Impact of Biofilm Layer on Virus Removal Capacity and Integrity Testing of Passive Membrane Systems

by

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Impact of Biofilm Layer on Virus Removal Capacity and Integrity Testing of Passive Membrane Systems

submitted by Amanda Tobias Costa de Oliveira in partial fulfillment of the requirements for the degree of Master of Applied Science in Civil Engineering

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Abstract

Passive membrane filtration is a self-sustained process where an ultra-low permeate flux is maintained by hydrostatic pressure (i.e. gravity) and little to no fouling control measures are used. Due to their ultra-low permeate flux and ease of operation, passive membrane systems are particularly suited for small scale applications, such as decentralized water treatment in small and remote communities. A key aspect of passive membrane filtration is the development of a structurally loose and permeable biofilm layer that enables a sustainable flux to be achieved. The biofilm layer of passive membrane systems has also been associated with improved removal of humic acids, polysaccharides, proteins, assimilable organic carbon and microcystins.

The present study investigated the contribution of the biofilm layer to the virus removal capacity of passive membrane systems. Intact and breached passive membrane modules were studied. Breach sizes considered ranged from 20 to 180µm. Challenge tests (CTs) identified an increase of more than 2.0 log in the log removal value (LRV) for viruses of both intact and breached passive membranes when a biofilm layer was present. As a result, intact and breached passive membranes consistently achieved an LRV for viruses of more than 4.0 log.

The use of pressure decay tests (PDTs) to estimate the LRVs of passive membrane systems was also studied. The LRVs estimated using the standard PDT approach did not reflect the increase in LRVs detected in the CTs. Results suggested that the biofilm layer grows on top of the breaches, acting as a secondary barrier to contaminants that would otherwise bypass treatment by flowing through the breach. The standard PDT approach failed to estimate the LRVs of passive membranes because it was not developed to take into account the additional removal provided by the biofilm layer. An alternative integrity testing protocol for passive membrane systems is provided, which includes a modified PDT approach for estimation of LRVs.
Lay Summary

Passive membrane filtration is a simple and low-cost method of drinking water treatment particularly suited for small and remote communities. It uses gravity to push raw water through membranes and produce clean drinking water. The membrane pores are small enough to retain contaminants such as bacteria and protozoa. As the retained contaminants accumulate on the membrane surface, a microbial community develops and forms a biofilm layer. In the present study, the capacity of this biofilm layer to retain viruses was investigated. Viruses are too small to be retained by clean membranes. However, it was observed in the present study that membranes that have a biofilm layer are capable of retaining viruses, even if the membranes are damaged. This was attributed to the biofilm layer growing over damaged areas, providing a secondary barrier to viruses and other contaminants.
Preface

This dissertation is original, unpublished, independent work by the author, Amanda Tobias Costa de Oliveira.
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<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ASTM</td>
<td>American Society for Testing and Materials</td>
</tr>
<tr>
<td>ATP</td>
<td>Adenosine triphosphate</td>
</tr>
<tr>
<td>CDWQG</td>
<td>Canadian Drinking Water Quality Guidelines</td>
</tr>
<tr>
<td>CT</td>
<td>Challenge test</td>
</tr>
<tr>
<td>DI</td>
<td>Deionized</td>
</tr>
<tr>
<td>DL</td>
<td>Detection limit</td>
</tr>
<tr>
<td>DOC</td>
<td>Dissolved organic carbon</td>
</tr>
<tr>
<td>GDM</td>
<td>Gravity-driven membrane</td>
</tr>
<tr>
<td>LRV</td>
<td>Log removal value</td>
</tr>
<tr>
<td>LT2ESWTR</td>
<td>Long Term 2 Enhanced Surface Water Treatment Rule</td>
</tr>
<tr>
<td>MF</td>
<td>Microfiltration</td>
</tr>
<tr>
<td>MFGM</td>
<td>Membrane Filtration Guidance Manual</td>
</tr>
<tr>
<td>PDR</td>
<td>Pressure decay rate</td>
</tr>
<tr>
<td>PDT</td>
<td>Pressure decay test</td>
</tr>
<tr>
<td>PFC-GMD</td>
<td>Passive fouling control – gravity-driven membrane</td>
</tr>
<tr>
<td>TMP</td>
<td>Transmembrane pressure</td>
</tr>
<tr>
<td>TOC</td>
<td>Total organic carbon</td>
</tr>
<tr>
<td>UF</td>
<td>Ultrafiltration</td>
</tr>
<tr>
<td>VCF</td>
<td>Volumetric concentration factor</td>
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1.0 INTRODUCTION

Ultrafiltration (UF) membrane systems are a well-established technology for drinking water treatment. One of their main advantages is the capacity to remove up to 8.0 log of bacteria, 7.0 log of protozoa (i.e. *Giardia lamblia* and *Cryptosporidium parvum*) and 4.0 log of viruses (Crittenden, et al., 2012). However, their implementation in small and remote communities is highly impaired by the complexity and cost of conventional membrane systems (Peter-Verbanets, et al., 2009). This has led to the development of passive UF membrane systems as a simpler and cheaper alternative. Passive membrane systems are operated using gravity as the driving force for permeation, and employ simple mechanisms for fouling control, such as a daily 1h relaxation followed by a few minutes of air sparging (Oka, et al., 2017). These conditions allow for the growth of a structurally loose and permeable biofilm layer that enables a sustainable flux to be achieved (Peter-Varbanets, et al., 2011).

An important aspect in the operation of membrane systems for drinking water treatment is membrane integrity testing. The main goals of membrane integrity testing are (USEPA, 2005):

1. Quantification of the contaminant removal capacity of the membrane system; and
2. Detection of breaches that could allow contaminants to bypass treatment.

For compliance with the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR), membrane integrity testing should be performed at least daily, unless relevant health authorities approve less frequent testing (USEPA, 2005). Integrity testing adds to the cost and operational complexity of membrane systems, thus being incompatible with the philosophy behind passive membrane systems. For a membrane system to qualify for less frequent integrity testing under the LT2ESWTR, evidence must be provided of demonstrated process reliability, the use of multiple barriers, or reliable process safeguards (USEPA, 2005).

Previous studies have suggested that the presence of a foulant layer on a membrane can increase its contaminant removal capacity (Kreiβel, et al., 2012; Lu, et al., 2013; ElHadidy, et al., 2013; ElHadidy, et al., 2014). It has also been observed that the presence of a foulant layer on a breached membrane could mitigate the potential negative effects of the breach on contaminant removal capacity (ElHadidy, et al., 2014). Therefore, it is possible that the biofilm layer present on passive membranes could potentially contribute to the contaminant removal capacity. If it can be demonstrated that the biofilm layer present on passive membranes can consistently provide
effective treatment even if a breach is present, frequent integrity testing could be eliminated, facilitating the implementation of advanced treatment in small and remote communities.

In the present study, the impact of the biofilm layer on the virus removal capacity of passive membrane systems was investigated. Intact and breached passive membrane systems were studied. Indirect (i.e. challenge tests) and direct (i.e. pressure decay tests) integrity test methods were used to assess the impact of the biofilm layer on virus removal capacity. The results of the present study demonstrate that the biofilm layer present on passive membranes can consistently provide effective virus removal even in the presence of an integrity breach. The results also provide insight into the mechanisms by which the biofilm layer contributes to improved virus removal, as well as the appropriate approach to use to quantify the magnitude of the improved removal.
2.0 BACKGROUND INFORMATION AND LITERATURE REVIEW

2.1 Fundamentals of Membrane Filtration for Drinking Water Treatment

The first membranes were patented in the 1920s and were mainly used for laboratory procedures. From the 1950s, they started to be used for larger scale industrial applications, such as sterilization of pharmaceuticals; concentration and clarification of fruit juices, dairy products and alcoholic beverages; and industrial wastewater treatment and recovery.

It was only in the 1980s that attention was drawn to membrane filtration for drinking water treatment, due to the increasing concern of regulators about microbiological contamination. In 1993, a cryptosporidiosis outbreak in Milwaukee further accelerated the expansion of membranes for drinking water treatment. The outbreak demonstrated that C. parvum oocysts were not effectively removed by conventional granular media filtration, resulting in over 400,000 illnesses and 50 deaths. Furthermore, advancements in technology, mass production and new manufacturers have allowed a significant decrease in the cost of membrane systems. Membranes are currently considered a viable option for any kind of water treatment facility.

The membranes most commonly used for the production of drinking water from fresh water sources are often referred to as low-pressure membranes. These include microfiltration (MF) and ultrafiltration (UF) membranes. MF and UF membranes differ mainly in the size of the membrane pores. Low-pressure membranes work as very fine sieves, with pore diameters in the order of 0.1µm (MF) to 0.01µm (UF). During treatment, raw water permeates through the membrane. A pressure gradient, generally generated by a mechanical device such as a pump, provides the driving force for the permeation. The particles smaller than the membrane pores are able to pass and form the permeate stream, while larger particles are retained and form the waste stream (Figure 1).

Unless otherwise indicated, introductory material on membrane filtration presented in section 2.1 is based on Crittenden et al. (2012).
MF and UF membranes are able to remove particulate material such as algae, protozoa and bacteria. UF membranes can also remove small colloidal material such as viruses. The capacity of membranes to remove contaminants of interest is often defined based on log removal value (LRV). The LRV can be calculated using Equation 1.

\[
LRV = \log(\dot{M}_f) - \log(\dot{M}_p) = \log \left( \frac{\dot{M}_f}{\dot{M}_p} \right)
\]

For the above, \( \dot{M}_f \) is the mass flux of a contaminant of interest in the feed water and \( \dot{M}_p \) is the mass flux of a contaminant of interest in the permeate.

The mass flux of a contaminant in the feed water (\( \dot{M}_f \)) is the product of the water flow through the membrane module and the concentration of the contaminant in the feed water (Equation 2); while the mass flux of a contaminant in the permeate (\( \dot{M}_p \)) is the product of the water flow through the membrane module and the concentration of the contaminant in the permeate (Equation 3).

\[
\dot{M}_f = Q C_f
\]

For the above, \( Q \) is the water flow through the membrane module and \( C_f \) is the concentration of contaminants in the feed water.

\[
\dot{M}_p = Q C_p
\]

For the above, \( Q \) is the water flow through the membrane module and \( C_p \) is the concentration of contaminants in the permeate.
Thus, Equation 1 can be rewritten as Equation 4.

\[
LVR = \log \left( \frac{M_f}{M_p} \right) = \log \left( \frac{Q}{Q_C} \frac{C_f}{C_p} \right) = \log \left( \frac{C_f}{C_p} \right)
\]

For the above, \( Q \) is the water flow through the membrane module; \( C_f \) is the concentration of contaminants in the feed water; and \( C_p \) is the concentration of contaminants in the permeate.

UF membranes typically have an LRV of more than 8.0 log for bacteria, 7.0 log for *Giardia lamblia* cysts and *C. parvum* oocysts, and approximately 0.5 to 3.0 log for viruses (Jacangelo, et al., 1995; Kreißel, et al., 2012; Lu, et al., 2013). The Canadian Drinking Water Quality Guidelines (CDWQG) require a minimum LRV of 4.0 log for viruses. Because UF membranes generally are not able to consistently achieve 4.0 log of removal for viruses, it is often necessary that a secondary disinfection treatment be applied after UF treatment.

The low-pressure membranes used throughout North America for drinking water treatment are mostly polymeric and configured as hollow fibres (Figure 2). Hollow fibre membranes can be described as a very thin straw or tube, with an outside diameter ranging from 0.6 to 2.0mm and a wall thickness ranging from 0.1 to 0.6mm. The outside of the fibre is called shell and the inside is called lumen. Hollow fibres can be designed to filter water from the shell to the lumen (outside-in) or in the opposite direction (inside-out).

**Figure 2 – Images of hollow fibre membranes.**
(a) Scanning electron microscope image of end view of a hollow fibre membrane (b) Water permeating hollow fibre membranes (Crittenden, et al., 2012).
Conventional membrane systems generally operate with repeating cycles consisting of two stages: a permeation stage, during which clean water is produced and retained material accumulates on the membrane; followed by a hydraulic cleaning stage generally consisting of backwash and scouring, during which clean water, generally coupled with air scouring, is used to flush out the accumulated retained material. The permeation stage of a cycle generally lasts for 30 to 90 minutes, while hydraulic cleaning (i.e. backwash and scouring) generally lasts for 1 to 5 minutes.

However, some material cannot be effectively removed by hydraulic cleaning and accumulates on the membrane over repeated cycles. Chemical cleaning is used to periodically remove accumulated material that cannot be removed hydraulically. Low intensity chemical cleanings are typically done every few days to weeks and generally last several minutes to a few hours. Low intensity chemical cleaning usually consists of soaking, permeating and/or backwashing the membranes with a solution of cleaning agents (e.g. sodium hypochlorite, citric acid). High intensity chemical cleanings are performed a few times per year and generally last several hours to a few days. High intensity chemical cleaning usually consists of soaking the membranes with a highly concentrated cleaning solution and can also be enhanced with heat.

The accumulation of retained material on the membrane is referred to as membrane fouling. Membrane fouling can be characterized as reversible or irreversible. The fraction of fouling that can be reversed by hydraulic cleaning is referred to as hydraulically reversible fouling. The fraction that cannot be reversed by hydraulic cleaning but can be reversed by chemical cleaning is referred to as chemically reversible fouling. The fraction of fouling that cannot be reversed by hydraulic or by chemical cleaning is referred to as irreversible fouling.

Membrane fouling can also be characterized according to its mechanism. Three fouling mechanisms are traditionally considered to occur during membrane filtration of fresh water: pore blocking, pore adsorption and cake layer formation (AWWA, 2005). Pore blocking occurs when the entrance of a pore is completely sealed by a particle that has a similar size to that of the pore. As pore blocking occurs, the number of pores per membrane area (i.e. pore density) decreases. Pore adsorption occurs due to the adsorption of particles smaller than the pores on the pore walls. As pore adsorption occurs, the diameter of the pores decreases. Cake layer formation occurs due to the deposition of particles that are too large to enter the membrane pores. These particles accumulate on the membrane surface and form a ‘cake layer’.
The productivity of low-pressure membranes is usually expressed in terms of volumetric flux (J). The volumetric flux can be calculated using Equation 5.

\[ J = \frac{Q}{A_m} = \frac{(TMP)B}{\mu_w} \]

For the above, Q is the water flow through the membrane module; \( A_m \) is the membrane filtration area of the membrane module; TMP is the transmembrane pressure; B is the membrane permeability; and \( \mu_w \) is the dynamic viscosity of water.

Membrane flux is usually expressed in Lm\(^{-2}\)h\(^{-1}\) and TMP is usually expressed in kPa. For conventional membrane systems, membrane flux generally ranges from 30 to 170 Lm\(^{-2}\)h\(^{-1}\) and TMP generally ranges from 20 to 100 kPa.

As represented in Equation 5, the flux through a membrane module depends on the membrane permeability (B). When filtering clean water, the membrane permeability is equal to the intrinsic membrane permeability (B\(_m\)). The intrinsic membrane permeability depends on parameters such as tortuosity, length, diameter and density of the membrane pores. These parameters are difficult to measure. Therefore, the intrinsic membrane permeability is typically estimated by performing a clean water filtration test on the clean membrane module.

When filtering raw water, the membrane permeability decreases over time as fouling occurs. This is caused by the fouling-induced decrease in diameter and/or density of the membrane pores; or by the formation of a cake layer.

Membrane permeability is often used to present the results of laboratory experiments. Different membrane systems might operate under constant flux and variable TMP or under constant TMP and variable flux. The use of membrane permeability enables an easier comparison between the performance of membrane systems with different operating modes.

Membrane permeability depends on the dynamic viscosity of water, which changes with temperature. This makes it important to correct the permeability to a standard temperature, when comparing permeabilities measured at different temperatures. The membrane permeability corrected to a standard temperature (B\(_s\)) can be calculated using Equation 6.
Equation 6

\[ B_s = B_t \left( \frac{\mu_{w,t}}{\mu_{w,s}} \right) \]

For the above, \( B_t \) is the permeability at the measured temperature; \( \mu_{w,t} \) is the dynamic viscosity of water at the measured temperature; and \( \mu_{w,s} \) is the dynamic viscosity of water at the standard temperature.
2.2 Gravity-Driven Membrane Filtration

Gravity-driven membrane (GDM) filtration is a self-sustained process where an ultra-low permeate flux is maintained by hydrostatic pressure (i.e. gravity). The concept of GDM also usually implies that no fouling control measures (e.g. backwash, chemical cleaning, aeration) are used. Due to their ultra-low permeate flux and ease of operation, GDM systems are particularly suited for small scale applications, such as decentralized water treatment in small and remote communities. This type of system has been studied since 2010 and multiple studies have been published on process characteristics (Peter-Varbanets, et al., 2010; Peter-Varbanets, et al., 2011; Derlon, et al., 2012; Derlon, et al., 2013; Akhondi, et al., 2015; Fortunato, et al., 2016; Desmond, et al., 2018) and optimization (Derlon, et al., 2012; Kus, et al., 2013; Tang, et al., 2016a; Tang, et al., 2016b; Oka, et al., 2017; Ding, et al., 2018).

In the first study published on GDM systems, gravity-driven dead-end filtration was operated without any flushing or cleaning (Peter-Varbanets, et al., 2010). It was observed that a stable low flux ranging from 4 to 10 L/m²/h could be maintained for an extended period of time, in contrary to general expectations. A second study followed where the mechanisms of fouling and flux stabilization were investigated (Peter-Varbanets, et al., 2011). The data from both studies led to the conclusion that flux stabilization is related to the development of a biofilm layer on top of the membrane. As filtration occurs, the biofilm layer undergoes biologically induced structural changes that lead to the formation of heterogenous structures and channel networks. Flux stabilization is then reached when the decrease in resistance due to structural changes balances the increase in resistance due to deposition and irremovable fouling.

Of particular importance to the present study is the work by Oka et al. (2017). An alternative GDM system was developed and operated with a daily 1-hour relaxation period followed by a few minutes of passive air sparging and backwash. Neither practice added substantial mechanical or operational complexity to the system. It was observed that, compared to the same system operated without any fouling control measures, the permeability that could be sustained more than doubled. This system is referred to in the presented study as a PFC-GDM (passive fouling control – gravity-driven membrane) system. The PFC-GDM system was successfully operated as a pilot plant for approximately 1 year at the Eagle Lake Membrane Filtration Facility, West Vancouver, BC (Jain, 2019).
A summary of the main characteristics of GDM filtration, PFC-GDM filtration and conventional membrane filtration is presented in Table 1.

Table 1 – Summary of main characteristics of GDM filtration, PFC-GDM filtration and conventional membrane filtration.

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>GDM Filtration$^1$</th>
<th>PFC-GDM Filtration$^2$</th>
<th>Conventional Membrane Filtration$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source of pressure gradient to drive permeation</td>
<td>Hydrostatic head (i.e. gravity)</td>
<td>Hydrostatic head (i.e. gravity)</td>
<td>Mechanical device (e.g. pumps)</td>
</tr>
<tr>
<td>Transmembrane pressure</td>
<td>2 to 6.5kPa (20 to 65cm)</td>
<td>2 to 3.5kPa (20 to 35cm)</td>
<td>20 to 100kPa (2 to 10m)</td>
</tr>
<tr>
<td>Permeate flux</td>
<td>1 to 20Lm$^{-2}$h$^{-1}$</td>
<td>3 to 5Lm$^{-2}$h$^{-1}$</td>
<td>30 to 170Lm$^{-2}$h$^{-1}$</td>
</tr>
<tr>
<td>Presence of biofilm layer</td>
<td>Biofilm layer is present</td>
<td>Biofilm layer is present</td>
<td>Biofilm layer is not present</td>
</tr>
<tr>
<td>Fouling control measures</td>
<td>None</td>
<td>Daily 1h relaxation Daily 5min passive backwash Daily 5min passive aeration</td>
<td>Relaxation, backwash, scouring and chemical cleaning</td>
</tr>
<tr>
<td>Application</td>
<td>Small scale decentralized treatment</td>
<td>Small scale decentralized treatment</td>
<td>Large scale centralized treatment</td>
</tr>
</tbody>
</table>

$^1$Information sourced from Pronk, et al. (2018)
$^2$Information sourced from Khadem (2016), Oka, et al. (2017) and Jain (2019)
$^3$Information sourced from Crittenden, et al. (2012)
2.3 Virus Removal by Fouled Ultrafiltration Membranes

The primary mechanism of contaminant removal by UF membranes is size exclusion, or in other words, straining (Crittenden, et al., 2012). Therefore, UF membranes are capable of effectively removing contaminants that are larger than the pore size of the membrane. UF pore sizes vary, but generally range from 0.01 to 0.1µm (Baker, 2004). With sizes as small as 0.025µm, viruses can be somewhat removed by UF membranes depending on the pore size of the membrane (Crittenden, et al., 2012).

Additional virus removal mechanisms have been reported in the literature, namely electrostatic repulsion (Huang, et al., 2012; ElHadidy, et al., 2013; Gentile, et al., 2018) and hydrophobic adsorption (Madaeni, et al., 1995; Jacangelo, et al., 1995; Kreiβel, et al., 2012). Electrostatic repulsion occurs when virus and membrane have a similar surface charge, generating repulsive forces that prevent the virus from passing through the membrane. Hydrophobic adsorption occurs when viruses exhibit hydrophobic properties, i.e., they ‘dislike’ water and tend to adsorb to the also hydrophobic membrane surface (Crittenden, et al., 2012). In some cases, hydrophobic interactions can be strong enough to overcome electrostatic repulsion and lead to adsorption (Schijven & Hassanizadeh, 2000).

Depending on membrane pore size, surface charge and hydrophobicity, clean UF membranes can achieve LRVs of 0.5 to 3.0 log for viruses (Jacangelo, et al., 1995; Kreiβel, et al., 2012; Lu, et al., 2013). When fouled, however, UF membranes can achieve LRVs of up to 6.0 log (Jacangelo, et al., 1995; Lv, et al., 2006; Kreiβel, et al., 2012; Lu, et al., 2013; ElHadidy, et al., 2014; Yin, et al., 2015). A summary of relevant studies on virus removal by fouled membranes is presented in Table 2.

<table>
<thead>
<tr>
<th>Type of Treatment</th>
<th>Membrane Pore Size</th>
<th>Clean LRV</th>
<th>Fouled LRV</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drinking water</td>
<td>0.2µm</td>
<td>1.0</td>
<td>4.0</td>
<td>Jacangelo, et al. (1995)</td>
</tr>
<tr>
<td>Wastewater</td>
<td>0.2µm</td>
<td>1.7</td>
<td>6.3</td>
<td>Lv, et al. (2006)</td>
</tr>
<tr>
<td>Drinking water</td>
<td>0.02µm</td>
<td>2.5 to 3.0</td>
<td>5.5 to 6.0</td>
<td>Kreiβel, et al. (2012)</td>
</tr>
<tr>
<td>Wastewater</td>
<td>0.04µm</td>
<td>2.5</td>
<td>3.7</td>
<td>Lu, et al. (2013)</td>
</tr>
<tr>
<td>Drinking water</td>
<td>0.002 to 0.056µm</td>
<td>3.5</td>
<td>&gt; 5.0</td>
<td>ElHadidy, et al. (2014)</td>
</tr>
<tr>
<td>Drinking water</td>
<td>0.04µm</td>
<td>2.3</td>
<td>2.8</td>
<td>Yin, et al. (2015)</td>
</tr>
</tbody>
</table>
Cake layer fouling, in particular, has been associated with an increase in virus removal by UF membranes (Jacangelo, et al., 1995; Lv, et al., 2006; Lu, et al., 2013; Yin, et al., 2015). The reported mechanisms of virus removal by a cake layer are size exclusion (Jacangelo, et al., 1995; Crittenden, et al., 2012), adsorption (Lv, et al., 2006) and electrostatic repulsion (Lu, et al., 2013), depending on cake layer properties. These are the same removal mechanisms reported for clean UF membranes. It could be said that the cake layer can act as a secondary, more selective membrane capable of removing smaller contaminants.

The biofilm layer of passive membrane systems is an example of cake layer fouling. Besides contributing to the stabilization of flux during passive filtration (Peter-Varbanets, et al., 2011), the biofilm layer has been reported to contribute to permeate quality by improving the removal of organic carbon (Derlon, et al., 2014; Chomiak, et al., 2015; Oka, et al., 2017) and even of algae toxins (Kohler, et al., 2014). The contribution of the biofilm layer to the removal of viruses in passive membrane systems, however, is yet to be investigated. It is expected that the biofilm layer can contribute to virus removal, as other types of cake layer and membrane fouling in general have been observed to contribute in conventional membrane systems (Table 2).

There is also a scarcity of reports in the literature on the impact of membrane fouling on virus removal when an integrity breach occurs on the membrane. ElHadidy et al. (2014) observed that, in the presence of a suspected minor integrity breach, a fouled UF membrane was still capable of maintaining an LRV of approximately 4.0 log. This suggests that a foulant layer may be able to ‘repair’ breaches and thus prevent decreases in virus removal even when membrane integrity is compromised. However, no studies have been published with focus on the effects of a breach on the virus removal capacity of a fouled UF membrane.
2.4 Membrane Integrity Testing

A key aspect that has contributed to the success of membrane technology for drinking water treatment is the ability to test and verify the integrity of membranes (AWWA, 2005). Integrity testing allows for the verification of the contaminant removal capacity of the membrane system (i.e. the LRV) and facilitates the diagnosis and repair of integrity breaches. The integrity is compromised when a breach occurs on the membrane, mainly due to one of four reasons (Guo, et al., 2010):

1. Chemical corrosion such as oxidation;
2. Faulty installation and maintenance;
3. Membrane stress and strain from operating conditions, such as backwashing or excessive movement due to vigorous aeration; and/or
4. Damage by sharp objects not removed by pre-treatment.

In general, membrane integrity test methods can be divided into two main categories: direct methods and indirect methods. The direct methods refer to tests applied directly to the membrane or membrane module (e.g., pressure decay test, diffusive airflow test, bubble point test). Indirect methods are those applied to water quality parameters in the permeate (e.g., challenge test, particle counting, turbidity monitoring). The appropriate integrity testing methods for a certain membrane system will depend on the membrane type, manufacturer, system setup and local regulation (Guo, et al., 2010).

One of the most adopted guidelines in Canada is the USEPA Membrane Filtration Guidance Manual (MFGM), which was developed as part of the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR). According to the MFGM, for compliance with the LT2ESWTR, it is required that the membrane system undergo challenge testing (a type of indirect integrity testing) at least once, at its implementation. The LT2ESWTR also requires that direct integrity testing be conducted at least daily during operation. This frequency can be decreased if approved by relevant authorities, based on demonstrated process reliability, use of multiple barriers, or reliable process safeguards (USEPA, 2005).
2.4.1 Challenge Testing

Challenge tests (CTs) are used to determine the capacity of a membrane module to remove a certain target contaminant. Even though they are classified as an indirect integrity test method, CTs are a direct and straightforward method of measuring the LRV of a membrane module. During a CT, the membrane module is fed with a solution containing a known concentration of the challenge material. The challenge material can be the target contaminant itself or a surrogate. The solution is then filtered through the membrane module in operating conditions similar to those used in regular operation. Samples are collected from both feed water and permeate, and the LRV of the membrane module is calculated using Equation 4.

The target contaminant is selected based on the treatment goals of the membrane system. For compliance with the LT2ESWTR, the target contaminant should be *C. parvum*. However, it might be of interest to test for more than one contaminant. In that case, the most conservative of the contaminants should be selected – usually the smallest due to the straining aspect of membrane filtration processes. Table 3 presents some common target contaminants and their size ranges.

<table>
<thead>
<tr>
<th>Target Contaminant</th>
<th>Size Range (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enteric viruses</td>
<td>0.03 to 0.1</td>
</tr>
<tr>
<td>Fecal coliform</td>
<td>1 to 4</td>
</tr>
<tr>
<td><em>C. parvum</em></td>
<td>3 to 7</td>
</tr>
<tr>
<td>Giardia</td>
<td>7 to 15</td>
</tr>
</tbody>
</table>

Table 3 – Potential target contaminants and their respective size ranges (USEPA, 2005).

Even though challenge testing with the target contaminant itself would be ideal, it might not always be feasible due to cost or health concerns. In that case, a surrogate may be used as the challenge material. The surrogate should have similar characteristics to those of the target contaminant, such as particle size, shape and surface charge. It should also be safe to handle, easy to measure and cost effective. A common surrogate used for enteric viruses is the MS2 bacteriophage, as it has similar size and shape to the poliovirus and hepatitis virus, it is easy to grow and count, and it is not harmful to humans or animals (Wu, et al., 2017). According to the MFGM, MS2 can also be used as a conservative surrogate for *C. parvum*, due to its small size (USEPA, 2005).

The concentration of the CT solution is based on the target LRV to be demonstrated and the detection limit (DL) of the challenge particulate. The target LRV is usually selected as 6.5 log, as
this is the maximum LRV that can be demonstrated under the LT2ESWTR. The maximum CT solution concentration (Cf max) for a target LRV of 6.5 log can be calculated using Equation 7 (USEPA, 2005).

Equation 7

\[ C_{f_{max}} = 3.16 \times 10^6 \times DL \]

For the above, DL is the detection limit of the challenge particulate.

The MFGM recommends that the CT solution be prepared with high quality water with low concentration of suspended solids, minimizing the interference of other substances that might be present in the water (USEPA, 2005). It is also recommended that the test be conducted at maximum design flux and that flux and TMP be monitored during the test (USEPA, 2005).

As previously mentioned, CTs are a straight forward method of measuring the LRV of a membrane module. However, one of the main disadvantages of the CT is that it cannot reflect the membrane integrity in real time, due to the time required for the microbial analysis (24h to 48h). Therefore, for routine integrity testing in full-scale operation, pressure decay tests (PDTs) are preferred as they can provide a real time estimation of the LRV (see section 2.4.2).

2.4.2 Pressure Decay Testing

The PDT is one of the most commonly used methods of direct integrity testing (Guo, et al., 2010). PDTs are used to detect the presence of breaches or defects on the membrane module. If a breach is present, the results from the PDT can be used to estimate the LRV of the breached membrane module. Unlike CTs, PDTs do not allow for a straight forward determination of the LRV. In order to estimate the LRV from the results of a PDT, several assumptions and calculations must be made.

A PDT is performed by pressurizing one side of a wetted membrane module with air while the other side remains submerged in water. The source of pressure is isolated and the pressure within the system is monitored. A pressure decay rate (PDR) is then calculated based on the initial and final pressures within the system and the duration of the test (Equation 8).

Equation 8

\[ PDR = \frac{P_i - P_f}{t} \]
For the above, \( P_i \) is the pressure at the beginning of the test; \( P_f \) is the pressure at the end of the test; and \( t \) is the test duration.

Typically, the test duration should be enough to allow for a 10% decrease in \( P_i \).

From the PDR, it is possible to estimate the air flow through the membrane module (\( Q_{\text{air}} \)) using Boyle’s Law (Equation 9).

\[
\text{Equation 9} \\
P_1 V_1 = P_2 V_2 \rightarrow \frac{P_i - P_f}{t} V_s = P_{\text{atm}} \frac{\Delta V_{\text{air}}}{t} \rightarrow (PDR)V_s = P_{\text{atm}}(Q_{\text{air}}) \rightarrow \\
Q_{\text{air}} = \frac{(PDR)V_s}{P_{\text{atm}}} 
\]

For the above, PDR is the pressure decay rate; \( V_s \) is the volume of the pressurized system; and \( P_{\text{atm}} \) is the atmospheric pressure.

During a PDT, a baseline air flow is expected due to air diffusion through the wetted membrane pores. If the observed air flow is above the baseline, it is likely that the membrane is breached. The diffusive air flow (\( Q_{\text{air,d}} \)) of a membrane module can be measured by performing a PDT on the intact membrane module.

When a PDT is performed on a breached membrane module, the \( Q_{\text{air}} \) measured during the test corresponds to the sum of the diffusive air flow (\( Q_{\text{air,d}} \)) and the air flow through the breach (\( Q_{\text{air,b}} \)). Therefore, \( Q_{\text{air,b}} \) can be calculated as the difference between total air flow and diffusive air flow through the membrane module (Equation 10).

\[
\text{Equation 10} \\
Q_{\text{air},b} = Q_{\text{air},t} - Q_{\text{air},d}
\]

For the above, \( Q_{\text{air},t} \) is the total air flow through a breached membrane module; and \( Q_{\text{air,d}} \) is the diffusive air flow (measured from the intact membrane module).

The air flow through a breach can change with air temperature. When comparing the results of PDTs performed at different temperatures, it is important to calculate the corrected air flow through the breach (\( Q_{\text{air,b,s}} \)) at a standard temperature using Equation 11 (Guibert & Colling, 2011).
Equation 11

\[ Q_{\text{air,b,s}} = \frac{Q_{\text{air,b,m}} T_s}{T_m} \]

For the above, \( Q_{\text{air,b,m}} \) is the air flow through the breach at the measured air temperature; \( T_s \) (in K) is the standard air temperature; and \( T_m \) (in K) is the measured air temperature.

The size of the smallest breach that can be detected by a PDT is directly related to the pressure applied during the test. The minimum pressure required to detect a breach of a certain size can be calculated using the bubble point theory. According to the bubble point theory, there is a minimum pressure \( (P_{\text{min}}) \) required to overcome the capillary forces and surface tension of a liquid in a fully wetted membrane, and force air flow through the membrane pores or through a breach. This minimum pressure can be derived from Jurin’s Law, which describes the height of a liquid column in a capillary. Thus, \( P_{\text{min}} \) can be calculated using Equation 12.

Equation 12

\[ P_{\text{min}} = 0.58 k \sigma \cos \theta \frac{D_{b,\text{min}}}{D_{b,\text{min}}} + B P_{\text{max}} \]

For the above, \( P_{\text{min}} \) is in psi; 0.58 is a unit conversion factor; \( k \) is a pore shape correction factor (dimensionless); \( \sigma \) is the surface tension at the air-liquid interface (dynes/cm); \( \theta \) is the liquid-membrane contact angle (degrees); \( D_{b,\text{min}} \) is the diameter of the smallest breach to be detected (\( \mu \)m); and \( B P_{\text{max}} \) is the maximum backpressure (e.g. static water head) on the membrane module during the test.

Recommendations for \( k \), \( \sigma \) and \( \theta \) values can be found in the MFGM or acquired from the membrane manufacturer (USEPA, 2005).

The test pressure should not exceed the \( P_{\text{min}} \) equivalent to the size of the membrane pores, nor the maximum pressure the membrane module can withstand according to the manufacturer. Therefore, PDTs are only capable of detecting breaches of a relatively large size. To meet the requirements of the LT2ESWT rule, the test pressure should be enough to detect a breach of at least 3\( \mu \)m (size of a \( C. \text{parvum} \) oocyst). Using Equation 12 and values commonly used by the industry, the minimum test pressure to detect a 3\( \mu \)m breach is 8psi, assuming a backpressure of zero (USEPA, 2005).
2.4.2.1 Estimation of Breach Diameter and Expected Air Flow Through Breach from Pressure Decay Tests

It is possible to estimate the diameter of a breach based on the air flow detected during the PDT. Wang et al. (2015) developed a mathematical model that considers the breach to behave like a thin-walled orifice and assumes that the air flow through a breach is an unsteady adiabatic flow process for high-pressure air flow. It combines the energy equation and the Ideal Gas Law to calculate the breach diameter ($D_b$). The model proposed by Wang et al. (2015) is represented by Equation 13.

$$D_b = 2 \frac{Q_{air,b} P_{atm} (P_{test}/P_{atm})^{1/\kappa}}{\pi P_{test} \sqrt{\frac{2\kappa}{\kappa - 1} RT \left[ 1 - \left( \frac{P_{atm}}{P_{test}} \right)^{\frac{\kappa - 1}{\kappa}} \right]}}$$

For the above, $Q_{air,b}$ is the air flow through the breach; $P_{atm}$ is the atmospheric pressure; $P_{test}$ is the average test pressure; $\kappa$ is the adiabatic constant; $R$ is the universal gas constant; and $T$ is the air temperature.

Equation 13 can be rearranged for $Q_{air,b}$ and used to estimate the expected air flow through a breach of a certain diameter during a PDT (Equation 14).

$$Q_{air,b} = \frac{D_b^2 \pi P_{test} \left[ \frac{2\kappa}{\kappa - 1} RT \left[ 1 - \left( \frac{P_{atm}}{P_{test}} \right)^{\frac{\kappa - 1}{\kappa}} \right] \right]}{4P_{atm} (P_{test}/P_{atm})^{1/\kappa}}$$

For the above, $D_b$ is the breach diameter; $P_{atm}$ is the atmospheric pressure; $P_{test}$ is the average test pressure; $\kappa$ is the adiabatic constant; $R$ is the universal gas constant; and $T$ is the air temperature.

An alternative approach for estimating breach diameter is to model the breach as a tube. The Darcy-Weisbach equation can then be used to convert air flow into breach diameter. A detailed description is provided in Appendix A, and a comparison between these two approaches is provided in Appendix B.
2.4.2.2 Estimation of Water Flow Through Breach from Pressure Decay Tests

It is possible to estimate the water flow through a breach ($Q_{\text{water},b}$) based on the air flow through the breach ($Q_{\text{air},b}$) measured during a PDT. The approach described in the American Society for Testing and Materials (ASTM) Standard models the breach as a tube and uses the Darcy-Weisbach equation to calculate the head loss through the breach for both air and liquid flows (ASTM Standard D6908, 2003). The resulting equation from the ASTM approach is Equation 15 (for derivation, see Appendix C).

\[
Q_{\text{water},b} = \frac{2(TMP)\mu_aP_{\text{atm}}Q_{\text{air},b}}{\mu_w(P_{\text{test}}^2 - P_{\text{atm}}^2)}
\]

For the above, TMP is the transmembrane pressure; $\mu_a$ is the dynamic viscosity of air; $P_{\text{atm}}$ is the atmospheric pressure; $Q_{\text{air},b}$ is the air flow through the breach; $\mu_w$ is the dynamic viscosity of water; and $P_{\text{test}}$ is the average test pressure.

Equation 15 is used in the estimation of the LRV from the results of the PDT (see section 2.4.2.3).

2.4.2.3 Estimation of Log Removal Value from Pressure Decay Tests

According to Equation 1 (see section 2.1), the LRV of a membrane module depends on the mass flux of contaminants in the feed water ($M_f$) and the mass flux of contaminants in the permeate ($M_p$).

\[
LRV = \log\left(\frac{M_f}{M_p}\right)
\]

For the above, $M_f$ is the mass flux of contaminants in the feed water; and $M_p$ is the mass flux of contaminants in the permeate.

According to Equation 2 (see section 2.1), $M_f$ is the product of the total water flow through the membrane module and the concentration of contaminants in the feed water.

\[
M_f = QC_f
\]

For the above, Q is the water flow through the membrane module; and $C_f$ is the concentration of contaminants in the feed water.
For a breached membrane module, contaminants enter the permeate stream by flowing through the breach and, for contaminants that are not effectively removed, also by flowing through the intact membrane. Therefore, $\dot{M}_p$ is the sum of the mass flux of contaminants through the breach and the mass flux of contaminants not removed by the intact area of the membrane (Equation 16).

Equation 16

$$\dot{M}_p = \dot{M}_b + \dot{M}_{nr}$$

For the above, $\dot{M}_b$ is the mass flux of contaminants through the breach; and $\dot{M}_{nr}$ is the mass flux of contaminants not removed by the intact area of the membrane.

$\dot{M}_b$ is the product of the water flow through the breach and the concentration of contaminants flowing through the breach (Equation 17).

Equation 17

$$\dot{M}_b = Q_{\text{water},b} C_b$$

For the above, $Q_{\text{water},b}$ is the water flow through the breach; and $C_b$ is the concentration of contaminants flowing through the breach.

$\dot{M}_{nr}$ is the product of the water flow being treated by the intact area of the membrane and the concentration of contaminants not removed by the intact area of the membrane (Equation 18).

Equation 18

$$\dot{M}_{nr} = Q_{\text{int}} C_{nr}$$

For the above, $Q_{\text{int}}$ is the water flow being treated by the intact area of the membrane; and $C_{nr}$ is the concentration of contaminants not removed by the intact area of the membrane.

Thus, the mass flux of contaminants in the permeate of a breached membrane module ($\dot{M}_{p,b}$) can be calculated by Equation 19, which combines Equation 16, Equation 17 and Equation 18.

Equation 19

$$\dot{M}_{p,b} = Q_{\text{water},b} C_b + Q_{\text{int}} C_{nr}$$
For the above, \( Q_{\text{water},b} \) is the water flow through the breach; \( C_b \) is the concentration of contaminants flowing through the breach; \( Q_{\text{int}} \) is the water flow being treated by the intact area of the membrane; and \( C_{nr} \) is the concentration of contaminants not removed by the intact area of the membrane.

Therefore, the LRV of a breached membrane module can be calculated using Equation 20.

\[
LRV = \log \left( \frac{M_f}{M_{p,b}} \right) = \log \left( \frac{\dot{M}_f}{M_b + M_{nr}} \right) = \log \left( \frac{Q_{\text{water},b}C_b + Q_{\text{int}}C_{nr}}{Q_{\text{water},b}C_b + Q_{\text{int}}C_{nr}} \right)
\]

For the above, \( \dot{M}_f \) is the mass flux of contaminants in the feed water; \( M_{p,b} \) is the mass flux of contaminants in the permeate of a breached membrane module; \( M_b \) is the mass flux of contaminants flowing through the breach; \( M_{nr} \) is the mass flux of contaminants not removed by the intact area of the membrane; \( Q \) is the water flow through the membrane module; \( Q_{\text{water},b} \) is the water flow through the breach; \( Q_{\text{int}} \) is the water flow being treated by the intact membrane; \( C_f \) is the concentration of contaminants in the feed water; \( C_b \) is the concentration of contaminants flowing through the breach; and \( C_{nr} \) is the concentration of contaminants not removed by the intact area of the membrane.

As previously discussed (see section 2.4.2), PDTs are only capable of detecting breaches of a relatively large size due to limitations in the pressure that can be applied during the test. As a consequence, PDTs can only estimate the LRV of contaminants that would be effectively removed by an intact membrane. Therefore, when estimating the LRV of a breached membrane module from the results of a PDT, the concentration of contaminants not removed by the intact area of the membrane (\( C_{as} \)) can be considered zero (ASTM Standard D6908, 2003). Thus, Equation 20 can be rewritten as Equation 21.

\[
LRV = \log \left( \frac{\dot{M}_f}{M_{p,b}} \right) = \log \left( \frac{Q_{\text{water},b}C_b}{Q_{\text{water},b}C_b + Q_{\text{int}}C_{nr}} \right)
\]

For the above, \( \dot{M}_f \) is the mass flux of contaminants in the feed water; \( M_{p,b} \) is the mass flux of contaminants in the permeate of a breached membrane module; \( Q \) is the water flow through the membrane module; \( Q_{\text{water},b} \) is the water flow through the breach; \( C_f \) is the concentration of contaminants in the feed water; \( C_b \) is the concentration of contaminants flowing through the breach; and \( C_{nr} \) is the concentration of contaminants not removed by the intact area of the membrane.
contaminants in the feed water; and \( C_b \) is the concentration of contaminants flowing through the breach.

For a clean breached membrane module, the concentration of contaminants flowing through the breach can be assumed to be equal to the concentration of contaminants in the feed water (ASTM Standard D6908, 2003). Therefore, Equation 21 can be rewritten as Equation 22.

\[
L_{RV} = \log \left( \frac{Q_C f}{Q_{water,b} C_f} \right) = \log \left( \frac{Q}{Q_{water,b}} \right)
\]

For the above, \( Q \) is the water flow through the membrane module; and \( Q_{water,b} \) is the water flow through the breach.

For some hydraulic configurations of membrane systems, the term \( Q_{water,b} \) in Equation 22 must be multiplied by a volumetric concentration factor (VCF). The VCF is a dimensionless term that accounts for the increase in the concentration of contaminants that can occur on the feed side of the membrane. However, for membrane systems that operate in deposition mode (i.e. dead-end filtration), such as GDM systems, the VCF is 1.0 (USEPA, 2005) and therefore was not included in Equation 22.

Equation 22, along with Equation 15 (see section 2.4.2.2), are part of the ASTM approach for estimating the LRV from the results of a PDT. It is important to note that the ASTM approach was developed for conventional membrane systems, which generally do not have a biofilm layer. As previously discussed (see section 2.3), it is expected that the biofilm layer can contribute to contaminant removal and therefore positively affect the LRV of passive membrane systems. However, there is a scarcity of reports in the literature on the impact of membrane fouling on pressure decay testing. It is not known whether or not the biofilm layer can affect the estimation of the LRV from the results of a PDT. Therefore, it is necessary to investigate if PDTs are an appropriate method of estimating the LRV of passive membrane systems.
3.0 OBJECTIVES

The objectives of the present study were to:

1. Quantify the contribution of the biofilm layer to the virus removal capacity of passive membrane systems, both intact and with integrity breaches of different sizes; and
2. Assess the use of PDTs as a method of estimating the LRV of breached passive membrane systems.
4.0 MATERIALS AND METHODS

4.1 Experimental Set-up

The experimental set-up used for the present study consisted of 3 identical laboratory scale UF membrane systems operated in parallel (see section 4.1.1). The set-up was built in the Filtration Technology Laboratory at the University of British Columbia. Each membrane system contained 1 membrane tank with 4 membrane modules (see section 4.1.2). Three out of the 4 modules were breached (see section 4.1.5). The 3 membrane systems were fed with a mixture of pond and tap water (see section 4.1.3). The membrane systems were operated continuously for 116 days (Dec. 17, 2018 to Apr. 11, 2019), each with a different set of operating conditions (see section 4.1.4).

4.1.1 Membrane Systems

The 3 membrane systems used in the present study were referred to as Passive System 1, Passive System 2 and Conventional System. A schematic representation of the membrane systems is presented in Figure 3. All membrane systems were physically identical, therefore only Passive System 1 is illustrated in detail in Figure 3. The membrane systems were fed by 4 interconnected feed water tanks (25L each). The backwash water was provided to all membrane systems by a backwash tank (5L) filled with distilled water. Each membrane system consisted of:

1. 1 cylindrical membrane tank with 4 UF membrane modules (see section 4.1.2);
2. 1 permeate tank (20L);
3. 1 drain tank (20L);
4. 1 compressed air line for aeration;
5. 1 pressure regulator on the compressed air line;
6. 1 air flow regulator on the compressed air line;
7. 1 level switch for control of water level in the membrane tanks;
8. 2 normally closed ball valves, 1 on the feed tubing and 1 on the drain tubing;
9. 1 normally closed solenoid valve on the compressed air line;
10. 2 Masterflex peristaltic pumps, 1 for permeation and 1 for backwashing;
11. 4 pressure transducers, located on the tubing between the membrane modules and the permeate and backwash pumps, connected to a laptop for TMP recording (see section 4.2.1.2); and
12. A cyclic timer connected to valves and pumps for control of operating conditions (see section 4.1.4).

Figure 3 – Schematic representation of membrane systems of experimental set-up.
4.1.2 Membrane Tanks and Modules

The membrane tanks used in the present study consisted of open-top acrylic cylinders with an internal diameter of 5cm and a height of 30cm (Figure 4). The water level in the membrane tanks was controlled by a level switch, which controlled the feed water valve. The level was maintained at a minimum of approximately 25cm, so that the membrane modules were always completely submerged.

![Image of one membrane tank used in the experimental set-up.](image)

Figure 4 – Image of one membrane tank used in the experimental set-up.

As illustrated in Figure 3, the membrane tank of each system contained 4 membrane modules: 1 membrane module was intact and 3 were breached. The breached membrane modules had 1 breach
each, with diameters of approximately 20, 40 and 180µm. The procedure used for membrane breaching is detailed in section 4.1.5.

Custom hollow-fibre UF membrane modules were used. The membrane specifications are presented in Table 4.

Table 4 – Specifications of membranes used in the experimental set-up.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manufacturer</td>
<td>Suez Water Technologies &amp; Solutions</td>
</tr>
<tr>
<td>Model</td>
<td>ZeeWeed 500®</td>
</tr>
<tr>
<td>Type</td>
<td>Ultrafiltration</td>
</tr>
<tr>
<td>Nominal Pore Size</td>
<td>0.04µm</td>
</tr>
<tr>
<td>Material</td>
<td>Polyvinylidene fluoride (PVDF)</td>
</tr>
<tr>
<td>Flow Path</td>
<td>Outside-in</td>
</tr>
<tr>
<td>Surface Properties</td>
<td>Non-ionic &amp; hydrophilic</td>
</tr>
<tr>
<td>Internal Diameter</td>
<td>0.8mm</td>
</tr>
<tr>
<td>External Diameter</td>
<td>1.9mm</td>
</tr>
</tbody>
</table>

Each membrane module was composed of 3 hollow fibres, each 21cm in length. The top 2cm of the fibres were inserted into a 10cm firm EVA tube (¼” OD, 0.17” ID) and sealed in place with epoxy, making sure that the fibres were not plugged; the bottom 1cm was inserted into a 1cm tube and completely sealed with epoxy (Figure 5). The final exposed fibre length was 18cm, resulting in a membrane filtration area of 32.2cm² per membrane module. Prior to potting, the hollow fibres were soaked in deionized (DI) water for 24h to remove the preserving agents. After potting, the membrane modules were cleaned as described in section 4.3.1.1.

Figure 5 – Image of potted membrane module used in the experimental set-up.
4.1.3 Feed Water

The feed water provided to all 3 membrane systems consisted of a mixture of tap water and pond water. The pond water was collected from Jericho Pond, located at Jericho Beach, Vancouver, BC. Jericho Pond was chosen to maintain consistency with the previous study by Oka, et al. (2017). Jericho Pond is a run-off water pond that serves as habitat for ducks, birds and fish and it is subjected to seasonal quality changes, making it a good representation of a typical surface water source.

The dates of pond water collection are presented in Table 5, as well as the total and dissolved organic carbon concentrations (TOC and DOC). At the time of collection, the water was filtered through a 1mm screen to remove large suspended solids. The collected water was then stored in the dark at T < 4°C until use. Prior to use, the water was filtered through a 1.5µm glass fibre filter (VWR, Grade 691), tested for TOC and DOC, warmed to room temperature and mixed with tap water. The mixing ratio was selected to achieve a target final DOC concentration of 4 to 5mg/L.

<table>
<thead>
<tr>
<th>Date of Collection</th>
<th>TOC (mg/L)</th>
<th>DOC (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oct. 19, 2018</td>
<td>9.94</td>
<td>9.86</td>
</tr>
<tr>
<td>Dec. 21, 2018</td>
<td>7.19</td>
<td>6.70</td>
</tr>
<tr>
<td>Jan. 15, 2019</td>
<td>6.30</td>
<td>6.26</td>
</tr>
<tr>
<td>Feb. 15, 2019</td>
<td>7.65</td>
<td>7.39</td>
</tr>
<tr>
<td>Mar. 19, 2019</td>
<td>7.74</td>
<td>6.94</td>
</tr>
</tbody>
</table>

4.1.4 Operating Conditions

In order to assess the impact of the biofilm layer on the integrity test performance of passive membrane systems, Passive System 1 and Passive System 2 were operated under conditions that simulate passive filtration. As a control, the Conventional System was operated under conditions that simulate conventional filtration. To assess the impact of frequent PDTs on the biofilm layer, Passive System 1 was subjected to weekly PDTs, while Passive System 2 was not (see section 4.2.2.2). To enable a direct comparison of passive and conventional operation, Passive System 1 and Passive System 2 were not designed and operated as gravity-driven membrane systems. Instead, Passive
System 1 and Passive System 2 were operated under constant flux with vacuum-driven permeation. This was not expected to affect the results of the present study, as it has been reported that, under low flux conditions, the fouling behaviour of systems operated under constant pressure and constant flux are similar (Miller, et al., 2014; Oka, 2015).

The operating conditions used throughout the present study are summarized in Table 6. The operating conditions used for Passive System 1 and Passive System 2 were chosen in accordance with those used for the PFC-GDM system (Oka, et al., 2017; Jain, 2019). Passive System 1 and Passive System 2 were operated on a 24h cycle. Permeation lasted 22h55min, with a permeation flux of 5Lm⁻²h⁻¹. After permeation, the membrane systems were subjected to a 1h relaxation period, followed by 5 minutes of aeration and backwash. The membrane tanks were then drained and the cycle restarted. The membrane systems were not subjected to any kind of chemical cleaning. The backwash flux was 20Lm⁻²h⁻¹ (see section 4.1.4.1) and the air flow was 4L/min (see section 4.1.4.2).

The operating conditions used for the Conventional System were chosen in accordance to what is normally used in the field for the ZeeWeed 500® membranes, as well as for conventional membrane systems in general (Crittenden, et al., 2012; SUEZ Water Technologies & Solutions, 2019). The Conventional System was operated on a 1h cycle. Permeation lasted 55min, with a permeation flux of 40Lm⁻²h⁻¹. After permeation, the membrane system was subjected to 5 minutes of aeration and backwash. The membrane tank was then drained and the cycle restarted. The membrane system was subjected to routine chemical cleaning 3 times per week (see section 4.3.1.2). The backwash flux was the same as for permeation (40Lm⁻²h⁻¹) and the air flow was the same as for the passive systems (4L/min).

To control the filtration cycles, each membrane system was equipped with a custom timer, which controlled the valves and pumps of the system. For timer programming, see Appendix D.
Table 6 – Operating conditions of Passive System 1, Passive System 2 and Conventional System.

<table>
<thead>
<tr>
<th>Operational Parameter</th>
<th>Membrane System</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Passive System 1&lt;sup&gt;1&lt;/sup&gt;</td>
</tr>
<tr>
<td>Mode of operation</td>
<td>Passive</td>
</tr>
<tr>
<td>Total cycle duration</td>
<td>24h</td>
</tr>
<tr>
<td>Permeation flux</td>
<td>5Lm&lt;sup&gt;-2&lt;/sup&gt;h&lt;sup&gt;-1&lt;/sup&gt;</td>
</tr>
<tr>
<td>Relaxation</td>
<td>1h at end of cycle</td>
</tr>
<tr>
<td>Aeration and backwash</td>
<td>5min after relaxation, at end of cycle</td>
</tr>
<tr>
<td>Aeration flow rate</td>
<td>4Lmin&lt;sup&gt;-1&lt;/sup&gt;</td>
</tr>
<tr>
<td>Backwash flux</td>
<td>20Lm&lt;sup&gt;-2&lt;/sup&gt;h&lt;sup&gt;-1&lt;/sup&gt;</td>
</tr>
<tr>
<td>Chemical cleaning</td>
<td>None</td>
</tr>
<tr>
<td>Pressure decay testing</td>
<td>Once per week</td>
</tr>
</tbody>
</table>

<sup>1</sup>Operating conditions chosen based on Oka et al. (2017) and Jain (2019).

<sup>2</sup>Operating conditions chosen based on Crittenden et al. (2012) and SUEZ Water Technologies & Solutions (2019).

4.1.4.1 Backwash Flux of Passive System 1 and Passive System 2

In passive membrane systems, a vacuum can be generated as liquid exits the membrane tanks during the process of draining (Jain, 2019). The vacuum provides the driving force to 'pull' permeate back into the membrane tank through the membrane fibres, thus performing a passive backwash. The magnitude of the vacuum depends on the head difference between the elevation of liquid in the permeate tank and the elevation of the discharge port in the membrane tank. Therefore, the
backwash flux varies during the backwash process and depends on the design of the membrane module.

Because the membrane systems used in the present study were not operated using gravity (see section 4.1.4), a pump was used to provide the driving force for backwashing. The backwash water was provided by a backwash tank filled with distilled water (see section 4.1.1).

For the present study, the backwash flux was chosen based on the pilot scale PFC-GDM system (Jain, 2019). For the pilot system, the height difference between the maximum elevation of permeate and the drain valve of the membrane tank is approximately 4 times greater than the head used for permeation. As presented in Equation 5 (see section 2.1), flux is directly proportional to TMP. Therefore, a 4-fold increase in TMP can be expected to result in a 4-fold increase in flux. Thus, a conservative constant backwash flux of 4 times the permeation flux (i.e. 4 x 5 = 20Lm⁻²h⁻¹) was chosen for the passive membrane systems of the present study.

4.1.4.2 Aeration of Passive System 1 and Passive System 2

Similar to the backwash, the aeration in passive membrane systems can be generated passively due to the vacuum that can be created inside the membrane tanks during the draining process (Jain, 2019). An easy analogy to this process is what happens when a full water bottle is inverted and drained. As the water leaves, air enters the bottle in ‘pulses’ forming air bubbles. The same principle can be applied to passive membrane systems in order to generate a passive aeration while the membrane tanks are draining. The air flow and the size of the bubbles created during aeration of passive membrane systems depend on elements such as the volume of the membrane tanks and the diameter of the pipes, tubes and valves that compose the system.

Because the membrane systems used in the present study were not operated using gravity (see section 4.1.4), it was not possible to create the aeration ‘passively’. Instead, air was added from an external source. The air flow was chosen based on the PFC-GDM systems previously studied by Oka et al. (2017) and on the pilot PFC-GDM system studied by Jain (2019). According to Serra (1999), the optimum air flow (Q_{air, opt}) of a membrane module can be estimated based on the packing density or the diameter of another membrane module, using Equation 23 or Equation 24, respectively.
\[
\frac{Q_{\text{air,opt}1}}{Q_{\text{air,opt}2}} = \frac{1 - \varphi_2}{1 - \varphi_1}
\]

Equation 23

\[
\frac{Q_{\text{air, opt,1}}}{Q_{\text{air, opt,2}}} = \sqrt{\frac{D_{V1}}{D_{V2}}}
\]

Equation 24

For the above, \(\varphi\) is the packing density and \(D_V\) is the vessel diameter.

Using the characteristics of the membrane modules used by Oka et al. (2017), the optimum air flow for the membrane modules of the present study was estimated to be 3.88L/min (Equation 23) or 2.04L/min (Equation 24). The air flow used in the pilot system studied by Jain (2019) was approximately 4L/min. Therefore, a conservative value of 4L/min was chosen for the present study.

### 4.1.5 Membrane Breaching

The membranes were breached using a Laser Micromachining Workstation (IX-280-ML, IPG Photonics) located at 4D Labs, at Simon Fraser University, Burnaby, BC. Previous studies that dealt with membrane integrity testing reported the successful use of either needle puncturing (Deluhery & Rajagopalan, 2008; Suh, et al., 2011; Choi, et al., 2011) or laser drilling (Giglia & Krishnan, 2008; Brehant, et al., 2010; Songlin, et al., 2014) to create breaches in the membranes. Laser drilling was the chosen method for the present study, because it allows for smaller breach diameters and higher precision than needle puncturing.

The two main laser parameters that could be adjusted to create breaches of different diameters were the laser aperture and drilling time. The laser aperture defines the diameter of the laser beam, which influences the diameter of the breach. The drilling time defines for how long the beam is directed at the membrane surface, which influences both the diameter of the breach and its depth.

For the present study, the goal was to completely breach one wall of the membrane fibre, from surface to lumen (Figure 6), and to create breaches of approximately 20, 40 and 180\(\mu\text{m}\) of diameter. To define the laser configurations (i.e. combination of aperture + time) that produced the desired breaches, two breaching experiments were performed and are detailed in Appendix E.
Figure 6 – Schematic representation of the cross-section of a breached membrane fibre.

The laser configurations selected from the results of the breaching experiments are presented in Table 7.

Table 7 – Laser configurations used for membrane breaching.

<table>
<thead>
<tr>
<th>Target breach diameter (µm)</th>
<th>Laser Aperture (µm)</th>
<th>Drilling Time (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td>40</td>
<td>500</td>
<td>1</td>
</tr>
<tr>
<td>180</td>
<td>3000</td>
<td>1</td>
</tr>
</tbody>
</table>

Prior to breaching, all membrane modules were submitted to PDTs to establish their diffusive air flows (see section 4.2.2.1). Nine membrane modules were then taken to 4D Labs for breaching (stored in leak-proof plastic containers filled with DI water). The remaining 3 membrane modules were kept intact. Microscope images (taken with the laser built-in microscope) of typical breaches of approximately 20, 40 and 180µm are presented in Figure 7. The dimensions of each breach (a x b according to Figure 7c) were measured using the laser built-in microscope.

After breaching, the 9 breached membrane modules were again submitted to PDTs. Using the air flow measured during the PDTs, the breach diameters were estimated using Equation 13 (see section 2.4.2.1). Table 8 summarizes the target, measured and estimated breach dimensions for each membrane module of Passive System 1, Passive System 2 and Conventional System.
Figure 7 – Microscope images of typical breaches.
(a) 20µm breach (b) 40µm breach (c) 180µm breach

Table 8 – Target, measured and estimated breach dimensions of membrane modules used in the experimental set-up.

<table>
<thead>
<tr>
<th>Membrane System</th>
<th>Membrane Module</th>
<th>Target Breach Diameter (µm)</th>
<th>Measured Breach Dimensions: a x b (µm)&lt;sup&gt;1&lt;/sup&gt;</th>
<th>Estimated Breach Diameter (µm)&lt;sup&gt;2&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Passive System 1</td>
<td>1</td>
<td>Intact</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>20</td>
<td>21 x 21</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>40</td>
<td>44 x 42</td>
<td>43</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>180</td>
<td>180 x 190</td>
<td>179</td>
</tr>
<tr>
<td>Passive System 2</td>
<td>1</td>
<td>Intact</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>20</td>
<td>21 x 21</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>40</td>
<td>47 x 48</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>180</td>
<td>150 x 170</td>
<td>178</td>
</tr>
<tr>
<td>Conventional System</td>
<td>1</td>
<td>Intact</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>20</td>
<td>21 x 21</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>40</td>
<td>44 x 40</td>
<td>43</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>180</td>
<td>168 x 170</td>
<td>177</td>
</tr>
</tbody>
</table>

NA: Not applicable.

<sup>1</sup>Dimentions measured according to Figure 7c.

<sup>2</sup>Diameters estimated from PDT results using Equation 13 (see section 2.4.2.1).
4.2 Analytical Methods

Several analytical methods were applied throughout the study in order to monitor the overall performance of the membrane systems and membrane integrity. Overall performance parameters that were monitored included membrane permeability (i.e. measurement of permeate flux and temperature and TMP monitoring), organic matter removal (i.e. total and dissolved organic carbon measurements) and ATP concentration in system components. Membrane integrity testing methods that were used included challenge testing and pressure decay testing.

4.2.1 Overall Performance Monitoring

4.2.1.1 Permeate Flux, Permeate Temperature and Permeate Sampling
Permeate flux was measured 2 times per week on Mondays and Thursdays. The permeate from each membrane module was collected throughout an entire cycle in previously weighted Erlenmeyer flasks (TP-1502 scale, Denver Instrument). The filled flasks were weighted again and the temperatures of the permeates were measured. The volume of permeate was calculated based on weight and density at the measured temperature. The flux was calculated by dividing the volume of permeate by the membrane area of each membrane module (32.2 cm\(^2\)) and by the permeation time (1374 min for Passive System 1 and Passive System 2; 54 min for Conventional System). After measuring flux, permeate samples were collected for TOC, DOC and/or ATP analyses.

4.2.1.2 TMP Monitoring
The TMP was continuously monitored using custom pressure transducers placed on the tubing between the membrane modules and the permeate pumps. The pressure transducers and permeate pumps were at similar elevations. The pressure transducers were connected to a laptop where the LabView SignalExpress software was used to record the electrical signals detected by the transducers and convert them into pressure. The pressure transducers were all calibrated prior to the beginning of the study to determine the scaling factors for conversion (for calibration, see Appendix F). The TMP was recorded at a frequency of 1 Hz.

4.2.1.3 Total and Dissolved Organic Carbon
TOC and DOC analyses were performed to monitor the organic matter removal of the membrane systems throughout the study, in order to compare to results of previous studies on PFC-GDM systems (Oka, et al., 2017). Samples were collected 2 times per week on Mondays and Thursdays from feed water, membrane tanks, drains and permeates. Samples collected from the membrane...
tanks were collected at the end of the filtration cycle, immediately before aeration and backwash. The samples were collected in 40mL TOC glass vials and stored in the dark at T < 4°C until analysis. Prior to analysis, the DOC samples were filtered through a 0.45µm membrane disc filter (Supor®, Plain, 47mm diameter, No. 60173, Pall Laboratory). The filters were rinsed with 1L of DI water prior to sample filtration to ensure that no organics from the filter dissolved into the sample.

The TOC/DOC analyses were performed using the NPOC method of the TOC-LCPH/CPN Total Organic Carbon Analyser by Shimadzu. The instrument was calibrated to provide accurate measurements from 0 to 20mg/L of TOC/DOC (for calibration, see Appendix G). Every batch analysed included a standard sample with a known concentration that was analysed at the beginning and end of every batch as a quality control measure. The standard and calibration samples were prepared using DI water and a 1,000ppm TOC stock solution (prepared with 0.5312g of Potassium Hydrogen Phthalate in 250mL of DI water and preserved with phosphoric acid).

After analysis, the samples were discarded and the vials were rinsed with tap water and baked at 450°C for 1 hour to burn off any remaining organics. The same vials were used for all sample batches collected throughout the study.

4.2.1.4 ATP Testing

Adenosine triphosphate (ATP) concentrations were measured in order to monitor the microbiological activity in the various components of the membrane systems. ATP measurements are an indirect indicator of total living biomass (LuminUltra Technologies Ltd., 2017). ATP tests were performed weekly during the first 80 days of study. One sample was collected from feed water, membrane tanks, drains and permeates. All samples were collected in clean glass beakers and were analysed immediately after collection.

The analyses were performed using the Quench-Gone™ Aqueous Test Kit by LuminUltra Technologies Ltd (Fredericton, Canada). The test kit uses the enzyme Luciferase (naturally occurring in the tails of fireflies) to produce light when in contact with a sample containing ATP. The light is detected by a luminometer as Relative Light Units (RLU), which can be converted into ATP concentration.

The ATP samples were prepared using the test kit according to LuminUltra instructions and the results were read by their PhotonMaster™ Luminometer connected to a laptop containing their
LuminCalc software. The software acquired the RLUs measured by the luminometer and converted them into concentration of ATP.

4.2.2 Membrane Integrity Testing

4.2.2.1 Challenge Testing

Challenge tests (CTs) were used in the present study to quantify the contribution of the biofilm layer to the virus removal capacity of passive membrane systems, both intact and with integrity breaches of different sizes.

Five CTs were performed on Passive System 1, Passive System 2 and Conventional System throughout the study. The 1st CT was performed prior to the beginning of the study to establish the initial LRVs of the membrane modules. The 2nd, 3rd and 4th CTs were performed on days 58, 85 and 106 of the study, respectively, to monitor the changes in LRVs. The 5th CT was performed at the end of the study, after a final extensive chemical cleaning (see section 4.3.1.3). The dates of challenge testing are summarized in Table 9.

<table>
<thead>
<tr>
<th>Challenge Test</th>
<th>Testing Date</th>
<th>Corresponding Day of Study</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st</td>
<td>Nov. 20, 2018</td>
<td>0</td>
</tr>
<tr>
<td>2nd</td>
<td>Feb. 12, 2019</td>
<td>58</td>
</tr>
<tr>
<td>3rd</td>
<td>Mar. 11, 2019</td>
<td>85</td>
</tr>
<tr>
<td>4th</td>
<td>Apr. 1, 2019</td>
<td>106</td>
</tr>
<tr>
<td>5th</td>
<td>Apr. 23, 2019</td>
<td>After Cleaning</td>
</tr>
</tbody>
</table>

The CTs were performed using MS2 bacteriophages as challenge material. The MS2 bacteriophage is considered a good surrogate for enteric viruses, as it has similar size and shape to the poliovirus and hepatitis virus, it is easy to grow and count, and it is not harmful to humans or animals (Wu, et al., 2017). MS2 bacteriophages have a diameter of approximately 0.03μm (Strauss Jr. & Sinsheimer, 1963), are negatively charged at the pH range typical of surface waters (Schijven & Hassanizadeh, 2000) and are hydrophobic (Attinti, et al., 2010).

During the CTs, the membrane systems were fed with a solution containing MS2 bacteriophage. The MS2 stock solution used to make the feed solution was purchased from GAP EnviroMicrobial
Services Ltd (GAPLAB - London, Ontario). The stock solution had an MS2 concentration of $4 \times 10^{11}$ cfu/mL. The stock solution was mixed with carbon filtered tap water to make the final feed solution. The tap water was filtered twice with a standard household carbon filter (EPW2C, EcoPure), according to recommendations from GAPLAB, to ensure that chlorine and most contaminants were removed but still leaving enough buffering capacity for the MS2 to survive.

The feed solution concentration for the 1st and 2nd CTs was $6 \times 10^6$ cfu/mL, calculated using Equation 7. However, the permeate concentrations of some membrane modules were close to zero on the 2nd CT. For these, the actual LRV could not be accurately estimated. For this reason, the MS2 concentration in the feed solution was increased to $6 \times 10^9$ cfu/mL for the remaining CTs.

The test solution was filtered through the membrane systems during one entire cycle to simulate normal operational conditions and to produce enough permeate for sampling. The CTs of Passive System 1 and Passive System 2 lasted approximately 24h and the CTs of the Conventional System lasted approximately 1h (i.e. durations of an entire cycle). The permeate fluxes used were the same as normal operation.

Two permeate samples were collected from each membrane module (total of 24 permeate samples). The permeate samples from Passive System 1 and Passive System 2 were collected during the first and last 3h of permeation in a cycle. The permeate samples from the Conventional System were collected throughout the first and last 25min of permeation in a cycle. Two feed water samples were collected from each membrane tank (total of 6 feed water samples), halfway through the collection of the permeate samples (i.e. for Passive System 1 and Passive System 2, after 1.5h and 21.5h of permeation; for the Conventional System, after 12.5min and 42.5min of permeation).

The samples were collected in 50mL sterile vials with sodium thiosulfate (final concentration of 0.1%), according to GAPLAB recommendations. The resulting 30 samples were then stored in a cooler at $T < 4^\circ$C and shipped overnight to GAPLAB for MS2 counting. The concentrations of MS2 in the feed water and permeates of the membrane modules were used in Equation 4 to calculate the LRVs of the membrane modules.
4.2.2.2 Pressure Decay Testing

Pressure decay tests (PDTs) were used in the present study to estimate the LRV of passive membrane systems.

PDTs were performed on Passive System 1 and Conventional System, but not on Passive System 2. This was done to investigate the potential impact of PDTs on the biofilm layer and therefore on the performance of passive membrane systems. The potential impact of PDTs was quantified using CTs (see section 4.2.2.1).

The PDT system consisted of a compressed air line with (Figure 8):

1. 1 pressure regulator;
2. 1 digital pressure gauge;
3. 2 cylindrical acrylic tanks, T1 and T2 (volumes of 11.26L and 9.68L, respectively); and
4. 4 manual ball valves.

Two tanks were used in parallel in the PDT system in order to increase the hold-up volume of the pressurized system. A greater hold-up volume enables a more accurate measure of the PDR (see Equation 9, section 2.4.2).

Figure 8 – Schematic representation of PDT system.
At the beginning of a PDT, BV1 was open and BV4 was closed. The membrane module to be tested was disconnected from the membrane system tubing (Figure 3) and connected to the PDT system tubing (Figure 8). BV4 was opened and after approximately 10s, BV1 was closed. The waiting period allowed the water inside of the membrane fibres to permeate into the membrane tank. Then, the air and water temperatures and the time required for the pressure gauge to register a decrease of 1psi were measured. The average test pressure used was 8psi, which is the minimum pressure required to detect a 3µm breach (see section 2.4.1). The backpressure on the membrane modules was negligible.

The PDTs were performed once a week on Wednesdays. Passive System 1 was tested first while the Conventional System was chemically cleaned (see section 4.3.1.2). Testing of Passive System 1 started 15 minutes after the beginning of a filtration cycle. The waiting period allowed water to completely fill the pores of the membrane modules. Testing at the beginning of the filtration cycle yielded the most conservative results, as the biofilm layer was likely looser after the relaxation and aerated backwash at the end of the previous filtration cycle. Testing of the Conventional System started 15 minutes after the beginning of the first filtration cycle after chemical cleaning. All membrane modules were tested inside the membrane tanks, submerged in regular feed water.
4.3 Quality Assurance and Control

4.3.1 Membrane Chemical Cleaning

Different chemical cleaning protocols were used throughout the present study in order to clean new membrane modules (see section 4.3.1.1), routine clean the membrane modules of the Conventional System (see section 4.3.1.2) and clean all membrane modules at the end of the study (see section 4.3.1.3). All chemical cleaning protocols used sodium hypochlorite and/or citric acid solutions as cleaning agents. The sodium hypochlorite solutions were prepared with DI water and bleach (Javex ® 12, Clorox Commercial Solutions). The citric acid solutions were prepared with DI water and citric acid monohydrate (FLA1045500, Thermo Fisher Scientific).

4.3.1.1 Chemical Cleaning Protocol for New Membrane Modules

Immediately after potting, all membrane modules were cleaned according to the following protocol (Robinson, 2018):

1. Soak membrane modules for 24h in 500ppm sodium hypochlorite solution;
2. Drain membrane tanks;
3. Refill with new 500ppm sodium hypochlorite solution and permeate membrane modules for 15min;
4. Drain membrane tanks;
5. Backwash membrane modules for 15min with DI water;
6. Drain membrane tanks;
7. Refill with DI water and permeate membrane modules for 30min;
8. Store membrane modules in Ziploc bags with DI water until use. If storing for more than one week, use a 5ppm sodium hypochlorite solution and store at T < 4°C.

4.3.1.2 Routine Chemical Cleaning of Conventional System

Routine chemical cleaning was performed on the membrane modules of the Conventional System 3 times per week on Mondays, Wednesdays and Fridays. The routine chemical cleaning protocol was adjusted throughout the study as decreases in permeability were observed (see section 5.1.2.2). The different routine chemical cleaning protocols used throughout the study are summarized in Table 10.
Cleaning Protocol A was chosen based on the protocol used in the Eagle Lake Membrane Filtration Facility, where membranes similar to those used in the present study are used. This protocol did not prove efficient in controlling fouling after a few weeks of operation (see section 5.1.2.2). To address this shortcoming, the concentration of sodium hypochlorite was increased on day 21 of the study (Cleaning Protocol B). After a few more weeks of operation, the permeability of the membrane modules continued to decrease (see section 5.1.2.2). Therefore, on day 63 of the study, the concentration of sodium hypochlorite was further increased and a citric acid soak was added.
after the sodium hypochlorite soak (Cleaning Protocol C). With Cleaning Protocol C, the permeability did not consistently decrease until the end of the study (see section 5.1.2.2).

### 4.3.1.3 Final Extensive Chemical Cleaning

At the end of the study, prior to the 5th CT, the membrane modules of Passive System 1, Passive System 2 and Conventional System were subjected to an extensive chemical cleaning according to the following protocol (Jain, 2019):

1. Soak membrane modules for 24h in 750ppm sodium hypochlorite solution;
2. Drain membrane tanks;
3. Refill with new 750ppm sodium hypochlorite solution and permeate membrane modules for 2h;
4. Drain membrane tanks;
5. Refill with DI water and aerate and backwash membrane modules with DI water for 1h;
6. Drain membrane tanks;
7. Repeat steps 1 to 6 using 750ppm citric acid solution;
8. Soak membrane modules in DI water for 24h until 5th CT.

### 4.3.2 Clean Water Permeability

The membrane modules were submitted to clean water permeability tests in order to establish and monitor the intrinsic membrane permeability. Four clean water permeability tests were performed: before breaching, after breaching, before the 1st CT and before the 5th CT (after final extensive chemical cleaning described in section 4.3.1.3).

The clean water permeability was tested by permeating the membrane modules with DI water for approximately 30min with all permeate pumps operating at 50rpm. Flux, TMP and temperature were measured according to sections 4.2.1.1 and 4.2.1.2. Permeability was calculated using Equation 5 and corrected to 20°C using Equation 6. The results of all clean water permeability tests are presented in Table II.
Table II – Clean water permeabilities of membrane modules used in the experimental set-up.

<table>
<thead>
<tr>
<th>Membrane System</th>
<th>Membrane Module</th>
<th>Clean Water Permeability Corrected to 20°C (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Passive System 1</td>
<td>1</td>
<td>4.19E-13</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>4.14E-13</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>4.35E-13</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>4.03E-13</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>4.34E-13</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>4.18E-13</td>
</tr>
<tr>
<td>Conventional System</td>
<td>1</td>
<td>4.10E-13</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>5.37E-13</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>5.65E-13</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>4.81E-13</td>
</tr>
<tr>
<td>Average and 99% confidence interval</td>
<td>4.47E-13 ± 4.71E-14</td>
<td>5.14E-13 ± 2.01E-14</td>
</tr>
</tbody>
</table>

The 99% confidence intervals of the clean water permeabilities varied less than 10% from the average for each test; and the clean water permeabilities of each test were not statistically different from each other according to the 99% confidence intervals. This indicates that the clean water permeabilities did not vary significantly within modules or within different tests. These permeability values are in accordance with previous studies done on the ZeeWeed 500® membranes (Oka, 2015; Khadem, 2016).
4.4 Statistical Analyses

The statistical analyses performed in the present study included paired and unpaired t-tests, two-factor ANOVAs without replication, Tukey’s Honestly Significant Difference tests and linear regressions. All statistical tests were performed with a confidence level of 99% (α = 0.01).

The paired and unpaired t-tests and the two-factor ANOVAs without replication were performed using the Data Analysis tool of Microsoft Excel. The Tukey’s Honestly Significant Difference tests were performed using the ‘multcompare’ function of MATLAB.

The linear regressions were performed using the Trendline tool of Microsoft Excel. The confidence intervals of the parameters of the linear regression (i.e. slope and intercept) were calculated according to Chapter 34 of Berthouex & Brown (2002).
5.0 RESULTS AND DISCUSSION

5.1 Overall Performance of Membrane Systems

Passive System 1, Passive System 2 and Conventional System were operated continuously for approximately 17 weeks. Analytical methods were employed to monitor the overall performance of the membrane systems during operation, as described in section 4.2.1. Monitoring was applied to ensure that the membrane systems were performing as expected for passive and conventional membrane systems, as well as to ensure that all system components were integral and fully functional.

5.1.1 Physical Aspect of Membrane Modules

Images of the membrane modules of Passive System 1, Passive System 2 and Conventional System on the last day of the study are presented in Figure 9.

It is possible to observe in Figure 9 that the membrane modules of Passive System 1 and Passive System 2 presented a visible foulant layer at the end of the study. The membrane modules of the Conventional System did not present a visible foulant layer. As previously discussed (see section 2.2), a biofilm layer is expected to develop on passive membrane systems (Peter-Varbanets, et al., 2010; Peter-Varbanets, et al., 2011).
The presence of the foulant layer, hereafter referred to as biofilm layer, on the membrane modules of Passive System 1 and Passive System 2 was visible after approximately 2 weeks of operation. The biofilm layer of the membrane modules of Passive System 1 was lighter in colour and had more patches where the biofilm layer was not as pronounced (i.e. dark) as the biofilm layer of the membrane modules of Passive System 2. Detachment of the biofilm layer was observed during the execution of PDTs on the membrane modules of Passive System 1. The detachment was caused by the slight movement of the modules when disconnecting the modules from the membrane system and connecting them to the PDT system (see section 4.2.2.2). Previous studies have also reported an easily detachable biofilm layer on passive hollow fibre membranes (Oka, et al., 2017). The membrane modules of Passive System 2 remained undisturbed throughout the entire study. It is likely that the difference between the biofilm layer on the membrane modules of Passive System 1 and Passive System 2 was caused by the application of PDTs on Passive System 1.

5.1.2 Membrane Permeability

The permeabilities of the membrane modules of Passive System 1, Passive System 2 and Conventional System were calculated using the TMP, permeate flux and permeate temperature measured throughout the study. TMP was monitored as described in section 4.2.1.2 (for TMP data, see Appendix H). Permeate flux and permeate temperatures were measured as described in section 4.2.1.1. The membrane permeability was calculated using Equation 5 (see section 2.1). Membrane permeabilities were corrected to 20°C according to Equation 6 (see section 2.1) and were normalized to the permeability values on the first day of the study.

5.1.2.1 Passive System 1 and Passive System 2

The normalized permeabilities of the membrane modules of Passive System 1 and Passive System 2 are presented in Figure 10.
As illustrated in Figure 10, the membrane modules of both Passive System 1 and Passive System 2 experienced a decrease in permeability as the study progressed. There was a greater decrease in the permeability of Passive System 2 than in that of Passive System 1. For both systems, the rate of decrease in permeability became slower over time, suggesting the establishment of steady-state conditions, as would be expected for GDM systems (see section 2.2).

In the previous study performed on the PFC-GDM system, similar feed water (mix of Jericho Pond water and tap water) was used, but the PFC-GDM system was operated under constant pressure (Oka, et al., 2017). Steady-state conditions were observed after approximately 2 weeks of operation. Also, the normalized permeability at steady-state was approximately 0.4. At steady-state, the permeate flux was approximately 5Lm⁻²h⁻¹, which is why, for the present study, a permeate flux of 5Lm⁻²h⁻¹ was selected for constant flux operation.

Although at low permeate flux operation the fouling mechanisms are not expected to differ for systems operated under either constant pressure or constant flux (Miller, et al., 2014; Oka, 2015), permeabilities can differ substantially. This is because the mass flux of foulants towards the membrane is variable in constant pressure systems, while it is fixed in constant flux systems. It is likely that the mass flux of foulants towards the membranes of the present study was different than that of the previous study, due to differences in feed water (water was collected from Jericho Pond at different times) and operating conditions (constant pressure vs. constant flux). This likely led to
the differences between the present and the previous studies in the timeline to establish steady-state conditions and in the magnitude of the permeability at steady-state.

Furthermore, the difference between the magnitudes of the normalized permeabilities of Passive System 1 and Passive System 2 at steady-state was potentially caused by the detachment of biofilm from the membrane modules that occurred during PDTs (see section 5.1.1). It is possible that this detachment of biofilm led to a looser and more permeable biofilm layer to develop on the membrane modules of Passive System 1 compared to the biofilm layer on the membrane modules of Passive System 2, as suggested by Oka, et al. (2017).

5.1.2.2 Conventional System

The normalized permeabilities of the membrane modules of the Conventional System are presented in Figure II.

As illustrated in Figure II, the membrane modules of the Conventional System experienced two significant decreases in permeability throughout the study. The first decrease was observed between days 0 and 21 of the study, while Cleaning Protocol A was applied to the membrane modules (see section 4.3.1.2). The cleaning protocol was adjusted to Cleaning Protocol B on day 21 of the study, causing an increase in normalized permeability to approximately 1.0. The second decrease was observed between days 21 and 63 of the study, while Cleaning Protocol B was applied
to the membrane modules. The cleaning protocol was adjusted to Cleaning Protocol C on day 63 of the study, causing an increase in normalized permeability to approximately 1.0.

Using Cleaning Protocol C, the normalized permeability of the membrane modules remained relatively constant (i.e. no consistent downward trend) until the end of the study.

5.1.3 Organic Matter Removal

The organic matter removal was monitored in the present study in order to compare to results of previous studies on PFC-GMD systems (Oka, 2015; Khadem, 2016). As these results do not relate to the objectives of the present study, only a brief summary of the results is provided in the present section. For all results, statistics and discussion, see Appendix I.

The soluble organic matter removal of the membrane modules of Passive System 1 ranged from 0 to 23%, with an average of 7%; of Passive System 2, also from 0 to 23%, with an average of 7%; and of the Conventional System, from 0 to 25%, also with an average of 7%.

Low organic matter removal for conventional UF membrane systems was expected (Schafer, 2001), as the pore size of UF membranes typically used in drinking water treatment applications is relatively large (see section 2.1). For passive membrane systems, previous studies using similar feed water (mix of Jericho Pond water and tap water) reported organic matter removal efficiencies of up to 50% (Oka, et al., 2017), likely due to biodegradation by the biofilm layer. However, the feed water preparation in the previous study did not include the pre-filtration with a 1.5µm filter (see section 4.1.3), as included in the present study. Also, the TOC/DOC concentrations in the previous study were approximately 1.5 times greater than the TOC/DOC concentrations in the present study. The differences in feed water preparation and concentration likely led to the differences in organic matter removal between the previous and the present study. A detailed discussion on the removal of organic material by PFC-GDM systems is presented in Oka et al. (2017).

5.1.4 ATP Concentration

The ATP concentrations were measured in order to monitor the microbiological activity in the various components of the membrane systems. As these results do not relate to the objectives of the present study, only a brief summary of the results is provided in the present section. For all results, statistics and discussion, see Appendix J.

For Passive System 1 and Passive System 2, the ATP concentration in the drains were greater than the ATP concentrations in the feed water and membrane tanks. The difference was likely caused by
the detachment of biofilm during the aeration and backwash period that occurred prior to draining. For the Conventional System, there was no statistical difference in the ATP concentration between the feed water and membrane tanks nor between the feed water and drain. This was expected, as the membrane modules of the Conventional System did not have a biofilm layer.

Furthermore, the ATP concentrations in the drain of Passive System 1 and Passive System 2 were greater than that of the Conventional System. This is further evidence that a biofilm layer was present on the membrane modules of Passive System 1 and Passive System 2, but not on the membrane modules of the Conventional System.
5.2 Contribution of the Biofilm Layer to Virus Removal Capacity of Passive Membrane Systems

The first objective of the present study was to quantify the contribution of the biofilm layer to the virus removal capacity of passive membrane systems, both intact and with integrity breaches of different sizes (see section 3.0). In order to do so, CTs were performed on the membrane modules of Passive System 1, Passive System 2 and Conventional System as described in section 4.2.2.1. The CT results are presented in Figure 12.

Figure 12 – LRVs measured from challenge tests.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System

LRV: Average of log removal values calculated from 1st and 2nd sampling (see section 4.2.2.1). Error bars indicate maximum and minimum values.

The LRVs of the membrane modules of the Conventional System on day 58 were considered outliers due to insufficient chemical cleaning (see sections 4.3.1.2 and 5.1.2.2).

At the start of the study (day 0), the LRVs of all membrane modules ranged from 2.1 to 3.0 log. These values are consistent with those from previous studies on virus removal by clean UF membranes with pore sizes of 0.02 to 0.04 µm (Kreiβel, et al., 2012; Lu, et al., 2013). Also, at the start of the study, the LRVs of the intact membrane modules and membrane modules with breaches of 20 and 40 µm ranged from 2.6 to 3.0 log. The LRVs of the membrane modules with breaches of 180 µm were all 2.1 log. This suggests that smaller breaches did not affect the LRVs of the membrane modules. Similar results have been reported for breached UF membranes, where a CT was able to detect a decrease in LRVs for membrane modules with breaches of 200 and 600 µm, but no
difference in LRVs was detected for membrane modules with breaches of 20, 40 and 60µm (Brehant, et al., 2010).

On day 58 of the study, the LRVs of Passive System 1 and Passive System 2 increased, ranging from 5.5 to 6.9 log. The LRVs of the Conventional System also increased, ranging from 4.1 to 4.9 log. The increase in the LRVs of the Conventional System was likely caused by the development of a foulant layer due to insufficient chemical cleaning (see sections 4.3.1.2 and 5.1.2.2). As previously discussed (see section 2.3), non-biological foulant layers can also cause an increase in the virus removal capacity of membrane systems. Thus, the LRVs of the Conventional System on day 58 were considered outliers. The LRVs of day 58 are consistent with previous studies on virus removal by fouled UF membranes (Jacangelo, et al., 1995; Lv, et al., 2006; Kreiβel, et al., 2012; Lu, et al., 2013; ElHadidy, et al., 2014; Yin, et al., 2015). There was also no obvious trend in the LRVs of membrane modules with different breach sizes, suggesting that the breach size did not have an impact on LRVs when the biofilm layer was present.

On days 85 and 106 of the study, the LRVs of Passive System 1 and Passive System 2 ranged from 4.1 to 6.7 log. As on day 58 of the study, there was no obvious trend in the LRVs of membrane modules with different breach sizes, suggesting that the breach size did not have an impact on LRVs when the biofilm layer was present. With more effective chemical cleaning, the LRVs of the Conventional System ranged from 2.3 to 2.9 log, values similar to those observed at the start of the study.

After cleaning, the LRVs of all membrane modules of Passive System 1, Passive System 2 and Conventional System were similar to those at the start of the study, ranging from 1.9 to 2.9 log. Also similar to the start of the study, after cleaning the intact membrane modules and the membrane modules with breaches of 20 and 40µm had greater LRVs than the membrane modules with breaches of 180µm.

To better compare the LRVs of the different membrane modules, a two-factor ANOVA without replication with a significance of 0.01 (99% confidence level) was performed on the LRV data of each day of challenge testing. The ANOVA assessed if the type of membrane system or the breach size had an impact on LRVs. When an impact was detected, a 0.01 significance (99% confidence level) Tukey test was performed to pinpoint which system(s) or breach size(s) was responsible for the impact. The results are summarized in Table 12.
### Table 12

Results of two-factor ANOVA and Tukey tests (significance of 0.01) performed on LRVs measured from challenge tests.

<table>
<thead>
<tr>
<th>Day of Study</th>
<th>Membrane System</th>
<th>Breach Size</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Is there an impact? (ANOVA)</td>
<td>Where? (Tukey)</td>
</tr>
<tr>
<td>0</td>
<td>No ( \text{(p=0.472)} )</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>58</td>
<td>No ( \text{(p=0.010)} )</td>
<td>NA</td>
</tr>
<tr>
<td>85</td>
<td>Yes ( \text{(p=0.0004)} )</td>
<td>Conventional ≠ Passive 1 ( \text{(p=0.0004)} )</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Conventional ≠ Passive 2 ( \text{(p=0.003)} )</td>
</tr>
<tr>
<td>106</td>
<td>Yes ( \text{(p=0.0004)} )</td>
<td>Conventional ≠ Passive 1 ( \text{(p=0.0005)} )</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Conventional ≠ Passive 2 ( \text{(p=0.001)} )</td>
</tr>
<tr>
<td>After Cleaning</td>
<td>No ( \text{(p=0.260)} )</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**NA**: Not applicable.

On days 0 and 58 of the study and after cleaning, there was no statistical difference in LRVs between Passive System 1, Passive System 2 and the Conventional System. On days 58, 85 and 106 of the study, there was no statistical difference between the LRVs of Passive System 1 and Passive System 2. On days 85 and 106 of the study, the LRVs of Passive System 1 and Passive System 2 were statistically greater than the LRVs of the Conventional System.

At the start of the study and after cleaning, there was a statistical difference in the LRVs of membrane modules with different breach sizes. On those days, the membrane modules with a breach of 180µm had statistically lower LRVs than the membrane modules with no breaches or
breaches of 20 and 40\(\mu\)m. On days 58, 85 and 106 of the study, the different breach sizes did not have a statistical impact on LRVs.

These results suggest that the presence of the biofilm layer caused an increase in the LRVs for viruses of the membrane modules of Passive System 1 and Passive System 2. The results also suggest that the biofilm layer was able to offset any possible decreases in LRVs due to the presence of breaches on the membrane modules. Both intact and breached membrane modules of Passive System 1 and Passive System 2 were able to consistently achieve LRVs of more than 4.0 log for viruses (minimum required by the CDWQG) when fouled by the biofilm layer.

It is also important to note that there was no statistical difference between the LRVs of the membrane modules of Passive System 1 and Passive System 2. These systems differed by two factors: the membrane modules of Passive System 1 were subjected to PDTs while those of Passive System 2 were not; and the membrane modules of Passive System 1 had a greater steady-state permeability than those of Passive System 2 (see section 5.1.2.1), suggesting that the biofilm layer of Passive System 2 had a greater density and/or thickness. The similarity between the LRVs of Passive System 1 and Passive System 2 suggests that the capacity of the biofilm layer to remove viruses is not significantly affected by PDTs; and that a denser and/or thicker biofilm layer does not contribute to a greater virus removal capacity.

It can be concluded from the CT results that the biofilm layer contributes with more than 2.0 log to the LRV for viruses of passive membrane systems. As a consequence, passive membrane systems are capable of constantly achieving an LRV for viruses of more than 4.0 log. It can also be concluded from these results that the biofilm layer is capable of mitigating the impact of integrity breaches in the range of breach sizes considered. As a consequence, the LRV for viruses of breached passive membrane systems is in the same range as the LRV of intact passive membrane systems (i.e. > 4.0 log). It can also be concluded that the virus removal capacity of the biofilm layer is not affected by the density and/or thickness of the biofilm layer for the conditions investigated, nor by the application of weekly PDTs.
5.3 Assessment of PDTs for Estimation of LRVs of Passive Membrane Systems

The second objective of the present study was to assess the use of PDTs as a method of estimating the LRV of passive membrane systems (see section 3.0). In order to do so, PDTs were performed on the membrane modules of Passive System 1 and Conventional System as described in section 4.2.2.2. The LRVs were then estimated from the PDT results using the standard ASTM approach (see section 2.4.2.3) and compared to the LRVs measured from the CTs (Figure 13). For the detailed LRV calculations, including air and water flows through the breaches, see Appendix K.

It is important to note that the LRVs from CTs and PDTs do not correspond to the retention of material of same size. LRVs from CTs are based on the removal of viruses and LRVs from PDTs are based on the removal of material larger than 3µm (see section 2.4). Regardless, for low-pressure membranes, it can be expected that the removal of larger material will be equal to or greater than the removal of small material, as the primary removal mechanism of low-pressure membranes is size exclusion (USEPA, 2005; Crittenden, et al., 2012). Thus, the LRVs for material larger than 3µm (LRVs from PDTs) should be similar or greater than the LRVs for viruses (LRVs from CTs).

Figure 13 – LRVs from standard PDT approach vs. LRVs from CTs.
(a) Passive System 1 (b) Conventional System

Confidence intervals of slopes and intercepts were determined based on a 99% confidence level.

The LRVs from PDTs presented in Figure 13 are for the same days as LRVs from CTs. For all LRVs from PDTs, see Appendix K.
If the LRVs from PDTs are similar to the LRVs from CTs, the linear regression between these two parameters is expected to have a slope of approximately 1.0 and an intercept of approximately zero (i.e. linear regression should be $y = x$). As illustrated in Figure 13, for Passive System 1, the slope of the linear regression was statistically different from 1.0 (i.e. $0.12 \pm 0.33$); and the intercept was statistically different from zero (i.e. $1.58 \pm 1.51$). For the Conventional System, the slope of the linear regression was not statistically different from 1.0 (i.e. $1.89 \pm 2.31$); and the intercept was not statistically different from zero (i.e. $-2.67 \pm 5.83$).

These results indicate that the LRVs from the standard PDT approach were similar to the LRVs from CTs for the Conventional System, but were different for Passive System 1. For the Conventional System, the LRVs estimated from the standard PDT approach ranged from 1.0 to 3.0 log, and the LRVs from CTs ranged from 2.0 to 2.9 log. For Passive System 1, the LRVs estimated from the standard PDT approach ranged from 1.0 to 3.0 log, while the LRVs from CTs ranged from 2.0 to 6.9 log. This indicates that the standard PDT approach provides a satisfactory estimation of the LRVs for viruses for conventional membrane systems. However, for passive membrane systems, the LRVs estimated based on the standard PDT approach do not reflect the LRVs for viruses measured from the CTs.

To attempt to reconcile the LRVs from PDTs and the LRVs from CTs for Passive System 1, a mass balance approach was used to convert the LRVs from the standard PDT approach to a modified PDT approach. The modified PDT approach combines the results of PDTs and CTs to recalculate the LRVs from PDTs in terms of virus removal.

As outlined in section 2.4.2.3, the standard PDT approach only considers larger material that would normally be removed by the membrane, but bypasses treatment by flowing through a breach (see derivation of Equation 22, section 2.4.2.3). However, for smaller material that is not completely removed by the membrane, the fraction of material that flows through the membrane must also be considered. For the modified PDT approach, Equation 20 was used to estimate the LRV rather than Equation 22 (see derivation of Equation 20, section 2.4.2.3).

$$LRV = \log \left( \frac{M_f}{M_{p,b}} \right) = \log \left( \frac{M_f}{M_b + M_{nr}} \right) = \log \left( \frac{QC_f}{Q_{water,b}C_b + Q_{int}C_{nr}} \right)$$

Equation 20
Equation 20 is the numerical representation of the mass balance used to estimate the LRV based on the modified PDT approach. The parameters used in Equation 20 were determined as follows:

1. The mass flux of viruses in the feed water ($M_F$) was calculated as the product of the total water flow through the membrane ($Q$) and the concentration of viruses in the feed water ($C_f$), as measured during CTs.

2. The mass flux of viruses in the permeate ($M_{p,b}$) was estimated based on the sum of the mass flux of viruses flowing through the breach ($M_B$) and the mass flux of viruses not removed by the intact area of the membrane ($M_{nr}$).
   a. The mass flux of viruses flowing through the breach ($M_B$) was estimated based on the product of the water flow through the breach ($Q_{water,b}$) and the concentration of contaminants flowing through the breach ($C_b$).
      i. The water flow through the breach ($Q_{water,b}$) was estimated based on the results of the PDTs using Equation 15 (see section 2.4.2.2).
      ii. The concentration of contaminants flowing through the breach ($C_b$) was estimated according to item 3 of the present list.
   b. The mass flux of viruses not removed by the intact area of the membrane ($M_{nr}$) was estimated based on the product of the water flow through the intact area of the membrane ($Q_{int}$) and the concentration of contaminants not removed by the intact area of the membrane ($C_{nr}$).
      i. The water flow through the intact area of the membrane ($Q_{int}$) was estimated based on the difference between the total water flow through the membrane ($Q$) and the water flow through the breach ($Q_{water,b}$).
      ii. The concentration of contaminants not removed by the intact area of the membrane ($C_{nr}$) was estimated based on the extent of treatment achieved by the intact membrane module, as measured during CTs.

3. The concentration of contaminants flowing through the breach ($C_b$) was estimated based on one of two hypotheses:
   a. Hypothesis I (Figure 14a): the biofilm layer developed on the walls of the breach, causing a decrease in breach diameter. Contaminants were not prevented from flowing through the breach and into the permeate stream. Therefore, $C_b$ was estimated to be equal to the concentration of contaminants in the feed water ($C_f$).
b. Hypothesis 2 (Figure 14b): the biofilm layer developed on top of the breach. Part of the contaminants were prevented from flowing through the breach and into the permeate stream. Therefore, $C_b$ was estimated to be equal to the concentration of contaminants not retained by the biofilm layer ($C_{nr,bio}$).

i. The concentration of contaminants not retained by the biofilm layer ($C_{nr,bio}$) was estimated based on the extent of treatment achieved by the biofilm layer of the intact membrane module, as measured during CTs (Equation 25).

$$C_{nr,bio} = \frac{C_f}{(LRV_{intact,x} - LRV_{intact,0})^{10}}$$

For the above, $LRV_{intact,x}$ is the LRV measured from the CT of the intact module on a certain day ‘x’ when biofilm was present; and $LRV_{intact,0}$ is the LRV measured from the CT of the intact module on day 0 when biofilm was not present; and $C_f$ is the concentration of viruses in the feed water measured during the CT.

Figure 14 – Hypothesis of biofilm growth and respective concentrations of contaminants not removed during permeation.

(a) Hypothesis 1 (b) Hypothesis 2

$C_f$: Concentration of contaminants in the feed water;

$C_{nr}$: Concentration of contaminants not removed by the intact area of the membrane; and

$C_{nr,bio}$: Concentration of contaminants not removed by the biofilm layer.

The two hypotheses of biofilm growth were considered in order to verify which hypothesis best reflects the actual behaviour of the biofilm layer. The LRVs from PDTs were recalculated using the modified PDT approach based on Hypothesis 1 and based on Hypothesis 2. The hypothesis that was able to better reflect the LRVs from CTs was considered the most likely to be true.
The comparisons between LRVs from CTs and LRVs from the modified PDT approach based on Hypothesis 1 and based on Hypothesis 2 are presented in Figure 15 and Figure 16, respectively.

Figure 15 – LRVs from modified PDT approach based on Hypothesis 1 vs. LRVs from CTs.
(a) Passive System 1 (b) Conventional System

Confidence intervals of slopes and intercepts were determined based on a 99% confidence level.

Figure 16 – LRVs from modified PDT approach based on Hypothesis 2 vs. LRVs from CTs.
(a) Passive System 1 (b) Conventional System

Confidence intervals of slopes and intercepts were determined based on a 99% confidence level.
For Hypothesis 1 (Figure 15), the slope of the linear regression between the LRVs from modified PDTs and the LRVs from CTs for Passive System 1 was statistically different from 1.0 (i.e. 0.16 ± 0.28); and the intercept was statistically different from zero (i.e. 1.32 ± 1.31). For the Conventional System, the slope of the linear regression was not statistically different from 1.0 (i.e. 0.97 ± 0.90); and the intercept was not statistically different from zero (i.e. -0.16 ± 2.30).

These results indicate that the LRVs from the modified PDT approach based on Hypothesis 1 were similar to the LRVs from CTs for the Conventional System, but different for Passive System 1. Therefore, the modified PDT approach based on Hypothesis 1 provides a satisfactory estimation of the LRVs for viruses for conventional membrane systems. However, for passive membrane systems, the LRVs estimated from the modified PDT approach based on Hypothesis 1 do not reflect the LRVs for viruses measured from the CTs.

For Hypothesis 2 (Figure 16), the slopes of the linear regressions between the LRVs from the modified PDTs and the LRVs from CTs for both Passive System 1 and Conventional System were not statistically different from 1.0 (i.e. 1.10 ± 0.30 and 0.97 ± 0.90, respectively); and the intercepts were not statistically different from zero (i.e. -0.68 ± 1.39 and -0.16 ± 2.30, respectively).

These results indicate that the LRVs from the modified PDT approach based on Hypothesis 2 were similar to the LRVs from CTs for both the Conventional System and Passive System 1. Therefore, the modified PDT approach based on Hypothesis 2 provides a satisfactory estimation of the LRVs for viruses for both conventional and passive membrane systems.

For the Conventional System, the LRVs estimated from the modified PDT approach based on Hypothesis 1 (Figure 15) were equal to those based on Hypothesis 2 (Figure 16). This is because the difference between hypotheses is only related to different mechanisms of biofilm growth, and the membrane modules of the Conventional System do not have a biofilm layer. Therefore, for the Conventional System, the LRVs estimated from the modified PDT approach are not affected by the hypothesis considered, as the concentration of contaminants flowing through the breach is always equal to the concentration of contaminants in the feed water due to the absence of a biofilm layer.

Furthermore, for the Conventional System, the modified PDT approaches based on Hypothesis 1 and Hypothesis 2 provided a more accurate estimation of LRVs than the standard PDT approach (Figure 13). This is likely because the modified PDT approaches consider the removal of smaller material (i.e. viruses), as do the LRVs from CTs. Regardless, the standard PDT approach and the
modified PDT approaches based on Hypothesis 1 and Hypothesis 2 all provided a satisfactory estimation of the LRVs for viruses.

For Passive System 1, the LRVs estimated from the modified PDT approach based on Hypothesis 2 (Figure 16) reflected the LRVs from the CTs, while those based on Hypothesis 1 (Figure 15) did not. This suggests that Hypothesis 2 is true. Moreover, it was observed from the results of the CTs that the biofilm layer is capable of mitigating the impact of breaches of up to 180µm of diameter (see section 5.2). These results suggest that, when a breach is present on a passive membrane module, the biofilm layer grows on top of the breach, covering it completely. The biofilm layer then acts as a secondary barrier to contaminants that would otherwise bypass treatment by flowing through the breach.

Due to the porous nature of the biofilm layer, air and water are still allowed to flow through the breach, making it possible to detect the breach during a PDT. The standard PDT approach assumes that, if there is a breach through which air and water are able to flow, contaminants are also able to flow through without restriction. However, if a biofilm layer is present, a fraction of the contaminants is prevented from flowing through the breach. Therefore, the additional removal provided by the biofilm layer must be taken into account in order to estimate the LRV of breached passive membrane systems from the results of a PDT.

It can be concluded from these results that both the standard and the modified PDT approaches can adequately estimate the LRV for viruses of conventional membrane systems. However, the standard PDT approach cannot adequately estimate the LRV for viruses of passive membrane systems. In order to estimate the LRV of breached passive membrane systems, a modified PDT approach that takes into account the additional removal provided by the biofilm layer is required.
5.4 Alternative Integrity Testing Protocol for Passive Membrane Systems

The results of the present study indicate that, due to the presence of the biofilm layer, passive membrane systems are capable of constantly achieving an LRV for viruses of more than 4.0 log (minimum required by the CDWQG). The results also suggest that the biofilm layer is capable of covering integrity breaches, acting as a secondary barrier to contaminants that would otherwise bypass treatment by flowing through the breach. The regulatory implication of these results is that, given the greater removal reliability provided by the biofilm layer, passive membrane systems could be allowed less frequent integrity testing in full scale applications.

An alternative integrity testing protocol for passive membrane systems could consist of periodic but infrequent pressure decay testing (e.g. weekly). If a breach is detected, the LRV of the breached membrane module should be estimated using a modified PDT approach that takes into account the additional removal provided by the biofilm layer. If using the modified PDT approach proposed in the present study, challenge testing must also be incorporated into the integrity testing protocol. A CT should be performed shortly after implementation, once the biofilm layer has established on the membrane modules. In the occasion that a breach is detected, the results from the CT could then be used for the estimation of an LRV based on the modified PDT approach. CTs could be performed every six to twelve months in order to monitor the contaminant removal capacity of the biofilm layer.

The proposed integrity testing protocol is a simpler alternative to the standard protocol of performing direct integrity tests daily. For the PFC-GDM systems, it might be possible to perform PDTs hydraulically, eliminating the need for extra equipment and energy consumption. Moreover, performing CTs once or twice per year should not add to the cost or operational complexity of the membrane system. MS2 bacteriophages are easily cultivated in the laboratory, are non-pathogenic to humans and animals (see section 2.4.1), and are easily inactivated by the residual chlorine dose typically applied for secondary disinfection. Therefore, challenge testing with MS2 could be done without interruption of operation.
6.0 CONCLUSIONS

From the results of the present study, it was concluded that:

1) The biofilm layer contributes with more than 2.0 log to the LRV for viruses of passive membrane systems. As a consequence, passive membrane systems are capable of constantly achieving an LRV for viruses of more than 4.0 log.

2) The biofilm layer is capable of mitigating the impact of integrity breaches in the range of breach sizes considered. When a breach is present, the biofilm layer likely grows on top of the breach, acting as a secondary barrier to contaminants that would otherwise bypass treatment by flowing through the breach.

3) The virus removal capacity of the biofilm layer is not affected by the density and/or thickness of the biofilm layer, for the range of conditions investigated.

4) The virus removal capacity of the biofilm layer is not affected by the application of weekly PDTs.

5) The standard PDT approach cannot adequately estimate the LRVs of passive membrane systems for small material such as viruses. In order to estimate the LRVs of breached passive membrane systems for small material, a modified PDT approach that takes into account the additional removal provided by the biofilm layer is required.
References


Appendix A – Estimation of Breach Diameter Based on the Darcy-Weisbach Equation

It can be assumed that a breach behaves as a tube with a determined length and with entrance and exit losses. According to the Darcy-Weisbach equation, the head loss \( h_L \) through a breach can be described using Equation 26.

\[
h_L = f \frac{L}{D_b} \frac{v_a^2}{2g} + k_\text{in} \frac{v_a^2}{2g} + k_\text{out} \frac{v_a^2}{2g}
\]

For the above, \( f \) is the Darcy friction factor; \( L \) is the length of the breach; \( D_b \) is the diameter of the breach; \( v_a \) is the air velocity through the breach; \( g \) is the acceleration of gravity; and \( k_\text{in} \) and \( k_\text{out} \) are the entrance and exit loss coefficients, respectively.

Assuming laminar flow and cylindrical breach, the Darcy friction factor can be calculated according to Equation 27.

\[
f = \frac{64}{Re}
\]

For the above, \( Re \) is the Reynolds number, which can be calculated according to Equation 28 for flow in a pipe.

\[
Re = \frac{v_a D_b}{\gamma_a}
\]

For the above, \( v_a \) is the air velocity through the breach; \( D_b \) is the diameter of the breach; and \( \gamma_a \) is the air kinematic viscosity.

Equation 26 can be rewritten using Equation 27 and Equation 28, resulting in Equation 29.

\[
h_L = \frac{32L\gamma_a v_a}{D_b^2 g} + (k_\text{in} + k_\text{out}) \frac{v_a^2}{2g}
\]
For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( v_a \) is the air velocity through the breach; \( D_b \) is the diameter of the breach; \( g \) is the acceleration of gravity; and \( k_{in} \) and \( k_{out} \) are the entrance and exit loss coefficients, respectively.

Assuming that the air velocity through the breach can be represented by the average air velocity, \( v_a \) can be calculated using Equation 30.

\[
\text{Equation 30}
\]

\[
v_a = v_{avg} = \frac{Q_{\text{air},avg}}{A_b} = \frac{4Q_{\text{air},avg}}{\pi D_b^2}
\]

For the above, \( v_{avg} \) is the average air velocity through the breach; \( Q_{\text{air},avg} \) is the average air flow through the breach; \( A_b \) is the cross-sectional area of the breach; and \( D_b \) is the diameter of the breach.

Equation 29 can be rewritten using Equation 30, resulting in Equation 31.

\[
\text{Equation 31}
\]

\[
h_L = \frac{128L\gamma_a Q_{\text{air},avg}}{\pi D_b^4 g} + \frac{8(k_{in} + k_{out})Q_{\text{air},avg}^2}{\pi^2 D_b^4 g}
\]

For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( Q_{\text{air},avg} \) is the average air flow through the breach; \( D_b \) is the diameter of the breach; \( g \) is the acceleration of gravity; and \( k_{in} \) and \( k_{out} \) are the entrance and exit loss coefficients, respectively.

Using Boyle’s Law and assuming that the average air flow \( (Q_{\text{air},avg}) \) occurs half-way through the breach, the \( Q_{\text{air},avg} \) during a PDT can be calculated using Equation 32.

\[
\text{Equation 32}
\]

\[
P_1 V_1 = P_2 V_2 \Rightarrow \frac{(P_{\text{test}} + P_{\text{atm}})}{2} Q_{\text{air},avg} = P_{\text{atm}} Q_{\text{air},b} \Rightarrow
\]

\[
Q_{\text{air},avg} = \frac{2P_{\text{atm}} Q_{\text{air},b}}{P_{\text{test}} + P_{\text{atm}}}
\]

For the above, \( P_{\text{atm}} \) is the atmospheric pressure; \( Q_{\text{air},b} \) is the air flow out through the breach; and \( P_{\text{test}} \) is the average test pressure during the PDT.

Equation 31 can be rewritten using Equation 32, resulting in Equation 33.
Equation 33

\[
h_L = \frac{128L\gamma_a}{\pi D_b^4 g} \frac{2P_{atm}Q_{air,b}}{P_{test} + P_{atm}} + \frac{8(k_{in} + k_{out})}{\pi^2 D_b^4 g} \left( \frac{2P_{atm}Q_{air,b}}{P_{test} + P_{atm}} \right)^2
\]

For the above, \(L\) is the length of the breach; \(\gamma_a\) is the air kinematic viscosity; \(P_{atm}\) is the atmospheric pressure; \(Q_{air,b}\) is the air flow out through the breach; \(D_b\) is the diameter of the breach; \(g\) is the acceleration of gravity; \(P_{test}\) is the average test pressure during the PDT; and \(k_{in}\) and \(k_{out}\) are the entrance and exit loss coefficients, respectively.

The head loss through the breach \((h_L)\) during a PDT can also be calculated using Equation 34

Equation 34

\[
h_L = \frac{P_{test} - P_{atm}}{\rho_a g}
\]

For the above, \(P_{test}\) is the average test pressure during the PDT; \(P_{atm}\) is the atmospheric pressure; \(\rho_a\) is the air density; and \(g\) is the acceleration of gravity.

Equation 33 and Equation 34 can be equated, resulting in Equation 35.

Equation 35

\[
\frac{P_{test} - P_{atm}}{\rho_a g} = \frac{128L\gamma_a}{\pi D_b^4 g} \frac{2P_{atm}Q_{air,b}}{P_{test} + P_{atm}} + \frac{8(k_{in} + k_{out})}{\pi^2 D_b^4 g} \left( \frac{2P_{atm}Q_{air,b}}{P_{test} + P_{atm}} \right)^2
\]

For the above, \(P_{test}\) is the average test pressure during the PDT; \(P_{atm}\) is the atmospheric pressure; \(L\) is the length of the breach; \(\rho_a\) is the air density; \(\gamma_a\) is the air kinematic viscosity; \(Q_{air,b}\) is the air flow out through the breach; \(D_b\) is the diameter of the breach; \(g\) is the acceleration of gravity; and \(k_{in}\) and \(k_{out}\) are the entrance and exit loss coefficients, respectively.

Equation 35 can be rearranged for \(D_b\), and the breach diameter can be estimated using Equation 36.

Equation 36

\[
D_b = \left\{ \frac{\rho_a P_{atm}Q_{air,b}}{\pi(P_{test}^2 - P_{atm}^2)} \left[ \frac{256L\gamma_a}{\pi(P_{test}^2 - P_{atm}^2)} + \frac{32(k_{in} + k_{out})P_{atm}Q_{air,b}}{\pi(P_{test} + P_{atm})} \right] \right\}^{1/4}
\]

For the above, \(k_{in}\) and \(k_{out}\) are the entrance and exit loss coefficients, respectively; \(P_{atm}\) is the atmospheric pressure; \(\rho_a\) is the air density; \(Q_{air,b}\) is the air flow out through the breach; \(P_{test}\) is the
average test pressure during the PDT; \(L\) is the length of the breach; and \(\mu_a\) is the air dynamic viscosity.

If entrance and exit losses are disregarded, the breach diameter can be estimated using Equation 37.

\[
D_b = \left[ \frac{256L\mu_aP_{atm}Q_{air,b}}{\pi(P_{test}^2 - P_{atm}^2)} \right]^{1/4}
\]

For the above, \(Q_{air,b}\) is the air flow out through the breach; \(L\) is the length of the breach; \(\mu_a\) is the air dynamic viscosity; \(P_{atm}\) is the atmospheric pressure; and \(P_{test}\) is the average test pressure during the PDT.

If the loss through the length of the breach is disregarded, the breach diameter can be estimated using Equation 38.

\[
D_b = \sqrt{\frac{P_{atm}Q_{air,b}}{\pi} \left[ \frac{32(k_{in} + k_{out})\rho_a}{(P_{test}^2 - P_{atm}^2)(P_{test} + P_{atm})} \right]^{1/4}}
\]

For the above, \(P_{atm}\) is the atmospheric pressure; \(Q_{air,b}\) is the air flow out through the breach; \(k_{in}\) and \(k_{out}\) are the entrance and exit loss coefficients, respectively; \(\rho_a\) is the air density; and \(P_{test}\) is the average test pressure during the PDT.
Appendix B – Comparison of Approaches for Estimating Breach Diameter

The Darcy-Weisbach and the Wang et al. (2015) approaches were used to estimate the breach sizes of the breached membrane modules used in the present study. As described in section 4.1.5, the membrane modules were submitted to PDTs prior to breaching to establish their diffusive air flows. After breaching, the modules were again submitted to PDTs and the air flows through the breaches were determined using Equation 10. The air flows were then converted into diameters and the results are presented in Table 13.

Table 13 – Estimated breach sizes based on air flow through the breaches using the Darcy-Weisbach and the Wang et al. (2015) approaches.

<table>
<thead>
<tr>
<th>Measured breach dimensions (µm)</th>
<th>Estimated breach diameter (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(1) Darcy-Weisbach (complete)</td>
</tr>
<tr>
<td>21 x 21</td>
<td>18</td>
</tr>
<tr>
<td>21 x 21</td>
<td>18</td>
</tr>
<tr>
<td>21 x 21</td>
<td>18</td>
</tr>
<tr>
<td>44 x 40</td>
<td>28</td>
</tr>
<tr>
<td>44 x 42</td>
<td>28</td>
</tr>
<tr>
<td>47 x 48</td>
<td>28</td>
</tr>
<tr>
<td>180 x 190</td>
<td>77</td>
</tr>
<tr>
<td>150 x 170</td>
<td>76</td>
</tr>
<tr>
<td>168 x 170</td>
<td>76</td>
</tr>
</tbody>
</table>

As illustrated in Table 13, approaches 1 and 2 were able to accurately estimate the diameters of breaches of approximately 20µm, but failed to estimate the diameters of breaches of approximately 40µm and 180µm. Approach 3 failed to estimate the diameters of all breaches. Approach 4 was able to accurately estimate the diameters of all breaches.

As breach size increases, the length/diameter ratio decreases. The breaches become closer to an orifice, making approach 4 the most appropriate. For diameters of 20µm or less, approaches 1 and 2 can be used since the length/diameter ratio is high and the breach is similar to a tube. Approach 3 is not appropriate for any breach size, as the loss through the length of the breach is the most significant loss and cannot be disregarded.
Appendix C – Estimation of Water Flow Through Breach from Pressure Decay Tests (ASTM Approach)

It is possible to estimate the water flow through a breach ($Q_{water,b}$) based on the air flow through the breach ($Q_{air,b}$) measured during a PDT. The approach described in the American Society for Testing and Materials (ASTM) Standard models the breach as a tube and use the Darcy-Weisbach equation to calculate the head loss through the breach for both air and liquid flows (ASTM Standard D6908, 2003).

For air flow through a breach, according to the Darcy-Weisbach equation, the head loss ($h_L$) can be calculated using Equation 39.

$$h_L = f \frac{L}{D_b} \frac{v_a^2}{2g} + k_{in} \frac{v_a^2}{2g} + k_{out} \frac{v_a^2}{2g}$$

For the above, $f$ is the Darcy friction factor; $L$ is the length of the breach; $D_b$ is the diameter of the breach; $v_a$ is the air velocity through the breach; $g$ is the acceleration of gravity; and $k_{in}$ and $k_{out}$ are the entrance and exit loss coefficients, respectively.

Assuming laminar flow and cylindrical breach, the Darcy friction factor can be calculated using Equation 40.

$$f = \frac{64}{Re}$$

For the above, $Re$ is the Reynolds number.

For flow in a tube, the Reynolds number can be calculated using Equation 41.

$$Re = \frac{v_a D_b}{\gamma_a}$$

For the above, $v_a$ is the air velocity through the breach; $D_b$ is the diameter of the breach; and $\gamma_a$ is the air kinematic viscosity.

Equation 39 can be rewritten using Equation 40 and Equation 41, resulting in Equation 42.
Equation 42

\[ h_L = \frac{32LY_a v_a}{D_b^2 g} + (k_{in} + k_{out}) \frac{v_a^2}{2g} \]

For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( v_a \) is the air velocity through the breach; \( k_{in} \) and \( k_{out} \) are the entrance and exit loss coefficients, respectively; \( D_b \) is the diameter of the breach; and \( g \) is the acceleration of gravity.

According to the ASTM protocol, entrance and exit losses can be disregarded (ASTM Standard D6908, 2003). Thus, Equation 42 can be rewritten as Equation 43.

Equation 43

\[ h_L = \frac{32LY_a v_a}{D_b^2 g} \]

For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( v_a \) is the air velocity through the breach; \( D_b \) is the diameter of the breach; and \( g \) is the acceleration of gravity.

Assuming that the air velocity through the breach (\( v_a \)) can be represented by the average air velocity (\( v_{avg} \)), \( v_a \) can be calculated using Equation 44.

Equation 44

\[ v_a = v_{avg} = \frac{Q_{air,avg}}{A_b} = \frac{4Q_{air,avg}}{\pi D_b^2} \]

For the above, \( v_{avg} \) is the average air velocity through the breach; \( Q_{air,avg} \) is the average air flow through the breach; \( A_b \) is the cross-sectional area of the breach; and \( D_b \) is the diameter of the breach.

Equation 43 can be rewritten using Equation 44, resulting in Equation 45.

Equation 45

\[ h_L = \frac{128LY_a Q_{air,avg}}{\pi D_b^4 g} \]

For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( Q_{air,avg} \) is the average air flow through the breach; \( D_b \) is the diameter of the breach; and \( g \) is the acceleration of gravity.

Using Boyle’s Law and assuming that the average air flow (\( Q_{air,avg} \)) occurs half-way through the breach, the \( Q_{air,avg} \) during a PDT can be calculated using Equation 46.
Equation 46

\[ P_1 V_1 = P_2 V_2 \rightarrow \frac{(P_{test} + P_{atm})}{2} Q_{air,avg} = P_{atm} Q_{air,b} \rightarrow \]

\[ Q_{air,avg} = \frac{2P_{atm} Q_{air,b}}{P_{test} + P_{atm}} \]

For the above, \( P_{atm} \) is the atmospheric pressure; \( Q_{air,b} \) is the air flow out through the breach; and \( P_{test} \) is the average test pressure during the PDT.

Equation 45 can be rewritten using Equation 46, resulting in Equation 47.

Equation 47

\[ h_L = \frac{128 L \gamma_a 2P_{atm} Q_{air,b}}{\pi D_b^4 g \left( P_{test} + P_{atm} \right)} \]

For the above, \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( P_{atm} \) is the atmospheric pressure; \( Q_{air,b} \) is the air flow out through the breach; \( D_b \) is the diameter of the breach; \( g \) is the acceleration of gravity; and \( P_{test} \) is the average test pressure during the PDT.

The head loss through the breach (\( h_L \)) during a PDT can also be calculated using Equation 48.

Equation 48

\[ h_L = \frac{P_{test} - P_{atm}}{\rho_a g} \]

For the above, \( P_{test} \) is the average test pressure during the PDT; \( P_{atm} \) is the atmospheric pressure; \( \rho_a \) is the air density; and \( g \) is the acceleration of gravity.

Equation 47 and Equation 48 can be equated, resulting in Equation 49.

Equation 49

\[ \frac{P_{test} - P_{atm}}{\rho_a} = \frac{128 L \gamma_a 2P_{atm} Q_{air,b}}{\pi D_b^4 \left( P_{test} + P_{atm} \right)} \]

For the above, \( P_{test} \) is the average test pressure during the PDT; \( P_{atm} \) is the atmospheric pressure; \( \rho_a \) is the air density; \( L \) is the length of the breach; \( \gamma_a \) is the air kinematic viscosity; \( P_{atm} \) is the atmospheric pressure; \( Q_{air,b} \) is the air flow out through the breach; and \( D_b \) is the diameter of the breach.
Equation 49 can be rewritten in terms of water flow through the breach \((Q_{\text{water,b}})\) during permeation, instead of air flow through the breach during a PDT. The pressure difference \(P_{\text{test}} - P_{\text{atm}}\) becomes transmembrane pressure \((\text{TMP})\); the terms that represent \(Q_{\text{air,avg}}\) become water flow through breach \((Q_{\text{water,b}})\); and the air density and air kinematic viscosity become water density and water kinematic viscosity \((\rho_w\) and \(\gamma_w\)). This results in Equation 50.

\[
\frac{(\text{TMP})}{\rho_w} = \frac{128L\gamma_w}{\pi D_b^4} Q_{\text{water,b}}
\]

For the above, \(\text{TMP}\) is the transmembrane pressure; \(L\) is the length of the breach; \(\gamma_w\) is the water kinematic viscosity; \(Q_{\text{water,b}}\) is the water flow through the breach; \(\rho_w\) is the water density; and \(D_b\) is the diameter of the breach.

Equation 49 and Equation 50 can be rearranged, resulting in Equation 51 and Equation 52, respectively.

\[
\frac{128L}{\pi D_b^4} = \frac{(P_{\text{test}}^2 - P_{\text{atm}}^2)}{2P_{\text{atm}}Q_{\text{air,b}}\rho_a \gamma_a}
\]

Equation 52

\[
\frac{128L}{\pi D_b^4} = \frac{(\text{TMP})}{Q_{\text{water,b}}\gamma_w \rho_w}
\]

Equation 51 and Equation 52 can be equated and rearranged for \(Q_{\text{water,b}}\), resulting in Equation 53.

\[
\frac{(P_{\text{test}}^2 - P_{\text{atm}}^2)}{2P_{\text{atm}}Q_{\text{air,b}}\rho_a \gamma_a} = \frac{(\text{TMP})}{Q_{\text{water,b}}\gamma_w \rho_w}
\]

Equation 53

\[
Q_{\text{water,b}} = \frac{2(\text{TMP})\gamma_a \rho_a P_{\text{atm}} Q_{\text{air,b}}}{\gamma_w \rho_w (P_{\text{test}}^2 - P_{\text{atm}}^2)}
\]

For the above, \(\text{TMP}\) is the transmembrane pressure; \(\rho_a\) is the air density; \(\gamma_a\) is the air kinematic viscosity; \(P_{\text{atm}}\) is the atmospheric pressure; \(Q_{\text{air,b}}\) is the air flow through the breach; \(\gamma_w\) is the water kinematic viscosity; \(\rho_w\) is the water density; and \(P_{\text{test}}\) is the average test pressure during the PDT.
The product of density and kinematic viscosity is the dynamic viscosity. Therefore, Equation 53 can be rewritten as Equation 54.

\[
Q_{\text{water},b} = \frac{2(TMP)\mu_a P_{\text{atm}} Q_{\text{air},b}}{\mu_w (P_{\text{test}}^2 - P_{\text{atm}}^2)}
\]

For the above, TMP is the transmembrane pressure; \(\mu_a\) is the dynamic viscosity of air; \(P_{\text{atm}}\) is the atmospheric pressure; \(Q_{\text{air},b}\) is the air flow through the breach; \(\mu_w\) is the dynamic viscosity of water; and \(P_{\text{test}}\) is the average test pressure during the PDT.

Equation 54 can be used to estimate the water flow through a breach based on the air flow through the breach measured during a PDT.
Appendix D – Timer Programming for Control of Membrane Systems

The timer programming for conventional and passive membrane systems is presented in Table 14 and Table 15, respectively. The elements of the systems that are connected to the timers are presented in Table 16.

Table 14 – Timer programming for passive systems (24h cycle).

<table>
<thead>
<tr>
<th>Time point</th>
<th>Outlet ID</th>
<th>Stage</th>
<th>Duration (min)</th>
<th>Interval (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>2</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>On</td>
</tr>
<tr>
<td>3</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>4</td>
<td>Off</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>5</td>
<td>Off</td>
<td>On</td>
<td>On</td>
<td>Off</td>
</tr>
<tr>
<td>6 = 1</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
</tbody>
</table>

Table 15 – Timer programming for conventional system (1h cycle).

<table>
<thead>
<tr>
<th>Time point</th>
<th>Outlet ID</th>
<th>Stage</th>
<th>Duration (min)</th>
<th>Interval (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>2</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>On</td>
</tr>
<tr>
<td>3</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>4</td>
<td>Off</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
</tr>
<tr>
<td>5</td>
<td>Off</td>
<td>On</td>
<td>On</td>
<td>Off</td>
</tr>
<tr>
<td>6 = 1</td>
<td>On</td>
<td>Off</td>
<td>Off</td>
<td>Off</td>
</tr>
</tbody>
</table>

Table 16 – Outlet IDs and correspondent system components.

<table>
<thead>
<tr>
<th>Outlet ID</th>
<th>System component</th>
</tr>
</thead>
<tbody>
<tr>
<td>T1</td>
<td>Feed valve</td>
</tr>
<tr>
<td>T2</td>
<td>Compressed air valve</td>
</tr>
<tr>
<td>T3</td>
<td>Drain valve</td>
</tr>
<tr>
<td>T4</td>
<td>Permeate pump</td>
</tr>
<tr>
<td>T5</td>
<td>Backwash pump</td>
</tr>
</tbody>
</table>
Appendix E – Membrane Breaching Experiments

E.1 Breaching Experiment I: Breach Diameter

The goal of the first breaching experiment was to define the laser configurations that achieved the desired breach diameters of 20, 40 and 180µm. Twenty-four membrane modules were potted, cleaned and taken to 4D Labs for breaching (stored in leak-proof plastic containers filled with DI water). After breaching, the breach dimensions were measured using the laser built-in microscope. All configurations tested and the resulting breach dimensions are presented in Table I7.

Table I7 – Breaching Experiment I: laser configurations tested and resulting breach dimensions measured with microscope.

<table>
<thead>
<tr>
<th>Target breach diameter (µm)</th>
<th>Module #</th>
<th>Aperture (µm)</th>
<th>Time (s)</th>
<th>Measured breach dimensions (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>1</td>
<td>50</td>
<td>1</td>
<td>24 x 24</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>50</td>
<td>5</td>
<td>25 x 25</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>100</td>
<td>0.5</td>
<td>21 x 18</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>100</td>
<td>1</td>
<td>24 x 21</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>100</td>
<td>1</td>
<td>24 x 22</td>
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<td>6</td>
<td>100</td>
<td>1</td>
<td>24 x 24</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>100</td>
<td>2</td>
<td>24 x 24</td>
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<tr>
<td></td>
<td>8</td>
<td>100</td>
<td>5</td>
<td>24 x 24</td>
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<tr>
<td>40</td>
<td>9</td>
<td>300</td>
<td>0.1</td>
<td>35 x 35</td>
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<tr>
<td></td>
<td>10</td>
<td>300</td>
<td>1</td>
<td>36 x 36</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>300</td>
<td>1</td>
<td>36 x 36</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>500</td>
<td>1</td>
<td>41 x 44</td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>600</td>
<td>1</td>
<td>57 x 51</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>600</td>
<td>2</td>
<td>63 x 51</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>700</td>
<td>1</td>
<td>63 x 60</td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>1000</td>
<td>1</td>
<td>90 x 88</td>
</tr>
<tr>
<td>180</td>
<td>17</td>
<td>2000</td>
<td>1</td>
<td>123 x 120</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>3000</td>
<td>0.05</td>
<td>143 x 144</td>
</tr>
<tr>
<td></td>
<td>19</td>
<td>3000</td>
<td>1</td>
<td>205 x 216</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>3000</td>
<td>1</td>
<td>210 x 230</td>
</tr>
<tr>
<td></td>
<td>21</td>
<td>3000</td>
<td>2</td>
<td>286 x 265</td>
</tr>
<tr>
<td></td>
<td>22</td>
<td>3200</td>
<td>0.5</td>
<td>265 x 215</td>
</tr>
<tr>
<td></td>
<td>23</td>
<td>3200</td>
<td>0.5</td>
<td>266 x 315</td>
</tr>
<tr>
<td></td>
<td>24</td>
<td>3200</td>
<td>1</td>
<td>246 x 219</td>
</tr>
</tbody>
</table>
Table 18 summarizes the laser configurations that resulted in the approximate desired breach sizes.

<table>
<thead>
<tr>
<th>Target breach diameter (µm)</th>
<th>Aperture (µm)</th>
<th>Time (s)</th>
<th>Measured breach dimensions (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>100</td>
<td>1</td>
<td>24 x 21</td>
</tr>
<tr>
<td>40</td>
<td>500</td>
<td>1</td>
<td>41 x 44</td>
</tr>
<tr>
<td>180</td>
<td>3000</td>
<td>1</td>
<td>205 x 216</td>
</tr>
</tbody>
</table>

E.2 Breaching Experiment II: Breach Depth

The goal of the second breaching experiment was to guarantee that the breaches were piercing entirely through the fibre wall. The approach used in the present study was to study the air flow through breaches ($Q_{air,b}$) of different depths. As breach depth increases, $Q_{air,b}$ is expected to go through 4 stages:

(I) $Q_{air,b}$ increases until the lumen is reached;
(II) $Q_{air,b}$ remains constant until the opposite fibre wall is reached;
(III) $Q_{air,b}$ increases again until the outside of the fibre is reached; and
(IV) $Q_{air,b}$ remains constant.
Based on this, the second breaching experiment was designed to find the drilling times required to produce breaches of 20, 40 and 180µm of diameter and 0.55mm of depth (stage II). Twenty-four membrane modules were potted, cleaned and submitted to PDTs to establish their diffusive air flows. The modules were then taken to 4D Labs for breaching (stored in leak-proof plastic containers filled with DI water). The modules were separated in 3 groups:

1. 8 modules breached with 100µm of aperture and time ranging from 1 to 35s;
2. 8 modules breached with 500µm of aperture and time ranging from 0.5 to 25s; and
3. 8 modules breached with 3,000µm of aperture and time ranging from 0.05 to 10s.

The resulting breach dimensions are presented in Table 19.
Table 19 – Breaching Experiment II: laser configurations tested and resulting breach dimensions measured with microscope.

<table>
<thead>
<tr>
<th>Target breach diameter (µm)</th>
<th>Module #</th>
<th>Aperture (µm)</th>
<th>Time (s)</th>
<th>Measured breach dimensions (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>1</td>
<td>100</td>
<td>1</td>
<td>21 x 21</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>100</td>
<td>5</td>
<td>21 x 24</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>100</td>
<td>10</td>
<td>30 x 24</td>
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<tr>
<td></td>
<td>4</td>
<td>100</td>
<td>15</td>
<td>33 x 27</td>
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<td></td>
<td>5</td>
<td>100</td>
<td>20</td>
<td>39 x 39</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>100</td>
<td>25</td>
<td>42 x 39</td>
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<tr>
<td></td>
<td>7</td>
<td>100</td>
<td>30</td>
<td>39 x 33</td>
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<td></td>
<td>8</td>
<td>100</td>
<td>35</td>
<td>42 x 44</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>500</td>
<td>0.5</td>
<td>33 x 33</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>500</td>
<td>1</td>
<td>41 x 42</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>500</td>
<td>3.5</td>
<td>50 x 45</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>500</td>
<td>6</td>
<td>54 x 54</td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>500</td>
<td>8.5</td>
<td>66 x 72</td>
</tr>
<tr>
<td></td>
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<td>0.05</td>
<td>143 x 144</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>3000</td>
<td>0.1</td>
<td>153 x 149</td>
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<td></td>
<td>19</td>
<td>3000</td>
<td>0.25</td>
<td>116 x 108</td>
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<td>3000</td>
<td>0.5</td>
<td>166 x 200</td>
</tr>
<tr>
<td></td>
<td>21</td>
<td>3000</td>
<td>0.75</td>
<td>209 x 175</td>
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<td>22</td>
<td>3000</td>
<td>1</td>
<td>224 x 222</td>
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<td>23</td>
<td>3000</td>
<td>5</td>
<td>239 x 242</td>
</tr>
<tr>
<td></td>
<td>24</td>
<td>3000</td>
<td>10</td>
<td>224 x 233</td>
</tr>
</tbody>
</table>

After breaching, the modules were again submitted to PDTs to measure their total air flows. The air flows through the breaches were calculated Equation 10 and are presented in Figure 18.
Figure 18 – Air flow through the breaches drilled with 100, 500 and 3000µm laser aperture and variable drilling times. 
(a) 100µm laser aperture (20µm breach) (b) 500µm laser aperture (40µm breach) (c) 3,000µm (180µm breach)
For the 100µm laser aperture (Figure 18a), $Q_{\text{air,b}}$ was initially constant (from 0 to 10s) and then started to increase (from 10 to 35s). For the 500µm laser aperture (Figure 18b), $Q_{\text{air,b}}$ initially increased (from 0 to 1s), became constant (from 1 to 8.5s) and increased again (from 8.5 to 25s). For the 3000µm laser aperture (Figure 18c), $Q_{\text{air,b}}$ initially increased (from 0 to 1s), became constant (from 1 to 5s) and increased again (from 5 to 10s). This indicates that $Q_{\text{air,b}}$ went through stages II and III for the 100µm laser aperture, and through stages I, II and III for the 500 and 3000µm laser apertures. Thus, the drilling times required to breach through one wall of a membrane fibre were in the ranges of 1 to 10s for a 100µm aperture, 1 to 8.5s for a 500µm aperture and 1 to 5s for a 3000µm aperture.

However, it is important to note that the measured breach dimensions increased as drilling time increased, despite the constant laser aperture (Table 19). The longer exposure time led to increased temperatures on the membrane surface around the laser beam, increasing the breach size. Therefore, it is unclear if the increasing $Q_{\text{air,b}}$ presented in Figure 18 is caused by an increase in breach depth, diameter or a combination of both.

In order to select the drilling time that would be applied to the membrane modules of the experimental set-up, the results from both membrane breaching experiments were evaluated together. It was decided that a drilling time of 1s would be applied to all modules, as this value resulted in breaches with the desired diameters and it is included in the ranges required to drill through an entire fibre wall.
Appendix F – Pressure Transducers Calibration

The pressure transducers were calibrated to provide accurate measurements ranging from 0.5 to 6 psi. The calibration was performed by connecting the pressure transducers to a pressure calibrator and setting it to the desired pressure in psi. The electrical signal generated by the pressure transducer was read with the LabView SignalExpress software. This was done for 3 pressures within the calibration range. The values recorded were used to calculate the slope and intercept of the line that best fit the 3 points. The slope and intercept values were then programmed into the LabView SignalExpress project so that the software automatically converted the electrical signals into pressure in psi. The signals, pressures, slopes and intercepts for Passive System 1, Passive System 2 and Conventional System are presented in Table 20, Table 21 and Table 22, respectively.

Table 20 – Values used for calibration of pressure transducers of Passive System 1.

<table>
<thead>
<tr>
<th>Module 1</th>
<th>Module 2</th>
<th>Module 3</th>
<th>Module 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Signal (V)</td>
<td>Pressure (psi)</td>
<td>Signal (V)</td>
<td>Pressure (psi)</td>
</tr>
<tr>
<td>-0.0027</td>
<td>0.53</td>
<td>-0.0051</td>
<td>0.87</td>
</tr>
<tr>
<td>-0.0170</td>
<td>3.03</td>
<td>-0.0137</td>
<td>2.44</td>
</tr>
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<td>-0.0324</td>
<td>5.93</td>
<td>-0.0220</td>
<td>3.95</td>
</tr>
<tr>
<td>Slope</td>
<td>Intercept</td>
<td>Slope</td>
<td>Intercept</td>
</tr>
<tr>
<td>-181.5</td>
<td>0.012</td>
<td>-182.4</td>
<td>-0.054</td>
</tr>
</tbody>
</table>

Table 21 – Values used for calibration of pressure transducers of Passive System 2.

<table>
<thead>
<tr>
<th>Module 1</th>
<th>Module 2</th>
<th>Module 3</th>
<th>Module 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Signal (V)</td>
<td>Pressure (psi)</td>
<td>Signal (V)</td>
<td>Pressure (psi)</td>
</tr>
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<td>-0.0042</td>
<td>0.74</td>
<td>-0.0045</td>
<td>0.78</td>
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<td>-0.0108</td>
<td>1.94</td>
<td>-0.0124</td>
<td>2.25</td>
</tr>
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<td>-0.0265</td>
<td>4.77</td>
<td>-0.0220</td>
<td>3.99</td>
</tr>
<tr>
<td>Slope</td>
<td>Intercept</td>
<td>Slope</td>
<td>Intercept</td>
</tr>
<tr>
<td>-180.5</td>
<td>-0.011</td>
<td>-183.1</td>
<td>-0.033</td>
</tr>
</tbody>
</table>
Table 22 – Values used for calibration of pressure transducers of the Conventional System.

<table>
<thead>
<tr>
<th>Module 1</th>
<th>Module 2</th>
<th>Module 3</th>
<th>Module 4</th>
</tr>
</thead>
<tbody>
<tr>
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<td>Pressure (psi)</td>
<td>Signal (V)</td>
<td>Pressure (psi)</td>
</tr>
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<td>0.76</td>
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<td>2.04</td>
</tr>
<tr>
<td>0.0224</td>
<td>4.13</td>
<td>0.0223</td>
<td>4.08</td>
</tr>
<tr>
<td>Slope</td>
<td>Intercept</td>
<td>Slope</td>
<td>Intercept</td>
</tr>
<tr>
<td>184.2</td>
<td>-0.005</td>
<td>184.2</td>
<td>-0.025</td>
</tr>
</tbody>
</table>
Appendix G – TOC Analyser Calibration

To create the TOC Analyser calibration curve, 5 standard samples were prepared with concentrations of 0, 5, 10, 15 and 20mg/L of TOC. The samples were placed in the instrument and analysed in the calibration curve mode to create a calibration curve file. This same file was used for all subsequent sample analyses. The calibration curve analysis results are presented in Figure 19 and Figure 20.

Figure 19 – Calibration curve graph generated by the TOC Analyser.

![Calibration Curve Graph](image1)

Figure 20 – Calibration curve table generated by the TOC Analyser.

<table>
<thead>
<tr>
<th>Sl. No.</th>
<th>Std. Conc.</th>
<th>Inv. No.</th>
<th>Area</th>
<th>Mean Area</th>
<th>Remarks</th>
<th>SD Area</th>
<th>CV Area</th>
<th>Val</th>
<th>Excluded</th>
<th>Inv. Vol.</th>
<th>Auto Dil.</th>
<th>Date / Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
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<td>1</td>
<td>1.496</td>
<td>1.102</td>
<td></td>
<td>0.04172</td>
<td>3.75</td>
<td>47</td>
<td>E</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
</tr>
<tr>
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<td>5.000</td>
<td>1</td>
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<td>0.3883</td>
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<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
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</tr>
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<td>3</td>
<td>10.00</td>
<td>1</td>
<td>89.74</td>
<td>89.84</td>
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<td>1.138</td>
<td>1.28</td>
<td>49</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
<td></td>
</tr>
<tr>
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<td>100</td>
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<td></td>
</tr>
<tr>
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<td>51</td>
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<td>1.000</td>
<td>13/12/2018</td>
<td></td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Sl. No.</th>
<th>Std. Conc.</th>
<th>Inv. No.</th>
<th>Area</th>
<th>Mean Area</th>
<th>Remarks</th>
<th>SD Area</th>
<th>CV Area</th>
<th>Val</th>
<th>Excluded</th>
<th>Inv. Vol.</th>
<th>Auto Dil.</th>
<th>Date / Time</th>
</tr>
</thead>
<tbody>
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<td>1.072</td>
<td></td>
<td></td>
<td>0.04172</td>
<td>3.75</td>
<td>47</td>
<td>E</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
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<td>5.000</td>
<td>2</td>
<td>44.20</td>
<td></td>
<td></td>
<td>0.3883</td>
<td>0.87</td>
<td>50</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
<td></td>
</tr>
<tr>
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<td>10.00</td>
<td>2</td>
<td>88.13</td>
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<td>1.138</td>
<td>1.28</td>
<td>49</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>15.00</td>
<td>2</td>
<td>132.0</td>
<td></td>
<td></td>
<td>0.3536</td>
<td>0.27</td>
<td>50</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>20.00</td>
<td>2</td>
<td>176.0</td>
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<td>0.07071</td>
<td>0.04</td>
<td>51</td>
<td>100</td>
<td>1.000</td>
<td>13/12/2018</td>
<td></td>
</tr>
</tbody>
</table>

Figure 20 – Calibration curve table generated by the TOC Analyser.
Appendix H – TMP Results

The transmembrane pressures of the membrane modules of Passive System 1, Passive System 2 and Conventional System measured throughout the study are presented in Figure 21.

Figure 21 – Transmembrane pressures of membrane modules during study.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System
Appendix I – TOC and DOC Results

The organic matter removal in Passive System 1, Passive System 2 and Conventional System was monitored via measurements of total and dissolved organic carbon (TOC and DOC) as described in section 4.2.1.3.

The TOC and DOC concentrations in the feed water, membrane tanks and drain of Passive System 1, Passive System 2 and Conventional System measured throughout the study are presented in Figure 22 and Figure 23, respectively. The DOC/TOC ratios in the feed water, membrane tanks and drain of Passive System 1, Passive System 2 and Conventional System are presented in Figure 24.

Figure 22 – TOC concentrations in feed water, membrane tanks and drain of membrane systems.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System

Figure 23 – DOC concentrations in feed water, membrane tanks and drain of membrane systems.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System
The TOC and DOC concentrations in the feed water ranged from 2.7 to 5.2mg/L and 2.7 to 5.1mg/L, respectively. The DOC/TOC ratio in the feed water remained relatively constant at approximately 1.0, indicating that most of the organic material in the feed water was soluble. This was expected as the Jericho Pond water used for the feed solution was filtered prior to use (see section 4.1.3).

For Passive System 1 and Passive System 2, paired t-tests (99% confidence level) indicated that the TOC concentrations in membrane tanks and drain were greater than that in the feed water (for membrane tanks and drain of Passive System 1, p-values were 2.34E-06 and 2.15E-08, respectively; for Passive System 2, p-values were 3.56E-08 and 2.92E-08). This suggests that there was accumulation of TOC in the membrane tanks during the filtration cycle. Similarly, for Passive System 1 and Passive System 2, paired t-tests (99% confidence level) indicated that the DOC concentrations in membrane tanks and drain were greater than that in the feed water (for membrane tanks and drain of Passive System 1, p-values were 4.05E-06 and 4.12E-06, respectively; for Passive System 2, p-values were 2.72E-06 and 6.97E-07).

However, paired t-tests (99% confidence level) indicated that the TOC concentrations in membrane tanks and drain of Passive System 1 and Passive System 2 were greater than the DOC concentrations (for membrane tanks and drain of Passive System 1, p-values were 5.42E-05 and 1.14E-03, respectively; for Passive System 2, p-values were 9.96E-05 and 4.23E-08). This suggests that the organic matter that accumulated was particulate material.

Moreover, paired t-tests (99% confidence level) indicated that the TOC concentrations in membrane tanks and drain of Passive System 1 were greater than the TOC concentrations in membrane tanks and drain of Passive System 2 (for membrane tanks, p-values were 1.17E-05; for drain, p-values were 2.57E-03). This suggests that there was a greater accumulation of suspended...
organic material in the membrane tank of Passive System 1 than in the membrane tank of Passive System 2.

The accumulation of TOC in the membrane tanks of Passive System 1 and Passive System 2 was likely caused by the long duration of the filtration cycle (24h). It is likely that, over time, TOC concentration in the membrane tank increased due to TOC removal by the membrane modules and/or detachment of biofilm from the surface of the membrane modules. The detachment of biofilm is a possible reason for the greater TOC accumulation in the membrane tank of Passive System 1, since this membrane system had a looser biofilm layer due to the application of pressure decay tests, as previously discussed (see section 5.1.1).

For the Conventional System, paired t-tests (99% confidence level) indicated that the TOC and DOC concentrations in the feed water, membrane tank and drain were similar (for membrane tanks and drain, p-values were 5.52E-03 and 1.20E-02, respectively). This suggests that there was no significant TOC accumulation in the membrane tank of the Conventional System, likely due to the short duration of the filtration cycle (1h).

The DOC removal efficiencies of the membrane modules of Passive System 1, Passive System 2 and Conventional System are presented in Figure 25. The DOC/TOC ratio in the permeates of the membrane modules of Passive System 1, Passive System 2 and Conventional System is presented in Figure 26.

Figure 25 – DOC removal efficiency per membrane module.  
(a) Passive System 1 (b) Passive System 2 (c) Conventional System
The DOC removal efficiency of all membrane systems was highly variable throughout the study. The DOC removal efficiency of the membrane modules of Passive System 1 ranged from 0 to 23%, with an average of 7%. The DOC removal efficiency of the membrane modules of Passive System 2 also ranged from 0 to 23%, with an average of 7%. The DOC removal efficiency of the membrane modules of the Conventional System ranged from 0 to 25%, with an average of 7%. These values suggest that there was no significant difference in DOC removal between membrane modules of Passive System 1, Passive System 2 and Conventional System.

The DOC/TOC ratio in the permeates was approximately 1.0 for all membrane modules. This suggests that particulate organic material was completely removed by all membrane systems. This is in accordance with what is expected for UF membrane systems, as particulate material is defined as any material with size larger than 0.45µm (approximately ten times larger than the pore size of the ZeeWeed 500® membranes used in the present study).

Low organic matter removal for conventional UF membrane systems is expected (Schafer, 2001), as the pore size of UF membranes typically used in drinking water treatment applications is relatively large (see section 2.1). On the other hand, previous studies on GDM filtration using similar feed water (mix of Jericho Pond water and tap water) reported organic matter removal efficiencies of up to 50% (Oka, 2015), likely due to biodegradation by the biofilm layer. The feed water preparation, however, did not include the pre-filtration with a 1.5µm filter (see section 4.1.3) included in the present study. Also, the TOC/DOC concentrations were approximately 1.5 times greater than the TOC/DOC concentrations used for the present study. The differences in feed water preparation and concentration likely led to the differences in organic matter removal between the previous and the present study.
Appendix J – ATP Results

The ATP concentration in the various components of Passive System 1, Passive System 2 and Conventional System was measured weekly during the first 80 days of the study. The measurements were done as described in section 4.2.1.4.

The ATP concentrations in the feed water, membrane tanks and drain of Passive System 1, Passive System 2 and Conventional System are presented in Figure 27.

Figure 27 – ATP concentration in the feed water, membrane tanks and drain of the membrane systems.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System
cATP: cellular adenosine triphosphate.

For Passive System 1 and Passive System 2, paired t-tests (99% confidence level) indicated that the ATP concentration in the feed water and membrane tanks was similar (for Passive System 1, p-value was 0.099; for Passive System 2, p-value was 0.046). Paired t-tests (99% confidence level) also indicated that the ATP concentration in the drain was greater than that in the feed water (for Passive System 1, p-value was 0.004; for Passive System 2, p-value was 0.004). The greater ATP concentration in the drain was likely caused by the detachment of biofilm during the aeration and backwash period that occurred prior to draining.

For the Conventional System, paired t-tests (99% confidence level) indicated that there was no statistical difference in the ATP concentration between the feed water and membrane tanks nor between the feed water and drain (p-values were 0.691 and 0.157, respectively). This was expected, as the membrane modules of the Conventional System did not have a biofilm layer.

Paired t-tests (99% confidence level) also indicated that the ATP concentrations in the drain of Passive System 1 and Passive System 2 were greater than the ATP concentration in the drain of the Conventional System (p-values were 0.0030 and 0.0047, respectively). This is further evidence that
a biofilm layer was present on the membrane modules of Passive System 1 and Passive System 2, but not on the membrane modules of the Conventional System.

The ATP concentrations in the permeates of Passive System 1, Passive System 2 and Conventional System are presented in Figure 28.

Figure 28 – ATP concentration in the permeates of the membrane systems.
(a) Passive System 1 (b) Passive System 2 (c) Conventional System

For Passive System 1, paired t-tests (99% confidence level) indicated that, for the intact membrane module and the membrane modules with breaches of 20 and 40µm, ATP concentrations in the membrane tank and permeates were similar (p-values of 0.014, 0.031 and 0.011, respectively). For the membrane module with a breach of 180µm, ATP concentration in the permeate was greater than in the membrane tank (p-value of 0.001). For Passive System 2, paired t-tests (99% confidence level) indicated that, for the intact membrane module, ATP concentrations in the membrane tank and permeate were similar (p-value of 0.007). For the membrane modules with breaches of 20, 40 and 180µm, ATP concentration in the permeate was greater than in the membrane tank (p-values of 0.001, 0.001 and 0.003, respectively).

For the Conventional System, paired t-tests (99% confidence level) indicated that, for the intact membrane module and the membrane modules with breaches of 20 and 40µm, ATP concentration in the permeate was lower than in the membrane tank (p-values of 0.0003, 0.001 and 0.001, respectively). For the membrane module with a breach of 180µm, ATP concentrations in the membrane tank and permeate were similar (p-value of 0.102).

For Passive System 1 and Passive System 2, permeate collection lasted approximately 23h. During that period, it is possible that contamination and/or re-growth occurred. That is likely what caused
the high ATP concentrations in the permeates of Passive System 1 and Passive System 2. For the Conventional System, permeate collection lasted approximately 50 minutes. Therefore, it is unlikely that substantial contamination and/or regrowth occurred. Thus, the low ATP concentrations in the permeates of the membrane modules of the Conventional System was likely caused by retention of microorganisms by the membranes. The exception was the membrane module with a breach of 180µm, whose relatively higher ATP concentration was likely caused by microorganisms bypassing treatment through the breach.
Appendix K– Results of the Calculation of LRVs from PDTs using the ASTM Approach

K.1 Total Air Flow Through the Membrane Modules

The first step in calculating the LRV from the results of a PDT is to calculate the total air flow through the membrane module ($Q_{\text{air}}$) using Equation 9. The $Q_{\text{air}}$ of an intact membrane module corresponds only to the diffusive air flow through the wetted membrane pores; the $Q_{\text{air}}$ of a breached membrane module corresponds to both the diffusive air flow and the air flow through the breach (see section 2.4.2). The $Q_{\text{air}}$ results for the membrane modules of Passive System 1 and Conventional System are presented in Figure 29.

Figure 29 – Total air flow through the membrane modules.
(a) Passive System 1 (b) Conventional System

As expected, the membrane modules with larger breach sizes had greater $Q_{\text{air}}$ values than the membrane modules with smaller breach sizes or no breaches ($180\mu m > 40\mu m > 20\mu m > \text{Intact}$), as expected. At the start of the study, the $Q_{\text{air}}$ of membrane modules with same breach size but from different systems had similar values. However, as the study progressed, the $Q_{\text{air}}$ of the breached membrane modules from Passive System 1 experienced a greater decrease than the $Q_{\text{air}}$ of the breached membrane modules of the Conventional System. This suggests that the biofilm layer had an impact on the air flow through the breaches. A more detailed discussion on the air flow through the breaches is presented in section K.2.
At the start of the study, the $Q_{air}$ values of the intact membrane modules of Passive System 1 and Conventional System were 6.02E-07 and 5.96E-07 m$^3$/s, respectively. These values are consistent with the diffusive air flow value of 5.76E-07 m$^3$/s (corrected for test pressure and membrane area) reported in a previous study for the ZeeWeed® 500 membranes (Farahbakhsh & Smith, 2004). Over time, the $Q_{air}$ of both passive and conventional intact membrane modules experienced a slight, linear decrease, reaching approximately 90% of its initial value by the end of the study.

Because the decrease occurred in a similar manner for both membrane modules, it was likely not caused by the development of the biofilm layer. Variations in temperature are also not likely to have caused the decrease, as there was not a significant variation in water temperature between the start and the end of the study (18.3°C and 21.8°C, respectively).

As previously discussed, the $Q_{air}$ of the intact membrane modules represents only the diffusive air flow through the membrane pores. The diffusive air flow of a membrane module ($Q_{air,d}$) can be estimated using Equation 55 (Farahbakhsh & Smith, 2004).

Equation 55

$$Q_{air,d} = \frac{DHRT}{P_{atm}} \left( \frac{P_{test}A\pi r^2 \rho_{pore}}{\tau L} \right)$$

For the above, D is the diffusivity constant for air-water system; H is Henry’s law constant for air-water system; R is the universal gas constant; T is temperature; $P_{atm}$ is the atmospheric pressure; $P_{test}$ is the average test pressure; A is the membrane surface area; r is the membrane pore radius; $\rho_{pore}$ is the pore density (number of pores per membrane area); $\tau$ is the pore tortuosity factor; and L is the pore length.

The diffusive air flow through a membrane module depends on temperature, test pressure, membrane area and pore characteristics (see Equation 55). For the present study, temperature, test pressure and membrane area remained relatively constant. It is also unlikely that changes occurred in pore length and tortuosity, as these are intrinsic characteristics of the membrane. It is possible, however, that changes occurred in pore radius and/or pore density due to fouling.
K.2 Air Flow Through the Breaches

The second step in calculating the LRV from the results of a PDT is to calculate the air flow through the breach \( Q_{\text{air, b}} \) using Equation 10. The \( Q_{\text{air, b}} \) values (corrected to 20°C using Equation 11) of the membrane modules of Passive System 1 and Conventional System are presented in Figure 30. The diffusive air flow of the breached membrane modules was assumed to be equal to the diffusive air flow of the intact modules, as all modules were subjected to the same operational conditions (ASTM Standard D6908, 2003).

![Figure 30 – Air flow through the breaches of the membrane modules.](image)

The \( Q_{\text{air, b}} \) of membrane modules with same breach size but from different systems had similar values at the start of the study. As the study progressed, the \( Q_{\text{air, b}} \) of all membrane modules decreased. However, the \( Q_{\text{air, b}} \) of the membrane modules of Passive System 1 experienced a greater decrease than the membrane modules of the Conventional System. Membrane modules with larger breach sizes experienced a greater decrease in \( Q_{\text{air, b}} \) than membrane modules with smaller breach sizes. Overtime, the \( Q_{\text{air, b}} \) reached steady-state values for all breached membrane modules.

To assess if the initial air flows through the breaches \( Q_{\text{air, b, 0}} \) were as expected, the \( Q_{\text{air, b, 0}} \) values of the breached membrane modules of Passive System 1 and Conventional System were compared (Figure 31a). The \( Q_{\text{air, b, 0}} \) values of the breached membrane modules of Passive System 1 and Conventional System were also compared to the expected \( Q_{\text{air, b, 0}} \) values for breaches of 20, 40 and 180µm (Figure 31b), calculated based on Equation 14 (see section 2.4.2.1).
Figure 31 – Initial air flow through breaches for breached membrane modules of Passive System 1 and Conventional System.
(a) Comparison between systems (b) Comparison to expected values

\( Q_{\text{air,b,0}} \): Air flow through breach corrected to 20°C at start of study.

As illustrated in Figure 31a, the \( Q_{\text{air,b,0}} \) values of membrane modules with same breach sizes were similar. Also, as illustrated in Figure 31b, the measured \( Q_{\text{air,b,0}} \) values were similar to those expected according to Equation 14.

Since the measured \( Q_{\text{air,b,0}} \) values were similar, the \( Q_{\text{air,b}} \) values throughout the study were normalized to \( Q_{\text{air,b,0}} \) to facilitate the comparison between the membrane modules. The results are presented in Figure 32.

Figure 32 – Normalized air flow through the breaches of the membrane modules.
(a) Passive System 1 (b) Conventional System.

\( Q_{\text{air,b}}/Q_{\text{air,b,0}} \): Air flow through breach corrected to 20°C and normalized to air flow through the breach on first day of study.
As illustrated in Figure 32, all membrane modules experienced a decrease in $Q_{air,b}$ over time. However, the membrane modules of Passive System 1 experienced a greater decrease in $Q_{air,b}$ than the membrane modules of the Conventional System. Furthermore, the membrane modules with larger breach sizes experienced a greater decrease in $Q_{air,b}$ than the membrane modules with smaller breach sizes.

The decrease in $Q_{air,b}$ observed for the membrane modules of the Conventional System was likely caused by the development of some degree of fouling, even though chemical cleaning was performed. As discussed in section 5.1.2.2, the permeabilities of the membrane modules of the Conventional System were not completely recovered, even after the implementation of Cleaning Protocol C.

As for Passive System 1, the decrease in the air flow through the breaches was likely caused by the development of the biofilm layer on the membrane modules. This suggests that there was development of biofilm either in or on the breaches of the passive membrane modules.

### K.3 Water Flow Through the Breaches

The third step in calculating the LRV from the results of a PDT is to calculate the water flow through the breach ($Q_{water,b}$) using Equation 15 (see section 2.4.2.2). The $Q_{water,b}$ values of the membrane modules of Passive System 1 and Conventional System are presented in Figure 33.

![Figure 33 – Water flow through the breaches of the membrane modules.](image)

(a) Passive System 1 (b) Conventional System

$Q_{water,b}$: Water flow through breach corrected to 20°C.
As expected, the $Q_{\text{water,b}}$ values of the membrane modules with larger breach sizes were greater than the $Q_{\text{water,b}}$ values of the membrane modules with smaller breach sizes. The $Q_{\text{water,b}}$ values of the membrane modules of Passive System I were lower than the $Q_{\text{water,b}}$ values of the membrane modules of the Conventional System. The $Q_{\text{water,b}}$ values of the membrane modules of Passive System I remained relatively constant over time. The $Q_{\text{water,b}}$ values of the membrane modules of the Conventional System were more variable.

According to Equation 15, $Q_{\text{water,b}}$ depends on the TMP and on the air flow through the breaches ($Q_{\text{air,b}}$). At the start of the study, the $Q_{\text{air,b}}$ values for the membrane modules of both systems were similar (see section K.2), but the TMP of the membrane modules of the Conventional System was greater due to its greater permeate flux (see Appendix H). This caused greater $Q_{\text{water,b}}$ values for the membrane modules of the Conventional System than for the membrane modules of Passive System I. As the study progressed, the membrane modules of the Conventional System experienced a decrease in $Q_{\text{air,b}}$ until approximately day 60 of the study (section K.2). However, there was also a substantial increase in TMP between days 0 to 20 and 40 to 60 of the study (see Appendix H). The increase in TMP was enough to overcome the decrease in $Q_{\text{air,b}}$, causing corresponding increases in $Q_{\text{water,b}}$ between days 0 to 20 and 40 to 60 of the study. After day 60 of the study, both TMP and $Q_{\text{air,b}}$ of the membrane modules of the Conventional System remained relatively constant. As a consequence, the $Q_{\text{water,b}}$ of the membrane modules of the Conventional System also remained relatively constant.

For the membrane modules of Passive System I, there was also a decrease in $Q_{\text{air,b}}$ due to the growth of biofilm in/on the breaches (see section K.2). However, there was no decrease in the water flow through the breaches. As the biofilm layer developed on the membrane modules, TMP increased in order to maintain a constant permeate flux (see Appendix H). Therefore, the increase in TMP was sufficient to offset the increase in resistance through the breach generated by the biofilm growth. As a consequence, the water flow through the breaches remained constant.

**K.4 Log Removal Values**

Once the water flows through the breaches ($Q_{\text{water,b}}$) of the membrane modules have been calculated, it is possible to determine the LRVs of the membrane modules using Equation 22 (see section 2.4.2.3). The LRVs of the membrane modules of Passive System I and Conventional System are presented in Figure 34.
As presented in Equation 22 (see section 2.4.2.3), the LRVs are proportional to the ratio between the total water flow through the membrane modules and the water flow through the breaches. For both Passive System 1 and Conventional System, the total water flow was constant because the membrane systems were operated under constant permeate flux (see section 4.1.4). The Conventional System had a greater permeate flux (i.e. 40Lm⁻²h⁻¹) than Passive System 1 (i.e. 5Lm⁻²h⁻¹). Therefore, the total water flow through the membrane modules of the Conventional System was greater than that of Passive System 1. However, the water flows through the breaches of the membrane modules of the Conventional System were also greater than that of Passive System 1 (see section K.3). As a consequence, the LRVs of the Conventional System were similar to those of Passive System 1, for membrane modules with breaches of same size, as illustrated in Figure 34.

Because the total water flow through the membrane modules was constant, the behaviour of the LRVs was predominantly determined by the behaviour of the water flow through the breaches. Increases in the water flow through the breaches generated respective decreases in LRVs throughout the study (e.g. between days 0 and 20, for the Conventional System), and vice-versa. However, similarly to the water flows through the breaches, the LRVs of both membrane systems remained relatively constant over time. For both systems, the LRVs of the membrane modules with breaches of 20µm were approximately 3.0 log; the LRVs of the membrane modules with breaches of 40µm were approximately 2.0 log; and the LRVs of the membrane modules with breaches of 180µm were approximately 1.0 log.