

## Blending membrane treated WTP waste residuals with finished water: impacts to water quality and biofilm formation

M. E. Walsh and G. A. Gagnon

### ABSTRACT

The purpose of this study was to evaluate the impact of blending filtered water with treated waste residuals streams from a water treatment plant on finished water quality and biofilm formation in a modelled distribution system. A mixed waste residual stream composed of an equalized blend of filter backwash water (FBWW) and clarifier sludge blowdown was treated with a pilot-scale ultrafiltration (UF) membrane. The UF treated residuals stream was blended with plant finished water to model a recycle return location to the clearwell of a drinking water treatment train. Field evaluations involved the use of bench-scale annular reactors (ARs) to investigate two parallel model distribution systems: (i) a control system representing 100% plant filtered water (AR-filtered) and (ii) a blended system representing 10% by volume of the UF permeate blended with plant filtered water (AR-blended). To evaluate the impact of chemical disinfection on bacterial regrowth and the fate of inorganic constituents, free chlorine, monochloramine and chlorine dioxide were fed to the ARs during three distinct disinfection trials. An initial disinfection trial consisted of a control run during which no disinfectant was added. The results of the field experiments showed that blending 10% by volume of the UF permeate with plant finished water did not significantly impact regrowth of heterotrophic bacteria in the bulk or biofilm phases. Throughout each disinfection trial, the largest difference in HPCs noted between the two systems was 1 log in the bulk and biofilm samples. Free chlorine and monochloramine disinfection resulted in the greatest mitigation of microbial biofilm in both AR-filter and AR-blended systems. Aluminum, manganese, and iron in the bulk phase were found to be below Canadian Drinking Water Guideline levels. However, the detection of manganese-oxidizing bacteria (MOB) and elevated manganese in biofilm samples from AR-blended could hold potential implications for blending UF treated residual streams on biofilm development and inorganic post-precipitation occurrences in systems treating source waters containing elevated concentrations of manganese.

**Key words** | bacteria regrowth, biofilm, filter backwash water, inorganic post-precipitation, manganese oxidizing bacteria, ultrafiltration

### INTRODUCTION

The two largest waste residual streams generated from conventional water treatment are filter backwash water (FBWW) and clarifier sludge blowdown. FBWW is typically defined as a high volume, low solids waste stream that is

produced when clean water is directed in a counterflow configuration through the filter bed to purge particulate matter that has accumulated in the filter media. Depending upon plant design, clarifier sludge is generated in sedimentation

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basins or up-flow clarifiers and represents a low volume, higher solids waste residual stream in comparison to the FBWW stream (10,000–50,000 mg/l and 50–400 mg/l, respectively) (MWH Inc., 2005). The physico-chemical characteristics of FBWW and clarifier sludge blowdown have shown to be highly variable due to differences in raw water quality and treatment plant design and operation (Arora *et al.* 2001; Cornwell *et al.* 2001; Edzwald *et al.* 2001; Bourgeois *et al.* 2004). However, these water treatment plant (WTP) waste residuals streams are generally characterized as having appreciable levels of naturally occurring colloidal and particulate matter removed in the water treatment processes (e.g., clay, silt, natural organic matter (NOM), microorganisms and metals), as well as chemical precipitates formed during coagulation and flocculation (e.g.,  $\text{Al}(\text{OH})_3$ ,  $\text{Fe}(\text{OH})_3$ ). Various studies have also reported that elevated concentrations of *Cryptosporidium parvum* and other disinfection-resistant pathogens occur within the WTP waste residual stream (LeChevallier *et al.* 1991; Cornwell & Lee 1994; States *et al.* 1995; Cornwell *et al.* 2001).

In the United States, recycling FBWW within the main treatment train to alleviate challenges associated with discharge limits or for conservation purposes is a critical management option for many water utilities. A recent analysis of the U.S. EPA Information Collection Rule (ICR) database found that of 362 drinking water filtration plants, 62.4% of the facilities reported recycling a waste stream (Hamele & Bonner 1998). A similar survey of 333 water utilities in the U.S. found that 20 to 40% of the participating plants had no alternative to recycling, with direct discharge to sewer or the ability to obtain a discharge permit was not possible (Cornwell *et al.* 2001). Although treatment of recycle streams is not mandated by specific legislation in the United States or Canada, many utilities choose to provide treatment of residuals streams.

In general, sedimentation is the most common method for treating waste residuals recycle streams (Hamele & Bonner 1998; Cornwell *et al.* 2001). However, several high-rate processes such as dissolved air flotation (DAF), microsand-assisted settling, and low-pressure membrane filtration have been investigated at bench- and pilot-scale for the treatment of FBWW (Thompson *et al.* 1995; Kawamura 2000; Shealey *et al.* 2000; Taylor *et al.* 2000;

Cornwell *et al.* 2001; Bourgeois *et al.* 2004). The collective results of these studies suggest that the use of high-rate processes can potentially offer both capital and operational advantages over conventional sedimentation systems in terms of the ability of these technologies to achieve higher loading rates, reduced footprint, reduced chemical dosages, and improved adaptability to variable influent water quality.

In particular, results of pilot-scale microfiltration (MF) and ultrafiltration (UF) membrane studies have consistently shown that the treatment of FBWW streams with this technology can result in significant reductions in particulate matter as measured by turbidity, particle counts, total suspended solids (TSS), microbial pathogens and particulate inorganic material (Thompson *et al.* 1995; Shealey *et al.* 2000; Taylor *et al.* 2000; Cornwell *et al.* 2001; MacPhee *et al.*, 2002; Bourgeois *et al.* 2004; LeGouellec *et al.* 2004). Effective removal of suspended or colloidal particles from feed streams with MF and UF membranes is accomplished *via* a sieving mechanism based on the size of the membrane pores relative to that of the particulate matter. Based on this removal mechanism, UF membrane filtration with a rated pore size of 0.01 to 0.1  $\mu\text{m}$  can effectively remove particulate matter and microorganisms. The results of several challenge studies using pilot-scale MF/UF membranes have reported significant removals of protozoan and bacterial organisms (Dwyer *et al.* 1995; Jacangelo *et al.* 1995; Hagen 1998; MacPhee *et al.* 2002; LeGouellec *et al.* 2004) and further support the application of this technology for FBWW treatment. However, based on the principle of pore size exclusion, organic and inorganic material that is dimensionally smaller than the nominal pore size would be expected to pass through the membrane surface.

The potential downstream impacts on finished water quality from dissolved material not removed from residuals recycle streams with UF membrane filtration has not been widely reported in the literature. In particular, the biodegradable fraction of natural organic matter (NOM) in water has been shown to play a critical role in the proliferation of bacteria and increased incidences of biofouling on pipe surfaces within distribution systems (LeChevallier *et al.* 1987; van der Wende *et al.* 1989; Mathieu *et al.* 1993). The most readily biodegradable fraction of organic carbon has been termed assimilable organic carbon (AOC) and is generally composed of small molecular weight compounds.

In full-scale plant evaluations conducted by Cornwell and Lee (1994), recycling FBWW containing elevated AOC concentrations was found to increase the filtered water AOC levels, although the potential impact on microbial regrowth in the distribution system was not investigated. As well, the precipitation and assimilation of inorganics into microbial biofilm matrices by biological or chemical oxidation pathways have been shown to influence biofouling occurrences and corrosion rates of pipe surface (Sly *et al.* 1990; Dickinson *et al.* 1996; Murdoch & Smith 2000). Thus, it is plausible that biodegradable organic matter (BOM) and dissolved metals originating from residuals streams could have deleterious impacts on chemical and microbial quality in distribution systems.

The purpose of this paper was to critically examine microbial and chemical water quality impacts of blending UF treated waste residuals streams with plant finished water in a modelled distribution system. In the United States, the *Filter Backwash Recycle Rule* requires systems that recycle FBWW, thickener supernatant or liquids from dewatering processes to return these flows through the processes of a system's existing conventional (i.e. coagulation, flocculation, sedimentation and filtration) or direct (i.e. coagulation, flocculation and filtration) filtration systems (U.S. EPA 2001). The recycle return location requirement ensures that at least 2-log removal of *Cryptosporidium* is obtained, and is a logical choice for untreated residuals or recovered residuals streams that are treated using conventional technology (i.e. sedimentation or enhanced sedimentation with plates or tubes).

Previous challenge studies have demonstrated that MF and UF membranes are capable of achieving greater than 4.8-log removal of *Cryptosporidium* (Dwyer *et al.* 1995; Jacangelo *et al.* 1995, 1997; Schneider *et al.* 1999; MacPhee *et al.* 2002). These studies also demonstrated the wide range in removal efficiencies for viruses by MF and UF systems, from marginal (i.e. less than 0.5-log) to below detection limits (i.e. >6.5-log). As such, this project evaluated the feasibility of blending UF treated residuals streams upstream of a drinking water plant's clearwell as presented in Figure 1. This recycle design would account for the potential presence of certain viruses in the permeate stream due to reduced removal with UF treatment of the mixed residuals stream. For plants that are regulated by

concentration x time (Ct) requirements, this recycle return location would provide for inactivation of viruses through proper disinfection strategies.

The specific objectives of this study were to investigate this recycle return location in terms of microbial regrowth and the fate of specific inorganic contaminants (e.g. aluminum, manganese and iron) in the bulk and biofilm phases using bench-scale annular reactors (ARs). Two ARs were operated in parallel to simulate (i) a control system representing 100% plant filtered water (AR-filtered), and (ii) a blended system representing 10% by volume of UF permeate blended with plant filtered water (AR-blended). In addition, disinfection trials with chlorine, monochloramine, and chlorine dioxide were conducted in series to investigate regrowth and post-precipitation occurrences under various chlorine-based disinfection strategies.

## MATERIALS AND METHODS

### Description of field test site

This project was conducted on-site at the Lake Major WTP in Dartmouth, Nova Scotia, Canada. The Lake Major WTP is a 45 MLD surface water plant with a treatment train consisting of coagulation with alum (11 mg/L typ. as  $\text{Al}_2(\text{SO}_4)_3 \cdot 18 \text{H}_2\text{O}$ ) followed by two parallel high-rate upflow solids-contact clarifiers and four parallel dual-media anthracite sand filters. Chlorine is added prior to the dual-media filters to provide enhanced oxidation for manganese removal as well as to the clearwell to maintain a 1 mg/l free chlorine residual at the entry point to the distribution system. The raw water quality is characterized by low turbidity (<0.3 NTU) and low TOC levels (<3.0 mg/l), with slightly elevated levels of total aluminum (0.21 mg/l) and manganese (0.08 mg/l) (Bourgeois *et al.* 2004). During the project, the filters were backwashed on average every 105 to 120 hours with effluent turbidity measurements typically less than 0.1 NTU. The average daily backwash flow was calculated to be approximately 0.34 MLD and clarifier sludge blowdown was reported to be wasted at a rate of approximately 0.37 MLD (Bourgeois *et al.* 2003). Thus the plant typically loses slightly less than 1.5%, by volume, of its process water to waste streams.

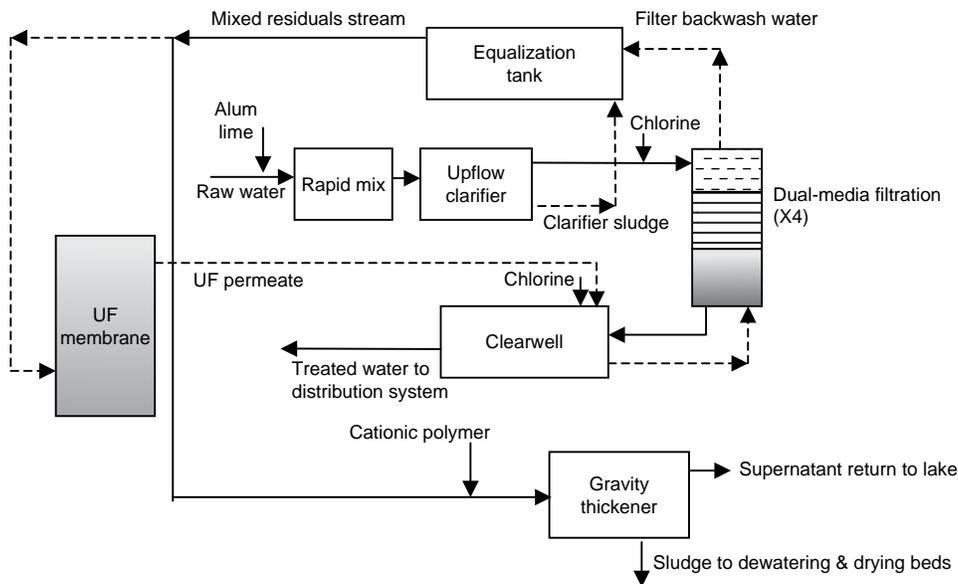


Figure 1 | Proposed UF treated residuals return location in conventional drinking water treatment plant.

The plant combines FBWW and clarifier sludge in a continuously mixed equalization tank to homogenize the two residuals streams. A cationic polymer (Novus CE2667, GE Infrastructure Water and Process Technologies,) is added at a dosage of 0.1 mg/l to the outlet of the equalization tank prior entering to two parallel gravity thickeners. The supernatant from the thickening tanks is discharged back to the source water.

### Bench- and pilot-scale test equipment

The field test equipment consisted of a pilot-scale UF membrane (ZeeWeed-10, ZENON Environmental Ltd, Burlington, ON) and two bench-scale biofilm annular reactors (BioSurface Technologies Corp., Bozeman, MT). The ZW-10 module utilizes a ZeeWeed™ 500 hollow-fiber membrane model with a total membrane surface area of 0.93 m<sup>2</sup> (10 ft<sup>2</sup>) and rated 0.04 μm nominal and 0.1 μm absolute pore size. The system was operated in an “immersed”, outside-in dead-end configuration with continuous air injection at the bottom of the membrane module to reduce fouling on the fibre surface. ARs have been used extensively in model distribution system experimental work due to their adaptable configuration which allows for input and operational variables to be manipulated for specific investigative needs (Camper 1996; Gagnon *et al.* 2004, 2005).

The reactors were constructed of a stationary outer cylinder that encloses a rotating inner cylinder on which removable coupons composed of standard pipe material are flush-mounted.

### Experimental design

For this research project, the ZW-10 module was operated in a dead-end configuration at a constant flux of 0.023 m/hr. An equalized blend of FBWW and clarifier sludge blow-down from the plant equalization tank was periodically pumped into a satellite storage tank to provide a dedicated feed supply to the UF process tank. The pilot system was operated in batch mode to supply sufficient permeate volume to feed to the ARs, and was routinely cleaned as per the manufacturer's specifications once the transmembrane pressure (TMP) reached 17 kPa (2.5 psi). Cleaning procedures for the pilot-scale UF system included a 5 hour soak in 1,000 mg/l sodium hypochlorite (NaOCl), followed by operations in a forward flow configuration for 30 minutes and a backward flow configuration for 30 minutes. The process tank was then drained, recharged with tap water and operated in a forward flow configuration for a period of 60 minutes.

The bench-scale model distribution system involved the operation of two ARs equipped with PVC coupons in

parallel to evaluate the impact of blending the UF treated mixed residual stream with plant finished water. The ARs were operated at a fixed rotational speed of 50 rpm, which equates to a shear stress of  $0.25 \text{ N/m}^2$  at the outer wall of the reactor and a flow rate of  $0.30 \text{ m/s}$  in a  $100 \text{ mm}$  ( $4 \text{ in.}$ ) diameter smooth pipe (Camper 1996). Prior to entering the ARs, the plant finished water was dechlorinated using an up-flow granular activated carbon (GAC) filter. The removal of DOC in the GAC filter averaged  $0.6 \pm 0.1 \text{ mg/l}$  through each of the disinfection trials. A retention time of 1 hour in each reactor was established for the duration of the tests. AR-filtered was continuously fed 100% plant finished water ( $15.8 \text{ ml/min}$ ) while AR-blended received a blend of 10% ( $1.6 \text{ ml/min}$ ) of UF treated waste residuals and 90% ( $14.2 \text{ ml/min}$ ) plant finished water. A blend ratio of 10% for the UF permeate was chosen due to the fact that full-scale water treatment plants typically recycle FBWW streams at a rate of 3 to 10% of raw water flows (Edzwald *et al.* 2001).

To evaluate the impact of chemical disinfection on microbiological regrowth and the fate of metals in drinking water, free chlorine, chloramines, and chlorine dioxide were fed to the ARs during three distinct disinfection trials. Each disinfection trial lasted for a period of 4 weeks, during which bulk water and biofilm samples were collected weekly from the AR-filtered and AR-blended systems. Target disinfectant residual concentrations at the effluent of each AR were  $1.0 \text{ mg/l}$  for chlorine and monochloramine and  $0.2 \text{ mg/l}$  for chlorine dioxide. The target disinfectant values were chosen to be within minimum and maximum limits that have been established by North American regulatory agencies. As well, a control trial was run where no disinfectant was added. The ARs were operated for a 4-week period prior to the commencement of each disinfection trial to establish a pseudo-state biofilm on the reactor coupons. Bulk water quality parameters were monitored through sampling from the effluent port, with biofilm analysis achieved by sampling the removable coupons mounted on the inner cylinder. Throughout each of the disinfection trials, samples were taken weekly from the UF process tank, UF permeate tank and bench-scale clearwell. Samples collected were characterized for the following parameters: pH, turbidity, colour, UV<sub>254</sub>, DOC and HPC (suspended and biofilm), and aluminum, manganese and iron (total and dissolved fractions).

## Analytical methods

Heterotrophic bacteria in the bulk water and biofilm phases were measured using the heterotrophic plate count (HPC) spread plate technique on R2A agar as described in *Standard Methods for the Examination of Water and Wastewater* (APHA *et al.* 1995). Biofilm samples from the ARs were taken by removing a test PVC coupon and placing in a sterile test tube containing a 25 ml phosphate buffered saline (PBS) and 0.1% w/v sodium thiosulfate solution. The attached microbiological cells were removed by transferring the PBS solution and sample coupon to a sterile stomacher bag and stomaching at 230 rpm for 2 minutes as described by Gagnon and Slawson (1999). Serial dilutions were prepared from each sample analyzed to yield 30 to 300 colonies per plate. HPC plates were incubated at room temperature for 7 days and then enumerated using a colony counter. Manganese-oxidizing bacteria (MOB) were monitored in the suspended and biofilm samples by plating on R2A agar that had been modified through the addition of  $50 \text{ mg MnSO}_4 \cdot \text{H}_2\text{O}$  per liter of original medium, as described by Smith (1992). MOB plates were incubated at room temperature in the dark for 28 days and enumerated by identification of black colonies formed in the modified agar media. All HPC and MOB samples were plated in duplicate.

Bulk water samples for dissolved organic carbon (DOC) analysis were filtered through a  $0.45 \mu\text{m}$  pore-size membrane filter (Cole-Parmer® Nylon Membranes) rinsed with deionised water as described in *Standard Methods for the Examination of Water and Wastewater* (APHA *et al.* 1995). The filtered samples were transferred headspace-free in 40-ml glass vials and preserved with concentrated phosphoric acid to a  $\text{pH} < 2$ . Measurements were performed with a TOC-V CHP analyzer (Shimadzu Corporation, Kyoto, Japan). Apparent colour was measured using a spectrophotometer (HACH DR/4000, HACH Co., Loveland, CO) with the American Dye Manufacturers Institute (ADMI) Weighted Ordinate method. UV<sub>254</sub> was measured with a UV/VIS spectrophotometer (Hach DR/4000, Loveland, CO) according to method 5910B in *Standards Methods for the Examination of Water and Wastewater* (APHA *et al.* 1995).

Bulk water samples for metal analysis were collected in amber glass bottles that had been washed, soaked in a 1 N

hydrochloric acid (HCl) solution for 24 hours, and then rinsed three times with deionised water. These bulk water samples were acidified to a pH <2 with 1 N nitric acid (HNO<sub>3</sub>) for a 24-hour period to allow for sample digestion prior to analysis. Biofilm samples for metal analysis were taken by removing a test PVC coupon and soaking the extracted coupon in 70 ml of deionised water adjusted to a pH of <2 with a 1 N nitric acid (HNO<sub>3</sub>) for 24-hours. The coupons were then removed from the pH adjusted deionised water soak, and manually scraped in the soaking water medium to remove any remaining attached inorganic material on the surface of the coupon. The pH of all samples was adjusted back to 7.0 with a 1 N sodium hydroxide (NaOH) solution prior to analysis. Total aluminum, manganese, and iron concentrations were measured using a HACH DR/4000 Spectrophotometer according to USEPA Method #200.7. The dissolved fraction of these metals were analyzed with the same procedure after filtering the samples through a 0.2 µm filter as described by Carlson *et al.* (1997).

## RESULTS AND DISCUSSION

### AR influent water quality

Throughout each of the disinfection trials, samples were taken weekly from the UF process tank, UF permeate tank and bench-scale clearwell. Each sample was characterized for the following parameters: pH, turbidity, colour, UV254, DOC, aluminum, manganese and iron. Table 1 summarizes the average results of the monitored parameters for each of the sample locations. Collectively, these results illustrate the concentration of organic and inorganic contaminants within the mixed residuals stream and significant reductions achieved with UF membrane filtration. DOC concentrations in the mixed residuals stream varied from 20 to 153 mg/l during the disinfection trials. Other studies have shown DOC concentrations in FBWW streams can vary within the range of 0.8 to 191 mg /l (Arora *et al.* 2001; Cornwell *et al.* 2001). UV254 measurements ranged between 0.028 and 3.5 cm<sup>-1</sup> in the mixed residuals stream during the disinfection trials. UV254 measurements of FBWW streams in other studies have been reported to range between 0.014 and 0.079 cm<sup>-1</sup> (Edzwald *et al.* 2001).

UF treatment of the mixed residuals stream resulted in significant reductions in turbidity (from 123.4 ± 70.8 to 0.17 ± 0.05 NTU), producing a permeate stream with lower turbidity than the plant filtered water samples. Natural organic matter (NOM) as measured by DOC, UV254 and colour was reduced greater than 95% with UF treatment of the mixed residuals stream. Similar reductions were achieved for total aluminum and iron. Approximately three quarters of the total manganese was removed with a UF permeate concentration of 0.11 ± 0.06 mg/l. However, with the exception of iron and turbidity, statistical analysis of the test results showed that the UF permeate samples contained significantly ( $P < 0.05$ ) higher levels of DOC, UV-254, colour and aluminum and manganese (total and dissolved) compared to the plant filtered water samples.

The limited ability of UF membranes to effectively remove NOM has often been related to the fundamental mechanism for contaminant removal (i.e. pore size exclusion). Previous studies have shown that in the absence of coagulant, polymer or adsorbent addition, the passage of organic material smaller in dimension to the rated pore size of the membrane can be expected (Laine *et al.* 1990; Maatens *et al.* 1999; Mourato *et al.* 1999; Siddiqui *et al.* 2000; Bérubé *et al.* 2002). Similarly, results of pilot-scale UF studies presented by Bourgeois *et al.* (2004), showed reduced removal of soluble inorganic contaminants in that approximately 39% of the total manganese in the UF feed stream present in the dissolved form passed through the membrane barrier into the permeate. However, the concept of pore size exclusion as the principal mechanism for contaminant removal does not support other work which has shown that removal of smaller sized contaminants (i.e. viruses) can be influenced by a number of factors including adsorption onto the membrane surface, adsorption onto particles in the cake layer, or sieving as a result of physical constriction of the membrane pores due to irreversible fouling (Jacangelo *et al.* 1995).

As the primary objective of this study was to evaluate the impacts of blending membrane treatment residuals streams on finished water quality, investigations into pre-treatment of the UF feed stream were not incorporated into the experimental design. Therefore, while high removal of particulate inorganic matter was achieved with UF treatment, the removal of material in the

**Table 1** | Summary of monitored water quality parameters

Parameter	Units	UF process tank <sup>1</sup>	UF permeate tank	Plant filtered water
pH	units	6.9 ± 0.4 (N = 11)	6.9 ± 0.4 (N = 15)	7.2 ± 0.6 (N = 16)
Turbidity	NTU	123.4 ± 70.8 (N = 10)	0.17 ± 0.05 (N = 20)	0.27 ± 0.10 (N = 22)
Colour	TCU	1500 ± 100 (N = 3)	6 ± 3 (N = 10)	4 ± 2 (N = 11)
DOC	mg/l	107 ± 80 (N = 12)	4.8 ± 1.3 (N = 23)	2.2 ± 0.5 (N = 24)
UV-254	cm <sup>-1</sup>	1.59 ± 1.39 (N = 12)	0.078 ± .041 (N = 18)	0.023 ± 0.013 (N = 20)
Aluminum total	mg/l	29.4 ± 12.2 (N = 9)	0.06 ± 0.05 (N = 20)	0.01 ± 0.01 (N = 21)
Dissolved	mg/l	0.24 ± 0.36 (N = 3)	0.05 ± 0.03 (N = 13)	0.02 ± 0.01 (N = 13)
Manganese total	mg/l	0.46 ± 0.09 (N = 10)	0.11 ± 0.06 (N = 20)	0.03 ± 0.02 (N = 21)
Dissolved	mg/l	0.22 ± 0.12 (N = 4)	0.06 ± 0.02 (N = 13)	0.02 ± 0.01 (N = 14)
Iron total	mg/l	5.9 ± 0.9 (N = 4)	0.05 ± 0.04 (N = 13)	0.06 ± 0.03 (N = 14)
Dissolved	mg/l	0.38 ± 0.45 (N = 4)	0.03 ± 0.02 (N = 13)	0.03 ± 0.01 (N = 14)

<sup>1</sup>Equalized blend of FBWW and clarifier sludge from WTP Equalization Tank.

dissolved fraction was found to be more challenging. In particular, total aluminum and iron were reduced by 99% with UF membrane filtration of the mixed residuals stream while dissolved aluminum and iron were reduced by 79 and 92%, respectively. A comparison of the dissolved manganese removal results of this study (e.g. 73%) to those reported by Bourgeois *et al.* (2004) (e.g. 39%) further emphasizes the conclusions of Jacangelo *et al.* (1995) that removal of smaller sized contaminants can be influenced by a number of factors. Overall, the contaminant reductions found in this project present interesting findings, and would warrant targeted investigations into the specific organic and inorganic fractions present in residuals streams and impacts to membrane filtration removal efficacy.

## Microbiological regrowth

### Acclimation period

AR-filtered and AR-blended were operated without the addition of disinfectants for a 4-week acclimation period

prior to each disinfection trial to allow for a steady-state biofilm to develop on the reactor coupons. During each acclimation period, the number of suspended heterotrophic bacteria ranged from  $2.8 \times 10^4$  to  $6.9 \times 10^5$  CFU/ml in AR-filtered, and  $1.9 \times 10^4$  to  $4.3 \times 10^5$  CFU/ml in AR-blended in samples. Biofilm formation was also similar in both ARs, with heterotrophic bacteria densities ranging from  $2.5 \times 10^5$  to  $7.3 \times 10^6$  CFU/cm<sup>2</sup> in both systems. Collectively, HPCs measured in the bulk and biofilm phases of AR-filtered and AR-blended were not found to be significantly ( $P > 0.05$ ) different during the acclimation periods.

The number of heterotrophic bacteria in the bulk water phase (suspended) and attached (biofilm) measured in this study were found to be similar to steady-state HPC levels reported in other bench-scale studies with annular reactors (Camper 1996; Ollos *et al.* 2003; Gagnon *et al.* 2004, 2005). The bacterial data presented in this study are also in the range of data found in pilot- and full-scale investigations. Specifically, suspended bacteria concentrations have been reported to range between  $10^3$  to  $10^5$  bacteria/ml (Mathieu *et al.* 1993; Eisnor & Gagnon 2004) and biofilm

concentrations have been found to range between  $10^5$  to  $10^7$  bacteria/cm<sup>2</sup> (Butterfield *et al.* 1999; Volk & LeChevallier 1999) in studies at the pilot- and full-scale.

### Disinfection trials

Disinfectant residuals measured on effluent samples from AR-filtered and AR-blended during the disinfection trials are presented in Table 2 along with calculated Ct values. Figure 2 illustrates the suspended and biofilm heterotrophic bacteria counts measured from AR-filtered and AR-blended samples collected during each of the disinfection trials. The number of suspended heterotrophic bacteria measured in effluent samples from both AR systems was not statistically significantly different ( $P > 0.05$ ) through each of the disinfection trials. The application of free chlorine and monochloramine at concentration x time (Ct) values of 58 and 56 mg/l-min, respectively, resulted in significant ( $P < 0.05$ ) reductions in suspended HPCs from values measured during the control trial. In particular, free chlorine disinfection resulted in 4.5- and 4.1-log reductions in suspended HPCs in AR-filtered and AR-blended, respectively. During the monochloramine disinfection trial, suspended bacteria were reduced by 0.7 and 1.0-log in AR-filtered and AR-blended, respectively. These results were consistent with other bench-scale studies that reported similar log inactivation of suspended HPCs at equivalent Ct values (Gagnon *et al.*, 2004).

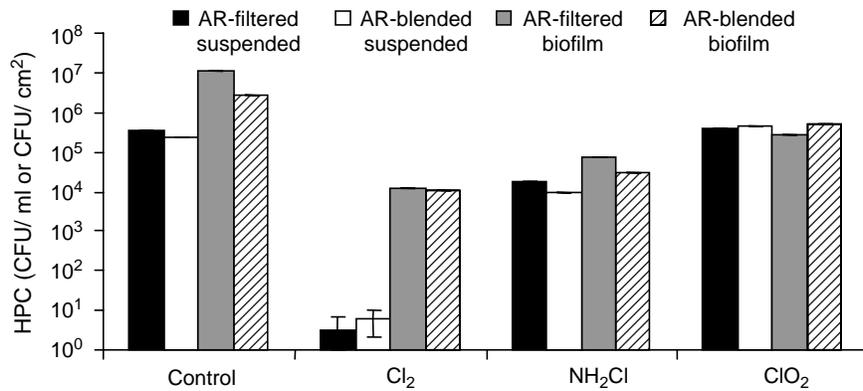
During the control trial, bacteria densities measured from biofilm samples were found to be significantly ( $p = 0.003$ ) lower in the blended AR. Similarly, throughout each acclimation period when the ARs were operated without the addition of disinfectant, AR-blended had lower biofilm HPCs as compared to AR-filtered. However,

the geometric mean of HPCs measured in the UF permeate feed stream were found to be significantly ( $p = 0.004$ ) lower than measurements taken from the plant filtered water samples ( $1.4 \times 10^4$  CFU/ml and  $1.2 \times 10^5$  CFU/ml, respectively) and would have resulted in a diluted feedstock of bacteria to the blended AR. During each of the subsequent disinfection trials, HPCs in the biofilm samples were not shown to be significantly different between the two model distribution systems ( $P > 0.05$ ) suggesting that under disinfection conditions, the dynamics of biofilm development in both systems were equalized.

In both AR systems, free chlorine disinfection resulted in higher inactivation of suspended heterotrophic bacteria than in the biofilm. Specifically, log inactivation of suspended HPCs were 4.8- and 4.3-log in AR-filtered and AR-blended, respectively, during chlorine disinfection. Biofilm HPCs in AR-blended and AR-filtered were only reduced by 3.2 and 2.7-log with chlorine disinfection. Previous studies have found that disinfection is less effective for biofilm bacterial cell inactivation as compared to suspended colonies, due to adhesion (LeChevallier *et al.* 1988; Huang *et al.* 1995; Camper *et al.* 1997) and/or incomplete penetration of the disinfectants into the biofilm layer (Gagnon *et al.* 2004). In contrast, monochloramine disinfection resulted in a higher log inactivation in the biofilm phase (2.5- and 2.0-log in AR-filtered and AR-blended, respectively) as compared to HPC log inactivation in the suspended phase (1.1-log in AR-filtered and AR-blended). Other studies have suggested that chloramines can penetrate deeper into the biofilm layer to limit its growth (Kirmeyer *et al.* 2003), while consumption of free chlorine with other constituents may limit its ability to penetrate the biofilm (LeChevallier *et al.* 1990; Camper *et al.* 1997).

**Table 2** | Disinfectant residuals and Ct Values for AR-Filtered and AR-Blended

Disinfectant	Target residual concentration (mg/l)	Average measured residual concentration (mg/l)		Ct (mg/l-min)	
		AR-filtered	AR-blended	AR-filtered	AR-blended
Cl <sub>2</sub>	1.0	0.98 ± 0.19 (N = 9)	0.95 ± 0.16 (N = 9)	59	57
NH <sub>2</sub> Cl	1.0	0.89 ± 0.16 (N = 10)	0.98 ± 0.16 (N = 10)	53	59
ClO <sub>2</sub>	0.2	0.18 ± 0.03 (N = 6)	0.21 ± 0.01 (N = 6)	11	13



**Figure 2** | Suspended and biofilm HPCs in AR-filtered and AR-blended during disinfection trials. (Error bars represent standard deviation from mean).

Chlorine dioxide disinfection at a lower Ct value of 12 mg/l-min did not reduce suspended HPCs in either AR system. However, heterotrophic bacteria densities in biofilm samples were reduced by 1.6 and 0.8 log in AR-filtered and AR-blended, respectively, with chlorine dioxide disinfection. Previous studies have reported chlorine dioxide to be an effective disinfectant for both suspended and biofilm HPC control (Gagnon *et al.* 2004), with a log inactivation of 1.6 to 1.8 for suspended cells at a Ct value of 14 mg/l-min. However, biofilm HPC concentrations on PVC test coupons were reduced by 1.0 log in the same studies, and correspond with the results of the present investigation.

Collectively, the results of heterotrophic bacteria measurements in samples taken from AR-filtered and AR-blended during the disinfection trials are consistent with data collected during the acclimation periods. Specifically, HPC data collected during the disinfection trials further demonstrate that the introduction of a 10% blend of UF treated waste residuals to the plant finished water did not impact regrowth of heterotrophic bacteria in the model distribution system. The overall average DOC concentration in the plant finished water feed stream to the two ARs was  $2.2 \pm 0.5$  mg/l through each of the disinfection trials. In contrast, DOC concentrations in the UF permeate feed stream to AR-blended were significantly ( $P < 0.05$ ) higher during each of the disinfection trials, and ranged from 4.1 to 7.1 mg/l. However, as presented in Table 3, influent DOC concentrations to AR-filtered and AR-blended were not significantly ( $P > 0.05$ ) different due to dilution of the UF permeate blended with the plant finished water at a ratio of 1:10. Similarly, ANOVA results showed

that effluent DOC concentrations were not significantly ( $P > 0.05$ ) different between the two AR systems during each disinfection trial. These results indicate that both model distribution system water matrices contained biologically available NOM that supported similar numbers of heterotrophic bacteria. Thus, it is hypothesized that the organic carbon introduced into the blended AR system with the UF permeate feed contained recalcitrant NOM and the extent to which it affected heterotrophic bacteria growth was minimal.

### Visual observation of biofilm formation

Weekly visual inspection of the PVC coupons sampled from AR-filtered and AR-blended was conducted to qualitatively evaluate deposition and biofouling on the surface of the test coupons. Within the first weeks of the control trial, a discoloration of the coupons in AR-blended was visible, with a thin brownish layer developed on the surface of the coupons. This visual discoloration was not apparent on the test coupons sampled from the AR-filtered system. This discoloration of the AR-blended coupons did not occur during subsequent disinfection trials. Scanning Electron Microscopic (SEM) micrographs of the surface of the PVC coupons were taken and are presented in Figure 2.

The SEM images revealed the presence of initial bacterial growth on the coupons in AR-blended system in addition to a considerable coating of deposited material on the surface of the test coupon. In comparison, the test coupon from AR-filtered showed a more developed bacterial biofilm in the early stages of development, and

**Table 3** | Average DOC concentrations through disinfection trials

Disinfection trial	Plant finished water	UF permeate	AR influent DOC <sup>1</sup>		AR effluent DOC	
			AR-filtered	AR-blended	AR-filtered	AR-blended
Control	1.9 ± 0.6 (N = 9)	4.1 ± 1.2 (N = 11)	1.9 ± 0.6	2.1 ± 0.9	1.8 ± 0.6 (N = 9)	1.9 ± 0.4 (N = 9)
Cl <sub>2</sub>	2.4 ± 0.6 (N = 8)	4.5 ± 0.7 (N = 8)	2.4 ± 0.6	2.6 ± 0.6	2.0 ± 0.4 (N = 8)	2.4 ± 0.3 (N = 8)
NH <sub>2</sub> Cl	2.4 ± 0.2 (N = 7)	7.1 ± 2.3 (N = 7)	2.4 ± 0.2	2.7 ± 0.4	2.4 ± 0.1 (N = 7)	2.6 ± 0.3 (N = 7)
ClO <sub>2</sub>	1.1 ± 0.4 (N = 3)	6.6 ± 1.6 (N = 3)	1.1 ± 0.4	1.6 ± 0.4	1.1 ± 0.7 (N = 3)	1.8 ± 0.7 (N = 3)

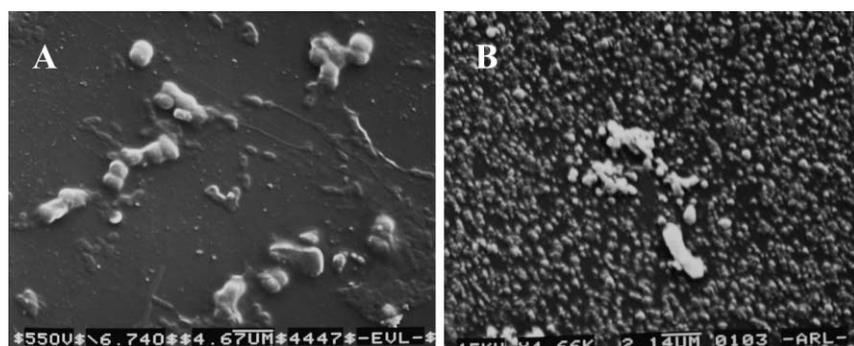
<sup>1</sup>AR -filtered based on 100% plant finished water DOC concentration, AR-blended influent calculated from 90% plant finished water and 10% UF permeate DOC concentration.

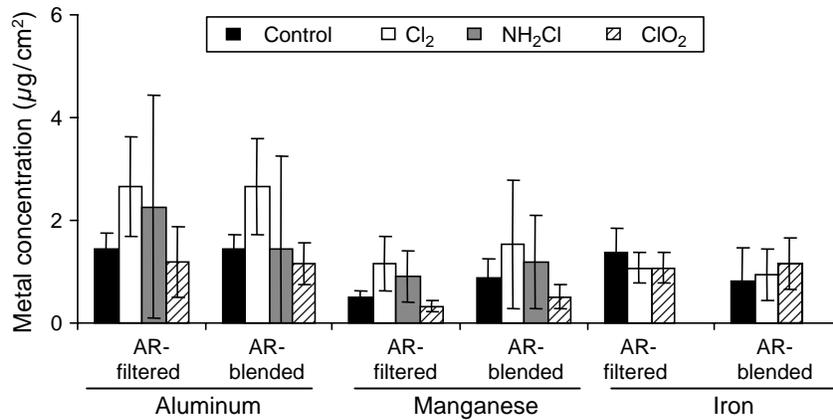
the absence of the deposited material. Results of metal analysis on biofilm samples taken from the test coupons when no disinfectant was applied (Figure 3) showed manganese concentrations were significantly higher on test coupons from AR-blended as compared to test coupons sampled from AR-filtered ( $p = 0.023$ ). However, aluminum and iron concentrations were not found to be statistically different between the two AR systems. ANOVA results of biofilm metal measurements taken during the remaining disinfection trials did not show significant differences between AR-filtered and AR-blended for aluminum, iron, or manganese concentrations (Figure 4).

Inspection of the SEM micrographs of the test coupon from AR-blended in combination with elevated manganese in biofilm samples suggested that the deposited material on the coupon was chemically deposited manganese oxide (MnO<sub>2</sub>). Specifically, chemically deposited MnO<sub>2</sub> coatings have been found to be generally free of microorganisms that characterize microbial manganese deposits (Sly *et al.* 1990). Although disinfection with chlorine, monochloramine or

chlorine dioxide had not started at this point in the experiments, sodium hypochlorite (NaOCl) cleaning procedures utilized for the pilot membrane unit may have resulted in oxidation of soluble manganese into its precipitated form of MnO<sub>2</sub>. The deposition of inorganic and organic particles in an aqueous suspension is influenced by surface charge and electrostatic interactions (Bowen & Epstein 1979). The surface charge of particles in natural waters is dependent on the solution pH, with the zero point of charge (ZPC) corresponding to a surface charge of zero. Manganese oxide has a low zero point charge (ZPC) in range with that of humic acid (2–4.5 and 3, respectively) (MWH Inc., 2005). Humic acids are the major natural organic matter in drinking water distribution systems and tend to be adsorbed onto distribution system pipe surfaces. Therefore, the similar ZPC of manganese precipitates would concur with increased potential for the sorption of MnO<sub>2</sub> onto the AR coupon surfaces.

To evaluate the possible influence of biologically induced manganese deposition, biofilm samples were

**Figure 3** | SEMs of (A) AR-filtered and (B) AR-blended coupon surfaces during control trial.



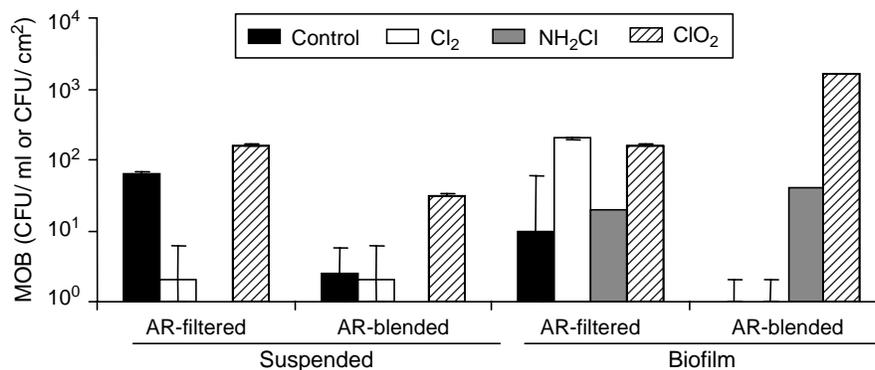
**Figure 4** | Metal concentrations in biofilm samples from AR-filtered and AR-blended during disinfection trials. (Error bars represent standard deviation from mean).

enumerated for indigenous manganese-oxidizing bacteria (MOB). Figure 5 presents MOB counts enumerated from bulk water and biofilm samples from both ARs during the disinfection trials. During the control trial, the number of suspended MOB was  $6.3 \times 10^1$  and  $2.5 \times 10^0$  CFU/ml in AR-filtered and AR-blended effluent samples, respectively. Analysis of biofilm samples showed similar low MOB densities of  $1 \times 10^1$  and  $1 \times 10^0$  CFU/cm<sup>2</sup> in AR-filtered and AR-blended, respectively. Although these oxidizing bacteria were detected within the biofilm samples of both test ARs, their particular influence on the precipitation of manganese observed within AR-blended was likely minimal. Chemical deposition of manganese has been shown to be a more acute pathway than microbially induced manganese deposition, with rapid build-up of MnO<sub>2</sub> occurring over a short period (Sly *et al.* 1990). Subsequent biofilm MOB concentrations during the other disinfection trials were found at elevated concentrations ( $10^2$  and

$10^3$  CFU/cm<sup>2</sup>) when the presence of the MnO<sub>2</sub> micronodules on the test coupon surfaces was not detected. However, the detection of manganese-oxidizing bacteria (MOB) and elevated manganese within biofilm samples of AR-blended was significant in terms of the potential implications of blending UF treated waste residual streams on biofilm development and biofouling.

## CONCLUSIONS

This project evaluated the impact of blending 10% (by volume) UF treated waste residuals streams with plant finished water on biological and chemical water quality using a model distribution system. In particular, a UF treated residuals stream was blended with plant finished water to model a recycle return location to the clearwell of a drinking water treatment train. Microbial and chemical



**Figure 5** | Suspended and biofilm MOB in AR-filtered and AR-blended during disinfection trials. (Error bars represent standard deviation from mean).

parameters were evaluated in the bulk and biofilm phases using parallel bench-scale annular reactors (ARs) to compare a control system receiving 100% by volume of plant finished water with a blended system receiving 10% by volume of UF treated residuals. The results of this study present new information on the impacts of blending UF treated residuals with plant finished water from microbial regrowth (e.g. heterotrophic bacteria) and chemical fate in the distribution system (e.g. DOC, aluminum, manganese and iron) perspectives. However, additional work should be conducted to investigate potential impacts of this design on pathogen passage and DBP formation. Although this study involved only one test location with one set of water and waste residuals conditions, it provides a constructive framework for gaining a better understanding of waste residual management strategies involving low-pressure membrane filtration technology.

The significantly higher DOC concentrations measured in the UF permeate samples as compared to the plant finished water was of particular interest in this study. Specifically, the potential to augment biodegradable organic matter concentrations within the blended model distribution system (AR-blended) and impact heterotrophic bacteria growth trends through the introduction of the UF treated waste residual stream was evaluated through analysis of influent and effluent biological and chemical water quality parameters. The collective results of the field experiments demonstrated that blending 10% UF permeate with plant finished water did not result in significant proliferation of heterotrophic bacteria in the bulk (suspended) water or biofilm (attached) matrices as compared to a control model distribution system receiving 100% plant finished water (AR-filtered). Free chlorine and monochloramine disinfection resulted in significant log reductions of bulk and biofilm HPCs in both AR-filtered and AR-blended. At a lower Ct value, chlorine dioxide was more effective at reducing biofilm heterotrophic densities, although absolute HPC reductions in the suspended phase were less than for the other disinfectants.

During the control trial, heterotrophic bacteria densities in biofilm samples from AR-blended were found to be significantly lower than samples from AR-filtered. These results coincide with the formation of a thin, brown layer on the surface of the test coupons in the blended AR stream

and suggest that chemical oxidation of elevated manganese levels in the UF permeate had occurred after routine membrane cleaning procedures with sodium hypochlorite. However, the detection of manganese-oxidizing bacteria in biofilm samples and the presence of elevated manganese within the UF permeate stream suggests that inorganic post-precipitation occurrences are possible through biological reaction pathways, and would warrant further investigation in waste residuals studies.

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