Gray Water Recycle: Effect of Pretreatment Technologies on Low Pressure Reverse Osmosis Treatment

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Abstract

Gray water can be a valuable source of water when properly treated to reduce the risks associated with chemical and microbial contamination to acceptable levels for the intended reuse application. In this study, the treatment of gray water using low pressure reverse osmosis (RO) filtration after pre-filtration and aerobic biological filtration pretreatment was explored. Initially, nine commercial RO membranes were compared in dead end filtration tests for water flux and salt rejection at low pressures of operation (100-400 psi). Based on the dead end filtration results, one commercially-available membrane was subjected to further evaluation using a cross flow flat sheet test unit and gray water that had undergone various levels of pretreatment. The effects of gray water pretreatment with cartridge filtration, microfiltration, and aerobic biofiltration were compared in terms of membrane flux, chemical oxygen demand reduction, and salt rejection. Results from these RO filtration tests suggest that cartridge filtration in combination with RO treatment can achieve adequate reduction of gray water chemical oxygen demand (COD), but at the cost of progressively decreasing water flux through the RO membrane. Flux decline during cartridge filtration is mainly attributed to the presence of colloids $>0.2\ \mu\text{m}$ in size. In comparison, microfiltration maintains the water permeation capacity of the membrane, but permeate COD levels are higher. Biofiltered/microfiltered gray water shows RO permeate flux similar to microfiltered gray water and achieves permeate COD levels similar to cartridge filtered gray water.

Keywords: reverse osmosis, microfiltration, ultrafiltration, gray water recycle, biofilter, water reuse

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37 **Abbreviations:** reverse osmosis (RO), cartridge filtered (CF), microfiltered (MF), biofiltered
38 (BF), biological oxygen demand (BOD), chemical oxygen demand (COD), deionized (DI),
39 Environmental Protection Agency (EPA), American National Standards Institute (ANSI),
40 nanofiltrated (NF), Nephelometric Turbidity Unit (NTU), granular activated carbon (GAC),
41 volume concentration factor (VCF), Silt Density Index (SDI)

42 **Introduction**

43 Affordable technologies to allow reuse of gray water can significantly improve the water
44 supply of geographical regions that have limited freshwater resources. Energy efficient gray
45 water treatment technologies that can produce water suitable for non-potable reuse applications
46 are being explored. This study explores low pressure RO filtration of gray water using different
47 pretreatment measures to alleviate permeate flux decline associated with direct filtration of these
48 complex water matrices. Herein the low pressure RO filtration is defined as membrane systems
49 operating at or below 300 psi but capable of rejecting approximately 99% of sodium and
50 chloride ions at the design membrane flux.

51 The definition of gray water can vary but a common interpretation is domestic wastewater
52 that has not come into contact with toilet waste. Previous studies report that up to 75% of
53 household wastewater may consist of gray water, with the relative quantity and composition of
54 gray water differing based on cultural practices and water use patterns (Ghaitidak and Yadav,
55 2013). Depending on the source of the water, gray water composition can include soaps and
56 detergents, personal care products, oils and grease, fecal coliforms and other microbes, and
57 turbidity from food residue and clothing fibers (Ghaitidak and Yadav, 2013).

Gray water reuse generally requires treatment to decrease turbidity, organic and inorganic contaminants, and pathogenic microorganisms (Li et al., 2009). The treatment criterion varies based on the intended reuse application of the treated gray water. For example, the United States Environmental Protection Agency's (US EPA's) water reuse guideline for unrestricted non-potable reuse in impoundments, which allows human contact with the treated water, recommends achieving pH 6-9, biological oxygen demand (BOD) less than 10 mg/L, turbidity less than 2 NTUs, no detectable coliform counts per 100 mL, and maintaining 1 mg/L equivalent Cl₂ residual, in addition to meeting acceptable aesthetic levels of odor and color reduction (USEPA, 2012). In comparison, the guidelines for restricted use in impoundments, where human contact is limited, calls for less stringent controls on BOD, turbidity, and fecal coliform levels in the treated water. A combination of physical, chemical, and biological treatment options can be employed to meet the treatment levels desired for each type of reuse scenario (Li et al., 2009).

Physical filtration measures alone, such as microfiltration and ultrafiltration, can achieve adequate reduction of turbidity and pathogens, but poor removal of organics, makes the product water less microbiologically stable and leads to excessive formation of disinfection byproducts on addition of chemical disinfectants (Li et al., 2009). On the other hand, aerobic biological treatment combined with physical filtration has been shown to adequately reduce organic and nutrient loads in gray water (Leal et al., 2010, Li et al., 2009). Aerobic biological treatment eliminates much of the biologically oxidizable organics, making the product water more microbiologically stable and therefore more suitable for long term storage and distribution (Li et al., 2009).

Although a biological treatment/filtration sequence can achieve adequate reductions of COD/BOD values, the effluent may still contain personal care products and household chemicals

that are incompletely biodegraded (Leal, 2010). It is, therefore, advisable to implement a polishing step such as RO filtration to achieve more complete removal of hazardous chemicals from the gray water effluent stream. Previous water reclamation studies suggest that biological filtration treatment ahead of RO filtration reduces rates of RO flux decline (Chinu et al., 2009, Hu et al., 2005, van der Hoek et al., 1999).

In this study, physical and biological pretreatment measures to minimize membrane fouling during low pressure RO operation have been evaluated. To identify a candidate for low pressure RO treatment, a suite of commercial membranes were screened using dead end filtration studies to identify membranes that achieved the highest water flux and salt rejection at low pressure (100-400 psi) operation. While this list of membrane materials was not comprehensive, it generally represented a broad range of materials and sources for low pressure RO membrane treatment. After selecting a membrane based on dead end filtration studies, cross flow filtration experiments were conducted to establish baseline values for permeate flux at applied pressures, osmotic pressures, and cross flow velocities. Finally, using synthetic gray water, the effect of gray water pretreatment using pleated prefiltration, micro- and ultrafiltration, and biological filtration (biofiltration) was evaluated on RO membrane fouling, solute rejection, and product water recovery. Pleated prefiltration (10 μ m cutoff), herein referred to as ‘cartridge filtration’ and biologically-active GAC filtration, herein referred to as ‘biofiltration.’

The use of low pressure RO treatment for gray water reuse could enable a broader range of reuse activities than those commonly practiced at present (USEPA, 2012). For example, the military has an interest in the use of shower water recycling systems in remote arid environments (Army Public Health Center, 2013).

Materials and methods

2.1 Membranes

Table 1 summarizes the material composition of the RO and nanofiltration (NF) membranes utilized in this study. With the exception of the NanoH₂O and HRLE 440 membranes, all other membranes were purchased as pre-cut flat sheets for use in a Sepa CF cross flow unit (Sterlitech Corporation, Kent, WA, USA). The NanoH₂O (NanoH₂O, Coral Spring, FL, USA) and **DOW FILMTEC** HRLE 440 (DOW Chemical Company, USA) membranes were cut to size for the intended use (dead end filtration or cross flow filtration unit) from a larger flat sheet and spiral wound unit, respectively.

Table 1. Membranes and properties		
Filtration membranes	Product line	Composition
BW 30 FR	DOW RO	Polyamide
BW 30 LE	DOW RO	Polyamide
NF45	DOW FILMTEC NF	Polypiperazine amide
Polyamide AG	GE Osmonics	Polyamide
ACM 5	TriSep RO	Polyamide
XLE	DOW RO	Polyamide
NanoH ₂ O	NanoH ₂ O SWRO	Polyamide
HRLE 440	DOW FILMTEC RO	Polyamide
UTC-70 UB	Toray RO	Polyamide

2.2 Gray water

Simulated gray water was prepared in local tap water with initial pH of 8.8 ± 2 , alkalinity of 65 ± 10 , and monochloramine present at 1-2 mg/L as Cl₂. The simulated gray water used in this study was adapted from **the National Sanitation Foundation/American National Standards Institute** (NSF/ANSI) 350 standard (ANSI, 2011). The water was prepared to simulate a mixture of 67% shower water (20 g body wash, 2 g toothpaste, 1.33 g deodorant, 26.7 g shampoo/conditioner, 2 g lactic acid, 15.3 g liquid hand soap per 100 L) and 33% laundry water

(13.2 mL liquid laundry detergent, 7 mL liquid fabric softener, 1.33 g Na₂SO₄, 0.667 g NaHCO₃, 1.33 g Na₂HPO₄, 10 g test dust per 100 L). Water quality was consistent with NSF/ANSI guidance and a previously published study (Ward et al., 2015).

2.3 *Gray water pretreatment*

Simulated gray water was subjected to one or a combination of biological and prefiltration measures prior to RO filtration. Two prefilters were tested: a 10 µm polypropylene, single-open end cartridge filter (Cole-Parmer) and a 0.2 µm microfilter (MF, Fiberflo 200).

Biofiltration was conducted using a granular activated carbon (GAC) based aerobic biological filter (BF) initially seeded with wastewater to establish an active bacterial community. The biofilter configuration consisted of three parallel biological filters operated at 4.15 L per minute/m² in 4 h on, 8 h off cycles, the off period being the biological regeneration time for the filters. The average influent COD was 374 mg/L and the average effluent COD ranged from 124-151 mg/L. All biofiltered gray water samples were microfiltered prior to RO filtration, hence these feed waters are represented by the notation BF/MF. The age of the BF during RO testing of BF/MF-treated gray water feed was 3, 5, and 5 weeks, for BF/MF replicates 1, 2, and 3, respectively.

The conductivity and COD values for the pretreated gray water used in RO cross flow experiments (in triplicate) are summarized in Table 2.

Table 2. Properties of cross flow filtration feed waters					
RO feed		Conductivity (μS/cm)	COD (mg/L)		Appearance
Cartridge filtered (CF)	1	285	271		Turbid (~25 mg/L total suspended solids), foamy
	2	288	a		
	3	316	293		
Microfiltered (MF)	1	306	194		Clear, negligible turbidity, foamy
	2	292	228		
	3	303	220		
Biofiltered/microfiltered (BF/MF)	1	268 ^c	163 ^b	185 ^c	Clear, no foam
	2	246 ^c	188 ^b	104 ^c	
	3	283 ^c	192 ^b	125 ^c	

^a Analysis error.

^b COD values of biofiltered gray water prior to microfiltration.

^c Conductivity and COD measurements for the BF/MF recycled feed water were taken post-RO filtration experiment.

Tests using chlorine test strips (Hach Company, Loveland, CO)) indicated that the chlorine (free and combined) concentrations of all feed waters were below 0.5 ppm at the time of RO filtration testing (Molk et al., 2013).

2.4 Dead end filtration

Dead end filtration studies using each of the nine commercial RO membranes were conducted using a Sterlitech HP4750 (Sterlitech Corporation, Kent, WA, USA) stirred dead end filtration unit loaded with 300-mL of deionized (DI) water or 50 ppm NaCl. The experiment was initiated by pressurizing the system to desired feed pressure using a compressed nitrogen feed. The permeate flow was calculated by recording the volume of permeate expressed per unit time. NaCl rejection of the system was tracked by measuring the conductivity of the permeate

solutions and comparing against NaCl standards. Permeate flux and solute rejection were tracked as a function of time and volume concentration factor ($VCF = \text{Initial feed volume} / \text{retentate volume}$).

2.5 Cross flow filtration

Cross flow filtration studies were conducted using a Sepa CF (Sterlitech Corp. Kent, WA, USA) filtration unit. The cell was outfitted with 65 mil spacer on the feed side and 17 mil permeate distributor on the permeate side, separated by a custom cut section of the HRLE 440 RO membrane obtained from a spiral wound unit. A pressure gauge on the feed inlet side was used to monitor feed pressure inside the cell. The transmembrane pressure in this system is expected to be approximately equal to the inlet feed pressure as little pressure drop is expected between the concentrate inlet and outlet lines of this unit. A flow restriction needle valve on the feed outlet was used to adjust the transmembrane pressure. Increasing flow restriction resulted in higher feed pressure and consequently higher water flux through the membrane. All experiments using the cross flow filtration system were conducted in a batch recirculating mode such that the permeate was redirected to the feed tank, with the exception of small volumes removed for COD testing (for gray water experiments only).

During the cross flow RO experiments with DI water and NaCl solutions (Section 3.2.1), the pressure was manually adjusted to the desired system pressure using the outlet restriction valve. Cross flow velocities ranging from 0.09 to 0.14 m/s were tested by varying pump frequency to test the effect of cross flow velocity on water flux and salt rejection. Permeate flux and NaCl rejection (based on conductivity readings) were monitored in these experiments.

Gray water RO filtration and pretreatment efficacy (Section 3.2.2) were tested at a constant inlet flow velocity producing a cross flow velocity of approximately 0.14 m/s within the cell. This cross flow velocity was chosen based on the manufacturer's recommended feed flow rates for the HRLE 440 spiral wound units. In the gray water RO experiments, the system feed pressure was adjusted to 100 psi only at the beginning of the experiment; subsequent increases in pressure occurred naturally, presumably due to increasing resistance to water flow through the RO membrane as a result of progressive fouling by gray water constituents. The experiments were terminated when the pressure exceeded 400 psi or by the end of 2 h. Each of the three pretreated feed waters, CF, MF, and BF/MF, were tested in triplicate using fresh feed (initial conductivity and COD values presented in Table 2). Prior to each gray water experiment, the membrane was hydrated/conditioned by maintaining DI water feed at 0.09 m/s at 100 psi pressure for 30 min. The experimental variables tracked during pretreated gray water filtration experiments were concentrate inlet pressure, permeate flux, COD, and conductivity. Detailed results for gray water RO experiments are provided in the Supporting Information.

2.6 Chemicals and analyses

With the exception of simulated gray water, all other experimental solutions were prepared using DI water. NaCl (>99%) was purchased from Acros Organics (Morris Plains, NJ, USA). Chemical oxygen demand (COD) low range (3-150 mg/L) and high range (150-3000 mg/L) test kits were obtained from Hach Company (Loveland, CO, USA).

The conductivity of permeate was measured against the conductivity of the feed solution to estimate rejection of ionic solutes at each condition. For gray water experiments, a small portion (2 mL per test) of the initial feed and resulting permeate was used to test the COD rejection at various conditions. COD testing was carried out following the USEPA-approved reactor

digestion method 8000 (Hach, 2009). The BOD does not capture all of the organic matter in a given sample. Current EPA guidance for unrestricted reuse generally covers non-potable applications, but not near-potable or potable reuse applications. This study is focused on high tier gray water reuse, which is more similar to Direct Potable Reuse, in which both biodegradable and non-biodegradable organic matter are of potential concern. Thus COD is a more appropriate parameter than BOD as it is also used commonly with the industry and sufficient for this study.

Results and Discussion

3.1 Membrane down-selection through dead end filtration studies

Experiments were first performed at small scale to down-select a membrane material for testing with gray water. The relative down-selection criteria to be maximized were water flux and sodium chloride rejection. Figure 1 shows water flux through each of the nine (9) flat sheet test membranes as measured in dead end filtration of DI water at different feed pressures as a function of VCF. It should be noted that the comparative results reported herein were derived from a limited set of filtration conditions that may not represent the optimal performance range for all membranes that the manufacturer recommends.

At target operational pressures for the low pressure gray water RO system (100-400 psi), four RO membranes, XLE, UTC-70 UB, ACM 5, and HRLE 440, achieved similar levels of DI water flux, scaling from 0.1 to 0.45 L/m².min. Surprisingly, the nanofiltration membrane, NF45, showed lower or similar rates of permeate fluxes for the same pressures. For some of the membranes, permeate flux declined slightly with increasing volume concentration.

Figure 2 shows the effect of increased osmotic pressure on the water permeability of the membranes during filtration of 50 ppm NaCl solution. Of the four membranes identified as having similar permeate fluxes for DI water, permeate flux was decreased the most for the UTC - 70 UB membrane upon introducing NaCl feed water. XLE and ACM 5 also exhibited relative suppression of permeate flux values, particularly at higher feed pressures, whereas HRLE 440 exhibited decreased permeate flow at low pressure (200 psi), but with permeate flows comparable to DI water feed at higher feed pressures (300, 400 psi).

The rejection of NaCl during filtration of 50 ppm NaCl feed solution is shown in Figure 3. (same experiments for which water flux is reported in Figure 2). Only the GE polyamide and the HRLE 440 membrane consistently achieved better than 80% salt rejection at feed pressures between 200 and 400 psi. The HRLE 440 membrane maintained better than 90% salt rejection under most of the conditions tested. For most of the membranes, salt rejection appeared to increase with increasing VCF. As the feed concentration increased due to volume reduction of the feed, the percentage of this salt concentration diffusing into the permeate decreased, offsetting any decrease in rejection we may expect from the increased osmotic pressure of the feed at higher VCF.

Figure 1. DI water flux in dead end filtration mode for nine test membranes.

Figure 2. Water flux as a function of volume concentration factor (VCF) during dead end filtration of 50 ppm NaCl for nine test membranes

Figure 3. NaCl rejection during dead end filtration of 50 ppm NaCl for nine test membranes (NaCl rejection = Fraction of NaCl Removed)

Based on the relatively higher salt rejection and permeate flux of the HRLE 440 membrane within the 100-400 psi pressure range in the dead end filtration experiments, this membrane was selected for cross flow filtrations studies using higher salinity water and gray water reported in Section 3.2.

3.2 Cross flow RO filtration

3.2.1 DI water and NaCl

Prior to conducting gray water RO filtration experiments, baseline values for water flux and NaCl rejection of the HRLE 440 membrane were assessed in a cross flow filtration system and compared to values reported by the manufacturer.

Figure 4. Permeate flux as function of transmembrane pressure for DI water (closed symbols) and 2000 ppm NaCl (open symbols) feed at different cross flow velocities (A). NaCl rejection as a function of transmembrane pressure at different cross flow velocities (B) for the HRLE 440 membrane.

Figure 4 shows the results of cross flow filtration experiments using a flat sheet HRLE 440 membrane with DI water and NaCl feed. Permeate flux increased linearly with applied pressure in DI water experiments (Figure 4A), consistent with widely-accepted theory that permeate flux

is directly proportional to net applied pressure. When 2000 ppm NaCl was introduced into the feed water, permeate flux decreased, particularly at the higher pressures. Some reduction in permeate flux can be expected based on the higher osmotic pressure of the bulk solution which decreases the driving force for water permeation across the membrane. Additionally, concentration polarization intensifies at higher permeate fluxes (at higher pressures), presenting further resistance to water flux at higher pressures.

Cross flow velocity did not have an effect on water permeation in any of the experiments, including the NaCl feed experiments. These data suggest that this range of cross flow velocities did not achieve differences in salt accumulation at the boundary layer significant enough to affect the overall driving force for water permeation. Based on data interpolation (assuming linear trend), permeate flux obtained in this study for the 2000 ppm NaCl feed was about 30% lower than the permeate flux reported by the manufacturer for similar conditions (0.81 L/m².min at 150 psi for 2000 ppm NaCl feed) (DOW). The cross flow velocity used by the manufacturer was not reported. It is possible that the higher permeate flux observed by the manufacturer corresponds to a higher cross flow velocity testing condition or differences in velocity profiles between spiral wound units and the flat sheet RO setup used in this study.

Greater than 95% salt rejection was achieved under all pressure and cross flow velocity conditions tested for the HRLE 440 membrane, as shown in Figure 4B. In general, salt rejection improved at higher applied pressures. Improved rejection values at higher pressures likely result from greater dilution of salt in the permeate water due to higher permeate flows, offsetting any increase in salt permeation at these same pressures due to greater concentration polarization. Increasing cross flow velocity also resulted in some improvement in salt rejection. This effect may be rationalized on the basis of a thinner boundary layer formed at higher cross flow

velocities, which encourages faster back diffusion of salts concentrated at the membrane surface due to concentration polarization (Cheryan, 1998). The highest salt rejection observed in this study was 98.2% at 400 psi at the highest cross flow velocity setting.

Cross flow filtration experiments using the HRLE 440 membrane indicated that membrane flux and rejection values obtained during this study were similar to the manufacturer's reported values. The baseline numbers for water flux and rejection obtained in this section are used to compare system performance during gray water RO filtration experiments using the HRLE 440 membrane.

3.2.2 Pretreated gray water

RO filtration of gray water was assessed after pretreatment using cartridge filtration (CF), microfiltration (MF), and sequential biofiltration/microfiltration (BF/MF). Membrane flux and pressure were monitored over time at a fixed feed flow rate. Pressure was set to 100 psi at the beginning of each experiment and allowed to increase as flow resistance built up inside the membrane vessel.

Figure 5. Change in pressure over time during RO cross flow filtration of cartridge filtered (A), microfiltered (B), and biofiltered/microfiltered (C) gray water through the HRLE 440 membrane at a cross flow velocity of 0.14 m/s.

Figure 5 shows the change in feed pressure within the cross flow filtration unit as a function of experiment time for the three different pretreated gray water feeds. The triplicate experiments for each pretreated feed water condition showed considerable variability in the time scales over which pressure increased in the RO cross flow filtration system, but all show a trend towards increased system pressure.

The system pressure is a very sensitive measure of the resistance of the membrane to water permeation. Any increase in membrane (and the fouling layer) resistance results in an observable pressure increase within the filtration cell as the concentrate inlet flow is fixed and outlet flow is restricted. As the system pressure increases, compaction of a pre-formed fouling layer can pose additional resistance to water flow through the fouling layer (Faibish et al., 1998, Potts et al., 1981), feeding into the trend towards increasing system pressure. The mechanism(s) due to which resistance to water flux builds up on the membrane is likely different for each type of pretreated gray water and based on its composition discussed further in Section 3.3).

The CF-treated waters showed a general trend towards an initial rapid pressure increase to ~300 psi followed by a second stage of slower pressure increase. Also, in general, CF-treated gray water maintained systems pressures less than 400 psi longer than the MF- and BF/MF-treated water. MF and BF/MF did not show any clear advantages over CF from the perspective of stabilizing system pressure changes. An exception to this was RO filtration of BF/MF #1, which exhibited the slowest rate of pressure increase of all the pretreated gray water feeds, not exceeding 250 psi during the duration of the experiment.

Figure 6. Permeate flux as a function of feed pressure during cross flow filtration experiments with pre-treated gray waters. Scatter plots for each pretreatment condition combine data points obtained from triplicate experiments shown in Figure 5.

While there is considerable variability in the time scales over which pressure increases were observed for the same pretreated gray water feeds, Figure 6 shows that all three treatment conditions initially achieved similar permeate fluxes that increased linearly with pressure. MF and BF/MF feed waters achieved very similar responses in RO water flux with increasing system pressure over the entire pressure range (100-400 psi) considered. Water flux rates for MF and BF/MF water are in the range of values observed for 2000 ppm NaCl (Figure 4, .2-1.5 L/m²/min between 100 and 400 psi), but lower than permeate flux recorded for cross flow filtration of DI water through the same membrane. Although it is not clear how the osmotic pressure of the pretreated gray water compares with that of 2000 ppm NaCl, these results suggest minimal impact on the membrane's original flux rates upon exposure to these pretreated gray waters.

In contrast, CF waters showed an initial linear increase with increasing pressure and subsequent deviation (at approximately 280 psi, corresponding to 0.8 L/m²/min flux) to decreasing permeate fluxes with further pressure increases. These data indicate that although all of the feed waters resulted in increased resistance to water flow through the RO membrane, only the CF-treated waters significantly impacted the water flux through the RO membrane at higher pressure. The pressure range over which flux decline is observed in CF-treated gray water coincides with the stage of slower pressure increase (above 300 psi) seen in 5A. It should be noted that the manufacturer recommends a silt density index (SDI) of five or less for waters treated with this particular membrane. The CF-treated gray waters may have exceeded this criterion.

Figure 7. Average RO permeate COD (left-axis) and conductivity (right-axis) for each of the pretreated gray waters. Error bars represent 95% confidence intervals.

Figure 7 shows average permeate COD and conductivity during RO filtration of the three different types of pretreated gray water. The average COD and conductivity values for RO effluents were lower for BF/MF waters in comparison to MF-only treated gray water. Surprisingly, CF/RO gray water effluent also achieved similar COD values as BF/MF/RO permeate. The conductivity values for each pretreatment process are very similar.

3.3 Discussion

The results presented in this study provide practical information about RO membrane performance for the treatment of gray water after various levels of pretreatment. Three gray water pretreatment methods were assessed: cartridge filtration (CF), microfiltration (MF), and biofiltration plus microfiltration (BF/MF). The primary goal of this investigation was to develop an integrated portable/deployable graywater filtration system with various components (cartridge filtration, biofiltration, GAC filtration, microfiltration and RO filtration system) which can be operated based on the feed water analysis and filtrate requirements. Based on the data generated, the pretreatment regimes resulted in different levels of organics removal. The highest COD removal levels for combined pretreatment and RO ranged from 90-96% and were achieved with BF/MF or CF pretreatments. MF alone plus RO resulted in lower levels of COD removal, highlighting that RO is not an absolute barrier to organic contaminants. The additive effect of biofiltration and RO on COD reduction indicates that GAC adsorption and biodegradation targets organics that might permeate through an RO membrane. For the CF studies, higher COD removal levels may be due to a distinct difference in fouling layer that is less permeable to hydrophobic contaminants than that formed when MF is used for pretreatment. In all of the gray water tests, conductivity values in the effluent were consistent, suggestive of high levels of desalination during RO treatment of gray water.

Trends observed for RO membrane flux in this study, in addition to observations from previous studies on membrane fouling, may provide insight into fouling mechanisms during gray water treatment. Previous studies suggest that dissolved organic matter and colloidal matter can lead to significant membrane fouling and flux reduction (Potts et al., 1981). Larger particulates ($>5\ \mu\text{m}$), such as test dust in the simulated gray water, are likely to be retained in the bulk fluid due to shear forces (Winfield, 1979) and are therefore unlikely to contribute to membrane fouling. In contrast, the deposition of smaller particulates and colloidal matter has been shown to be a function of permeate flux, a critical flux being necessary to deposit and retain these components on the membrane surface (Faibish et al., 1998, Zhu and Elimelech, 1997). Permeate flux decline due to colloid deposition also depends on the size of the colloids, smaller colloids forming less permeable cake layers resulting in greater flux declines (Faibish et al., 1998, Park et al., 2008). Kumar et al. (2006) demonstrated that pre-filtration of colloids in the $0.1 - 1\ \mu\text{m}$ range can significantly decrease RO membrane flux decline during seawater desalination. The interaction of dissolved organic matter and salts with deposited colloidal matter has also been shown to play an important role in increasing osmotic pressure within the fouling layer (Winfield, 1979). Reduced back diffusion of salts and dissolved organic compounds trapped within the stagnant fouling layer can exacerbate concentration polarization at the membrane surface, reducing the net driving force for water passage through the membrane (Hoek and Elimelech, 2003).

The effect of surfactants on membrane permeability and solute rejection can be a complex one, dependent on the chemical and charging properties of the membrane and that of the surfactant (Childress and Elimelech, 1996). Anionic surfactants (such as sodium dodecyl sulfate) have been shown to decrease water flux and increase organic rejection in thin film

composite polyamide membranes such as the one used in these studies (Childress and Deshmukh, 1998). On the other hand, surfactants (such as those used in membrane cleaning) can also reduce deposition of organic compounds by micelle formation and increase shearing of the cake layer from the membrane surface.

Of the three types of pre-treated gray water evaluated in this study, the CF-treated water had the highest initial COD, conductivity, turbidity, and surfactant loading. As a result, the pressure and flux trends observed during RO filtration of CF-treated water may be ascribed to a combination of mechanisms, including adsorption of dissolved organics, salt deposition, cake formation by deposition of colloidal matter, cake enhanced concentration polarization of salts and organic matter at the membrane surface, and effect of surfactants on membrane permeability and organic deposition.

Although pressure continued to rise in the CF-treated gray water experiments, no decrease in permeate flux is observed until a critical flux ($\sim 0.8 \text{ L/m}^2/\text{min}$) is reached, beyond which water flux begins to decline. Considering the rapid time scale over which this flux decline occurs (within a few minutes of critical flux being reached) and the relative consistency at which it occurs during CF-treated gray water filtration, the flux decline probably arises mainly from colloid deposition and cake enhanced concentration polarization upon formation of the colloidal cake layer. The high initial COD of these waters combined with the high COD rejection makes it possible for a very high osmotic pressure to build up at the membrane surface. An interesting feature of the CF-treated gray water experiments is the relatively slow rise in system pressure (Figure 5) after the onset of permeate flux decline is observed (above 280 psi, Figure 6). It is plausible that the decrease in permeate flux decreases the rate at which additional colloids/organics are deposited into the fouling layer, slowing the rate of fouling.

MF pretreatment, which sieved for particulates and colloids greater than $> 0.2 \mu\text{m}$, eliminated the permeate flux decline observed with CF-treated water. In light of these data, the flux decline observed for CF-treated water can be mainly attributed to membrane fouling due to colloidal aggregates in the size range above $0.2 \mu\text{m}$ and associated interactions of other gray water constituents with these colloidal deposits. This finding is not entirely surprising, considering one of the most reliable predictors of the RO membrane fouling potential is the SDI, which measures the rate of plugging of a $0.45 \mu\text{m}$ filter (Potts et al., 1981). The MF-treated gray water, however, also shows an increase in RO system feed pressure at constant feed flow, indicating that the surface of the membrane builds up resistance due to adsorption or deposition of other gray water constituents, such as dissolved organic matter. Over longer filtration times, these same constituents may result in progressive RO flux decline.

BF/MF treatment did not result in improvements in membrane flux or in slowing the rates of feed pressure increase relative to MF treatment only. Further studies are needed to look at membrane flux over longer operating conditions at constant pressure, rather than the constant feed flow conditions tested herein. Specifically, the tradeoff between reducing the total organic load being introduced to the RO feed versus factors such as increased extracellular polymeric substances introduced during biological treatment needs to be evaluated.

No clear effect of surfactants on altering or maintaining water flux can be surmised from these experiments. Both CF- and MF-treated gray water are expected to retain a significant quantity of surfactants (as evidenced by their foamy appearance), but CF-treated water exhibited progressive flux decline at higher permeate fluxes while MF-treated water did not. Furthermore, BF/MF-treated water, in which the surfactants appeared to have been digested, showed similar permeate flux to surfactant-containing MF-treated gray water.

This paper highlighted some of the complexity encountered when treating gray water using RO membranes. While potential benefits of pre-treatment technologies applied upstream of RO membrane treatment, further studies of long term performance, fouling, and optimal cleaning processes are needed. Additionally, because the product water from RO treatment could be considered for higher tier reuse applications like showering or even human consumption when accompanied by advanced oxidation processes, the fate of regulated and emerging micropollutants should also be studied. Such micropollutants may include disinfection byproducts (DBPs), personal care products, hormones, and pharmaceuticals.

Conclusions

This study compares the effects of gray water pretreatment by cartridge filtration, microfiltration, and biofiltration on RO membrane performance. Major findings from this study are as follows:

- Colloidal constituents of gray water in the size range $> 0.2 \mu\text{m}$ contribute to rapid flux decline during RO filtration of cartridge filtered gray water.
- In RO filtration of CF-treated gray water, permeate flux decline due to colloidal fouling requires a critical permeate flux, below which no adverse effects on permeate flux are observed.
- Microfiltration of gray water to remove colloids in the $> 0.2\mu\text{m}$ size range eliminates rapid flux decline observed above the critical flux value.

- No positive or negative effect of biofiltration is observed over microfiltration alone in terms of water flux, but biofiltration improves reduction of gray water COD both in the RO concentrate and resultant RO permeate.
- Cartridge filtration alone achieves COD reduction in the RO permeate similar to biofiltration/microfiltration. The fouling layer formed in the presence of gray water colloidal matter may improve rejection of organic compounds.

Life cycle cost analyses should also be performed to determine the economic viability of high tier gray water reuse compared to current gray water reuse practices. Potential scenarios where high tier gray water reuse may be viable include remote environments (such as military or emergency response operations) or even building scale systems for dormitories or other facilities with high gray water generation intensity. For the latter, the coupling of wastewater heat recovery technology may be required to achieve economic viability. In all cases, careful consideration of relative health risks and end-user acceptance would be essential.

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