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**REVERSIBILITY OF FOULING ON ULTRAFILTRATION MEMBRANE
BY BACKWASHING AND CHEMICAL CLEANING:
DIFFERENCES IN ORGANIC FRACTIONS BEHAVIOUR**

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ABSTRACT

Membrane fouling is an inherent phenomenon in UF membrane processes, making it necessary to periodically perform backwashes (BW) and chemical “cleanings in place” (CIP) to restore the initial permeability of the membrane. The objective of this study was 1) to explore systematically the effect of distinct BW-related variables (BW transmembrane pressure, duration, frequency, composition) on the reversibility of UF membrane fouling and on the permeate quality (in terms of total organic carbon, turbidity and UV absorbance) over successive filtration/BW cycles; and 2) to identify which organic fractions were most removed by the membrane and, of these, which were most detached after BW, alkaline and oxidant CIP and acid CIP episodes. For this purpose, a bench-scale outside-in hollow fibre module operated under dead-end filtration mode at constant transmembrane pressure and treating settled water from a drinking water treatment plant was employed. Dissolved organic carbon fractionation was performed by high performance size exclusion chromatography (HPSEC). Results showed that in general the more intensive a BW was (in terms of high transmembrane pressure, shortened frequency and prolonged duration) the more effective it was in removing fouling from the membrane. Concerning the composition of the water used for the BW, the addition of NaClO led to maximum fouling reversibility, closely followed by the combination of NaOH+NaClO, while citric acid and NaOH contributed little compared to water alone. However, results also showed that irreversible fouling was never completely avoided whatever the BW regime applied, leading to a gradual increase of the total resistance over time. Larger

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3 differences in the behaviour of the different organic fractions were observed. UF membrane
4 preferentially retained the heaviest fraction of biopolymers (BP), while the intermediate fraction
5 of humic substances (HS) was removed at a lower percentage and the lighter fractions seemed
6 to entirely pass through the UF membrane. The successive application of BW and CIPs resulted
7 in the detachment from the membrane of a significant percentage of the retained BP, whereas
8 only a modest percentage of the retained HS.
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13 **Keywords:** backwashing; fouling reversibility; organic fractions; surface water treatment;
14 ultrafiltration
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20 21 1. INTRODUCTION 22 23

24 Application of pressure-driven membrane processes as microfiltration (MF) and
25 ultrafiltration (UF) has expanded in recent years as an alternative technology for wastewater
26 treatment and drinking water production. This expansion is due to the fact that UF has proved to
27 be an effective physical barrier to particles, colloids, bacteria and certain viruses that are larger
28 than the UF membrane pores and, hence, are retained by size-exclusion mechanisms, among
29 others. Furthermore, UF provides extra advantages over conventional treatments such as small
30 footprint, low energy consumption, limited chemical dosing, capability of coping with wide
31 fluctuations in feed quality and delivering permeate of relatively constant quality, and reduced
32 scale-up risks [1-5].
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38 The retained particles accumulated on the feed side of the membrane (and within the
39 membrane pores), however, give rise to the major drawback of UF systems: fouling formation.
40 Fouling leads to additional hydraulic resistance to permeate flow, increase of the energy
41 consumption of the process, lowering of the productivity, worsening of the product quality and
42 eventually premature replacement of membranes [2,4,6-8].
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46 In order to remove fouling, UF membranes are periodically subjected to physical
47 cleaning such as backwashing (BW). BW is performed by reversing the direction of flow
48 through the membrane to dislodge and remove foulants from it and restore the initial
49 permeability [6,8-10]. Fouling removed by a hydraulic cleaning such as BW is referred to as
50 “physically reversible fouling”, in opposition to the “irreversible fouling” made of substances
51 strongly adhered on or within the membrane and not flushed out by any physical cleaning
52 procedure. It is this irreversible fouling that leads to a long-term increase of the resistance (with
53 the subsequent increase of the operational costs) and to a progressive deterioration of the
54 membrane.
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The operation of a UF membrane consists, then, of a succession of cycles each comprising a filtration step (in which membrane resistance gradually increases due to fouling) and a BW step (in which membrane resistance is lowered as foulants are removed from the membrane). Figure 1 schematically represents the evolution of the total membrane resistance during its operation, showing all its components, i.e. resistance of clean membrane (R_m), resistance due to the reversible fouling (R_{rev}) and resistance due to the irreversible fouling (R_{irrev}).

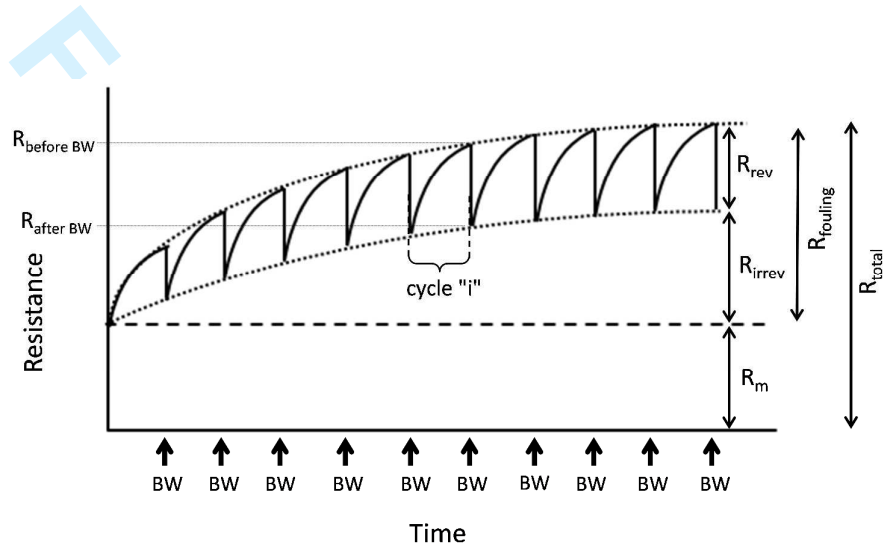


Figure 1: Qualitative representation of the evolution of membrane resistance over a succession of filtration and backwashing (BW) cycles.

The removal of the irreversible fouling can be partially achieved only through aggressive chemical cleaning (“cleaning in place” (CIP)), which is usually burdensome and requires the shutdown of the unit being cleaned for several hours. Chemical cleaning causes both a reduction of the overall production plant capacity and a deterioration of the membranes, wherefore it should be minimised wherever possible [3,11]. A strategy that helps minimise chemical cleaning is the addition of chemical cleaners into the water used for BW, giving rise to the so-called “chemically-enhanced backwash” (CEB) [12]. This cleaning operation does not require an extended shutdown, since it is conducted on line, and the chemicals’ concentration and/or their nature tend to be less aggressive compared to the CIP ones. As a consequence, typically CEBs are less effective than CIPs.

The extent and reversibility of membrane fouling is largely dependent on multiple variables such as membrane characteristics, feed water properties, filtration conditions, module configuration, BW regime... making the control of membrane fouling a complex phenomenon. Among these, the effect of feed water composition and membrane operating conditions have been most researched [8,13-15], while less studies have dealt with the effect that BW variables

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3 (duration, frequency...) or different organic components of the dissolved organic matter (DOM)
4 exert on UF membrane fouling.
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6 Comparison of results from studies on BW variables is, moreover, not entirely reliable
7 and needs to be treated with caution for several reasons. First, these studies treat different types
8 of water: wastewater [2,5,6,11,12,16], seawater [1,17,18] and surface water [3,10], each with
9 different fouling behaviour potential under a given BW regime. For instance, it has been
10 reported that a high salt concentration in the seawater might influence the interaction forces
11 between membrane and foulants [1]. Second, the configuration of UF modules in these studies
12 is very variable: flat-sheet [10], spiral wound [17], pressurized (inside-out) [5,6,12,18] and
13 submerged (outside-in) [1,10] hollow fibre membrane systems, also affecting the propensity to
14 fouling [1,16,19]. Furthermore, most of them are focused on the evolution of the membrane
15 resistance and fouling rates [2,3,5,6,16] and only a few quantify the reversible and irreversible
16 fouling after each backwash cycle [1]. Within this context, more research is still needed on
17 quantitatively determining the effect of BW-related variables on the reversibility of fouling on
18 UF membranes for all scenarios and, in particular, for the outside-in hollow fibre UF for surface
19 water.
20

21 Fouling by DOM components or fractions is also gaining increasing attention of
22 researchers. Indeed, it is acknowledged that different constituents of DOM do not necessarily
23 foul UF membranes on the same way or degree [20-22]. Characterizing such DOM fractions is
24 thus essential for a better understanding of which constituents contribute most in the fouling of a
25 UF membrane. Among the methods developed to characterize DOM, high performance size
26 exclusion chromatography (HPSEC) (whereby DOC fractions are separated according to their
27 hydrodynamic size) has gained much attention as a powerful method for quantitative and
28 qualitative characterization of DOC [23].
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30 The objective of this study was 1) to explore systematically the effect of distinct BW-
31 related variables on the reversibility of UF membrane fouling and on the permeate quality over
32 successive filtration/BW cycles in the treatment of surface water; and 2) to identify which
33 organic fractions were best removed after backwashing (BW) and cleaning-in-place (CIP)
34 episodes. For this purpose, a bench-scale outside-in hollow fibre module operated under dead-
35 end filtration mode at constant transmembrane pressure (TMP) was employed. The variables of
36 study comprised BW TMP, duration and frequency as well as composition of CEBs. Permeate
37 quality was monitored in terms of total organic carbon (TOC), turbidity and UV absorbance
38 (UV_{254}). For the second objective, DOC fractionation was performed by HPSEC.
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2. METHODS

2.1. Feed water characteristics

The feed water to be filtered by the UF module was decanted water from the settling basin of the drinking water treatment plant in Sant Joan Despí (Barcelona, Spain). The average composition of this water during the course of the study is given in Table 1.

Table 1: Average feed water quality

Parameter	Concentration
pH	7.6
Conductivity ($\mu\text{S}/\text{cm}$)	1490
Turbidity (NTU)	1.76
Abs ₂₅₄ (abs/m)	0.080
TOC (mg/L)	4.1
Al ($\mu\text{g}/\text{L}$)	364
Fe ($\mu\text{g}/\text{L}$)	23
P ($\mu\text{g}/\text{L}$)	43

2.2. UF device and membrane characteristics

All experiments conducted in this study were carried out employing a bench-scale outside-in hollow fibre module (Polymem UF0808M3) operated under dead-end filtration mode at constant TMP. The main characteristics of the UF module provided by the manufacturer are shown in Table 2.

Table 2: Characteristics of the UF membrane module provided by the manufacturer.

Manufacturer	Polymem
Membrane type	UF0808M3
Filtration mode	Out-in
Membrane material	Polysulfone
Potting material	Polyurethane
Vessel material	U-PVC
Fibre diameter (mm)	1.4
Surface area (m ²)	0.01
Module external diameter (mm)	20
Module length (mm)	200
Nominal MWCO (Da)	300000
Nominal Pore size (µm)	0.08
Maximum feeding pressure (bar)	3.0
Maximum TMP (bar)	1.5
Maximum TMP during backwash (bar)	2.0
Maximum temperature (°C)	35
pH range	2-12

The decanted feed water was directed to the UF module by means of a centrifugal pump (IML S.A.U., Model MS100M). Valves in the system were opened and closed such that the direction of flow was out-in during the filtration step and reversed to in-out during the BW step. During filtration the feed solution passed through the UF membrane and permeate was collected for flux measurements and chemical analysis. TMP was measured by a pressure gauge (Keller Group, model Leo 3). During BW, UF permeate was pumped using a peristaltic pump (Heidolph, model Pump drive PD5001) and the backwash stream discharged into a separate tank. A schematic diagram of the experimental set-up is shown in Figure 2.

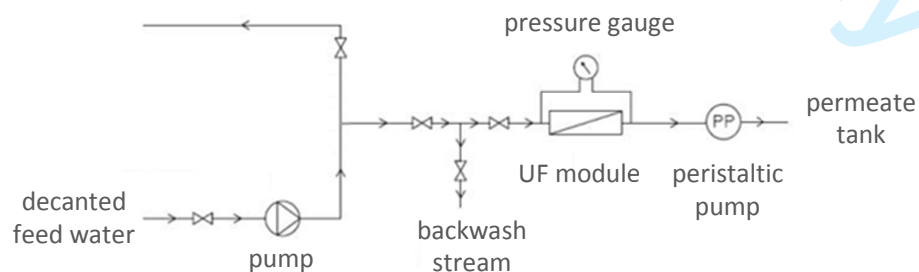


Figure 2: Schematic diagram of the experimental UF system setup.

2.3. Filtration procedure

Prior to each fouling experiment, the cleaned membrane resistance (R_m) was measured through a filtration test with deionised water. These tests were conducted in dead-end mode at a TMP of 1.2 bar without recirculation of permeate for 15 min and then with recirculation until constant permeate production. At this point, R_m was calculated according to the well-established Darcy's equation described in Section 2.6.

Each filtration experiment was carried out at a filtration TMP of 1.2 bar and one combination of BW-related variables, i.e.: backwash TMP (BW_{TMP}), backwash duration (BW_d), backwash frequency (BW_f) and chemically enhanced backwash composition (BW_{CEB-c}). For the carrying out of the experiments with CEBs the dose of NaClO was 7 mg/L, while NaOH and citric acid were added to bring pH to 10-11 and to 3-4, respectively. In order to isolate the effect of these variables on fouling reversibility, each of them was varied (as shown in Table 3) while setting all other variables at fixed values (marked in bold in Table 3).

Table 3: Summary of the experimental conditions and variation of each set of experiments conducted during the study.

Variable of study	Tested values
BW_{TMP} (bar)	0.7, 1.0, 1.5, 1.8
BW_f (min)	10, 20 , 40, 60
BW_d (min)	0.5, 1.0 , 2.0
BW_{CEB-c}	Blank* , NaClO, NaOH, NaOH+NaClO, citric acid

* Blank means BW with UF permeate

The pre-fixed value is marked in bold

Permeate flux and quality was monitored all over each filtration experiment, which lasted 200 min and was generally conducted in duplicate to ensure consistency of results. Flux was measured by the timed collection of permeate in a volumetric flask. Because feed water for all experiments was taken from the same location in the treatment train of the DWTP in Sant Joan Despí, the differences observed in fouling reversibility could be attributed to the investigated BW-related variables.

2.4. Removal and reversibility of organic fractions

Further efforts were devoted to investigate which organic fractions were most removed by the UF membrane and which were most detached from it after the successive application of BW (assisted with intermittent CEB), a CIP based on alkaline and oxidant agents (CIP-B) and finally a CIP based on an acid agent (CIP-A). This allowed quantify the reversible fouling after each backwashing and cleaning step and eventually the irreversible fouling on the UF membrane.

For this purpose, a filtration experiment was conducted similarly to those described above at a constant TMP of 1.2 bar and a BW regime optimised from the previous set of experiments, i.e. BW was performed every 20 min of filtration at a TMP of 1.8 bar and with a duration of 1.0 min. Additional CEBs based on a combination of NaClO (7 mg/L) and NaOH (pH 10-11) were applied every 3 BW. A total volume of 3.945 L of feed water was filtered, of which 0.337 L was used for BW. On completion of the filtration experiment, the UF membrane was consecutively subjected first to the CIP-B with the addition of NaOH (pH between 11 and 12) in combination with 200 mg/L NaClO (volume 50 mL, contact time 90 min) and, second, to the CIP-A with the addition of citric acid (pH between 3 and 4, volume 50 mL, contact time 30 min). The reagents used for CIP-B and CIP-A were selected in accordance with the ones used in the drinking water treatment plant of Sant Joan Despí.

Feed and permeate over the experiment were collected for analysis of DOC and its fractions (see below analytical techniques) by HPSEC. Organic fractionation was also performed for the successive BW streams (collected as a composite sample) and CIP-B and CIP-A solutions.

2.5. Chemical analysis of water samples

Feed water and UF permeate quality for the first set of experiments was analysed in terms of turbidity, TOC and UV₂₅₄. The samples were collected in sterile vials and stored in cold conditions until analysis in the laboratory. Turbidity was analysed by nephelometry (Hach 2100 AN IS Turbidimeter), Absorbance was analysed by spectrophotometry (Hach DR 5000) and TOC by oxidative combustion and infrared-detection (Shimadzu V CPH).

Fractionation of dissolved organic carbon (DOC) was performed by HPSEC using a Toyopearl TSK HW-50S column (250x20 mm) coupled to on-line UV254, organic carbon (OC) and organic nitrogen (ON) detectors by DOC-Labor (Karlsruhe). The principles of the technique are reported in depth by Huber et al. [23]. Briefly, it is based on size exclusion liquid chromatography whereby organic compounds are fractionated into five sub-fractions according to their molecular weight (MW): (1) biopolymers (BP, with MW>20000 g/mol, basically

constituted by polysaccharides and proteins), (2) humic substances (HS, with MW of approx. 1000 g/mol, constituted by fulvic and humic acids), (3) building blocks (BB, with MW between 300 and 500 g/mol, constituted by breakdown products of humics), (4) low molecular weight acids (LMWA, with MW<350 g/mol, constituted by alcohols, aldehydes, ketones, sugars and amino acids) and (5) low molecular neutrals (LMWN, with MW<350 g/mol, constituted by alcohols, aldehydes, ketones and amino acids). The organic carbon retained in the chromatographic column (i.e. non-chromatographic DOC) is termed hydrophobic fraction. Based on the differences in UV-active components or nitrogen content, HPSEC can also determine the content of proteins within the BP fraction.

2.6. Data treatment for the membrane hydraulic performance evaluation

Fouling was determined by the increase of resistance posed by the fouled membrane, which was in turn calculated from the decline of permeate flux according to the well-established Darcy's equation:

$$J = \frac{\Delta P}{\mu \cdot R_{\text{total}}} \quad (\text{eq. 1})$$

where J is the permeate flux ($\text{m}^3 \cdot \text{m}^{-2} \cdot \text{s}^{-1}$), ΔP is the TMP (Pa), μ is the permeate viscosity (Pa·s) (corrected to 20°C) and R_{total} is the total resistance of the fouled membrane (m^{-1}). According to Darcy's law, a decrease in J under constant ΔP during membrane filtration process (or equivalently an increase in ΔP under constant J) is indicative of membrane fouling. The total resistance can be described by the resistance-in-series model and expressed as [4,10,13]:

$$J = \frac{\Delta P}{\mu \cdot (R_m + R_{\text{rev}} + R_{\text{irrev}})} \quad (\text{eq. 2})$$

where R_m is the cleaned membrane resistance (measured before each experiment with deionised water) and R_{rev} and R_{irrev} are the hydraulically reversible and irreversible fouling resistances, respectively.

For each filtration cycle "i" R_{rev}^i was calculated as the difference of resistance measured before and after backwashing (as shown in Figure 2).

$$R_{\text{rev}}^i = R_{\text{before BW}}^i - R_{\text{after BW}}^i$$

The contribution of R_{rev}^i over the total fouling of the membrane (R_{fouling}) can then be calculated as follows (see Figure 1):

$$\text{Reversible fouling (\%)} = \frac{R_{\text{rev}}^i}{R_{\text{fouling}}^i} = \frac{R_{\text{before BW}}^i - R_{\text{after BW}}^i}{R_{\text{before BW}}^i - R_m} \quad (\text{eq. 3})$$

In this study, averaged reversible fouling percentages over all filtration cycles and duplicates under the same experimental conditions are reported for comparison between different BW regimes.

It must be remarked here that most published studies report experimental data on a dimensionless basis (e.g. normalised flux, pressure, permeability or resistance). While this facilitates indeed comparison of experiments carried out under different experimental conditions, it also masks the possible effects of e.g. flux and pressure values on fouling. For this reason, measured fouling-related variables were not normalised and reported as measured.

3. RESULTS

Plotted in the next figures are a) the total resistance curves obtained for each set of BW conditions, b) the degree of membrane fouling reversibility calculated from the resistance profile and according to eq. 2, and c) the quality of permeate in terms of turbidity, UV₂₅₄ and TOC. In all cases, resistance profile showed a pattern as described in Figure 1, i.e. an increase in resistance during the filtration step and a decrease during BW. The resistance was however not completely restored to the initial value, indicating that, regardless the BW regime, irreversible foulants slowly accumulated onto and into the membrane.

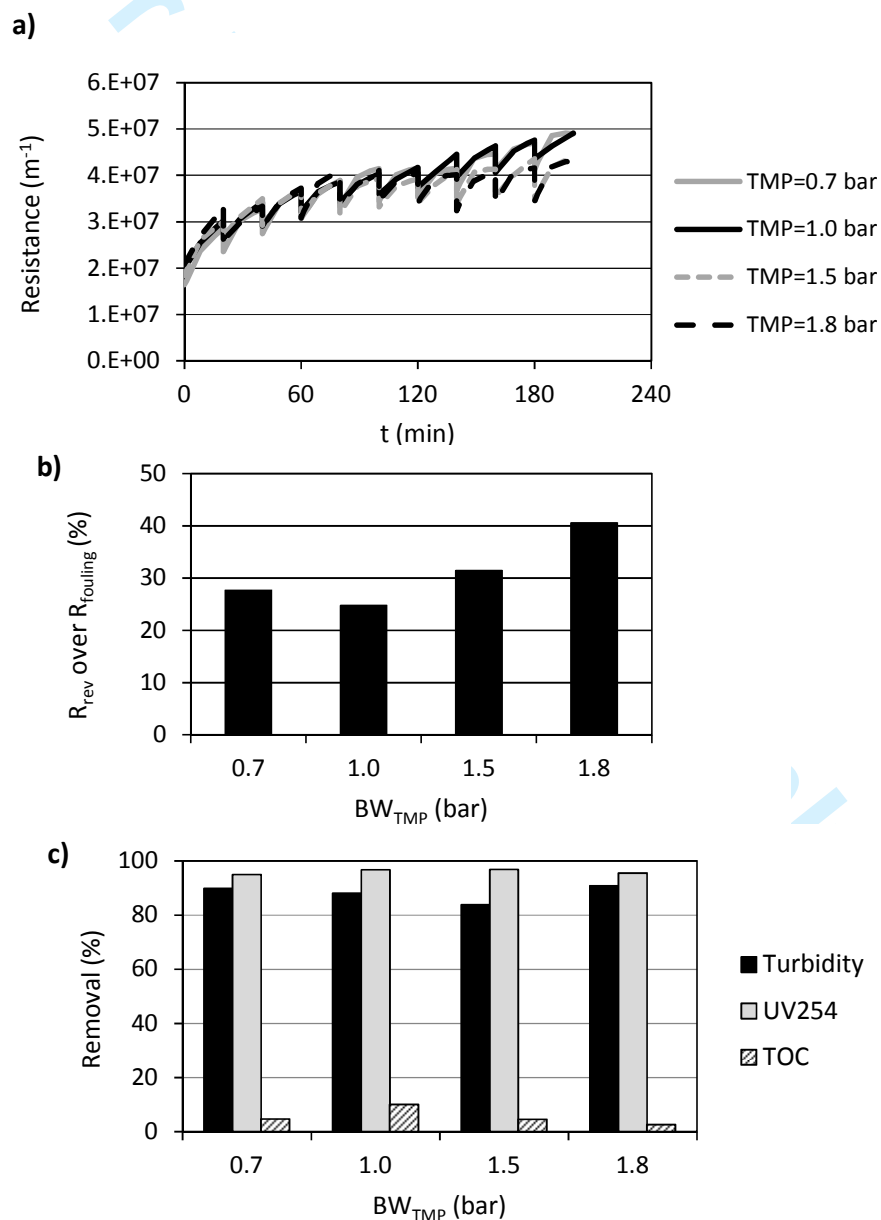
3.1. Effect of backwashing transmembrane pressure (BW_{TMP})

As shown in Figure 3, higher BW_{TMP} provided a lower resistance increase (i.e. a better permeability restoration) over the experiment (Figure 3a) and a higher degree of fouling reversibility (Figure 3b). R_{rev} percentage was below 30% at BW_{TMP} of 0.7 and 1.0 bar, but it increased to 31% at BW_{TMP} of 1.5 bar and up to 41% at BW_{TMP} of 1.8 bar. This trend is likely due to the fact that shearing stress can more efficiently wash out tightly bound foulants from the membrane that would not be removed by lower BW_{TMP}.

This finding is partially in accordance with that reported by Remize et al. [10], who observed that increasing BW_{TMP} from 1.2-2.0 bar in the filtration of surface water with UF membranes resulted in an increase of the foulants removed from the membrane (from 25% to 44%). Interestingly, and in opposition to our study, this trend did not translate into an increase of permeability recovery with BW_{TMP}, highlighting that measurement of permeability (or

resistance) recovery may not be sufficient to identify fouling removal and that measurement of matter removed may be also necessary.

With regards to the permeate quality, removal of turbidity, UV_{254} and TOC were comparable within the experimental error whatever the BW_{TMP} applied (Figure 3c). Turbidity and UV_{254} were decreased at high degrees (average removals of 88% and 96%, respectively), while TOC was only decreased by <10% for all BW_{TMP} values. The low retention of TOC by the UF membrane may be explained by the predominance of small molecular weight (MW) organic fractions with $MW \leq 1000$ Da (much smaller than the nominal MWCO of the UF membrane of 300000 Da, see Table 2) present in the decanted water [24].



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4 Figure 3: Effect of backwash transmembrane pressure (BW_{TMP}) on the a) total resistance of the
5 fouled membrane, b) percentage of reversible fouling over the total membrane fouling and c)
6 removal of turbidity, UV_{254} and TOC by the UF membrane (confidence intervals $\leq 4.5\%$ at a
7 confidence level of 95% for all cases).
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10 11 12 **3.2. Effect of BW frequency (BW_f)**

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15 The effect of BW_f on the total resistance, fouling reversibility and permeate quality
16 during the process of membrane filtration is shown in Figure 4. It is noticeable in Figure 4a that
17 the initial resistance for $BW_f=10$ min was slightly higher than that corresponding for all other
18 BW_f , suggesting that permeability membrane before that experiment had not been completely
19 restored. Even so, backwashing every 10 min resulted in a lower fouling rate, in contrast to
20 backwashing at stretched frequencies (20, 40 and 60 min), which led to a more severe increase
21 in fouling resistance (i.e. accumulation of irreversible fouling) during membrane operation. As
22 shown in Figure 4b, the more frequent the BW the higher the reversibility of fouling: fouling
23 reversibility decreased from 50% for BW_f of 10 min to 41% for BW_f of 20 min and below 37%
24 for both BW_f of 40 and 60 min.
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29 Similar trends on lowered fouling accumulation with more frequent BW have been
30 reported by other researchers, although the degree of dependence differ considerably if other
31 types of feed water or UF configurations are used as it is commonly the case [2,5,12]. There is,
32 however, consensus that stretched BW frequencies allow more material to be accumulated on
33 the membrane surface during a filtration cycle, forming a fouling layer more tightly attached
34 and compacted and exhibiting thus a lower degree of reversibility under a given BW [1,6,16].
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39 Concerning the permeate quality, no significant differences were observed neither under
40 the different BW_f tested nor compared with the previous set of experiments under different
41 BW_{TMP} . Turbidity and UV_{254} removals were 88% and 95%, respectively, whereas TOC removal
42 was only 5%.
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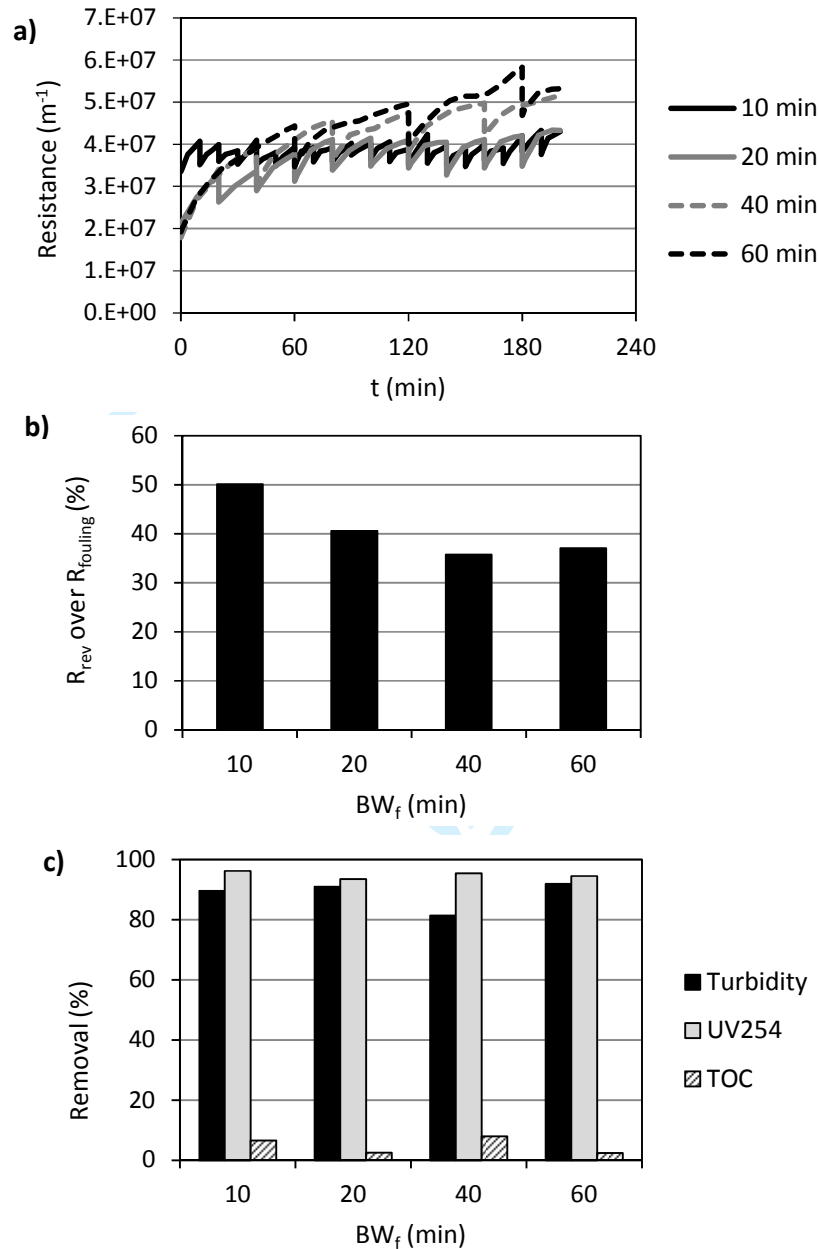


Figure 4: Effect of backwash frequency (BW_f) on the a) total resistance of the fouled membrane, b) percentage of reversible fouling over the total membrane fouling and c) removal of turbidity, absorbance and TOC by the UF membrane (confidence intervals $\leq 7.6\%$ at a confidence level of 95% for all cases).

3.3. Effect of BW_d duration (BW_d)

The effect of BW_d on the total resistance, fouling reversibility and permeate quality during the process of membrane filtration is shown in Figure 5. As in the previous set of experiments, an experiment showed an initial membrane resistance slightly higher than the corresponding to the other experiments, suggesting again that the membrane was not completely cleaned prior to the filtration experiment. Despite the different starting point, the evolution of resistance over all experiments is comparable.

Increasing BW_d from 0.5 min to 1 min increased the fouling reversibility from 32% to 41%, indicating that foulants are more easily washed out away from the membrane pores during a longer backwash (Figure 5b). In fact, better fouling removal from UF membrane at increased BW_d has been reported in the scientific literature for very variable filtration scenarios in terms of feed water characteristics and UF configurations [1,2,5,6,16-18], including other types of membrane systems such as microfiltration [9], ceramic membranes [25] and membrane bioreactors [26].

In our study, lengthening the BW_d to 2 min was not accompanied by any increase of fouling reversibility. The existence of a threshold in BW_d beyond which no further improvement is observed has also been observed by other researchers [1,23]. Ye et al. [1] attributed this trend to the fact that “excess backwash volume might also foul the membrane or the remaining fouling cake due to the impurities in the backwash flux”.

Similarly to the previous set of experiments, turbidity and UV₂₅₄ removals were removed at high extents (83% and 94%, respectively), whereas TOC removal was only 2% (with the exception at BW_d of 0.5 when a TOC removal of 22% was attained).

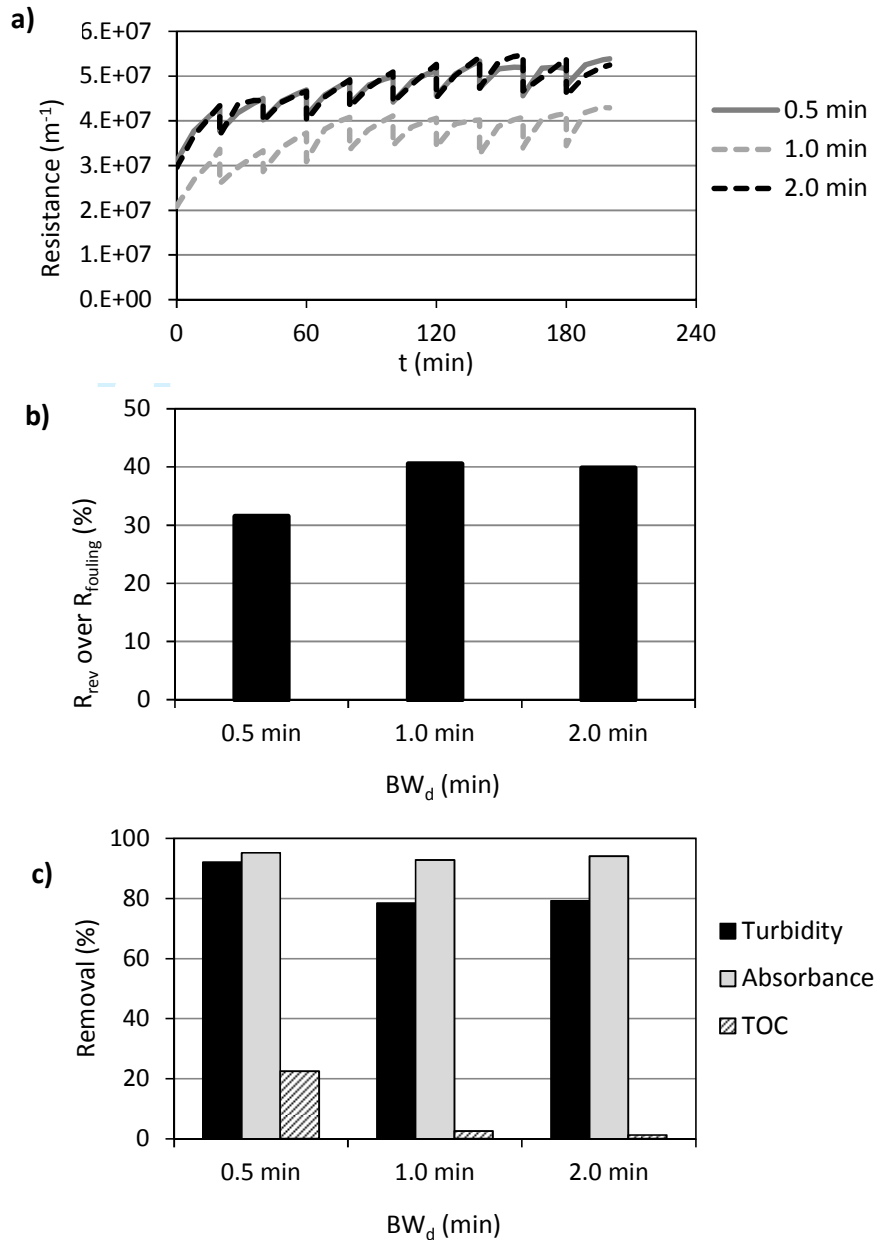


Figure 5: Effect of backwash duration (BW_d) on the a) total resistance of the fouled membrane, b) percentage of reversible fouling over the total membrane fouling and c) removal of turbidity, UV_{254} and TOC by the UF membrane (confidence intervals $\leq 4.5\%$ at a confidence level of 95% for all cases).

3.4. Effect of the chemically enhanced BW composition (BW_{CEB-c})

The fouling rate and reversibility degree differed depending on the chemical cleaners used for the CEB (Figure 6). NaClO performed the best, exhibiting the lowest fouling rate and the maximum fouling reversibility degree (approx. 38%), closely followed by the combination of NaOH+NaClO (approx. 34%). Acidic and alkaline cleaning solutions are commonly employed to remove inorganic and organic foulants, respectively, but the use of citric acid and NaOH in this study contributed little to the reversibility of fouling (approx. 28-27%) compared to the blank (26%) (Figure 6b).

These results compare well with those reported by Arnal et al. [7] and Zheng et al. [12], who found that NaClO as a cleaner added to the BW water performed the best at restoring the permeability of a UF membrane fouled after treatment of surface water [7] and wastewater [12], while NaOH had less influence compared to water. Similar results were observed by Espinasse et al. [29] after treating coupons of nanofiltration membrane with various cleaning agents. The benefits of using NaClO are explained by the fact that NaClO can oxidise the organic foulants accumulated on the membrane, generating more oxygen-containing functional groups (such as ketone, aldehyde and carboxylic acids), which due to their increased hydrophilicity are less attached to the membrane [12,29,30]. To exemplify the disinfection power of some chlorine-based compounds, Laine et al. [31] reported that ceasing the dosage of chlorine in backwash water after 20 days of operation resulted in severe fouling of the membranes within 5 days. Alkaline agents have also been reported to be effective at detaching foulants (particularly organic ones) since at high pH many organic compounds are hydrolysed presenting, under their dissociated form, increased solubility and propensity to be detached from the membrane [29].

Beyond the use of oxidant and alkaline agents separately, their combination has also been reported to be more effective at removing foulants from the membrane [29,30,32]. However, the combination of NaClO and NaOH in this study did not perform better than NaClO alone (Figure 6). The low performance of citric acid, which is effective for the removal of inorganic foulants via dissolution of salts and complexation of certain metals, is indicative that the fouling layer formed on the membrane was made up of organic materials rather than inorganic salts.

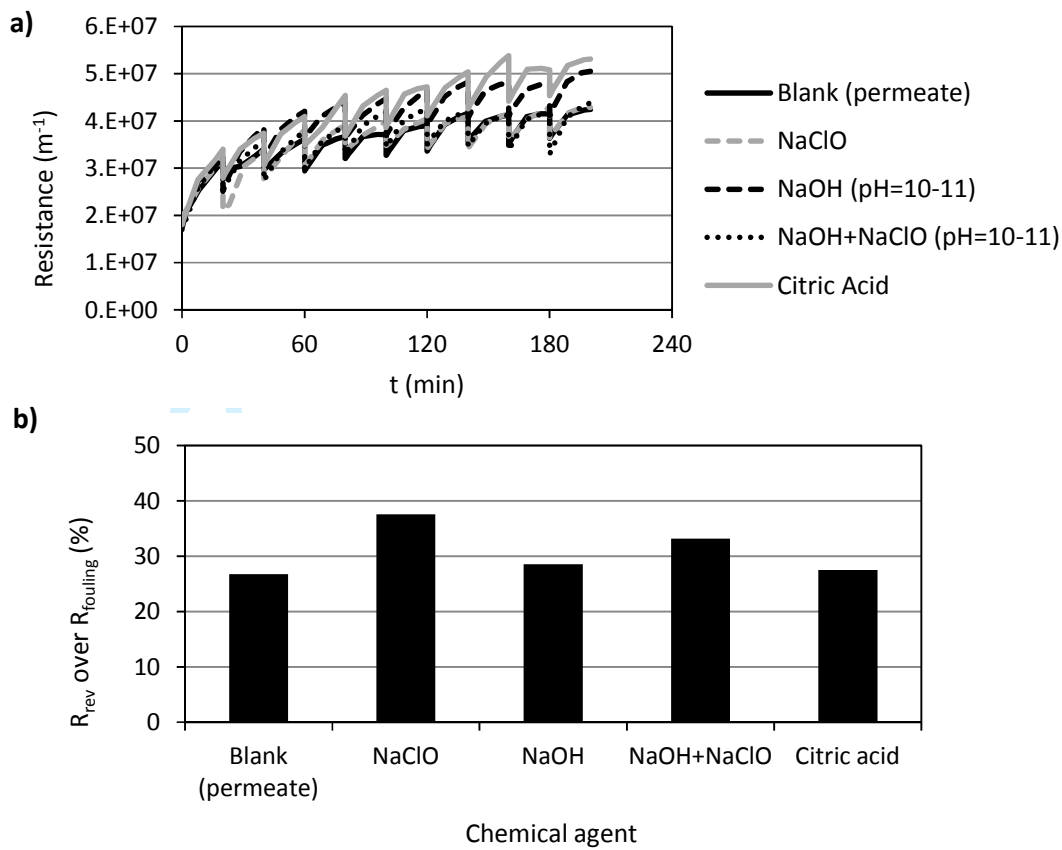


Figure 6: Effect of the CEB composition (BW_{CEB-c}) on the a) total resistance of the fouled membrane, b) percentage of reversible fouling over the total membrane fouling by the UF membrane (confidence intervals $\leq 4.0\%$ at a confidence level of 95% for all cases).

3.5. Organic fouling composition on the UF membrane

Figure 7 compares the concentration of TOC, DOC and each of the organic fraction in feed and permeate (with removal percentages in brackets) as analysed by HPSEC.

Feed water showed TOC and DOC values of 4.0 and 3.3 mg/L, respectively. This difference (of 15%) indicated that even this water had passed through a sand filtration stage a fraction of the organic load was still in the form of particular or colloidal organic carbon. With regards to the organic fractions, HS was always the most predominant one, accounting for 56% of the total DOC, followed by the lighter BB and LMWN (20%) fractions, while the heavier BP fraction averaged only 4%. The LMWA fraction was always found below limit detection. This composition is in accordance with previous studies that also applied HPSEC for the fractionation of NOM after sand filtration of river water [33,34].

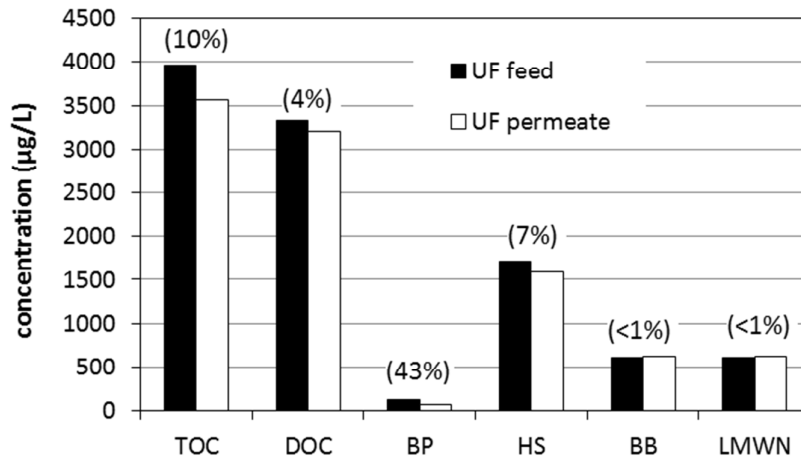


Figure 7: Concentration of TOC, DOC and its fractions BP, HS, BB and LMWN in both feed and permeate streams (in brackets removal percentages by UF the membrane)

The removal of TOC by the UF membrane was 10%, while the corresponding one for DOC was fairly low (4%). Larger differences in the removals were observed for the different organic fractions. UF membrane preferentially removed the heaviest (and biggest) fraction BP (removal percentage of 43%), while intermediate HS was removed at a percentage of 7% and lighter (and smaller) BB and LMWN seemed to entirely pass through the UF membrane. This finding is in agreement with Peter-Varbanets et al. [35], who also observed that “only biopolymers and the humic acid fraction of NOM were retained to some extent during filtration” of different types of water. The fact that BP was in percentage the most retained material was expected based on size exclusion effects [22,36,37] and was in accordance with other studies that have observed a flux decline through UF membrane when the concentration of BP was increased within a range of concentrations similar to that in the present study [35,38,39].

Based on a comparison between the composition of feed and permeate streams, fouling on UF membranes was anticipated to be made up of 67% HS (amounting 469 µg) and 33% BP (amounting 229 µg) (percentages referred to the total DOC removed by UF). It is of note that 20% of the 229 µg of BP retained on the UF membrane consisted of protein-like compounds, indicating preferential removal of polysaccharides over proteinaceous substances. This is in agreement with other researchers that have applied HPSEC in the ultrafiltration of water [22].

3.6. Fouling detachment after each backwashing and cleaning

UF membranes are periodically backwashed with ultrafiltered water to remove deposited matter from the membrane and restore its original permeability as much as possible.

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3 Detachment of organic matter from the UF membrane was evident as the backwash stream was
4 richer in absolute TOC concentration (4.7 mg/L) than ultrafiltered water used for the backwash
5 (3.6 mg/L). The composition of such backwash stream was 9% BP, 51% HS, 20% BB and 20%
6 LMWN. In comparison with the ultrafiltered permeate, it was found to be enriched in BP (by
7 5%) and impoverished in HS (by 5%), while the concentrations of BB and LMWN were
8 essentially the same.
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11 Figure 9 compares the initial organic mass (in μg) fouling the UF membrane with the
12 mass remaining after applying BW (+CEBs), CIP-B and CIP-A calculated through a mass-
13 balance from the concentration of each organic fraction within each volume stream.
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15
16 It can be seen that BW(+CEBs) was able to detach 33% of the initial BP but only 9% of
17 the initial HS. The enrichment in BP suggested that components within this fraction, in
18 particular polysaccharides rather than proteins, were not rigidly attached to the membrane but
19 amenable to be washed out. This behaviour is likely due essentially to their size relative to that
20 of the membrane pores: organic substances within the BP fraction much larger than the
21 membrane pores lead to cake formation, which is more readily detached, while lighter fractions
22 such as HS can cause pore blocking, build-up a denser cake layer less readily washed out or be
23 adsorbed onto the membrane material [8,32]. The remaining BP and HS on the membrane
24 would explain the irreversible fouling (never completely avoided whatever the BW regime
25 applied) that resulted in the gradual increase of total resistance over time (Figures 3-6). Which is
26 of these fractions has a larger impact on the membrane resistance is not clear.
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29 HS was found in this study to be the most retained fraction in terms of amount (but not
30 of percentage) (Figure 8). HS is considered by some studies of minor relevance in terms of
31 fouling due to their high transmission through the mesoporous UF membrane [22,38,39],
32 whereas it is considered a detrimental foulant causing severe hydraulically irreversible fouling
33 by some others [40].
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35
36 Opposite to HS, BP was found to be the most retained fraction in terms of percentage
37 (but not of amount) (Figure 8), in agreement with other previous studies [20,22]. Its impact on
38 fouling depends however upon its components: polysaccharides are believed to cause only
39 hydraulically reversible fouling, while protein-like substances are thought to induce
40 hydraulically irreversible fouling [22,39]. The major impact of proteins on fouling may be
41 caused to the fact that they are more compact and can be retained at or inside the pores, thus
42 resulting in the constriction and/or blockage of the membrane pores [39]. The finding in this
43 study that 20% of the BP retained by the UF membrane was made of protein-like substances
44 may explain why BP was not completely detached after the application of BW(+CEB) (Figure
45 9).
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48 In this study, because a portion of both BP and HS fractions were attached on the UF
49 membrane, it could not be elucidated whether fouling was mainly caused by one or another (or
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both) fraction. Clearly, more investigations are required to identify if HS or BP contributes most to hydraulically (ir)reversible membrane fouling during UF of different waters.

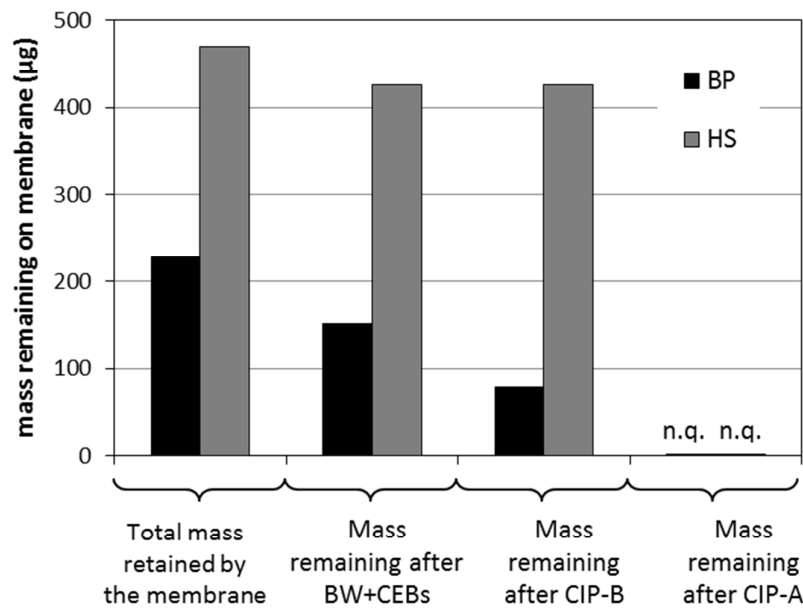


Figure 9: Evolution of mass of BP and HS remaining on the UF membrane (μg) after the successive application of BW (+CEBs), CIP-B and CIP-A.

Soaking the membrane with the CIP-B solution resulted in a detachment of a further 32% of the initial BP retained by the membrane, but on contrary no HS was detached at all, corroborating that this fraction was rigidly tight to the membrane and not easily detached by NaOH either NaClO under the experimental conditions of this study. As mentioned above in this study, the detachment of BP may be explained by the fact that the constituents of the BP fraction (polysaccharides, proteins) are hydrolysed at high pH (even the weakest phenolic groups dissociate at such a high pH) and oxidised, increasing their solubility and therefore being more prone to be detached from the membrane [29]. Finally, the performance of an acid solution (CIP-A) could not be quantified because the organic fractions detached, if any, might be in the HPSEC chromatograms overwhelmed by the very high concentration of citric acid employed as cleaning agent. However, and also according with what was discussed in previous sections, organic fouling detachment is expected to be of minor importance since acid cleanings are applied commonly to eliminate inorganic foulants from the membrane (e.g. Fe and Mn) [32]. This is in qualitative agreement with Strugholtz et al., who found that NaOH and in particular NaClO were effective at removing both BP and HS while HCl was not [32]. The fact that organic fractions were analysed in their study only in the cleaning solution did not allow

determine how much BP and HS were remaining on the membrane and, hence, compare results with the ones obtained in this study.

4. CONCLUSIONS

Reversibility of fouling on a bench-scale UF membrane treating decanted water under different BW regimes was assessed in this study. The BW regimes were varied in terms of backwash TMP (BW_{TMP}), backwash duration (BW_d), backwash frequency (BW_f) and chemically enhanced backwash composition (BW_{CEB-c}). Permeate quality was monitored in terms of turbidity, UV_{254} and TOC.

Although differing in their efficiency, all BW regimes proved to contribute to control fouling on UF membranes. Nevertheless, results also showed that irreversible fouling was never completely avoided whatever the BW regime applied, resulting in a gradual increase of total resistance over time. Splitting the $R_{fouling}$ into its components, it was found that R_{irrev} was always higher than R_{rev} .

As expected, the degree of reversibility depended on the BW-related variables. Higher BW_{TMP} provided higher degrees of fouling reversibility, with R_{rev} percentage below 30% at BW_{TMP} of 0.7 and 1.0 bar, but of 31% at BW_{TMP} of 1.5 bar and up to 41% at BW_{TMP} of 1.8 bar. Concerning the BW_f , the more frequent the BW the higher the reversibility of fouling: fouling reversibility decreased from 50% at BW_f of 10 min to 41% at BW_f of 20 min and below 37% at both BW_f of 40 and 60 min. Similarly, increasing BW_d from 0.5 min to 1 min increased the fouling reversibility from 32% to 41%, but lengthening the BW_d to 2 min was not accompanied by any further increase of fouling reversibility. To sum up, the more intensive a BW was (in terms of higher BW_{TMP} , shortened BW_f and prolonged BW_d) the more effective it was in removing foulants from the membrane. This was so because less intensive BW allowed more material to be accumulated on the membrane surface during a filtration cycle, forming a fouling layer more tightly attached and compacted and exhibiting thus a lower degree of reversibility under a given BW.

Concerning the composition of CEB, NaClO performed the best, exhibiting the maximum fouling reversibility percentage (approx. 38%), closely followed by the combination of NaOH+NaClO (approx. 34%), while citric acid and NaOH contributed little (approx. 28-27%) compared to the blank (26%).

With regards to the permeate quality, no significant differences were observed whatever the BW regime applied. Turbidity and UV_{254} removals were always above 80% and 90%, respectively, whereas TOC removal was generally below 10%. The low retention of TOC may

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3 be explained by the predominance of small molecular weight (MW) organic fractions with
4 $MW \leq 1000$ Da (much smaller than the nominal MWCO of the UF membrane of 300000 Da)
5 present in the decanted water as determined in the subsequent experiments.
6

7 While applying more intensive BW (in terms of higher BW_{TMP} , shortened BW_f and
8 prolonged BW_d) results in reduced membrane fouling, it also leads to higher water losses,
9 energy consumption and chemicals requirements if CEB is applied, bringing down the
10 operational efficiency of the UF treatment. A compromise solution must be taken to establish
11 the optimal BW conditions that minimise both membrane fouling and total costs.
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13

14 Besides optimising the BW efficiency, new approaches to minimise membrane fouling
15 have been researched. These include other physical methods to remove foulants from a
16 membrane surface such as flushing, vibrations, air assisted BW, CO_2 back permeation or
17 combination of them [41]. Examples of such approaches are the works carried out by Bessiere et
18 al. [42], who reported that coupling flushing with air assisted BW significantly reduced
19 membrane fouling leading to energy savings of 65% compared to conventional BW when
20 treating surface water with hollow-fibre UF modules; and Chen et al. [11], who found by
21 employing a statistical factorial design that the optimal UF physical cleaning protocol consisted
22 of a combination of BW followed by forward flushing when treating wastewater with a spiral
23 wound UF system. All these studies conclude that minimising membrane fouling is still an open
24 research field and that further efforts are required to develop optimised cleaning procedures.
25
26

27 With regards to the fouling potential and reversibility of the organic fractions as
28 analysed by HPSEC, UF membrane preferentially retained the heaviest (and biggest) fraction
29 BP (removal percentage of 43%), while intermediate HS was retained at a percentage of 7% and
30 lighter (and smaller) BB and LMWN seemed to entirely pass through the UF membrane. The
31 pattern was expected from size exclusion effects [22,36,37]. Based on a mass-balance over the
32 UF membrane, fouling was anticipated to be made up of 67% HS and 33% BP.
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35 The application of BW(+CEBs) resulted in the detachment of 33% of the initial BP but
36 only 9% of the initial HS. This revealed that HS was more rigidly attached to the membrane
37 whereas BP (in particular polysaccharides rather than protein-like substances) was more
38 amenable to be washed out. Which of the fractions (BP or HS) remaining on the membrane
39 contributed most on the irreversible fouling could not be elucidated in this study, but recent
40 studies have concluded that protein-like substances represent a detrimental foulant that induce
41 severe hydraulically irreversible fouling. This agrees with the finding that 20% of the BP
42 fouling the UF membrane in this study consisted of proteinaceous materials.
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45 Soaking the membrane with an alkaline and oxidant solution (CIP-B) resulted in the
46 detachment of a further 32% of the initial BP retained by the membrane, but on contrary no HS
47 was detached at all. Finally, the performance of an acid solution (CIP-A) could not be quantified
48 in this study because the organic fractions detached, if any, might be in the HPSEC
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3 chromatograms overwhelmed by the very high concentration of citric acid employed as cleaning
4 agent. However, organic fouling detachment is expected to be of minor importance since acid
5 cleanings are applied commonly to eliminate inorganic foulants from the membrane (e.g. Fe and
6 Mn).
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10 11 12 **5. ACKNOWLEDGEMENTS**

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