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Ultrafiltration for environmental safety in shellfish production: A case of bloom emergence

Clémence Cordier ^a, Alexandra Voulgaris ^b, Christophe Stavrakakis ^b, Patrick Sauvade ^c, Franz Coelho ^c,
Philippe Moulin ^{a,*}

^a Aix-Marseille Université, M2P2-UMR 7340, Équipe Procédés Membranaires, Laboratoire de Mécanique Modélisation et Procédés Propres,
Aix en Provence 13545, France

^b Plateforme expérimentale Mollusques Marins, Station Ifremer de Bouin, Polder des Champs, Bouin 85230, France

^c Suez-Aquasource, Toulouse 31029, France

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Abstract

The process of ultrafiltration (UF) of natural seawater often encounters the problems of variation in water quality and coastal blooms. To validate the feasibility of UF in shellfish farms, this study compared the hydraulic performance and pollutant removal efficiency of the UF process with those of the commonly used treatments that combine several filtration steps with UV disinfection. The comparison was conducted in the cases of natural seawater and a coastal bloom. Given that the UF process encountered this specific pollution, this study evaluated the filtration performance of the UF process and the retention of total suspended solids (TSS), bacteria, phytoplankton, and zooplankton. A real coastal bloom was considered in the case study of an experimental shellfish hatchery/nursery in France. The results show that both treatments were able to eliminate approximately 50% of TSS. However, in contrast with UV treatment combined with filtration, the UF process retained total amounts of phytoplankton, zooplankton, and bacteria in the bloom. Although the hydraulic performance of the UF process was impacted by the coastal bloom, the fouling was eliminated through chemical cleaning conducted at a frequency less than once per 12 h. Despite the severe pollution, this study confirmed the pollution resistance and treatment performance of the UF process, indicating that UF has the potential to enhance the biosecurity level.

Keywords: Ultrafiltration; Environmental safety; Aquaculture; Shellfish culture; Coastal bloom

1. Introduction

Ultrafiltration (UF) is widely used for seawater purification. It is commonly applied in pre-treatment of desalination by reverse osmosis membranes, and it is able to produce water with constant quality regardless of the turbidity of the feed water (Prihasto et al., 2009; Greenlee et al., 2009; Wolf et al., 2005). The production of potable water from lakes is another application of membrane processes in natural water treatment, leading to abatement of turbidity, colloids, and microorganisms (Bottino et al., 2001; Karakulski et al., 2002). UF of natural water is challenging because of the instability of water quality and composition, especially in the case of seawater treatment under the impacts of seasons, tides, temperature, and weather. Given that filtration processes encounter a diversity of fouling species, their resistance and stability over time should be proven. In addition to water quality variation, punctual and severe water quality degradation, such as uncontrolled increase of plankton concentrations caused by blooms, is a common phenomenon and causes operational problems in the production of drinking water with reverse osmosis because of the generation of irreversible fouling (Richlen et al., 2010). Moreover, blooms are potentially deleterious to aquaculture activities, including shellfish production, as microalgae produces toxins harmful to shellfish and their consumers (Brand et al., 2012). In 1995, *Cochlodinium polykrikoides* spread as a bloom in Korean coastal waters and caused an economic loss over 9.5×10^7 USD (Richlen et al., 2010). It is necessary to investigate filtration and operation conditions that can produce water with constant quality and adapt to natural water variations and acute degradations. In aquaculture, clay can be used as an algicide in water (Beaulieu et al., 2005; Sengco and Anderson, 2004). This treatment is practical in low sea currents, but not suitable near coasts. Although the treatment is efficient and inexpensive, the environmental impacts of clay are still unknown. The accumulation of these impacts on the sea floor affects other planktonic species in the water column and other organisms living in the environment. The use of natural algicides in the ecosystem to avoid environmental impacts has been studied,

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* Corresponding author.

E-mail address: philippe.moulin@univ-amu.fr (Philippe Moulin)

but their available quantities are not sufficient for microalgae removal (Anderson et al., 2001; Doucette et al., 1999; Jeong et al., 2008).

With the aim of implementing membrane processes in shellfish ponds for shell protection during harmful algal blooms, the treatment of simulated algal blooms at the laboratory scale demonstrated the efficiency of microfiltration in retaining toxic microalgae, with a retention ratio superior to 99% (Castaing et al., 2010). At the laboratory scale, Massé et al. (2015) investigated the UF fouling during the filtration of synthetic seawater composed of humic acids, alginic acids, inorganic particles, and numerous salts at high concentrations. Most of the fouling is reversible (80%), and the backwash efficiency drops during the operation, which induces fewer organics into backwash water. Membrane processes conducted at the industrial scale commonly encounter blooms, and it is necessary to conduct membrane cleaning to ensure the resistance capacity. Schurer et al. (2012) conducted a study at a seawater desalination plant with UF pre-treatment using hollow fiber membranes for dead-end filtration and demonstrated that turbidity peaks in the range of 50–500 FTU had no impact on membrane performance. However, in the case of algal blooms appearing in spring, the fouling rate was severe, and in such circumstances coagulation as a pre-treatment measure should be employed. Coagulation before UF and membrane cleaning in place after episodes of blooms were necessary to ensure the stability of the process (Schurer et al., 2013, 2017). Instead of employing pre-treatment before UF, Pearce et al. (2004) conducted a study on the UF pre-treatment of seawater and highlighted the efficiency of air-enhanced backwashes. They concluded that algal blooms led to an increase of the silt density index (SDI) after UF from 2 to 4, and increasing the frequency of this cleaning procedure avoided the SDI increase during algal blooms. Indeed, the membrane fouling caused by algal blooms is particularly problematic because compounds including polysaccharides, proteins, humic substances, and other molecules exacerbate membrane fouling. In particular, the generated shear stress on membranes causes the release of algogenic organic matters (Ladner et al., 2010; Villacorte et al., 2015). Given that deterioration is expected to escalate with increased bloom frequency, it is essential to consider the impact of blooms (Delpla et al., 2009; UN-Water, 2018).

The UF process is efficient in providing water in shellfish hatcheries and nurseries (Cordier et al., 2019a, 2019b, 2020a, 2020b). In this study, the impact of a coastal bloom on the performance of UF was studied. Given that the UF process encountered this specific type of pollution, the impact was evaluated based on the filtration performance of the UF process and the retention of total suspended solids (TSS), bacteria, phytoplankton, and zooplankton. The reasons for choosing UF in this study are as follows: (1) UF has a higher removal efficiency of pathogens, especially viruses, than microfiltration (Bazargan, 2018); (2) UF is less impacted by fouling generated by silt particles suspended near water intakes (Voutchkov, 2010); and (3) sustainable conditions are obtained through seawater filtration (Guilbaud et al., 2018, 2019). The objective of this study was to evaluate the resistance capacity of the UF process during a bloom composed of phytoplankton and zooplankton. To achieve this objective, the performance of the UF process was evaluated in terms of the hydraulic efficiency, TSS, and bacteria retention, and compared to those of a classical water treatment approach and an optimized water treatment method (ultraviolet (UV) disinfection preceded by filtration steps) for shellfish culture.

2. Materials and methods

2.1. Water treatment equipment

2.1.1. UF process

The UF membranes used in the tests were Aquasource polyether sulfone hollow fibres, including seven channels with a 0.9-mm inside diameter. Their molecular weight cut-off (MWCO) was 200 kDa, and the initial seawater permeability was 800 L/(h·m²·bar). A membrane module with a total area of 8 m² was able to treat 20 m³ seawater per day (Cordier et al., 2018, 2019a, 2019b, 2020b). All the tests were performed for dead-end filtration, the seawater to be treated was continuously introduced in the pilot feed tank, and the treated water, called permeate, was recovered in a buffer tank to perform backwashes (Fig. 1) (Moll et al., 2007). The experiments lasted for more than two months. Considering that water temperature influenced the membrane flux, the permeability estimated at 20°C.

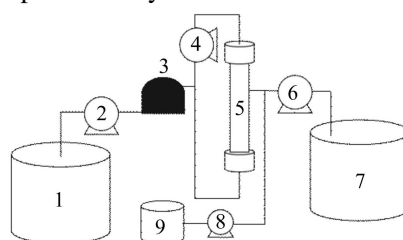


Fig. 1. Schematic diagram of UF unit (1–Feed tank; 2–Feed pump; 3–Pre-filter (130 μm); 4–Recirculating pump; 5–Membrane module; 6–Backwashing pump; 7–Purified water tank for backwash; 8–Chlorination pump; 9–Tank).

Three membrane cleanings were automatically carried out by the UF unit to eliminate fouling: classical backwash (CB), air backwash (AB), and chemically enhanced backwash (CEB). During a CB, an injection of permeate at a flow rate of 2.5 m^3/h was pumped from the permeate tank to the membrane with reverse flow. During an AB, water was discharged from the membrane module, and hollow fibres were filled with air until the pressure reached approximately 300 mbar. This was followed by a backwashing operation. An AB was performed every three or five CBs, and the frequency of backwash was determined by the duration of filtration cycles between 20 and 100 min (Cordier et al., 2018). With a limited permeability fixed at 300 $\text{L}/(\text{h}\cdot\text{m}^2\cdot\text{bar})$, a CEB was carried out. First, the permeate with additions of sodium hydroxide (NaOH, 400 mg/L and $\text{pH} = 10$) and sodium chlorine (NaCl, 150 mg/L) was injected into the membrane, and an AB was performed after 30 min contact. Then, a mixture of permeate and sulfuric acid (H_2SO_4 , 800 mg/L and $\text{pH} = 2$) was injected into the module for 30 min, and a final AB was performed at the end of the CEB. Chemicals were injected using the pilot-controlled dosing pumps. 30% NaOH, 13% NaCl, and 37.5% H_2SO_4 used for CEB of membranes were purchased from Quaron (Rennes, France). To evaluate the hydraulic performance, membrane permeability (L_p) and transmembrane pressure (TMP) were calculated and continuously recorded at one-minute intervals, with consideration of the temperature variation in analysis. Filtration conditions such as the permeate flux (J), filtration time (t_f), and ratio of frequencies of AB to CB (r) were selected according to Guilbaud et al. (2018, 2019) and Cordier et al. (2018, 2019a, 2019b, 2020a, 2020b), with $J = 60$ $\text{L}/(\text{h}\cdot\text{m}^2)$, $t_f = 60$ min, and $r = 1/5$.

2.1.2. Treatments of seawater

In the case of UF, seawater from Bourgneuf Bay (France) was decanted in a settling pond, and particles with a size greater than 20–30 μm were prefiltered with a sand filter. The classical treatment to provide water to shellfish hatcheries is sand filtration, followed by a 5- μm filtration and UV disinfection. In the case at the Ifremer experimental station (Bouin, France), before water was supplied for oyster cultures, an optimized treatment was performed first with a 5- μm zeolite filtration and UV disinfection. When water was used for oyster breeding in the laboratory, a second UV treatment and two filtrations at sizes of 5 and 1 μm were performed for further purification. Moreover, in the case of a bloom, another 1- μm filtration was performed. Each UV treatment was carried out with a Bio UV device, which can deliver a dose of 35 mJ/cm^2 with a flow rate of 6 m^3/h . It is important to note that this succession of treatments used for experimental breeding was more severe than the one implemented in professional hatcheries and nurseries. Indeed, three treatment steps, prefiltration, 5- and 1- μm filtrations, and UV disinfection, are more likely to be found in real conditions (Wallace et al., 2008).

2.2. Analyses

The concentration of TSS was measured by filtering samples through a glass fiber paper (fisher brand), which retained particles with diameters over 0.7 μm . The filter was rinsed and dried, and the concentration of TSS was obtained by calculating the difference between weights before and after filtration. Turbidity was measured and recorded every minute in the feed tank of the UF unit using the probe VisoTurb 700 IQ (WTW). In shellfish culture, some *Vibrio* species are pathogens of oysters, which should be controlled in water-feeding animals. Thus, the total bacterial load and *Vibrio* in water samples were analyzed. Microbiologic analyses were conducted twice a week in the feed tank and in permeate at the beginning (initial permeate) and end (final permeate) of a filtration cycle when the pathogen concentration inside the membrane reached its maximum value. These analyses aimed to validate the absence of *Vibrio* and to estimate the total bacterial retention by the UF membrane. *Vibrio* analyses were carried out in a thiosulfate-citrate-bile salts-sucrose (TCBS) agar medium and total bacterial load in a marine agar medium. Each sample of 50 μL was deposited in Petri dishes and then incubated at 20 $^\circ\text{C}$ for 48 h. Samples were partitioned in triplicate for suspended solid, *Vibrio*, and turbidity analyses, and at least three microscopic observations for each sample were conducted.

3. Results and discussion

3.1. Treatment efficiency

3.1.1. TSS measurement

To characterize the bloom and analyze its impact on water treatment processes, TSS concentrations in different samples were measured after different steps of water treatment. To investigate the natural effects of blooms, one bloom on the first and second days was studied. The results obtained during the bloom and in normal conditions are presented in Figs.

2 and 3. The TSS concentration in the UF pilot feed (109 mg/L) was higher than that before classical treatment (80 mg/L). This result can be explained by the accumulation of TSS in the pilot feed tank where settlement occurred. Despite this, the comparison of bloom treatment efficiency indicated a strong performance of the UF process, and the treatment was able to effectively eliminate more than 60% of the inlet TSS. This result was confirmed by the filter observations (Fig. 2). Moreover, it was found that, regarding the classical treatment feed with a lower TSS concentration, the succession of treatments (Fig. 2), including a 5- μm prefiltration, two UV disinfections, a 5- μm filtration, and two 1- μm filtrations, led to a lower TSS removal rate (only 44%) than the UF process alone. Importantly, the average values of turbidity of the permeate were, respectively, lower than 0.25 NTU and 0.65 NTU during the bloom and in normal conditions, while the values in the feed tank were higher than 9 NTU and between 2 and 3 NTU, respectively. However, comparison of the UF performance during the bloom and in normal conditions revealed the impact of water degradation on the UF performance. The TSS concentration in the permeate during the bloom was approximately twice as high as that in normal conditions. However, the permeate quality in terms of turbidity was not affected, with a turbidity lower than 1 NTU. The TSS elimination was accomplished through backwashes, and the use of air backwashes led to improved TSS removal. When the process encountered the bloom, classical backwashes effectively eliminated TSS, with the TSS concentration in backwash water being approximately twice as high as that in the pilot feed tank. However, air injection did not lead to better TSS removal. The bloom was mainly composed of organic materials, phytoplankton, and zooplankton. Membrane draining and a two-phase flow occurred when the membrane was air-backwashed, which had the potential to destroy the integrity of microorganisms and partition biological materials into particles smaller than the pore sizes of filters used for TSS measurement (0.7 μm). These findings on the cleaning efficiency of air backwashes in seawater filtration and its impact on organic materials were in agreement with the results obtained in the treatment of oyster gametes (Cordier et al., 2019a, 2019b).

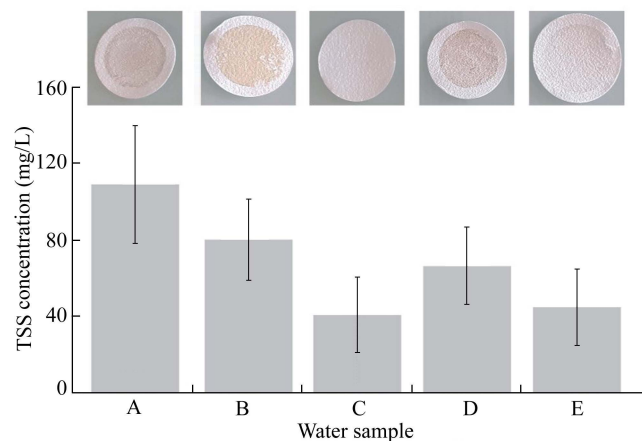


Fig. 2. TSS concentrations for different steps of water treatment and pictures of filters during blooms (A means UF pilot feed; B means classical treatment feed; C means permeate; D means a 5- μm prefiltration and UV disinfection; and E means a 5- μm prefiltration, two UV disinfections, a 5- μm filtration, and two 1- μm filtrations).

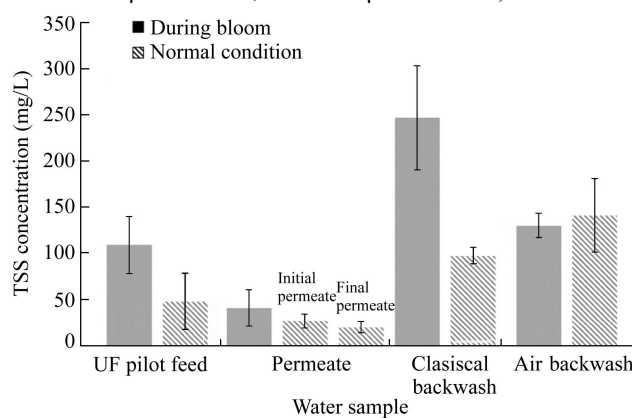


Fig. 3. TSS concentrations in different water samples during bloom and in normal conditions.

According to TSS measurements, particles composing the bloom can be categorized according to their sizes: 36% of the particles had diameters less than 0.02 μm , 21% of the particles had diameters between 0.02 and 1 μm , 26% of the particles had diameters between 1 and 5 μm , and 17% of the particles had diameters larger than 5 μm . Therefore, more than 40% of TSS had particle sizes exceeding 1 μm . It should be noted that the value of 1 μm was selected based on two

successive 1- μm filtrations, with an additional 1- μm filtration conducted during the bloom. Particles with sizes smaller than 1 μm were not retained by filtration before UV disinfection. Their presence at a high concentration might impact water transmittance, thereby leading to a weak UV performance in terms of the inactivation of microorganisms (Gullian et al., 2012).

3.1.2. Microscopic observations

To characterize the composition of the bloom and compare the efficiency of treatment processes, samples from untreated and treated seawater and backwash water from UF unit were observed under a microscope. An array of microorganisms was observed, from different species of microalgae to oyster larvae and sponge spicules (Fig. 4). Among these planktons, some of the species are known to be predators in shellfish culture, such as copepods and nematodes, and some of them might be harmful to human health (Leibovitz, 1978). Their presence was found in every sample of treated water after four filtrations and two UV disinfections, performed for oyster breeding. The presence of TSS decreased the efficiency of UV treatment and limited the inactivation of microorganism. For instance, UV treatment is not efficient in the case of high turbidity and low transmittance in the water. The influence of water quality on the efficiency of UV disinfection was demonstrated by comparing the results obtained from different water samples under similar experimental conditions (Huff et al., 1965; Hijnen et al., 2006).

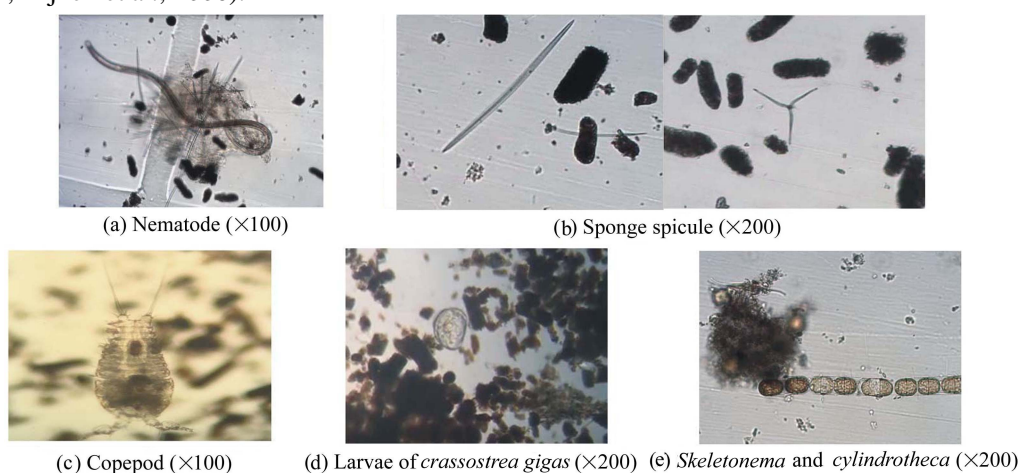


Fig. 4. Microscopic observations of samples after three filtrations and two UV disinfections.

Regarding the UF process, no microalgae or plankton were observed in different permeate samples (Fig. 5). The UF process demonstrates its effectiveness in eliminating phytoplankton and zooplankton from seawater in the case of blooms. Moreover, in contrast to UV disinfection, the treatment performance of the UF process is not influenced by the presence of TSS.

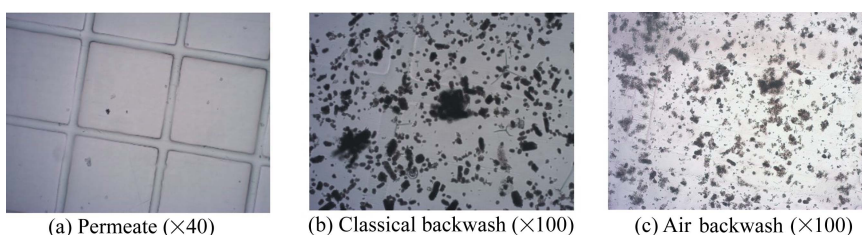


Fig. 5. Microscopic observations of permeate and backwash water.

3.1.3. Bacteria removal efficiency

Table 1 shows the concentrations of bacteria measured before and after UF and UV treatments. The retention of *Vibrio*, a bacteria genus that has a potential negative impact on aquaculture, was validated for both treatments because no *Vibrio* was detected in treated water (permeate as well as after filtration and UV disinfection). These results also highlighted a better control of total bacteria by the UF process, with the concentration of the total bacteria lower than the detection limit, corresponding to a retention rate over 99.76%. This value was calculated with a total bacteria concentration in permeate equal to the detection limit, but not equal to the measured value. Therefore, it was an underestimation of the removal rate.

The bacteria concentration after the combination of two UV disinfections and three filtrations was merely three times lower than that before treatment.

Table 1

Concentrations of total bacterial and *Vibrio* before and after UF and UV treatments.

Bacteria	Concentration (CFU/mL)			
	Settling pond (before UV)	After filtration and UV disinfection	Pilot feed (before UF)	Permeate (after UF)
Total bacteria	3 600	1 200	8 200	< detection limit
<i>Vibrio</i>		< detection limit	307	< detection limit

The treatment efficiency of the UF process was investigated based on a two-month bacteria measurement. During the measurement period, no *Vibrio* was detected in the permeate (Fig. 6(b)) with different inlet concentrations. UF effectively protected oysters by eliminating potentially harmful microorganisms. Moreover, the change in the total bacteria concentration (C_b) in this period (Fig. 6(a)) highlighted that, in most cases, the total bacteria concentration in the permeate was below the detection limit (20 CFU/mL), and the retention rate exceeded 99%. The obtained removal rates for *Vibrio* and total bacteria during the filtration process varied between 2.4log and 4.8log and between 2.6log and 5.3log, respectively. They were calculated as functions of the volumetric concentration, with the bacteria concentration in permeate equal to the detection limit (20 CFU/mL), but not equal to the measured value (0 CFU/mL). In the case of the algal bloom, the treatment efficiency of the UF process was confirmed by the absence of *Vibrio* and total bacteria in the permeate. Therefore, oyster breeding protection can be achieved even in the case of accidental water quality degradation.

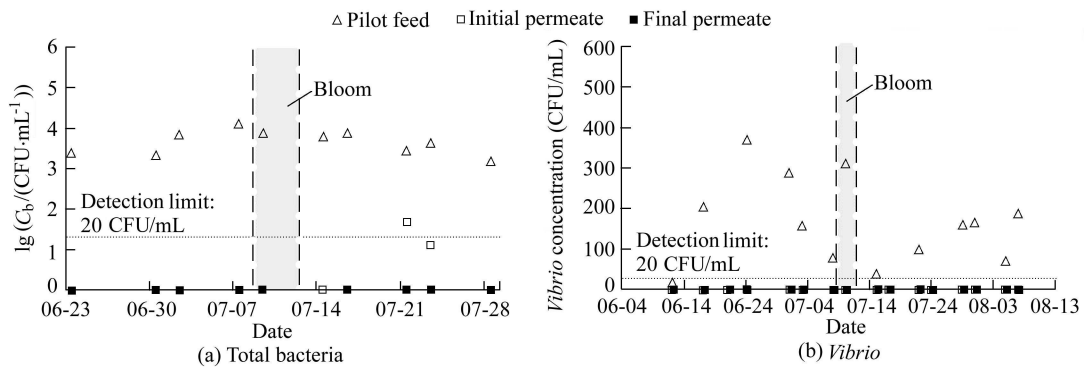


Fig. 6. Changes in total bacteria concentration and *Vibrio* concentration.

3.2. Hydraulic performance of UF treatment

This study proved the efficiency of UF membranes in removal of the bloom-generated pollution in seawater. This punctual water contamination caused high concentrations of TSS and microorganisms, and it might impact hydraulic performance of the UF process, inducing possible significant fouling. In this study, the change in permeability of UF membranes was investigated (Fig. 7). Water quality had a significant impact on membrane performance, leading to a decrease rate of permeability. However, as shown in Fig. 7, no significant permeability decrease was found during the bloom, due to the retention time of pollution in the hydraulic system before UF. The impact of the bloom was found with a one-day delay and reflected in a fast permeability drop.

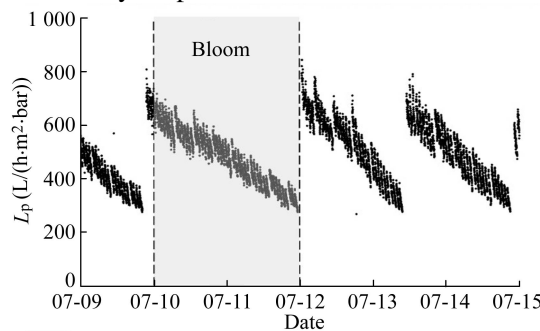


Fig. 7. Change of permeability.

To evaluate the impact of water degradation on membrane fouling, the time between two CEBs was estimated by determining the time that was required to decrease the membrane permeability from the minimum value after the first CEB (600 L/(h·m²·bar)) to the threshold before the second CEB (300 L/(h·m²·bar)). The result from Fig. 8 confirmed previous

observations. When the UF unit was fed with seawater, more than 47 h were needed for this permeability loss, and shorter time durations were obtained during and after the bloom.

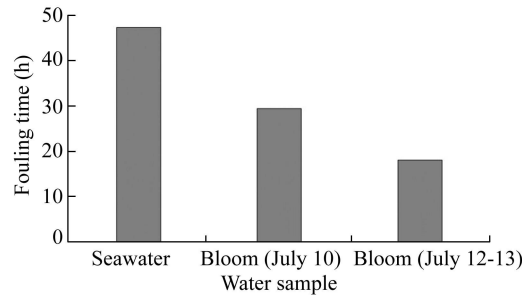


Fig. 8. Fouling time between filtration cycles for different water qualities under condition of $r = 1/5$, $J = 60 \text{ L}/(\text{h}\cdot\text{m}^2)$, and $t_f = 60 \text{ min}$

To evaluate membrane fouling, potential reversible and irreversible fouling resistances were calculated. In the case of seawater (Fig. 9(a)), the variation of irreversible resistance (R_i) with time can be categorized into two parts. In the first part, R_i versus time demonstrated a straight line, indicating a proportional fouling elimination during filtration. In contrast, the reversible resistance (R_r) was not a constant and tended to increase at a lower rate than R_i , and the resistance of the membrane (R_m) ranged from 4/6 to 4/8 of the total resistance. For the second part, R_i versus time presented an exponential relationship, indicating that fouling removal became increasingly difficult, with a remarkable increase of irreversible resistance due to the accumulation of compounds on the membrane surface after several filtration cycles. This accumulation generated a high density of cake and affected reversible resistance as well, and the ratio of the membrane resistance to the total resistance reached the lowest value (4/11) at the end of filtration for 2 500 min.

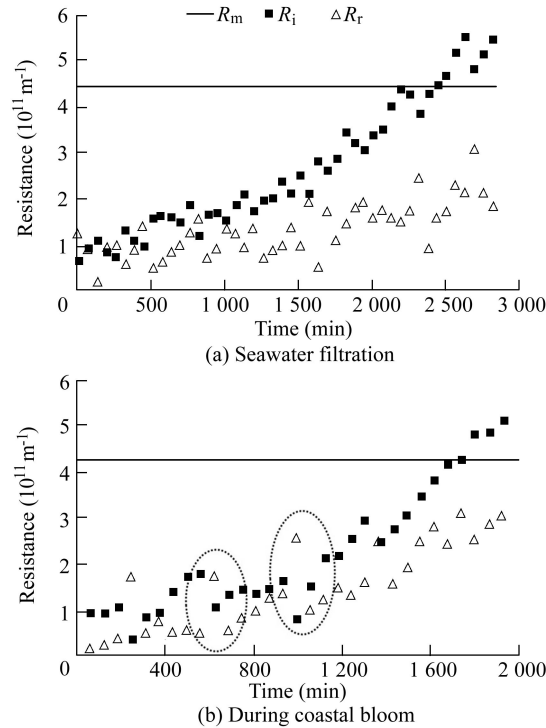


Fig. 9. Variations of irreversible, reversible, and membrane resistances versus time for seawater filtration and during coastal blooms under condition of $r = 1/5$, $J = 60 \text{ L}/(\text{h}\cdot\text{m}^2)$, and $t_f = 60 \text{ min}$.

During the bloom and as expected in dead-end filtration mode, it is important to note that the fouling is severe for large-scale desalination plants even for a backwash each hour. The time between two CEBs was reduced from 29 h to 18 h. This reveals that supplying the pilot feed tank with contaminated water including living organisms and a high amount of TSS had no impact on the filtration performance, but had an impact on the frequency of CEBs. The value of 18 hours was close but exceeded the limit interval of CEBs (every 12 h) for the sustainability of the UF process (Field and Pearce, 2011). Regarding the reversible and irreversible resistances, fouling generated during the bloom was greater and faster than that in the case of seawater filtration, by an increase factor of 2 and 1.25, respectively. In comparison with CB, the efficiency of AB was higher due to the stronger fouling, as indicated by the circles in Fig. 9(b). To reach a membrane resistance with a ratio of 4/11 to the overall resistance during the bloom, only a duration of 1 600 min was required against the duration of 2 500 min in the standard seawater operation.

The efficiency of CEBs in eliminating the fouling resistant to backwashes was also investigated. Fig. 10 shows the obtained permeability after each CEB in the two-month period. In the case of seawater with different water qualities, CEBs performed with the UF unit led to a permeability of approximately 750 L/(h·m²·bar). This cleaning procedure eliminated resistant fouling even in the case of a bloom with high concentrations of TSS and plankton inside membranes. After the bloom, the permeability was recovered with a lightly lower efficiency at first, and then the following CEBs allowed a total recovery. The stability of the permeability and the resistance of the process were confirmed.

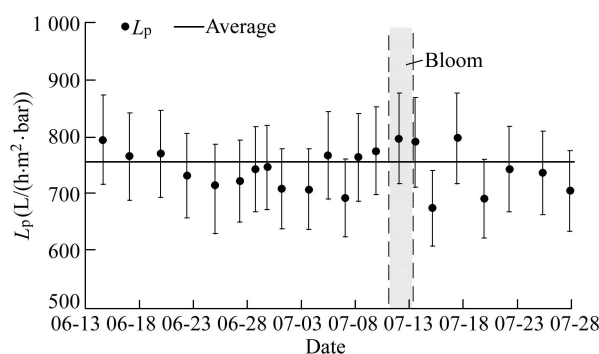


Fig. 10. Permeability after CEBs (averaged $L_p = 754 \text{ L}/(\text{h}\cdot\text{m}^2\cdot\text{bar})$).

4. Conclusions

The objective of this study was to demonstrate the resistance of UF membranes when encountering natural blooms that highlight the limitation of conventional water treatment processes used in shellfish production. UF is the only process that can deliver water with a low TSS concentration, free from living phytoplankton, zooplankton, *Vibrio*, and total bacteria. The hydraulic performance of the UF process was impacted to a greater degree by coastal blooms, with a higher CEB frequency than in the case of seawater filtration, but the CEB frequency in the case of blooms was less than the limit frequency of CEBs for the sustainability of the UF process. In addition, the permeability of the UF membrane could be recovered efficiently through CEBs, with a permeability of approximately 750 L/(h·m²·bar), confirming the resistance of the UF process in the case of severe water quality degradation. These results confirmed the ability of the UF process to treat natural seawater and produce water with constant quality even in the case of a bloom. In contrast to the optimized process combining filtration and UV disinfection, protection of oyster from harmful bacteria and parasites was achieved by use of the UF process. This study also confirmed that the UF process is useful to industry due to its ability to secure facilities and industrial plants from algal blooms. Researchers could build upon these findings to study the retention of toxins from microalgae. In fact, the high retention rate of phytoplankton by UF membranes creates a stress, which could lead to the release of potentially harmful toxins. Given that their sizes are smaller than the pore sizes of UF membranes, these toxins can pass through the membranes.

Declaration of competing interest

The authors declare no conflicts of interest.

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