Biofouling Control of a Forward Osmosis Membrane during Single-pass Pre-concentration of Wastewater

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Abstract

Pre-concentration of wastewater using a forward osmosis (FO) membrane prior to processing by an anaerobic digester can enhance biogas production. However, biofouling caused by microbes in wastewater remains a challenge. The study aimed to evaluate the efficacy of chloramination in mitigating the biofouling of an FO membrane during a single-pass concentration of primary wastewater effluent. Pre-disinfection at a chloramine dose of 22–121 mg/L successfully alleviated membrane fouling. Bacterial cell counts in the feed and concentrate showed that most of the bacterial cells in the wastewater were trapped on the membrane surface or spacer. The FO membrane surfaces in non-chloraminated/chloraminated systems were fully-covered by intact/damaged bacterial cells, respectively, indicating that chloramination effectively mitigated biofouling. However, due to high permeate-recovery and low cross-flow velocity in a single-pass concentration process, organic fouling on the membrane surface (and possibly on the interior wall of the membrane-pores) appeared to cause a gradual reduction in permeate-flux. This study demonstrated successful biofouling control using chloramination during a single-pass and high-recovery pre-concentration of primary wastewater effluent.

Keywords: Biocide; biofouling; FO membrane; chloramine; wastewater treatment.

1. Introduction

Reclamation of biogas by treating municipal wastewater is an attractive approach in reducing overall energy consumption. Anaerobic biogas at a wastewater treatment plant is typically produced through a digester that can generate methane gas from organic substances in wastewater (Appels et al., 2011). High concentrations of organic substances in wastewater lead to higher methane gas production, whereas typical municipal wastewater has low organic concentrations. Therefore, deploying a pre-concentration process prior to anaerobic digestion is crucial in improving feasibility of biogas production from municipal wastewater. As an effective pre-concentration technology, forward osmosis (FO) membrane treatment has attracted considerable research interest (Lutchmiah et al., 2011; Lutchmiah et al., 2014; Ansari et al., 2015; Onoda et al., 2016b; Ansari et al., 2017; Onoda et al., 2017). FO membranes, typically made of polyamide (PA) or cellulose triacetate (CTA), allow water transportation from a low-salinity solution (feed solution, e.g., wastewater) to a high-salinity solution (draw solution), concentrating the low-salinity solution (Wang et al., 2014). Cities near the ocean have an advantage of utilizing abundant seawater as the draw solution. Pre-concentration of wastewater using FO membranes has been successfully demonstrated in literature (Chen et al., 2014; Zhang et al., 2014; Ansari et al., 2018), but it still faces a challenge of reduction in permeate-flux caused by membrane fouling. In addition to organic fouling (Mi and Elimelech, 2008, 2010; Onoda et al., 2016a; Chun et al., 2017; Ly et al., 2019), biofouling is a dominant fouling mechanism (Qasim et al., 2015).

Biofouling of the FO membrane during pre-concentration of wastewater can be controlled by disinfecting feed-water prior to the FO process (Firouzjaei et al., 2019), e.g., continuous chlorination of wastewater effectively slows down biofouling on the FO membrane surface (Xue et al., 2016). However, in ammonia-containing wastewater, continuous chlorination

43 with free chlorine can only be performed at high chlorine doses, as free chlorine can be
44 present only after exceeding breakpoint chlorination, which depends on ammonia
45 concentrations. Other potential chemicals include hydrogen peroxide (H_2O_2) and ozone,
46 which oxidize and destroy the microbial community in wastewater (Wang et al., 2017;
47 Firouzjaei et al., 2019). All these strong oxidants (i.e., chlorine, H_2O_2, and ozone) need to be
48 quenched prior to the FO membrane to avoid oxidative damage; thus, disinfecting effects
49 cannot remain on the FO membrane surface.

50 Chloramine, a weak disinfectant, is known to be continuously applied to both PA- and CTA-
51 based membranes without notable damage to their performance. In water recycling systems
52 comprising reverse osmosis (RO) membranes, chloramination has been established as a
53 standard pre-disinfection process for biofouling mitigation (Fujioka et al., 2012; Farhat et al.,
54 2018). Our previous study (Fujioka et al., 2018) applied chloramination to alleviate FO
55 membrane fouling during the concentration of primary wastewater effluent. Despite its
56 successful application, biological growth in closed-loop recirculation systems could differ
57 considerably from full-scale systems, as they are typically based on single-pass
58 configurations to treat the continuous inflow of wastewater. Almost all other studies have
59 used similar recirculating systems to address the limited volume of actual wastewater
60 available in laboratories (Yang et al., 2019; Jung et al., 2020). Therefore, the feasibility of
61 chloramination as a pre-disinfection technique for wastewater pre-concentration must be
62 evaluated to simulate a single-pass treatment system.

63 This study aimed to evaluate the efficacy of chloramination in mitigating biofouling of a
64 CTA FO membrane during a single-pass concentration of primary wastewater effluent. The
65 effect of chloramination on biofouling mitigation was evaluated based on permeate-flux, feed
66 water quality (bacterial concentrations and organic characteristics), and bacterial state on the

membrane surface. The ultimate aim of this study was to establish a feasible disinfection approach for wastewater pre-concentration prior to anaerobic digestion.

2. Materials and methods

2.1 Chemicals

The study made use of the chemicals NaCl, NH_4Cl, NaOH, NaOCl, and sodium bisulfiteused in this study were purchased from FUJIFILM Wako Pure Chemical Corporation (Osaka, Japan). Artificial seawater was used as the draw solution (DS), prepared at 3.5 % weight by weight (w/w) NaCl in pure water. Pure water was produced by filtering tap water by a reverse osmosis treatment system (RTA-200W, AS ONE, Osaka, Japan). We used pre-filtered primary wastewater effluent as feed solution (FS). The pre-filtered primary wastewater effluent was prepared by filtering the primary effluent, collected at a municipal wastewater treatment plant in Japan, using a cellulose filter-paper with retention particle size of 7 μm (No. 5A, Advantec; Tokyo, Japan). Electrical conductivity, total organic carbon concentration, and pH of the pre-filtered primary wastewater effluent were 1.4 mS/cm, 6.6 mg/L, and 7.5, respectively.

2.2 Membrane treatment system

The flat sheet FO membranes used in this study were a CTA active layer embedded on a polyester (PES) screen mesh (Fluid Technology Solutions; Albany, OR, USA). One CTA FO membrane with an effective surface area of 60 cm^2 was enclosed in an acrylic membrane cell (C10-T, Nitto Denko, Japan). The DS stream had an effective gap of 5.0 mm without a spacer. The FS stream had a spacer between the membrane surface and membrane cell to maintain a channel gap of 1.2 mm. A bench-scale FO system used in this study **(Figure 1)** comprised a quantitative liquid-feed pump for feeding FS (MP-2000, Tokyo Rikakikai; Tokyo, Japan); a

diaphragm pump for feeding DS (DCP 8800, Aquatec International; CA, USA); an electromagnetic metering pump (EH-B10VC, IWAKI, Tokyo, Japan) with a control unit for dosing a NaCl stock solution in DS; a two hanger quantitative liquid-feed pump for feeding chemicals used for the formation of chloramine (MP-2000, Tokyo Rikakikai; Tokyo, Japan); two digital balances (EK-4100i and EK-610i, A&D Company; Tokyo, Japan) to measure the weight of feed and concentrate solutions; an 8-L styrofoam DS reservoir; a 2-L glass beaker as a feed storage; a 500-mL glass beaker for concentrate storage; and a temperature circulator (Thermax TM-1A, AS ONE; Osaka, Japan). Chloramine was pre-formed by mixing a solution of NH_4Cl and another containing NaOCl and NaOH, which were kept at 4 °C using a chiller unit (ACE-1100, Tokyo Rikakikai Co. Ltd.; Tokyo, Japan). The monochloramine stock solution thus formed was dosed into the FS stream located prior to the entry of the membrane cell.

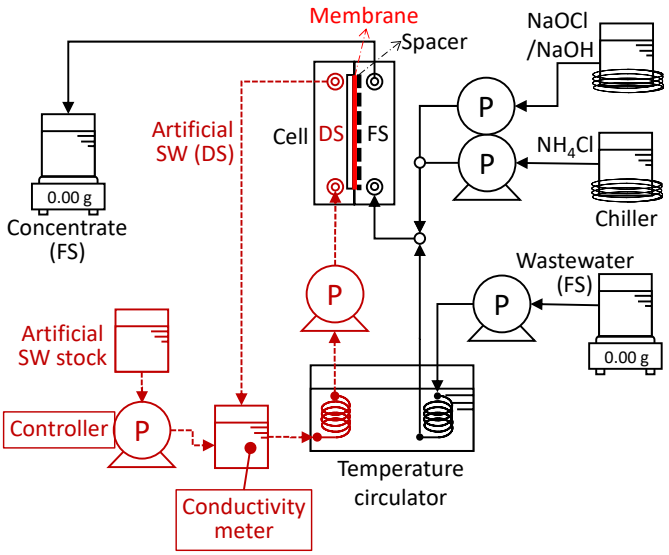


Figure 1. Schematic diagram of the Forward osmosis (FO) system.

2.3 Experimental protocols

Disinfection tests at various chloramine doses (5, 10, 20, and 40 mg/L) were performed using the pre-filtered primary wastewater effluent at room temperature (20 °C). After 10 min of reaction time, a sodium bisulfite solution was dosed at 2 mM to quench the residual

chemicals. Thereafter, the samples underwent heterotrophic plate counting of bacteria. The chloramination period was determined to be 10 min, as it was the minimum retention time of the feed solution in the membrane cell that achieved over 50 % permeate recovery in the FO system.

Similar to previous studies (Yoon et al., 2013; Yang et al., 2019), this study evaluated the effect of chloramination on membrane fouling during forward osmosis membrane treatment in terms of permeate flux, feed water quality, and bacterial state on the membrane surface. Prior to each treatment experiment, the system was operated to ensure stability using pure water in FS and artificial seawater (3.5 % NaCl) in DS for 2 h. During stabilization, the pure water permeate-flux was recorded. Thereafter, the FO treatment test was carried out by replacing the FS from pure water with pre-filtered primary wastewater effluent. Unless specified, each test was continued for 4 days. Throughout the tests, the cross-flow rates in the FS and DS were maintained at approximately 1.1 mL/min and 500 mL/min, corresponding to a cross flow velocity of approximately 0.5 and 46 mm/s in FS and DS, respectively. Throughout the tests, conductivity in the DS was maintained constant at 57 ± 0.5 mS/cm by dosing the NaCl stock solution into the DS reservoir. The temperature of the FS and DS was maintained at 25 ± 1 °C using a temperature circulator. Pre-formed chloramine solution (or pure water for control test) was continuously dosed at 0.1 mL/min, intended to achieve a varied monochloramine dose of 0, 22, 51, or 121 mg/L in the FS stream. The chloramine solution flow rate accounted for approximately 10 % of the overall FS flow rate. The FS in the FS reservoir (i.e., pre-filtered primary wastewater effluent) was supplemented once a day. After the end of each test, the FS was replaced from the treated wastewater with pure water, and the system was operated for 1 h to measure pure water flux. Thereafter, each membrane was removed from the membrane-cell for membrane surface characterization.

The bulk reverse salt flux J_S (g/m^{2}h), a salt passage from the DS to the FS, was calculated using the following formula:

$$J_s = 0.46[(C_{FSc} \times F_{FSc}) - (C_{FS} \times F_{FS}) - (C_M \times F_M)]/A, \quad (1)$$

where C_{FSc}, C_{FS}, and C_M (mS/cm) are the conductivities while F_{FSc}, F_{FS}, and F_M (L/h) are the flow rates of the FS concentrate, FS, and chloramine solution, respectively; A (m^2) is the membrane surface area; and 0.46 (g/L) is the conversion coefficient of conductivity (mS/cm).

2.4 Analysis

Conductivity and pH were measured using an Orion Star™ A325 (Thermo Fisher Scientific, MA, USA). Total organic carbon (TOC) concentrations were analyzed using a TOC-VSH analyzer (Shimadzu, Kyoto, Japan). Monochloramine concentrations were determined using a portable colorimeter (DR900, Hach, Loveland, CO, USA) with Monochlor F reagent pillows.

The heterotrophic plate count (HPC) method was used to determine the number of total viable bacteria. A diluted sample (1 mL) was added to the R2A medium (Nissui Pharmaceutical Co., Tokyo, Japan). The plates were incubated at 35 °C and counted after four days. Intact and damaged bacterial cells in the feed and concentrate in the FS stream were counted using a fluorescence microscope (BZ-X800, Keyence Co., Osaka, Japan). Each sample was stained with the LIVE/DEAD BacLight Bacterial Viability Kit (Thermo Fisher Scientific, Waltham, MA, USA) for 15 min. The stain-kit used in this study contained SYTO®9, a green fluorescent nucleic acid that only stains cells with intact membrane, and propidium iodide, a red fluorescent nucleic acid that only stains cells with damaged membranes. The stained sample was filtered using a track-etched polycarbonate MF filter with 0.2 μm pore size (Merck, Tokyo, Japan). The filter sample was analyzed using a

fluorescence microscope using a green or red filter with Ex wavelengths of 470±40 nm and 545±25 nm and absorption wavelengths of 525±50 nm and 605±70 nm, respectively.

Fouled FO membranes were analyzed using a fluorescence microscope (BZ-X800, Keyence Co., Osaka, Japan). The FO membranes were submerged in 2 mL of diluted LIVE/DEAD BacLight Bacterial Viability Kit (Thermo Fisher Scientific, Waltham, MA, USA) for 15 min, and analyzed using the same green and red filters as described above. Organics in the FS and DS were evaluated using the excitation emission matrix (EEM) fluorescence spectra using an RF-6000 spectrophotometer (Shimadzu Co., Kyoto, Japan). Solutions of FS, FS concentrate, and DS were directly analyzed without pre-treatment. EEM fluorescence spectra of membrane foulants were obtained after removing the foulants from the membrane surface and rigorously mixing with a 10 mL solution for analysis.

3. Results

3.1 Pre-disinfection

Chloramination of the pre-filtered primary wastewater effluent at monochloramine doses of 5–40 mg/L showed that chloramination is a powerful disinfection technique for treating primary wastewater effluent. Chloramination at the lowest chloramine dose (10 mg/L) reduced viable bacterial counts from 2.4×10^5 to 2.0×10^2 CFU/mL, achieving > 99.9 % (3-log) inactivation efficiency **(Figure 2)**. Higher chloramine doses of 20 or 40 mg/L chloramine doses achieved further reduction in bacterial counts to 1.6×10^2 or 58 CFU/mL, respectively. Thus, a chloramine dose of > 20 mg/L was determined as the minimum chloramine dose in this study, and a varied chloramine dose of 22, 51, or 121 mg/L was applied during a single-pass concentration of the pre-filtered primary wastewater effluent in the following section. It should be noted that elimination of bacteria (i.e., 0 CFU/mL) was not achieved with decent

concentrations of chloramine in water treatment (< 40 mg/L). The biofilm layer on the membrane surface can form as a result of bacterial attachment and their multiplication (Mansouri et al., 2010). Unless complete bacterial inactivation flowing into the membrane surface is continuously achieved, the remaining bacteria can trigger biofilm formation (Flemming et al., 1997). As chloramine, which damages nucleic acids or DNA of bacterial cells, inactivates bacteria slowly (Fetner, 1962; Shih and Lederberg, 1976), rapid proliferation of bacteria and consequent biofouling is expected to be inhibited by maintaining residual chloramine concentrations at the FO membrane surface. Therefore, the effect of chloramine concentrations on biofouling mitigation during a pre-concentration of primary wastewater effluent was evaluated at three different chloramine doses (i.e., 22, 51, and 121 mg/L).

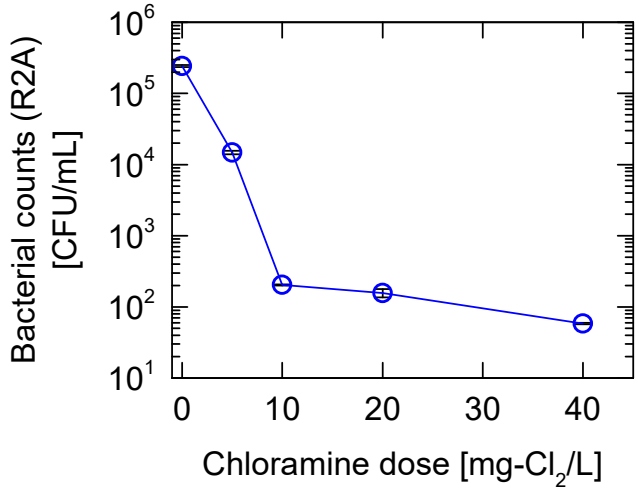


Figure 2. Bacterial counts as a function of chloramine doses (reaction time of 10 min).

3.2 Fouling mitigation during single-pass concentration

Fouling mitigation levels by chloramination were evaluated by tracking a reduction in permeate-flux (i.e., permeate volume per unit area and time) in the non-chloraminated and chloraminated FO systems. As the speed of membrane fouling can change according to the volume permeated through the membrane rather than the treatment time, the permeate-flux

reduction levels were evaluated at the cumulative permeate volume. Over the course of the four-day test, membrane fouling in the non-chloraminated FO system progressively occurred **(Figure 3)**. After a cumulative volume permeation of 2.5 L, the permeate-flux decreased from 7.5 to 4.2 L/m^{2}h, which accounted for a 44 % flux reduction. In contrast, the FO system at a chloramine dose of 22 mg/L showed a lesser reduction in permeate-flux from 7.5 to 4.9 L/m^{2}h, which accounted for 35 % flux reduction.

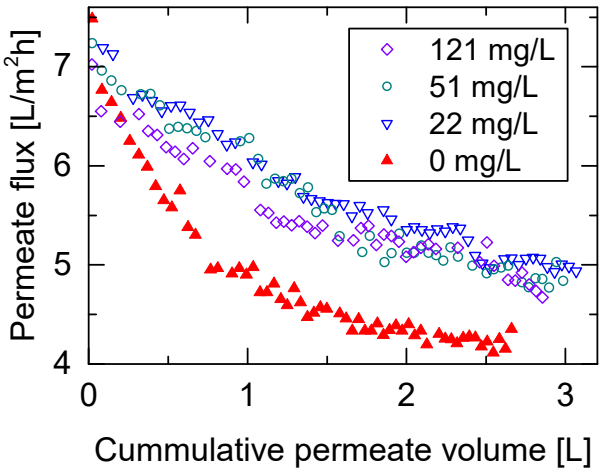


Figure 3. Changes in permeate-flux as a function of cumulative permeate volume during the pre-concentration of the pre-filtered primary effluent by the forward osmosis (FO) membrane with different chloramine doses (plot every 2 h).

Membrane fouling inhibited the permeation of water through the FO membrane; thus, permeate recovery in the non-chloraminated and chloraminated FO systems progressively decreased over the course of the four-day test **(Figure S1)**. The decreased permeate recovery from 80 % to 45–47 % corresponds to a decreased concentration ratio from 4–5-fold to approximately 2-fold. Higher chloramine doses of 51 and 121 mg/L showed fouling trends similar to that of 22 mg/L, indicating that a 22 mg/L chloramine dose may be sufficient for controlling membrane fouling for this specific primary wastewater effluent. The reduction in permeate-flux by membrane fouling was likely caused by the reduced effective driving force of water transport from the FS to DS—osmotic pressure difference ($\Delta\pi$). The fouling layer deposited on the membrane surface can induce the external concentration polarization of salts

due to the hindered back-diffusion from the membrane surface to bulk FS solution (Lee et al., 2010; Li et al., 2012). It enhances the osmotic pressure at the interface between the fouling layer and membrane surface; thus, the effective osmotic pressure difference is reduced **(Figure 4)**.

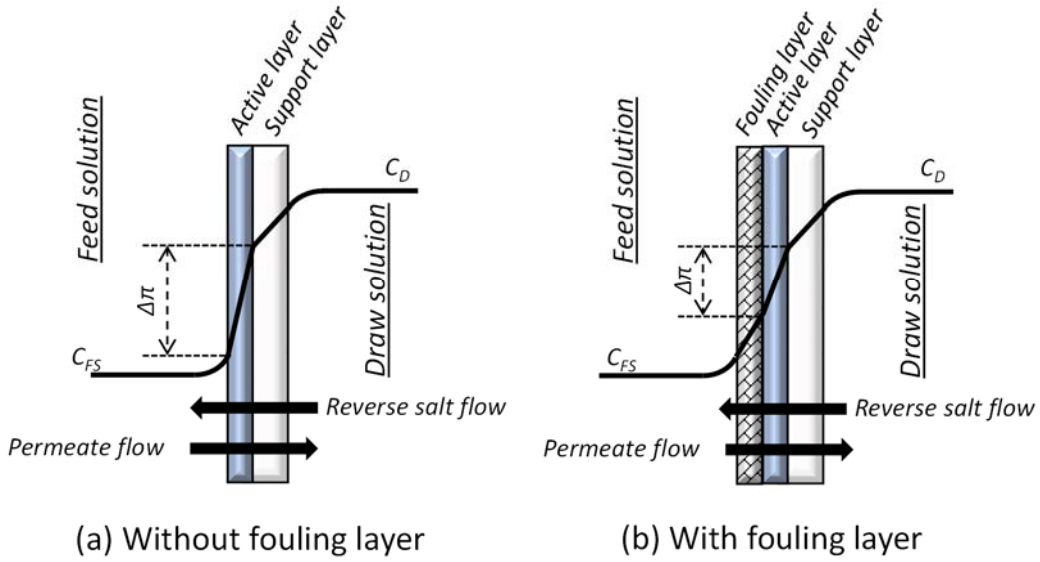


Figure 4. Conceptual images of osmotic pressure differences (a) Without fouling layer and (b) With fouling layer (adapted from (Li et al., 2012)).

To evaluate the impact of chloramine doses on fouling mitigation, the ratio of permeate-flux recorded at a cumulative permeate volume of 0.1 L ($J_{v=0.1}$) and 2.5 L ($J_{v=2.5}$), corresponding to initial and late stages of the test, respectively, was also compared **(Figure 5)**. The fouling levels at chloramine doses of 22 and 51 mg/L were almost equivalent. The highest chloramine dose of 121 mg/L resulted in the highest normalized permeate-flux of 0.8 (i.e., least fouling). Although chloramine doses of less than 22 mg/L may effectively inhibit biofouling, the optimization of chloramine doses for this specific pre-filtered primary wastewater effluent was beyond the scope of this study. Disinfection efficiency of chloramination can vary depending on the operating conditions such as contact time, concentration, and temperature (Wolfe et al., 1984). Therefore, the optimization of chloramine doses will be conducted at the pilot scale in our future study.

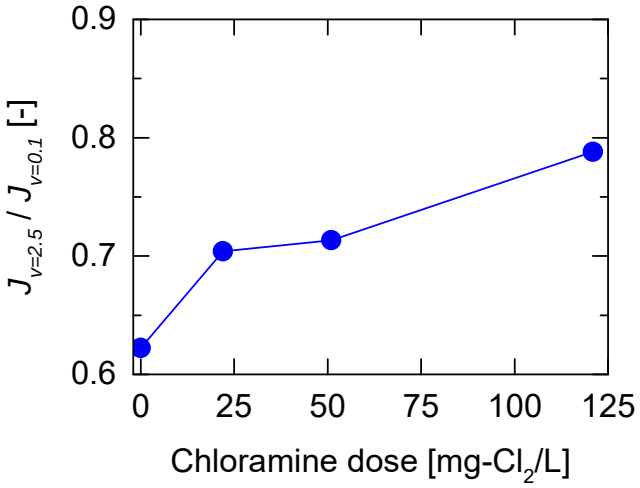


Figure 5. Normalized permeate-flux expressed as the ratio between the cumulative permeate volume (v) of 2.5 L ($J_{v=2.5}$) and 0.1 L ($J_{v=0.1}$) as a function of chloramine dose.

3.3 Water quality in the concentrate

The conductivity of the FS concentrate is dependent on the concentration effect of the FS and the reverse salt flux. The chloraminated FO system showed a higher permeate recovery (i.e., more concentrated) and underwent an addition of chloramine; thus, the FS concentrate conductivity in the chloraminated FO system was expected to be higher than that in the non-chloraminated FO system. However, at 23 h when the fouling development had already occurred, FS concentrate conductivity in the non-chloraminated FO system (5.3 mS/cm) was higher than that in the chloraminated FO system with a chloramine dose of 22 mg/L (4.4 mS/cm) **(Figure 6a)**. The non-chloraminated FO system showed a reverse salt flux of 5.4 g/m^{2}h, higher than the chloraminated FO system (1.7 g/m^{2}h) at 23 h **(Figure 6b)**. Minor changes in reverse salt flux were observed over the course of the last three days of the test. It is important to note that salinity in the FS concentrate can negatively affect biological activity in the following biogas digester. Thus, the chloraminated FO system has two advantages over the non-chlorinated FO system: higher permeate-flux (i.e., higher concentration of organics) and lower salinity in the FO concentrate.

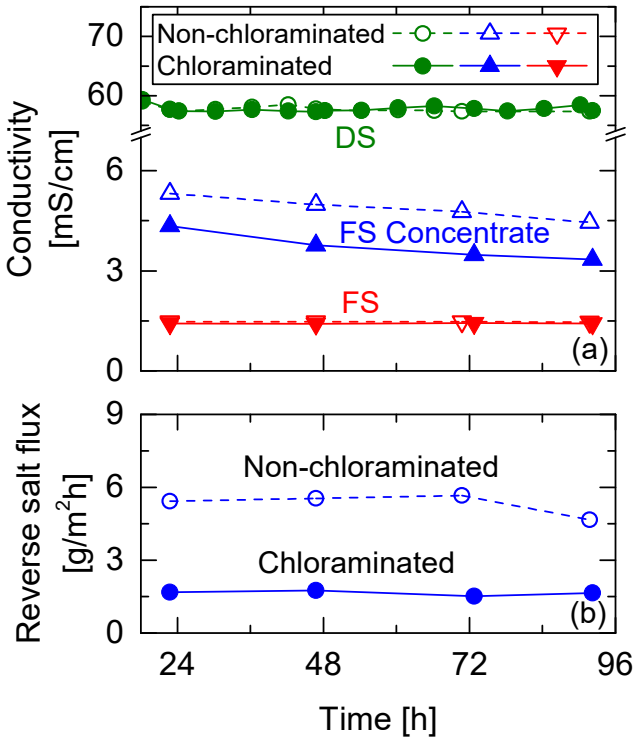


Figure 6. (a) Conductivity in the feed solution (FS), FS concentrate, and draw solution (DS) in the non-chloraminated and chloraminated (dose = 22 mg/L) forward osmosis (FO) systems, and (b) Their reverse salt flux.

3.4 Membrane fouling

The formation potential of a biofouling layer on the FO membrane surface was evaluated based on the difference in bacterial counts between the inflow (FS feed) and outflow (FS concentrate) of the membrane cell. For this evaluation, another set of tests with non-chloraminated and chloraminated FO systems (chloramine dose = 21 mg/L) were conducted **(Figure S2)**, and the samples for bacterial counts were collected at a sampling period of 18 h. As a result, viable bacterial counts in the non-chloraminated FO system considerably decreased from 2.4×10^5 (FS feed) to 45 CFU/mL (FS concentrate) **(Figure 7a)**. Viable bacterial counts in the FS concentrate of the chloraminated FO system (70 CFU/mL) were equivalent to that of the non-chloraminated FO system. The results indicate that a majority of viable bacteria in their feed stream were trapped in the membrane cell. It should be noted that the plate counting method covers only bacteria forming a colony on the R2A medium, but a

vast majority of bacterial species do not form a colony. To cover almost all bacteria in water, epifluorescence microscopy was also used. As a result, the concentrations of damaged and intact bacterial cells exiting from the membrane cell (in the FO concentrate) were equivalent at 1.0–4.1×10^6 counts/mL **(Figures 7b and 7c)**. Considering the concentration factor of two in both systems, the results indicate that an equivalent number of bacteria had been trapped on the membrane surface or spacer implying that the occurrence of biofouling in FO systems depend on the bacterial state (active or inactive) on the membrane surface.

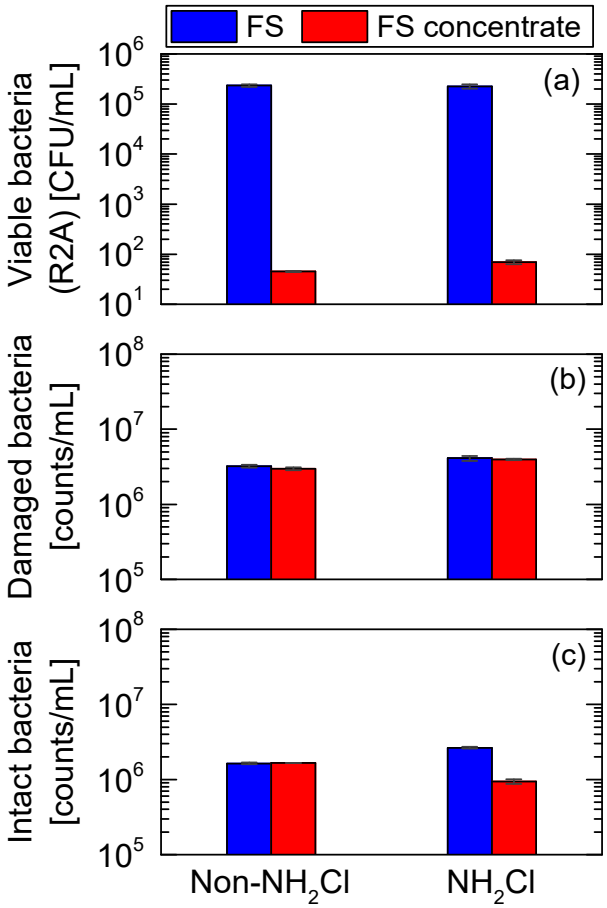


Figure 7. Concentrations of (a) viable bacteria, (b) damaged bacteria, and (c) intact bacteria in the feed solution (FS) and FS concentrate that were collected during the pre-concentration of primary wastewater effluent in the non-chloraminated and chloraminated (21 mg/L dose) forward osmosis (FO) systems at 18 h. FS feed in the chloraminated FO system was collected prior to chloramine addition.

The state of bacteria on the membrane surface was analyzed by differentiating intact and damaged bacterial cells deposited on the membrane surface. The FO membrane surface

collected from the non-chloramine FO system was fully covered by intact bacteria (green) with a minor number of damaged bacteria (red) across the membrane surface **(Figure 8a)**. Although the pre-filtered primary wastewater effluent contained both intact and damaged bacteria at similar ratios **(Figure 7)**, intact bacteria were more apparent than damaged bacteria, indicating the potential of bacterial growth (i.e., proliferation of intact bacteria) on the membrane surface. In contrast, the FO membrane surface collected from the chloramine system was fully covered by damaged bacteria (red) across the surface **(Figure 8a)**, indicating that chloramine effectively damaged the membrane cells of bacteria deposited on the membrane surface.

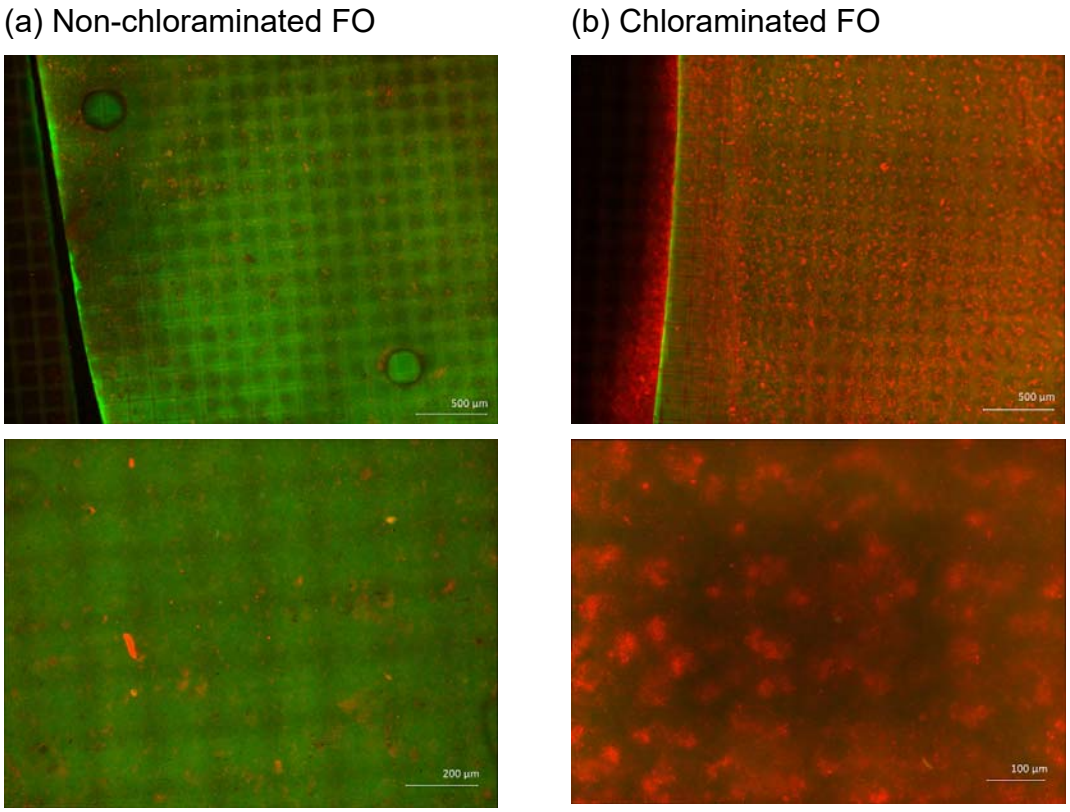
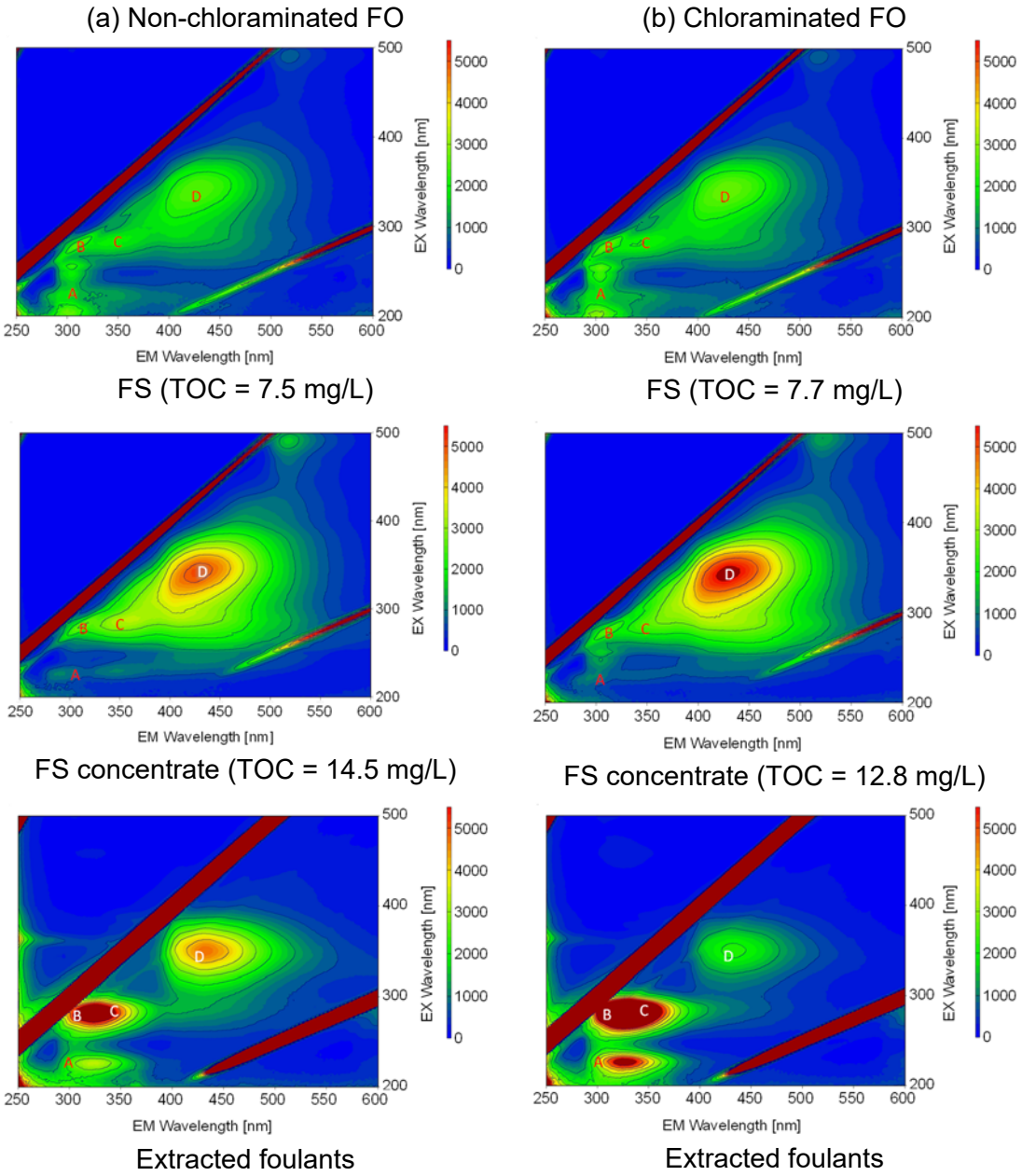


Figure 8. Surface images of forward osmosis (FO) membranes stained by BacLight staining: (a) Non-chloraminated and (b) Chloraminated (21 mg/L dose) systems after 40 h of treatment. The black area on the left side is the membrane surface without staining.

Despite the effective disinfection of the membrane surface, a gradual reduction in permeate-flux (i.e., membrane fouling) occurred in the chloraminated FO system, indicating the

296 occurrence of membrane fouling other than biofouling. Thus, the potential of organic fouling
297 was evaluated through characterization of the wastewater **(Figure 9)**. The FS feed (i.e., pre-
298 filtered primary wastewater effluent) showed four major peaks: Ex/Em of 230/300 nm
299 (aromatic protein including tyrosine, denoted by "A"), Ex/Em of 270/300 nm (protein-like
300 substances containing tryptophan, denoted by "B"), Ex/Em of 280/340 nm (protein-like
301 substances related to microbes, denoted by "C"), and Ex/Em of 340/425 nm (humic-like
302 substances, denoted by "D") (Chen et al., 2003; Nam and Amy, 2008). The peaks of regions
303 B, C, and D were intensified in the FS concentrate, meaning that organics in these regions
304 were concentrated by the FO membrane. However, fractions of these organics, particularly in
305 regions B and C, remained on the membrane surface, as can be seen in the EEM of the
306 extracted foulants **(Figure 9)**. The results indicate that foulants on the membrane surface may
307 be composed of protein-like substances. It is important to note that a small fraction of
308 protein-like substances in regions A and B were detected in the DS of the non-chloraminated
309 FO system, although the DS of the chloraminated FO system showed no peaks in the same
310 regions **(Figure S3)**. Substances in these regions include small hydrophilic chemicals such as
311 tryptophan ($C_{11}H_{12}N_2O_2$) and tyrosine ($C_9H_{11}NO_3$) with molecular weights of 204 g/mol and
312 181 g/mol, respectively which are equivalent to the molecular weight cut off of typical CTA
313 FO membranes (200 g/mol) (Valladares Linares et al., 2011). Thus, these small chemicals
314 may have been deposited on the interior surface of the membrane pore in both non-
315 chloraminated and chloraminated FO systems, inducing pore-blocking and consequently
316 reducing membrane permeability.



317 **Figure 9.** Excitation emission matrix (EEM) fluorescence spectra and total organic carbon
318 (TOC) concentrations of the feed solution (FS) and FS concentrates, and foulants extracted
319 from the fouled membrane surface: (a) Non-chloraminated and (b) Chloraminated (21 mg/L
320 dose) forward osmosis (FO) systems.

321 ### *3.5 Implications for full-scale systems*

322 This study successfully demonstrated the effectiveness of pre-chloramination in mitigating
323 biofouling during a single-pass concentration (i.e., without recirculation) of the pre-filtered
324 primary wastewater effluent. For long-term operation at full scale, the permeate-flux

reduction observed in this study may deteriorate the feasibility of this pre-concentration technique. The occurrence of organic fouling could be attributed to the low cross-flow velocity of FS (0.2–0.5 mm/s), applied to attain a high recovery of >50 % in a single-pass orientation with a small membrane coupon. In theory, the reduction in organic fouling can be achieved by increasing the cross-flow velocity that provides shear force and inhibits organic deposition (Boo et al., 2013; Blandin et al., 2016; Lotfi et al., 2017; Lotfi et al., 2018). An additional test conducted in this study showed that a high cross-flow velocity of FS (0.23 m/s) with a typical recirculation system showed minor membrane fouling **(Figure S4)**, although the momentary permeate recovery was reduced to only 2 %. The advantage of the high cross-flow velocity was the increase in the baseline of the permeate-flux (>10 L/m^2h over the course of 20 h), induced by the reduction in salt concentration polarization at the membrane interface.

At full scale, high cross-flow velocity with high-recovery can be achieved through a so-called tree configuration typically employed in water recycling applications **(Figure 10)** which has multiple stages, in which the number of membrane vessels are reduced in the following stages in proportion to the reduction of feed flow, so that cross-flow velocity is maintained and high-recovery is achieved. As spiral-would membrane elements have a limitation in the acceptable level of particles in the intake (e.g., silt density index of <5), the pre-concentration of primary wastewater effluent using typical spiral-would FO membrane elements may not be viable. Therefore, different orientations of membrane elements (e.g., high feed spacer thickness or courser mesh) may be needed to maintain a high-recovery of wastewater by FO membranes.

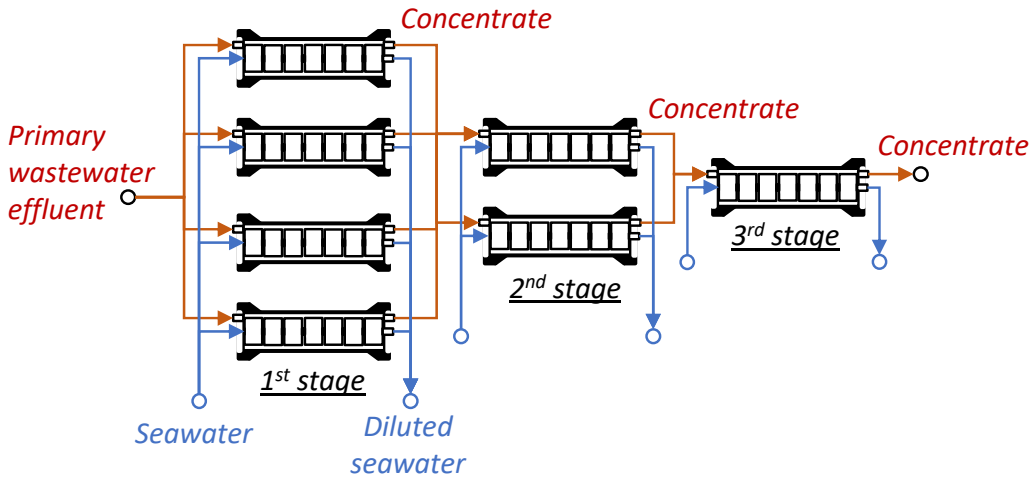


Figure 10. Conceptual flow diagram of a high concentration (recovery) system in tree configuration.

4. Conclusions

Chloramination during the single-pass concentration of pre-filtered primary wastewater effluent by a CTA FO membrane was found to alleviate biofouling. A chloramine dose of 22 mg/L was found to mitigate membrane fouling, confirmed by the decreased reduction of permeate-flux and coverage of the membrane surface with damaged bacteria. However, other types of membrane fouling appeared to occur even with chloramination, and it gradually decreased permeate-flux. The permeate-flux reduction was attributed to organic fouling that occurred very likely due to the low cross-flow velocity. This study suggested the need to develop a new treatment configuration to maintain a high cross-flow velocity and achieve a high recovery.

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