Pilot scale evaluation of biofiltration as an innovative pre-treatment for ultrafiltration membranes for drinking water treatment


ABSTRACT

Fouling remains one of the major constraints on the use of low pressure membranes in drinking water treatment. Work over the last few years has shown the importance of biopolymers (carbohydrates and protein-like material) as foulants for ultrafiltration (UF) membranes. The purpose of this study was to investigate at pilot scale the use of rapid biofiltration (without prior coagulation or ozone addition) as an innovative pretreatment to reduce fouling of UF membranes. The investigation was carried out on a water with a higher than average DOC and significant temperature variation. The biofilters, each operated at a hydraulic loading of 5 m/h, had empty bed contact times of 5, 10 and 15 minutes. The membrane unit was operated at a flux equivalent to 60 LMH at 20°C. The investigation confirmed the encouraging results obtained in an earlier smaller scale study with essentially the same water. Increased biofiltration contact time (i.e. increased bed depth) led to lower rates of hydraulically irreversible fouling. The initial biofiltration backwash procedure, involving air scour as is common in chemically assisted filtration, led in some cases to an increased rate of membrane fouling immediately after the backwash. An alternative backwashing strategy was developed, however the feasibility of operating with this approach over very long periods of time needs to be confirmed. To assist in full-scale implementation of this “green” and simple pretreatment, the design and operating conditions for the biofilters should be optimized for various types of waters. It is expected that biofiltration pretreatment will be of particular interest for small and/or isolated systems where a higher initial capital cost may be acceptable because of operational simplicity and reduced chemical requirements.

Key words | backwashing, biofiltration, biopolymers, drinking water, organic fouling, ultrafiltration

INTRODUCTION

The fouling of low pressure membranes used for drinking water treatment is an important factor affecting their technical and economic feasibility. Although particulate matter is an important foulant, recent investigations have shown the importance of specific natural organic matter (NOM) fractions as foulants. Her et al. (2007) identified protein- and polysaccharide-like substances as the major foulants on nanofiltration membranes. Lee et al. (2004) and Kimura et al. (2004) have implicated colloidal and/or high molecular weight macromolecules such as polysaccharides in causing
irreversible fouling on low pressure membranes. Howe & Clark (2002) reported that particulates and dissolved organic matter (DOM) were relatively unimportant in the fouling of microfiltration and ultrafiltration (UF) membranes using natural surface water, and that the greatest contributors to fouling were small mostly organic colloids (3–20 nm). In investigating ultrafiltration of secondary wastewater effluent, Haberkamp (2008) showed the importance for fouling of biopolymers (polysaccharides and protein-like substances) and/or large humic substances. In treating an agriculturally and municipally impacted river water, Hallé et al. (2009) have also demonstrated the importance of the biopolymer fraction in both hydraulically reversible and hydraulically irreversible fouling.

The work reported by Hallé et al. (2009) involved the use of a novel membrane pretreatment, i.e. biofiltration without prior coagulation or ozone addition, to reduce fouling. Although biofiltration as a membrane pretreatment has been previously studied (e.g. Persson et al. 2006; Hu et al. 2005) and biofiltration preceded by ozonation is being used in a large full scale ultrafiltration plant in the Region of Peel, near Toronto, Canada (Farr & Stampone 2007), detailed investigations have not been reported.

Biological processes have a long history of use in drinking water treatment, in the form of processes such as slow sand filtration and bank filtration. In more recent decades, biological rapid filtration processes have come into use. Key early work in this regard was the development of the Mülheim process in Germany (Sontheimer et al. 1978). Basically a filter will operate biologically whenever there is no disinfectant residual throughout its depth, in that a biofilm will develop on the surface of the media. Although biological filtration in drinking water treatment can also be used for other applications such as nitrogen removal (e.g. Rittmann & Huck 1989) the most common use is for the removal of organic carbon.

Although biofiltration has the potential to remove the types of organic matter identified above as important for low-pressure membrane fouling, biofilters also produce some of the same materials. Therefore if they are to be applied as a membrane pretreatment, it is important that they be designed and operated to maximize the net removal of dissolved and particulate organic matter (Huck & Sozański 2008).

In modelling biofiltration, Zhang & Huck (1996a, b) introduced the parameter X° (dimensionless empty bed contact time or EBCT). X° is proportional to EBCT and also incorporates other factors important for biofiltration performance: media surface area for biofilm growth, biomass density, and BOM diffusivity and biodegradation kinetic parameters. For easily biodegradable organic matter, expressed as assimilable organic carbon or AOC (e.g. van der Kooij et al. 1982), Zhang & Huck (1996a) showed that removals were approximately proportional to X° for lower values of X°, gradually tapering off at higher values of X°. Huck & Sozański (2008) discussed the applicability of X° to other biofiltration applications, including as a membrane pretreatment to reduce fouling. Although noting the greater complexity of this application, they concluded that X° should be useful.

In order to make X° more applicable to biofilter design and operation, Huck & Sozański (2008) also introduced a new practically-oriented parameter, the Biofiltration Factor (BF). BF is directly proportional to X° and essentially numerically equal to it. However it is directly tied to practice and to “concrete” conditions because a biofilter is defined as having a BF value of 0.5 if it is achieving 50% AOC removal at approximately 20°C. The potential applicability of BF for biofiltration as a membrane pretreatment is discussed at the end of the paper.

Hallé et al. (2009) (see above) provided “proof of concept” that biofiltration without prior coagulation or ozonation could provide effective reduction in fouling for a UF membrane. The work was conducted using relatively small-scale (50 mm diameter) filtration columns and a bench-scale UF unit. The work reported herein was conducted at pilot scale using essentially the same water to investigate the process at a larger scale and using longer membrane run times.

**MATERIALS AND METHODS**

**Feedwater**

The Grand River is located in southern Ontario in Canada and is impacted by agricultural, industrial and municipal activities. It serves as a drinking water source for several municipalities. As is evident from Table 1, there are wide variations in both temperature and turbidity. Although DOC levels are higher than average, there is only moderate
variation. However, specific UV absorbance (SUVA) values had large variations probably in part due to associated high turbidities. The temperature variation is essentially seasonally-based; however large turbidity peaks can occur at numerous times throughout the year, except in the coldest months. The variations in alkalinity, hardness and conductivity reflect a higher percentage of groundwater in the river during the winter. Before being pumped to the treatment plant where the pilot plant was located, the river water passes through several raw water storage basins (no chemical addition), with a retention time on the order of several days. Thus although the pilot plant was fed with untreated treatment plant influent, it did not experience the same turbidity peaks shown in Table 1. The maximum influent turbidity to the pilot plant during the period reported in this paper was approximately 25 NTU.

Biofilters

Three different biofilter empty bed contact times (EBCTs) (A: 5 min, B: 10 min and C: 15 min) were investigated, using parallel dual-media filters (anthracite/sand). These EBCTs are within the range used for rapid granular media filtration and granular activated carbon contactors. The different EBCTs were achieved through different bed depths (Filter A: 15 cm diameter, top to bottom, 20 cm anthracite, 20 cm sand, 15 cm gravel support; Filter B: 20 cm in diameter, top to bottom, 20 cm anthracite, 63 cm sand, 15 cm gravel support, Filter C: 15 cm in diameter, 40 cm sand over 15 cm gravel support). Filter C was fed by gravity by Filter B and the combined EBCT for the effluent of C was 15 min. All filters were operated in downflow mode at 5 m/h. Each filter was preceded by a small roughing filter to exclude debris and plant material. During the period for which data are presented in this paper all filters were backwashed using collapse pulsing with air scour and 50% bed expansion approximately every 5 days unless performance parameters indicated that an earlier backwash was necessary.

Ultrafiltration

The pilot-UF membrane module (Zeeweed 10 Pilot, GE/Zenon, Oakville, Canada) was equipped with polyvinylidene (PVDF) hollow fibres which are operated in outside-in mode. This module was installed vertically in a 15 L tank. The nominal surface area was 0.93 m² and the nominal pore size is 0.04 μm (manufacturer’s information). The pilot unit was operated in a dead end mode using a constant permeate flux with an overall recovery of 90%. Every 40 minutes the UF module was automatically backpulsed for 60 seconds using the operating flux while simultaneously sparged with air. Next the membrane tank was drained completely and then refilled before normal operation was resumed. The UF pilot unit was fed with the effluent of one biofilter at a time as only one pilot unit was available.

Analyses

Turbidity, temperature, flux and transmembrane pressure (TMP) were monitored on-line. General water quality parameters were monitored weekly for raw water, each biofilter effluent and permeate. Total organic carbon (TOC), dissolved organic carbon (DOC), UV absorbance (254 nm, 1 cm cuvette), pH and conductivity were measured using procedures from Standard Methods (2005). Specific UV absorbance (SUVA) was calculated as follows: SUVA = (DOC/UV_254) × 100.

RESULTS AND DISCUSSION

A total of four runs were conducted between late September and mid-December 2008, during a time of steadily decreasing water temperature (Table 2). Because only one membrane
When the UF unit was available, the experiments were conducted sequentially. For the experiment with Filter C and the second one with Filter A (A2), membrane flux was adjusted using a temperature (viscosity) correction factor to be equivalent to 60 LMH at 20 °C (this procedure was not yet in place for the experiment with Filter B). The increase in flux partway through the experiment with Filter C was undertaken to assess the effect of this change. Changes in flux did not affect the flow rate through the biofilters, as there was always some excess biofilter effluent that was run to waste.

The information in Table 2 is discussed more fully following the presentation of the transmembrane pressure (TMP) results (hydraulically irreversible fouling) as a function of time for the four runs (Figure 1). The designations A, B and C in the figure refer to the particular biofilter effluent being used to feed the membrane during that run. Each experiment was run for at least 200 hours and the biofilters were backwashed as indicated in the figure. Extensive (chemical) cleaning of the UF unit (i.e. soaking in hypochlorite solution for at least 6 h) was performed between experiments to remove hydraulically irreversible fouling and the original performance was recovered as confirmed by clean water permeability tests.

It is evident from Figure 1 that increased EBCT has a beneficial effect on the rate of irreversible fouling. In general, the two runs using Filter A effluent (A and A2) are reproducible, as indicated by the general similarity in the slopes of the TMP versus time curves. (Run A2 is a replicate of run A, with membrane flux adjusted as described above to account for the increased water viscosity at lower temperature.) It is also

### Table 2 | General parameters for each run (2008)

<table>
<thead>
<tr>
<th>Feed - biofilter</th>
<th>Duration</th>
<th>Flux (l/m²h)</th>
<th>TMP (kPa)</th>
<th>Temp. (°C)</th>
<th>UF feed turbidity (NTU)</th>
<th>UF feed DOC (mg C/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Sept. 26–Oct.11</td>
<td>60</td>
<td>21–68</td>
<td>13–18</td>
<td>0.29–0.62</td>
<td>7.5–7.7</td>
</tr>
<tr>
<td>B</td>
<td>Oct. 11–22</td>
<td>60</td>
<td>26–49</td>
<td>12–15</td>
<td>0.20–0.32</td>
<td>6.7–7.5</td>
</tr>
<tr>
<td>A2</td>
<td>Nov.12–26</td>
<td>50</td>
<td>32–80</td>
<td>3–9</td>
<td>0.84–3.4</td>
<td>7.1–7.4</td>
</tr>
<tr>
<td>C</td>
<td>Nov.28–Dec.8</td>
<td>42</td>
<td>20–27</td>
<td>4–6</td>
<td>0.55–1.9</td>
<td>7.0–8.1</td>
</tr>
<tr>
<td>C</td>
<td>Dec.8–11</td>
<td>53</td>
<td>37–40</td>
<td>4</td>
<td>0.81–1.1</td>
<td>6.0–6.3</td>
</tr>
</tbody>
</table>

**Figure 1** | TMP vs. time for membrane fed with effluents of biofilters A, B and C (5, 10 and 15 minutes EBCT respectively).
evident that there was only a very slight increase in slope following the increase in flux partway through the experiment with Filter C.

Maintenance cleaning, i.e. soaking of the UF unit for 30 min in a dilute hypochlorite acid solution, was performed once per experiment after 8 or 9 days with the exception of the run with biofilter C. Good TMP recoveries ranging from 17 to 38 kPa were achieved (Figure 1). However, the operating TMP after maintenance cleaning was always somewhat higher than the original starting TMP, indicating that maintenance cleaning will have to be performed more frequently. Even if maintenance cleaning has to be performed 2-3 times per week this is still less frequent than in full-scale practice where it is often performed daily. Hence, this would still amount to substantial savings in terms of downtime and chemical costs.

Turbidity was monitored on-line for the biofilter influent and effluents and the UF permeate. For the investigation reported herein, raw water (biofilter feed) turbidity ranged from 1.38 to 24.6 NTU with an arithmetic mean of 3.6 NTU and a standard deviation of 3.0 NTU. Generally effective removal was provided by the biofilters, with mean effluent turbidities below 1 NTU (Figure 2). As expected, for a given raw water turbidity, effluent turbidity decreased as EBCT (i.e. bed depth) increased. It will be recalled that the filters operate without prior coagulation, and since they are operating as a pretreatment, they do not have to meet turbidity regulations. In some cases TMP increased following biofilter backwashing, and optimum backwashing strategies for these filters were investigated, as discussed later.

During the experiment with Filter C, which was conducted later in the fall, the raw water turbidity was higher, and the effluent turbidity of Filter C was higher (between 0.5 and 1 NTU) than the effluent turbidities of Filters A and B when the membrane was fed by them during the earlier runs. Since the rate of increase in TMP during the experiment with Filter C was lowest, it is evident that the rate of hydraulically irreversible fouling is not directly related to turbidity. These results imply that the longer EBCT in Filter C resulted in effective removal of the biopolymer fraction responsible for irreversible fouling, supporting the earlier work using 5 cm diameter biofilter columns and a laboratory scale membrane unit (Hallé et al. (2009)). Although results are not shown for reasons of space, Liquid Chromatography-Organic Carbon Detection (LC-OCD) analyses performed in the summer and early fall of 2008 showed that the biopolymer concentration in the biofilter effluent decreased as EBCT increased.

As is evident in Figure 1, in most cases the slope (i.e. the rate of increase in membrane TMP) increased following filter backwashing. This increase was more pronounced for runs with higher rates of fouling and is most evident for the backwashes taking place at approximately 120 hours. Initially, a standard backwashing procedure used for chemically assisted filtration (including air scour) was employed, followed by a 30 min filter-to-waste period before directing the filter effluent to the UF. However, given that the filters are operated without prior coagulation, it may be that too rigorous a cleaning impairs particle attachment in the early part of the immediately-following filter cycle. In fact, typical filter ripening curves were not observed when monitoring turbidity during filter-to-waste periods. Towards the end of 2008, experimentation with different backwashing strategies was carried out. Eventually the procedure adopted involved interspersing standard backwashing with a less rigorous procedure - employing 50% bed expansion without air scour. Although this was found to be beneficial as is evident at 200 h in the experiment with Filter C where the slope of the TMP curve remains essentially the same after backwash, the most appropriate procedure may need to be determined for different types of waters, and may be seasonally dependent. The procedure would need to be validated over a long enough
period to ensure that the inclusion of less rigorous cleaning for some filter cycles does not lead to long-term filter operational problems, such as mudballing. In a large treatment plant with a number of parallel filters, if the membranes are fed with blended filter effluent, an increase in turbidity of an individual filter following backwashing may be less of an issue.

Additional runs were carried out during the winter and early spring of 2009, under cold water conditions (temperatures ranging from 2 to 14 °C). The results showed excellent performance of the UF membrane, with a very low fouling rate over several weeks when using effluent of Filter C. However, regular maintenance cleaning proved to be important, as otherwise dramatically increased fouling rates were observed. As expected, repeat runs with Filters A and B in spring 2009 displayed a higher fouling rate than the Filter C run, albeit to a much lesser degree than in the fall of 2008. This seasonal difference is consistent with bench-scale results observed a year earlier where fouling rates were lower in the spring than in the fall. Although it is possible that the concentrations of the principal organic foulants in the pilot plant influent were lower in the spring than in the fall, it is also possible that the fact the biofilters had been in operation for an additional several months contributed to better performance. Further investigation is required to determine the cause of the need for regular maintenance cleaning. It is possible that, after operation of the membranes for several months throughout the fall, biofouling (as opposed to organic and particulate fouling) became more established.

In evaluating the results from this study, it may be important that Filter C, for practical reasons, actually consisted of two separate filters in series (this was also the case for the longer EBCT filter operated in the previous small-scale studies reported by Hallé et al. (2009)). This prevented complete mixing of the bed during backwashing and may have facilitated the development of a succession of microbial communities, and that could have impacted performance. This aspect should be investigated in future studies.

CONSIDERATIONS FOR IMPLEMENTATION

Although it will be important to test the application of biofiltration (without coagulation) prior to ultrafiltration on other waters, and to evaluate the backwashing procedures over a longer period of time, the good performance obtained at pilot scale in this investigation on a challenging water indicates that this technology has promise at full scale. Although its capital cost would certainly be higher than for the usual alternative (coagulation/flocculation and possibly sedimentation) biofiltration may be particularly promising for small and/or remote systems, where greater value may be placed on simple and robust systems. It may also have particular value in other situations where there may be a desire to reduce the use of chemicals for coagulation and/or cleaning.

Ultimately it will be important to determine the necessary design and operating conditions for biofiltration as a membrane pretreatment, to make the process as economically viable as possible. The biofiltration factor (BF) introduced by Huck & Sozański (2008) and discussed earlier provides a potential framework for doing this. The current investigations showed that with relatively standard anthracite/sand filtration media, an EBCT of five minutes was able to give good performance, and progressively better performance was achieved at the two longer EBCTs. A biofiltration factor could not be determined for the filters because AOC measurements could not be made. In future work it is our intention to include these measurements. For example, AOC measurements could be made for Filter A to determine its Biofiltration Factor. The BF values for the other filters could then be obtained by a simple ratio of the contact times. Ultimately it should be possible to develop a table or nomograph giving the necessary BF values for representative types of waters to achieve a given reduction in membrane fouling, or alternatively a given interval between chemical cleanings for a given operating flux. These BF values could then be translated into contact time for a given media. X* and BF themselves do not need to be adjusted for temperature, because this is taken care of by temperature-related variations in factors such as the biodegradation kinetic parameter that go into calculating X*. However because of slower kinetics at lower temperatures, a given X* value requires a longer contact time at lower temperatures to compensate. Therefore the table or nomograph would have to provide values for parameters such as contact times (for typical media diameters) to achieve a given X* (and BF) at different temperatures.
CONCLUSIONS

This pilot scale investigation confirmed an earlier smaller scale study in demonstrating the feasibility of rapid biofiltration (without prior coagulation or ozonation) as pretreatment to reduce fouling in UF membranes. The work was conducted in a challenging water with significant temperature variations. It was demonstrated that increased biofiltration contact time (i.e. increased bed depth) led to lower rates of hydraulically irreversible fouling. The design and operating conditions for the biofilters, including backwashing and contact time, should be optimized for various types of waters. In terms of lifecycle costing and overall environmental impact, an optimization criterion could be the length of time required between chemical cleanings of the membrane. This simple, robust and “green” pretreatment technology is likely to be of special interest for small systems.

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