



## Bench-scale assessment of membrane pre-treatment and seasonal fouling potential variations

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### ABSTRACT

Fouling is widely recognized as an important challenge to the widespread use of membrane filtration technologies in water and wastewater treatment. Unfortunately fouling by natural waters is complex and its mechanisms are not presently well understood. Pre-treatment of feed water is part of a successful fouling control strategy. The aim of this study was to gain a better understanding of membrane fouling by characterizing two natural waters seasonally using the modified fouling index-ultrafiltration (MFI-UF) and other various traditional water quality parameters (turbidity, NOM, UV<sub>254</sub>, etc.). In addition, fouling prevention by automatic backwash filtration was studied at a bench scale using samples of filter screens and thread filters. Apparent relationships between typical water parameters and fouling potential were explored through partial least square regression and pre-treatment performance was evaluated via particle counts and MFI-UF measurements. Results showed that it is not feasible to identify foulants using traditional water quality parameters as they lack the precision to specifically describe the actual foulants and also pre-treatment that only removes large particles (>2µm) only is ineffective at reducing fouling potential.

**Keywords:** Membrane; Fouling; Pre-treatment; MFI-UF; Particles

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### 1. Introduction

Membrane filtration is a versatile solution for many of the water quality issues that drinking water producers are faced with today. Nanofiltration (NF) and ultrafiltration (UF) membranes can be used for a variety of applications including the removal of natural organic matter (NOM) [1-4], cyanotoxins [5-7], pathogenic organisms [8,9] and endocrine disrupting

compounds (EDC) [10]. Membrane systems are particularly interesting for smaller systems due to their small foot-print, high level of automation and relative independence from chemical additives. However, the most critical challenge to a more widespread application of membrane technology remains membrane fouling [8].

Understanding membrane fouling is necessary for the advancement of membrane technologies and numerous studies have been conducted to gain a greater knowledge of the physical and chemical factors

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controlling fouling of NF and UF membranes [11–18]. However, many of these studies have focused on observing individual fouling mechanisms either by using synthetic waters [17–19] or by fractioning natural waters into different components [12,13,16] before studying each component individually. This does not provide a comprehensive picture of fouling since there is often an overlap between the fouling types [20] and since synthetic waters do not necessarily behave the same way natural waters do [19,21].

Estimation of the fouling potential of feed water based on traditional water quality parameters has not been successful [22–24]. Fouling indexes have been developed to meet the need for a fast and accurate measurement of fouling. Of the many fouling indexes proposed in the literature [25–29] the modified fouling index-ultrafiltration (MFI-UF) [30] was found to be the most interesting option. This index was developed based on cake filtration, which is thought to be an important fouling mechanism for NF and UF membranes [31].

Measuring fouling potential is essential to gauging the performance of physical and/or chemical pre-treatments which are integral parts of a complete fouling control strategy. While there are a wide variety of potential pre-treatment options, the schemes most often proposed in the literature to control fouling in NF and UF membrane systems [32–36] are methods that require either continuous chemical addition, construction of important infrastructure or additional low pressure membrane systems upstream. These constraints are often prohibitive for smaller systems. A treatment technique which does not have these constraints is automatic backwash filtration. Successful operation of NF systems that implemented this pre-treatment technology has been reported [37,38] but results did not elucidate the actual improvements made by introducing the pre-treatments. There has also been mention of the use of this type of technology upstream of UF membranes but with a less than stellar performance [39].

The present study aims at gaining a better understanding of fouling by natural surface waters and its prevention through physical pre-treatment in order to facilitate the use of spiral wound high pressure membrane filtration systems in small communities. MFI-UF measurements and various traditional water quality parameters were used to describe the fouling potential of raw natural waters over an 8-month period. Meanwhile, laboratory filtrations using samples of filters screens and thread filters were used to simulate the performance of automatic backwash filter technology. Industrial filter samples were used rather than laboratory grade filter discs which are more

commonly used in bench-scale experiments. This was done to gain a better idea of the performance of full scale filters. Pre-treatment performance was evaluated via particle counts and MFI-UF measurements.

## 2. Materials and methods

### 2.1. Feedwaters

Two natural surface waters were used in this study. Analyses of water quality variations were carried out on samples taken from the raw water intakes of the drinking water treatment plants of the city of Laval (source water Des Prairies River) and the city of Lachute (source water Barron Lake) from February to October 2007. Both locations are situated in the Province of Québec, Canada. Only the more heavily particle charged Des Prairies River feed water was used in the pre-treatment analysis section as it represented the greater challenge in terms of particle loading. The samples were stored in a dark cold room (4 °C) on arrival and were kept there until the experiments were performed; samples were conserved for a maximum of 24 days.

### 2.2. MFI-UF measurements

#### 2.2.1. MFI-UF theory

As mentioned previously, the MFI-UF is a fouling index which is used to measure fouling potential based on the cake filtration theory. This theory states that the increase in hydraulic resistance is caused by the formation of a cake layer formed by particles retained on the external membrane surface. This increase in resistance is proportional to the cumulated volume of filtered water,  $V$  [40]. Thus the MFI-UF is a function of the concentration and nature of the particles in the feed water. The MFI-UF is measured at a constant pressure gradient through a UF membrane and it corresponds to the slope of the linear portion of the  $t/V$  vs  $V$  curve (see Eq. (1)), where  $t$  is the filtration time and  $V$  is the volume of permeate produced. Measurements were taken at standard conditions; room temperature (20 °C) and 2.1 bar 30 psi [30]. If the actual conditions departed slightly from these reference conditions, the MFI-UF values were standardized using Eq. (2).

$$\frac{t}{V} = \frac{\eta R_m}{\Delta P A_m} + \left( \frac{\eta C_b \alpha}{2 \Delta P A_m^2} \right) V = \frac{\eta R_m}{\Delta P A_m} + (\text{MFI-UF}) V \quad (1)$$

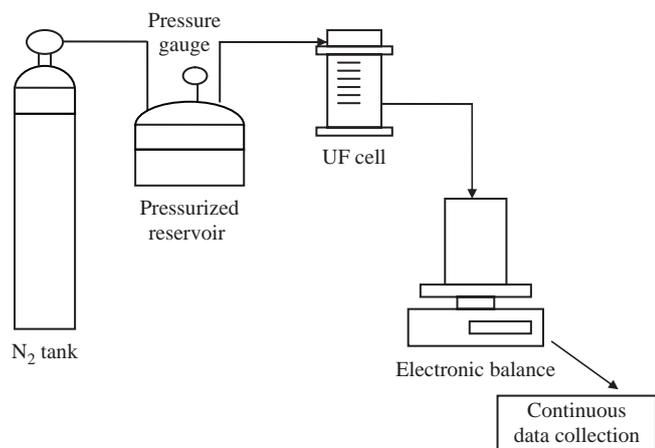


Fig. 1. MFI-UF measurement set-up.

$$MFI - UF_{std} = \frac{\eta_{20}}{\eta_T} \left( \frac{\Delta P}{206.84 \text{ kPa}} \right) MFI - UF_{measured} \quad (2)$$

where  $t$  is the filtration time (s);  $V$  is the permeate produced ( $\text{m}^3$ );  $\Delta P$  is the transmembrane pressure ( $\text{N m}^{-2}$ );  $\eta$  is the dynamic viscosity of water ( $\text{N s m}^{-2}$ );  $A_m$  is the membrane area ( $\text{m}^2$ );  $R_m$  is the membrane resistance ( $\text{m}^{-1}$ );  $\alpha$  is the specific cake resistance ( $\text{m kg}^{-1}$ ); and  $C_b$  is the particle concentration ( $\text{kg m}^{-3}$ ).

### 2.2.2. Set-up

The MFI-UF was measured using the bench-scale set-up presented in Fig. 1. The feed water samples were placed in a stainless steel reservoir and maintained under constant pressure by means of a nitrogen gas tank whose pressure was controlled by a pressure regulator (Tescom) with an accuracy of  $\pm 0.5$  psi. The pressure was monitored with a digital pressure gauge (Ashcroft) with a full scale of 60 psi, a resolution of  $\pm 0.01$  psi and an accuracy of  $\pm 0.3$  psi. The membrane filtrations were conducted in unstirred dead-end filtration cells with an internal volume of 50 mL (Amicon-Millipore Corporation). Permeate flux was calculated from cumulative gravimetric mass measurements which were continuously recorded using an electronic balance (Sciencetech) with a full scale of 3000 g, a readability of 0.01 g and a repeatability of 0.01 g. Data were collected automatically every 5 min. Experiments were carried out at room temperature ( $20 \pm 3$  °C) and at 2 bar ( $30 \pm 1$  psi), temperature and pressure were noted at the beginning and the end of every experiment. Pressure was adjusted at the beginning of the experiment and was not significantly altered during the rest of the experiment.

### 2.2.3. Membrane preparation

The membrane used for all MFI-UF measurements was a flatsheet polyacrylonitrile (PAN) membrane with a MWCO of approximately 10 kDa (Sepro Membranes, USA). This membrane was chosen as it was very similar to the reference membrane recommended for MFI-UF measurement by Boerlage et al. [31]. A new membrane coupon was used for every experiment in order to avoid the effects of irreversible fouling. Conditioning has been shown to produce more homogenous fluxes in membranes with values closer to those quoted by manufacturers [41]. Therefore, membrane coupons were soaked in laboratory grade water (Milli-Q) for a minimum of 12 h previous to the experiments to remove any preservation agents. Membranes were then stabilised by pressurized filtration first at 45 psi for 15 min and then at 30 psi until stable clean water flux was attained (between 3 and 4 h). In order to insure a certain homogeneity, coupons were rejected if they had an initial flux which varied by more than 20% from the pure water permeability quoted by the manufacturer ( $70 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ ) or if they failed to attain a stable clean water flux (flux deviation of less than 3%) during the last 1.5 h of filtration. Typically this second criteria was verified after 3 h and if it had not been attained, the filtration was allowed to continue for up to 4 h, after which if the criteria had still not been attained, the coupon was rejected.

### 2.3. Bench-scale pre-treatment

In order to develop an adequate filtration protocol which was representative of particle removal by full scale automatic backwash filters at a laboratory scale, samples of filters screens and textile filters employed by Amiad Filtration Systems ([www.amiadusa.com](http://www.amiadusa.com)) in full scale installations were used. The automatic backwash filter options proposed by this company were interesting since they included a thread filter technology with filtration degrees that went down to  $2 \mu\text{m}$ , which was by far the finest particle removal available found for self-backwashing filters. In addition to this, an article published by Nemirovsky [37] described the successful use of this technology upstream of NF membranes. Even though laboratory filters have been previously used in bench-scale studies to model the behaviour of different treatment technologies [12,42], it is highly unlikely that these filters will perform in the same manner as filters produced for large scale applications.

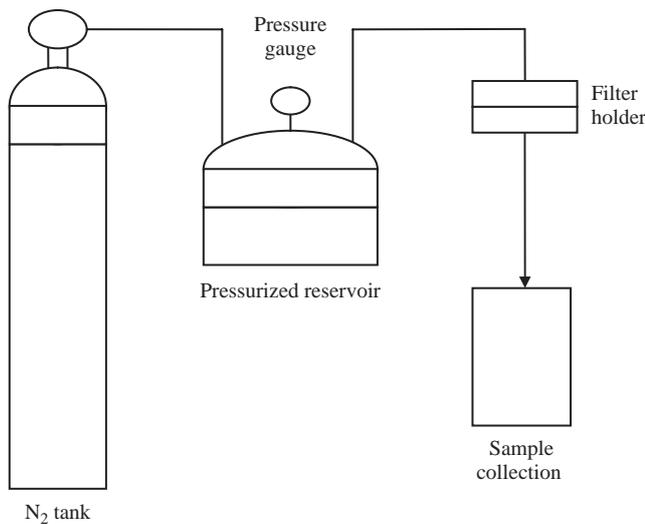


Fig. 2. Screen filtration set-up.

### 2.3.1. Pre-filtration – role of filter porosity

Four pre-filters of different porosities and different technologies were used in the study, 25 and 10  $\mu\text{m}$  stainless steel screen filters and 3 and 2  $\mu\text{m}$  thread filter cassettes. The 10 and 25  $\mu\text{m}$  stainless steel screens were cut into 40 mm diameter coupons and placed in the pre-filter housing of a stainless steel filter holder (Millipore Corporation) in order to filter the sample at 22 psi (minimum operating pressure for the full scale filter) (Fig. 2). Due to the high flux through the screens and the small volume of filtrate necessary ( $\sim 2$  L), the duration of the filtration run was very short (a matter of seconds) and it was not practical to record the variation of flux throughout the run. The first liter that was filtered through the screen was discarded and the second liter was collected for MFI-UF measurements and particle counting. The 2 and 3  $\mu\text{m}$  cassette filters, which are made of high strength polyester thread wound around a plastic cassette (active filtration area of  $102\text{ cm}^2$ ), were first soaked for 10 min in the water sample that was to be filtered and filtrations were then done at a constant flowrate of  $500\text{ mL/min}$  ( $2.95\text{ m}^3\text{ m}^{-2}\text{ h}^{-1}$ ) as suggested by the manufacturer. The initial and final differential pressures were 1.4 and 2.4 psi, respectively for the 2  $\mu\text{m}$  cassette and 0.5 and 1.3 psi for the 3  $\mu\text{m}$  cassette. The pressure of the influent was measured and the outflow was at atmospheric pressure (Fig. 3). As done previously, the first liter filtered was discarded and the second liter was collected for MFI-UF measurements and particle counting.

### 2.3.2. Pre-filtration – role of cake formation

An experiment with a 2  $\mu\text{m}$  cassette filter was done to investigate the role that cake formation plays on

pre-filter performance in terms of particle removal and MFI-UF reduction. The filter cassette was soaked for 10 min in the raw sample and the filtration was done at a constant flowrate of  $500\text{ mL/min}$  ( $2.95\text{ m}^3\text{ m}^{-2}\text{ h}^{-1}$ ). The pressure differential across the cassette was initially 0.6 psi and by the time a cumulative volume of 12 L had been filtered, it had surpassed 15 psi (the full scale on the pressure gage on the set-up). Samples of filtrate were collected at different specific volumes (volume filtered/cassette filter area) during the filtration run; they were collected at 0.2, 0.7 and  $1.2\text{ m}^3/\text{m}^2$  for particle counts and MFI-UF measurements.

### 2.4. Additional water quality analysis

Several analytical methods were used to measure traditional water quality parameters to complement the information on fouling potential. Temperature and pH (AB15 Accumet basic, Fisher Scientific) were measured but were not included in the data analysis as they did not vary significantly for either source. Ultraviolet absorbance at 254 nm (UVA), color (Cary 100, Varian) and total organic carbon (TOC) (5310C Laboratory TOC analyser, Sievers) were measured to characterize the amount and nature of the organic matter in the samples. The ionic content of the samples was characterized through conductivity (Conductivity Testr, Oakton instruments) and total, Fe, Mg and Ca concentrations (AAAnalyser200, PerkinElmer). Particle counts (DPA 4100, Brightwell technologies) and turbidity (2100AN, HACH) were measured to characterize the particulate/colloidal content. It should be noted that the DPA 4100 can only measure particles in the 2.25–300  $\mu\text{m}$  range. The specific ultraviolet absorbance (SUVA) was calculated based on the TOC concentrations and  $\text{UV}_{254}$  absorbance. Preliminary TOC and DOC analyses showed that these two parameters had very similar values (results not shown). It was decided to continue only with the TOC analyses in the interest of reducing the number of variables measured.

### 2.5. Data analysis – seasonal water quality variations

Principal component analysis (PCA) and partial least squares (PLS) regression were done on the data from both sampling sites, first in conjunction and then separately. PCA is a multivariate exploratory technique that is useful for the classification of variables and observations and thus also for data interpretation [43]. PLS regression is a linear regression method that can be used in situations where traditional multivariate methods cannot be applied such as when there are too few observations and many collinear variables [43,44]. This was the case for the data in this study with 10 predictor

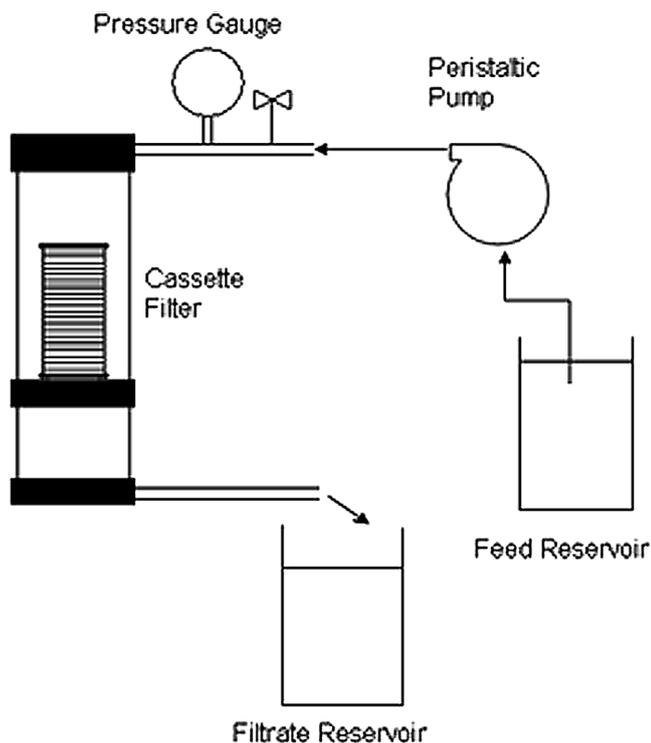


Fig. 3. Thread filter set-up.

variables and 16 observations per water source. Statistical analyses were performed with Statistica 7.1 [43].

### 3. Results

#### 3.1. Seasonal water quality variations

As mentioned previously water quality parameters were measured from February to October 2007 at two different sampling locations (SLs) throughout different seasonal conditions (precipitation, snowmelt, temperature variations, etc.); one source was a lake the other a river. The range of the values found for the quality parameter measured at Barron Lake and the Des Prairies River are presented in Figs. 4–6. As expected the lake water had a different profile than the river water. These differences will be discussed in greater details further on in this section.

It should be noted that particles with diameters between 2.5 and 3  $\mu\text{m}$  represented the most important size fraction of the total counts for both for Barron Lake and the Des Prairies River (averages of  $63 \pm 6\%$  and  $68 \pm 4\%$  of total counts, respectively). Particles larger than 10  $\mu\text{m}$  were consistently the least important fraction with averages of  $5 \pm 4\%$  and  $3 \pm 1\%$  for Barron Lake and the Des Prairies River, respectively. Particles

larger than 30  $\mu\text{m}$  represented on average less than 0.5% of the total particle concentration for both sources.

A PCA was carried out using all observations of traditional water quality parameters collected at both sites (with the exception of the MFI-UF) to better gauge if it was appropriate to analyse the results from both sampling sites together. The PCA results showed that the samples collected from the different sources varied predominantly along two different factors which are graphically represented by the vertical and horizontal axes (Fig. 7). A PLS regression was done to further investigate the role water source plays in result interpretation. It was found that SL was the variable with the greatest influence as measured by the regression coefficients on the model produced by this method (Table 1). Therefore, the statistical analyses that followed were done separately for each of the SLs.

PCAs for each of the sources elucidated the multicollinearity between several of the variables measured. TOC,  $\text{UV}_{254}$ , color and turbidity all varied in the same direction along the same factor for the samples collected at Barron Lake (Fig. 8). All parameters had peak values during the beginning of June and end of October (see Fig. 9). Turbidity, conductivity, Ca, Mg and Fe concentrations as well as the particle counts all varied in the same direction along factor 1 for the samples collected at the Des Prairies River (Fig. 10). These variables had peak values mid April, during the period of snowmelt (see Fig. 11). Color, UVA and SUVA also varied similarly at the Des Prairies River (no marked peaks during the sampling period).

In general the parameters associated with particulate and colloidal matter (turbidity and particle counts) were significantly higher for the river samples than for the lake samples. The parameters associated with ionic content (Fe, Mg, Ca) had slightly higher values for the river water samples and the organic matter parameters had comparable values for both SLs, but lake samples tended to have slightly more marked minimum and maximum values. Surprisingly, the ranges of the MFI-UF values were nearly the same for both water sources despite the significant differences in the signatures of the two waters (Figs. 4–6).

A PLS regression was done for each of the water sources in order to investigate any apparent relationships between the MFI-UF and the traditional water quality parameters that were measured. The models produced were able to predict MFI-UF values to coefficients of determination ( $R^2$ ) close to 0.90 for both sampling sites. For the model built from measurements at Barron Lake the variables with the greatest influence on the variability of the MFI-UF were turbidity, TOC and Fe concentration. Color and  $\text{UV}_{254}$  absorbance also played a role but to a somewhat lesser extent. For the

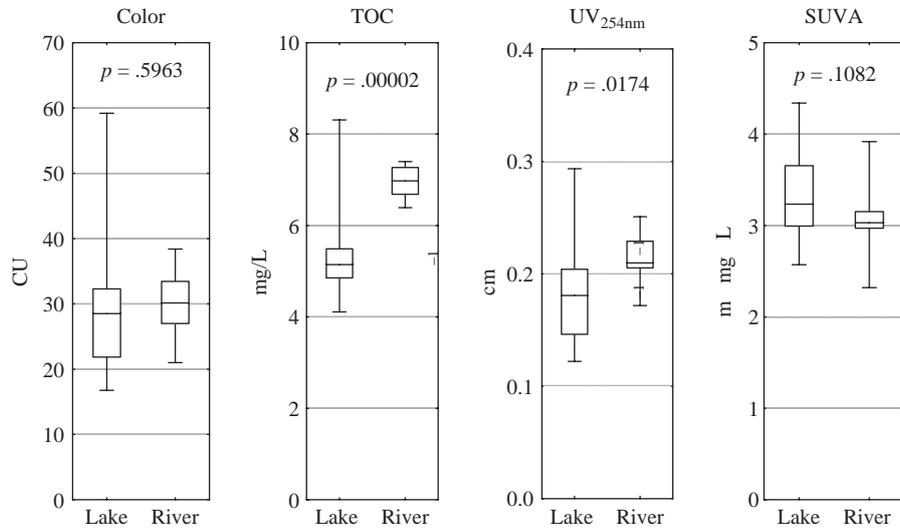


Fig. 4. Range of NOM parameters measured during seasonal monitoring: —, median; □, 25–75%; and I, min–max.

model built from the Des Prairies River observations, the variables that were the most influential on the prediction of the MFI-UF were, respectively, turbidity particle counts and Fe concentrations and to a slightly lesser degree conductivity and Mg concentrations. The models' scaled regression coefficients are presented in Tables 2 and 3.

not reported since the filters used were expected to affect parameters measuring particle content only (the minimum porosity studied was 2 μm). The UV<sub>254</sub>, color and conductivity of filtrate samples from the 2, 3, 10 and 25 μm filters were measured as indicators of changes in NOM and ionic content. No appreciable changes were noted (data not shown).

### 3.2. Pre-treatment

Both the effects of pore size and cake formation on pre-filter particle removal and fouling potential reduction were investigated. Traditional water quality parameters other than particle counts and turbidity were

#### 3.2.1. Effects of pre-filter porosity on MFI-UF and particle counts

As mentioned previously four pre-filters (rated 2, 3, 10 and 25 μm) were used to determine the role pre-filter porosity plays on MFI-UF reduction and particle

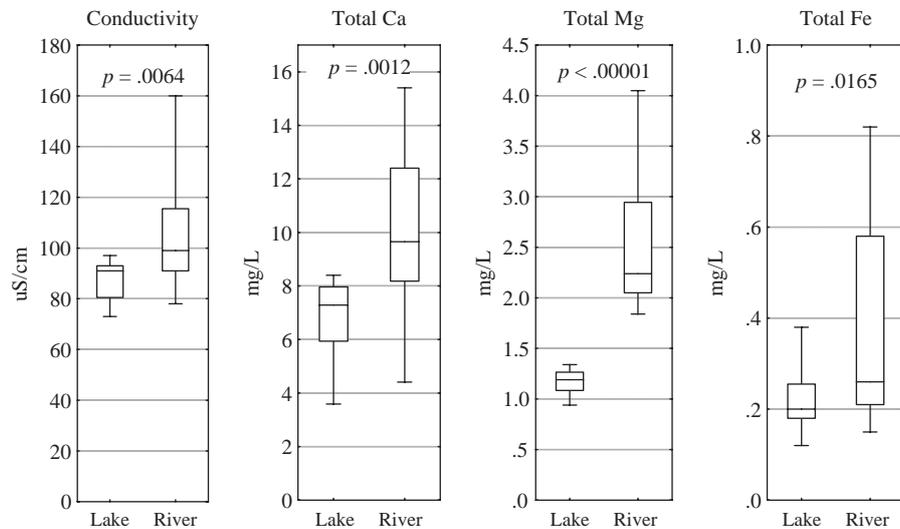


Fig. 5. Range of ionic species measured during seasonal monitoring: —, median; □, 25–75%; and I, min–max.

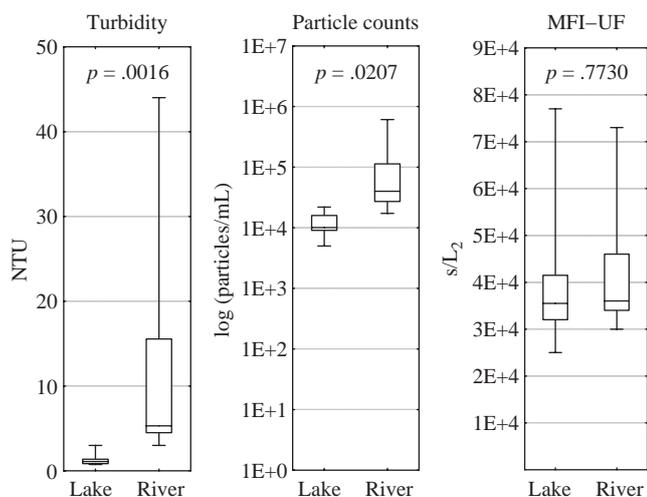


Fig. 6. Range of solids parameters measured during the seasonal monitoring: —, median; □, 25–75%; and ⊥, min–max.

removal. The pre-filters ability to remove particles of different diameters was evaluated (Fig. 12). The 2, 3 and 10  $\mu\text{m}$  filters were found to be capable of removing 74% (0.6 log), 82% (0.7 log) and 66% (0.5 log) of particles with diameters greater than the “reported pore size”, respectively. The results for the 25  $\mu\text{m}$  filter are not presented as the concentration of particles of diameters greater than 25  $\mu\text{m}$  in the raw water used for this experiment was negligible (<50 particles/mL). It is also important to mention that the concentration of particles

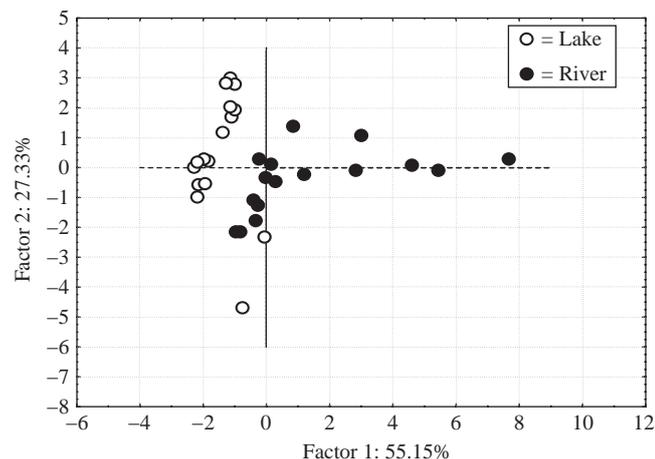


Fig. 7. PCA case projection – all observations.

Table 1  
PLS scaled regression coefficients - combined

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV <sub>254</sub>	SUVA	PC	SL
MFI-UF	0.188	0.065	0.351	0.134	−0.200	0.40236	0.213	0.109	−0.214	0.253	0.571

greater than 10  $\mu\text{m}$  in diameter in the filtrate were also extremely low (150 particles/mL).

Pre-filters reduced turbidity marginally, even at lowest porosity (2  $\mu\text{m}$ ). The raw water sample had a turbidity of 3.12 NTU while the 2, 3, 10 and 25  $\mu\text{m}$  filtrates had turbidities of 2.36, 2.55, 2.66 and 2.67 NTU, respectively. The filtrations were found to have little to no effect on the MFI-UF values (Fig. 12); variations that are less than 10% are considered as marginal [36], and all measurements were found to be within  $\pm 10\%$  of MFI-UF of the raw water sample used for this set of tests (30,000  $\text{s/L}^2$ ).

### 3.2.2. Effects of cake layer formation on pre-filter on MFI-UF and particle counts

The effects that the formation of a cake on a 2  $\mu\text{m}$  cassette filter has on MFI-UF and particle counts are presented in Fig. 13. The removal of particles with diameters greater than 2.25  $\mu\text{m}$  decreased slightly from 89% (0.95 log) to 87% (0.88 log) to 83% (0.76 log) as the filtration run progressed. The raw water sample had a turbidity of 9.6 NTU while samples collected at the specific volumes of 0.2, 0.7 and 1.2  $\text{m}^3/\text{m}^2$  had turbidities of 4.70, 3.83 and 3.34 NTU, respectively. Although the removal of larger particles (>2.25  $\mu\text{m}$ ) was declining, the improved reduction in turbidity probably reflects the higher removal of smaller particles due to the cake layer. Nevertheless, even though a cake layer formed visibly on the cassette during the filtration, the MFI-UF of the samples taken at specific volumes of 0.2 and 0.7  $\text{m}^3/\text{m}^2$  did not vary importantly from that of the raw water sample used for this set of tests (33,000  $\text{s/L}^2 \pm 12\%$ ). Even though the variations are above the 10% threshold value, they are not significantly higher and cannot be classified as major variations.

## 4. Discussion

### 4.1. Water quality variation data

The fact that the observations from the two SLs varied distinctly along different factors in the PCA suggests that it is not possible to analyse data from different types of sources together. This conclusion was also supported by the model produced by PLS

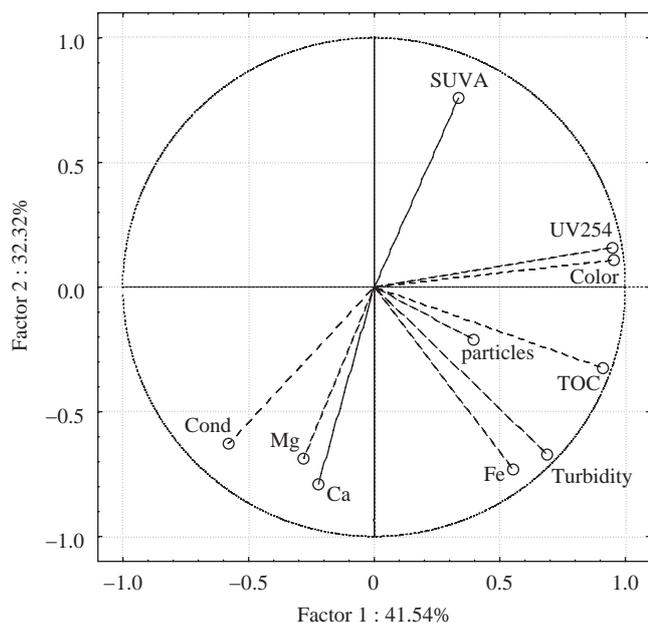


Fig. 8. PCA variable projections – Barron Lake.

regression using all the observations from Barron Lake and the Des Prairies River in conjunction. The model featured SL as the variable that contributed most heavily to explaining the variability of the fouling potential reduction.

Despite the different signatures that these waters had, the range of the MFI-UF values found were highly comparable (average of 40,000 s/L<sup>2</sup> with standard deviation of 14,000 s/L<sup>2</sup> for Barron Lank and average of 41,000 s/L<sup>2</sup> with standard deviations of 14,000 s/ L<sup>2</sup> for the Des Prairies River), leading us to believe that there was an unmeasured parameter or

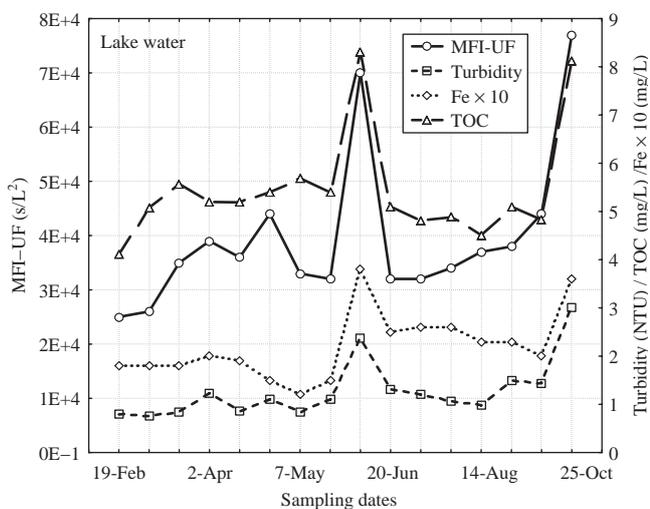


Fig. 9. Barron Lake – fouling potential and related variables.

parameters that were responsible for flux decline. While turbidity was found to be strongly correlated to the MFI-UF both for Barron Lake and Des Prairies River samples, the vastly different turbidity values associated with similar MFI-UF values suggest that it is most likely a specific fraction of the particulate/colloidal matter which is responsible for the fouling observed. The TOC concentration of the Barron Lake samples was strongly correlated to the MFI-UF while TOC concentration for the Des Prairies River samples was not found to be particularly relevant to relationship developed to model fouling for that source water. It is possible again that there is a specific fraction of TOC which is responsible for fouling, and this fraction varied proportionately with TOC concentrations for Barron Lake but did not for the Des Prairies River. Since no additional characterization of the NOM of the raw waters was done, it is no possible to conclude on the fraction that might be responsible for the observed fouling.

The presence of Fe as a relatively important parameter in both models could be explained by iron oxide precipitation on the membrane and/or by increasing iron concentrations being indicative of the increased presence of another fouling agent no characterized here. Since no analyses were done on the membrane coupons it is not possible to exclude the presence of iron oxides as a fouling source. It should be noted, however, that iron concentrations in both sources were consistently below 1 mg/L and that no oxidizing agents were used during the measurement of the MFI-UF.

The same parameters did not have the same importance in the different models. This highlights the importance of using natural waters for fouling studies, since using laboratory solutions that are composed of single model elements of turbidity or organic matter for example do not capture the variety of the characteristics that these potential foulants have.

All this implies that when speaking about fouling, traditional water quality parameters lack the precision to describe and quantify the foulants which are causing the flux reduction amongst different water sources. However, the high coefficients of determination ( $R^2 > 0.90$ ) for the models produced by using PLS regression for each water sources indicate that traditional water quality indicators could be used to easily follow seasonal variations in fouling potential once the signature of a specific water source has been adequately described. It should be noted that since PLS regressions were used instead of simple linear regressions to explore the relationships between the different variables, correlations between the MFI-UF and individual water quality parameters were determined based on the importance or scaled regression coefficients of the parameter in the

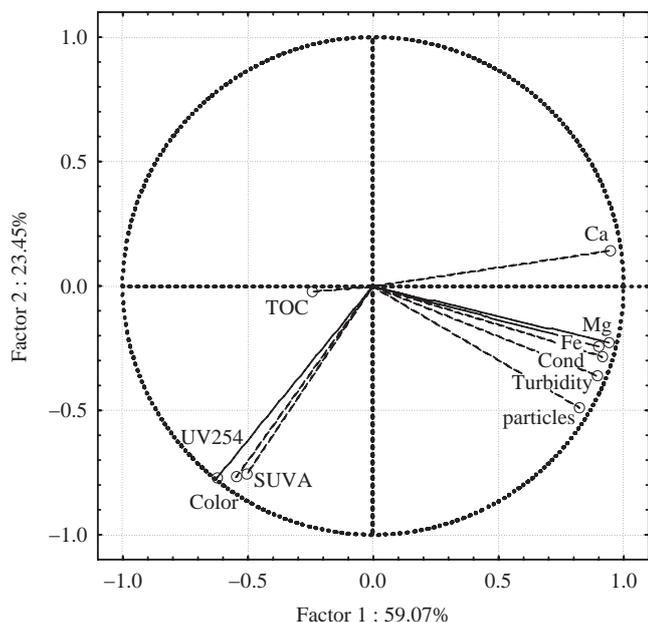


Fig. 10. PCA variable projections – Des Prairies River.

overall model instead of the global  $R^2$  values. These values are presented in Tables 2 and 3.

As mentioned previously the focus of many of the studies presented in the literature has been to observe and dissect a single fouling phenomenon, often this by means of synthetic or fractionated waters which simplify the control of certain parameters. This kind of approach has led to the identification of possible foulants such as hydrophilic, high molecular weight/colloidal NOM [13,16,45,46], calcium – NOM complexes [11,47], iron oxides [22] and inorganic colloid – NOM

complexes [18]. However, this kind of approach does not typically consider the effects of the seasonal variations that can occur with a raw natural water source. And even in the case where these variations have been considered the number of samples used remained small [48].

The work presented here attempted create a context within which fouling potential can be interpreted by considering two different raw water sources over a prolonged period of time. Also, the difficulties associated with the analysis of many correlated variables were addressed by using a linear regression method that circumvents many of the limitations of more traditional linear regression methods.

#### 4.2. Pre-treatment

The samples of filtration screens/thread filters tested did not completely remove all particles which were larger than the porosities reported by the manufacturer. This incomplete particle removal by commercial cartridge filters has also been noted by other researchers [12]. However, the thread filters consistently removed more than 70% ( $\approx 0.5$  logs) of particles which were of greater size than the announced porosities. Therefore, the removal of particles with diameters greater than  $2 \mu\text{m}$  did not significantly affect fouling potential of the natural waters. This was illustrated by the little to no change in MFI-UF values of raw water samples and pre-filtered samples. This result is in concurrence with findings published by Koyuncu et al. [49] who also found that larger particles played a negligible role in fouling.

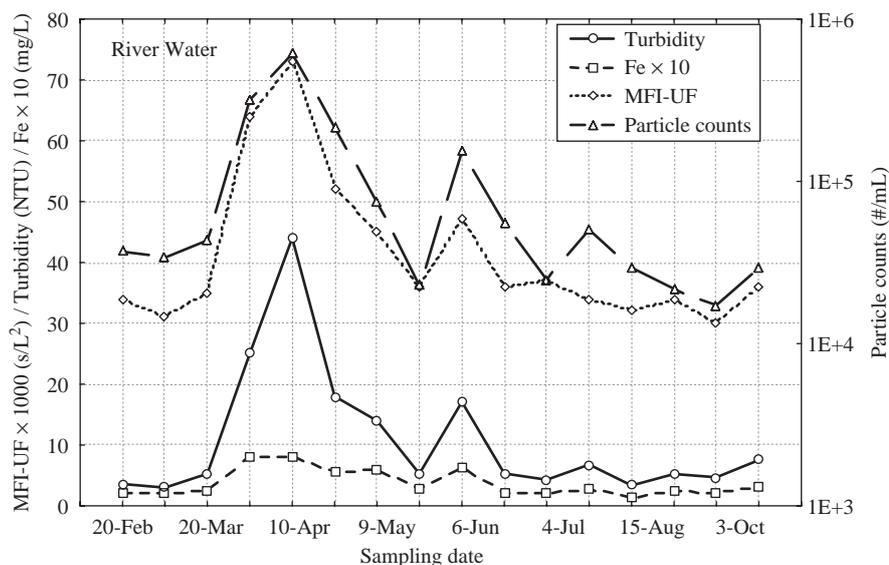


Fig. 11. Des Prairies River – fouling potential and related variables.

Table 2  
PLS scaled regression coefficients – Barron Lake

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV <sub>254</sub>	SUVA	PC
MFI-UF	0.240	-0.067	0.167	0.050	-0.006	0.194	0.231	0.165	-0.047	0.102

Table 3  
PLS scaled regression coefficients – Des Prairies River

	Turbidity	Conductivity	Color	Ca	Mg	Fe	TOC	UV <sub>254</sub>	SUVA	PC
MFI-UF	0.172	0.160	0.030	0.138	0.161	0.166	-0.024	-0.046	-0.034	0.169

MFI-UF changes due to pre-treatment reported in the literature are in the range of 1 log reduction [50,51] while the changes that were observed here were very close to 10%, a value which changes can be considered marginal considering that MFI-UF can vary on a log scale. It seems likely then that the culprits behind fouling are colloids which are in the sub-micron range. Organic colloids in particular have been identified by several researchers as being strong contributors to the fouling of MF/UF membranes [14,52] as well as NF membranes [13,16,53,54]. Evidence suggesting certain inorganic colloids such as iron oxides have a linear correlation to fouling has also been presented [22].

It should be noted that the raw water samples that were used to evaluate the efficiency of the pre-filters had very few particles with diameters greater than 10  $\mu\text{m}$ . While particles larger than 10  $\mu\text{m}$  were consistently the least important size fraction for all samples collected from both the Des Prairies River and Barron Lake, they were still sometimes present in non-negligible numbers. The average concentration of

particles in this size range for Barron Lake was 500 particles/mL while for the Des Prairies River it was 2000 particles/mL. Thus 10  $\mu\text{m}$  pre-filtrations would provide some protection against plugging with water sources like the Barron Lake and the Des Prairies River. Filtration coarser than 30  $\mu\text{m}$ , however, would most likely be ineffective since particles larger than this porosity were typically found in negligible concentrations.

The 5  $\mu\text{m}$  particle removal which is a standard specification requested by membrane manufacturers protects the feed spacer in spiral wound modules from obstruction by larger particles during events of high particle loading, but its effects on the protection of the membrane itself from fouling are most likely negligible as illustrated by the results presented here.

The formation of cake layer at a constant flowrate and increasing differential pressure did not significantly improve the performance of the filter in terms of MFI-UF reduction or particle removal. The pre-filter effluent collected at different specific volumes did not have MFI-UF values that varied by important amounts ( $\pm 13\%$ ) from the raw water sample. Indeed, the removal of large particles decreased as the filtration run progressed. This is possibly the result of the increasing shear forces that developed in the filter due to the reduction of the open pore area, an effect reflected by the increase in differential pressure. Thus, the cake layer formed under constant pressure did not succeed at progressively reducing fouling potential or the number of larger particles by virtue of lower cake layer porosity.

As mentioned previously the majority of the studies presented in the literature have focused on the low pressure membranes, conventional treatment [32,55,56], biological filtration [33], ozonation [35] and a combination of the above [36]. There is therefore no wealth of information regarding simpler pre-treatment schemes to which to compare the present results.

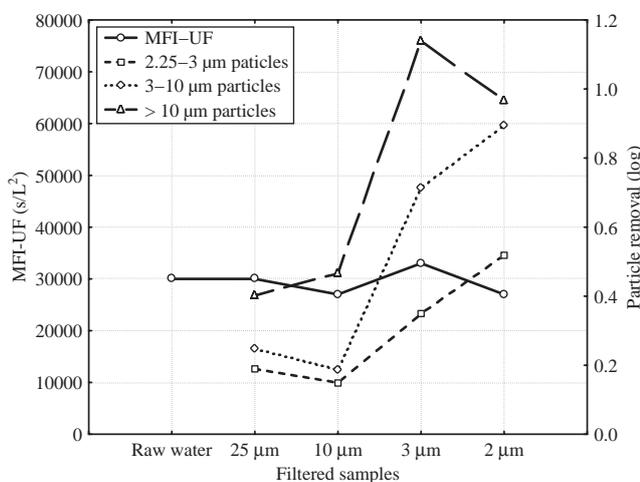


Fig. 12. Pre-filter performance pore size effects.

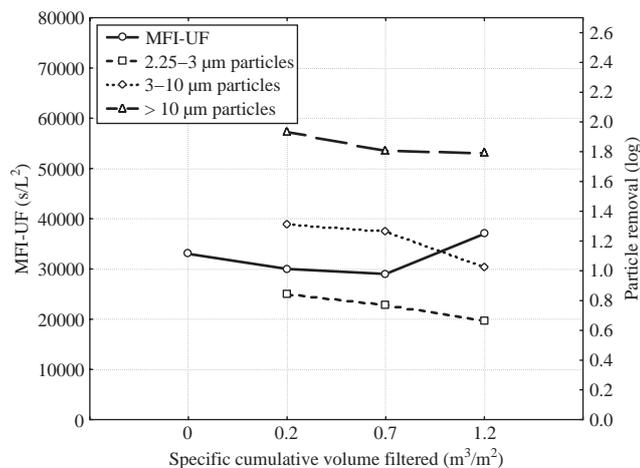


Fig. 13. Pre-filter performance – cake layer effects.

Since colloids appear to play a key role in fouling potential, future works should focus on developing better methods for colloid quantification (possibly colloidal counts) and applying these methods to natural waters to better understand the role colloids play in fouling in conjunction with larger particles, specific NOM fractions and ionic parameters.

## 5. Conclusion

In conclusion, it is not possible to analyse data from different sources together since traditional water quality parameters lack the precision to accurately describe the actual foulants present in natural water samples. However, if a single water is studied, and comparisons between sources are not made, these parameters can be used to model fouling events and easily follow seasonal variations in fouling potential associated with climatic events (spring turnover, heavy precipitation, algal blooms, etc.) which could be useful when planning membrane piloting schedules to agree with the time when a pilot would face the greatest challenge in terms of fouling potential.

The use of MFI-UF as the fouling index allowed for the effects of particles and colloids smaller than 0.45 µm on fouling to be taken into consideration during this study. Despite the fact it is not as widely used as the SDI, and that the time required for its measurement is longer than the SDI's, for the purpose of this work (fouling evaluation), the MFI-UF provided more information than the SDI would have.

While an improved detailed understanding of complex fouling mechanisms was not attained, the inadequacy of traditional water quality as general fouling indicators was shown. This is information that is

valuable to the combined scientific community's efforts to achieving a better overall picture of membrane fouling.

The automatic backwash filter samples tested did not completely remove particles from the feed, but there was a minimum reduction of 0.5 logs of particles larger than the reported thread filter porosity. The formation of the cake layer on the pre-filter improved neither the removal of particles nor the fouling potential reduction. The removal of large particles only (>2 µm) has a marginal effect on fouling potential reduction, and it is suspected that colloids in the sub-micron range are responsible for the observed fouling.

Future works should focus on the quantification and qualification of colloids in natural raw waters, and the comparison of this colloidal signature to the fouling potential over time.

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