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Enhancing stormwater treatment through ultrafiltration: Impact of cleaning chemicals and backwash duration on membrane efficiency

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ABSTRACT

The effect of chemical cleaning and regular backwashing on the efficiency of an ultrafiltration membrane fouled during stormwater treatment was studied. Increasing backwash time from 30 to 60 s resulted in an increase in productivity by 20%. However, the productivity was highest when a backwash time of 45 s was used (3% higher than using 60 s). Chemical cleaning was carried out using an alkaline solution (NaOH with or without NaOCl) followed by acid washing with HCl. The addition of NaOCl to the cleaning chemical did not significantly increase the efficiency of chemical cleaning, and the average pure water permeability increase was 97 ± 13 LMH bar^{-1} after chemical cleaning with NaOH followed by HCl and 117 ± 15 LMH bar^{-1} after chemical cleaning with NaOH + NaOCl followed by HCl, on average. In addition, reversibility after chemical cleaning was 96 ± 67%, on average. The result from scanning electron microscopy showed that at the end of the experiments, inorganic foulants existed in both the inner layer (feed side) and the outer layer (permeate side) of the membrane.

Key words: dead-end filtration, fouling, permeability, pulsatile fluid flow, runoff, stormwater treatment

HIGHLIGHTS

- Longer backwash time resulted in higher permeability and backwash efficiency.
- Chemical cleaning using NaOH with/without NaOCl followed by acidic cleaning by HCl could recover pure water permeability and reversibility of the ultrafiltration membrane.
- A significant amount of inorganic material was found on the fouling layer.

INTRODUCTION

Stormwater runoff is known to be a contributor of pollutants discharged into water bodies. Stormwater management is necessary for a variety of reasons, particularly because of the environmental damage stormwater can cause, such as disturbing ecosystems and endangering the lives of various species (Prudencio & Null 2018; Levin *et al.* 2020). Due to climate change and its related problems, such as water scarcity, treating stormwater for reuse as a water resource is becoming increasingly important (Barbosa *et al.* 2012). Controlling, storing, treating, and reusing stormwater runoff could result in mitigating water scarcity and also prevent the pollution of natural water sources (Rupak *et al.* 2010). For industries that have high water demands, natural water resources are currently the main water resource, but it might be practical to reuse treated stormwater for industry processes, reducing the pressure on natural waters. This issue is particularly of interest to regions where such resources are limited.

Various centralized or decentralized stormwater treatment systems have been studied at laboratory and pilot scales, as well as being implemented in a full scale (Saraswat *et al.* 2016). The most common systems are blue-green infrastructure, such as ponds, wetlands, and bioretention systems, and infiltration (Karlsson *et al.* 2010; Lange *et al.* 2020). Treatment ability, capacity, area requirement, costs as well as legal requirements are important factors for decision makers to consider when choosing a treatment method.

Membrane treatment, which is known to be an efficient treatment method in the water and wastewater industry (Arévalo *et al.* 2009; Yadav *et al.* 2021; Bilal *et al.* 2022), is a promising option for stormwater treatment. The membrane process has the potential to treat stormwater to a high degree and produces a high-quality permeate, which could be reused for industrial purposes or as a water source for non-potable and potable water applications.

Recently, a number of research studies have been carried out on applications of membranes for the treatment of stormwater. Forward osmosis (Li *et al.* 2014), a gravity-based membrane reactor (Du *et al.* 2019), as well as microfiltration (MF) and ultrafiltration (UF) (Ortega *et al.* 2019) have been tested. In a study by Ortega *et al.* (2019), both MF and UF processes removed total and settleable suspended solids by more than 90%, and fecal and total coliforms were reduced to below 300 colony forming units (CFUs)/100 mL. In comparison to MF, a more stable flux was achieved using UF, and flux decline occurred faster in the MF membrane process. It has been suggested that stormwater treated with UF could be used for irrigation, flushing toilet, or washing streets.

UF membranes can be used for stormwater treatment because of two reasons. First, it has a higher removal efficiency in comparison to MF for the separation of pollutants from stormwater (Ortega *et al.* 2019). Second, UF membranes can separate stormwater pollutants, i.e., total suspended solids (TSS), turbidity, biochemical oxygen demand, and oils, well enough to meet the regulations for reuse (Ortega *et al.* 2019; Kaykhaii *et al.* 2023). However, research about membrane applications for stormwater treatment is still limited. During the membrane filtration process, the particles attach to the membrane surface and very small particles can stick inside the pores, which result in fouling. There are some common methods to disturb and partially remove the fouling layer on membranes, e.g., by physical methods (e.g., backwashing with gas or liquid, and forward flushing) and in combination with chemical cleaning. Depending on the water quality of the feed, the chemicals needed for membrane cleaning may differ (Cardew & Le 1999). Stormwater often contains large and fine-grained mineral particles, metals (Lindfors *et al.* 2020), oils, poly-aromatic hydrocarbons, and other organic micro-pollutants (Müller *et al.* 2020). However, the proportion of particulate organic matter is low in comparison to municipal wastewater (Lindfors *et al.* 2020; Philip *et al.* 2021). Previous studies mainly focused on the application of membranes for drinking water, industrial water, or wastewater treatment. Since the quality of stormwater is significantly different from other types of (waste)water, our study is highly relevant, especially with respect to stormwater reuse for different purposes. The insights that the study at hand provides on methods for membrane cleaning will help to design membrane processes for stormwater treatment, which will open new opportunities and facilitate the application of stormwater as a water resource. Effective membrane cleaning has a positive effect on the flux recovery and membrane lifetime, and it is important to find an efficient method to remove fouling substances from the membrane surface and pores.

The aim of this research study was to evaluate the effects of different backwash durations (varying from 30 to 60 s) and different combinations of cleaning chemicals (NaOH with or without NaOCl followed by HCl) on the efficiency of the membrane process used for stormwater treatment. Filtration productivity, backwash efficiency, as well as pure water reversibility and permeability of the membrane were the parameters assessed. The fouling layer on the membrane surface after the experimental runs was characterized using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX).

MATERIALS AND METHODS

Stormwater characteristics

The stormwater used for the experiments was collected from a manhole of a stormwater sewer located in a commercial and light industrial area in Luleå, Sweden (65°37′09.8″N and 22°03′23.6″E). The catchment area included roads, pavements, parking lots, and industrial buildings. Stormwater runoff samples were collected during two precipitation events in two 1 m^3 tanks, transported to the laboratory, and kept at room temperature until used in the experiments. The recorded stormwater quality values are shown in Table 1. Prior to feeding the stormwater into the UF membrane module, the stormwater in 1 m^3 tank was stirred for 10 min and then a feed sample of approximately 15 L was collected and sieved through a stainless-steel sieve with a pore size of 315 μm. The stormwater was sieved to remove particles bigger than 300 μm as these particles could lead to faster clogging and harm the structure of the UF membrane.

Experimental design

To investigate the effect of backwash duration and different cleaning chemicals on membrane productivity, backwash efficiency, and pure water reversibility and permeability, a total of 10 experiments were carried out (Table 2), four experiments with a backwash duration of 30 s, four experiments with a backwash duration of 45 s, and two experiments

Table 1 | Quality of the pretreated stormwater that was used as the feed in the experiments

Parameter	Concentration range	
TSS (mg/L)	172–226	
pH	6.8–7.5	
Turbidity (NTU)	86–134	
Conductivity (µS/cm)	48–55	
TOC (mg/L)	11–21	
Cl^- (mg/L)	4–26	
Metal content	**Total**	**Dissolved**
Al (µg/L)	4,160–8,600	0.1–50
As (µg/L)	0.7–1.3	0.1–19
Ba (µg/L)	71–129	4–20
Ca (mg/L)	5–13	0.01–12
Cd (µg/L)	<0.05–0.1	0.006–0.03
Co (µg/L)	4–6	0.006–1.4
Cr (µg/L)	7–26	0.04–3.5
Cu (µg/L)	11–34	0.004–5.5
Fe (mg/L)	6–15	0.003–0.4
Hg (µg/L)	<0.02	<0.002–1.6
K (mg/L)	4.6–5.5	0.8–2.8
Mg (mg/L)	4–5.6	0.5–2.5
Mn (µg/L)	195–276	0.3–124
Mo (µg/L)	0.87–1.2	0.3–2.3
Na (mg/L)	3–16	0.8–15
Ni (µg/L)	6–13	0.7–12
Pb (µg/L)	3–10	0.01–1.1
V (µg/L)	19–39	0.9–15
Zn (µg/L)	107–245	15–60

NTU, nephelometric turbidity unit.

with a backwash duration of 60 s. The run order of the 10 experimental runs was fully randomized. The backwash duration was chosen in a range that was reasonable in relation to the membrane treatment time of 1 h. In preliminary experiments, it was found that a backwash duration of 15 s was not sufficient to adequately rinse the membrane surface and remove particles that had weaker bonds with the adsorptive layer on the membrane surface. This was because the transmembrane pressure (TMP) rise was rapid and this backwash duration was not sufficient to remove the contaminants from the membrane surface and the pipes.

To remove the foulants on the membrane surface, membrane chemical cleaning was evaluated using two chemical combinations. NaOH with or without NaOCl was evaluated as a chemical for cleaning of membrane and removing the organic pollutants followed by membrane acidic washing with HCl to remove inorganic pollutants and for neutralization. The alkaline solution was a 1 M solution of NaOH and adjusted to the pH of 12. When chlorine alkaline solution was used, a 200-ppm chlorine solution was prepared with NaOCl in water and NaOH was added to adjust the pH to 12. An HCl solution with a pH of 2 was used for acidic cleaning of the membrane. Table 2 shows which chemical combination is used for each experiment. NaOH and HCl were supplied by Merck and NaOCl by FF-Chemicals AB.

Experimental setup of the UF membrane

The pretreated stormwater was filtered through a UF membrane using a laboratory-scale setup equipped with a hollow fiber membrane module (Figure 1) run in dead-end mode. A hydrophilic UF membrane, consisting of a combination of polyether

Table 2 | Design of backwash duration and chemical cleaning experiments

Exp. no.	1	2	3	4	5	6	7	8	9	10
Backwash duration	30	30	30	30	45	45	45	45	60	60
Chemical	NaOH, NaOCl, HCl	NaOH, HCl	NaOH, HCl	NaOH, NaOCl, HCl	NaOH, NaOCl, HCl	NaOH, HCl	NaOH, HCl	NaOH, HCl	NaOH, HCl	NaOH, NaOCl, HCl

sulfone and polyvinylpyrrolidone, provided by PENTAIR was used in the experiments. Urban runoff can contain small colloids to which metals and small particles can attach, and to separate these small colloids, membrane with molecular weight cut-off of 1 kDa with an active area of 0.07 m^2 was chosen. Three pressure sensors, a data logger, and a computer system were used to store the pressure variation data during the experiment (Figure 1). The stormwater was fed to the membrane system using a peristaltic pump and pulsatile flow at a pulse frequency of 4 Hz. Pulsatile flow at this frequency was found to prolong UF operation efficiently in a previous study (Kaykhaii *et al.* 2023).

Each experiment started by cleaning the membrane with one of the chemical combinations (Table 2). One of the alkaline solutions was added to the feed stream and the membrane was filled with cleaning chemical and left standing for 10 min before the membrane was acid washed using HCl with the same cleaning process. The temperature of the cleaning solution was kept constant (20 °C) for all experiments. After chemical cleaning, the membrane was backwashed for 1 min followed by 2 min of forward washing with Milli-Q water, which has been recommended after chemical cleaning (Chen *et al.* 2003). Pure water flux (PWF) was measured before and after chemical cleaning. Three different pump flow rates were determined, and flux was recorded manually using a beaker and scale 10 times during 10 min (once per minute). The pressure was recorded by pressure sensors during this time. This process was repeated two more times with higher flow rates.

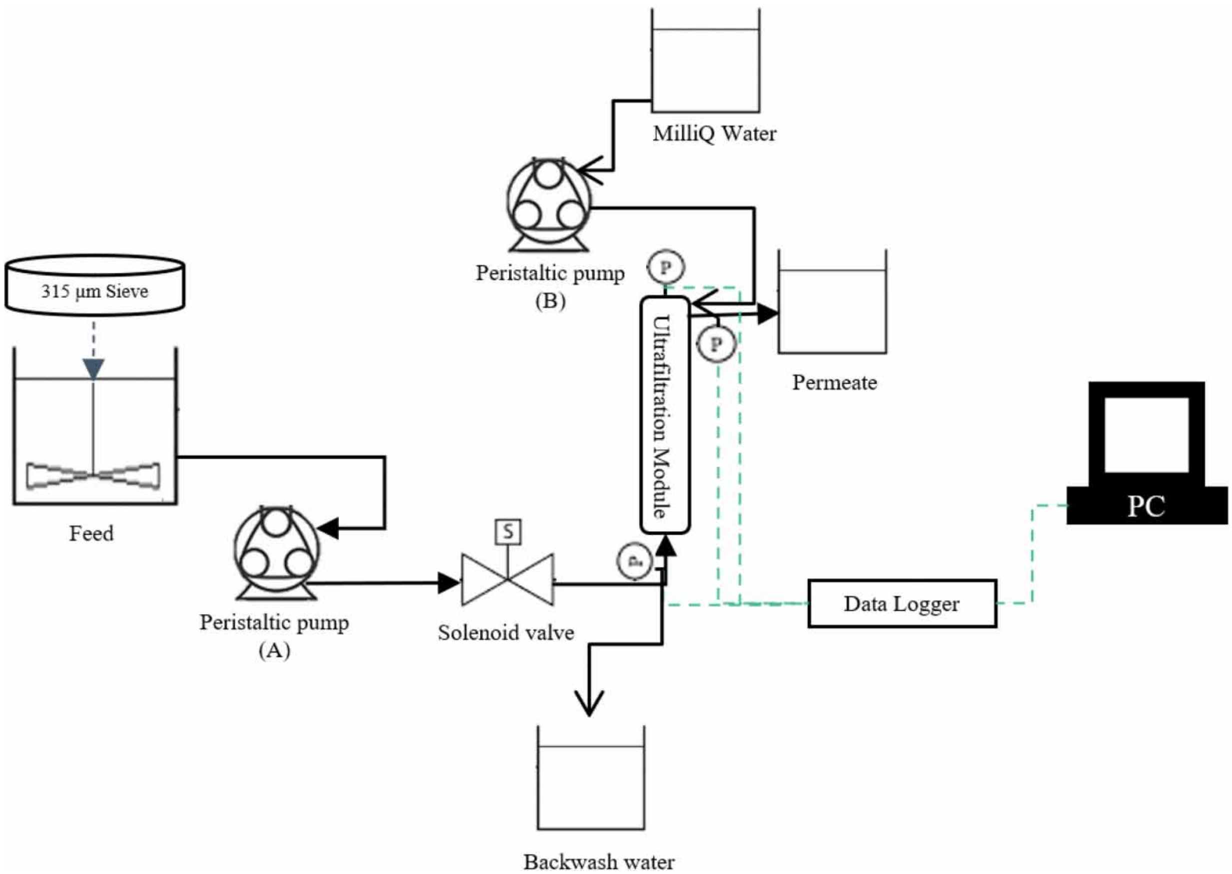

Figure 1 | Experimental setup of the UF membrane process. *P* stands for pressure sensor.

Following this, a TMP of 1.8 bar was set and the experiment was started. After 1 h of stormwater treatment, the permeate volume was measured, and if the average flux had declined by 30% compared to the start of the experiment, the TMP was increased by 0.1 bar. Every hour, the membrane was backwashed using a peristaltic pump (150 ml/min) and Milli-Q water for either 30, 45, or 60 s. These 1-h cycles were repeated until the TMP had increased by 0.3 bar (i.e., from a TMP of 1.8 bar at the beginning to a TMP of 2.1 bar). The permeate flux was measured at the beginning, middle, and end of each 1-h cycle.

Physical and chemical analyses

Feed samples were taken at the beginning of each of the 10 experimental runs and were analyzed with respect to TSS, turbidity, pH, conductivity, total and dissolved metal concentrations, total organic carbon (TOC), and Cl^-. In addition, before starting the experiment, a pretest was carried out using Milli-Q water and a permeate blank sample, which was analyzed for the same parameters as previously described. Samples for analysis of dissolved metals were passed through a 0.45 μm filter. TSS, conductivity, pH, and turbidity were analyzed immediately after sampling. For TOC and Cl^-, samples were stored at −18 °C, and for total and dissolved metals, the samples were stored at 8 °C until taken to the external laboratory for analysis.

The standard method SS-EN 872:2005 was used to determine the concentrations of TSS. Total and dissolved metal concentrations (except Hg) were analyzed using inductively coupled plasma sector filed mass spectrometry (ICP-SFMS) according to ISO 17294-2:2016. For Hg, the standard SS-EN ISO 17852:2008 was used. The reporting limits (RLs) for the total and dissolved metal concentrations were 10 and 0.2 μg/L for Al, 0.5 and 0.05 μg/L for As, 1 and 0.01 μg/L for Ba, 0.2 and 0.1 μg/L for Ca, 0.05 and 0.002 μg/L for Cd, 0.2 and 0.005 μg/L for Co, 1 and 0.1 μg/L for Cu, 0.9 and 0.01 μg/L for Cr, 0.01 and 0.0004 μg/L for Fe, 0.002 and 0.02 μg/L for Hg, 0.4 and 0.4 μg/L for K, 0.2 and 0.09 μg/L for Mg, 0.9 and 0.03 μg/L for Mn, 0.5 and 0.05 μg/L for Mo, 0.5 and 0.1 μg/L for Na, 0.6 and 0.05 μg/L for Ni, 0.5 and 0.01 μg/L for Pb, 1 μg/L for P, 0.2 and 0.005 μg/L for V, and 4 and 0.2 μg/L for Zn. The atomic fluorescence spectroscopy (AFS) method (ISO 17852:2008) was used for Hg analysis, and RLs for total and dissolved Hg concentrations were 0.02 and 0.002 μg/L, respectively. TOC was analyzed according to DIN EN 1484(H3) and the RL was 0.5 mg/L. Turbidity was measured using the turbidity meter 2100N (Hach, Loveland, Colorado), pH was measured using a WTW pH 330 electrode (WTW, Wielheim, Germany), and the conductivity was measured using a CDM210 conductivity meter. Cl^- concentration was measured by ion chromatography according to the method CSN EN ISO10304-1.

The scanning electron microscope (SEM instrument) was provided by PENTAIR, Sweden, to characterize the fouling layer on the membrane at the end of the experiment and was a combined system for SEM and EDX. The scanning electron microscope was JSM-IT500HR type from JEOL.

A module integrity test was carried out by PENTAIR, Sweden, on the module before autopsy. The permeate side was pressurized with air (1–3 bar), while the module was submerged in water. Any leak would have become visible because any compromised fiber would cause a bubble-train.

Data analysis

The productivity (P) of the process, which reflects the balance of operational and backwash duration, was calculated using the following equation:

$$P_{\text{experiment}} = \frac{V_{\text{filtrate}} - V_{\text{backwash}}}{(t_{\text{filtrate}} + t_{\text{backwash}})*A} \tag{1}$$

where P is the productivity (LMH = L/m^2/h) determined for each experiment, V is the volume (L) produced during the experiment, A is the membrane active area (m^2), and t is the time (h). Equation (2) is used to assess the efficiency of backwash (Chellam *et al.* 1998):

$$\eta_{\text{experiment}} = \frac{1}{n}\sum_{k=1}^{n} \frac{P_{\text{final},k} - P_{\text{after backwash},k}}{P_{\text{final},k} - P_{\text{initial},k}} \tag{2}$$

where $\eta_{\text{experiment}}$ is the average backwash efficiency achieved during an experiment, n is the number of 1-h cycles, $P_{\text{initial},k}$ and $P_{\text{final},k}$ are the initial and final feed pressures (bar) for filtration cycle k, and $P_{\text{afterbackwash},k}$ is the pressure after backwashing which is the initial pressure for cycle $k+1$ and averaged throughout each experiment. The pressure sensors recorded pressure 10 times per second, and for calculating feed pressure, the pressure values were averaged over a minute. Reversibility (RF) is a parameter used to compare the flux before and after chemical cleaning with the initial flux for the clean membrane and was calculated according to the following equation (Qu *et al.* 2011):

$$\mathrm{RF} = \frac{J_a - J_b}{J_0 - J_b} \tag{3}$$

where RF is the reversibility, J_a and J_b are the average PWFs after and before cleaning (LMH), and J_0 is the average initial PWF (averaged over three different feed pressures) of the membrane before initiating each experiment.

To assess the effect of chemical cleaning and backwashing on foulant removal, permeability was calculated in two different ways. First, the average permeability for each experiment was calculated using the following equation (Chang *et al.* 2017):

$$L_{p,\,\mathrm{experiment}} = \frac{\bar{J}}{\overline{\mathrm{TMP}}} \tag{4}$$

where L_p is the mean membrane permeability (LMH bar^{-1}) for each experiment, J is the mean permeate flux (LMH) determined by dividing the total volume of permeate by the total filtration time of an experiment, active area of the membrane, and TMP is the mean transmembrane pressure (bar) during the experiment. Using data of PWF to calculate the permeability after chemical cleaning, it was possible to compare the effect of each chemical. The data were used for calculating pure water permeability using the following equation:

$$\bar{L}_{p,\,\mathrm{experiment}} = \frac{1}{n}\sum_{k=1}^{n} \frac{\bar{J}_k}{\mathrm{TMP}_k} \tag{5}$$

where J_k is the PWF after chemical cleaning at cycle k, $n = 3$ (as the PWF was measured at three different pressures), and TMP_k is the TMP measured during 10 min of flux measurement at pressure k.

A partial least square (PLS) model was derived to find the statistical relationship between factors (backwash duration and type of chemical cleaning) and responses (productivity, backwash efficiency, pure water reversibility, and permeability).

RESULTS AND DISCUSSION

Effect of backwash duration

Increasing the backwash duration from 30 to 60 s resulted in a 20% increase in productivity, on average. Optimum productivity (58 LMH), however, was achieved with a backwash duration of 45 s, which was 3% higher than the productivity with a backwash duration of 60 s. Although the experiment could run for a longer time using 60 s of backwash (Table 3),

Table 3 | Backwash efficiency, reversibility, permeability, and productivity determined under different experimental conditions

Backwash duration (s)	Chemical used for cleaning	Productivity (LMH)	Backwash efficiency (%)	Pure water reversibility after chemical cleaning (%)	Permeability (LMH bar^{-1})	Average TMP (bar)
30	NaOH, NaOCl, HCl	39	59	–	29	1.9
30	NaOH, HCl	43	62	83	29	1.8
30	NaOH, HCl	52	63	42	29	1.8
30	NaOH, NaOCl, HCl	50	64	256	34	1.9
45	NaOH, NaOCl, HCl	59	75	54	35	1.7
45	NaOH, HCl	60	88	37	38	1.6
45	NaOH, HCl	58	80	126	39	1.6
45	NaOH, HCl	55	85	83	37	1.6
60	NaOH, HCl	57	89	66	46	1.5
60	NaOH, NaOCl, HCl	56	94	113	48	1.5

the total volume of backwash water that was needed for 60 s of backwash was large and the increase in permeate volume was not considerably higher than when 45 s of backwash took place, which resulted in lower productivity compared to the experiments with a backwash duration of 45 s. According to the PLS model, an increase in backwash duration had a positive effect on productivity ($R^2Y = 0.42$, $Q^2 = 0.2$).

Backwashing and membrane productivity are closely interconnected, and a trade-off between productivity and backwashing is challenging. Increasing the frequency of backwash, backwash duration, or pressure can potentially have a positive or negative effect on membrane productivity. Therefore, it is important to optimize the backwash duration to remove the fouling layer without reducing productivity. The experiments using 60 s of backwash duration were continued for 50 h and stopped at the end of the 50th hour due to not observing TMP increase. If they had been continued until a TMP increase of 0.3 bar (as the other experiments), the productivity would have been even lower. However, a backwash duration of 60 s resulted in a higher backwash efficiency which is beneficial and results in an improved ability to control permeate flux and keep it constant for a longer time. Efficient backwashing helps to reduce the number of chemical cleaning runs required, which is important because chemical cleaning reduces membrane lifetime (Park *et al.* 2018). Therefore, a longer backwash duration can be beneficial despite the trade-off with productivity.

The backwash efficiency (Equation (2)) increased with increasing backwash duration (PLS; $R^2X = 1$, $R^2Y = 0.74$, $Q^2 = 0.3$). The variation of backwash efficiency over time is shown in Figure 2. For 30, 45, and 60 s of backwash, the mean backwash efficiency was 62 ± 2, 82 ± 6, and 92 ± 4%, respectively. As stated above, a backwash duration of 45 s resulted in the highest productivity. Although the backwash efficiency was highest for 60 s of backwash (92 ± 4%), the mean backwash efficiency for 45 s of backwash was high (82 ± 6%) as well. As the experiments with 60 s of backwash duration were stopped after 50 h (without reaching the 0.3 bar of TMP increase), the backwash efficiency for these experiments is probably overestimated. If the 60 s backwash experiments had been continued, the backwash efficiency would have probably been lower. Backwash efficiency is affected by feed water characteristics and temperature (Zhang *et al.* 2020). In our experiment, however, the stormwater was kept at room temperature with very small temperature variations, and the stormwater was thoroughly mixed before use so that the variation in TSS and turbidity was small (Table 1). The effect of turbidity and TSS on the backwash efficiency was detected. To summarize, the positive effect of backwash duration on the backwash efficiency was observed as was observed by the PLS model.

The mean permeability (Equation (4)) was positively affected by an increasing backwash duration ($R^2Y = 0.9$, $Q^2 = 0.65$). For 30, 45, and 60 s, the mean permeability was 30 ± 3, 37 ± 3, and 47 ± 2 LMH bar^{-1}, respectively. This effect might be different for 60 s of backwash if the experiment had been allowed to continue.

The experimental results showed that increasing the duration of regular backwash (every hour) with Milli-Q water from 30 to 60 s gave a better control of the permeate flux through the membrane, which was seen through the better control of TMP,

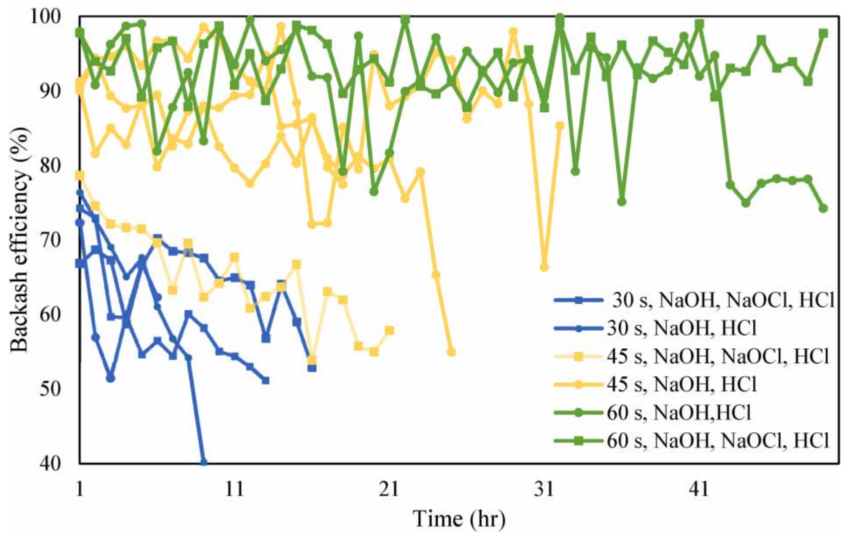

Figure 2 | The variation of backwash efficiency over time using backwash durations 30, 45, and 60 s.

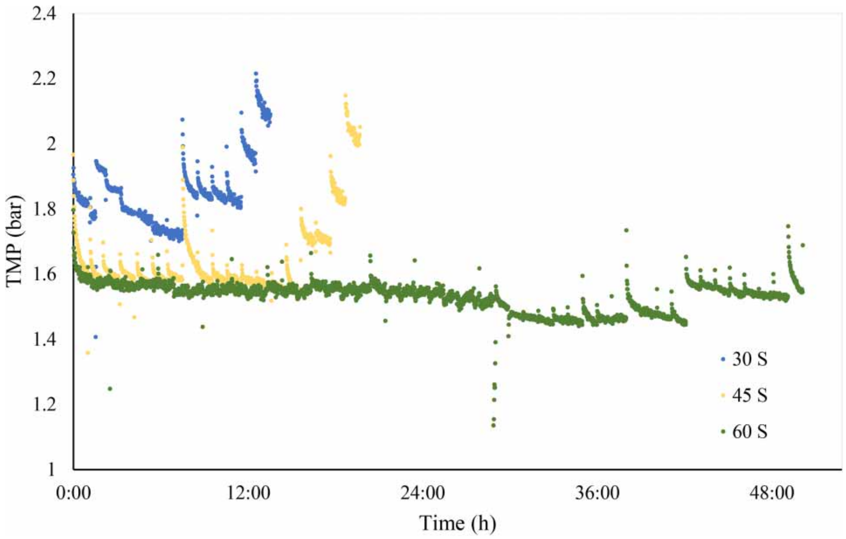

Figure 3 | TMP variation over time during three experiments with 30, 45, and 60 s of backwash. Three experimental runs were chosen randomly for this plot: 30 s with NaOH + NaOCl followed by HCl, 45 s with NaOH followed by HCl, and 60 s with NaOH followed by HCl.

as well. The duration of the experiments with backwash durations of 30 and 45 s averaged 11 and 24 h, respectively. A faster TMP increase because of shorter backwash durations indicated that the backwash was less effective in controlling the formation of a fouling layer on the membrane surface and pores. For the longest investigated backwash duration (60 s), the TMP need not be increased for a long time, up to 50 h (Figure 3), after which the experiment was stopped due to practical reasons, which is an indirect indicator of better control of the pore blockage in the membrane. This is in line with the results of Lüken *et al*. (2020) who showed that during membrane treatment, particles and clusters accumulated at the pore edges and over time, these particles formed a bridge in the pore. This process of pore blockage led to an increase in TMP. In addition, they showed that longer backwash duration was more efficient in removing the particles from the pores and in preventing this bridge formation and eventually pore blockage, something shown by this work as well.

Chemical cleaning

Two chemical combinations were tested for membrane cleaning: NaOH with or without NaOCl, followed by HCl. According to the PLS model, the comparison between the efficiencies of chemical cleaning when the alkaline solution did or did not contain NaOCl showed that the addition of NaOCl had no significant effect (PLS) on chemical cleaning efficiency. Efficiency was evaluated by comparing the pure water permeability (Equation (5)) after chemical cleaning and average reversibility (Equation (3)). Reversibility after chemical cleaning (determined from the PWF after chemical cleaning) varied widely, ranging from 37 to 255% (average 96 ± 67%, $R^2Y = 0.8$, $Q^2 = 0.75$) for the two types of cleaning tested (Table 2). The performance of chemical cleaning depends on the particles adsorbed on the fouling layer, as well as the structure of the fouling layer and how compact this layer was (Chen *et al.* 2003). During chemical cleaning, chemical reactions take place between the chemical and foulants, detaching the particles from the membrane surface.

Pure water permeability was calculated after using the two types of chemical cleaning (NaOH with or without NaOCl followed by HCl). The pure water permeability after cleaning with NaOH followed by HCl averaged 97 ± 13 LMH bar^{-1} and after cleaning with NaOH + NaOCl followed by HCl averaged 117 ± 15 LMH bar^{-1}. Stormwater contains microorganisms, and the addition of chlorine solution to the chemical treatment could be suitable to prevent biological growth on the membrane surface, which is crucial for prolonging the membrane lifetime, especially in applications where biological growth is unavoidable (Decarolis *et al.* 2001). However, chlorine is harmful to the environment, and its use in stormwater treatment should be avoided whenever possible (Parveen *et al.* 2022).

Looking at the contour plot, it can be seen that the aging of the membrane during the experiments negatively affects the ability of the chemical cleaning on recovering pure water permeability. It should be mentioned that according to the PLS model, reversibility was positively correlated with productivity, backwash efficiency, and permeability.

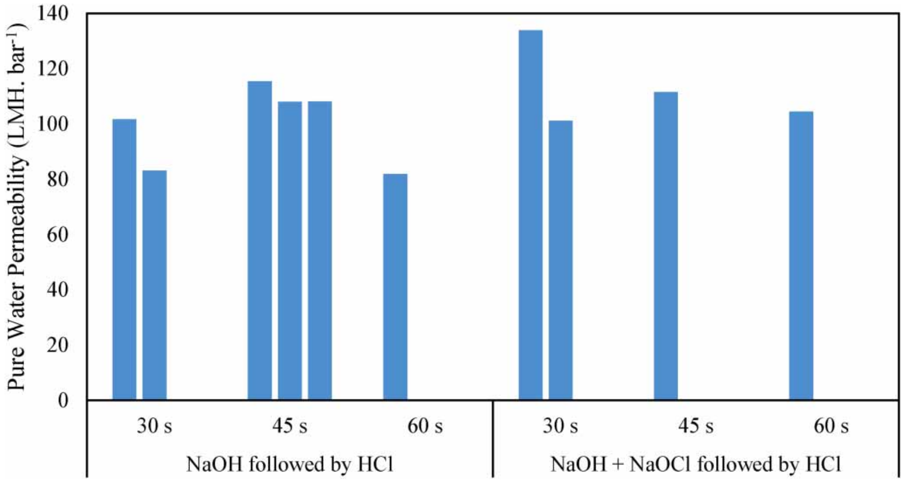

Figure 4 | Pure water permeability after chemical cleaning using the two different chemical combinations.

The efficiency of chemical cleaning depends on the residence time of the chemicals on the membrane surface during the cleaning process, with a longer residence time increasing the efficiency. In the experiments, the duration of the membrane cleaning process was limited to 10 min, because longer cleaning with chlorine could change the properties and lifetime of the membrane. The temperature of the chemical solution was kept constant (room temperature) in all experiments. Jolis *et al.* (1995) and Bartlett *et al.* (1995) showed that increasing the temperature of the chemical solution to 50–60 °C was more effective than using chemicals at room temperature. However, this was not investigated in this study. Figure 4 shows the average pure water permeability after each chemical cleaning. As can be seen, NaOH in combination with NaOCl, followed by HCl, resulted in better pure water permeability after cleaning than cleaning without NaOCl. However, this effect was not statistically significant ($R^2Y = 0.6$, $Q^2 = 0.4$).

SEM–EDX analysis

The integrity test after the experiments showed that the membrane used for these experiments was not leaking and that the pretreatment method chosen was sufficient (in terms of stormwater quality) to prevent damage that could be caused by pollutants or large particles in the stormwater.

Figure 5 shows the inner (feed side), outer (permeate side), and cross-sectional surfaces of the fouled and new membranes at various magnifications. The inner surface of the used membrane was mostly fouled (Figure 5(a)). The membrane had been used in experiments with stormwater for more than 380 h and had been chemically cleaned 22 times during this period (including preliminary stormwater treatment experiments and snowmelt treatment experiments). The adsorption layer on the feed side of the membrane was very compact (Figure 5(a)), which could be due to the fact that the membrane was operated at a high pressure near the maximum allowable TMP for this membrane in this series of experiments. In addition, the SEM image of the outer surface of the membrane was compared to the outer layer of the new membrane (Figure 5(b)). Surprisingly, some foulants were also found on the permeate side of the used membrane, possibly due to precipitation of salts that had passed through the pores of the membrane. No significant changes were observed in the cross-sectional area of the membrane (Figure 5(c)). The EDX data showed that the inner and outer layers of the membrane were fouled with a significant amount of Si and Fe. The amount of O was significantly increased in both the inner and outer layers of the membrane compared to the new membrane (Table 4), indicating accumulation/precipitation of iron oxides and/or hydroxides. This was consistent with the color of the inner membrane surfaces turning brown, also indicating iron precipitation. Iron enters stormwater runoff from a variety of sources such as surface corrosion (Galfi *et al.* 2017; Melliti *et al.* 2019).

From the EDX results, it appears that the foulants on the membrane surface were metals belonging to the group of inorganic substances. In the case of inorganic foulants, the use of citric acid, sodium metabisulfite, sulfamic acid, or sodium metaphosphate may be another option for chemical cleaning of the membrane (Cardew & Le 1999). However, the application of these chemicals and the effects on this type of membrane need further study. It could also be interesting to

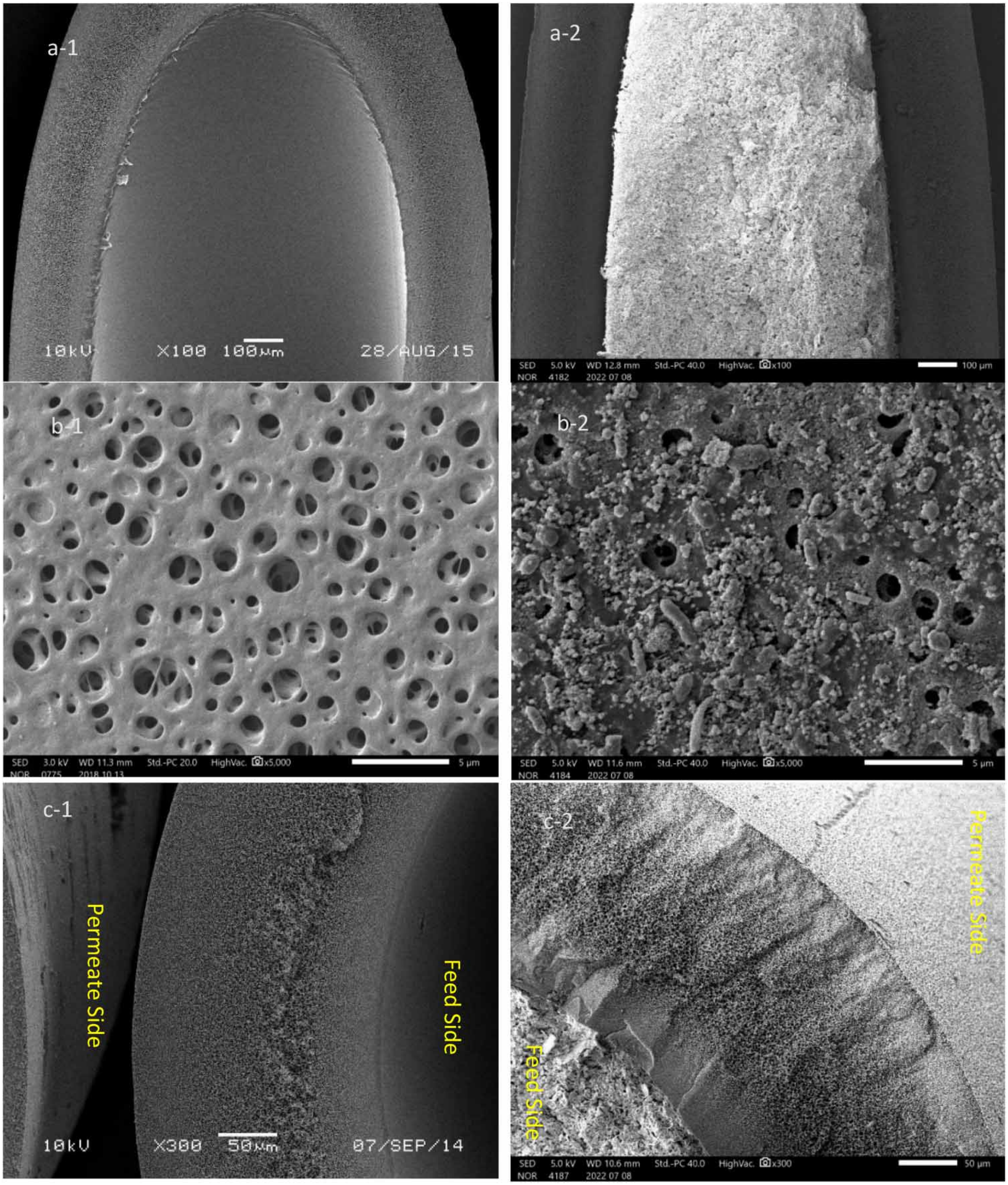

Figure 5 | SEM images from (a) inner surface (feed side), (b) outer surface (permeate side), and (c) cross-section of the UF membrane with magnification (a) 100, (b) 5,000, and (c) 300. The images marked with −1 show the new membrane and −2 the used membrane. The membrane is categorized as the in-to-out membrane, and feed and permeate sides of the membrane are shown in the cross-sectional images (C-1 and C-2).

Table 4 | Proportion (%) of different elements in the fouling layer on the inner and outer membrane surfaces

	Elements (%)												
Parameter	**C**	**O**	**Na**	**Al**	**Si**	**S**	**Cl**	**Ca**	**K**	**Mg**	**Fe**	**Cu**	**Zn**
Inner surface (new membrane)	67	14	–	–	–	19	–	–	–	–	–	–	–
Feed side (used membrane)	21	45	2	5	17	–	–	1	2	1	5	–	–
Permeate side (used membrane)	46	35	–	–	11	–	–	–	–	–	7	–	–

characterize and compare the composition of the different foulants on the membrane surface before and after chemical cleaning and backwashing.

CONCLUSIONS

In this study, the effect of chemical cleaning and regular backwashing on the efficiency of a UF membrane used for stormwater treatment was investigated. The results of the experiments showed that increasing the backwash duration resulted in a more stable TMP over time, higher backwash efficiency (62 ± 2, 82 ± 6, and 92 ± 4% for 30, 45, and 60 s backwash duration, respectively), and permeability (the mean permeability was 30 ± 3, 37 ± 3, and 47 ± 2 LMH bar^{-1} for 30, 45, and 60 s, respectively). Productivity was highest at a backwash duration of 45 s and was 3% higher than at 60 s. Since productivity is affected by backwash duration and flow rate, a balance between productivity, permeability, and energy consumption must be found to find the optimal operating conditions.

The membrane was chemically cleaned before each experiment and two different combinations of chemicals were used. The use of NaOH with or without NaOCl, followed by HCl, resulted in an average pure water membrane reversibility of 96 ± 67%, restoring flux and reducing TMP. The pure water permeability was 97 ± 13 LMH bar^{-1} after cleaning with NaOH and HCl and 117 ± 15 LMH bar^{-1} after cleaning with NaOH + NaOCl followed by HCl, although the difference was not statistically significant according to the PLS model.

The surface of the fouled membrane was examined with a scanning electron microscope. This showed that the fouling layer consisted of mineral particles, e.g., significant amounts of Si and Fe. In addition, small amounts of Si, Al, Na, K, Ca, and Mg were found on the inner surface of the membrane.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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