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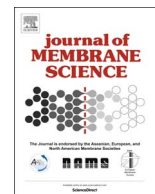
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Application of monochloramine for wastewater reuse: Effect on biostability during transport and biofouling in RO membranes

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ABSTRACT

The rising demand for clean and safe water has increased the interest in advanced wastewater treatment and reuse. Reverse osmosis (RO) can provide reliable and high-quality water from treated wastewater. Biofouling inevitably occurs, certainly with wastewater effluents, resulting in RO performance decline and operational problems. Chlorination of feed water has been commonly applied to limit biological growth. However, chlorine use may lead to a loss of membrane integrity of RO systems. In this study the potential of monochloramine as an alternative for chlorine was studied by (i) evaluating the biological stability of a full-scale wastewater membrane bioreactor (MBR) effluent during transport over 13 km to a full-scale RO plant and (ii) assessing the biofouling control potential in membrane fouling simulator (MFS) and pilot-scale RO installation. Microbial water analysis was performed on samples taken at several locations in the full-scale water reuse system (MBR effluent, during transport, and at the RO inlet and outlet) using a suite of tools including heterotrophic plate counts (HPC), adenosine triphosphate (ATP), flow cytometry (FCM), and 16 S rRNA gene pyrosequencing. Growth potential tests were used to evaluate the effect of monochloramine presence and absence on bacterial growth. Results showed limited changes in the microbial water quality in the presence of monochloramine. MFS studies showed that membrane biofouling could be effectively repressed by monochloramine over prolonged time periods. The normalized salt passage in a pilot RO system with monochloramine dosage was constant over a one year period (data of last 130 days presented), demonstrating that no membrane damage occurred. From this study, it can be concluded that monochloramine dosage in wastewater applications is effective in controlling biofouling in RO systems and maintaining a monochloramine residual during water transport provides biologically stable water.

1. Introduction

The Environmental Protection Agency 2012 guidelines [1] for water reuse reported that "Treated wastewater is increasingly being seen as a resource rather than simply waste". Reclaimed water can fulfill most water demands, as long as it is satisfactorily treated to ensure water quality suitable for the intended use [2]. Reverse osmosis (RO) and nanofiltration (NF) membranes produce high-quality water from sources such as brackish or seawater and secondary treated wastewater effluent [3]. The pre-treated feed water or secondary wastewater effluents still contain dissolved organic compounds, microorganisms, and colloidal particles, contributing to membrane fouling [4]. Therefore, membrane fouling is a major constraint for the operation and cost effectiveness of membrane systems [5]. Fouling, severely limits membrane performance, leading to a reduction in permeate quality and quantity, and eventually causing membrane damage. Several types of fouling can occur simultaneously and affect each other [6]. In practice, an extensive pre-treatment can eliminate scaling and particulate fouling but to a lesser extent organic and biological fouling [7,8].

Biofouling or biological fouling is the excessive growth of a biofilm that results in an unacceptable performance decline [7]. In practice, a 10–15% increase in feed channel pressure drop or reduction in permeate flux is considered operationally unacceptable [8,9]. Reducing biological growth and biofouling in transport pipes and membrane

systems is normally achieved by limiting the essential nutrients for bacterial growth, mainly (but not exclusively) organic carbon [10], and/or dosing disinfectants. Free chlorine is the most commonly used disinfectant to prevent biological growth in water because of its ability to rapidly inactivate most pathogenic microorganisms [2,11]; however residual chlorine has to be removed from the water before entering RO systems with chlorine-intolerant polyamide membranes. Moreover, the reaction of chlorine with organics present in the water results in the formation of halogenated organic by-products such as trihalomethanes (THMs), which are classified as suspect carcinogens for humans [12,13]. Several studies have reported that RO membranes reduce up to 80% of the THM concentration present in the feed water [14,15]; however, using an alternative disinfectant with a lower halogenated organic by-product formation is desirable. Monochloramine dosage was introduced in an attempt to abide by the new THM regulations, due to its weaker tendency to produce halogenated organic reaction products [16]. Monochloramine, the most stable form of chloramine, although a more slowly acting and weaker disinfectant than free chlorine, can be more effective in penetrating and inactivating biofilms [17,18]. In aqueous solution, naturally present ammonia (NH_3) or ammonium ions react with chlorine or hypochlorite to form inorganic chloramines [19] through the reaction:

$$HClO + NH_3 \leftrightarrow NH_2Cl \text{ (monochloramine)} + H_2O. \quad (1)$$

The stability and the type of chloramine formed depends on the ratio ammonia/chlorine. Monochloramine formation is a function of the pH and occurs most rapidly at a pH value of approximately 8.3.

For wastewater reuse applications and further treatment of secondary wastewater effluents, preventing bacterial regrowth during transport to tertiary treatment facilities is essential for an optimal RO treatment performance. During transport, several biological processes can occur including biofilm formation on the pipe walls and biofilm detachment [11,20], microbial growth in the bulk water [21], bio-corrosion of pipe material [22,23], and proliferation of pathogenic bacteria [24] deteriorating the water quality. Ideally, the goal is to transport biologically stable water where microbial growth is restricted [25]; however, due to the development of more sensitive and accurate microbial analysis techniques changes in microbial presence can be detected without necessarily having a negative impact on the water quality rebuking the earlier definition of biological stability [26,27].

In this study, monochloramine was used to disinfect a membrane bioreactor (MBR) permeate and provide biologically stable effluent water during transport to an RO treatment facility. The treated effluent was transferred by a 13 km long pipe to the RO facility, where mono-chloramine residual was removed before the water entered the RO membranes. The RO treatment plant suffered from performance decline due to fouling development in the membrane modules. The aim of this study was to investigate the effect of monochloramine dosage on the biological stability of the water during transport. The effectiveness of monochloramine dosage in biofouling control and possible consequences for membrane damage were assessed in lab-scale membrane fouling simulator (MFS) experiments and pilot-scale membrane module experiments. The effect of monochloramine removal and RO filtration on microbial water quality was also examined. A suite of microbial analysis techniques including flow cytometry and pyrosequencing was applied.

2. Material and methods

2.1. Analysis of water samples from practice

#### 2.1.1. Site description

The RO water treatment facility (DECO) produces demineralized water, cooling tower supply water, and ultrapure water for industrial usage in Terneuzen, the Netherlands (51°20′08″ N, 3°49′40″ E). Since

Table 1
Average MBR effluent quality.

Parameter	Unit	Value
Specific conductivity at 25 °C	μS/cm	1600 ± 450
pH after acid dosage	–	7.4 ± 0.2
Temperature	°C	15 ± 4
O_2	mg/L	9.7 ± 0.2
Total COD	mg/L	34 ± 8
Total BOD	mg/L	< 3
Ca^{+2}	mg/L	71 ± 19
Mg^{+2}	mg/L	22 ± 8
Bicarbonate (HCO_3^-)	mg/L	280 ± 80
TSS	mg/L	0.3 ± 0.1
TKN	mg/L	1.9 ± 0.6
NH_4^+	mg/L	0.9 ± 0.1
Ortho-P	mg/L	0.8 ± 0.9
SO_4^{-2}	mg/L	88 ± 22

COD: chemical oxygen demand, BOD: biological oxygen demand.

2010, a membrane bioreactor (MBR) constructed and operated by Evides Industriewater on the site of the municipal wastewater treatment plant (WWTP) (51°17′49″ N, 3°50′14″ E) has been used to produce feed water for the DECO water treatment facility. Table 1 summarizes the average MBR effluent quality. The effluent from the MBR is disinfected using monochloramine and transported over a 13 km long pipe to the DECO facility (residence time of 4 h). Monochloramine was formed by dosing ammonium chloride (NH_4Cl - a 20% solution) and sodium hypochloride ($NaClO$ - a 12.5% solution) according to the following stoichiometric equation (Molar ratio = 1).

$$NH_4Cl + NaClO \rightarrow NH_2Cl + H_2O + NaCl \quad (2)$$
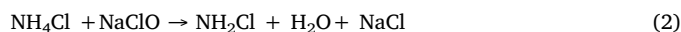
At the DECO facility, the water is consecutively treated by sulphuric acid dosage (H_2SO_4), 50 μm screens, an antiscalant, and sodium bisulfite dosage for monochloramine removal (residual monochloramine ≈ 1 ppm). The water is then fed into the reverse osmosis (RO) system. DOW FILMTEC BW30-400/34i membranes were used. The plant is operated at a minimum capacity of 210 m³ h⁻¹. The recovery of the RO system is 75%. The DECO RO installation has performance decline problems, and membrane cleanings have to be carried out frequently. Cleaning the modules in place (CIP) is done by dosing NaOH up to a pH of 12.

2.1.2. Sampling scheme

The schematic diagram of the treatment train between the MBR and the RO water treatment facility with an overview description of the sampling locations is schematized in Fig. 1. Triplicate samples were taken at each location. Microbial analysis and bacterial community analysis were performed on the samples.

2.1.3. Microbial analysis

2.1.3.1. Heterotrophic plate counts (HPC) and adenosine triphosphate (ATP) measurements. For HPC and ATP measurements, water was collected in high-density polyethylene (HDPE) plastic bottles containing 2 mL L⁻¹ of a mixed solution of sodium thiosulfate (20 g L⁻¹) and nitrilotriacetic acid (25 g L⁻¹). HPC was measured by Aqualab Zuid (Werkendam, NL), according to the Dutch standard procedure (NEN-EN-ISO 6222, 1999) [28]. ATP was measured by Het Waterlaboratorium (Haarlem, NL) using a luminometer (Celsis Advance). ATP was first released from suspended bacterial cells with nucleotide-releasing buffer (LuminEX, Celsis) for total ATP measurement, while this step was not performed for assessment of free ATP. Bacterial ATP concentrations were calculated by subtracting free ATP from total ATP concentrations. The detection limit of the method was 1 ng ATP L⁻¹.

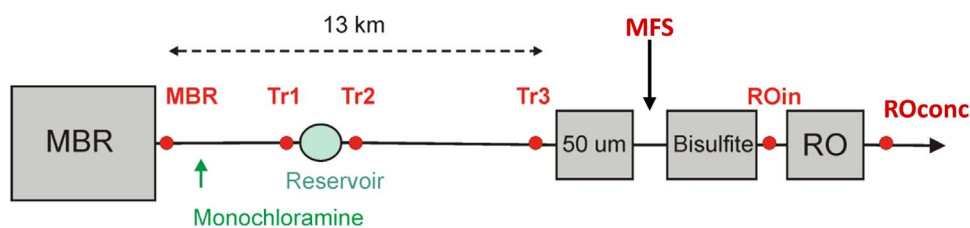*2.1.3.2. Flow cytometry.* Measurements of the total and intact bacterial

Location	Description
MBR	MBR permeate before monochloramine dosing
Tr1	MBR permeate after monochloramine dosing, before buffer tank
Tr2	MBR permeate after monochloramine dosing, after buffer tank
Tr3	Influent to DECO plant (MBR pearmeate dosed with monochloramine and transported over a 13 km pipe)
ROin	RO feed water after sodium bisulfite dosage
ROconc	RO concentrate

cell concentrations in the water samples taken at the six locations shown in Fig. 1 were done using flow cytometry according to the protocol reported by Prest et al., 2013 [29]. For the determination of the total bacterial cell concentration, 500 μL samples were preheated to 35 °C for 10 min, stained with 10 μL mL^{-1} SYBR Green I (Molecular Probes, Eugene, OR, USA), then incubated in the dark at 35 °C for 10 min. For the determination of the intact bacterial cell concentration, propidium iodide in combination with SYBR Green I was used according to the same protocol used for total bacterial cell concentration. Measurements were performed using a BD Accuri C6 flow cytometer (BD Accuri Cytometers, Belgium) equipped with a 50 mW laser having a fixed emission wavelength of 488 nm. Fluorescence intensity was collected at FL1 = 533 ± 30 nm, FL3 > 670 nm, sideward and forward scattered light intensities were obtained as well. All data were processed with the BD Accuri CFlow® software, and electronic gating was used to select SYBR green labelled signals for quantifying total bacterial cell count following the procedure described by Hammes and Egli (2005) [30]. The same electronic gating was used to quantify intact bacterial cells when the mixture of propidium iodide and SYBR Green I staining was used. Additional gates on the green fluorescence histogram were applied to differentiate low (LNA) and high (HNA) nucleic acid containing bacterial communities [31,32]. The percentages of LNA and HNA are used as basic flow cytometric fingerprinting strategy [29].

2.1.3.3. Growth potential of water. Water samples were collected in triplicates in assimilable organic carbon (AOC)-free glass vials for growth potential assessment. No sample treatment and no chemical or bacterial dosage was performed. After that, the vials were incubated at 30 °C for 11 days [30]. Flow cytometry measurements were performed at the start and after 1, 2, 3, 4, 7, 9 and 11 days of incubation for a comparison of bacterial growth potential at the different water sampling locations.

2.1.3.4. 16S rRNA gene pyrosequencing. Water samples (2 L) were collected in HDPE bottles containing 2 mL L^{-1} of a mixed solution of sodium thiosulfate (20 g L^{-1}) and nitrilotriacetic acid (25 g L^{-1}). Each sample was filtered through a 0.2 μm-pore-size Isopore membrane filter (Merck Millipore, Billerica, MA) on the same day of sampling. The filters were stored at minus 20°C until processing. Genomic DNA was extracted from the collected biomass using the Fast DNA SPIN Kit (MP Biomedicals, Santa Ana, CA) according to the manufacturer's instructions. Bacterial 16S rRNA genes were amplified with the bacteria-specific forward primer 515F (5′-LinkerA-Barcode-GTGYCAGCMGCCGCGGTA-3′) and reverse primer 909R (5′-LinkerB-

CCCCGYCAATTCMTTTRAGT-3′). A single-step 28-cycle PCR using the HotStarTaq Plus Master Mix Kit (QIAGEN, Valencia, CA) was performed for each DNA sample (triplicate reactions) under the conditions described in El-Chakhtoura et al. (2015) [26]. Pyrosequencing was carried out on the Roche 454 FLX Titanium genome sequencer, and sequence data was processed as described in El-Chakhtoura et al. (2015) [26]. OTUs were defined by clustering at 3% divergence. Final OTUs were taxonomically classified using BLASTn against a curated database derived from NCBI and Greengenes. Multidimensional scaling (MDS) was performed with the Bray-Curtis matrix using the R statistical package to ordinate the sequencing operational taxonomic unit (OTU) data (samples with similar community structure cluster together, taking into account the relative abundance of each OTU).

2.2. Lab-scale monochloramine studies

2.2.1. Experimental setup

Four membrane fouling simulator (MFS) setups [33] were installed at the DECO water treatment facility after the 50 μm screens to assess the RO feed water biofouling potential. The feed water entering the DECO water treatment facility contained 1 ppm of monochloramine. The MFS setup consisted of a diaphragm pump, temperature and differential pressure transmitter (Delta bars, Endress + Hauser, PMD75), pressure-reducing valve, chemical dosing diaphragm metering pump, and the MFS flow cell. The MFS contained a 20 cm × 4 cm coupon of a membrane and a feed spacer. The spacer and membrane sheets were taken from virgin spiral wound membrane elements (DOW FILMTEC LE-440i). The feed spacer consisted of a sheet of 28 mil (711 μm) thick, diamond-shaped polypropylene spacer.

2.2.2. Operating conditions

Different operating conditions were applied for each MFS setup (Fig. 2). Two MFS setups were placed directly after the 50 μm screeners (MFS 1 and 2). The other two MFS setups were placed after a 1 μm cartridge filter that was installed after the 50 μm screeners (MFS 3 and 4). Excess sodium bisulfite (SBS – 8.2 mg L^{-1}) was dosed using a diaphragm metering pump (STEPDOS 03S, Knf NEUBERGER) to two MFS setups (MFS 2 and 4) at a rate of 0.4 mL min^{-1} to remove residual monochloramine. The MFS setups were operated without permeate production at a pressure of one bar for a period of 50 days. Earlier studies done with membrane elements in the same parallel position in an NF installation, with and without permeate production, showed the same feed channel pressure drop increase and biofilm formation [8,33–35]. Mass transfer calculations supported the observations that
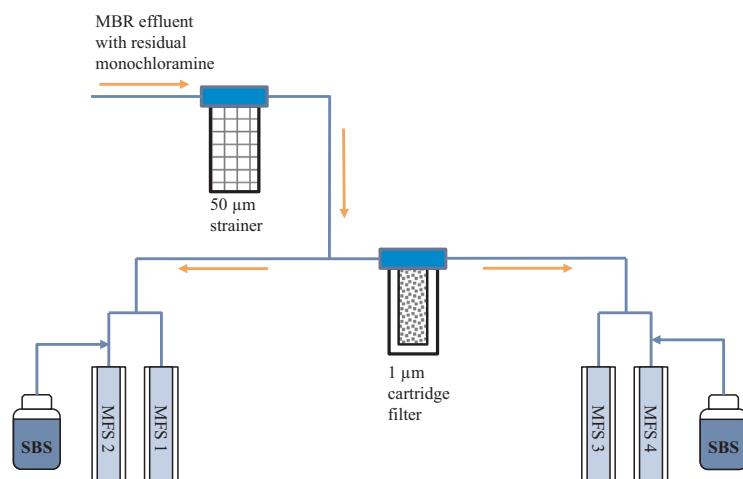
MFS	Code	Experiment description	Particles > 1 μm	Monochloramine residual
1	-CF +M	Feed water passing 50 μm strainer	+	+
2	-CF -M	Feed water passing 50 μm strainer, dosed with sodium bisulphite (SBS) to remove residual monochloramine	+	-
3	+CF +M	Feed water passing 50 μm strainer, filtered using 1 μm cartridge filter to remove particles	-	+
4	+CF -M	Feed water passing 50 μm strainer, filtered using 1 μm cartridge filter to remove particles, and dosed with SBS to remove residual monochloramine	-	-

(+) present (-) absent CF: cartridge filter; M: monochloramine

the permeate flux is not playing a significant role in the nutrient supply to the fouling layer. Hydrodynamic conditions in the MFS were similar to spiral wound membrane modules as applied in practice for water treatment [33]. A flow rate of $16 \, \mathrm{L \, h^{-1}}$ equivalent to a linear flow velocity of $0.16 \, \mathrm{m \, s^{-1}}$, representative for practice, was used [36]. Pressure drop (ΔP) development in time was recorded for all the MFS systems and the increase in ΔP was used as an indication of fouling development [37].

2.3. Pilot-scale monochloramine studies

Two pilot-scale membrane filtration units (P1 − P2) were operated in parallel to the full-scale DECO water treatment facility to assess the effect of monochloramine dosage on membrane performance. DOW FILMTEC LE-440i elements, containing a 28 mil (711 μm) thick feed spacer with a membrane surface area of 41 m²/module, were used for the two pilots (P1 − P2). The DECO full-scale installation is equipped with DOW FILMTEC BW30-400/34i elements; containing a 34 mil (863 μm) thick feed spacer with a total membrane surface area of 37 m²/module. The pilot research was carried out with DOW FILMTEC LE-440i membranes as in that stage the original DOW FILMTEC BW30-400/34i membranes were not available. DOW FILMTEC LE-440i membranes have a slightly higher permeate flow, with a similar permeate quality as DOW FILMTEC BW30-400/34i membranes. The feed water was passed through a 10 μm pore size cartridge filter before flowing into P1. The 10 μm pore size cartridge filter was not used after the feed water to P2. Both pilot units received monochloramine dosage while the full-scale facility had monochloramine (1 ppm) removed by dosing sodium bisulfite. The pilot modules were operated with the same average permeate flux ($14.7 \, \mathrm{L \, m^{-2} \, h^{-1}}$) and recovery (10% per module) as the full-scale plant. Normalized salt passage was recorded for the two pilot units as well as the full-scale plant for a period of 12 months. The normalized salt passage is used to describe the efficiency of salt rejection by the membrane and is normalized for temperature and permeate flow rate according to the method described by Huiting et al., 2001 [38]. The salt passage data presented in the manuscript is for a period of 130 consecutive days in between two cleanings performed for the full-scale installation.

3. Results

3.1. Monochloramine dosage and biological stability

Microbial analysis was performed on water samples that were taken from six different locations starting from the MBR outlet, along the transport pipe to the DECO water treatment facility, as well as the RO treatment train at the DECO plant (Fig. 1). The samples can be differentiated into two groups (i) the samples during transport: the MBR effluent and samples containing monochloramine (MBR, Tr1, Tr2, and Tr3), and (ii) the samples at the RO plant: after removal of the monochloramine residual with sodium bisulfite and the RO concentrate (ROin and ROconc).

ATP measurements showed relatively high ATP concentrations in all water samples ($\approx 100 \, \mathrm{pg \, ATP \, mL^{-1}}$). Limited changes in ATP (5–15% increase) were seen in the water samples during transport (MBR, Tr1, Tr2, and Tr3, Fig. 3A). No increase in HPC was seen in sample Tr2 compared to the MBR sample (Fig. 3B); however, the HPC in sample Tr3 was approximately 800% higher than the MBR sample, inconsistent with the ATP measurements. Compared to sample ROin, sample ROconc showed a relatively high HPC (700% increase) (Fig. 3B). Furthermore, compared to ROin, an increase in bacterial ATP ($\approx 430\%$) was observed in the ROconc samples (Fig. 3A); the increase was higher than the expected concentration yield (factor of 4 increase) due to 75% recovery of the RO modules. Both ATP and HPC results suggest bacterial growth in the membrane modules.

Total, intact, and damaged bacterial cell concentrations were determined for samples taken at the six locations using flow cytometry (Fig. 4). An average bacterial cell concentration of $\approx 11.7 \times 10^3$
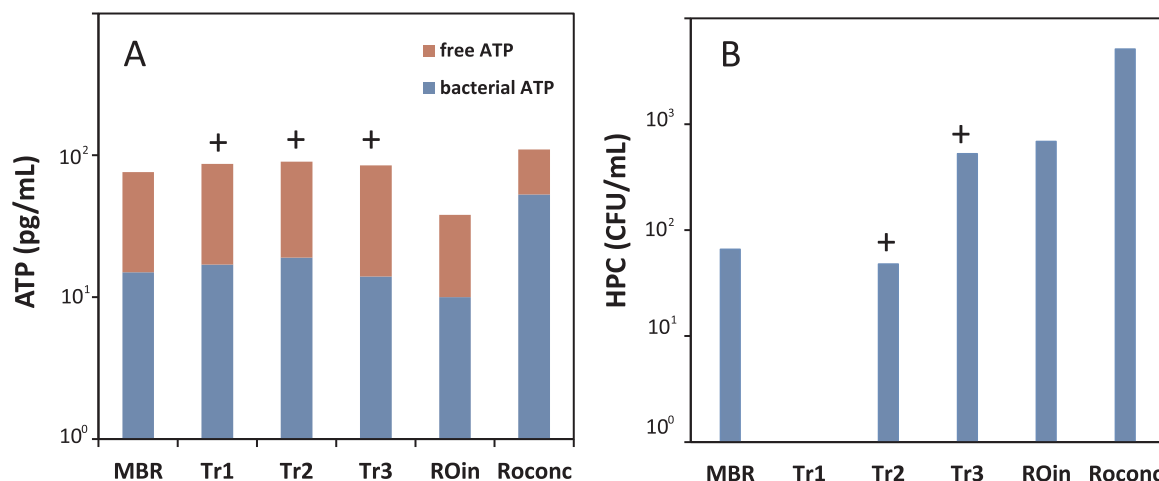
Fig. 3. (A) ATP concentrations (pg ATP/mL) and (B) Heterotrophic Plate Counts (CFU/mL) of water samples taken at the locations shown in Fig. 1. Samples containing monochloramine are marked with + sign.

cells mL^{-1} was found at the MBR outlet, before monochloramine dosage. The total cell concentration increased to an average of $\approx 19 \times 10^3$ cells mL^{-1} during water transport (Tr1, Tr2, and Tr3, Fig. 4A); however, a decrease in the intact cell fraction was seen (from 83% to 59%) indicating cell damage due to the disinfection by monochloramine (Fig. 4B).

After dosing of sodium bisulfite to quench residual monchloramine, an increase in total cell concentration in ROin (37 × 10^3 cells mL^{-1}) and ROconc (88 × 10^3 cells mL^{-1}) samples (Fig. 4A) was observed. An increase in the intact cell fraction (Fig. 4B) and HNA concentration was observed compared to the transport samples (Fig. 5A). Both addition of sodium bisulfite and RO filtration induced large changes in total cell numbers (ROin and ROconc) and cell composition (Figs. 4 and 5). The flow cytometric fingerprint plot (Fig. 5B) clearly showed the samples after the MBR and during transport (monochloramine residual) clustered together signifying their similarity; the samples after sodium bisulfite dosage (ROin) and RO filtration (ROconc) appeared as outliers clearly demonstrating the bacterial changes that occurred in these samples.

The limited increase in HPC except for sample Tr3, ATP, cell concentration and the similarity in flow cytometric fingerprints for the

MBR sample and the samples during transport all indicate biologically stable water after monochloramine dosage, while the highest biological growth was observed after monochloramine removal.

3.1.1. Growth potential tests

In parallel to HPC, ATP, and bacterial cell concentration analysis, growth potential tests were performed on the water samples taken at the six different locations to evaluate the impact of monochloramine dosage and the impact of monochloramine removal on the water bacterial growth potential (Fig. 6). Faster growth occurred in the sample before monochloramine dosage (MBR) compared to samples after monochloramine dosage (Tr1, Tr2, and Tr3) showing the delaying effect of monochloramine on microbial growth (Fig. 6A). The total cell concentration for samples Tr1, Tr2, and Tr3 only started to increase after day four, reaching the same plateau value as the MBR effluent sample. The dosage of sodium bisulfite before samples ROin and ROconc apparently reduced the bacterial growth lag phase compared to the samples when monochloramine was present. Compared to Tr1, Tr2, and Tr3, a faster growth occurred in ROin and ROconc (Fig. 6B). A higher growth potential for ROconc compared to ROin was measured (Fig. 6B). After six days, approximately the same plateau value in total

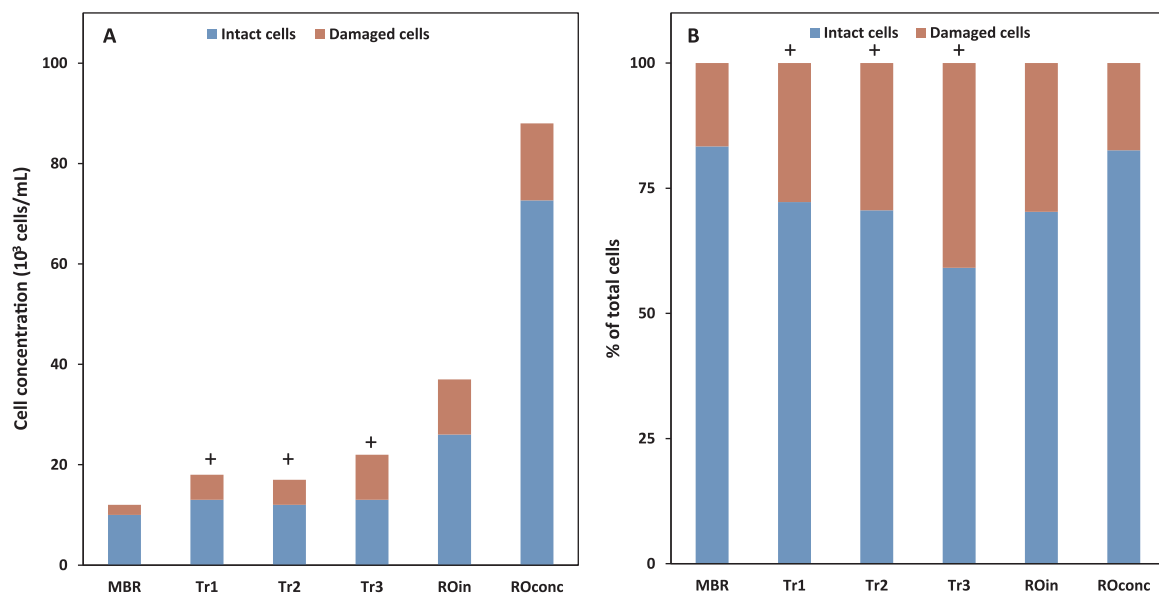

Fig. 4. Comparison between (A) cell numbers (total, intact and damaged) and of (B) percentages of intact and damaged cells, of samples taken at the locations shown in Fig. 1. Samples containing monochloramine residual are marked with + sign.
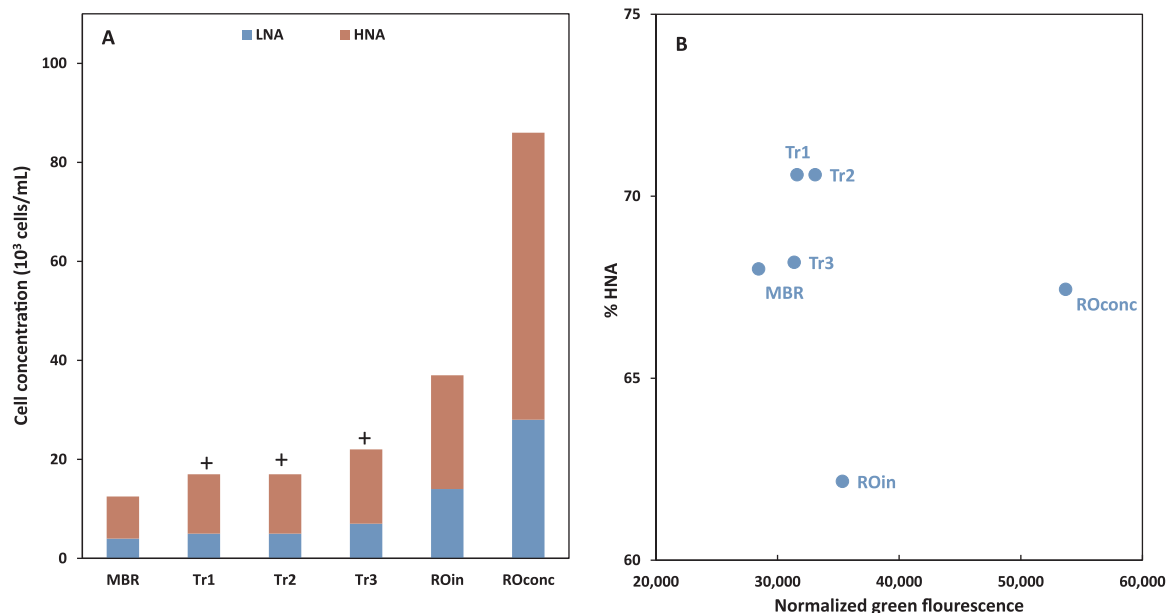
Fig. 5. Comparison between (A) proportions of LNA and HNA cells and (B) flow cytometric fingerprints of samples taken at the locations shown in Fig. 1. HNA = high nucleic acid containing cells. LNA = low nucleic acid containing cells. Samples containing monochloramine residual are marked with + sign. In (B) the closer the samples are located to each other, the higher the similarity in bacterial composition.
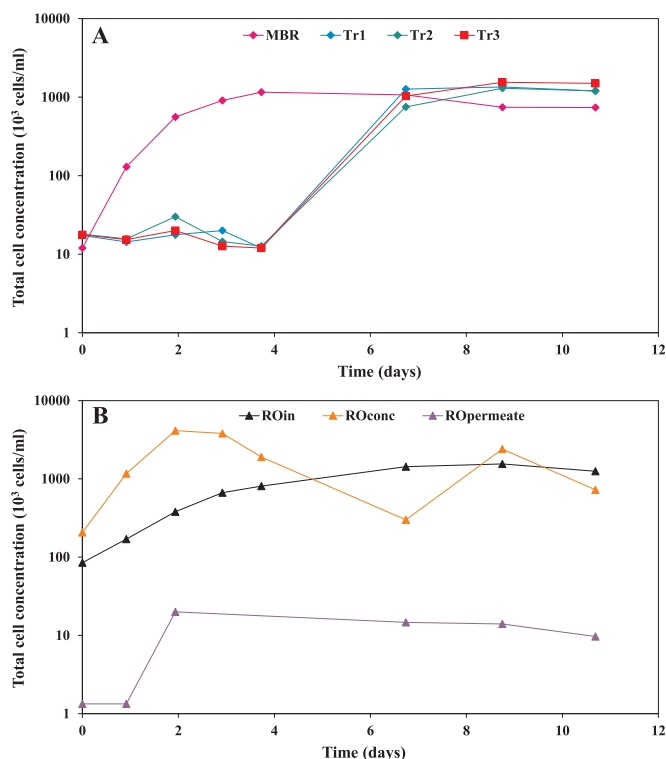

Fig. 6. (A) and (B) Development in time of the total bacterial cell concentration during growth potential tests performed on the water samples taken at the locations shown in Fig. 1.

cell numbers was reached for all six samples indicating equal nutrient concentrations in the six samples. As expected, the RO permeate had the lowest cell concentration and the lowest growth potential.

3.1.2. Bacterial community structure

Bacterial community analysis using 16S rRNA gene pyrosequencing showed a diverse bacterial community in the water samples. At phylum level classification, bacteria with a relative abundance below 1% across

all samples were grouped together under the "other" category. After this percentage cutoff, 12 phyla (including three candidate phyla) were identified in all water samples. The bacterial community in the MBR effluent sample and the samples along the transport pipe which contained monochloramine residual were relatively comparable (samples MBR, Tr1, Tr2, and Tr3), as seen in the bacterial community structure in Fig. 7 and the multidimensional scaling (MDS) plot in Fig. 8. Fig. 8 clearly shows that these samples which contain monochloramine are clustered together with the MBR effluent sample indicating a similar bacterial community structure in agreement with the flow cytometric fingerprint results (Fig. 5B). *Proteobacteria* was the dominant phylum in all the samples (with and without monochloramine residual) with a relative abundance of 49.6 ± 5.1%. The other detected phyla in addition to *Proteobacteria* in the MBR effluent and samples containing monochloramine were *Bacteroidetes* (14.9 ± 4.8%), *Planctomycetes* (7.2 ± 2.3%), *Chloroflexi* (5.1 ± 1.9%), *Actinobacteria* (4.9 ± 1.0%), *Firmicutes* (3.9 ± 0.9%), *OP3* (3.8 ± 0.5%), and *WS3* (1.1 ± 0.6%). *Acidobacteria*, *Verrucomicrobia*, *Spam*, and *Spirochaetes* were found at a low relative abundance, average less than 1% in the samples containing monochloramine residual. The addition of sodium bisulfite to remove residual monochloramine before the RO installation resulted in changes in the bacterial community composition and structure (Fig. 7A and B). At a phylum level, *Spirochaetes* (10.8%) and *Verrucomicrobia* (3.0%) increased in relative abundance. Passing across the RO feed spacer channel induced another shift in the relative abundance where *Acidobacteria* fraction substantially increased from 1.4% to 31.5%. Samples ROin and ROconc appeared as outliers in the MDS plot (Fig. 8) when compared with the samples that contained monochloramine and the MBR effluent sample, and this further validates the difference of the bacterial community structure of ROin and ROconc. At a class level, very minor changes in the classes of the dominant phylum *Proteobacteria* were observed. Fig. 7B compares a sample containing monochloramine (Tr3) to samples without monochloramine before RO filtration (ROin) and in the RO reject stream (ROconc) at a genus level and shows that the relative abundance of genera evidently changed between these samples. Removal of monochloramine resulted in an increase in extremophilic genera like *Planctomycetaceae*, *Spirochaeta*, *Phyllobacteriaceae*, *Caldilinea*, and nitrifiers like *Nitrosopumilus*. Passing across the RO feed spacer channel caused another shift in the relative
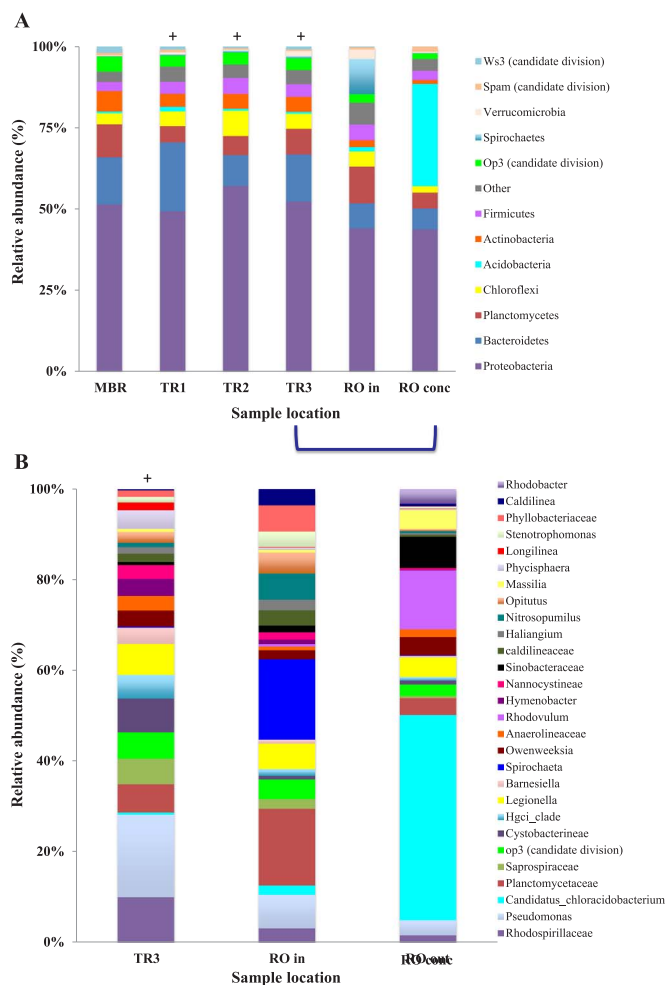
Fig. 7. Relative abundance of bacterial phyla (A) and genera (B) in samples collected at the locations shown in Fig. 1. Samples containing monochloramine residual are marked with + sign.
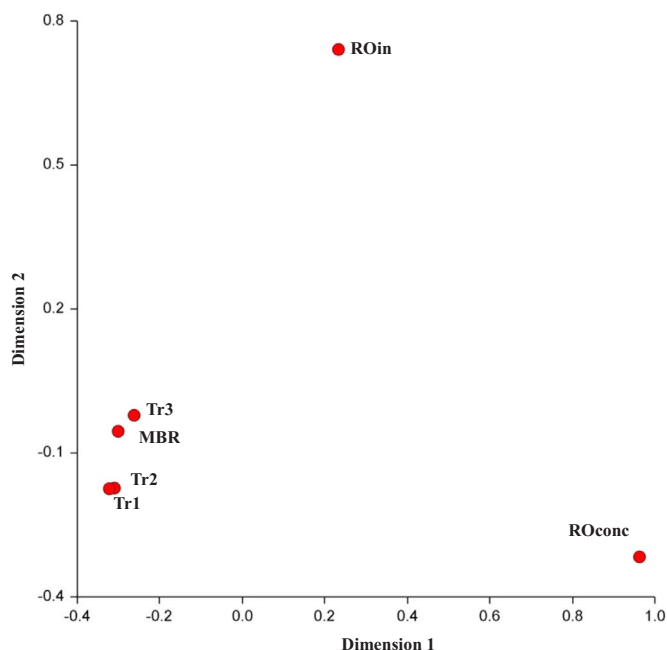

Fig. 8. Pyrosequencing multidimensional scaling (MDS) plot for the samples taken at the locations shown in Fig. 1. An MDS plot is used to visualize the level of similarity between samples. The closer the samples are located to each other, the higher the similarity in bacterial community structure.

abundance of bacterial genera showing high microbial diversity and activity in the RO system. Compared to RO feed, genera like *Candidatus_Chloracidobacterium*, *Rhodovulum*, *Sinobacteraceae*, *Massilia*, and *Rhodobacter* became more abundant in the RO concentrate. Based on the above reported HPC, ATP, flow cytometry, and pyrosequencing results, the use of monochloramine as a residual disinfectant resulted in a stable microbial water quality during water transport, as long as monochloramine is present.

3.2. Monochloramine effectiveness for biofouling control: MFS research

Monochloramine efficiency for biofouling control was tested by running four MFS setups in parallel at the DECO water treatment facility using the same feed water source as the full-scale installation. The effect of using a 1 μm cartridge filter to remove particles on fouling development was evaluated. Fig. 2 summarized the different operating conditions for each MFS setup. No increase in *ΔP* and a minor increase in *ΔP* were observed for MFS3 (+CF +M) and MFS1 (-CF +M) respectively (Fig. 9). Both MFS1 and MFS3 had a monochloramine residual; however, a 1 μm cartridge filter was used before MFS3, resulting in no increase in *ΔP* at all. MFS2 (-CF –M) and MFS4 (+CF –M) displayed a major increase in *ΔP*. MFS2 had the highest increase in *ΔP* during the 50 day experiments (Fig. 9); the *ΔP* increase was exponential indicative of biomass formation. This highest *ΔP* development in MFS2 was caused by the absence of both the 1 μm cartridge filter and monochloramine residual.

A closer look at the 50 μm strainer and the 1 μm cartridge filters showed the deposition of black particles on the cartridge filters. Analysis of the filter deposits revealed, in addition to a majority of organics, the presence of manganese, therefore, explaining the black color (Supplementary material Figs. S2, S3, S4 and Tables S1, S2). The MFS studies revealed that maintaining a monochloramine residual enabled control of biofilm development. The addition of a 1 μm cartridge filter to the pretreatment train further reduced *ΔP* development in the MFS suggesting that besides biofouling, particulate fouling was playing a role in performance decline at the DECO water treatment facility.

3.3. Monochloramine suitability for biofouling control: pilot research

Two pilot spiral wound elements (P1 and P2) were operated in parallel to the full-scale installation and monochloramine was continuously dosed to these pilot elements for a period of one year to assess the effect of monochloramine on membrane performance (data shown for the last 130 days). The primary concern was whether monochloramine would result in an increase in salt passage caused by membrane damage. Normalized salt passage development in time was monitored for the full-scale installation and the two pilots (Fig. 10). Normalized salt passage remained constant throughout the 130 day period, so it can be concluded that no membrane damage was caused by monochloramine dosage.

4. Discussion

4.1. Monochloramine enables biological stability and biofouling control

Disinfection is a commonly applied practice to achieve biological stability during transport of both drinking water and treated wastewater. Water utilities have been urged to switch to monochloramine disinfection as an alternative to chlorination to limit the production of the two regulated groups of disinfection by-products (DBPs): trihalomethanes (THMs) and haloacetic acids (HAAs) [39]. Monochloramine has been applied as a disinfectant for drinking water distribution [40–44]. In this study, monochloramine was dosed to an MBR effluent
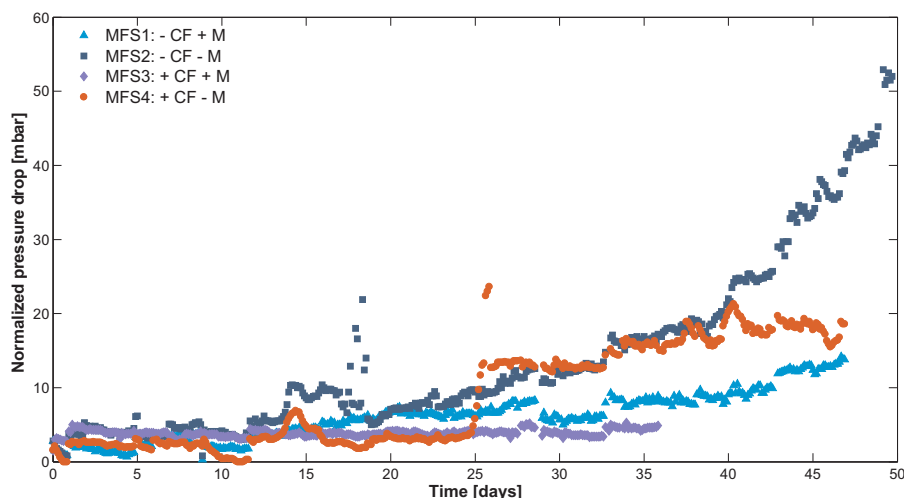
Fig. 9. Pressure drop (mbar) development in time for the 4 MFS units run in parallel under the operating conditions described in Fig. 2. CF: 1 μm cartridge filter, M: monochloramine; (+) present (-) absent.

before transport to an RO treatment facility.

Various microbial analysis techniques including flow cytometry and pyrosequencing were applied. Flow cytometry measurements enabled detection of variations in the total bacterial cell concentration as well as changes in the intact and damaged bacterial cell concentration (Fig. 4), providing information on cell integrity [28,45]. Information on intact and damaged bacterial cell concentration was particularly useful to evaluate the efficiency of monochloramine disinfection [46]. Results showed that monochloramine was efficient in controlling microbial growth during water transport based on measurements of ATP concentration (Fig. 3), total cell concentration (Fig. 4) and bacterial community structure (Figs. 6 and 7). Monochloramine affected the bacterial cell integrity (increase in damaged cell concentration) and delayed microbial growth under controlled conditions (growth tests). In this study, monochloramine use did not result in a major change in the bacterial community structure of the MBR effluent during transport. The phyla with the highest relative abundance were *Proteobacteria* and *Bacteroidetes* similar to MBR effluent bacterial community structure reported in other studies [47,48]. In this study, the bacterial community structure after monochloramine dosage was similar to the MBR effluent sample. Removal of monochloramine immediately resulted in bacterial growth, changes in bacterial community structure (Figs. 7 and 8) and to a similar growth potential as observed in the MBR effluent before monochloramine addition (Fig. 6). In this study, maintaining adequate monochloramine concentration in the transport pipe to avoid bacterial growth was relatively easy, due to the short residence time from the MBR to the RO plant (approximately 4–8 h).

Few studies are reported in the literature addressing monochloramine application in RO membrane systems [49–51]. Most of these studies mainly focused on the type of DBPs forming when monochloramine was used and membrane rejection ability of the formed DBPs. This study focused on monochloramine application for biofouling control and possible membrane damage due to monochloramine use and demonstrated that monochloramine could control biofouling development in RO membranes without causing membrane damage.

4.2. Balancing pros and cons of using chlorine versus monochloramine to achieve biological stability and biofouling control

Chlorine, the most commonly used oxidizing agent for water disinfection, has shown to react with the RO polyamide membrane active layer, resulting in membrane degradation and performance decline [52,53]. Chlorine reacts rapidly with organics which are typically in higher concentrations in MBR effluents and results in higher assimilable organic carbon (AOC) formation [54] leading to increased biofouling potential in the RO modules. Monochloramine, a less aggressive oxidizing agent than free chlorine, has been recommended to minimize biofilm formation in RO polyamide membranes [49–51]. In this study, monochloramine was effective in achieving biological stability during water transport. Results of the MFS experiments revealed that monochloramine had great potential in controlling biofilm formation. Normalized salt passage during the pilot membrane module experiments remained constant throughout the 130 day period, indicating no
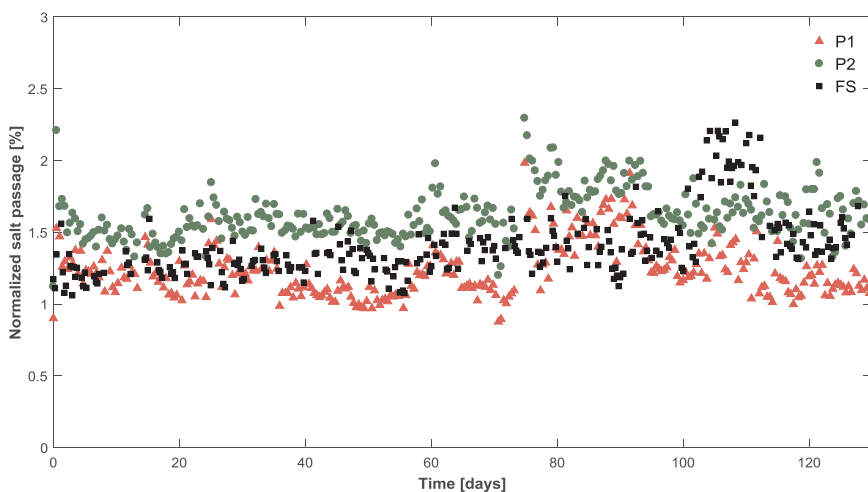

Fig. 10. Normalized salt passage (%) development in time over the two pilot units P1 and P2 and the full-scale installation (FS) for a period of 130 days. P1 with a 10 μm cartridge filter before the RO module. P2 without a 10 μm cartridge filter before the RO module.

membrane damage. In contrast, some studies in the literature reported that monochloramine resulted in an increase of permeate flux and salt passage as a result of membrane damage [51,55–57]. Different feed water types, longer monochloramine contact time, monochloramine application techniques, and the presence of some metals such as ferrous and aluminium can be factors affecting membrane performance, resulting in membrane damage during monochloramine application in these studies. Concerning membrane performance loss, special attention should be given to the formation of free chlorine and of secondary oxidizing agents from the reactions of monochloramine with organics and inorganics present in the water. For example, if monochloramine is applied in seawater, two species of particular concern are bromide and iodide, as monochloramine can react with these ions to form bromine and iodine species that have been found to be reactive towards polyamide [58,59].

In addition to membrane damaging potential, the use of disinfectants is also of concern due to the undesirable formation of DBPs. DBPs are considered a potential human health risk [12,60] and DBP concentrations been regulated to reduce the associated health risks. Chlorination produces the highest amount of THMs and HAAs [60]. Many other halogenated DBPs are also formed during chlorination, however, in lower concentrations. Although monochloramine has been introduced as a better option than chlorine due to its lower THMs and HAAs formation potential, studies on the use of monochloramine disinfection showed an increase in a different set of toxic DBPs, namely the nitrogenous disinfection by-products (N-DBPs) such as N-nitrosodimethylamine (NDMA) [61–64]. N-DBPs, although formed at considerably lower concentrations than regulated DBPs, may pose a greater health risk [65,66]. Disinfection of water rich in nitrogen-containing compounds, specifically in the case of treated wastewater effluents, has been associated with the formation of N-DBPs [67,68]. Monochloramine can also be a source of nitrogen when used as a disinfectant, therefore, increasing N-DBP formation potential. In addition to nitrogen as a precursor, Le Roux et al. (2016) [39] demonstrated that aromatic dissolved organic compounds can play a role as precursors for N-DBP formation and Chu et al. (2010) [63] revealed that protein-like organic matter in certain hydrophilic fractions (acids and bases) played a role in the formation of haloacetamides, an emerging class of N-DBPs. A limited number of studies has investigated N-DBP rejection by RO membranes. Rejection of N-nitrosodimethylamine (NDMA) by RO membranes has been reported to be between 10% and 50% [69–71] while rejections of above 50% have been found for haloacetonitriles (HANs) [49,72]. Doederer et al. (2014) [73] emphasized the influence of feed water quality, membrane properties, and operational conditions on the rejection of DBPs in general and N-DBPs in particular.

In this study, wastewater was treated to produce demineralized water for industrial applications and not as potable reuse; nevertheless, DBPs would still be concentrated in the brine and later discharged to the environment posing human health risks emphasizing the importance of reducing DBP formation. In conclusion, the application of monochloramine as a disinfectant especially for wastewater effluents should be implemented with emphasis on pretreatment and better removal of N-DBP precursors.

4.3. Considerations for monochloramine use (practical implications)

Feed water quality will have a major effect on DBP formation during monochloramination and on monochloramine decay. Providing sufficient pre-treatment for removing DBP precursors helps in restricting DBP formation and consequently DBP concentrations in the permeate; although this might sometimes be technologically or economically impractical especially in wastewater treatment. A better understanding of N-DBP precursors and mechanisms of formation is another step forward toward a better DBP control [39].

During monochloramine use, chlorine residuals should always be monitored in membrane systems. Special attention should be given to

monochloramine application techniques [74]. When dosing preformed monochloramine, prevention of monochloramine decay is essential as free chlorine may be released through dissociation of monochloramine, which may be the cause of polyamide membrane damage and membrane performance loss [75]. Another application technique, specifically used for wastewater where the effluent already contains ammonia, is the separate addition of free chlorine to achieve a certain Cl:N ratio and therefore form monochloramine at a controlled pH. In this case, monitoring that all chlorine is converted into monochloramine is crucial [76].

5. Conclusions

The main study findings can be summarized by:

- Monochloramine was effective in controlling microbial growth during transport and in biofouling control in RO systems of treated wastewater.
- After removal of monochloramine, microbial growth in the RO membrane modules occurred.
- Monochloramine did not affect the bacterial growth potential of the water.
- No increase in salt passage was observed during the operation of two pilot-scale units with monochloramine dosage indicating that in this study no membrane damage has occurred and therefore the suitability of monochloramine use for biofouling control during RO filtration of the MBR effluent.

Acknowledgements

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.memsci.2018.01.060.

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