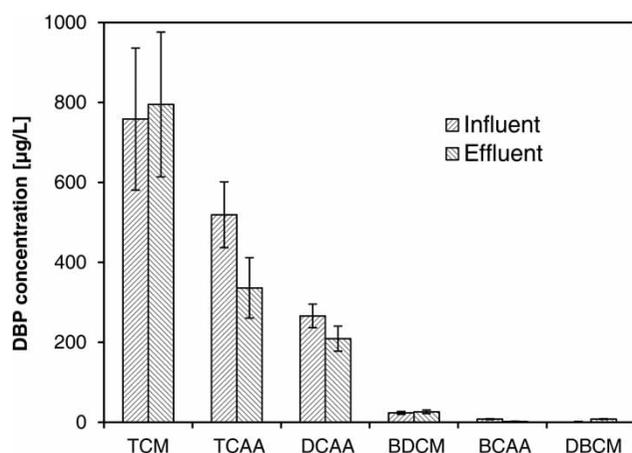


Figure 2 | Concentrations of DBP species in chlorinated MBR influent and effluent.

compared to DCAA were typical for surface water. Thus, the starting water possessed the following DBP precursors at high concentrations: TCM precursors ( $758 \pm 177 \mu\text{g/L}$ ), TCAA precursors ( $519 \pm 82 \mu\text{g/L}$ ), and DCAA precursors ( $266 \pm 30 \mu\text{g/L}$ ). Other DBPs were all brominated DBPs (Br-DBPs) including BDCM ( $24 \pm 4 \mu\text{g/L}$ ), BCAA ( $7.8 \pm 1.0 \mu\text{g/L}$ ) and DBCM ( $1.4 \pm 0.7 \mu\text{g/L}$ ) at much lower levels, indicating a low level of bromide as an inorganic DBP precursor in the surface water. The MBR treatment substantially reduced the formation of TCAA followed by DCAA. The TCAA and DCAA precursors decreased to  $336 \pm 76$  and  $209 \pm 31 \mu\text{g/L}$ , which were equivalent to average removal percentages of 35% and 21%, respectively. On the other hand, removals of other DBP precursors were not detected, and it was even noted that TCM precursors slightly increased to  $795 \pm 181 \mu\text{g/L}$  after the MBR treatment.

Mechanisms that lead to the preferential removal of TCAA over DCAA precursors and the non-removal of TCM precursors by MBR are complex. TCM and TCAA precursors are believed to be those organics that are more hydrophobic and recalcitrant to biodegradation than DCAA precursors (Liu *et al.* 2016). The mechanisms of MBR treatment involve size exclusion as well as electrostatic and hydrophobic effects (Siegrist & Joss 2012). A conventional idea would be that the TCM and TCAA precursors would be rejected by the membrane due to their hydrophobic nature, allowing more time for biological degradation by the slow-growing bacteria (Li & Chu 2003). In this MBR system, this theory could explain the preferential removal of TCAA over the more hydrophilic DCAA precursors, but could not explain the non-removal of TCM precursors. There must be other concurrent processes.

Further explorations were needed by taking into account the associated DOM that contributes to DBP precursors.

### DBP yields in chlorinated MBR influent and effluent

DBP yield was measured as the amount of formed DBPs per gram of DOC, and the parameter was able to describe the reactivity of DOM in forming DBPs. As shown in Figure 3, TCM yield increased from  $220 \pm 70$  to  $259 \pm 56 \mu\text{g/mg C}$ , while the TCAA and DCAA yields decreased from  $146 \pm 27$  to  $110 \pm 27 \mu\text{g/mg C}$  and from  $76 \pm 14$  to  $69 \pm 15 \mu\text{g/mg C}$ , respectively. It was obvious that the MBR treatment changed the composition of the DOM by making it more reactive in forming TCM and less reactive in forming TCAA and DCAA. The changed reactivity could be ascribed to the production of soluble microbial products (SMPs) in the MBR. SMPs are composed of utilization associated products (UAPs) and biomass associated products (BAPs). It is known that BAPs have high molecular weight polysaccharides and hydrophobic humic substances while UAPs are considered to contain more proteins and low molecular weight polypeptides and amino acids that are subject to further biodegradation (Ma *et al.* 2015). At a long SRT (180 days) in the MBR, the growth of bacteria that could prey on UAPs were enabled (Meng *et al.* 2012; Xie *et al.* 2012), leaving a majority of hydrophobic and humic BAPs in the effluent DOM that were more reactive in forming TCM.

### Bromine substitution factors (BSFs) of DBPs in chlorinated MBR influent and effluent

In addition to organic DBP precursors, their confounding effects with bromide – an important inorganic DBP

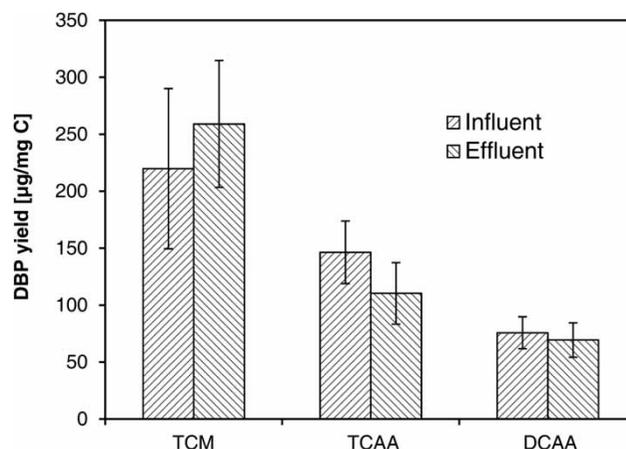


Figure 3 | DBP yields of chlorinated MBR influent and effluent.

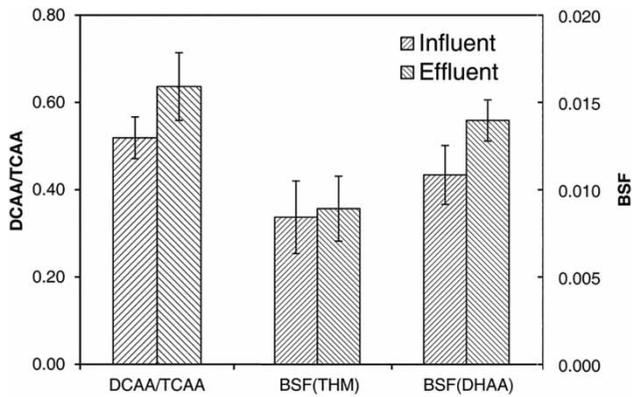


Figure 4 | DCAA/TCAA ratios and BSFs of DBPs in chlorinated MBR influent and effluent.

precursor – were explored by investigating the degree of bromine incorporation into DBPs following [Hua & Reckhow \(2012\)](#)'s method. Since the Br-DBPs are more toxic than their chlorinated analogues ([Hua et al. 2006](#)), less bromine incorporation is preferable as an indicator of reduced toxic potency. BSF, a fraction expressed between 0 and 1, was calculated by dividing the molar concentration of bromine incorporated into a given class of DBPs by the total molar concentration of chlorine and bromine in that class. Equations (1) and (2) present the algorithms for BSFs of THM and dihaloacetic acid (DHAA) calculations, and the results are shown in [Figure 4](#).

$$\text{BSF(THM)} = \frac{[\text{BDCM}] + 2[\text{CDBM}] + 3[\text{TBM}]}{3([\text{TCM}] + [\text{BDCM}] + [\text{CDBM}] + [\text{TBM}])} \quad (1)$$

$$\text{BSF(DHAA)} = \frac{[\text{BCAA}] + 2[\text{DBAA}]}{2([\text{DCAA}] + [\text{BCAA}] + [\text{DBAA}])} \quad (2)$$

Both BSFs showed increases, and the most significant increase was observed for BSF (DHAA). The MBR treatment increased the BSFs for DHAAs from  $0.011 \pm 0.0017$  to  $0.014 \pm 0.0012$ . This could be ascribed to the non-removal of bromide, the removal of DOM, and the resultant increased Br/DOM ratio. Since bromide is too small to be blocked by the pores of the membrane, it could flow unobstructed out of the MBR. As the rate of bromine-DOM reaction was approximately an order of magnitude higher than the chlorine-DOM reaction ([Hua & Reckhow 2012](#)), there would be more bromine substitution at higher Br/DOM ratios.

The DCAA/TCAA ratios shown in [Figure 4](#) also show the change of DOM relating to the tendency towards bromine incorporation. As the bromine incorporation into DHAAs was easier than that into trihaloacetic acids (THAAs) ([Hua & Reckhow 2012](#)), the increased

DCAA/TCAA ratio by the MBR treatment suggested preferential THAA precursor removal from DOM, leaving greater a proportion of DHAA precursors in the DOM that are more susceptible to bromine incorporation.

#### DBP fractions of chlorinated MBR influent and effluent

[Figure 5](#) presents the arithmetic means of 12 samplings relating to the fractions of individual DBP species after chlorination of MBR influent and effluent. Two increasing and two decreasing fractions are noted. The fractions of TCM and Br-DBPs increased from 48% and 2% to 58% and 3%, respectively, while the fractions of TCAA and DCAA decreased from 33% and 17% to 24% and 15%, respectively. The results were not surprising as they aligned well with previous discussions on the preferential removal of TCAA over DCAA precursors, non-removal of TCM precursors, and the generally increased Br-DBPs. Increased formation of Br-DBPs by MBR treatment was also reported by [Ma et al. \(2014b\)](#), who attributed this to a long SRT and the associated production of SMPs. Since the DBP profiles during MBR treatment were under the influence of SMP-excreting biomass, it would be beneficial to quantify such an effect to differentiate it from the effect of naturally occurring DOM in surface water. In the following section, we temporarily changed the MBR influent to distilled water to eliminate the contribution of DBP precursors from naturally occurring DOM in surface water.

#### Effect of biomass on MBR treatment of surface water

[Figure 6](#) presents the impact of biomass on DBP profiles of MBR effluent. All experimental conditions were kept the

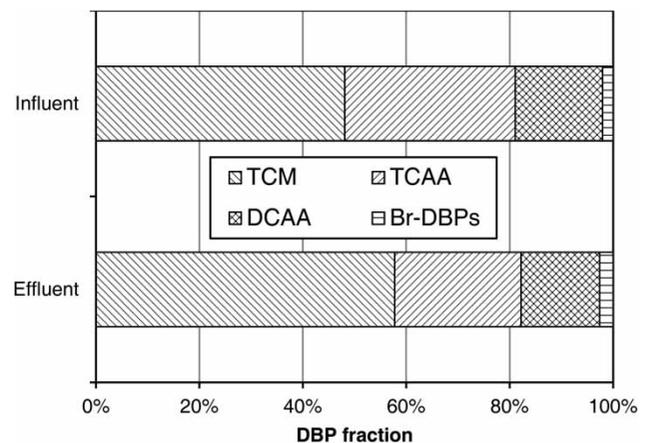


Figure 5 | DBP fractions of chlorinated MBR influent and effluent.

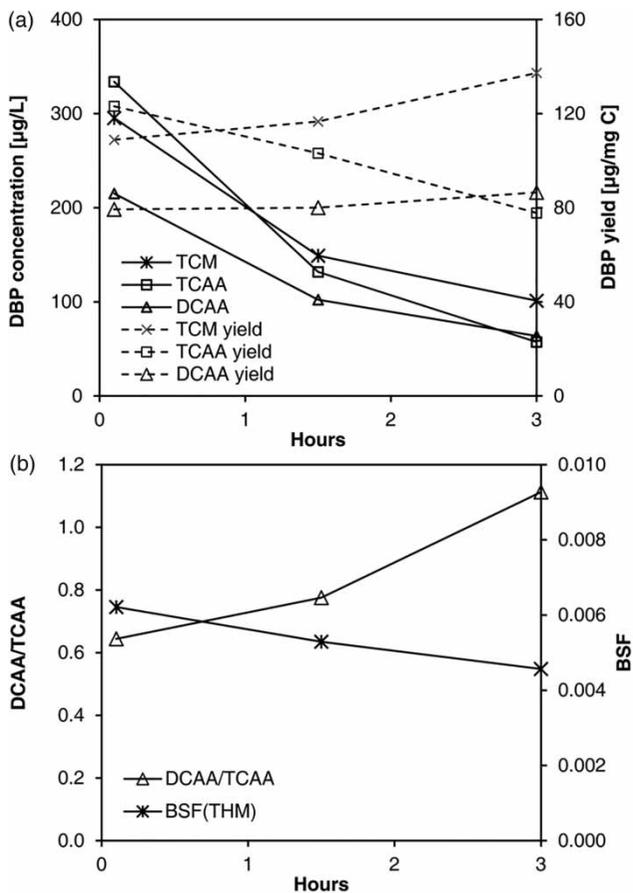


Figure 6 | DBP profiles of MBR effluent under the impact of biomass.

same (e.g. HRT = 1.5 hours) except the influent was changed from reservoir water to distilled water to eliminate the influence of naturally occurring DOM in surface water. It is clear from Figure 6(a) that the biomass contributed to significant amounts of TCM, TCAA and DCAA precursors. Their concentrations decreased consistently as time progressed, which was attributable to a dilution effect by the influent distilled water. However, the DBP yields gave clear indications of the increase in TCM yield and decrease of TCAA yield, which could be ascribed to neither dilution nor the naturally occurring DOM in surface water. It was deduced that either new TCM precursors were generated from the biomass or the non-TCM precursors (e.g. TCAA precursors) were preferentially removed from the DOM as a result of SMP production during biomass activities. Similar observations were observed by Liu & Li (2010) who reported increased DBPFP as a result of SMP production. It is interesting to note that the change of DCAA yield was marginal, suggesting the proportion of DCAA precursors in DOM

was less impacted by the SMP production. Although there could also be generation of new DCAA precursors with SMP production, there was neither accumulation nor deficit to imply a significant change of its proportion in effluent DOM.

The BSF for THMs decreased (Figure 6(b)), which was ascribed to decreased bromide concentration by dilution with the influent distilled water. The DCAA/TCAA ratio increased significantly from 0.64 to 1.11, suggesting preferential removal of TCAA over DCAA precursors from the SMP-DOM by the MBR as a result of extended treatment time for TCAA precursors.

### Implications

The results of the study are meaningful for the water industry considering process upgrades for effective removal of DBP precursors from surface water. By enriching bacteria from wastewater organic matter with an MBR, it was our hypothesis that the refractory DOM in surface water, especially with an EfOM origin, could be degraded, and the removals of DBP precursors was possible. At the experimental settings described in this study, satisfactory DBP precursor removal, however, was not obtained. Instead, increased TCM and Br-DBP precursors were observed, along with certain removal of TCAA and DCAA precursors. The change of DBP profile by the MBR treatment suggests the necessity to re-explore the mechanisms of MBR treatment regarding different classes of DBP precursors. As a result of the hydrophobic, aromatic, and high molecular natures of TCM and TCAA precursors, the MBR treatment did lead to extended treatment time on the two precursors by membrane rejection compared to DCAA precursors. In the meantime, the MBR treatment also led to the production of SMPs, which eventually contributed to more TCM precursors as constituents of BAPs, while others were probably present as biodegradable intermediates and were associated with constituents of UAPs. In addition, the toxic potency of the MBR effluent was not alleviated due to the limitations of MF membranes on removal of bromide and its associated Br-DBPs. Therefore, if MBRs are considered for the removal of readily biodegradable DOM and the control of DBP precursors from surface water, one needs to factor in the production of SMPs. Moreover, to reduce the toxic potency of the formed DBPs, additional processes (e.g., capacitive deionization (Liu *et al.* 2016), electrolysis and volatilization (Kimbrough *et al.* 2012), etc.) that are capable of bromide removal prior to disinfection would be preferable.

## CONCLUSIONS

Operation of a wastewater-organic-matter-acclimatized MBR with a low pressure MF membrane module for surface water treatment was able to achieve 35% removal of TCAA precursors and 21% removal of DCAA precursors. The removal of TCM and Br-DBP precursors was unsatisfactory. The TCM yield and BSFs for THMs and DHAAs increased. The phenomena could be attributed to an extended treatment time for hydrophobic DOM, production of SMPs from biomass activities, and an increased Br/DOM ratio by the MBR. Since the MBR treatment would lead to a production of some new DBP precursors and a change of DOM composition, the toxic potency of the formed DBPs needs to be taken into consideration if this process is employed for surface water treatment.

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