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Ultrafiltration as tertiary treatment for municipal wastewater reuse

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ABSTRACT

Water reuse is an enduring topic that benefits the society and future generations of mankind. Ultrafiltration (UF) is one of the most cost-effective treatment technologies for improving water quality. In this study, a semi-industrial UF pilot plant with periodical classic backwash (CB) and air backwash (AB) was operated automatically to evaluate its feasibility and sustainability for municipal wastewater reuse and find out the optimized filtration condition. This study carried out 15 filtration conditions to investigate the impacts of flux (J in $\text{L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$), filtration cycle time (t in min), and air backwash frequency (BW) on membrane hydraulic filtration performance and membrane fouling management. Through comparative analysis of all conditions in water quality, permeability variation, irreversible fouling management, and water recovery rates, the sustainable conditions J80t40BW1/3 (flux of $80 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, filtration cycle time of 40 min, 1 AB followed with 3 CBs), J60t60BW1/4 (flux of $60 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, filtration cycle time of 60 min, 1 AB followed with 4 CBs), and J60t60BW1/3 (flux of $60 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, filtration cycle time of 60 min, 1 AB followed with 3 CBs), stood out from the others with higher overall performances. Additionally, air backwash showed excellent reversibility on membrane fouling control, which was around 1.25–2 times that of CB in average. After all, long term operation on condition J60t60BW1/3 in winter and in summer confirmed that the UF system could provide sustainable and adaptable filtration performance regardless of the temperature and feed water quality. The UF permeate quality is good enough to be reused in non-potable applications as it met reuse guidelines of the World Health Organization, reuse standards of France and the most recent EU regulation for agricultural irrigation. This work confirms the great interest of UF as tertiary treatment for water reuse and gives operational indications for future industrial-scale production of reclaimed water.

1. Introduction

Water scarcity has long been a worldwide crisis under the increasing pressure of population growth, climate change, and various pollutions. Water reuse, defined as using wastewater or reclaimed water from one application to another application, is a promising approach to alleviate water crisis sustainably. However, the total volume of reused water all over the world, which is approximately $14.2 \text{ billion m}^3\cdot\text{y}^{-1}$, represents less than 4% of the total volume of domestic wastewater [1]. Moreover, the reuse proportion differs a lot among different countries and regions. In Europe, approximately 2.4% of the total treated wastewater effluents was reused in 2015 [2], while France only reused around 1% [3]. Water reuse market is still young and limited, which needs to be highly improved in the future. Nowadays, more and more advanced treatment technologies for municipal wastewater reuse have emerged, and

membrane filtration is one of the main unit technologies. Generally, ultrafiltration (UF) has been widely accepted as one of the most cost-effective technologies to deal with secondary treated effluent on non-potable reuses, such as agricultural irrigation, landscape irrigation, urban reuse, and process water for industries [4–7]. UF enables high removal efficiency on total suspended solids (TSS), turbidity, organic matters, and microorganisms, etc [4,8]. On reuse purpose, the studies by Muthukumaran *et al.* [9], and Pollice *et al.* [10] or Falsanisi *et al.* [11] all confirmed that UF (with prefilter) can support qualified permeate water for reuse, all with respect to World health organization (WHO) reuse guidelines. However, regulations are becoming stricter in terms of concentrations of *E. coli*, biological oxygen demand (BOD_5), TSS and turbidity such as in the higher reclaimed water quality class (A) of the Regulation (EU) 2020/741 of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse [12].

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Therefore, there is a need to confirm if UF can meet these new standards.

In addition, fouling formation limits the economical operation of UF process on wastewater treatment, [13]. Generally, the total fouling resistance in UF filtration is mainly composed of hydraulic reversible resistance and irreversible resistance [14]. The macro-solute or particle deposition on top of membrane surface mainly caused by suspended solids is usually reversible and non-adhesive [15]. The pore blocking and internal pore adsorption on membranes caused by organic matters is usually irreversible and adhesive [16]. To control fouling, filtration conditions (flux, filtration time, cleaning) need to be optimized because they can directly affect either the formation and nature of the fouling layer, or the removal mechanism of the foulants. For example, the filtration flux and filtration cycle time will affect the density and the thickness of the fouling layer deposited on the membranes [17]. Besides, frequent cleanings for membranes are required, including physical and chemical washing, to maintain good filtration performance. Chemical washing is effective on irreversible foulants removal through the reaction between fouling and chemicals. The choice of chemical agents, mostly using acids, alkalis, or oxidants, is based on the membrane materials and fouling types [18]. Physical cleanings such as classic backwash (CB) and air assisted backwash (AB) are commonly used methods in membrane fouling control [19]. CB can loosen and detach fouling cake partially from membrane surface, and AB is a more effective anti-fouling method compared to CB [15,20,21]. Normally, AB can be performed either by air sparing in backwash water, or by air injection into membrane fibers. A previous study by Y. Ye *et al* [20] on a model mixture of bentonite and alginate demonstrated that the membrane system using air bubble backwash in moderate air flow rate (e.g. 154 mL·min⁻¹) could remove more foulants without recompression potential while the loosen and residual foulants after CB would be recompressed on membrane surface when filtration restarts. P.J. Remize *et al* [22] also stated that AB with air injection enhanced particles removal during long-term filtration in drinking water production. Additionally, C. Cordier *et al* [23] confirmed the higher removal ability of AB with air injection, and it also destroyed the cells' integrity by air injection, such as oysters' oocytes and spermatozoa in seawater. Frequent AB can bring some other disadvantages such as pore blocking inside membranes [20]; partial drying which can cause membrane integrity problems [19]; more energy and equipment cost requirements. Considering the pros and cons of AB and CB, it is therefore interesting to alternate cleaning by CB and AB during filtration and it is very important to study the optimum backwash frequency of AB and CB as it has never been done on municipal secondary wastewater effluent. In addition to optimum AB frequency, filtrations conditions such as flux and filtration time have also never been optimized in the scientific literature for this effluent whereas these are crucial to control fouling and enable the wide development of water reuse by UF.

In this context, a semi-industrial UF pilot plant was set-up on-site with real secondary effluent for reuse application and was operated with periodic CB and AB. The targets of this study are: (i) to check the permeate quality for reuse according to EU regulation adopted in May 2020 among others, (ii) to find out the sustainable filtration conditions (flux, filtration cycle time, AB frequency) enabling sustainable flux and low operating costs, (iii) to check the UF performances as tertiary treatment for municipal wastewater reuse in the long term.

2. Material and methods

2.1. Feed water quality

The feed water is the secondary effluent of a municipal wastewater treatment plant (WWTP) located in *Châteauneuf-les-Martigues*, France. The WWTP uses a conventional activated sludge process to treat raw wastewater. Two experimental campaigns were conducted in winter and summer. **Table 1** shows the quality of the inlet and outlet effluent of the WWTP. The outlet effluent of the WWTP was the UF feed of this work.

Table 1
Raw wastewater and UF feed quality.

Parameters	Campaign 1 (October 2019 – March 2020)		Campaign 2 (July 2020)	
	WWTP influent	Outlet effluent (UF feed)	WWTP influent	Outlet effluent (UF feed)
<i>E. coli</i> (CFU 100 mL ⁻¹)	1.6 × 10 ⁸	(3.4 ± 2.6) × 10 ⁴	> 1.4 × 10 ⁶	(8.5 ± 9.4) × 10 ³
Enterococci (CFU 100 mL ⁻¹)	2.2 × 10 ⁷	(1.3 ± 1.0) × 10 ⁴	(3.1 ± 3.4) × 10 ⁷	(8.6 ± 9.5) × 10 ²
Anaerobic sulphite-reducers (spores) (CFU 100 mL ⁻¹)	5.6 × 10 ³	268 ± 253	(1.3 ± 1.7) × 10 ⁴	(9.6 ± 5.1) × 10 ²
Specific F-RNA bacteriophages (PPF 100 mL ⁻¹)	4.5 × 10 ³	< 30	(1.7 ± 1.8) × 10 ³	< 30
COD (mgO ₂ ·L ⁻¹)	1124	20 ± 9	552 ± 61	45 ± 21
BOD ₅ (mgO ₂ ·L ⁻¹)	n.m.	n.m.	205 ± 82	< 3
TSS (mg·L ⁻¹)	77	4 ± 2	367 ± 166	12.1 ± 8
TOC (mgC·L ⁻¹)	n. m.	18 ± 9	n.m.	n.m.
Turbidity (NTU)	n. m.	2.3 ± 0.9	208 ± 38	2.1 ± 1.6
pH	n. m.	7.2 ± 0.4	7.4 ± 0.3	7.5 ± 0.1
Conductivity (µS·cm ⁻¹)	n. m.	1 168 ± 128	n.m.	n.m.

CFU: colony-forming unit; PFP: Polyhedral, filamentous, and pleomorphic; n. m.: not measured.

COD: Chemical oxygen demand; BOD₅: Biochemical oxygen demand TSS: Total suspended solids; TOC: Total organic carbon.

The standard deviations (SD) were measured with 2–4 samples.

2.2. Ultrafiltration pilot plant description

A semi-industrial UF pilot plant manufactured from Aquasource (France) has been established for this study. The nominal capacity of the pilot plant is 20 m³ water per day. The pilot plant can be performed in manual mode, half automatic mode and automatic mode. Operating parameters shown in the flow diagram of the pilot plant in **Fig. 1** are recorded automatically every minute. The secondary effluent of WWTP as feed will be drawn into a feed tank T3 when the feed water drops to the lowest settled level in the pilot plant system. Tanks T1 and T2 are containers for permeate and backwash water. A concentration of 5 ppm of chlorine was added in T1 while in T2, corresponding to the water production tank, no chlorine was added. A 130 µm disk prefilter is designed before UF membrane to avoid UF hollow fiber clogging.

2.3. UF membrane and module

The membrane module is an ALTEONTM I (Aquasource, France) multichannel UF hollow fiber, with detailed information listed in **Table 2**. The module is operated in dead-end filtration mode with an inside-out configuration. Dead-end ultrafiltration is considered as an energetic efficient operation mode in large-scale water treatment systems [24]. The hydraulic resistance of new membrane modules was measured with pure water to be 4 × 10¹¹ m⁻¹ at 20 °C (pure water permeability = 900 L·m⁻²·h⁻¹·bar⁻¹). The maximum permeability (L_p) of UF membrane when filtrated with the feed water is around 600 L·m⁻²·h⁻¹·bar⁻¹ at 20 °C. Therefore, each filtration condition was started from initial L_p at around 600 L·m⁻²·h⁻¹·bar⁻¹, and the corresponding membrane resistance of 6 × 10¹¹ m⁻¹ at 20 °C was considered for future calculations.

2.4. Membrane cleaning

There are four types of membrane cleanings in the system: (1) Classical backwash (CB) using permeate water from T2 without chlorine

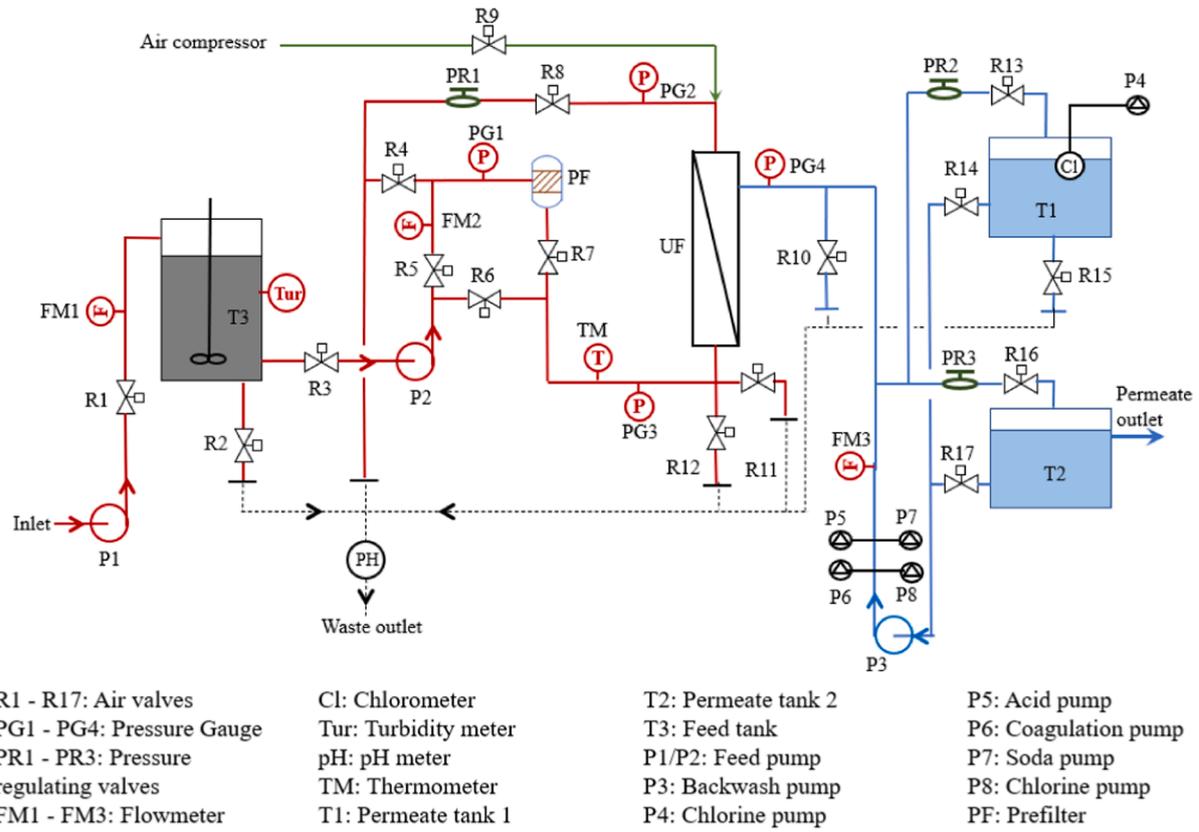


Fig. 1. Diagram of semi-industrial pilot plant.

Table 2
 UF membrane and module characteristics.

Characteristics	Data
Material	Polyethersulfone
MWCO	200 kDa
Pore size	0.02 μm
Length	1.2 m
Internal diameter	0.9 mm
Number of channels	7
Filtration Surface	9 m^2
Volume of fibers	2.0 L
Maximum TMP	2.5 bar
pH tolerant value	1-13

with a flow rate of $2.5 \text{ m}^3 \cdot \text{h}^{-1}$. The duration and the volume of water used during one CB is 52 s and 36 L, respectively. (2) Air backwash (AB) includes 3 steps: empty fibers, dry the fouling layer with air injection, and CB using permeate water from T1. This type of backwash was described by Cordier *et al.* [23,25]. The duration and the volume of water used during an AB is 67 s and 52 L, respectively. (3) Before new operating conditions start or if the permeability dropped below $200 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$ at real temperature, the chemical enhanced backwash (CEB) was conducted. Before CEB, the system carries out a backwash related to the backwash sequence that is in turn. The CEB starts with chemical injection into membrane modules and then soaking happens with chemicals to degrade foulants. Chemicals are either sulfuric acid ($[\text{H}^+] = 1000 \text{ ppm}$), or sodium hydroxide ($[\text{OH}^-] = 800 \text{ ppm}$) and chlorine ($[\text{active Cl}] = 50 \text{ ppm}$). The duration of soaking is 1200 s. After soaking, the system discharges the soaking solutions by gravity and then starts with a long-term CB without chlorine until pH of discharge water comes back to neutral. The duration and the volume of water used during a CEB is 1560 s and 250 L, respectively. (4) In addition, a forward flushing happens in the middle of each filtration cycle for about 10 s

with $2.0 \text{ m}^3 \cdot \text{h}^{-1} - 3.0 \text{ m}^3 \cdot \text{h}^{-1}$ of feed water from bottom to top of the UF module. The forward flushing can assist to decrease aggregation or dense attachment of particulates from the membrane surface [26].

2.5. Filtration conditions

The experiments were operated under constant flux mode (20 to $100 \text{ L} \cdot \text{h}^{-1} \cdot \text{m}^{-2}$). During three months' operation, the study conducted various filtration conditions to investigate the impact of filtration cycle time, AB frequency, and flux on hydraulic filtration performances. The AB frequency, represented by "1/n" in the following contents, such as 1/3, 1/4, 1/6, and 1/9, represents (n) times of CBs followed with 1 AB. 15 Different designed conditions were studied in this test and were separated into 3 main parts to study the impact of filtration cycle time, flux, and AB frequency on filtration performance, respectively (Table 3). Each condition was conducted continuously for >40 h for stable behavior. After each condition, several CEBs were operated manually to clean the membrane until the initial permeability reached to $600 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$, so as to maintain the similarly initial membrane state for all filtration conditions. To be noted, the parameters including flux and L_p , that could be affected by the temperature have been normalized to a standard temperature (20°C) to account viscosity fluctuations with these parameters.

2.6. UF performance analysis

2.6.1. Permeability

As the flux J is constant in the semi-industrial system, the relationship between J ($\text{L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) and L_p ($\text{L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$) can be expressed as Eq. (1).

$$L_p = \frac{J}{TMP} \quad (1)$$

Table 3

Filtration conditions.

	Flux (J) (L·h ⁻¹ ·m ⁻²)	Filtration time (t) (min)	AB frequency (Air Backwash/ Classic Backwash)
Impact of Filtration cycle time on filtration performance	60	20	1/3
		30	
		40	
		60	
		80	
Impact of Flux on filtration performance	30	30	1/3
		60	
		80	
		100	
		60	60
Impact of AB frequency on filtration performance	60	60	1/3
			1/4
			1/6
			1/9
			No AB

The pressure difference over a membrane is called the Transmembrane Pressure (TMP), unit *bar*, which is calculated by Eq. (2)

$$TMP = \frac{P_{feed} + P_{concentrate}}{2} - P_{permeate} \quad (2)$$

2.6.2. Fouling resistances

According to Darcy's law [27], hydraulic resistance of the fouled membrane was measured with Eq. (3):

$$R_t = R_{irr} + R_{re} + R_m = \frac{TMP}{\mu \cdot J} = \frac{1}{\mu \cdot L \cdot P} \quad (3)$$

where, TMP is the transmembrane pressure (Pa), μ is the viscosity of permeate (Pa·s) and J is the applied flux (m·s⁻¹). The total membrane resistance (R_t) includes three parts of resistances (m⁻¹): irreversible fouling resistance (R_{irr}) which cannot be removed by backwashes, reversible fouling resistance (R_{re}) which can be removed by backwashes, and membrane resistance (R_m).

The flux at 20°C is calculated through Eq. (4) [28]:

$$J(20^\circ C) = \frac{\mu(T)}{\mu(20^\circ C)} \cdot J(T) = J(T) \times e^{0.0239 \times (20-T)} \quad (4)$$

In two adjacent filtration cycles, it can be considered that the composition of fouling resistance in end of cycle (n-1) is:

$$R_{end}(n-1) = R_m + R_{irr}(n-1) + R_{re}(n-1) \quad (5)$$

While after a CB, the total resistance in the beginning of cycle (n) is:

$$R_{ini}(n) = R_m + R_{irr}(n-1) \quad (6)$$

Through difference in Eq. (7) (8), values of R_{irr} and R_{re} in the n-1 filtration cycle can be found.

2.6.3. Reversibility

Filtration performance can be evaluated by parameters such as backwash effectiveness, fouling rate, and fouling resistance [29]. Backwash effectiveness can be indicated by fouling reversibility which was calculated after each filtration cycle (n) according to [19,30]. Reversibility after each filtration cycle could then be calculated using the initial TMP and final TMP values of the cycle (n) (TMP_{end}^n and TMP_{ini}^n) as well as the initial TMP of the next filtration cycle (TMP_{ini}^{n+1}). As described above, the flux and viscosity at 20 °C are both constant during each filtration condition. Therefore, the reversibility can be calculated as follow Eq. (7).

$$Reversibility(n) = \frac{TMP_{end}^n - TMP_{ini}^{(n+1)}}{TMP_{end}^n - TMP_{ini}^n} = \frac{R_{end}^n - R_{ini}^{(n+1)}}{R_{end}^n - R_{ini}^n} \quad (7)$$

2.6.4. Water recovery rate

The water recovery rate represents the proportion of net produced water quantity when compared to the total inlet feed water quantity. This was calculated with Eq. (8).

$$Recovery \text{ rate} = \frac{V_{net}}{V_{inlet}} \times 100\% = \frac{V_{inlet} - V_{CB} - V_{AB} - V_{CEB} - V_{Con}}{V_{inlet}} \times 100\% \quad (8)$$

where V_{inlet} refers to the volume of inlet feed water (m³), V_{net} refers to the net volume produced (m³). The calculation of V_{inlet} is related to the filtration flux and filtration time, as in Eq. (9). The volume unit is the liter.

$$V_{inlet} = J \times t_f \quad (9)$$

V_{Con} refers to the concentrate volume, which is known to be 2 L (volume of fibers presented in Table 2). V_{CB} , V_{AB} , and V_{CEB} represent the volume of permeate consumed for CB, AB and CEB which are 36 L, 52 L and 250 L, respectively.

2.6.5. Impact of air backwash on irreversible resistance increase

To find out the influence of air backwash frequency on UF membrane filtration performance, the variation of irreversible fouling during operation can be an interesting parameter to follow. The relationship of R_{irr}/R_m variation with feed water turbidity integration (Eq. (10)) will be considered to investigate the influence of AB frequency on fouling formation whatever the quality of the feed water:

$$\frac{(R_{irr}/R_m)_n}{(R_{irr}/R_m)_{initial}} = f\left(\sum \int Turbidity \cdot dt\right) \quad (10)$$

where the turbidity integration with time is shown as follows with n = number of filtration cycle, and t the duration of a filtration cycle:

$$f\left(\sum \int Tur \cdot dt\right) = \sum_{i=1}^n \left[\int_0^t Turbidity \cdot dt \right]_i \quad (11)$$

2.7. Water quality assessment

Total suspended solids (TSS) were measured with a 47 mm glass-fiber-filter (Whatman). The filter was firstly rinsed with distilled water and dried during minimum 2 h at 105 °C after filtration of the sample. The TSS concentrations were then obtained by calculating the difference of weight before and after filtration divided by the volume of sample. The turbidity of feed was measured and recorded every minute by the pilot plant using a probe TurbiMax W CUS31 (Endress Hauser). Besides, the turbidity of permeate was also tested punctually in laboratory with a turbidity meter (Turb 550 IR, WTW, Germany). The electrical conductivity value was determined with a conductometer (Sension + EC7, Hach, USA). The pH was measured with a pH-meter (Sension + pH31, Hach, USA). During Campaign 1, COD analysis was conducted through reagent vials (COD cell test C3/25), with test range from 10 to 150 mg·L⁻¹. A volume of 3 mL of a homogeneous water sample was injected into a COD digestion reagent vial. Then, the vial contents were stirred and subsequently placed in the COD reactor at 148 °C for 2 h. Finally, the samples after cooling were tested by a spectrophotometer (190 – 1100 nm, Photolab 6600 UV-Vis, WTW). A TOC-L machine (Shimadzu, Japan) based on the 680 °C combustion catalytic oxidation method was used to measure the concentration of total organic carbon (TOC). The non-purgeable organic carbon (NPOC) method with a detection limit of 4 µg·L⁻¹ was used. During Campaign 2, COD, BOD₅, microorganisms including SARS-CoV-2 were all tested by an external laboratory (Carso –

Laboratoire Santé Environnement Hygiène de Lyon) certified by the French Ministry of Health for water analysis. The methods for COD and BOD₅ are respectively ISO 15,705 and NF EN ISO-5815-1. The microbiological analyses were also tested by Carso laboratory and consisted in the measurements of *E. Coli* (NF EN ISO 9308-1, NF EN ISO 9308-3), *Enterococci* (NF EN ISO 7899-1, NF EN ISO 7899-2), anaerobic sulphito-reducers (spores) (NF EN 26461-2), specific F-RNA bacteriophages (NF EN ISO 10705-1), and SARS-CoV-2 by real-time quantitative polymerase chain reaction (qPCR). The detection methods NF EN ISO 9308-1 for *E. coli*, and NF EN ISO 7899-2 for *Enterococci* were used in samples during Campaign 1 with both detection limits at < 1 CFU·100 mL⁻¹. The detection methods NF EN ISO 9308-3 for *E. coli*, and NF EN ISO 7899-1 for *Enterococci* are used in samples of Campaign 2 with both detection limits at < 56 CFU·100 mL⁻¹. For SARS-CoV-2 analysis, the envelope protein gene E is first tested and the nucleoprotein gene N is tested only if the envelope protein gene E is detected. The detection and identification of microplastics were analyzed by Spotlight 400 Fourier-transform infrared spectroscopy (FT-IR) imaging system from PerkinElmer after filtration of the samples through a 3 µm gold-coated filter. Before filtration, the water samples are first mixed with hydrogen peroxide solution (30 % w/v, fisher chemical) in a ratio of 1:1 for one day to remove microorganisms. Data treatment is realized by siMPLE (Systematic Identification of MicroPlastics in the Environment) software developed by Aalborg University, Denmark and Alfred Wegener Institute, Germany. For all analyses described previously, inlet and outlet (UF feed) of the WWTP and permeate were sampled and analyzed almost at the same time.

3. Results and discussion

3.1. Variation of feed water quality

The flowrate of raw wastewater in the WWTP changes regularly between day and night. Generally, periods with higher flowrate mean higher concentration of pollutants for the activated sludge process in the WWTP potentially decreasing its treatment efficiency [31]. Besides, higher flowrates lead to shortened sedimentation time resulting in decreased particles' removal rates [32]. Additionally, the weather can also affect the flowrate and the quality of the wastewater [33]. After all, the increase of raw wastewater flowrate will largely lead to deteriorated quality of WWTP effluent/UF feed, which is the main cause of UF membrane fouling. Turbidity can be an indicator of feed water quality

because it is related to the concentration of suspended solids. Fig. 2 shows the variations of feed water turbidity and raw wastewater flow-rate versus time for a standard day. The feed water quality variation seems positively correlated to the variations of raw wastewater flow rate as the two curves follow similar trends. Therefore, the feed water turbidity varies regularly everyday which has the similar peak and trough periods with flow rate variation of raw wastewater: this was undergone during all experiments.

The variations of permeability and feed water turbidity obtained in condition J60t20BW1/3 ($J = 60 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, $t = 20 \text{ min}$, 1 AB with 3 CB) are shown in Fig. 3. The variations of L_p showed several unregular over-increase periods, such as in circles (a) and (b) which correspond to the moments when the turbidity became lower (trough periods). It can be considered that trough periods with lower feed turbidity (Fig. 2) brought less fouling load on membrane thus the permeability decreased slower than in peak periods. In trough periods, the feed water contains less turbidity, suspended solids, and probably also fewer organic matters which results in lower foulant potential. Therefore, with periodical backwash (1 AB with 3 CB), the UF membrane would be cleaned more thoroughly in trough periods than that in peak periods, resulting in higher permeability recovery. The above analysis demonstrates that the over-increase of permeability during long term filtration is normal and regular, and usually occurs at the trough periods during a day, like the periods (a) and (b) in Fig. 3. The variations shown in Fig. 2 and Fig. 3 for two conditions are representatives from the others. All the conditions are faced with the same or similar impacts from external environment.

4. Permeate quality

Permeate quality analysis is critical to the realization of water reuse. To ensure the safety and reliability of reused water in the frame of this study, the permeate quality was compared to the three kind of guidelines and standards. Table 4 shows the UF permeate quality during both Campaign 1 (October 2019 – March 2020) and Campaign 2 (July 2020) with comparison to WHO guidelines, French reuse standard, and European parliament regulations for water reuse [12,34,35]. The removal rates between feed and permeate were calculated to evaluate the benefit of UF. Moreover, as required by the standards, the removal rates between WWTP feed and UF permeate were also calculated when possible. As a result, the UF system can greatly retain the microorganisms, including *E.coli*, *Enterococci*, spores of anaerobic sulphito-reducers, and bacteriophages, all under the detection limitation of the analysis

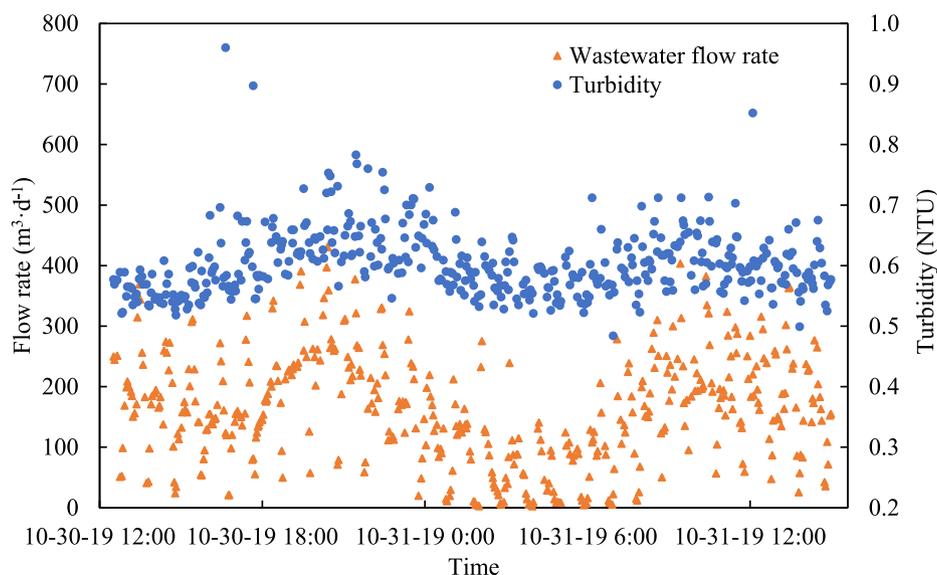


Fig. 2. Variation of flow rate and turbidity as the function of time.

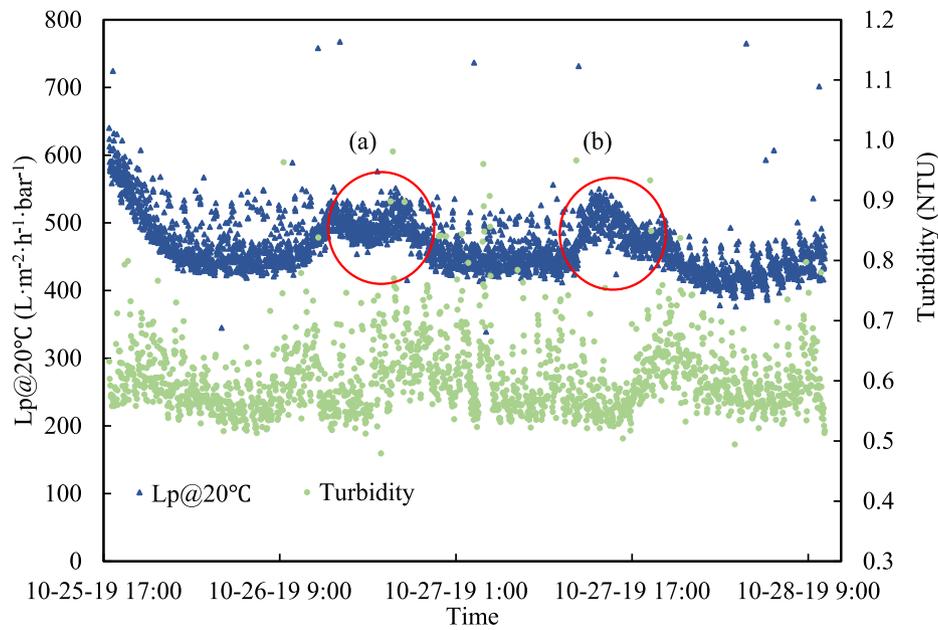


Fig. 3. Permeability and feed water turbidity variations.

Table 4

Mean UF permeate qualities compared to WHO guidelines for water reuse, French reuse standard, and European parliament regulation.

Parameters	Campaign 1 (October 2019 – March 2020)			Campaign 2 (July 2020)					WHO guidelines	French standard (A)*	European parliament regulation (A) *
	UF permeate			UF permeate							
	Mean ± SD	RRs* by WWTP + UF (%)	RRs* by UF (%)	6 th July 2020	16 th July 2020	28 th July 2020	RRs* by WWTP + UF (%)	RRs* by UF (%)			
<i>E. coli</i> (CFU-100 mL ⁻¹)	<1	>6.7 (log*)	>4.8 (log*)	<56	<56	<56	>6.5 ± 1.4 (log*)	>2.0 ± 0.5 (log*)	≤ 200	≤ 250	≤ 10 / ≥ 5 (log*)
<i>Enterococci</i> (CFU-100 mL ⁻¹)	<1	>6.2 (log*)	>4.4 (log*)	<56	<56	<56	>5.4 ± 1.4 (log*)	>1.1 ± 0.3 (log*)	-	≥ 4 (log*)	-
Anaerobic sulphito-reducers (spores) (CFU-100 mL ⁻¹)	<1	>4.1 (log*)	-	<1	<1	<1	>3.5 ± 0.3 (log*)	>2.8 ± 0.1 (log*)	-	≥ 4 (log*)	≥ 5 (log*) / Absence
Specific F-RNA bacteriophages (PFP-100 mL ⁻¹)	<1	>3.6 (log*)	-	<30	<1	<1	>2.6 ± 0.8 (log*)	-	-	≥ 4 (log*)	≥ 6 (log*) / Absence
COD (mgO ₂ -L ⁻¹)	<10	>98%	>44%	<30	<30	<30	>95.4 ± 2.0% (log*)	-	-	<60	-
BOD ₅ (mgO ₂ -L ⁻¹)	-	-	-	<3	<3	<3	>98.1 ± 0.3% (log*)	-	-	-	≤ 10
TSS (mg-L ⁻¹)	<2	>97%	>56%	-	<2	<2	>98.5 ± 0.8% (log*)	>84 ± 6%	≤ 30	<15	≤ 10
TOC (mgC-L ⁻¹)	4.61 ± 0.61	94 ± 3%	70 ± 11%	-	-	-	-	-	-	-	-
Turbidity (NTU)	0.42 ± 0.35	-	85 ± 7%	<0.1	<0.1	0.31	>99.9 ± 0.0% (log*)	>86 ± 11%	≤ 2	-	≤ 5
pH	7.46 ± 0.07	-	-	7.6	7.4	7.5	-	-	-	-	-
Conductivity(μS-cm ⁻¹)	1056 ± 70	-	-	-	-	-	-	-	-	-	-

CFU: colony-forming unit; PFP: Polyhedral, filamentous, and pleomorphic; log*: log removal calculated from the raw wastewater quality; - : not measured.

(A)*: There are ABCD four different levels of water quality in French water reuse standards and EU regulation. Level A being the best.

RRs*: Removal rates.

methods. Besides, the COD, BOD₅, TSS concentrations and turbidity of permeate all fulfill the three standards. However, because of the detection limit, the log removals of the spores of anaerobic sulphito-reducers and virus (Specific F-RNA bacteriophages) cannot fully reach to 4. In fact, the concentrations of the spores and virus in permeate were very low and under the detection limits which can be considered

absence. In addition of the parameters in Table 4, the parameters of *Salmonella*, *Legionella spp.*, and viable pathogenic helminth eggs, were tested during Campaign 2 as mentioned in the EU regulation, and they were not found in any of the permeates. Therefore, regardless of the filtration conditions and the variation of feed water quality during campaign 1, UF as tertiary treatment can provide high-quality water for

reuse as it reaches the WHO guidelines for reuse and the French reuse standards with quality “A”. According to the results of campaign 2, UF permeate can also reach the EU regulation for reuse with quality “A”.

Because of the international Covid-19 pandemic, coronavirus SARS-CoV-2 was measured in the raw wastewater, UF feed, and UF permeate in November 2020. In results, the raw wastewater was tested positive with the envelope protein gene E above the detection limit (17 000 genome units per liter) but below the quantification limit (170 000 genome units per liter) and the nucleoprotein gene N was not detected. The UF feed and UF permeate were tested negative to SARS-CoV-2. While the limit of detection of the PCR method is quite high, it is uncertain to say that the virus is completely absent in the feed and permeate, or not. According to recent publications in this field, the findings indicate that secondary wastewater treatment can largely remove SARS-CoV-2 from wastewater, and tertiary treatment or disinfection can enhance the inactivation of SARS-CoV-2 in WWTPs [36,37]. Bogler *et al.* [38] stated briefly in their review that SARS-CoV-2 is expected to be removed reliably by UF. Based on the above analysis and detection, the UF permeate after UF treatment in this test would have no infectivity of SARS-CoV-2 and could be reused safely. Besides, in order to fully assess UF as a barrier to SARS-CoV-2, lower detection limits in PCR should be sought or new analytical tools should be developed to quantify infectious SARS-CoV-2 for example.

Recently, microplastic contamination have attracted worldwide attention as a new emerging pollutant, especially because of its ecotoxicological effects in marine environments and accumulation in bodies through food chain transmission [39]. Therefore, it is necessary to investigate the microplastics removal by UF in this study. Fig. 4(a) shows the types and quantity of polymers in feed water which includes 45.8% Polyethylene (PE), 45.8% Polypropylene (PP), 4.8% Polyethylene terephthalate (PET), 2.4% Polystyrene (PS), and 1.2% Polyamide (PA). Fig. 4(b) shows the distribution of microplastics in feed and permeate. Fig. 5 shows the capacity of UF on microplastics removal at different size of the microplastic particles (MPs) surface morphologies. The result showed the excellent removal capacity of UF membrane on MPs with side dimension > 70 μm , which is > 95%. However, the MP retention rates by UF will decrease as the particle size becomes smaller. When MPs' side dimension is $\leq 40 \mu\text{m}$, the retention rate is <17%. In macro views, there are about 207.5 $\text{MPs}\cdot\text{L}^{-1}$ in the feed and 17.5 $\text{MPs}\cdot\text{L}^{-1}$ in the permeate, resulting in 91.6% removal of MPs. Therefore, the microplastics in feed water can be largely removed by UF membrane especially on particles with side dimension > 70 μm . Nevertheless, despite the apparent larger size of the microplastic particles compared to the UF pore size (0.02 μm), the retention is not complete especially for the smallest particles and this suggests a need of further research in this field to understand why.

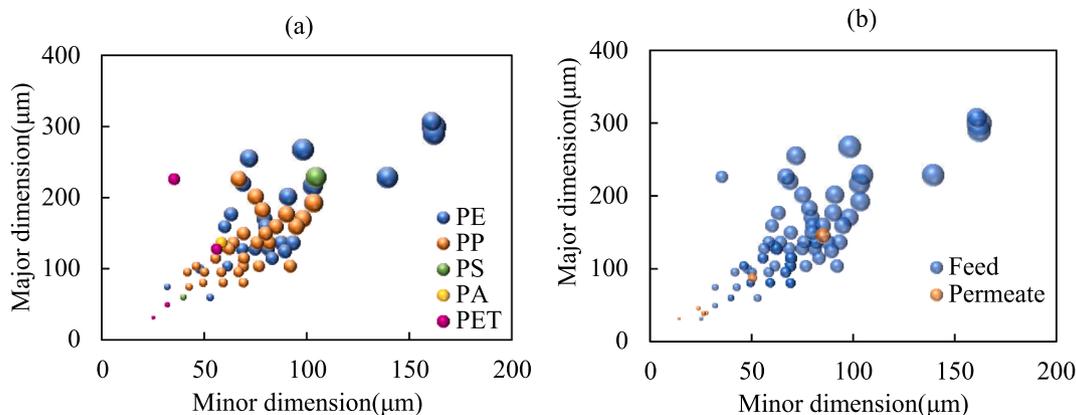


Fig. 4. (a) The polymer types in feed water and their size distribution. (b) The quantity and size distribution of microplastics in feed and permeate. The size of the data circles in both (a) and (b) represents the relative surface area of the microplastic particles.

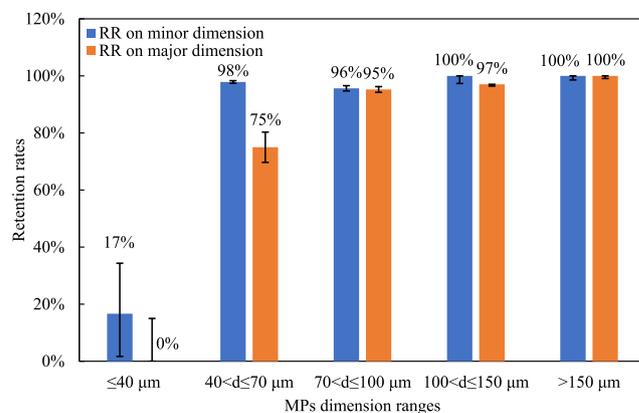


Fig. 5. The retention rates of microplastics by UF membrane at different size ranges of particles based on major dimension/minor dimension. The detection limit value of dimensions is 6.25 μm for the permeate and 25 μm for the feed.

4.1. Impact of filtration time and flux on hydraulic performances

Making the assumption that the feed water quality variation is cyclical for one operating condition, permeate flux and filtration time are the key operating parameters [40]. According to Table 3, different filtration cycle time (t) and feed water flux (J) were studied to analyze their impact on membrane hydraulic performance. Filtration performance was evaluated through backwash effectiveness, CEB frequency and fouling resistances. To evaluate the influence of flux and filtration cycle time, conditions with gradual increase of filtration cycle time (t) at same constant flux, and with gradual increase of filtration flux (J) at constant filtration cycle time (t) of 30 min (or 60 min) were designed and operated.

The average reversibility of CB and AB obtained under different conditions is shown in Fig. 6, which is indicative of the overall backwash effectiveness. From the figure, reversibility of AB fluctuates from 110% to 180% in all conditions which was conspicuously higher than reversibility of CB (between 70% and 90%). It is therefore very interesting to use AB in long-term fouling management.

A classification of all conditions is proposed in Table 5 based on the fouling reversibility, the total filtrated volume during a cycle ($V_{f,cyc}$), the initial TMP of a cycle and the number of CEBs in 40 h operation. From Fig. 6, J30t30BW1/3 and J60t20BW1/3 were performed in higher AB removal capacity than the others with reversibility values of 150% and 180%, respectively. These conditions with low flux or low filtration time had the smallest ultrafiltrated volume and initial TMP as well as no CEB needed during 40 h operation. Consequently, these conditions are referred as soft conditions. Some other conditions, such as J80t60BW1/

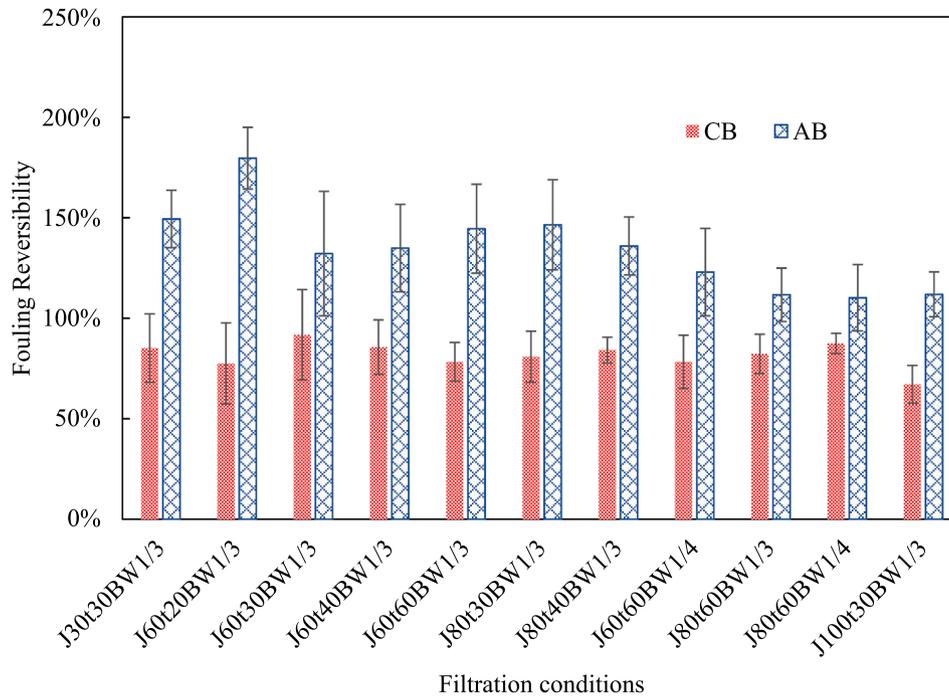


Fig. 6. Fouling reversibility by CB and AB in different conditions.

Table 5

Classification of the different operating conditions considering the total filtrated volume during a cycle, the initial TMP and the number of CEB in 40 h-operation.

Conditions	V_f , cyc (L)	Initial TMP* (bar)	CEB times in 40 h operation	Types
J30t30BW1/3	135	0.05	0	Soft conditions
J60t20BW1/3	180	0.1	0	
J60t30BW1/3	270	0.1	0	Standard conditions
J60t40BW1/3	360	0.1	0	
J60t60BW1/3	540	0.1	0	
J60t60BW1/4	540	0.1	0	Harsh conditions
J80t30BW1/3	360	0.133	0	
J80t40BW1/3	480	0.133	0	
J80t60BW1/3	720	0.133	6	
J80t60BW1/4	720	0.133	5	
J100t30BW1/3	450	0.167	10	
3				

Initial TMP*: calculated when I_p is $600 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$ at 20°C .

3, J80t60BW1/4, and J100t30BW1/3, were performed in relatively weaker removal ability of CB and AB. These conditions with high flux or high filtration times need at least 5 CEBs during 40 h operation. Thus, these conditions are referred as harsh conditions. Despite the soft and harsh conditions, the rest is considered as standard conditions with CB reversibility ranging between 80% and 90%, and AB reversibility ranging between 120% and 145%. The harsh conditions of J80t60BW1/3, J80t60BW1/4 could filtrate the highest volume (720 L) during one cycle which also means the large quantity of fouling load would be retained by membranes. The highly increased fouling resistance thereby reduced the fouling reversibility of CB and AB. This phenomenon can be explained by the higher filtration volume over a filtration cycle (V_{s-cyc}) as a combined result of flux and time, which means more solids, particles and organic matters retained by membrane thus increasing both irreversible and reversible fouling. For J100t30BW1/3 condition, even if the total filtrated volume in a cycle V_{f-cyc} (450 L) was lower than in standard conditions (such as J60t60BW1/3 and J80t40BW1/3), the reversibility of both CB and AB were still lower, and with more frequent CEBs. According to Darcy's law, the increase of flux ($100 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) under

constant filtration cycle time can directly increase TMP (0.167 bar) from the beginning of the filtration process. The increase of TMP may compress the fouling layer (i.e. cake density) which will promote the smallest particles penetration into membrane pores thus enhancing irreversible fouling [40]. In this case, although backwashes could flow away the cake foulant on membrane surface significantly, they could not remove the smallest compounds adsorbed onto the membrane material effectively, thus decreasing CB and AB reversibility [20]. As the irreversible fouling is mainly caused by increased TMP, the irreversible fouling was more sensitive to flux increase than filtration cycle time extension in these tests: this is in agreement with [15].

In order to confirm the above results, the variations and composition of fouling resistances before backwash versus time are proposed for each condition (Fig. 7). J60t20BW1/3, J60t60BW1/3, and J100t30BW1/3 are taken as representatives of the 3 types of conditions.

- In soft condition J60t20BW1/3, membrane resistance (R_m) was in dominant position during whole filtration and R_{irr} even not reached to one-third of R_m .
- In standard condition J60t60BW1/3 with 40 min filtration time longer than J60t20BW1/3 in a cycle, the irreversible fouling R_{irr} gradually reached the value of R_m after 35 h's operation and stayed in the same order of magnitude than R_m .
- In harsh condition J100t30BW1/3 with highest flux among all tested conditions, the irreversible fouling R_{irr} increased fastest from beginning and exceeded the value of R_m . Higher values of reversible resistances than in soft or standard conditions are also to be noted.

Through comparison, increase of flux and filtration cycle time both contribute to the increasing rate of R_{irr} during filtration process. The increase of filtration flux (comparison between J60t60 and J100t30) seems to have a higher impact on R_{irr} increasing rate than the increase of filtration time (comparison between J60t20 and J60t60).

From above, the harsh conditions all resulted in occurrence of CEB during 40 h's operation, especially in condition J100t30BW1/3 with the most frequent CEB needed. In practical uses, the CEB interval usually ranges from hours to weeks among different UF processes [42]. Because one CEB consumes more chemical agents, permeate water, and energy

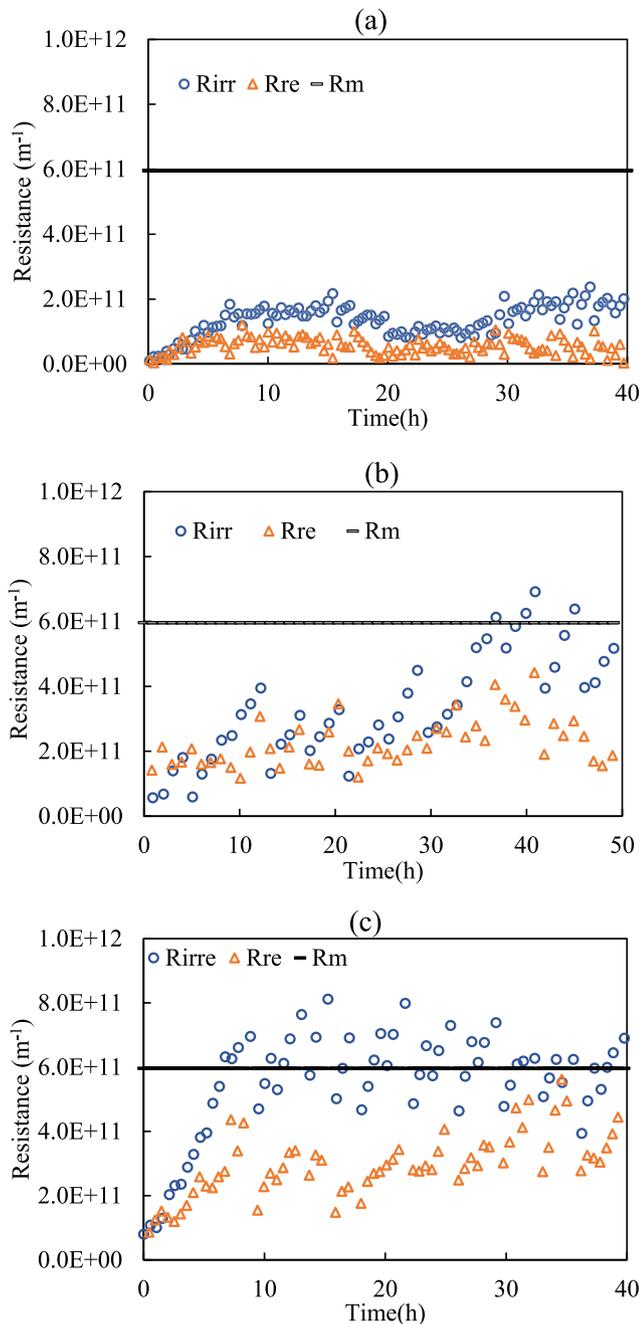


Fig. 7. Resistances' variations versus time at conditions: (a) J60t20BW1/3, (b) J60t60BW1/3 and (c) J100t30BW1/3.

compared with an AB or a CB, it is necessary to prolong the CEB interval during filtration. It has been previously estimated that sustainable operating conditions lead to less than 2 CEB occurrences per day [41]. Therefore, the harsh conditions cannot be considered as the optimized conditions for long term operation.

Furthermore, the permeate water was produced for reuse, but it was also used for cleanings. The permeate consumption in CB, AB, and CEB conversely decreased the productivity of the system. Therefore, it is necessary to investigate the water recovery rate in different conditions to make a comprehensive comparison, shown in Fig. 8 with conditions of AB frequency of 1/3. Through calculation, the highest water recovery rates occurred in the standard conditions, especially in conditions of J60t60BW1/3 and J80t40BW1/3 which resulted in 92.7% and 94.5% water recovery, respectively. In addition, the soft conditions showed the lowest water recovery rates between 70.7% and 72.2% because of

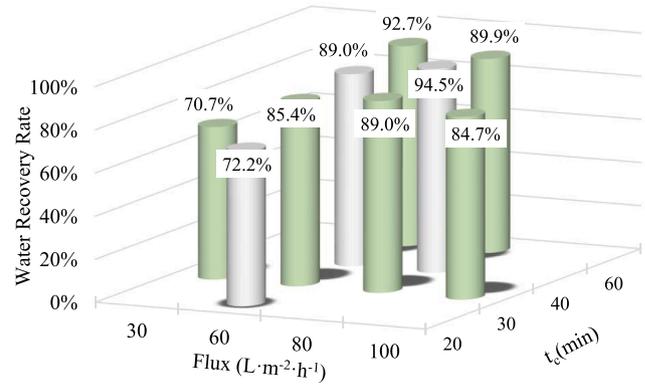


Fig. 8. Water recovery rates: conditions at backwash frequency of 1/3. *The uncertainty of each column is between 1 and 2%.

frequent AB and CB, and low flux. The water recovery rates of harsh conditions J100t30BW1/3 and J80t60BW1/3, were at an intermediate level, ranging from 84.7% to 89.9%. Except the conditions in backwash frequency of 1/3, the conditions J60t60BW1/4 and J80t60BW1/4, resulted in water recovery rates of 92.8% and 90.8% respectively. The condition J60t60BW1/4 showed almost the same water recovery rate with J60t60BW1/3, with only 0.1% difference because of different permeate consumption between AB and CB. Therefore, the higher water recovery rates among these conditions were obtained from the conditions of J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3 with potential to be the optimized operating conditions in this study.

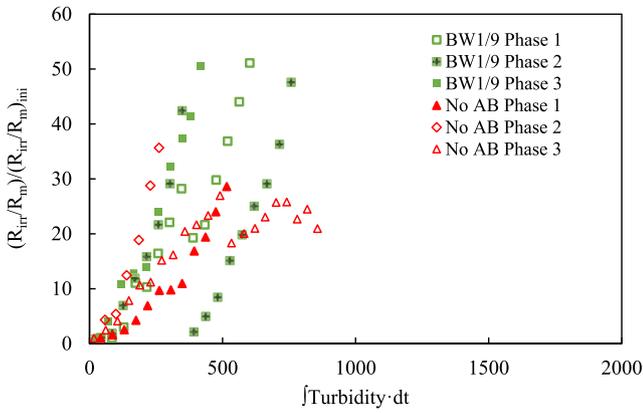
4.2. Impact of air backwash frequency

To evaluate the impact of AB frequency on filtration performance and confirm that the frequencies 1/3 and 1/4 previously studied were the best, 5 different backwash frequencies (1/3, 1/4, 1/6, 1/9, and No AB) (Table 3) were tested, all under the constant flux and filtration cycle time ($J = 60 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and $t = 60 \text{ min}$). The variation of $(R_{irr}/R_m)_n / (R_{irr}/R_m)_{ini}$ versus turbidity integration based on Eq. (10) reflecting the R_{irr} increasing rate was calculated and is shown in Fig. 9. The turbidity integration enables to consider feed water quality variation over time considering turbidity as a main indicator of feed quality. It should be noted that the filtration process between 2 CEBs was considered as a new and independent filtration phase to be calculated. Therefore, turbidity integration will be re-calculated from 0, and corresponding to the re-calculated $(R_{irr}/R_m)_{ini}$. This explains why some conditions present several phases.

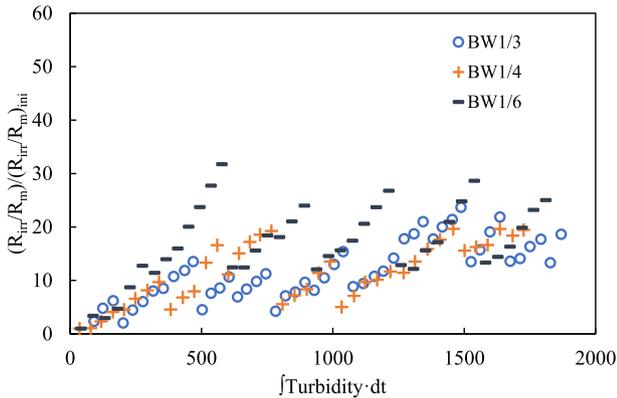
In Fig. 9(a) and (b), two main variation can be described. On one hand, Fig. 9(a) with AB frequency of 1/9 and no AB presents very high $(R_{irr}/R_m)_n / (R_{irr}/R_m)_{initial}$ increase with turbidity integration and 2 CEBs occurring during 2 days of filtration. On the other hand, Fig. 9(b) with AB frequencies of 1/3, 1/4 and 1/6 where $(R_{irr}/R_m)_n / (R_{irr}/R_m)_{initial}$ slowly increased with turbidity integration. AB frequency in conditions of BW1/3, BW1/4 are the best with excellent R_{irr} removal abilities, which can maintain membrane permeability over $200 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$ in 2 days of filtration. No CEB occurred in these conditions. Therefore, optimum conditions BW1/3 and BW1/4 used in 3.3 are confirmed as they are more suitable for long term operation at industrial scale. This result is similar to the result on seawater [17], who concluded that AB was more effective to control fouling resistance in conditions of BW1/3 and BW1/5, than that in conditions with lower AB frequency BW1/7 and BW1/9, all under the same $J = 60 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and $t = 60 \text{ min}$.

4.3. Hydraulic filtration performance in long-term operation in optimized conditions

From the above sections, the three best operating conditions on short



(a)



(b)

Fig. 9. (a) Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs of 1/9, and No AB; (b) Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs of 1/3, 1/4, 1/6; all at flux = $60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and $t = 60 \text{ min}$.

term experiments are J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3. To verify the feasibility and sustainability of the optimized conditions, long term tests were conducted on condition J60t60BW1/3 both in winter (from 26/2/2020 to 18/3/2020) and summer (from 1/7/2020 to 27/7/2020). Fig. 10 and Fig. 11 show the permeability variation for more than 20 days, respectively, in winter and in summer.

From Fig. 10 and Fig. 11, the UF pilot plant showed stable and continuous filtration performance, and great permeability recoveries (by CB, AB, and CEBs) during long-term operation, both in summer and winter. The occurrence of CEB were all less than once per day which meets sustainable objectives. However, the frequency of CEB is unstable, which is found to be related to the feed water quality and temperature changes. In summer, the permeability drops seem faster than in winter, but it also seems that permeability recoveries by CEB are higher than in winter. Faster permeability drops in summer can be related to more algae observed in the effluent of WWTP resulting in more suspended solids and organic matters. Therefore, the fouling potential on membrane in summer was relatively higher than in winter but at the same time the mean water temperature in summer was $27.7 \pm 1.3 \text{ }^\circ\text{C}$, while in winter it was $20.2 \pm 3 \text{ }^\circ\text{C}$ which could lead to better reversibility of cleanings including AB, CB, and CEB [43–45]. Indeed, the mean reversibility of AB in summer and in winter are respectively $144 \pm 46\%$ and $135 \pm 21\%$. Besides, the mean permeability recoveries by CEB in summer and in winter are respectively $308 \pm 15\%$ and $246 \pm 30\%$. However, the water temperature increase had little effects on CB performance: the mean reversibility of CB in summer and winter are all around 80%. Although there was higher fouling potential on membrane in summer, the corresponding temperature increase improved cleaning efficiency of AB and CEB, finally resulting in sustainable permeability recovery and reasonable CEB frequency.

5. Conclusion

In this study, ultrafiltration is considered as tertiary treatment for municipal wastewater reuse in semi-industrial operation and for the first time some conclusions can be given:

- (i) The UF permeate quality was detected to be good enough to be reused in non-potable applications as it met reuse guidelines of

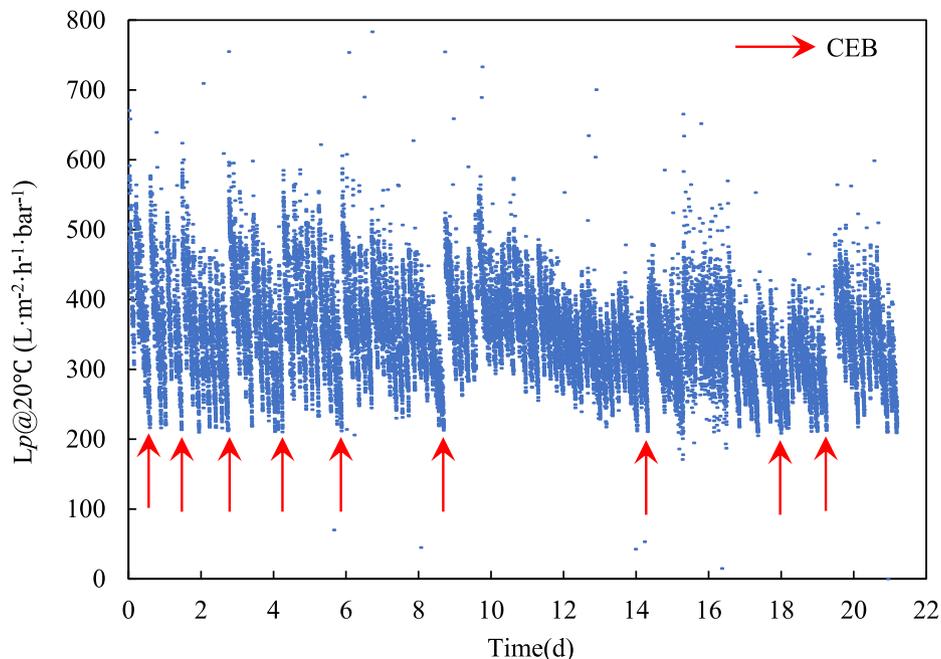


Fig. 10. Permeability variation in winter from 26/2/2020 to 18/3/2020.

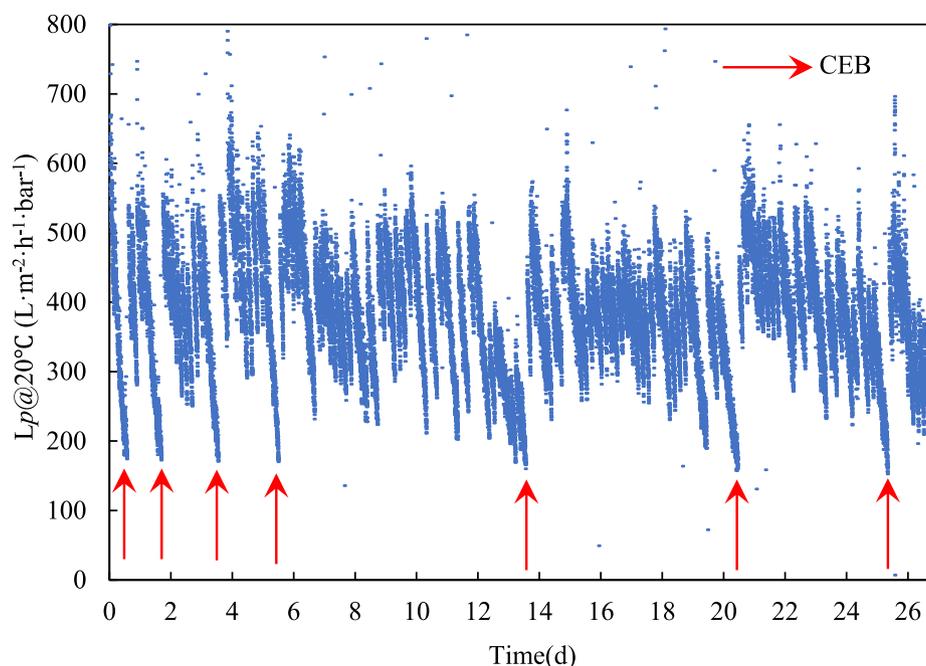


Fig. 11. Permeability variation in Summer from 1/7/2020 to 27/7/2020.

the WHO, reuse standards of France and the most recent EU regulation for agricultural irrigation. In addition, the UF membrane was confirmed to be effective on most microplastic particles' removal.

- (ii) Concerning the most suitable operating conditions for UF: soft conditions with lower flux or frequent physical cleaning (short filtration time) resulted in too little productivity and were discarded. Harsh conditions with higher flux or longer filtration time were also discarded because of high occurrence of CEB which led to difficult permeability stabilization and high consumption of permeate and chemicals. As the permeate quality was not affected by filtration conditions, the performance comparison was based on AB and CB reversibility, irreversible fouling resistance increase, occurrence of CEBs and water recovery rates. Finally, standard conditions J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3 stood out from the others with higher overall performances. Of course, these conditions are function of the WWTP and feed water quality, but still, this study defined some ranges of operating conditions for future water managers and engineers for municipal wastewater tertiary treatment before reuse.
- (iii) By analyzing the influence of AB frequency on irreversible fouling management, the irreversible fouling resistance increased faster with the decrease of AB frequency during operation. Optimum AB frequencies of BW1/3 and BW1/4 under constant flux of 60 $L \cdot m^{-2} \cdot h^{-1}$ and t of 60 min found in (ii) were confirmed.
- (iv) Concerning the selected sustainable conditions, more than 20 days of operation in condition J60t60BW1/3 in winter and in summer confirmed that the UF pilot plant could provide sustainable and adaptable filtration performance regardless of the temperature and feed water quality. This works confirms the great interest of UF as tertiary treatment of conventional WWTP for water reuse.

CRedit authorship contribution statement

J. Yang: Investigation. **M. Monnot:** Supervision. **T. Eljaddi:** Investigation. **L. Ercolei:** Resources. **L. Simonian:** Investigation. **P. Moulin:** Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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