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Ultrafiltration membranes for drinking-water production from low-quality surface water: A case study in Spain

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Abstract. Ultrafiltration membranes have several advantages over conventional drinking-water treatment. However, this technology presents major limitations, such as irreversible fouling and low removal of natural organic matter. Fouling depends heavily on the raw-water quality as well as on the operating conditions of the process, including flux, permeate recovery, pre-treatment, chemical cleaning, and backwashing. Starting with the premise that the optimisation of operating variables can improve membrane performance, different experiments were conducted in a pilot plant located in Granada (Spain). Several combinations of permeate and backwashing flow rates, backwashing frequencies, and aeration flow rates were tested for low-quality water coming from Genil River with the following results: the effluent quality did not depend on the combination of operating conditions chosen; and the membrane was effective for the removal of microorganisms, turbidity and suspended solids but the yields for the removal of dissolved organic carbon were extremely low. In addition, the threshold transmembrane pressure (-0.7 bar) was reached within a few hours and it was difficult to recover due to the low efficiency of the chemical cleanings. Moreover, greater transmembrane pressure due to fouling also increased the energy consumption, and it was not possible to lower it without compromising the permeate recovery. Finally, the intensification of aeration contributed positively to lengthening the operation times but again raised energy consumption. In light of these findings, the feasibility of ultrafiltration as a single treatment is questioned for low-quality influents.

Keywords: economical feasibility; fouling; natural organic matter; transmembrane pressure; ultra-filtration

1. Introduction

Membrane technology was born at the beginning of the 20th century for several applications (Baker 2004), water treatment being one of the most notable. In particular, ultrafiltration membranes are frequently used for drinking-water production from surface or groundwater as they offer a real alternative to conventional treatment: they constitute a physical barrier for the retention of microorganisms (Jacangelo *et al*. 1997), preventing disinfection by-products (DBPs) generated after chlorination.

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Furthermore, in terms of operability, this technology has great advantages such as smaller space requirements and ease of control and maintenance (Kimura *et al*. 2004, Xia *et al*. 2005). Not only are the facilities simpler to operate but they also provide clear water free from biological contamination, which explains the use of mobile ultrafiltration plants for drinking-water production in emergency situations, for example in developing and transition countries (Arnal *et al*. 2007, Peter-Varbanets *et al*. 2009). However, these membranes have two main limitations: low retention of natural organic matter (NOM) and fouling caused mainly by the former.

Laîné *et al*. (2000) contended that UF alone is not sufficient enough to guarantee the required effluent quality for surface waters with high organic contents. The membranes are effective in removing turbidity and pathogens but not for precursors of DBPs or organic micropollutants (Huang *et al*. 2009). In fact, very poor results have been reported when evaluating NOM retention, i.e., between 20 and 50% (Guo *et al*. 2008, Mijatovic *et al*. 2004, Rojas *et al*. 2008). On the other hand, low effectiveness in NOM removal has a direct impact on organoleptic properties such as colour, smell, and taste (Aoustin *et al*. 2001).

Fouling can be categorised according to the type of foulant: inorganic, particulate, organic and biological fouling, (Li 2008) and more frequently as concentration polarization and irreversible fouling, which are two related but different processes (Cui and Taha 2003). Concentration polarization is largely reversible and is characterised by the accumulation of the rejected macromolecules near the membrane surface, equivalent to the build-up of the mass-transfer boundary layer. Membrane fouling, on the other hand, is largely irreversible. It is caused by the adsorption of the macromolecules on the membrane surface as well as within the pores. In any case, the main consequence of both is the rise of the transmembrane pressure (TMP) or the reduction of the flux, depending on whether the process is TMP or flux constant. The result is greater energy consumption and a shorter membrane lifespan, and hence some strategies need to be followed in order to minimise this problem.

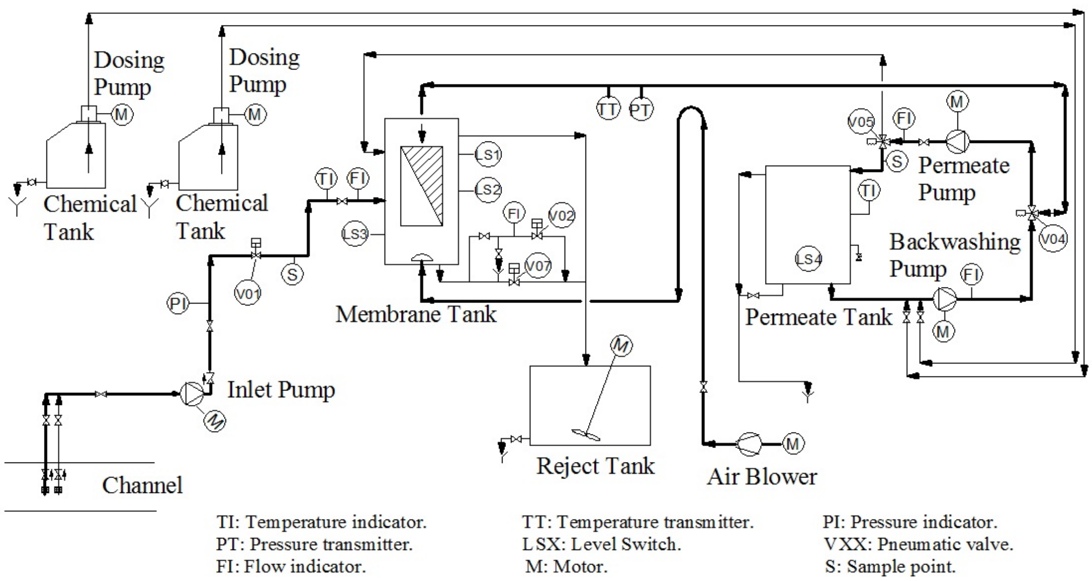

Fig. 1 Pilot plant flow diagram

It is commonly accepted that irreversible membrane fouling depends heavily on the raw-water quality, with the presence of NOM potentially adsorbable on the membrane surface being especially important; however, other operating parameters can be adjusted in order to limit this phenomenon (Crozes *et al*. 1997). Operating conditions influencing NOM fouling of membranes include flux, permeate recovery, pre-treatment, chemical cleaning and in the case of low-pressure membranes, backwashing (Amy 2008). Consequently, the optimisation of operating variables can improve ultrafiltration membrane performance. Aeration flow rates should be included among the variables to optimise, as some studies have demonstrated its efficiency to reduce fouling in hollow fibres or tubular membranes (Cui and Taha 2003, Ji and Zhou 2006) and even flat-sheet membranes (Cheng and Lee 2008). Nevertheless, very few data have been reported for spiral-wound membranes, i.e., only some preliminary works where the flux enhancement by air sparging ranged up to 25% (Cui *et al*. 2003).

Considering all the factors mentioned above, a study was developed for a pilot plant to treat river water in Granada (Spain). Given that the quality of the raw water is the only out-of-control variable, the present study seeks to evaluate spiral-wound ultrafiltration membrane performance for a low-quality influent. The specific objective of this paper is to determine whether drinking-water production by ultrafiltration is feasible when treating low-quality river water, taking into account the effluent quality, fouling generation, and operability of the plants for different operating conditions together with the effect of the latter on the energy consumption.

2. Materials and methods

2.1 Description of the pilot-scale plant

The facility was composed basically of an inlet pump, membrane tank, permeate pump, backwashing pump, permeate tank and air blower. The membrane used was polyvinyliden fluoride (PVDF) SpiraSep 960 (TriSep Corporation), spiral-wound configuration with an effective pore size of 0.03 μm and 20.9 m^2 filtration area. This type of membrane is highly resistant to chlorine (2000 mg Cl_2/L) and can operate at pH values of between 2 and 11. Clean-water membrane resistance for 1.0 m^3/h permeate flow rate at 20°C is 3.25 10^{11} m^{-1}.

The influent was drawn from the inlet channel by a centrifugal pump, JEXM/A 120, Ebara (Italy), which discharged the raw water into the membrane tank. The water was filtrated from outside to inside by a permeate pump, Koral KS 75, Kripsol (Spain), creating a vacuum (-1.6 bar maximum), while the permeate was periodically reversed through the membrane with a backwashing pump, CHI2-20, Grundfos, (Germany). In addition, the membrane was continuously aerated by means of an air blower, SCL 15 DH, FPZ (Italy). The operation was dead-end and normally without a reject. However, a reject tank was installed for draining, overflow control, collection of cleaning spills, etc., and sampling points were provided for the influent and effluent (Fig. 1).

The plant could be operated either manually or automatically by the touch screen of the control panel. Besides, dosing pumps and chemical tanks (Fig. 1) were also available for cleaning in place (CIP), initially carried out every 24 h. During these operations, the membrane tank was emptied and filled with permeate while NaClO was dosed, followed by 20 min of recirculation. Additionally, the membrane remained soaked in clean water for several hours with an extra dose of NaClO so that TMP was recovered more quickly.

2.2 Experimental procedure

The plant treated surface water from the Genil River, Granada (Spain). The river receives some waste spills, resulting in medium to low-quality influent. Working conditions consisted of constant flux-production periods with backwashing phases in between. Likewise, in order to have enough data to evaluate the fouling, TMP and permeate temperature were registered every two min (RSG30 Endress Hauser) every working day. The flow rates were adjusted manually as the facility had rotameters (Stübbe) together with manual gate valves located in the discharge of each pump.

The study was made in two stages:

- Stage I: The main operating variables were tested for 10 weeks so that optimum values were found for different combinations. Every week, the permeate flow rate, backwashing flow rate, and duration were fixed, whereas 3 possible cycle times were tested. The backwashing duration could also be 30 or 60 s. The air blower flow rate was the only unmodified variable during this stage (Table 1). Optimum values were achieved according to the following criteria: longer operation times before reaching threshold pressure, higher permeate recovery, and better operability for the plant. The membrane fouling was also evaluated over the period.
- Stage II: Once the main variables had been optimised in the previous stage, they were fixed, varying only the aeration flow rates, which were increased daily from 9 m^3/h to the maximum allowed by the blower, 21 m^3/h (Table 1). The optimum value was the one that under the same conditions generated the least membrane fouling, evaluated in terms of fouling slopes. TMP immediately before backwashing was represented vs. operation time for each air flow rate, in order to assess the membrane fouling in each case. Moreover, the achievement of long operation times was no longer the goal, so the maximum experimental time was fixed at 600 min.

The energy consumption was calculated for both stages.

2.3 Analytical methods

Physico-chemical and microbiological analyses together with particle-size distribution were performed daily for the influent and effluent. The samples were integrated, 100 mL being collected every 30 min for the whole operation time. Suspended solids, turbidity, colour, total and dissolved organic carbon (TOC and DOC, respectively), $UV_{254\ nm}$ absorbance, and total aerobic bacteria were analysed for a total of 50 samples.

Suspended solids concentration (SS) was determined by using glass-fibre filters (Millipore AP4004705), according to UNE-EN 872. Turbidity was determined by measuring the scattered light (DINKO D-112) while TOC and DOC were measured using a combustion TOC Analyser (Formacs HT, SKALAR). For the analysis of colour (436, 525 and 620 nm) and $UV_{254\ nm}$ absorbance, UV-visible spectrophotometer Heλios γ (Spectronic Unicam) was used.

For the determination of the molecular size of the organic matter present in the influent and the effluent, both were fractioned monthly by means of Amicon Ultra-15 centrifugal filters units (Millipore Corp.). Cellulose membranes with 3, 10, 30, 50, and 100 kDa nominal molecular weight cut-offs were used. Centrifugation was performed for 10 mL volumes at 5000 rpm (Eppendorf 5804 Centrifuge), after which TOC was measured for the filtrates. The membranes were preserved with an organic solution which needed to be removed so as not to cause

Table 1 Operating variables used within each stage of the study

	Period	Permeate flow rate, m^3/h	Backwashing flow rate, m^3/h	Cycle time, min	Backwashing time, *s*	Air flow rate, m^3/h
STAGE I	Week 1	1	1.5	30-20-10	30	9
	Week 2	1	1.5	30-20-10	30	9
	Week 3	1	1.5	30-20-10	30	9
	Week 4	0.8	2	30-20-10	30	9
	Week 5	0.8	1.5	30-20-10	30	9
	Week 6	1	2	30-20-10	60	9
	Week 7	0.8	2	30-20-10	60	9
	Week 8	0.8	1.5	30-20-10	60	9
	Week 9	0.7	2	30-20-10	60	9
	Week 10	0.7	1.5	30-20-10	60	9
STAGE II	Day 1	0.7	2	10	30	9
	Day 2	0.7	2	10	30	12
	Day 3	0.7	2	10	30	15
	Day 4	0.7	2	10	30	18
	Day 5	0.7	2	10	30	21
	Day 6	0.7	2	10	30	9-12-15-18-21[a]
	Day 7	0.7	2	10	30	9-12-15-18-21[b]

[a] For tap water; [b] For raw water

Table 2 Summary of influent and effluent characteristics

Parameters	Influent				Effluent				Efficiency (%)
	Max.	Min.	Average	S.D	Max	Min,	Average	S.D	
SS, mg/L	243.7	1.3	25.3	45.8	0.0	0.0	0.0	0.0	100.0
Turbidity, NTU	172.0	4.4	23.2	31.3	4.4	0.1	1.3	0.8	94.5
Colour $_{436}$, m^{-1}	1.1	0.1	0.3	0.2	0.4	0.0	0.2	0.1	30.0
Colour $_{525}$, m^{-1}	0.8	0.1	0.2	0.1	0.3	0.1	0.2	0.1	11.0
Colour $_{620}$, m^{-1}	0.7	0.1	0.2	0.2	0.6	0.0	0.2	0.1	0.0
TOC, mg/L	44.6	5.0	11.5	7.9	11.8	3.5	7.5	1.9	34.3
DOC, mg/L	11.5	4.3	7.5	1.7	10.4	4.2	7.3	1.6	1.8
UV $_{254}$, m^{-1}	6.8	2.2	3.3	1.1	5.5	1.7	3.0	0.9	7.6
SUVA, L/mg·m	0.6	0.3	0.4	0.1	0.6	0.2	0.4	0.1	-[a]
Total aerobic bacteria (22°C), CFU/mL	2839.1	54.6	1075.9	910.5	2.00	0.0	0.3	0.6	99.9[b]

[a] Not relevant; [b] 3.5 log removal

interference, and thus prior to fractionation all the ultrafiltration membranes were soaked and centrifuged with 10 mL of NaOH (0.25 N) solution.

Particle-size distribution (PSD) was determined daily using a particle counter (LiQuilaz-S02-E20, Particle Measuring Systems), measurements being based on laser-light extinction. A volume of 5 mL set at a fixed rate was analysed for each sample. Particles were detected in two size ranges, 0.2 to 2.0 μm (S02) and 2.0 to 125 μm (E20). The system was calibrated using inert latex particles of defined size.

Regarding microbiological analyses, the samples were collected in 120-mL sterile specimen containers. Total aerobic bacterial analyses were made at 22°C, according to ISO standard 6222:1999.

2.4 Statistical methods

Statistical analyses consisted mainly of building regression models and/or comparing different samples determining the existence of statistically significant differences among them with a significance level of 5%, $p < 0.05$. The tests used were analysis of variance (ANOVA).

3. Results and discussion

3.1 Effluent quality

Microbiological analyses revealed that despite the continuous presence of bacteria in the influent, the membrane was almost 100% efficient at removing the total aerobic bacteria (Table 2), with only a few exceptions that could be attributed to contamination in the permeate zone. Admitting recycling of backwash water is considered a potential source of contamination (Betancourt and Rose 2004), and the frequent contact between the inside membrane wall and the treated water from the permeate tank during backwashing hases, the presence of bacteria in the effluent can be easily understood. Poor membrane performance is negligible, especially taking into account the size of bacteria (0.5 to 10 μm) is considerably higher than the membrane pore (0.03 μm). Regarding physico-chemical analyses, it bears emphasizing that the raw water was not only of low quality but was also quite variable, with each parameter presenting very wide ranges.

The ultrafiltration process is highly efficient for the removal of turbidity and SS, (Jacangelo *et al*. 1997, Laîne *et al*. 2000), as confirmed by the results for both parameters (Table 2). However, the yields found for colour were very poor and rarely exceeded 50%. In general, the effluent quality was high and it was not affected by the characteristics of the influent except for NOM removal. TOC-removal efficiency values were medium although considerably higher than those for DOC and UV_{254}, indicating that most of the dissolved organic matter could pass through the membrane (Table 2). In fact, there were no statistically significant differences between influent and effluent for either UV_{254} nm or DOC, this confirming that DOC can barely be removed by the membrane. On the other hand, the quality of the effluent proved to be independent of the operating conditions chosen, as ANOVA tests indicated that there were no statistically significant differences in effluent quality reached under different operating conditions.

Regarding the particle-size distribution, the yields were in general very high, although they varied depending on the range analysed. For the first range measured, particles between 0.2 and

Table 3 Summary of particle-size distribution

	Influent			Effluent		
	Max.	Min.	Average	Max.	Min.	Average
0.2-2.0 μm						
Particle size, μm	2.0	0.2	1.1	2.0	0.2	1.1
MCS, μm	1.0	0.5	1.6	0.2	1.9	0.5
MCS distribution, counts/mL × 10^3	14.8	0.3	8.2	8.5	$1.0 \cdot 10^{-2}$	0.9
2.0 -125 μm						
Particle size, μm	58.0	2.0	30.0	30.0	2.0	16.0
MCS, μm	2.0	55.0	9.0	2.0	28.0	6.0
MCS distribution, counts/mL × 10^3	59.4	$3.3 \cdot 10^{-5}$	2.2	0.1	$4.0 \cdot 10^{-5}$	$3.7 \cdot 10^{-3}$

2.0 μm, null efficiency proved to be below 0.5 μm, with a constant presence of those particles in the effluent, far more significant than in the influent. This finding can be attributed to the possible sweep of particles from the permeate tank to the permeate circuit during backwashing or chemical cleaning. The origin of those particles could be biofilms, incrustations, accumulation of organic matter, etc., removed by the action of the chemical reagent, which could be retained on the clean side of the membrane, later appearing in the effluent (Rojas *et al*. 2008). On the other hand, some of the particles in the influent could break up due to the turbulence caused by aeration (Braak *et al*. 2011, Liu *et al*. 2014) or even as a result of the interactions with the membrane, which would also explain the high content of small-sized particles in the effluent. Nevertheless, for sizes over 0.5 μm the yields were higher, 98.5% being the average value. In addition, the particle removal for the 2-125 μm range was excellent, the average yield being 99.6% and the minimum 94.0%.

Maximum, minimum, and average particle sizes for influent and effluent are presented in Table 3, together with maximum count size, MCS (Fernández *et al*. 2012), the size where the major

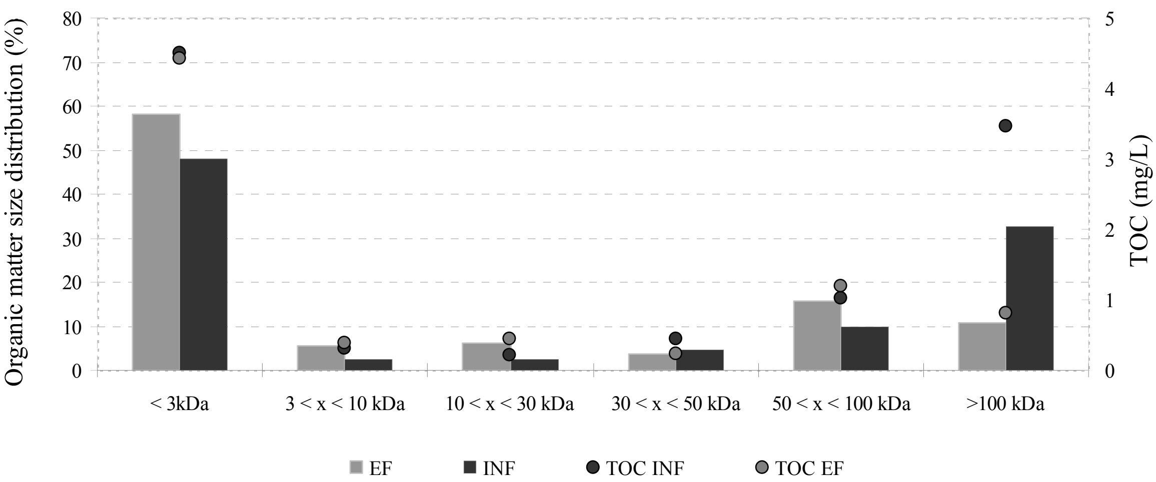

Fig. 2 Average molecular sizes for influent and effluent particles

count of particles was found. Maximum size for particles present in the effluent was 30 μm whereas the largest particles detected in the influent were 58 μm. The most frequent sizes were small, i.e., 2.0 μm in the influent with $59.4\cdot10^3$ counts/mL and 0.2 μm in the effluent, with $8.5\cdot10^3$ counts/mL (Table 3).

With respect to organic-matter fractionation, the results showed particles of between 3-10 kDa, 10-30 kDa, and 30-50 kDa were almost absent in the influent (the three bands represented less than 10% of the total in all the analyses), suggesting they can form aggregates, since particles over 100 kDa were very common (35% average). Nonetheless, the majority size for the influent was below 3 kDa (45% average). In addition, the organic content of this size was frequently higher in the effluent (59% average), in consistency with the particle-size-distributions results. Most of the analyses presented null yields for 3 kDa size. On the contrary, particles over 100 kDa were considerably reduced in the effluent (10% average), presenting the highest efficiency. TOC content for every band together with the percentage distribution is shown in Fig. 2.

According to the results, the membrane was very effective in removing particles over 100 kDa. However, intermediate size bands such as 3-10 or 10-30 kDa, barely present in the influent, had a more significant organic content in the effluent, the fraction below 3 kDa being especially remarkable. In view of the results for both particle-size distribution and fractionation, it appeared that the turbulence from the aeration could have fragmented the particles in the raw water.

3.2 Operation of the plant and optimisation of operating variables

Though operating conditions did not affect the effluent quality, they did influence the membrane behaviour, being related to how quickly the fouling was generated. The combinations presented in Table 1 were assayed weekly, although from the first day it was clear that the membrane could not work continuously due to the low quality of the influent, as the threshold pressure (-0.7 bar) was reached within a few hours (Fig. 3).

According to the membrane supplier recommendations, this pressure should not be exceeded,

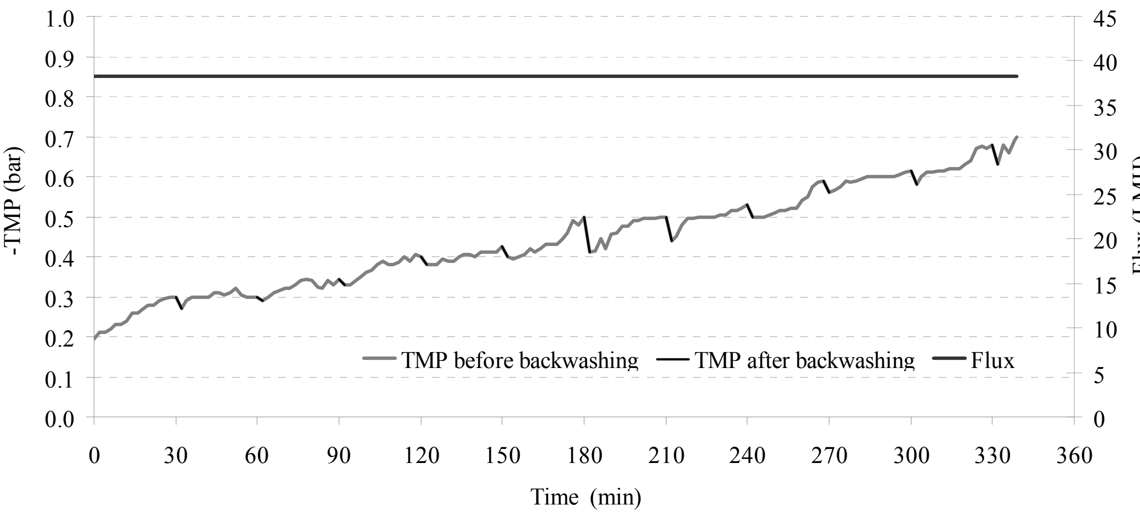

Fig. 3 TMP and Flux profiles with time lapse for a normal working day

and therefore the operation was interrupted and NaClO chemical cleaning was undertaken. Nevertheless, with time, membrane fouling became more and more severe and the starting TMP increased for every working day, so that it was deduced that the chemical cleanings applied were not effective enough.

According to the literature, chemical action during membrane cleaning is usually not sufficient to effectively reduce biofouling and particulate-fouling problems (Cornelissen *et al*., 2009). Particularly, in the case of spiral-wound membranes, biofouling and particulate fouling of the feed spacers is especially remarkable (Cornelissen *et al*. 2007). By contrast, membrane relaxation encourages diffusive back transport of foulants away from the membrane surface under a concentration gradient (Shi *et al*. 2014). As increasing the NaClO rates during the first chemical cleanings failed to induce any TMP recovery, the former were suspended. Thereafter, only relaxation periods were applied. The NaClO concentration in the membrane tank during relaxation periods increased up to 1000 mg/L at the end of the study, 10-fold higher than initially expected.

Despite the operation difficulties, the results for the optimisation of operating variables were conclusive. First, from all the combinations tested, it was deduced that the lower the permeate flow rates the longer continuous operation times for the membrane. However, the critical flux for the membrane seemed to be substantially below the range of flow rates chosen as the TMP increased very fast, even for the lowest values.

On the other hand, higher backwashing flow rates had a positive influence on the operation times as well but not as notable as the other backwashing parameters. Double backwashing times did not lead to longer operation times but they were certainly responsible for higher TMP recoveries, statistically significant differences being found for average TMP recovered after 30 and 60 s of backwashing. This signifies that over the medium and long term, increasing backwashing time can help improve the membrane performance by delaying the appearance of fouling.

Regarding cycle times, all other variables being equal, 10-min cycles allowed longer operation times and lower fouling velocities than 30 min, which in turn presented better results than 20 min. There were statistically significant differences for the fouling velocities with 10- and 20-min cycle times, but no differences were found between either 10 and 30 or between 20 and 30 min, 30 min being an intermediate value among the three of them. Some authors have reported better performance of ultrafiltration membranes with cake formation, with particles deposited on the membrane surface, facilitating the filtration process (Mosqueda-Jimenez *et al*. 2008). It may be possible that the material deposited on the membrane surface after 30 min had formed an additional layer, improving the membrane permeability while 20-min cycles would have been insufficient for layer formation. 10-min cycles would have been insufficient as well but this would have been compensated for by the higher frequency of the backwashing, enabling a lower fouling velocity. Nevertheless, further research is needed before drawing any conclusion in this respect.

3.3 Evaluation of fouling: Membrane resistance and fouling velocities

Fouling can be evaluated in different ways. Firstly, fouling velocity (bar/min) is given by the pressure difference TMP and TMP_0 divided by the time elapsed, Eq. (1). In particular, as TMP was registered every two min, fouling velocity was calculated for the same interval, and afterwards the average values were calculated for every working period. This parameter indicates how quickly TMP increases in relation to the initial value. Comparing average V_f for different experiments, it can be concluded when the fouling generation has been faster.

$$v_f = \frac{|TMP| - |TMP_0|}{2} \tag{1}$$

In general, the lowest fouling velocities were found for 0.7 m^3/h (0.0054 bar/min average), followed by 0.8 m^3/h (0.0060 bar/min), with the highest values for 1.0 m^3/h (0.0095 bar/min). It is noteworthy to remark that the lowest fouling velocities occurred at the end of the period (Weeks 9 and 10), when the membrane was already fouled and working under worse conditions. Therefore, if the order of the experiments had been the opposite, starting with the lowest flow rates instead, the differences between the lowest and the highest fouling velocities could have been more significant. However, certain peak values (0.013 bar/min) were reached for 0.7 m^3/h permeate flow rate, confirming the severity of irreversible fouling. For the same permeate flow rate, the higher the backwashing flow rate, the lower the fouling velocities. For low permeate flow rates such as 0.7 m^3/h and less frequently for 0.8 m^3/h, fouling velocities sometimes took negative values, indicating that a degree of TMP recovery could be achieved. In any case, these flow rates were less fluctuating and easier to control. According to the fouling velocities, the best combination of variables was certainly 0.7 m^3/h permeate flow rate, 2.0 m^3/h backwashing flow rate and 60 s backwashings performed every 10 min, which improved considerably the operability of the process.

Besides, the fouling of the membrane can also be quantified in terms of total membrane resistance (R_t, m^{-1}). Considering the relationship between flux, *J*, and *R*, this parameter can be calculated with the register of TMP and temperature, Eq. (2)

$$R_t = \frac{TMP}{J\ \mu} \tag{2}$$

TMP is expressed in Pa, *J* in $m^3/s \cdot m^2$ and μ is the dynamic viscosity in Pa·s.

The resistance in series model was employed, Eq. (3)

$$R_t = R_0 + R_i + R_r \tag{3}$$

R_0 is the intrinsic membrane filtration resistance, R_r is the reversible resistance, and R_i is the irreversible resistance (Kimura *et al*. 2004).

Given that R_0 was known, J was fixed, and μ depends only on the temperature; under the assumption that all the reversible fouling could be removed with the chemical cleanings/relaxation periods, R_i could be determined by calculating R_t with TMP after each chemical-cleaning plus relaxation periods. The R_i time course was monitored for the experimental period, showing a clear increasing trend (Fig. 4).

Several fluctuations for consecutive working periods were found, confirming that the cleaning strategy had not been the most appropriate one, since part of what had been considered irreversible fouling could be removed during relaxation periods. CIP is relatively simple to apply but its effectiveness is limited (Shi *et al*. 2014). On the other hand, it is a complex process influenced by many factors: membrane composition, cleaning agents, fouling components, and interactions among the three (Porcelli and Judd 2010). Consequently, the tendency of R_i to increase can easily be attributed to the inefficiency of the cleaning periods together with the high contaminant load of the influent.

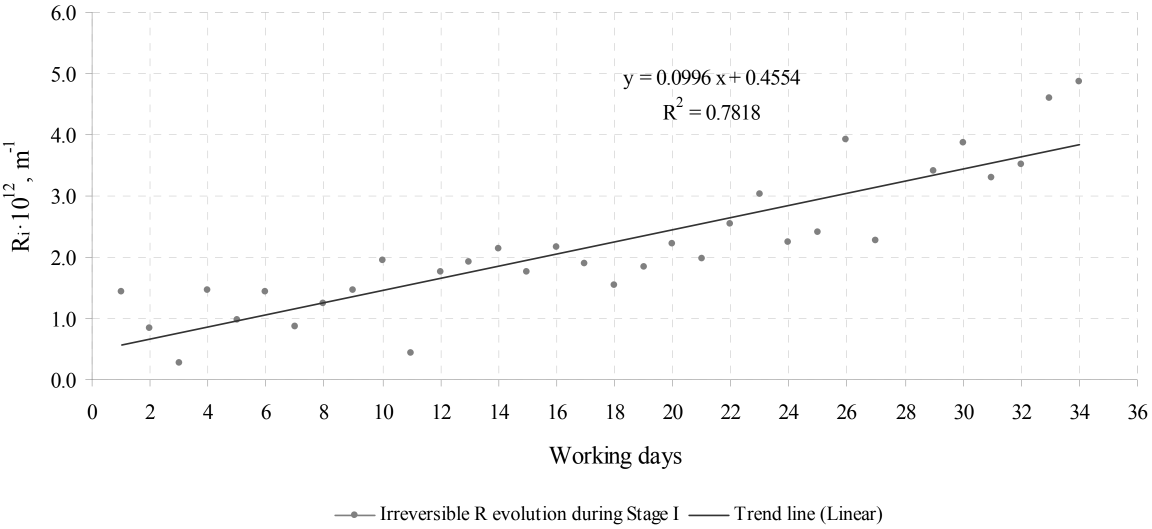

Fig. 4 Time course of irreversible membrane resistance, R_i, during stage I

For prospective experiments with such influent qualities, another combination of chemical agents shall be tested, e.g., NaClO together with NaOH as the combination oxidant-alkali has been shown empirically to be more effective than oxidant cleaning agents alone, especially where organic foulants dominate (Porcelli and Judd 2010).

3.4 Aeration experiments

After the optimisation of the rest of operating conditions, the best aeration flow rate was determined starting with the premise that aeration could enhance membrane performance: concentration polarization and fouling can be reduced by promoting mixing and turbulence in the fluid flow (Bellara *et al*. 1996). Gao *et al*. (2011) stated that aeration provides certain surface shear or enough mass transferring motion to avoid the foulants from compacting on the membrane surface. Consequently, fouling was evaluated for different air flow rates in terms of the so-called fouling slopes, which can be defined as the average fouling velocities for the corresponding operation time.

Linear regression was applied for TMP vs. time for 600 min of daily experiments but the results were not the expected ones. Despite that slopes progressively descended from day 1 (9 m^3/h) to day 5 (21 m^3/h), the slope for 9 m^3/h was lower than that for 12, 15, and 18 m^3/h, although higher than for 21 m^3/h so apparently no pattern could be found. However, given that the influent was natural water, its quality was unpredictable from one day to another, and it could have varied for each experiment. In fact, the raw-water analyses revealed that the influent DOC content was lower when testing 21 m^3/h than for 9 m^3/h, which at the same time was lower than for the intermediate flow rates, so the results could not be attributed to the aeration but to the water quality.

Bellara *et al*. (1996) reported an increase in the permeate flux as a consequence of gas sparging, although it was not very sensitive to dextrane feed concentration. On the contrary, Ghosh (2006) noted that the effectiveness of gas sparging increased with higher feed concentrations, again for dextrane solutions. Cabassud *et al*. (1997) found that for clay suspensions the enhancement of flux due to air injection gradually fell when the clay concentration rose, although there was a critical concentration for which the flux enhancement grew again. In all the cases, hollow-fibre ultrafiltration membranes were used but the composition of the respective feeds differed markedly, some of them being organic and other being inorganic, and the results proved quite different, too. In any case, none of the experiments were made with natural water. This fact, together with the few reported data on spiral-wound membranes underscored the need for experiments using the same feed-water composition prior to making an assessment. Therefore, a new test was conducted with tap water, in order to guarantee the operation of the membrane under fixed water quality. The experiment lasted 450 min, changing the air flow rate consecutively every 90 min. The results were satisfactory; for a normal working day with a fixed air flow rate, the fouling slope remained practically constant, but in this set of experiments, when the air flow rates were higher the slopes were slightly shallower, although the changes in TMP were very low (only 1%). The tap water could definitely not be compared to raw-water quality but at least it revealed that the TMP was somehow affected by the air flow rate.

Eventually, a final experiment was performed, exactly the same as the former but with raw water instead. Integrated influent samples were taken every 10 min for each 90-min period, giving ± 0.1 mg/L DOC differences among them so that the water quality could be considered constant. The results are shown in Fig. 5.

The order of magnitude found for the fouling slopes was clearly much higher than with tap water but the values decreased as the air flow rate rose. The TMP growth, in contrast to stage I, was asymptotic, indicating that the aeration had a positive effect, delaying the formation of the fouling layer on the membrane. The graph reflects only a slight difference between 18 and 21 m^3/h flow rates, in accordance with the results reported by other authors. Cabassud *et al*. (1997), Laborie *et al*. (1998) and Ghosh (2006) reported that the flux enhancement achieved by aeration presents a ceiling value, meaning a further increase in the air flow rate does not result in any significant improvement in the permeate flux. In this case, the operation was flux constant but the results were comparable — that is, once the air flow rate had been increased to a certain extent, a further increase did not lower the fouling slopes.

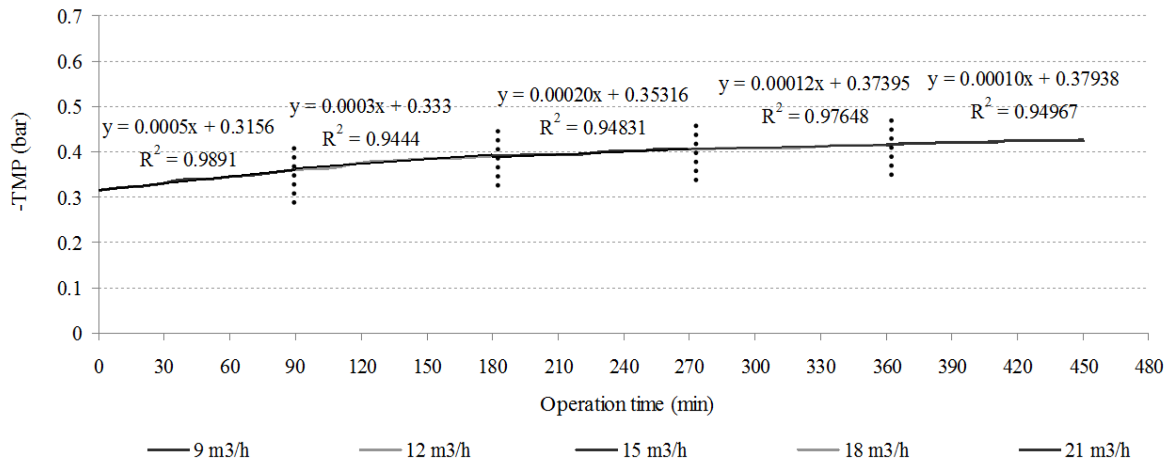

Fig. 5 Fouling slopes for different air flow rates with raw water

3.5 Evaluation of the energy consumption

The pilot plant used for the experiments had already worked within a different location and a different water quality. The transmembrane pressure then was -0.2 bar (Rojas *et al*. 2008, 2010), and therefore this value was initially expected and accepted as normal, but as stated in Section 3.2, the TMP rose quickly and the threshold pressure was reached daily. Consequently, after finishing stage I, the deviations from the expected energy demand were assessed for all the combinations of variables tested. In addition, the energy demand was also calculated when no operating variables were modified with the consequent increase in the TMP.

During stage I, permeate and backwashing pumps were the only equipment subject to changes, and therefore only the pumps involved were taken into account in the calculation.

The energy consumed by any pump can be calculated as the product of the hydraulic power and the operation time divided by the efficiency. Given that the hydraulic power is in turn the product of the pump flow rate and the differential pressure, the total energy consumption (kWh) was calculated according to Eq. (4).

$$E_{pumps} = \left(\frac{Q_p \ \Delta P_p \ t_p}{\eta_p} \right) + \left(\frac{Q_b \ \Delta P_b \ t_b}{\eta_b} \right) \tag{4}$$

where Q_p and Q_b are, respectively, the permeate and backwashing flow rates (m^3/s), t_p and t_b are the permeate production time (*h*) and the backwashing duration (*h*); ΔP_p and ΔP_b (Pa) are, respectively, the differential pressures for permeate production and backwashing periods, while η_p and η_b are the efficiency values for the two pumps, which were considered 0.7 in both cases (Massé *et al*. 2011).

Nonetheless, all the cases tested in the plant could not be compared in an exact way as the operation times were unpredictable and differed for each working day. However, they could certainly be compared by referring all the calculations to 1 hour.

Besides, the specific energy consumptions (kWh/m^3) were calculated considering the volume of permeate (m^3) produced in every case.

$$E_{pumps_p} = \frac{E_{pumps}}{(Q_p \ t_p - Q_b \ t_b)} \tag{5}$$

Moreover, the permeate recovery (*R*, %) is considered a good indicator of the process feasibility, at it denotes the ratio between the volume of permeate produced and the one spent during backwashing. Thus it was calculated according to the following expression.

$$R(\%) = \frac{Q_p \ t_p - Q_b \ t_b}{Q_p \ t_p} \tag{6}$$

Case 1 was considered the normal one and the energy consumptions together with their respective permeate recoveries for the rest of the cases were compared to it (Table 4).

From Table 4, it can be deduced that if the backwashing conditions were optimised in order not to have such high pressure increases, the energy consumptions shot up, especially the lower the permeate flow rate was . For example, for 1 m^3/h permeate flow rate, lowering the cycle time to 20 min involved increasing the energy consumption up to 3%, and 13% if the cycle time was 10 min.

Table 4 Summary of specific energy consumptions for all the cases tested in Stage I

Case	-TMP, bar	Q_p, m^3/h	Q_r, m^3/h	Cycle time, min	Backwashing duration, *s*	Specific energy consumption (Wh/m^3)	Energy difference (%)	Permeate recovery (%)
Normal/1	0.2	1	1.5	30	30	8.4	-	97.5
2	0.2	1	1.5	20	30	8.7	103.0	96.2
3	0.2	1	1.5	10	30	9.5	112.7	92.1
4	0.2	1	2	30	60	9.4	111.9	93.1
5	0.2	1	2	20	60	10.3	122.2	89.5
6	0.2	1	2	10	60	13.6	161.9	77.8
7	0.2	0.8	1.5	30	30	8.5	101.4	96.8
8	0.2	0.8	1.5	20	30	8.8	105.2	95.2
9	0.2	0.8	1.5	10	30	9.9	117.7	90.1
10	0.2	0.8	1.5	30	60	9.2	109.1	93.5
11	0.2	0.8	1.5	20	60	9.9	117.7	90.1
12	0.2	0.8	1.5	10	60	12.6	150.4	79.2
13	0.2	0.8	2	30	30	8.8	104.9	95.8
14	0.2	0.8	2	20	30	9.3	110.6	93.6
15	0.2	0.8	2	10	30	10.9	130.2	86.8
16	0.2	0.8	2	30	60	9.8	116.7	91.4
17	0.2	0.8	2	20	60	10.9	130.2	86.8
18	0.2	0.8	2	10	60	15.6	185.3	72.2
19	0.2	0.7	1.5	30	60	9.4	111.4	92.6
20	0.2	0.7	1.5	20	60	10.2	121.5	88.7
21	0.2	0.7	1.5	10	60	13.5	160.9	76.2
22	0.2	0.7	2	30	60	10.1	120.3	90.1
23	0.2	0.7	2	20	60	11.4	136.3	85.0
24	0.2	0.7	2	10	60	17.2	204.3	68.3
TMP 2	0.5	1	1.5	30	30	20.6	245.4	97.5
Threshold	0.7	1	1.5	30	30	28.8	342.3	97.5

The same changes for 0.8 m^3/h flow rates involved 5% higher energy consumptions for 20-min cycles and 18% more for 10-min cycles, (cases 8 and 9, respectively). The higher the number of variables that simultaneously changed, the greater the differences were. For example, if the backwashing duration was increased together with lowering the permeate flow rate to 0.7 m^3/h, the increment would be 22 and 61%, respectively for 20- and 10-min cycle times (cases 20 and 21).

On the other hand, delaying the fouling appearance by intensifying the backwashing and reducing the flux not only increased the energy demand but also compromised the permeate recovery. Thus, case 24 led to the longest operation times and the lowest fouling velocities, but also resulted in an energy consumption more than 2-fold higher and the lowest permeate recovery as well (68%).

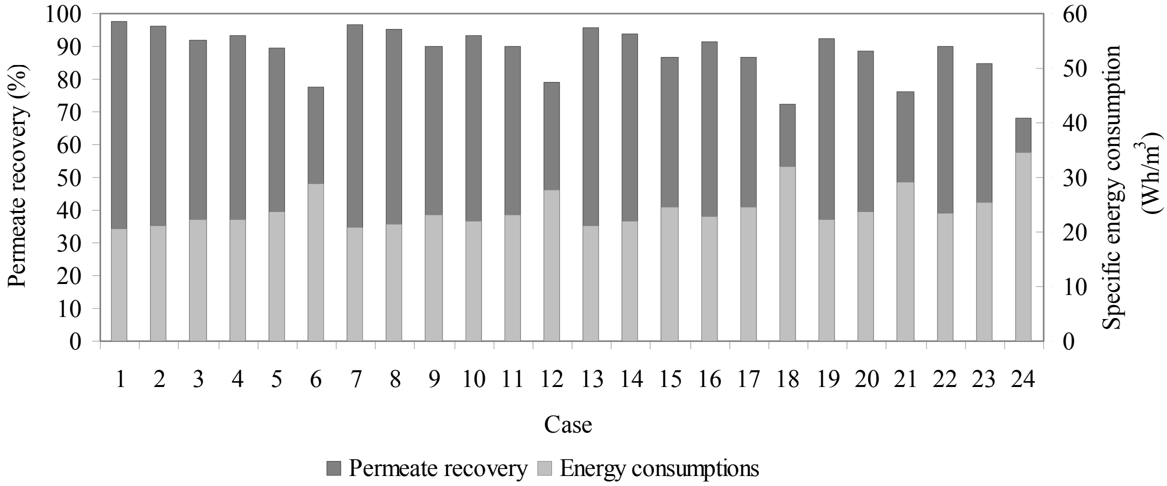

Fig. 6 Permeate recovery and energy consumption for all the cases tested in stage I

Finally, the energy needed to produce every m^3 of permeate was 245% higher due to the TMP increase from -0.2 to -0.5 bar, increasing up to 342% if threshold pressure was reached.

Fig. 6 summarises Table 4, making it possible to appreciate that the highest energy consumptions were consistently also those with the least permeate recoveries.

In stage II, the energy demand for the pumps was constant as the only variable changed was the air flow rate, the increase of which also had an impact on the energy consumption.

The power required for an air blower (kW) is given by Eq. (7) (Judd 2011)

$$P_{blower} = \frac{P_1\, T_1\, \lambda}{2.73\, x\, 10^5\, \eta_a\, (\lambda - 1)} Q_a \left[\left(\frac{P_2}{P_1} \right)^{1-\frac{1}{\lambda}} - 1 \right] \tag{7}$$

where P_1 and P_2 are the inlet and outlet pressure (Pa), respectively; T_1 is the inlet temperature (K); λ is the ratio of specific heat capacity at a constant pressure to specific heat capacity at a constant volume (c_p/c_v); Q_a is the blower flow rate (m^3/s); and η_a is the blower efficiency.

The specific energy demand (kWh/m^3) was calculated according to Eq. (8) (Massé *et al.* 2011), which can be deduced from Eq. (7), considering P_2 to be a function of P_1 and the membrane tank depth, d (m), and dividing the whole expression into the permeate flow rate (m^3/h)

$$E_{blower\, p} = \frac{P_1\, T_1\, \lambda}{2.73\, x\, 10^5\, \eta_a\, (\lambda - 1)} \left(\frac{Q_a}{Q_p} \right) \left[\left(\frac{P_1 + 10000\, d}{P_1} \right)^{1-\frac{1}{\lambda}} - 1 \right] \tag{8}$$

where P_1 was atmospheric, the inlet temperature for the air was 25°C; and the membrane tank depth was 1.3 m in our case. λ is normally assumed to be constant and equal to 1.4 for air whereas η_a was assumed to be 0.6 (Judd 2011).

Table 5 shows the differences for the range of flow rates tested.

It is clear that all other variables being equal, the difference in the energy demand for the

Table 5 Summary of specific energy consumption for the aeration flow rates tested in Stage II

Q_p, m^3/h	Q_a, m^3/h	E_{blower}, Wh/m^3	Growth factor
0.7	9	81.5	1.0
0.7	12	108.6	1.3
0.7	15	135.8	1.7
0.7	18	162.9	2.0
0.7	21	190.1	2.3

blower was proportional to the ratio between the air flow rates, and therefore once again the optimisation of the operating conditions had a heavy impact on the economy of the process.

In short, to improve the operability of the plant by optimising the operating conditions involved compromising the permeate recovery and augmenting the energy consumption. Nonetheless, the most significant factor affecting the energy consumption was still the growth of TMP as a consequence of fouling, so that it could be more convenient to try to lower it by other means, for example with the application of a pre-treatment that does not have such a high impact on the energy demand.

4. Conclusions

Potabilization by means of aerated spiral-wound ultrafiltration membranes enables high-quality effluents, especially regarding turbidity and microbiological displacement although the yields for NOM and colour removal are very low. In any case, effluent quality is independent of the operating conditions chosen, although they do have an influence on the performance of the membrane as they are related to how quickly the fouling is generated. The optimisation of operating variables such as decreasing the permeate flow rate, increasing the backwashing flow rates, shortening the production times and enlarging the backwashing duration contributes to slowing down the fouling generation and to lengthening the operation times but at the expense of augmenting the energy consumptions, which doubled in some of the cases hereby studied. On the other hand, not only was the energy demand higher but the permeate recovery also dropped considerably, the resulting minimum being 68%. In line with the former, increasing the aeration has proved to be very positive in order to lower the fouling velocity, tending to stabilize the fouling slope, with TMP presenting an asymptotic behaviour but again raising the energy consumption. Nonetheless, if normal operating conditions remained unchanged, the energy consumption of the pumps rose to 245% due to the TMP increase and even 342% when threshold pressure was reached (-0.7 bar).

These findings, together with the need of long relaxation periods after chemical cleanings, the increase in chemical doses and the reduction of the membrane lifespan make a single-treatment ultrafiltration not a recommendable option when the quality of the influent is low. Further research will be performed in order to assess whether the application of any pre-treatment can avoid the TMP growth without entailing such high rises in energy demands. This will help determine whether this technology is suitable for any type of water, and, if so, this would be promising as the use of low-quality surface waters is expected to become steadily more necessary in the future.

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